STUDIES OF HEAVY-ION REACTIONS AND TRANSURANIC NUCLEI

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I. Abstract

Progress has been made in understanding exotic modes of the disintegration of highly excited nuclei within an extended statistical model. This understanding encompasses the occurrence of multi-cluster decay and an increased nuclear stability due to expansion cooling. In these modes of nuclear transmutation, the nuclear surface is seen to play a dominant role. Within the framework of simple classical models, it is possible to demonstrate the importance of viscous properties of nuclei, in particular of their surface matter. A clear understanding of detailed properties assumed for colliding nuclei in using theoretical simulations of complex nuclear interactions turned out to be required for a realistic interpretation of model predictions.

The experimental exploration of heavy-ion reaction mechanisms in exclusive $4\pi$ studies has been sharpened through the development of realistic modeling of detector effects. Several specific correlations of experimental observables with the numbers of free neutrons observed in coincidence have been shown to distinguish between competing theoretical models of the collision dynamics and associated transport phenomena, thus providing a powerful analytical tool. The excitation function of reaction phenomena has been studied for the $^{209}$Bi+$^{136}$Xe reaction, spanning a large range of bombarding energies. The study reveals a gradual evolution of the dissipative reaction mechanism which is seen to dominate heavy-ion reactions still at and beyond 60 MeV per nucleon. Specification of experimental heavy-ion reaction studies using high resolution multi-detector arrays requires the development of new methods of data mining, which are pursued by the research group together with other segments of an international collaboration.

The production of light and intermediate-mass clusters in proton induced reactions has important applications, for example in the transmutation of nuclear waste and the generation of secondary particle beams. We have found that commonly used models exhibit an inability to account for important details of the nuclear disintegration process, a problem to be addressed in future research. Nuclear fission at high excitations and its statistical competition with other nuclear decay modes belongs to the processes that still need elucidation. We have likely observed a materialization of important new nuclear surface effects.

In addition to fundamental research of nuclear transport processes, applied studies have resulted in new instruments and methods for nuclear applications. New systematic work has been performed on tritium decontamination of materials used in fusion energy research. This work has important applications in education and training of scientific personnel in radio-chemistry.
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II. Introduction

This report describes the research activities and results obtained by the University of Rochester Nuclear Chemistry group over the period from September 2004 through August/September 2005. The research program of the UR Nuclear Chemistry group is supported by the United States Department of Energy under Grant DE-FG02-88ER40414. The group has active experimental programs at the Laboratorio Nazionali del Sud in Catania/Italy, at the COSY accelerator facility of the Forschungszentrum Jülich in Germany, and at the National Superconducting Cyclotron Laboratory of Michigan State University in Lansing/MI.

Research projects underway have been pursued jointly with nuclear science groups from the Universities of Catania and Milan, from the LNS Catania, from Washington University (St. Louis), Oregon State University (Corvallis), the German Hahn-Meitner-Institut Berlin, from the Forschungszentrum Jülich, the French Laboratory GANIL in Caen, as well as with nuclear physics groups at the Universities of Warsaw and Cracow, Poland. In addition, we have started a new technical collaboration with the University of Rochester Laboratory for Laser Energetics (LLE) on transport phenomena of radioactive species in materials important for fusion energy research. This latter program has significant educational aspects.

The main thrust of the group’s research has been directed toward the exploration of nuclear reaction and decay mechanisms, induced by intermediate-energy heavy ions, and of fission, induced by relativistic protons. The most important results obtained during the past grant period include, on the theoretical side,

- A further theoretical development of our ideas on the importance of the surface entropy for the statistical decay of hot nuclei;
- The prediction of increased nuclear meta-stability at high excitations through adiabatic cooling;
- The recognition of the importance of nucleon-nucleon correlations for the thermodynamics of finite nuclei, here modeled in terms of an effective nucleonic mass, which depends on surface properties;
- The natural and consistent explanation of a set of previously confusing or conflicting experimental observations, like the character of the apparent caloric curve, the “softening” of Coulomb barriers, the statistical independence of clusters emitted in heavy-ion reactions, the Arrhenius-like behavior of cluster multiplicities.
These theoretical ideas have now cumulated in the formulation of a unified view of nuclear decay phenomenology. It has been shown that a consistent incorporation of thermal excitation of surface oscillation modes into the traditional scenario of the compound nucleus decay leads to a unified description of the “classical” compound nucleus decay, fission-like processes, intermediate-mass fragment production, and multifragmentation. Furthermore, it is shown that such a unified compound nucleus phenomenology including surface oscillations leads to the same approximate technical or mathematical apparatus that is used (albeit with different justification) in other models and appears to reproduce a host of experimental observations on intermediate-mass cluster fragment (IMF) production.

In the context of this broader view, particle evaporation rates from excited nuclear systems at equilibrium matter density have been studied within the Harmonic-Interaction Fermi Gas Model (HIFGM) combined with Weisskopf’s detailed balance approach. It has been found that thermal expansion of a hot nucleus, as described quantitatively by HIFGM, leads to a significant retardation of particle emission, extending the validity of Weisskopf’s approach to the domain of excitation energies in excess of 8 MeV/u. The decay of such highly excited nuclei is strongly influenced by surface instabilities.

Nucleus-nucleus reactions are influenced by structure and response of the nuclear surface. In particular, as shown in a schematic study of (“row-on-row”) collisions between linear Lennard-Jones nuclei, the prompt dynamic disintegration of interaction partners depends very much on the internal structure of the produced clusters, here modeled in terms of nuclear viscosity. This realization suggests ways to improve current molecular-dynamics computer models (e.g., [Luk93]) which typically define complex reaction products in terms of criteria of random proximity of nucleons in phase space.

The viability of current transport models is tested by comparing the stabilities of a model $^{197}$Au nucleus in its ground state, as modeled in various realizations of the BUU/BNV transport theory, which is the most popular simulation tool in current heavy-ion research. Three different approaches in calculating nuclear ground state properties are compared: calculations based on pure mean field, on mean field with numerical s.p. population constraints [Bon00], and one based on the Boltzmann equation with mean field and nucleon-nucleon collision terms[Bau93, Bon00]. These exercises have provided realistic ideas of the range of applicability and the accuracy of the various approaches and have facilitated students’ exploration of corresponding Ph.D. topics.

Experimentally, the group has
- Demonstrated the dominance of the dissipative reaction mechanism for all heavy-ion reaction systems studied, via observed PLF energy-angle and other correlations;
- Reconstructed the total excitation energies of projectile-like (PLF) and target-like (TLF) interaction partners in $^{209}$Bi+$^{136}$Xe collisions at E/A $= 28$ MeV to 62 MeV;
- Demonstrated that, for $^{209}$Bi+$^{136}$Xe, at E/A = 28 to 62 MeV, the excitation energy division between PLF and TLF equilibrates gradually with decreasing impact parameter;
- Demonstrated unexpectedly long life times of hot reaction primaries against particle decay;
- Separated non-statistical from sequential cluster emission for different regions of impact parameters;
- Explained previously puzzling correlations between average sizes of clusters emitted in $^{209}$Bi+$^{136}$Xe reactions and thermal excitation in terms of statistical emission processes;
- Demonstrated an abrupt enhancement of fission relative to particle emission in p-induced reactions on heavy targets at excitations of app. 3-4 MeV/nucleon, likely caused by surface instability;
- Demonstrated a non-coalescence mechanism of complex-particle emission in in-tranuclear cascades induced by relativistic protons.

Emphasized in this report are experimental multidimensional joint distributions of neutrons and charged reaction products. These distributions, analyzed for the $^{136}$Xe+$^{209}$Bi reaction at E/A = 40 MeV, exhibit several different types of prominent correlation patterns. Some of these correlations have simple explanations in terms of, e.g., the overall excitation energy, and can be understood in the framework of several current statistical decay theories. In contrast, several of the observed correlation patterns clearly differentiate between possible reaction scenarios. This property makes these correlations a useful tool for probing reactions scenarios, which has a utility that is very different from the inclusive yields of individual reaction products that are available in typical experiments.

Included is also a relatively brief rendition of the status of analysis of the excitation function of the $^{209}$Bi+$^{136}$Xe reaction, which has now allowed us to follow the phenomenology of reaction features for the $^{136}$Xe+$^{209}$Bi reaction from the early experiments at E/A = 7, 8.3, 9.5, and 11 MeV to now 28, 40, (55) and 62 MeV. The smoothness of the observed trends is truly impressive. For example, the reaction cross section is domi-
nated by dissipative binary reactions of well-defined projectile and target-like fragments. Correlations between the kinetic energy and the deflection angle of projectile-like fragments (Wilczynski Plot) are seen to exhibit features characteristic of dissipative orbiting, commonly found at bombarding energies of a few MeV/nucleon above the interaction barrier. On the other hand, the Galilei-invariant velocity distributions of various charged reaction products demonstrate the presence of a third, intermediate-velocity source of emitted fragments, which is less conspicuous at low energies. While the production of light-charged particles can be attributed mainly to the process of evaporation from excited projectile- and target-like fragment, at high excitation energies the intermediate-velocity source appears responsible for a large fraction of the observed intermediate-mass fragment yields. Fragments emitted from the intermediate velocity source appear to be produced dynamically in the overlap zone of the projectile and target nuclei.

Our CECIL experiment series at the LNS/Catania utilizes the CHIMERA 4π charged-particle detector with the currently highest granularity and efficiency available anywhere. The object of the first experiment in the series is to test a proposed theoretical mechanism for statistical, surface entropy driven multi-fragmentation. The properties of the detector and electronics set-up allow for the detection of a wide range of reaction products. In recent years, the field of multi-fragmentation has come to rely increasingly on complex 4π detectors that provide a great amount of information but require an approach to data mining that is new to our field. To aid in this effort, software is being developed for the purpose of automating the necessary tasks of particle identification and energy calibration as much as possible. These significant efforts are invested by our group in collaboration with others to the benefit of several experiments already conducted and the upcoming next CECIL experiment.

Our recent 1.2-GeV p-A experiments with the double-4π NESSI setup has presented new challenges to theory to explain measured yields of complex nuclear clusters emitted from highly excited target nuclei. While fits for individual, light target nuclei are possible with a coalescence approach, this model fails to reproduce the observed trends for cluster emission from a range of heavier targets. In particular, the model fails to account for large emission probabilities of neutron rich clusters from heavy targets.

The same series of p-A experiments may have provided new evidence for a surface entropy driven type of fission instability predicted by our HIFG model for statistical nuclear decay. The experiments studied fission and particle emission induced by 2.5-GeV protons in targets of Au, Bi, and U. Consistently, for increasing excitation energy, both data and cascade simulation calculations show an increasing fission probability for Au, a
decreasing probability for U, and an intermediate behavior for Bi. Contrary to the systematics. Surprisingly, in these calculations no account needed to be taken of the usual finite transient times for fission. These observations indicate that at high excitation energies the familiar statistical competition between particle evaporation and fission is strongly altered. Such trends have been predicted by our HIFG model. Dedicated new experiments are required to investigate the effect further.

In addition to the experimental and theoretical work described above, the group has developed instruments and methods for nuclear physics research and applications. They include the design and testing of several methods of particle identification utilizing characteristic differences in the pulse shapes produced by these particles in large-area silicon detectors.

The performance of a simple method of discriminating slow pulses from charge-sensitive preamplifiers according to their rise times has been analyzed for the range of rise times from 10 ns – 500 ns. The method is based on measuring, using fast peak-sensing ADCs, peak values of two pulses derived from the raw preamplifier pulses by the way of using two amplifiers with largely differing shaping times. It is found that for the charges injected corresponding to energy deposits in silicon detectors of a few tens of MeV, the resolution in rise time that can be achieved is of the order of 1 ns. The method is expected to be useful in experiments involving large-area silicon detectors, whenever the mechanics of charge collection leads to a desired spread in pulse rise times for different species of impinging particles, such as is the case with $\Delta E$ detector constituents of the CHIMERA multi-detector array.

These developments have been based on preceding work on the pulse shape discrimination method applied to CHIMERA silicon detectors. Here, it has been found that in principle, it is technically feasible to achieve low thresholds for particle identification via pulse shape discrimination simultaneously with fast timing appropriate for time-of-flight measurement.

For a new R&D project, we have started a collaboration with the University of Rochester Laboratory for Laser Energetics, in which we set up and run several experiments addressing the transport of tritium gas, which is one of the major components in energy technology using laser-induced fusion. The first such experiments measured tritium adsorption at metallic surfaces and its transport through solid metal alloy lattices, as well as subsequent decontamination through desorption. This project has a typical radiation chemistry framework and complements the experimental training students in the group receive.
During the Academic Year 2004/05, the Principal Investigator spent approximately 30% of his time to research and student training supported by this grant, increasing to 90% during summer of 2005. The Senior Scientist (J.T.) and the Research Associate spent 100% of their efforts on these research projects.

During the Academic Year 2004/05 resident (Z.C., M.H., I.P.) graduate students and one undergraduate (M.Q.) have worked 100% on grant research. During summer of 2005, resident (Z.C., M.H., I.P.) and incoming (M.Q.) graduate students have worked on grant research. Mr. Mark Houck, and Ms. Iwona Pawelczak have worked full time on grant research. Both participated in a successful set of PSD-test experiments and in a collaboration meeting this last summer at LNS Catania. Both, Mr. Zachary Chambers and Mr. Michael Quinlan spent several weeks during summer of 2005 on the tritium transport and decontamination experiment, partially supported by an LLE fellowship.

To the best of our knowledge, we have complied with all grant requirements. It is a pleasure to acknowledge the direct support of our research program by the United States Department of Energy.

In addition, we are grateful to the European research community for giving us support and access to their COSY and LNS laboratories and accelerator facilities.

W. Udo Schröder
Rochester, September 2005
III. Research Program
A Unified Phenomenology of Sequential Compound Nucleus Decay, Intermediate-Mass Fragment Production, and Multifragmentation

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Abstract

It is shown that a consistent incorporation of thermal excitation of surface oscillation modes into a general scenario of the compound nucleus decay leads to a unified description of the “classical” compound nucleus decay, fission-like processes, intermediate-mass fragment production, and multifragmentation. Further, it is shown that such a unified phenomenology of a compound nucleus with surface oscillations leads to the same approximate technical or mathematical apparatus that is used (albeit on different grounds) by other models that appear to be successful in describing a host of experimental observations on intermediate-mass fragment (IMF) production.
I. INTRODUCTION

In the statistical treatment of the decay of excited nuclear systems, conceptually different classes of models are commonly applied for the low-energy domain, on one hand and for the higher-energy domain, on the other hand. One practical reason apparently necessitating the use of a distinctly different decay scenario or phenomenology at higher energies can be related to the experimentally observed copious production of intermediate-mass fragments (IMFs) at these elevated excitation energies and to the difficulty of explaining such a production in terms of the decay of a classical (metastable) compound system. The term “classical” compound nucleus refers here to a nuclear system characterized by ground-state matter distribution, subject to thermal shape oscillations responsible, among other things, for fission. The notion of such a compound nucleus proved sufficient for explaining the decay patterns, including fission, observed at low excitation energies, where IMF production was neither expected (on the grounds of high fragment separation saddles), nor observed.

The observation of a copious IMF production at higher excitation energies, where the temperatures involved were still significantly lower than the theoretical separation saddles for IMFs, has, thus, given rise to a large number of experimental and theoretical studies considering decay scenarios qualitatively different from the classical compound nucleus decay. Such scenarios often refer to liquid-gas transition and spinodal instabilities in bulk nuclear matter, believed to be brought to a desirable metastable “fragmentation” state in the course of the collision history. They do offer an attractive prospect of gaining an insight into the liquid-gas phase transitions in nuclear matter and in small systems, in general. However, these prospects come at the expense of the clarity of the proposed scenario, when sound notions of nuclear matter and charge distributions and their evolution (statistical or otherwise) are being replaced by abstract or mathematical notions of freezeout volume filled with rattling balls, statistical breakup, lattice, percolation, virtual gas, etc., for which no plausible or meaningful physical counterpart has been proposed.

For the reasons that become clear later below, the present paper takes as a reference point two types of models enjoying apparent successes in describing a large volume
of experimental observations. These are the SMM[1] and MMMC[2] multifragmentation models built on the concept of freezeout (dubbed here, freezeout-based models or FBM) on the one hand, and the Fisher’s droplet model [3] adapted for purposes of nuclear matter (dubbed here Nuclear Fisher Droplet Model NFDM) [4] on the other hand. Both classes of models part with the concept of a classical compound nucleus and while mutually exclusive, both refer to liquid-gas phase transition. Their apparent successes in reproducing quantitatively many salient characteristics of IMF production have contributed to their acceptance by the research community in spite of their many conceptual shortcomings, [5] ill-defined physical scenario, and mutually (FBMs vs. NFDM) exclusive assumptions.

It is not the purpose of this paper to revive or reinforce the criticism [5] of the above reference models on conceptual grounds. Rather, it is to show that the experimental observations that appear to support these models have a natural explanation within the framework of the recently proposed mechanism of massive fragment production relying on the role of surface entropy or surface free energy [6]. This latter model can be characterized as a compound nucleus model with surface entropy or CNSEM, in contrast to the classical compound nucleus model, where the role of surface entropy is customarily neglected (other than, perhaps, in fission).

The role of the surface entropy in general and in CNSEM, in particular, is to reduce surface tension with increasing excitation energy. As a result, the likelihood for the system to arrive at one of the very many fragment separation saddle points increases and, correspondingly, so does the fragment emission rate. Furthermore, as surface tension decreases with increasing temperature, more and more fragments are produced. Eventually, when surface tension vanishes, all multi-fragment saddle configurations become equally likely, resulting in statistical multifragmentation. So far, for the sake of simplicity, CNSEM has been formulated within the framework of a harmonic interaction Fermi gas model, but the basic premises hold for any model of nuclear matter with finite range nucleon-nucleon interaction.

The reason that the above two classes of established models, the freezeout-based models and the nuclear Fisher’s droplet model, are chosen here as reference models is a practical one. Namely, CNSEM can be shown to lead to essentially identical technical
or numerical expressions that are responsible for the apparent successes of the reference models and, hence, it will necessarily agree with the experiment to the same (impressive) extent to which the reference models agree. At the same time, CNSEM offers an alternative, yet clear physical interpretation or meaning for various critical notions of the reference models in terms of properties of a (metastable) compound nucleus undergoing thermal shape fluctuations. In the case of the freezeout-based models, among these notions are the freezeout volume, breakup and the breakup configuration, and the crucial (see Section IV) ”rattling” space - the excess part of the freezeout volume that allows any single fragment to move around and bounce off its neighboring fragments, as well as off the (unphysical) boundary of the freezeout volume. In the case of the Nuclear Fisher’s Droplet Model, CNSEM provides a simple interpretation of the particular form of the temperature-dependence of the surface free energy used by NFDM, and an explanation for the apparent vanishing of the Coulomb energy of touching spheres with increasing temperature, all essential ingredients of the model.

CNSEM lacks the appeal of novelty or exotics of phenomena such as, e.g., phase transitions and spinodal decomposition in small unconfined quantal systems interacting via finite-range forces. Accordingly, it does not offer any meaningful link of the observed fragment production patterns to the liquid-gas phase transitions in nuclear matter. What it does offer instead, is a clear and robust picture of the evolution of a compound nucleus with increasing excitation energy, free of unproven (ad-hoc) and abstract notions. It offers also a potentially important link between the observed fragment yields and the properties of hot and diffuse nuclear surface domain. Importantly, the picture offered by CNSEM unifies the description of all statistical decay modes of an excited (metastable) nuclear system, including particle evaporation, fission, IMF production and multifragmentation. Here, fragment production and multifragmentation are surface-related phenomena, reflecting the reduction and, eventually, the vanishing of surface tension with increasing excitation energy. Within the framework of CNSEM, their phenomenology is essentially the same as the phenomenology of fission, where the metastable system undergoes surface oscillations leading occasionally to saddle configurations.

It is important to stress that the description of the decay of excited nuclear system, offered by CNSEM, does not extend qualitatively beyond the well defined and understood
concepts of particle emission and fission. In particular, the notion of surface entropy that is at the crux of this description is not a new one. In the context of the present study, this notion has been used in fission studies to justify enhanced entropy of the system at saddle shapes with respect to spherical shapes. It is also at the crux of the Fisher's droplet model [3], albeit in reference to surface between the liquid and gaseous phases. Also the idea of a sequential emission of fragments is not a new one. It is, e.g., entertained in Expanding Emitting Source Model (EESM) [7], albeit with a different, non-thermal mechanics of overcoming fragment separation saddles [5].

The present paper is organized as follows. In Section II, the essentials of the Compound Nucleus Model with Surface Entropy are presented along with a discussion of the guiding role of the LeChâtelier Principle. In Section III, consistency of CNSEM with the NFDM parameterization of fragment yields is discussed and in Section IV, its technical equivalence to freezeout-based models (FBM) SMM[1] and MMMC[2] is revealed. Summary and Discussion is presented in Section V.

II. ESSENTIALS OF COMPOUND NUCLEUS MODEL WITH SURFACE ENTROPY

The concept of a compound nucleus is a time-proven one, going back to the early years of nuclear science. Its usefulness stems largely from the fact that it offers a general framework for modelling the decay of excited nuclear systems [8–10] produced under various scenarios, but believed to be thermally equilibrated. It has been commonly used not only in the domain of low to moderately-high excitation energies, but also at higher energies where multiple production of intermediate-mass fragments (IMF) is observed. In that latter case, however, its “classical” implementations fail to account for the IMF production, but often account reasonably well for the neutron and light charged particle yields.

Commonly, the compound nucleus is being thought of as an excited nucleus in a state of thermal equilibrium. It is also understood that the equilibrium is not complete, as the system is bound to decay over a period of time, with the decay rates and branching ratios being the quantities of interest.
In the present paper, the compound nucleus is defined more broadly as a system of interacting nucleons in a (transient) state of constrained microcanonical equilibrium, where all constituents are trapped within a family of possible shapes of the system. In contrast to models utilizing the concept of a freezeout volume (FBM), [1, 2] in CNESM the system is kept (transiently) together by nuclear forces and not by an unphysical, active boundary of a thought freezeout volume. In practical terms, CNSEM requires all particles to interact within a volume that maximizes entropy for the model family of density profiles. While at excitation energies in excess of nuclear binding energy no such volume exists (entropy increases indefinitely with increasing volume) and the proposed model is inapplicable, at lower excitation energies the “stable” (maximum entropy) volume reflects the effects of thermal expansion of nuclear matter and shape deformation degrees of freedom.

Within its constrained microcanonical logic, CNSEM considers explicitly effects of thermal expansion and thermal surface oscillations. The fact that excited nuclear matter expands plays an important role in stabilizing the compound nucleus with respect to (fast) neutron emission [11]. This is so, because a thermally expanded nucleus has lower temperature, compared to that kept at ground-state density and because the particle evaporation rate scales quadratically with temperature. The temperature is lower firstly, because part of the excitation energy is tied up in potential energy of expansion and, secondly, because (for Fermi gas) the level density parameter \( a \) scales with the matter density as \( \rho^{-2/3} \). For the infinite nuclear matter, a simple relationship between the excitation energy and the equilibrium matter density has been derived in the framework of the Fermi gas model and the harmonic approximation for the potential energy of expansion [6]

\[
\frac{\rho_{eq}}{\rho_o} = \frac{1}{4} \left(1 + \sqrt{9 - 8 \frac{E_{tot}}{E_{bind}}} \right). \tag{1}
\]

The trend described by Eq. 1 is illustrated in Fig. 1.

While thermal expansion plays a significant role in CNSEM, it is the role of the surface entropy or surface tension that makes the CNSEM quantitatively different from the “classical” compound nucleus as far as the description of IMF production or multifragmentation is concerned. This is so, because in the framework of the Fermi-gas model,
FIG. 1: Dependence of the equilibrium density of infinite nuclear matter on excitation energy, as calculated within the framework of harmonic interaction Fermi gas model [6].

on which CNSEM relies in its quantitative (but not qualitative) predictions, surface tension decreases with increasing excitation energy, vanishing altogether at a transition temperature given by [12]

\[ T_t = \sqrt{\frac{\epsilon_s}{\alpha_s}}, \]  

(2)

where \( \epsilon_s \) and \( \alpha_s \) are surface energy and surface level density parameters, respectively, both depending on temperature. Note that Eq. 2 follows simply from the requirement that surface free energy be zero

\[ F_s = (\epsilon_s - \alpha_s T^2)A^{\frac{2}{3}} = 0. \]  

(3)

The trends described by Eq. 3 under the assumption that the system expands thermally in selfsimilar manner, are illustrated in Fig. 2. As seen in this figure, surface
FIG. 2: Evolution of surface free energy with increasing excitation energy as predicted by Eq. 3.

tension is expected to vanish already at excitation energies below 8 MeV/nucleon, i.e., well within the range where copious production of IMFs is observed experimentally.

It is important to note that the logic of CNSEM is based on the assumption of the microcanonicity (albeit constrained) of the system and, thus, the proper physical quantity characterizing the state of the system is its total energy and not temperature. Accordingly, the population of possible macro-states of the system (shapes, matter density profiles, etc.) is governed by the entropy associated with such states, rather than free energy. Yet, the notion of free energy, with its Boltzmann factors and straightforward link to surface tension appears to allow one to assess more intuitively the effects of excitation on the behavior of the system and, especially, of its surface and the consequences for the stability of the system. Therefore, for didactic reasons, but also for the purpose of comparing the CNSEM formalism to the Nuclear Fisher Droplet Model [4] parame-
terization, references are made here often to free energy in general and to surface free energy, in particular.

Among the very many allowed states of the (constrained microcanonical) system are ones that are of special interest because they serve as transition states to exit channels. That is, whenever system arrives statistically at any of such states, it is driven dynamically apart by Coulomb and/or inertial forces into a particular exit channel. There are two distinct types of such configurations. In the first one, an individual nucleon or light charged cluster of nucleons has enough kinetic energy to leave the system, resulting in nucleon or light charged particle “evaporation”. The associated decay rate is here well described by Weisskopf’s formalism [13] based on the principle of detailed balance. The second type of decay configurations is characterized by a shape deformation to a point, where Coulomb or centrifugal forces drive fragments of the system apart. An example of such a configuration is the fission saddle-point configuration and, in the framework of CNSEM, IMF breakup-point configurations.

The probability for the system to arrive at a particular breakup configuration is related to the entropy associated with this macroscopic configuration and is given by ([6, 13])

\[ p \propto e^{S_{\text{Saddle}} - S_{\text{Eq.}}}, \]

where \( S_{\text{Saddle}} \) and \( S_{\text{Eq.}} \) are system entropies at the breakup saddle configuration and at equilibrium-density spherical shape, respectively. Note that Eq. 4 is meaningful only for \( S_{\text{Saddle}} < S_{\text{Eq.}} \).

It is important to note that at much reduced surface tension, such as is the case at high excitation energies, a distinction must be made between a classical transition or saddle state and what is here called a “break-up” state. This is so because at low excitation energies, arriving at a saddle configuration guarantees the separation of the system into the associated exit channel, i.e. the saddle is unconditional. The same is not true for high excitation energies where the behavior of the system is subject to large fluctuations. Here, saddle configurations are more compact (eventually, they become spherical) and arriving at any of them no longer guarantees decay into the associated exit channel, because the fluctuations may redirect the system into a different
FIG. 3: Conditional saddle configuration (left) will be driven to a multifragment scission point only when the fragment masses are fixed while the unconditional break-up configuration (right) will evolve to the respective scission with no conditions imposed.

exit channel. Hence, what would be an unconditional saddle at a low excitation energy, becomes a conditional saddle at high excitation energies, valid only for a “frozen” channel identity. It is not obvious how to deal with this problem and, specifically, how to find unconditional saddle or break-up configuration. At any rate, it is clear that such states must feature more prominently defined fragments than the compact conditional saddles. The relationship between the conditional saddle points and breakup configurations is illustrated schematically in Fig. 3. Note that the CNSEM breakup configurations differ from those implied by models based on the concept of freezeout (FBM) in an important way. Namely, in CNSEM, they are associated with shape (surface) fluctuations, while in FBMs they are thought to be associated with bulk instabilities, such as, perhaps, spinodal instabilities. In a stark contrast to CNSEM, FBMs do not spell out how exactly the system does break up.

While summarizing the evolution of the excited compound system with increasing excitation energy its is helpful to keep in mind the LeChâtelier’s Principle, stating that in response to a stimulus (excitation energy), the system will evolve macroscopically (adapt its matter distribution) so as to minimize the results of the stimulus, which is
in this case the increase in temperature. This principle is rooted in the Second Law of thermodynamics, which calls for an isolated system to maximize its entropy, with an obvious effect of keeping the temperature as low as possible and, as a byproduct, keeping neutron evaporation in check, thus, extending the decay time scales.

The way for the system to maximize entropy, i.e., to access more micro-states is by gaining access to a larger geometrical volume at the expense of potential energy. Thus, the excited system will expand thermally to reach the equilibrium density proper for the given excitation energy. But, it will also increase the diffuseness of its surface domain to extend even further out at the expense of potential surface energy [14]. And, importantly it will enter surface oscillations, with the same aim of reaching pockets of geometrical space at a possibly low cost in potential energy. The latter is also clear from the principle of equipartition of available excitation energy between all degrees of freedom, surface oscillations being one of them. Clearly, a thermally oscillating system, in agreement with the principle of equipartition, will have higher entropy than a system that is prevented from oscillating.

With increasing excitation energy, surface entropy increases, and surface tension decreases accordingly, inducing larger and larger thermal shape oscillations. And all of this happens while keeping the increase in temperature in check, thus allowing for a more complete equilibration of various shape degrees of freedom. Occasionally, and more so with increasing excitation energy, the system will arrive at any of the very many microstates corresponding to any of the (deformed) macroscopic unconditional (break-up) saddle configurations. Subsequently, the system is dynamically driven to scission and, possibly, to a multifragment scission at high excitation energies. The very essence of the surface entropy lies in the fact of multiplying the number of microstates corresponding to deformed macroscopic configurations.

One may speculate, in a spirit of the “universality” rule often cited with respect to phase transitions, that vanishing of surface tension will result in signatures characteristic of phase transitions. For example, as any breakup saddle shape becomes equally likely, one would expect fragment masses and charges to be distributed according to power laws.
III. CONSISTENCY OF THE CNSEM SCENARIO WITH THE NFDM PARAMETERIZATION OF FRAGMENT YIELDS

One notes that the Nuclear Fisher’s Droplet Model (NFDM) [4] postulates a successful parameterization of the fragment yields very similar to that arrived at by CNSEM, based on the Fermi gas model. Since NFDM does not provide a basis for this parameterization in terms of a perceived physical scenario with its characteristic distributions of nuclear matter and charges, it could be argued, for comparative purposes only (i.e., comparing the merits of CNSEM versus those of NFDM), that exactly the same parameterization is applicable to the CNSEM. However, as discussed further below in more absolute, and not comparative, terms CNSEM does, in fact, provide for a link between the above successful parameterization and the perceived decay scenario of CNSEM. On the other hand, the fact that NFDM does not provide a meaningful physical scenario for the same parameterization is also clear from the discussion below.

NFDM expresses [4] the abundance of cluster production nominally in terms of surface free energy of a droplet (which, supposedly, is identified with a detectable cluster), a very large (compared to the final yield) and strongly excitation energy dependent (many orders of magnitude over the excitation energy range of interest) Coulomb “correction”, and a minor chemical potential adjustment $A\Delta\mu$.

\[
n_A = n_0 A^{-\tau} e^{\left(\frac{\Delta\mu + E_{\text{Coul}}}{T} - c_0 \sigma A^\sigma\right)},
\]

where the Coulomb “correction” is postulated in the form of

\[
E_{\text{Coul}} = \frac{e^2}{4\pi \varepsilon_0 r_o} \frac{(Z_o - Z)Z}{(A_o - A)^{1/3} + A^{1/3}} (1 - e^{-x\varepsilon})
\]

and the surface free energy is represented by

\[
c_0 \varepsilon A^\sigma.
\]

In Eqs. 5-7, $A_o$ and $Z_o$ are system mass and atomic numbers, respectively, $A$ and $Z$ are fragment mass and atomic numbers, respectively, $\sigma$ is a dimensionality-related exponent, $\tau$ is a topological exponent, $x$ is a fit parameter, $r_o = 1.2 fm$ is a radius parameter, $\Delta\mu$ represents nominally the difference between the chemical potentials of
gaseous phase and the liquid phase of the formed droplet/cluster, and \( \varepsilon = (T_c - T)/T_c \) expresses the relative “distance” from the critical temperature \( T_c \).

While the original expression by CNSEM, relating the fragment yields to the cost in entropy for passing the fragment separation saddle does not resemble much of Eq. 5, the similarity becomes obvious when one approximates the cost in entropy by the relative (to the system temperature) cost in (Helmholtz) free energy, i.e.,

\[
\Delta S = S_f - S_i = \frac{E_{tot}^* - F_f}{T_f} + \frac{E_{tot}^* - F_i}{T_i},
\]

where \( E_{tot}^* \) is the total energy of the system and \( F_f \) and \( F_i \) are the transition-state and the compound nucleus free energies, respectively.

By assuming that the change in temperature is small compared to temperature (canonical approximation), i.e., \( T_f = T_i = T \) and by expressing the change in free energy in terms of associated differences in potential (Coulomb) energy and surface free energy, for spherical compound system, on the one hand, and the transition-state (saddle) configurations on the other hand, one obtains

\[
\Delta S = (E_{Coul}(A_o, Z_o) + E_{Coul}(A, Z) + E_{Coul}(A_o - A, Z_o - Z) - E_{Coul}^*) R_{Coul}^{Coul} \frac{\Delta F_{Surf}}{T} R_{Surf}^{Coul},
\]

where \( E_{Coul}(A_o, Z_o) \), \( E_{Coul}(A_o - A, Z_o - Z) \), and \( E_{Coul}(A, Z) \) are Coulomb energies of spherical compound and residual nuclei and the fragment, \( E_{Coul}^* \) is relative Coulomb energy of the fragment and residual nucleus in a touching-spheres configuration, \( \Delta F_{s} \) is the increase in surface free energy (while transiting from the spherical compound to touching-spheres configuration), and \( R_{Coul}^{Coul} \) and \( R_{Surf}^{Coul} \) are ratios of Coulomb energies and surface free energies taken at saddle and touching spheres configurations, respectively.

Surface free energy \( F_{Surf} \) of a spherical Fermi gas system of mass number \( A_o \) can be expressed by Eq. 3 and then, accordingly, the difference in surface free energy of a spherical compound system and the touching-sphere fragmentation configuration is

\[
\Delta F_{Surf} = (c_o - \alpha_{Surf} T^2)[A^{2/3} + (A_o - A)^{2/3} - A_o^{2/3}].
\]
By combining Eqs. 4, 3, and 10 one arrives at an expression for the relative fragment yields, structurally similar to Eq. 5, but different in several important details.

The similarity is in the presence of two leading terms, the surface free energy and Coulomb energy. Both these terms decrease with increasing temperature at rates that cannot be reliably calculated at the present time, and both are bound to vanish at some elevated temperature. Thus, the particular way the terms vanish must be left to a fitting process. The latter must, however, be subject to constraints derived from laws of thermodynamics and requirements of consistency with the perceived decay scenario.

Regarding the surface free energy term, one notes that the latter must be a strictly concave function of temperature, a requirement that is clearly violated by Eq. 7, often referred to as Fisher’s scaling. One notes in this respect that the origin and the authorship of Eq. 7 are, in fact, unknown as such an expression does not appear in the referenced publication, Ref. [3]. The requirement for the free energy to be a strictly concave function of temperature follows from the fact that the (partial) derivative of free energy over temperature is entropy taken with negative sign, i.e.

\[
\frac{\partial F}{\partial T} = -S
\]

and that entropy itself is an increasing function of temperature.

While within the NFDM description, surface free energy is a linear function of the NFDM temperature, its parameterization does represent a strictly concave function of the CNSEM temperature. This is illustrated in Fig. 4 and it is so because the CNSEM temperature increases with increasing excitation energy at rates consistent with the LeChâtelier Principle, that are lower than the rates postulated by NFDM. The latter are, in fact, in contradiction to that principle and to a body of recent experimental observations [15]. Hence, the NFDM parameterization of surface free energy (but not temperature) is consistent with CNSEM, such that the parameters of the actual fit are to be interpreted in terms of rates at which surface profile changes with temperature and at which the shape of the saddle configuration approaches spherical shape (see parameter \( R_{Saddle}^{Surf} \) in Eq. 9).

The caloric curves used by NFDM and CNSEM are compared to each other and to the limiting line associated with the LeChâtelier’s Principle, in Fig. 5.
FIG. 4: Dependence of surface free energy on temperature for the temperatures calculated using NFDM (solid line) and CNSEM (dashed line) temperature scale. In both cases, the free energy is calculated as a function of excitation energy using the NFDM parameterization, but only in the case of CNSEM is its dependence on temperature strictly concave.

As far as the NFDM parameterization of the Coulomb term is concerned, it strikes by the positive sign in front of what appears as the fragment-residue Coulomb interaction term calculated for two touching spheres. It strikes also by its large magnitude and its strong temperature dependence that is postulated in NFDM, but has not been justified by NFDM in terms of spacial distribution of charges for the perceived scenario and, as a matter of fact, in any meaningful terms. One notes that with its large magnitude and strong variation with temperature it is, in fact, the Coulomb “correction” that is responsible for the scaling claimed in Ref. [4]. This is illustrated in Fig. 6, where the final, quasi-linear “scaling” (heavy solid line, labelled “A=A(T)”) is compared with its
FIG. 5: Caloric curves as used by NFDM (solid line) and CNSEM (long-dashed line). The LeChâtelier’s limiting line is illustrated by the short-dashed line.

constituent Coulomb contribution (dash-dotted line), the latter varying by 6 orders of magnitude over the range of temperatures of interest.

While CNSEM places properly negative sign in front of the inter-fragment Coulomb interaction term, it is still consistent with the NFDM parameterization of the Coulomb correction, which uses the opposite sign. This is so because CNSEM includes additionally Coulomb energies of spherical individual fragments and of the spherical compound system as a whole. The resulting complete Coulomb correction shows then a dependence on the fragment mass very similar to that used by the NFDM parameterization, except for a normalization factor. The latter is illustrated in Fig. 7, where the two corrections are seen to differ to a good approximation only by a constant factor, that can be readily absorbed into an overall normalization factor.
FIG. 6: Anatomy of the Fisher-like scaling in NFDM. Heavy solid line illustrates the abundance of fragments with atomic number $Z = 11$, calculated using the NFDM parameterization (Eqs. 5 and 6) and assuming different fragment masses for different points along the scaling line. The magnitude of the Coulomb correction is illustrated by the dash-dotted line, while the result of calculations for fixed fragment mass number $A=22$ is shown by short-dashed line. Vertical dotted line indicates the critical temperature $T_c$. The long-dashed line illustrates the Boltzmann factor, meaningful only for $\Delta F > 0$, i.e. below the $\Delta F = 0$ limiting (dotted horizontal) line.

Regarding the temperature dependence of the Coulomb term postulated by NFDM, this has a natural explanation within the framework of CNSEM in terms of the saddle state configuration becoming more and more compact as the surface tension decreases. Eventually, with the surface tension reduced to zero, the saddle configuration is spherical, hence, Coulomb term is reduced to zero. Note, that the Coulomb interaction is inherently temperature independent and, therefore, any reduction in the Coulomb energy is possible
FIG. 7: Coulomb energy at zero temperature as used in NFDM (solid line) and CNSEM (long-dashed line). The short dashed line illustrates the quality of the agreement between the two models obtained when the SNSEM direct result is subjected to a suitable linear (with Z) transformation.

only via a respective change in the distribution of charges, a feature that appears difficult to reconcile with any conceivable scenario of Fisher’s droplet model.

In discussing the consistency of CNSEM with the NFDM parameterization and, by implication, with a large body of experimental data, it is important to note that NFDM assumes tacitly that primary fragments are formed mostly neutron rich, with the neutron “excess” varying with temperature, but without any penalty in terms of isospin asymmetry term. Such an assumption runs against what is known of isospin dependence of nuclear interactions and implies effectively reverse isospin fractionation. Yet, this assumption is essential for NFDM to be able to demonstrate what has been termed
FIG. 8: Dependence of fragment isospin asymmetry on inverse temperature for sodium fragments as postulated by NFDM. Note the very large cost of making highly iso-asymmetric fragments (dashed line and the associated rightmost abscissa labels).

“Fisher’s scaling”, with a little known fact that the many experimental points aligned along any single scaling line refer to different isotopes, albeit with a common atomic number. The significance of the variation of the isospin asymmetry of fragments with temperature for the “Fisher’s scaling” is illustrated in Fig. 6, where the “scaling” line (short dashes) for $Z = 11$ and fixed $A = 22$ is seen to deviate dramatically from the straight line obtained by assuming different mass numbers for different points along the “scaling line”. Fig. 8 illustrates the dependence of the fragment $N-Z$ asymmetry and the associated cost in isospin asymmetry energy $\Delta E_{iso}$ on inverse temperature for a representative sodium ($Z=11$) nuclide, as used by NFDM.

The reverse isospin fractionation as pertaining to fragment production would be
clearly inconsistent with CNSEM, as the latter must account for the isospin asymmetry energy in its energy balance and, as seen in Fig. 8, the energies involved are prohibitively high for the isospin asymmetries postulated by NFDM. Yet, CNSEM can readily adapt itself to the resulting scaling while keeping the fragment isospins consistent with maximum entropy (or minimum free energy). It can do this at the expense of adjusting the rate at which the surface free energy and the shape of the saddle configuration varies with temperature. Note, that without a detailed theory on how these quantities vary with excitation energy, one has to treat these rates as fit functions.

It may be tempting to deduce the dependence of the saddle-related factors $R_{\text{Coul}}^{\text{Saddle}}$ and $R_{\text{Surf}}^{\text{Saddle}}$ in Eq. 9, as well as the surface little-$a_S$, on temperature by requiring both, Coulomb and surface free energy terms to vanish at the same excitation energy where they change sign in NFDM, i.e. at $E^*/A_o = 3.8$ MeV. While possible (albeit ambiguous), such deduction is of little value in view of the facts that, on the one hand, CNSEM is valid only in the domain where $\Delta S < 0$ or $\Delta F > 0$ and that, on the other hand, the experimental data show no singularity around $E^*/A_o = 3.8$ MeV, where the two terms supposedly vanish according to NFDM. To secure the continuity of the predicted yield across the $T = T_c$ line (see Fig. 6), CNSEM must assume that the surface tension vanishes at a significantly higher excitation energy than the 3.8 MeV/nucleon deduced in ref. [4]. Note that by the same token, NFDM is meaningless at excitation energies in excess of approximately 3.4 MeV/nucleon, where $\Delta F$ crosses zero, turning negative.

To summarize the present chapter, CNSEM appears consistent with the NFDM parameterization of fragment yields and, by implication, with a large body of experimental data, while offering natural explanation for such seemingly strange phenomena as negative and temperature-dependent Coulomb interaction, linear (as opposed to strictly concave) dependence of free energy on temperature, and reverse isospin fractionation - production of increasingly iso-asymmetric fragments with increasing excitation energy.
IV. CNSEM AND FREEZEOUT-BASED MODELS OF NUCLEAR MULTI-FRAGMENTATION

As discussed further above, at elevated excitation energies the loss in entropy due to the deformation of the system is to a large extent offset by the gain in entropy due to enlarged volume of the diffuse surface domain. This translates into reduced surface tension, which in turn enhances the propensity of the system to large-amplitude surface oscillations. When the surface tension vanishes altogether (see Eq. 2), the system is almost equally likely to be at any saddle configuration and, more importantly, is quite likely to be in one of very many multi-fragment breakup configurations (see discussion in Section II). The latter are characterized by complex shapes, such that not one, but many individual portions of the nuclear matter will be driven by Coulomb forces toward scission and eventually separate from the rest of the system. The phenomenon will then manifest itself as a multi-fragment breakup and is beyond the scope of sequential fragment production models.

There is an obvious analogy of the above surface-instability based scenario and the scenarios of freezeout-based models (FBM), such as SMM [1] and MMMC [2], both envisioning a breakup configuration with a seemingly low matter density (freezeout density), when averaged over many configurations. In other words, the systems are spatially extended, with largely reduced Coulomb repulsion of individual fragments with the rest of the system, the matter distributions that can be probed experimentally by interferometric methods [16, 17].

Perhaps a more intriguing parallel to FBMs lies in the way CNSEM, on the one hand, and the FBMs, on the other hand arrive at high probabilities of realizing energetically costly and individually highly unlikely breakup configurations. In the case of CNSEM, it is the excess (as compared to bulk matter) entropy associated with increased surface area, or increased volume of low-density surface domain. This is what is here called surface entropy and its direct, but apparently under-appreciated, role is to immensely increase the number of micro-states corresponding to any individual macroscopic breakup configuration. While any individual micro-state is highly unlikely because of the prohibitively high cost in deformation energy, it becomes quite likely for one of the very
many microstates to become populated and, thus result in a multi-fragment decay.

There is an analogous mechanism behind the apparent successes of the freezeout-based models, except that the excess entropy now comes not from the increased surface area but from the availability of an excess volume, allowing the fragments to bounce off each other and, importantly, bounce back from the unphysical boundary of the freezeout volume. In the framework of FBMs, the system utilizes this excess volume by populating an immense number of “rattling” configurations, differing from each other only by spatial distribution of fragments and their individual momenta. It is the entropy associated with the “rattling” of fragments within the freezeout box that multiplies the number of highly unlikely individual multi-fragments states to the point where one of them might be populated on a reasonable time scale, i.e., before the system deexcites via particle evaporation. The importance of the rattling has largely gone unnoticed, or at least, its critical importance for the FBMs has not been discussed in published accounts on these models. Yet, it (the rattling) appears to contradict the customary narrative of FBMs presenting the freezeout configurations as the last points on possible system trajectories where the inter-fragment nuclear forces cease to act. Considering this crucial role of the rattling in FBMs, what is customarily termed as “freezeout volume” is, in fact, a “rattling volume”.

It is worth noting that rattling of fragments has an obvious effect on their effective matter distribution that mocks up surface diffuseness, when averaged over time. In this respect then, what CNSEM achieves by explicitly acknowledging the undisputable facts that (i) nuclei (and fragments) are endowed with diffuse surface domain and that they are (ii) bound by nuclear forces and (iii) are guaranteed the freedom to exercise thermal surface oscillation, when excited, freezeout-based models achieve by confining sharp-surface, noninteracting (other than Coulomb), ground-state-density clones of fragments to thought volumes adjusted in size so as to allow for (thermal) rattling. The size of the volume is chosen so as to produce agreement with experimental observations, but otherwise, this size has not been expressed in terms of any meaningful equations - a basic requirement for a quantity to be considered as a physical one.

It can be argued, that technically, CNSEM will reduce to what the computer codes of the freezeout-based models do - calculate the number of all possible breakup config-
urations along with their associated entropies. CNSEM may arrive at these numbers by asking how many different, but relevant multi-fragment breakup configurations can there be for contiguous nuclear matter. For a counting purpose then, CNSEM may approximate multifragment configurations by sets of touching spheres and chose a reasonably large volume to enclose all relevant configurations. With surface diffuseness of fragments substituting for the rattling space, surface entropy substituting for rattling entropy, and the “counting” volume substituting for the freezeout volume, the counting becomes technically identical to that performed by computer codes of the FBMs. Note, that unlike in the case of freezeout-based models, an oversized volume would not necessarily increase the number of breakup configurations in CNSEM. This is because of fundamentally different roles of the confining volume in the two classes of models. In the case of FBMs, the boundary of the freezeout volume is ideally reflecting, directing all the outgoing fragments back into the freezeout “furnace”, so as to populate all rattling configurations. This (unphysical) boundary is active in a sense that it alters the distribution of fragment momenta and, in fact, allows to populate states that would not be populated in its absence. Furthermore, this active boundary has no equivalent in nature. In contrast, the “counting” volume used (possibly) by CNSEM would be a purely passive and purely mathematical device, set up to filter out the most relevant configurations (in terms of costs in entropy), not affecting the behavior of nuclear matter in any meaningful way.

It is worth noting, that CNSEM would require a smaller “counting” volume at lower excitation energies, where forming of extended deformed shapes is costly in terms of entropy (or free energy) and can, hence, be a priori excluded. This is reminiscent of SMM [1] using smaller freezeout volumes at lower excitation energies and larger volumes at higher. In the context of this remark, it is then not surprising that, generally, SMM appears to provide for better fits to experimental data than does MMMC [2].

One important point to be kept in mind is that being nominally microcanonical ([2]) or pseudo-microcanonical ([1]), the FBMs should include the breakup configurations envisioned by CNSEM and, also, they should account for thermal expansion of nuclear matter and the presence of a diffuse surface domain. For example, a back-of-the-envelope calculation (using harmonic interaction Fermi gas model [6]) reveals that an $^{197}$Au
FIG. 9: Equivalence of counting of rattling states of sharp-surface spheres in FBM (left) and counting of breakup saddle configurations with enriched surface entropy in CNSEM (right).

A system excited to 8 MeV/nucleon would increase its entropy by staggering 35 units by just expanding to equilibrium density of $\rho = 0.5\rho_0$. Furthermore, calculations for boxed harmonic-interaction Fermi gas using the formalism outlined in Ref. [18], show that in the above case of the excited $^{197}\text{Au}$ system, the gaseous fraction would consist of only approximately 9 neutrons, when the system is placed in a “freezeout” box 8 times the size of the ground-state $^{197}\text{Au}$ nucleus, the most likely microcanonical behavior that, however, appears beyond the scope of FBM.

In view of the above discussion, and in view of large quantitative uncertainties as to the evolution of the surface domain with the excitation energy, it appears justified to use the existing computer code of SMM[1] to approximate CNSEM predictions. In that case, CNSEM provides for a definite meaning for breakup configurations, breakup phenomenon, the rattling entropy, the freezeout volume and the freezeout density, much different from the meaning suggested or left for speculation by FBM [1, 2].
V. DISCUSSION

It was shown that an extension of classical compound nucleus model, obtained by including explicitly the role of surface entropy in establishing the behavior of excited matter, leads to parameterizations of fragment yields similar to those shown to describe (but not to explain) a large volume of experimental data on statistical fragment production.

Within the accuracy to which the properties of surface domain of hot nuclei are known, it appears justified to use these existing parameterizations “as is”, treating the parameters as expressing, e.g., the evolution of the “softness” of the nuclear surface with increasing excitation energy. By adopting this approach, CNSEM is guaranteed a success in describing experimental observations. Unlike, however, the models for which the above parameterizations were developed, CNSEM offers a clear physical scenario consistent with the said parameterizations.

The scenario used by CNSEM does not, in fact, go beyond what has been generally accepted for the compound nucleus decay, including fission. In this sense, with the exception of the acknowledgment by CNSEM of the crucial role of the surface entropy at elevated excitations, CNSEM is a consensus model. What may be considered as debatable, is the magnitude of the surface entropy and whether the more accurate modelling than that by Fermi-gas model, would support vanishing of surface tension at temperatures near 5 MeV, or so, where statistical multi-fragment breakup is seen to set in. Yet, in view of its simplicity, continuity of the chain of reasoning, clarity, and absence of ad hoc or hidden assumptions, CNSEM is apparently superior to NFDM, and FBMs (SMM and MMC). At the same time, CNSEM explains why the latter, mutually exclusive models do produce quite impressive agreement with experimental observations.

One important lesson from the present study is that the agreement with experimental observations should not be used as a substitute for a scientific evaluation of the foundations of a model. Therefore, CNSEM as an immature model in parts relating to the properties of the surface domain of hot nuclear systems, begs for a scrutiny. Clearly an extensive theoretical study of the evolution of properties of diffuse surface domain with increasing excitation energy is warranted. At the same time, the foundations of both
NFDM and freezeout-based models (SMM and MMMC), should be subjected to a strict scrutiny, some of their significant deficiencies being pointed out in this paper.

The present study is not concerned with a possible dynamical production of IMFs as observed in many experimental studies [19–21]. Obviously, dynamical forces present at the interaction stage of massive nuclei, are quite capable of “tearing” fragments of nuclear matter apart from their original nuclei. Yet, the process of dynamically induced separation of fragments from residues, must be sensitive to the “softness” of nuclear surface, as well. In this respect, studies of dynamical fragment production may well provide necessary constraints for theories of hot nuclear surface domain and, by implication, to the statistical decay.

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Retardation of Particle Evaporation from Excited Nuclear Systems 
Due to Thermal Expansion

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ABSTRACT

Particle evaporation rates from excited nuclear systems at equilibrium matter density are studied within the Harmonic-Interaction Fermi Gas Model (HIFGM) combined with Weisskopf’s detailed balance approach. It is found that thermal expansion of a hot nucleus, as described quantitatively by HIFGM, leads to a significant retardation of particle emission, extending the validity of Weisskopf’s approach to the domain of excitation energies in excess of 8 MeV/u. The decay of such highly excited nuclei is strongly influenced by surface instabilities.
I. INTRODUCTION

Studies of nuclear heavy-ion reactions at Fermi bombarding energies (a few tens of MeV per nucleon) have faced conceptual and experimental challenges. This is the domain in which the low-energy dissipative reaction mechanism[1] is expected to morph into one that is characterized by a merging of time scales for collective motion and relaxation processes and the population of a new phenomenological realm [2]. For example, for nuclear temperatures of $T \sim 5\text{MeV}$, nucleon evaporation times of the order of $t_{\text{evap}} \approx 20\text{fm}/c$ have been estimated [2, 3]. Such short evaporation times describe a ”prompt” nuclear decay occurring during the nuclear interaction between projectile and target, thus preventing a complete equilibration of the system. Furthermore, mechanical and chemical instabilities [4] expected for highly excited nuclear systems should exceed the range of conventional statistical models and cause novel nuclear decay modes [5, 6].

On the other hand, experimental observations [7–9] of the kinematics of particle emission patterns in reactions at Fermi energies, induced by heavy projectiles, demonstrate the persistence of sequential decay of hot projectile-like fragments and their target-like reaction partners. Apparently, the primary hot fragments emit nucleons, light particles, and complex nuclear clusters with Maxwell-Boltzmann-type invariant velocity distributions that reflect the temperatures and somewhat reduced mean velocities of the emitter fragments emerging from a dissipative collision. Even though the situation is complicated by an additional intermediate-velocity component of probably dynamical origin [10–12], the existence of intense sequential evaporation components demonstrates unexpectedly high (meta-) stability of the primary hot projectile and target-like nuclei.

The present work investigates conditions for a consistency of the apparently long particle emission times with high nuclear excitations. It is shown that, consistent with the above observations, highly excited nuclear systems are indeed similar to a classical compound nucleus, once their expansion to the equilibrium matter density [13] is taken into account. The particle emission timescales are then long enough to allow for a meaningful equilibration of the system in the sense of Weisskopf’s detailed balance,[14]. Relative decay rates for various statistical decay channels are governed by the entropy at a respective “transition” state, in which the state of the surface plays an important role.
The transition state may be identified with an emission barrier for particle emission and a separation saddle for statistical fission-like decay.

In the following section, the Weisskopf evaporation model is briefly reviewed. The effects of nuclear expansion are described in Section III, followed by a presentation of calculations in Sect. IV and brief conclusions (Sect. V).

II. WEISSKOPF'S PARTICLE EVAPORATION TIME-SCALES

The evaporation time scales for (metastable) excited nuclear systems can be conveniently evaluated using Weisskopf’s detailed balance\[14\] approach. This approach relates the probability $W(E^*_A, \epsilon)$ for the emission of a particle with energy $\epsilon$ by a compound nucleus with mass number $A$ and excitation energy $E^*_A$ to the cross section $\sigma(\epsilon)$ for the reverse process of absorption of such a particle by a nucleus of mass number $B$ and corresponding excitation energy $E^*_B$,

$$W(E^*_A, \epsilon) d\epsilon = \frac{g m \epsilon}{\pi^2 \hbar^3} \frac{\omega_B(E^*_B)}{\omega_A(E^*_A)} \sigma(\epsilon) d\epsilon,$$

(1)

where $\omega_A$ and $\omega_B$ are the level densities of nuclei $A$ and $B$ at respective excitation energies, $m$ is the mass of the emitted particle, $\hbar$ is Planck’s constant divided by $2\pi$, and $g$ denotes the spin degeneracy.

In Eq. 1, excitation energies $E^*_A$ and $E^*_B$ of the compound nucleus $A$ and the residue $B$ are related via energy conservation

$$E^*_B = E^*_A - \epsilon - Q_{gg},$$

(2)

where $Q_{gg}$ is the $Q$ – value for the particle separation from the ground state of nucleus $A$, leaving the residue $B$ also in its ground state.

By expressing level densities $\omega$ in terms of entropy $S$

$$\omega_A(E^*_A) = e^{S_A(E^*_A)} \quad and \quad \omega_B(E^*_B) = e^{S_B(E^*_B)}$$

(3)

one rewrites Eq. 1 in a form

$$W(E^*_A, \epsilon) d\epsilon = \frac{g m \epsilon}{\pi^2 \hbar^3} e^{S_B(E^*_A-Q_{gg}-\epsilon)-S_A(E^*_A)} e^{S_B(E^*_B)-S_B(E^*_B)} d\epsilon,$$

(4)
which is well suited for use in conjunction with models such as HIFGM. Ultimately, the decay width $\Gamma$ for a particular particle channel is obtained by integrating Eq. 4 over particle energy $\epsilon$ and then by multiplying the result by $\hbar$, i.e.,

$$\Gamma(E_A^*) = \hbar \int_0^\infty W(E_A^*, \epsilon) d\epsilon = \frac{g_m}{\pi^2 \hbar^2} \int_0^\infty \sigma(\epsilon) \epsilon e^{S_B(E_A^* - Q_{gg} - \epsilon) - S_A(E_A^*)} d\epsilon.$$  (5)

To be able to make use of Eq. 5 one needs additionally a prescription for calculating the cross section as a function of bombarding energy. In the context of the Weisskopf’s approach, one assumes customarily that the absorption cross section is equal to the geometrical cross section of the nucleus. This corresponds to a constant value of $\sigma = \pi r_o^2$ for neutrons and an energy-dependent, Coulomb corrected expression for charged particles,

$$\sigma = \pi r_o^2 (1 - \frac{V_C}{\epsilon}),$$  (6)

where $r_o$ is the nuclear radius parameter and $V_C$ is the magnitude of the Coulomb barrier. Obviously, Eq. 6 is suited for both, charged particles and neutrons, with $V_C = 0$ for the latter.

A useful approximation for the right-hand side of Eq. 5 can be obtained for the neutron decay width. It assumes an identical functional dependence of the entropy on thermal excitation energy for both, mother and daughter nucleus, i.e., $S_A(x) = S_B(x)$. Then, expanding the entropy $S_B$ in a Taylor series about $E_A^*$ to first order results in

$$S_B(E_A^* - Q_{gg} - \epsilon) - S_A(E_A^*) = -\frac{dS_A}{dE_A^*} \*(Q_{gg} + \epsilon) = -\frac{1}{T_A}(Q_{gg} + \epsilon),$$  (7)

where use was made of the fact that the derivative of the entropy with respect to excitation energy, $dS_A/dE_A^*$, is equal to the inverse of the temperature, $T_A$, of nucleus A. Thus, for the neutron decay width one obtains an approximate expression

$$\Gamma_n(E_A^*) = \frac{g_m}{\pi^2 r_o^2 T_A^2 e^{-\frac{Q_{gg}}{T_A}}},$$  (8)

revealing the strong temperature dependence of the decay width. It is this exponential temperature dependence that has raised concerns regarding the applicability of Weisskopf’s equilibrium approach at higher excitation energies. Rationale given in Sect. III will demonstrate a much weaker effective temperature dependence of the decay width.
In figures shown further below, the decay rate is expressed in terms of the decay time scales parameter $\tau$, which is related to the decay width $\Gamma$ via Heisenberg’s uncertainty principle, i.e.,

$$\tau = \frac{\hbar}{\Gamma}. \quad (9)$$

III. EFFECTS OF THERMAL EXPANSION OF NUCLEAR MATTER

It is plausible that nuclear matter responds to thermal excitation by expanding, like other real gases and types of matter. One expects realistic nuclear transport theory to predict such an expansion to be driven towards the state of maximum entropy, for any given total excitation energy. A particularly simple expression for the dependence of the asymptotic, equilibrium matter density on total excitation energy is provided by the Harmonic Interaction Fermi Gas Model (HIFGM). The essentials of this model[13, 15] are described below.

The central notion of the HIFGM is entropy. The relation between this entropy and the thermal excitation energy $E^*_{\text{therm}}$ is assumed to be described by the regular Fermi-gas expression, such that

$$S(E^*_{\text{tot}}) = 2\sqrt{a(E^*_{\text{tot}} - E^*_{\text{compr}})}, \quad (10)$$

where $E^*_{\text{tot}}$ is the total excitation energy and $E^*_{\text{compr}}$ is its collective compressional (potential energy) part. The difference between these latter two energies defines the random, thermal excitation $E^*_{\text{therm}}$ and, hence, the entropy. Furthermore, the HIFGM adopts the matter density dependence of the level density parameter $a$ (“little-a”) germane to the Fermi-gas model,

$$a = a_o \left(\frac{\rho}{\rho_o}\right)^{-\frac{2}{3}}, \quad (11)$$

where $a_o$ is the ground-state value of the level-density parameter and $\rho_o$ is the ground-state matter density. For simplicity, the HIFGM assumes a quadratic dependence of the compressional energy, $E^*_{\text{compr}}$, on the relative matter density,[16]

$$E^*_{\text{compr}} = E_{\text{bind}}(1 - \frac{\rho}{\rho_o})^2, \quad (12)$$

where $E_{\text{bind}}$ is the ground-state binding energy of the system.
Given Eqs. 10-11, the equation for maximum entropy as a function of relative density can be resolved analytically, yielding [13]

\[
\frac{\rho_{eq}}{\rho_o} = \frac{1}{4} \left( 1 + \sqrt{9 - \frac{E_{\text{tot}}}{E_{\text{bind}}}} \right).
\]

(13)

Assessing qualitatively the effects of thermal expansion on the decay width (see Eq. 8), one notes that, according to Eq. 13, the matter density decreases with increasing total excitation energy. Consequently, the compressional part of the excitation energy and the level density parameter, both increase. The latter two trends reduce the rate of increase of the system temperature with increasing excitation energy, as compared to that without expansion. This follows from the Fermi-gas model relationship between temperature \((T)\), level-density parameter \((a)\), and thermal part \((E_{\text{therm}})\) of the excitation energy,

\[
E_{\text{therm}} = E_{\text{tot}}^* - E_{\text{compr}}^* = aT^2.
\]

(14)

This effect can be viewed as a manifestation of the LeChâtelier Principle. The reduction in effective temperature from the value that it would have at normal matter density leads to a reduction of the decay width, as approximated by Eq. 8. This reduction is partially offset by the effects of \(a)\) an increased value of the inverse capture cross section \((\sigma)\) and \(b)\) a neutron binding energy \((Q_{gg})\) that is reduced by an amount equal to the compressional energy per nucleon. Results of calculations performed using the “exact” Eq. 5 are discussed in the next section.

IV. RESULTS OF CALCULATIONS

Results of model calculations for the Weisskopf particle evaporation time scales are shown in Figs. 1 and 2. In Fig. 1, the evolution of the average nucleon evaporation time \(\tau\) (see Eq. 9) with total excitation energy per nucleon, predicted for expanded Fermi matter at equilibrium density (solid line), is compared to that resulting for Fermi matter forced to stay at ground-state density (dashed line). As seen in this figure, if allowed to expand to equilibrium density, a nucleus excited to \(E_{\text{tot}}^*/A \geq 8 MeV\) regains a degree of (meta)stability against nucleon evaporation comparable to that of a ground-state density nucleus at the much lower excitation of \(E_{\text{tot}}^*/A \sim 4.5 MeV\). The small
FIG. 1: Excitation-energy dependence of Weisskopf’s nucleon evaporation time scales for nuclei at equilibrium (solid line) and ground-state (dashed line) matter density. The dotted line illustrates results of calculations with $T = 0$ absorption cross section.

Increase in $\tau$ predicted to occur near the high-excitation end of the curve is related to the negative heat capacity predicted by the HIFGM in that energy domain. Both these effects are due the fact that the considered maximum in entropy is merely local and not absolute. Correspondingly, the considered state of equilibrium density is merely metastable against evaporation and not stable.

In Fig. 2, partial time scales are illustrated separately for neutron emission and for
FIG. 2: Excitation-energy dependence of Weisskopf’s neutron (bottom curves) and proton (top curves) partial evaporation time scales for nuclei at equilibrium (solid line) and ground-state (dashed line) matter density.

proton emission. In both cases, thermal expansion is seen to lead to a retardation of emission, extending the range of applicability of Weisskopf’s approach.
V. CONCLUSIONS

The present calculations have shown that thermal expansion of a nucleus to equilibrium density has a stabilizing effect on the excited system, such that the most likely statistical decay channels are suppressed. This may give the system time to reach a more complete statistical equilibrium, where various surface modes are excited as well. Such surface modes may be viewed as doorway states associated with cluster emission. It has been demonstrated elsewhere[13] that, when equilibrated at high excitation energies, such fission-like decay modes may compete successfully with nucleon emission. It should also be kept in mind that, with every neutron that escapes before these cluster emission modes are equilibrated, the probability for the emission of subsequent neutrons decreases, due to isospin effects, until it equals that for proton emission. This isospin effect adds additional credibility to the idea that Weisskopf’s approach may be applicable at much higher nuclear excitation energies than commonly thought.

It also worth noting that, in its present simple form, the HIFGM assumes that thermal expansion occurs in a self-similar fashion, i.e., that this expansion can be reduced to a rescaling of the radial coordinate. In addition, it is assumed that the effective nucleonic mass is equal to the free nucleon mass. A more complete modelling [17] also calls for a more thorough and detailed treatment of the diffuse nuclear surface and its evolution with increasing excitation. In such a treatment, it would be highly desirable to account for the finite range of the effective nuclear interaction. This finite range may alter the thermostatic properties of surface matter with respect to those of bulk matter at equal density and temperature. It is to be expected that, in line with LeChâtelier's Principle, the nuclear surface region will evolve in a manner that increases entropy and stabilizes the overall system even further.
Acknowledgments

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Numerical Simulation of Linear Chain Collisions with Dissipative Interactions

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Abstract

The combined effects of nucleon-nucleon strong, Coulomb, and dissipative forces on the prompt breakup of nuclei in energetic collisions have been simulated and studied in a simplified one-dimensional “row-on-row” collision model. In this model, projectile and target nuclei are each represented by a linear chain (row) of randomly distributed nucleons. The mass and charge distributions of reaction products have been studied as functions of bombarding energy and the strength of the dissipative interaction. The collision phenomenology includes processes ranging from elastic scattering to fusion, fusion/fission, and prompt cluster emission. The results show a strong dependence of the products mass distributions on the strength of the dissipative interaction.

I. Introduction

Study has been conducted on the equilibrium and non-equilibrium properties of nuclear matter and nuclear interactions [Bro60, Sch84, LiS04]. Both simulation and experiment show that dissipation of kinetic energy plays an important role in reaction dynamics. Kleinig et al. [Kle02] have developed a simple one-dimensional dynamics model that includes two-body dissipation generated by nucleon-nucleon collisions. Similar work by Dunkel et al. [Dun03] have modeled the same one-dimensional system, but with “intermediate energy storage”; the dissipated kinetic energy was only temporarily lost to excited states of the reaction partners before returning to relative motion. Both studies demonstrated an effect of the dissipative force strength on the fragmentation of target nuclei when hit by energetic projectiles. The present study was undertaken specifically to investigate the sensitivity of the distributions of clusters of nucleons produced in such collisions on details of the dissipation mechanism. This study will eventually allow one to
draw conclusions about the importance of a realistic modeling of the internal cluster structure in theoretical simulations of prompt nuclear breakup and other aspects of nuclear reaction dynamics.

It is well known that kinetic energy of relative projectile/target motion can be dissipated and transformed into excitation energy of the interacting projectile and target nuclei, as well as of other reaction products. With increasing interaction time, initial asymmetries in thermal energies of the interaction partners tend to equilibrate through nucleon exchange and particle-hole excitation processes, equivalent to macroscopic heat convection and conduction. The efficiency of such energy relaxation mechanisms depends on the thermal properties of the nuclei involved, which are a reflection of intrinsic nuclear structure. For example, the more complex the internal structure of a nucleus, the greater its heat capacity. The greater is its contribution to the effective viscosity (“stickiness”) in a nuclear collision. This realization establishes a connection between internal nuclear structure and the dissipative collision dynamics. Demonstration of a dependence of certain reaction features on energy dissipation immediately demonstrates a necessity to model realistically enough the internal structure in simulation of the reaction dynamics.

In this work, the dependence of prompt-fragment/cluster distributions on dissipation is studied. The particular task of the present study is to investigate characteristic differences in the cluster distributions obtained in a simple collision model under various assumptions about the balance between conservative and dissipative forces.

Besides their academic interest, the present calculations and their extensions to two- or three-dimensional models have potential practical applications for the design of more complex codes modeling the behavior of a new type of nuclear reactor [Nif03] based on spallation reactions. These reactors use accelerators to produce protons, which are then used to induce spallation reactions in any heavy material. The subsequent fragmentation of these heavy materials, which is more complex than the fission process, powers the reactor by releasing large amounts of energy. The amount of energy released is directly related to the size distributions of the fragments produced. This can be inferred from a diagram of the nuclear binding energy per nucleon as a function of mass number (see Fig.1).
On the right-hand-side of the dashed vertical line (Fe, Ni) in Figure 1 fragmentation of a nucleus into two (fission) or several smaller (clusters) fragments is exothermic.

![Figure 1. Binding energy per nucleon vs. Mass number](image)

For example, one calculates an energy release of 90 MeV for binary fission of an A=180 nucleus. If the same nucleus were to split into 3 equal fragments of A_f=60 each, an energy of 126 MeV is released. This illustrates how the fragment size distribution produced in nuclear reactions directly determines the energy released.

The present work adapts ideas presented by Kleinig et al. [Kle02] in a similar one-dimensional collision model. The same force form factors were used here as in their work, and an overall similar procedure was followed. However, in the present work calculations were performed for statistical ensembles of initial nucleonic conformations of the projectile-target system, and a more detailed analysis of the collision outcomes was performed. Kleinig et al. [Kle02] have simulated systems comprising evenly distributed protons and neutrons in each linear chain of particles and examined the probabilities for various exit channels. In contrast, the present work examines a more general class of reacting nuclei. It analyses the effects of different conformations and friction strength on the breakup or fusion of reaction partners. The complications involved with this addition, as well as those involved in the simulation of larger systems, are discussed further below.
II. The Collision Model

The coded model allows the user to simulate the dependence of fragmentation behavior on bombarding energy and the strength of the dissipative force. The three possible types of fragmentation are schematically depicted in scheme 1 below.

The particles interact through three forces: a short-range strong nucleon-nucleon force, a long-range Coulomb force, and a dissipative force. The nucleon-nucleon force is of short range and has negligible strength at inter-particle distances much larger than the diameter of a nucleon. In contrast, the Coulomb force is of long range, its strength cannot be neglected even at large inter-particle distances. The dissipative force is assumed to act within the same short range as the strong nucleon-nucleon force, because it is thought to arise from nucleus-nucleus surface interactions. The width of the nuclear surface is of the order of 1 fm, i.e., similar to the nucleonic diameter.

The three forces used in the calculations are of the form

\[ F_{\text{strong}}(r) = \frac{a}{r^{12}} - b \cdot e^{-c \cdot r} \]  \hspace{1cm} (1.1)

\[ F_{\text{Coulomb}}(r) = \frac{d}{r^2} \]  \hspace{1cm} (1.2)

\[ F_{\text{Visc}}(\vec{v}_j - \vec{v}_i) = k \cdot (\vec{v}_j - \vec{v}_i) \]  \hspace{1cm} (1.3)
The dimensionless parameters $a$, $b$ and $c$ are defined such that $r = r_0 = 1$ is the equilibrium distance of the nucleon-nucleon strong force and can be taken as the natural unit to scale all linear dimensions. The parameter values used in the calculations are given in Table 1 below.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$a$</td>
<td>5.0</td>
</tr>
<tr>
<td>$b$</td>
<td>100.0</td>
</tr>
<tr>
<td>$c$</td>
<td>$\ln(a/b) = \ln(20)$</td>
</tr>
</tbody>
</table>

Table 1. Nucleon-nucleon strong force parameters

The parameters for the Coulomb and viscosity force, $d$ and $k$ respectively, define the strength of these forces. The program user can vary the values of these two. In nature, the strength of the Coulomb force depends on the charge of the pair of interacting particles, but was allowed to vary in the calculations performed by Kleinig et al. [Kle02] to investigate the interplay between the repulsive Coulomb force and the largely attractive nuclear force. In the calculations performed in this work the strength of the Coulomb force was allowed to vary only initially, when attempting to reproduce the data reported by Kleinig et al. In all other cases, it was held constant. The parameter $k$ defining the strength of the dissipative interaction is varied in calculations below to test the effect of this force on fragmentation.

These parameters define the forces in terms of unit distances $r_0$ and allow one to introduce reduced units to simplify expressions to be coded. The reduced units and their conversion to normal nuclear science equivalents are given in Table 2 below.

<table>
<thead>
<tr>
<th>Reduced Variable</th>
<th>Reduced Value</th>
<th>Nuclear Equivalent</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$r_0$</td>
<td>1</td>
<td>1.13 fm</td>
<td>Distance</td>
</tr>
<tr>
<td>$m$</td>
<td>1</td>
<td>931.5 MeV/c$^2$</td>
<td>Nucleon Mass</td>
</tr>
<tr>
<td>$\tau$</td>
<td>1</td>
<td>28.9 fm/c</td>
<td>Time</td>
</tr>
<tr>
<td>$F$</td>
<td>$\frac{(mr_0^2)}{\tau^2}$</td>
<td>1.26 MeV/fm</td>
<td>Force</td>
</tr>
<tr>
<td>$\varepsilon$</td>
<td>$\frac{(mr_0^2)}{\tau^2}$</td>
<td>1.42 MeV</td>
<td>Energy</td>
</tr>
<tr>
<td>$P$</td>
<td>$\frac{(mr_0^2)}{\tau^2c}$</td>
<td>1.42 MeV/c</td>
<td>Momentum</td>
</tr>
</tbody>
</table>

Table 2. Reduced units and conversions
The calculation of the reduced unit for time is included in Appendix 1. The units for distance and mass were taken from the article by Kleinig et al. The other three result from combinations of the former.

The short-range nucleon-nucleon strong force is plotted in Figure 2 below.

![Figure 2. Short-range nucleon-nucleon strong force](image)

The total conservative force that acts between two protons is given below.

\[ F_d (r) = \frac{a}{r^{12}} - b \cdot e^{-cr} + \frac{1}{r^2} \]  \hspace{1cm} (1.4)

The addition of the Coulomb repulsion term makes the conservative force less attractive, as seen by comparison in Figure 3 below.

![Figure 3. Short-range nucleon-nucleon strong force (solid), addition of Coulomb force to short-range nucleon-nucleon strong force (dotted)](image)

At the start of this investigation, the parameterization of these forces was questioned. Kleinig et al. [Kle02] offer no explanation for their choice of parameters or functions. The true Hamiltonian of nucleons inside nuclei is not well known. For example,

---

1 Equation (1.1)
2 Equation (1.4)
correlation terms similar to those in the Hamiltonian describing the electronic structure of atoms are difficult to estimate. Thus, a perfect fit or parameterization is unreasonable to expect given this complex multi-body problem. One can hope, however, that important qualitative aspects of the underlying physics are captured in the simple approach followed here.

The total conservative potential felt by nearest neighbor protons as a function of their relative separation (r) is given below and illustrated in Figure 4.

\[
U(r) = \frac{a}{11 \cdot r^{11}} - \frac{b}{c} e^{-c \cdot r} + \frac{1}{r}
\] 

(1.5)

![Figure 4: Short-range nucleon-nucleon strong and Coulomb potential for nearest neighbor protons](3)

One further question arising from these nucleon-nucleon force parameterizations is the relative binding energy of protons vs. that of neutrons. Figure 5 below compares the potential energy of two protons separated by a distance r and two neutrons not interacting via the Coulomb potential.

---

3 Equation (1.5)
According to the model, there is a difference in the classical binding energy of protons with respect to neutrons in the bulk material. Protons interacting with neutrons are not affected by the Coulomb force and are therefore bound just as strongly as neutrons interacting with neutrons. The proton-proton effective potential matches the Coulomb potential at large inter-particle distances, \( r \), as it must. One can see the hump that develops after the bound region as the effective potential rises to meet the Coulomb potential. This is indicative of the Coulomb barrier that exists in real nuclei. This barrier results from the balance of nuclear and Coulomb potentials. A positively charged particle must have a certain amount of kinetic energy to overcome the Coulomb repulsion of another particle (cluster or nucleus) before it can be bound to it. Therefore, the parameterization that was at first questioned can be conditionally accepted because of its qualitative adherence to the underlying physics of the present model.

Since the Coulomb force has infinite range, the higher-order terms contributed by particles that are not nearest neighbors are important. Figure 4 only represents the first order approximation to the potential felt by particles in the simulation. More repulsive terms exist representing the effects of other protons in the linear chain, but these higher order terms have less effect because the inter-particle distances are greater than \( \sim 2r_0 \).
III. Model Implementation/Computer Code

This section provides a discussion of the methods and algorithms used to integrate the equations of motion. It also explains how the code was tested for validity and accuracy.

First, three different integration algorithms were evaluated for their utility in solving the numerical equations of motion for the model introduced previously. The three algorithms considered were 2nd order Verlet, 5th order Gear Predictor-Corrector, and 5th order Runge-Kutta methods. These three techniques were all adequate for the task. The 5th order integrator methods are more accurate techniques but use more computer resources. Therefore, the 5th order integrator methods were not used. The 2nd order Verlet algorithm is illustrated as a flow chart in Figure 6 below.

![Figure 6. Second order Verlet algorithm](image)

This algorithm was used with a time step of $\Delta t = 5 \times 10^{-4}$ in natural units of $\tau$ and run until time $= 100 \tau$. The code was checked for numerical accuracy, specifically with regard to energy and momentum balance. For simulations without dissipation of energy
the total energy is defined as the sum of kinetic and potential energies. If the dissipation interaction is included, then the sum of the kinetic energy dissipated every half time step must be calculated and taken into account, together with kinetic and potential energies. This sum gives the total energy at the end of the simulation. Without energy dissipation the error in total energy after an integration time of 100 $\tau$ was significantly less than 1%. The error in this sum was also less than 1% when the dissipative interaction was turned on.

The integration algorithm was also tested with respect to its conservation of momentum in the absence of dissipative interactions. If the algorithm conserves momentum then, after a particle moving with some velocity $v_1$ strikes a second particle of equal mass at rest, the first particle should come to rest. After the collision, the second particle should move in the original direction of the first particle and with the same velocity. In other words, momentum transfer from the “projectile” to a “target” nucleon should be numerically complete (~100%). Such a calculation was run and the algorithm fulfilled these expectations within less than 1% deviation.

Fortran 90 was used to numerically integrate the equations of motion of the particles in the linear chain. At the beginning of the simulation there are two linear chains of nucleons, one for the projectile and one for the target. These two chains are separated by some length greater than the range of the possible interaction forces. For the calculations in this report the distance was 5 $r_0$, unless otherwise noted. The projectile is given an initial user-defined amount of kinetic energy (EBEAM in natural units $\epsilon$) relative to the target; the later is considered at rest in the laboratory frame. All the particles within an initial chain are at rest with respect to one another, at the corresponding locations of potential energy minima defined by the nucleon-nucleon interaction. Thus, there is no initial potential energy. The model’s adherence to this ideal is discussed below.

The simulation of larger systems is difficult, if one wishes to sample a large fraction of all possible initial nuclear ground-state configurations. The combinatorial mathematics for a system of $^{12}$C colliding with $^{40}$Ca show that sampling all configurations is currently unfeasible, since there are $1 \times 10^{11}$ combinations to calculate. Thus, preliminary ideas for randomly sampling the ensemble of ground state configurations were explored.
The ensemble of ground state configurations for any given projectile or target is the set of all configurations of nucleons, corresponding to the projectile or target species. For example a $^{12}$C configuration must contain six protons and six neutrons. However, these nucleons can be arranged in any possible order. Each ground state configuration has a given amount of potential energy. This potential energy can be calculated by using equation (1.5) and summing over all pairs of particles in the chain, all while recognizing that the Coulomb force only acts between charged species. The calculated energy distributions of the ground state configurations for $^{12}$C are given in Figure 7 below.

![Figure 7](image.png)

**Figure 7. Ground state energy distribution of $^{12}$C configurations**

Figure 7 shows the initial energy distribution of all 924 possible configurations and a subset of 231 randomly chosen configurations of $^{12}$C. The figure shows that the averages of the distributions are fairly similar, such that the smaller set samples the ground state rather well.

As can be seen in Figure 7, the initial configurations used in the above simulations were not all in their exact classical ground state. Each contained a small degree of excitation due to the inherent Coulomb repulsion, which was not taken into account in the initial configurations assumed for projectile and target chains. The nucleons are all bound and none will spontaneously eject due to this excitation, but they will show harmonic oscillations within their local potential energy wells, similar to zero-point fluctuations. One could, of course, cool the initial configuration numerically by bleeding the kinetic energy, as the initial chain relaxes down to the minimum of the potential energy surface. There is no general analytical method to find this minimum. When the long-range Coulomb interaction is included, the potential energy minimum is a complicated function of the positions of all particles in the chain.
There is one other approximation worth keeping in mind: Only nearest neighbor short-range interactions are considered in these calculations. In addition, the force is assumed to be negligible at nearest neighbor distances greater than $2r_0$. Furthermore, the dissipative force acts only between those particles that would also interact through the short-range strong nucleon-nucleon force. Again, this means that only nearest neighbors are exposed to the dissipative force. The distance between these particles must be less than $2r_0$ for this force to influence their motion. In addition, the dissipative interaction is calculated with regard to the integrator but during the calculation of the forces. So for example, the 2$^{\text{nd}}$ order Verlet integrator described above calculates forces twice per time-step. Thus the dissipative interaction affects the appropriate particles twice per time-step, but each time it has half the strength.

In summary, the code was tested using energy and momentum balance. The algorithms chosen implement the model with satisfactory accuracy. Any numerical artifacts should have negligible effects on the results of the calculations because these errors are so small.

**IV. Results**

The output of the code described above was compared to the numerical results generated by Kleinig et al. These authors have modeled the fragmentation behavior of a very specific initial configuration of nucleons. They distributed protons between neutrons evenly throughout linear chains of particles and performed calculations for a range of kinetic energies, dissipation strength and Coulomb force strength. The results were displayed as contour diagrams of friction strengths $k$ vs. the energy per particle of the projectile chain such as that shown in Figure 7. Regions in $(k, E/\text{particle})$ space corresponding to different fragmentation channels (chain splits) are distinguished by different shading patterns.

In the calculations illustrated in Figure 8, the strength of the Coulomb force is assumed to be $d=1$. The gray region in the contour diagram corresponds to cases in which the projectile forces the last particle in the target chain to eject from the target, as the projectile is absorbed by the target chain on its opposite end. The white, uncolored region
corresponds to complete fusion. Here the dissipation is so strong that the projectile is absorbed by the target without fragmentation. A similar calculation was performed with the present code to compare results of actual calculations. The new calculations are illustrated in Figure 9 below.

A key for the color code scheme is given in Appendix 2. The results of the present calculations are qualitatively similar to those reported by Kleinig et al. but do not match the
latter quantitatively. The increased tendency for fusion or capture at higher energy observed in the present calculations has not been identified. Similar discrepancies are found when comparing other results with those reported by Kleinig et al. Figure 10 below shows results by Kleinig et al. for a three-particle projectile colliding with a ten-particle target.

![Figure 10. Results by Kleinig et al., a 3-particle projectile colliding with a 10-particle target](image)

The Coulomb force strength in this calculation is $d=0$. The regions corresponding to particular fragmentation patterns are outlined in the inset key. The white region again corresponds to capture or fusion of the projectile with the target. The corresponding results given by the present code are given in Figure 11 below.

If careful attention is paid to the scale when comparing the results presented in Figures 10 and 11, then one can see that discrepancies remain. The results do illustrate similar systematic behavior but the exact results are quantitatively somewhat different. Certain regions predicted to exist by Kleinig et al. are overlapped by others predicted by the present model. They are overlapped because these regions would appear at different total execution times for the code. In other words, the predicted fragmentations occur at certain times during the simulation but further evolution of the integration leads to a different result e.g. partial fusion or further fragmentation. However, no systematic way was
found to make the regions predicted by the different calculations match.

Figure 11. Results from code given in Appendix 2, 3-particle projectile colliding with 10-particle target; no Coulomb force

A favorable change in one region is usually accompanied by an unfavorable result in another.

The difference in results obtained by these two numerical implementations is here simply noted but cannot be resolved. It has not been possible to obtain further detailed information on the calculations by Kleinig et al., which could distinguish the two model calculations. Thus the validation of this code is left to its adherence to energy and momentum conservation as described earlier and the correct prediction of simple scattering scenarios.

The present code was used to numerically simulate the fragmentation behavior induced by collisions between many different configurations of the same projectile and target. Specifically, the relative positions of protons and neutrons within the linear projectile and target chains were varied. The fragmentation configuration produced after a time of 100 $\tau$ was recorded as the final result.

Using combinatorial mathematics, one can calculate the total number of possible configurations for a given projectile and target pair. For the simulation of a 1 particle projectile colliding with a 12 particle target, say a linear $^{12}$C nucleus, there are:

$$^{12}C_6 = 924$$ (3.1)
possible combinations, all due to permutations of positions of target nucleons in the chain.

The set of results for all permutations defines a statistical ensemble. Hence, there is a distribution of final fragmentations. The figures below show such un-normalized distributions vs. the fragment size. The fragmentation distribution produced by summing over all possible configurations of a linear $^{12}$C target being struck by either a proton or a neutron of energy EBEAM=3 is given in Figure 12. Included in this figure are the distributions produced with four different dissipative interaction strengths: $k=0$, $0.5$, $1.0$, and $1.5$ in natural units.

![Figure 12](image)

Figure 12. Fragment mass distribution of neutrons and protons colliding with $^{12}$C; EBEAM=3; $k$ varies from 0-1.5

From Figure 12, one can see that a collision with energy EBEAM=3 without dissipation leads almost always to elastic scattering. This is expected for a conservative system at low bombarding energy. However, as the strength of the dissipative interaction is increased, more complex fragmentation is observed. This behavior is expected from the
“wake” produced by the projectile momentum traversing the target chain. Medium-sized fragments are formed. When the dissipative force is increased even more, the system fuses into one chain (compound nucleus) and no fragmentation occurs.

The fragmentation patterns also depend on the bombarding energy. The present code was also used to calculate the results of protons and neutrons colliding with a linear $^{12}$C target at a higher energy of EBEAM=7 in natural units $\varepsilon$. The results are given in Figure 13 below.

![Figure 13](image.png)

**Figure 13.** Fragment mass distribution predicted by collisions of neutrons and protons with $^{12}$C; EBEAM=7; $k$ varies from 0-1.5

In these simulations, fusion does not occur for $K=1.5$. When the beam energy is equal to EBEAM=7 $\varepsilon$, fusion does not dominate until $K=2.5$ (Figure 14). Even then, it does not dominate to the same extent seen when EBEAM=3 and $K=1.5$. The scaling of fragmentation patterns is nonlinear in these forces.
The fragment mass distributions shown in Figure 13 are different in all cases from those at lower energies. Thus fragmentation depends on both the dissipative interaction strength and the beam energy.

The picture becomes more complex when even larger systems are considered. As mentioned above, the larger size of these systems requires the user to randomly select configurations to sample the ensemble under study. In the following calculations of collisions between alpha particles and \(^{12}\text{C}\), 924 random configurations were sampled and the results of their collisions analyzed. The fragment mass distributions resulting from these calculations are given in Figures 15-17 below.

Figure 15 illustrates the dependence of the fragment mass distribution of colliding \(^{4}\text{He}\) and \(^{12}\text{C}\) particle chains. The behavior is similar to that shown in Figures 12-14 above, but is more complex, especially for the non-dissipative case. The dependence of the fragment mass distribution on the bombarding energy for the \(^{4}\text{He}\) and \(^{12}\text{C}\) particle chain system is shown in Figure 16 below.

The amount of fusion produced with constant dissipation strength is inversely proportional to the bombarding energy. This behavior is similar to that illustrated in Figures 12-14. A comparison between calculations sampling only a portion of the system ensemble and calculations performed over the entire ensemble is given in Figure 17 below.

Figure 17 shows that the random sampling of the ensemble has little affect on the fragment mass distributions produced by the calculations.
Figure 15. Fragment mass distribution produced by collision of alpha particles with $^{12}$C; EBEAM=7; K varies from 0 to 20
Figure 16. Fragment mass distribution produced by collision of alpha particles with $^{12}\text{C}$; EBEAM varies between 3 and 7; $K=12.5$

Figure 17. Fragment mass distribution produced by collision of alpha particles with $^{12}\text{C}$; $K=0.1$; 924-configurations represent 1/6th of the ensemble and the fragmentation patterns produced are compared to the same calculation over the entire ensemble.

With this more complex system one can also investigate the sensitivity of the neutron and proton number of the fragments formed after the simulated reaction to the dissipative force strength. Figure 18 below shows an isometric plot of the fragmentation pattern calculated in the $\alpha$, $^{12}\text{C}$ simulation.

Figure 18 illustrates the complex nature of the fragmentation described in the previous figures. The marked columns are the same as the starting projectile and target pair. After the collision fragments may form or remain such that they are identical to the original projectile or target according to their charge and mass numbers. However, there are a great many other species present and the number of each individual species produced by the collision is sensitive to the dissipation force strength.
Figure 18. Isometric plot of fragment multiplicity as a function of neutron and proton number; α colliding with $^{12}$C; EBEAM=7; K ranges from 0-12.5; Marked columns signify the projectile and target.

There is also a curvature to the distributions illustrated in Figure 18 that can be better seen in a contour plot. Figure 19 below shows the results from the same calculations as in Figure 18, but in the form of a contour plot. The bend towards more neutron rich fragments can be understood as a result of the repulsive Coulomb force. Fragments with a lower neutron to proton ratios have higher conservative excitation energy as discussed in the model implementation section.

To investigate the isospin dependent behavior of the model further, two specific configurations were simulated and their results compared. To model the dependence resulting from different neutron to proton ratios, two calculations involving the same number of particles, bombarding energy, and dissipative interaction strength were chosen. One set of calculations involved the collision of $^{12}$C with neutron rich $^{40}$S, while the other
simulated collisions of $^{12}\text{C}$ with relatively neutron poor $^{40}\text{Ca}$. The results of these calculations are presented in Appendix 3 below.

Figure 19. Contour map of fragment multiplicity as a function of proton and neutron number; $\alpha$ colliding with $^{12}\text{C}$; EBEAM=7; $K$ varies from 0-12.5; Line indicates N/P ratio of system

The tables in Appendix 3 note the mean and variance of specific isotope distributions. This distribution is a mass distribution of all fragments having a particular number of protons, $Z$. Two examples of these distributions are plotted in Figure 20 below.

For plotting the distributions in Figure 20, the data was shifted with respect to the evenly split $^{40}\text{Ca}$ N/Z to illustrate increase in the number of neutron rich fragments produced in the $^{40}\text{S}$ reaction. The center of the $^{40}\text{Ca}$ distribution plotted in Figure 20 lies along $y=0$, while the center of the $^{40}\text{S}$ distribution is noticeably to the right of $y=0$ making it more neutron rich.

In sum the data from Appendix 3 shows that for a fixed $Z$ the $^{40}\text{S}$ reaction produces fragments that are on average 2-3 neutrons richer than the fragments produced by the relatively neutron poor $^{40}\text{Ca}$ target.
Figure 20. Z=8 isotope distributions for $^{40}$S and $^{40}$Ca simulations; $^{40}$S on the left, $^{40}$Ca on the right; distributions shifted on $^{40}$Ca A/Z

This indicates that fragments formed in collisions simulated by this model have a memory of the initial reaction partners. This is consistent with pre-equilibrium emission and reaction dynamics.

Thus there are two factors that affect the neutron to proton ratio of the fragments in the model. The first is the difference in binding energy of neutrons vs. protons arising from the different inter-particle potentials that govern their motion. The second is the initial configurations as the fragments formed after a collision have a memory of their initial target projectile pair. The first factor is much less important than the second in determining the isospin behavior of the model.

V. Discussion

The dependence of fragmentation emission patterns, induced by collisions between linear chains of nucleons, on the dissipative interaction strength and beam energy has been demonstrated by the calculations discussed in the previous section. The significance of the results derived consists in the realization that the fragmentation patterns depend on the strength of the dissipation of kinetic energy. Modeling the dissipation of energy is tantamount to modeling the transfer of kinetic energy to the internal excitation energy of the constituent particles. At the low energies of nuclear reactions considered here, internal excitation of individual nucleons is not possible. This means that the inter-
nal excitation of nuclear fragments (clusters) is the mode to which the kinetic energy of relative motion is transferred during the pre-equilibrium phase of nuclear reactions.

Hence, this report has shown that in a one-dimensional model the possibility of exciting clusters has a noticeable effect on the reaction dynamics. This excitation produces richer fragmentation patterns than those predicted by conservative dynamics alone in step with experimental results already published in the literature.

VI. Conclusions

Highly schematic models such as the linear collision model used in this work are useful primarily for exposing and amplifying important generic aspects of a process. Hence, the present study may serve to emphasize the importance of dissipation and viscosity in the modeling of prompt nuclear breakup reactions. More realistic (and time consuming) calculations in three dimensions are now justified and necessary to explore corresponding fragmentation effects in nuclear collisions. The present calculations have already pointed to the importance of accounting properly for the internal structure, i.e., the density of states, of all participants in a collision to model realistically nuclear reaction dynamics. In particular, the excitation of clusters has been found to influence the prompt disintegration and fragmentation pattern.

This work has been supported by the US DOE Grant No. DE-FG02-88ER40414

References:


[Sch84] W. U. Schröder and J. R. Huizenga, Damped Nuclear Collisions, in Treatise on Heavy-Ion Science, D.A. Bromley (Edt.), Plenum Press, 1984, p. 113

Appendices:

Appendix 1:

The nuclear-strong force has an approximate strength of 50 MeV around the equilibrium point \([\text{MeV}]\). Thus we will use this to calibrate our time unit. The force given by (1.1) has a potential given by:

\[
U(r) = -\int \frac{a}{r^{13}} - b \cdot e^{-c r} \, dr
\]

\[
U(r) = \frac{a}{11 \cdot r^{11}} + \frac{b}{c} \cdot e^{-c r} + D
\]  

(4.1)

We assume that \(D\) is zero and look at the minimum of the potential, which must occur at \(r = 1\). \(U(1) = -1.2145\) in natural units. Thus we take our expression for energy in natural units:

\[
E = \frac{(m\tau^2_0)}{\tau^2}
\]  

(4.2)

and combine with our ratio of reduced to literature value yielding:

\[
\frac{50\text{MeV}}{1.214} = \frac{m\tau^2_0}{\tau^2} = \frac{931.5\text{MeV/\(c^2\)} \cdot (1.13\text{fm})^2}{\tau^2}
\]  

(4.3)

\[\tau = 28.9\text{ fm/c}\]

This conversion then is used to define the other natural units.
Appendix 2:
A key is provided to the color-coding used in Figures 9 and 11 of this report.

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Table 3. Key to color-coding in Figs. 8 and 10
Appendix 3: Isotope Distributions:

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Table 4. $^{40}$S Isotope Distributions K=0

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Table 13. $^{40}$Ca Isotope Distributions K=20
Modeling Atomic Nuclei with a BUU Type Code.

I. Pawelczak, J. Tőke, and W.U. Schröder

Departments of Chemistry and Physics, University of Rochester, Rochester, NY 14627

Abstract

The viability of current transport models is tested by comparing the stabilities of a model $^{197}$Au nucleus in its ground state, as modeled in various realizations of the BUU/BNV transport theory. Three different approaches in calculating nuclear ground state properties are compared: pure mean field, mean field with numerical population constraints, and mean field plus nucleon-nucleon collisions.

I. Introduction

In past two decades, the nuclear Boltzmann-Uehling-Uhlenbeck (BUU) or Boltzmann–Nordheim-Vlasov (BNV) transport theory in several implementations has acquired the status of a standard model for the interpretation of experimental phenomenology in heavy-ion reactions at intermediate bombarding energies. It is therefore important to explore the limits of applicability of the numerical computer codes representing the basic transport theory. As basic entities whose evolution is modeled in the codes, the colliding model nuclei assumed in the codes are of particular interest.
All numerical models considered here employ a Monte Carlo method to sample the mean field and the phase space populated by nucleon-nucleon collisions and to solve the BUU/BNV equations of motion (described in detail in Section II). Specifically, the particle trajectories are calculated using the so-called test-particle method, where each of the A nucleons of a given nucleus is stochastically represented by a number \( N \) of test-particles. This method is similar to adopting an ensemble of \( N \) A-body micro states to calculate statistical averages or means. If \( N \) is a number of test particles per nucleon, the total number of particles propagated in the system is \( A \times N \).

Initialization of the system is done according to the Fermi gas model relation between matter density and momentum, again using a Monte Carlo method. Mean field potential. The nuclear density is calculated from the spatial distribution of the test-particles. Test-particles are assumed to move classically according to Hamilton’s equations for the forces assumed. They can collide and scatter in accordance with an average Pauli blocking scheme. Particular properties of the nuclear matter model are implemented through momentum, density, and isospin dependent forces defined independently in the computer codes.

In order to test the range of applicability of a theoretical model the first step is studying the properties of the model nuclei in their ground states. In this work the stability of nuclear ground states modeled by BUU and various distributions of BNV code is compared. The following approaches in ground state simulations are considered:

- mean field; BNVn model
- mean field and the constraint (explained in Sect.III); BNVc model
- mean field and collision; BUU
In the next section (II), theoretical description of the model is presented, followed by a discussion of properties of the ground states modeled by variations of transport codes (III).

II. Theoretical approach

The various distributions of transport model (BUU/BNV) describe time evolution of the average nucleonic probability distribution of the form

\[ f_i(\vec{r}, \vec{p}, t) = \frac{1}{N} \sum_{i=1}^{A\times N} \delta(\vec{r} - \vec{r}_i)\delta(\vec{p} - \vec{p}_i) \]  

where \( N \) is a number of test-particles per nucleon, \( \vec{r}_i \) and \( \vec{p}_i \) are positions and momenta of the test-particles.

Time evolution of such a distribution is governed according to the classical Boltzmann equation

\[ \frac{df_i}{dt} = \frac{\partial f_i}{\partial t} + \vec{v}\vec{\nabla} f_i + \vec{V}\vec{\nabla} f_i - (U f_i) + I_{\text{coll}} \]

with \( \vec{r} \) and \( \vec{p} \) denoting position and momentum, \( \vec{v} \) the velocity of the particle, \( U \) density dependent mean-field potential. The \( I_{\text{coll}} \) term incorporates collision into transport equation. Considering only a scattering process with two particles in an initial and a final state \((1 + 2 \longrightarrow 1' + 2')\), the collision integral is written as

\[ I_{\text{coll}} = -\int d^3p_1 d^3p_2 \frac{d^3p_1 d^3p_2}{(2\pi)^9} \sigma_{12} \times \]

\[ [f_i f_2 (1 - f_i)(1 - f_2) - f_1 f_2 (1 - f_1)(1 - f_2)](2\pi)^3 \delta^3(p_1 + p_2 - p_1 - p_2) \]
where $\sigma$ is the scattering cross section, $v_{12}$ is the relative velocity of colliding particles 1 and 2 and $f_i(1 - f_i)$ are the Pauli blocking factors. Assuming that such a scattering occurs at point $\vec{r}$, the phase-space around $(\vec{r}, \vec{p}_1)(\vec{r}, \vec{p}_2)$ loses particles while the phase-space around $(\vec{r}, \vec{p}_1')(\vec{r}, \vec{p}_2')$ gains particles. That’s why the collision term includes so-called loss and gain terms [1]. A more complex scenario, like three-body collision is described in [2].

If the transport equation is used without the collision integral $\mathcal{I}$ it is called Vlasov’s equation

$$\frac{df}{dt} = \frac{\partial f}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{r} f_i + \mathbf{v} \cdot \nabla \mathbf{p} f_i U = 0.$$  

The main components of a dynamical model: initialization, mean field propagation and collision are described in this section.

### 2.1 Initialization

Preparation of the nuclear ground state is done in the Monte Carlo fashion. The test-particles are randomly distributed in the phase-space within the radius of the nucleus, $R$, and the Fermi momentum, $p_F$. The positions of the test particles imply a certain density used to calculate nuclear forces.

A sharp, homogenously charged sphere is used in most cases as an initial distribution. However, other more realistic descriptions were considered [3], with a diffused nuclear surface, e.g. the complementary error function and the Fermi function. The test particles move inside a nucleus according to Fermi motion. Momenta are assigned up to the value of the Fermi momentum calculated from the density distribution using the formula:
Initially, the system is generated according to the Fermi gas model, so the Pauli principle is fulfilled.

\[
p_f(\rho) = \frac{3}{2} \pi^2 \rho^\frac{1}{3} \rho(r)
\]

2.2 Mean filed propagation.

Each test-particle feels the mean field potential, defined as a function of the density. The most common form is the Skyrme potential [4,5], given by equation

\[
U(\rho(\vec{r})) = A \left[ \frac{\rho}{\rho_0} \right] + B \left[ \frac{\rho}{\rho_0} \right]^\sigma
\]

where \( \rho_0 \) is the central density and parameters A, B and \( \sigma \) are calculated to maintain a certain compressibility.

There could also be other components of the potential, in addition to the volume term, such as the asymmetry and surface terms and the Coulomb interaction for protons. Also, the momentum dependent mean field \[6,7\] is incorporated in newer versions of BUU/BNV models.

Propagation of each test-particle is governed by the same mean field potential, according to the classical equations of motion

\[
\frac{d\vec{r}_i}{dt} = \frac{\vec{p}_i}{m} \quad \frac{d\vec{p}_i}{dt} = -\nabla_v U(\rho(\vec{r})) .
\]

2.3 Collision

The implementation of collision between test-particles is done using so-called “parallel ensemble method” [8]. If each nucleon is represented by N test-particles, then all test-particles are divided into N ensembles and only members of the same ensemble can collide.
In this case, all test-particles have properties of real nucleons. After each collision the test-particles are checked to ensure that they satisfy the Pauli principle: a classical method of applying the fermionic properties of the test-particles that require that the density in phase-space does not exceed $1/\hbar^3$.

III. Simulation results

In the following, a discussion of modeling the nuclear ground state by various distributions of transport model is included. We compare the stability of nuclear ground state calculated using Vlasov’s equation (BNVn), Vlasov’s eq. and the constraint (BNVc) and also, Vlasov’s eq. and collision between two test-particles (BUU).

First of all, we explain how the energy minimization procedure, described in [9], works and then compare results of calculations with and without the constraint. The constraint, included in BNV model, is a numerical method of finding the real ground state (state with minimum energy). Initialization is done in the same fashion as described in the previous section. After preparation, minimization process is applied to find the energy minimum by enhancing the fermionic properties of the system. For numerical reasons, a grid is put in phase space and initial occupation factors averaged over all test-particles are calculated. Test-particles are propagated with classical equation of motion containing some extra numerical factors, as follows:

$$\frac{d}{dt} r_i = \frac{p_i}{m} + \mu_0 \nabla r_i U$$

$$\frac{d}{dt} p_i = -\nabla r_i U + \mu(f) \frac{p_i}{E_i}$$
where $\mu_0$ - negative frictional coefficient, $\mu(f_i)$ – f-dependent frictional coefficient, U-mean field potential. If the occupation factor of each test particle, $f_i$, exceeds 1 then $\mu(f_i)$ is positive (energy of the test-particle increases), otherwise $\mu(f_i)$ is negative (energy of the test-particle decreases). The phase-space properties of fermionic system should be preserved with time under these conditions.

**3.1 Mean field approach; Boltzmann-Nordheim-Vlasov model, (BNVn)**

First, we will discuss some properties of $^{197}$Au nucleus in its ground state modeled with BNV code without the constraint (only mean field included). As can be seen in Figs. 1-7, total energy per nucleon, averaged over all test-particles, builds up very rapidly up to a time of approximately 150 fm/c, while rms neutron and proton radii increase roughly about $\Delta r=1$ fm and one nucleon is considered as emitted at that time. With time, energy continues to increase, but with lower rate, to reach the value of 3.2 MeV at 450 fm/c and stays constant beyond that time. Neutron and proton radii roughly double their initial values at the time and tend to diverge with longer time scale. Ejected protons are repelled by the residual nucleus. Number of emitted nucleons increases gradually with time and at 1000 fm/c, about 85% of neutrons and protons have density below 0.0017 fm$^{-3}$ and are considered as free particles. The occupation factor, $\bar{f}$, averaged over test-particles, decreases with time as a consequence of increasing number of the available states in the phase-space during expansion.

The scenario observed in ground state simulation by BNVn can be understood as follows: nuclear system initialized according to Fermi gas model whose evolution is done with classical Hamiltonian loses its fermionic features and becomes classical. It means that some particles prefer to occupy “forbidden for fermions energy states” while others can build up energy and are able to overcome the barrier. Classical simulation of a quantum
object such as nucleus, results in an undesirable and spurious disintegration of a nucleus in its “ground state”. To find the real ground state and increase its stability, the constraint was included in BNV code.

3.2 Mean field approach with the constraint:

Boltzmann-Nordheim-Vlasov model, (BNVc)

Initial fermionic distribution is reasonably preserved with time. The average occupation of the value of $f_i=1.06$ remains constant over 1000 fm/c. Oscillations observed at early stage of evolution up to the time when “temporal equilibrium” is established, are produced as an effect of adding or subtracting energy of the test-particles in the energy minimization procedure. Binding energy per nucleon reproduces Weizsacker’s results, BE=7.9 MeV/A, very well up to time of 750 fm/c and it slowly starts to diverge to the value of BE=7.0 MeV/A at 1000 fm/c. Rms neutron and proton radii (r=5.6 fm) remain constant for approximately 750 fm/c and they tend to increase to r=7 fm/c at 1000 fm/c. Density distributions don’t indicate long tails seen in BNVn. Ground state is stable for a relatively long time. Static properties are reproduced are preserved with time very well. However, some undesirable products of the constraint become present. Problems with nuclear geometry are noticeable. Figs. 8, 9 show a slice in 2d spatial and momentum distribution of a $^{197}$Au nucleus in its ground state shortly after preparation (20fm/c), modeled with BNV code without and with the constraint. If constraint is included then distribution of test-particles in the phase-space becomes cubical instead of the spherical one. The distinctive edges are formed by accumulation of test-particles at the borders of the grid. One approach of solving this problem was decreasing the cell sizes of the grid. That was an unsuccessful attempt, because decreasing the cell sizes means one has to increase
dramatically the number of test-particles per nucleon to maintain mean field. Sensitivity of number of test-particles on mean field decreases with increasing number of test-particles per nucleon. It means that if we increase number of test particles from 1000 to 5000 per nucleon we will see more significant difference in average occupation (fewer fluctuations) then if we change the number of test particles from 5000 to 9000. BNV with the constraint is a great approach of calculating a stable ground state that could be later used for simulation of collision dynamics. However, the problem with nuclear shape has to be understood first.

3.3 Mean field and collision approach; Boltzmann-Uehling-Uhlenbeck, (BUU)

Unlike in BNV, collision term is included in BUU calculations of the ground state. However, while collision integral contains the Pauli blocking factors, the decay problem of the “ground state” still remains to some extent. It means that the Pauli blocking is not 100% effective. By detailed comparison of number of emitted nucleons from the “ground state” in BUU with BNVn (Figs.4-6), we can draw the conclusion that collision suppresses the spurious emission, but not sufficiently enough to make the ground state stable for longer that 100 fm/c.

IV. Conclusions

Correct interpretation of simulation results of reaction dynamics and prediction of experimental results is only possible if ground state properties of modeled nuclei are sufficiently well known. All three approaches of calculating ground state have their problems and simulation results could be model dependent.
Even though the Fermi distribution is used in an initial state, time evolution through classical equation of motion produces Boltzmannic distribution in a final state. BNVn and BUU show significant emission of nucleons from their nuclear “ground states”. Such a process is not observed if the constraint is included, because the energy minimization procedure maintains pretty well the fermionic features of nuclear system. On the other side, nucleus modeled by BNVc has problems with nuclear geometry while predicting quite successfully most observables. BNV model with the constraint has great future potential, however better understanding is essential in order to solve the problem with nuclear shape.

Acknowledgments

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Figures and Captions

![Graph](image)

Fig. 1: Average occupation in phase-space as a function of time calculated with the constraint (blue dots) and without the constraint (red line).
Fig. 2: Binding energy per nucleon as a function of time calculated with the constraint (blue dots) and without the constraint (red line). The dashed black line represents energy calculated according to the Weizsacker formula.

Fig. 3: RMS radii of protons (blue dots, black dashed-doted line) and neutrons (red line, light blue dashed line) and as a function of time calculated with the constraint (light blue dashed line and black dashed-doted line) and without the constraint (red line and blue dots).
Fig. 4: Cumulative number of free nucleons as a function of time calculated with BUU (red line), BNV without the constraint (green dashed line) and BNV with the constraint (blue dotted line).

Fig. 5: Cumulative number of free protons as a function of time calculated with BUU (red line), BNV without the constraint (green dashed line) and BNV with the constraint (blue dotted line).
Fig. 6: Cumulative number of free neutrons as a function of time calculated with BUU (red line), BNV without the constraint (green dashed line) and BNV with the constraint (blue dotted line).
Fig. 7: Density profiles at various times for $^{197}$Au modeled by BUU (first row), BNV without the constraint (second row) and BNV with the constraint (third row).
Fig. 8: A section of 1-fm width in z direction of the spatial density distribution of a $^{197}$Au nucleus in its ground state at 20 fm/c, modeled with BNV with the constraint a), and BNV with the constraint b). The grid cell size is 1.7 fm.

Fig. 9: A section of 1-fm$^{-1}$ width in $p_z$ direction of the momentum distribution of a $^{197}$Au nucleus in its ground state at 20 fm/c, modeled with BNV with the constraint, a), and BNV with the constraint, b). The grid cell size is 0.48 fm$^{-1}$. 
References

Correlations between Reaction Product Yields
as a Tool of Probing the Reaction Scenario

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Abstract

Experimental multidimensional joint distributions of neutrons and charged reaction products
were analyzed for $^{136}Xe + ^{209}Bi$ reaction at $E/A = 40$ MeV, and were found to exhibit several
different types of prominent correlation patterns. Some of these correlations have a simple
explanation in terms, e.g., of the system excitation energy and pose little challenge to most
of statistical decay theories. However, several types of the observed prominent correlation
patterns are difficult to reconcile with some, but not other possible reaction scenarios, which
makes them a useful tool for probing reactions scenarios, different from the traditional approach
of interpreting inclusive yields of individual reaction products.

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I. INTRODUCTION

Over the last 20 years, or so, considerable effort has been made both, experimentally and theoretically, to explore the heavy-ion reaction dynamics and to understand the production scenarios of various products. As a result of this effort, a consensus has emerged as to the general collision scenario at low bombarding energies and, perhaps, also at the lower part of the Fermi-energy domain. In this “consensus” domain, the projectile and target are believed to proceed in a way of a dissipative collision, where they form transiently a revolving dinuclear complex and where an ever increasing (with time passing) part of the kinetic energy of their relative motion is converted into their intrinsic thermal and rotational energies by means of stochastic nucleon exchange between them (the, now, projectile-like and target-like constituents). Subsequently, the dinuclear complex reseparates under the combined action of Coulomb and centrifugal forces and the projectile- (PLF) and target-like (TLF) fragments are set free to proceed on their individual Coulomb trajectories. Furthermore, the PLF and TLF are believed to emerge from the dissipative collision excited and thermally equilibrated and, accordingly, decay statistically. Super-imposed on the above simple scenario, but not interfering with it to any significant extent, is pre-equilibrium emission of neutrons and light charged particles (LCP), occurring mostly at early stages of collision history. It is important to note that instrumental in arriving at the above “consensus” picture were observations of individual product yields and, specifically, of patterns in the yield distributions characteristic of the inferred “consensus” scenario.

At higher bombarding energies, up to $E/A = 62.5$ MeV, the underlying general dissipative collision scenario still appears to persist [? ], however, here, a consensus is still lacking as to the production mechanisms of the observed reaction products. To some extent, this is so because of the increased role of two-body interactions and increased role of preequilibrium emission, including emission from the dynamically unstable interfragment “neck-like” structure. But more importantly, it is so because of the observation of copious production of intermediate-mass-fragments (IMFs) that appeared difficult to reconcile with a classical scenario of a statistical decay of an equilibrated system. The failure to reach a consensus at these higher energies may well point to inherent limita-
tions of a "one-dimensional" analysis based on yields of individual species of reaction products and, thus, call for an extended analysis scheme involving various correlations between the production patterns of different species, such as neutrons, LCPs, IMFs, PLFs, and TLFs. With the potential probing value of such correlations in mind, series of experiments were performed [? ], in which all of the many types of products were measured simultaneously, event-by-event. More specifically, in these experiments both, neutrons and charged reaction products were measured with 4π angular coverage using Rochester Superball neutron calorimeter/multiplicity meter in combination with one of the available 4π charged-product detector arrays (MSU MiniBall, Washington University DwarfBall/Wall and MicroBall). Additionally, PLFs were measured at forward angles, also with high geometrical efficiency resulting from their strong kinematical focussing.

The present study focuses on experimental data on 209Bi + 139Xe reaction at E/A = 40 MeV and aims at evaluating the significance of three types of prominent correlation patterns observed in multidimensional joint distributions of neutrons, LCPs, IMFs, and PLFs, as a tool of probing the underlying reaction scenario and, by implication, the production mechanisms of these species. The correlations considered include those between the neutron and charged particle multiplicities, between the average size of IMFs and the joint neutron and LCP multiplicity, and between the average size of PLF and the joint neutron and LCP multiplicity.

II. ESSENTIALS OF THE EXPERIMENTAL SETUP

The experiment was performed at the National Superconducting Cyclotron Laboratory of the Michigan State University. The beam of 136Xe ions from the K200 cyclotron, with energy of E/A = 40 MeV was focussed on a self-supporting 3.5 mg/cm² thick 209Bi target [5] placed in the operational center of the detector setup. The latter consisted of two 4π detector systems, the Washington University charged-particle detector array, DwarfBall/Wall [6] and the University of Rochester neutron calorimeter/multiplicity meter, SuperBall [7], and two position-sensitive silicon-detector telescopes placed at very forward angles. The telescopes covered angular range forward of the anticipated
grazing angle ($\theta_{\text{Grazing}} \approx 3.90^\circ$). The Dwarf array provided for a reliable $Z$ identification for atomic numbers up to $Z=20$, but also for the energy and emission angle measurement. The SuperBall provided for a high-efficiency event-by-event measurement of neutron multiplicities and of summed kinetic energy of neutrons, in five angular bins. The forward-angle telescopes provided for $Z$ identification of projectile like fragments, along with the energy and angle measurement. They were also sensitive to intermediate-mass fragments. All in all, the experimental setup allowed one to obtain almost a complete characterization of individual reaction events in terms of product identification, and their yields, and kinematical parameters.

III. THEORETICAL MODELLING OF VARIOUS REACTION SCENARIOS

In the present study, several theoretical models are used to assess the consistency of observed correlations with various reaction scenarios. And so, the dynamical, interaction phase of the collision history is modeled either using the classical trajectory code CLAT [8], based on a stochastic nucleon exchange model NEM [9], or the Quantum Molecular Dynamics (QMD) code CHIMERA [12], accounting more fully for the two-body interactions. The subsequent statistical decay of primary reaction products is modelled using either the equilibrium-statistical, sequential decay code Gemini [10] or the (pseudo-microcanonical) statistical multifragmentation code SMM [11]. Note that modeling of a complete scenario always requires successive application of one of the two interactions phase models followed by the statistical modeling of decay of the products emerging from the interaction phase.

In the present study, a version of the CHIMERA code [?] was used that includes isospin dependence of nuclear interactions. Calculations were made for soft EOS ($K \approx 200\text{MeV}$) with symmetry energy strength coefficient corresponding to ASY-STIFF EOS ($C = 31.4 \text{ MeV}$). Calculations with the CHIMERA code were performed for a time interval from 0 to up to 300 fm/c.

When modeling sequential decay of primary products, a version of the GEMINI code [13] was used that incorporates mutual Coulomb interaction of all emitted particles and the residue. In such a modeling, excited primary fragments were allowed to decay in
flight with proper time constants, with trajectories of all products being calculated by solving numerically sets of equations of motion.

The results of model calculations were subsequently “filtered” by emulating numerically the response of the DwarfBall/Wall and SuperBall $4\pi$ detector systems to the impinging flux of reaction products.

IV. EXPERIMENTAL RESULTS AND ANALYSIS

In the following subsections, three types of prominent correlations between the yields of four distinct classes of reaction products, neutrons, LCPs, IMFs, and PLFs are discussed and analyzed in terms of various reaction scenarios, represented by pairs of theoretical models discussed further above.

A. Neutron and LCP Multiplicity Correlation Patterns

It is well known from the literature \cite{1,2} that the joint multiplicity distributions of neutrons and light particles can be used as measures of kinetic energy dissipation achieved in individual reaction events. Such a distribution for the $^{136}Xe + ^{209}Bi$ reaction at $E/A = 40$ MeV is shown in the upper panel of Fig.1 in a form of a (logarithmic) contour plot, while the respective distributions expected for different interaction/decay scenarios are shown in a series of panels below. As seen in Fig. 1, the two-dimensional joint multiplicity distribution features a prominent intensity ridge with a crest line running first parallel to the neutron multiplicity axis and then, at $m_n \approx 30$, turning away from this axis, to continue along a line running at an angle with respect to the coordinate axes. Such a behavior has a natural explanation in phase-space based decay models, which favor strongly neutron emission at low excitation energies, hence the section of ridge parallel to the $m_n$ axis. At higher excitation energies, when temperature becomes comparable to the height of the Coulomb barrier for the emission of LCPs, emission of the latter can successfully compete with neutron emission, aided additionally by a buildup of iso-asymmetry with every neutron emitted.

As seen in Fig. 1, all theoretical scenarios, represented here by combinations of model codes “CLAT + GEMINI” (one-body interaction and sequential decay), “CLAT+SMM”
FIG. 1: Logarithmic contour plots of joint distribution of neutron $(m_n)$ and light-charged particle $(m_{LCP})$ for $^{136}Xe + ^{209}Bi$ reaction at E/A = 40 MeV.
(one-body interaction followed by simultaneous decay of systems at “freezeout” configurations), and “QMD+GEMINI” (one- and two-body dynamics followed by sequential decay of primary products), tend to reproduce the general appearance of the joint multiplicity distribution. However, there are significant differences as to the quality of the resemblance between the experimental plot, on the one hand and various theoretical predictions, on the other hand.

It appears that the least complex calculations based on the codes CLAT [8] and Gemini [10] provide for the best agreement with experimental observations, as far as the location of the crest line of the yield ridge is concerned. This particular combination is seen, however, to overestimate the length of the ridge, i.e., to convert too much of the available kinetic energy of the relative projectile-target motion into the thermal excitation energy. This is so here, because the presence of an intermediate-velocity source (IVS) is neglected, which is known to emit particles with higher average kinetic energies than the equilibrated PLF and TLF sources. Thus, one expects that by including preequilibrium emission represented by an IVS, one would shorten the length of the theoretical yield ridge, bringing it to a better agreement with the experimental one.

As seen in the third panel from top of Fig. 1, a combination of codes CLAT and SMM, predicts a saturation in neutron multiplicity, not observed experimentally. This fact not necessarily disqualifies SMM as a viable model, but rather points to a model deficiency in describing the relative neutron and LCP yields and should help in devising corrections to the model that remedy this deficiency.

The QMD + GEMINI calculations (bottom panel) predict to much neutron emission compared with LCP emission and an early (on the excitation energy scale) onset of LCP emission. Obviously, the latter is due to an excessive dynamical component in the LCP yield. The fact that there are generally too many neutrons emitted for a given LCP multiplicity tends to suggest that also the neutron yield is too much affected by dynamical emission during the interaction stage of the collision, a deficiency that, apparently, cannot be remedied by simple ad-hoc corrections to the QMD code CHIMERA [? ]. Therefore, one may conclude tentatively that at $E/E = 40$ MeV, the collision dynamics is still largely governed by one-body interaction of nucleons with a mean field of a dinuclear complex and the effects of direct nucleon-nucleon interactions are still too small.
to justify a “universal” use of QMD logic for evaluating individual yields of all reaction products. However, the possibility that QMD provides a reasonably good description of the interaction stage for a limited range of impact parameters cannot be excluded.

B. Correlations between Average Fragment Sizes and the Joint Multiplicity of Neutrons and LCP

In recent years, the presence of prominent correlations between the average sizes of IMFs and the joint multiplicity of neutrons and protons has been discovered [? ], which was found also to exhibit non-thermal scaling, i.e., dependence on bombarding energy. An example of such a correlation is illustrated in the top panel of Fig. 2, in a form of a contour plot of average atomic number of IMFs ($Z_{IMF}$), plotted versus the associated neutron and LCP multiplicities. As seen in this plot, the size of produced IMFs is correlated with neutron and LCP multiplicities such that larger sizes of IMFs are associated with higher excitation energies. The clear contour lines running almost perfectly parallel to each other appear to coincide with lines of constant excitation energy. Note that this kind of correlation involves three independently measured quantities, neutron and LCP multiplicities and the atomic numbers of IMFs.

It has been shown in earlier studies [? ] that the contour lines of equal $<Z_{IMF}>$ shift prominently toward higher neutron and LCP multiplicities as bombarding energy increases, which means that they do not scale directly with thermal excitation. This tends to suggest also that a significant portion of IMFs is produced in dynamical processes. Indeed, it has been suggested earlier [? ] that this kind of trend is consistent with the size of IMF scaling with the size of neck-like structure formed transiently between the interacting PLF and TLF. This is so, because for higher bombarding energies, a smaller overlap region leads to the same excitation energy that requires larger overlap at lower bombarding energies, hence the shift of contour lines toward higher excitation energies (same overlap).

Results of attempts to reproduce the observed correlations by three reaction scenarios are illustrated in three pairs of contour plots, where the leftmost panels represent “raw” trends and the rightmost panels illustrate trends expected for the actual detector setup.
FIG. 2: Logarithmic contour plot of average atomic number of IMFs, $\langle Z_{IMF} \rangle$, as a function of associated neutron and LCP multiplicities as observed in $^{136}Xe + ^{209}Bi$ reaction at $E/A = 40$ MeV. Here, $3 \leq Z_{IMF} \leq 16$. 
Obviously, only the latter plots merit comparison with experimental data. As seen in the rightmost panel in the second row, the combined “CLAT + GEMINI” results only faintly resemble the experimental trends, consistent with the role of the dynamical component in IMF yield (see discussion in the paragraph further above). This is also not surprising in view of the fact that very few IMFs are expected within the framework of these two models.

The rightmost panel in the third row illustrates the gross failure of the simultaneous multifragmentation model SMM [11] to account for a prominent experimental correlation pattern. While based on the argument presented above (regarding the dynamical component) one would not expect a good agreement with experiment in this case, the predicted correlations for the CLAT + SMM scenario are to a good extent orthogonal to those actually observed. This may be taken as indicative of a simultaneous breakup playing little, if any role in IMF production in the bombarding energy range considered and the IMF production occurring, perhaps, with no meaningful competition from particle decay channels.

The rightmost panel in the bottom row illustrates predictions of the QMD code CHIMERA [12] complemented by the dynamical version of the code GEMINI [13]. Even though the CHIMERA scenario appears inconsistent with the correlation pattern seen in the joint distribution of neutron and LCP multiplicities, it may still be responsible for IMF production in a limited range of impact parameters. This is possible in view of a strong indication that the IMFs, unlike neutrons and LCPs are produced here in dominantly dynamical primary processes, expected to be well described by QMD type of codes. Indeed, as seen in the respective panel, CHIMERA is capable to correctly render the trends observed experimentally and, most notably, the increase of average IMF size with increasing neutron and LCP multiplicities. In this respect, it would be highly desirable to repeat the time-consuming CHIMERA calculations for a different bombarding energy to verify if such calculations are capable of accounting also for the experimentally observed trends in the bombarding energy dependence of the correlations discussed.
C. Correlations between the Size of PLF and the Joint Multiplicity of Neutrons and LCP

Figure 3 illustrates correlations between the average size of the PLF and the associated joint multiplicity of neutrons and LCPs. These correlations appear easy to understand within a prevailing scenario of a dissipative collision followed by statistical and sequential decay of primary PLF and TLF, as modelled by $CLAT + GEMINI$ combination. Yet, they can be also understood within the frameworks of the combined $CLAT + SMM$ and of the combined $QMD + GEMINI$ scenarios. It appears then that this type of correlation is of a lesser value as far as probing the more detailed interactions scenario. Yet the fact that they do not contradict conclusions reached further above is in itself encouraging.

V. SUMMARY

The present study has shown that certain prominent correlations observed between the yields of different reaction products can be used to probe the underlying collision and decay scenario and, hence serve as a guidance in devising models of heavy-ion collision and decay of the excited primary products. Already the simple correlation pattern between the multiplicities of neutrons and LCPs tends to exclude dynamical QMD models as describing the overall collision scenario in the bombarding energy domain considered. It appears that at $E/A = 40 MeV$ the dynamics is still largely dominated by one-body interaction with lesser propensity to a preequilibrium release of particles and fragments than exhibited by a two-body interaction dominated scenario.

Concerning the IMF production, it appears dominated in this case by dynamical processes, perhaps, including QMD-like scenarios. This conclusion is largely based on the bombarding-energy dependence of the correlations between the average size of IMFs and the joint neutron and LCP multiplicity, reported earlier, but corroborated here by the trends observed at a single bombarding energy. On the other hand a simultaneous break-up in a SMM-like freezeout scenario appears strongly contradicted by the prominent experimental trends and there seems to be little hint at this time as to how this gross deficiency of SMM could be remedied.
FIG. 3: Logarithmic contour plot of average atomic number of the detected PLF, $< Z_{PLF} >$, as a function of associated neutron and LCP multiplicities, as observed in $^{136}Xe + ^{209}Bi$ reaction at $E/A = 40$ MeV.
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The dissipative Reaction Environment at Intermediate Energies

Excitation Function of the Reaction $^{136}\text{Xe} + ^{209}\text{Bi}$
at E/A = 28, 40, and 62 MeV


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ABSTRACT

The phenomenology of reaction features was studied for $^{136}\text{Xe} + ^{209}\text{Bi}$ reactions at E/A = 28, 40, and 62 MeV. Correlations between the kinetic energy and the deflection angle of projectile-like fragments (Wilczynski Plot) are seen to exhibit features characteristic of dissipative orbiting, commonly found at bombarding energies of a few MeV/nucleon above the interaction barrier, but also reported in the Fermi-energy domain. The reaction cross section is dominated by dissipative binary reactions of well-defined projectile- and target-like fragments. On the other hand, the Galilei-invariant velocity distributions of various charged reaction products, show presence of a third, intermediate-velocity source of emitted fragments. While the production of light-charged particles can be attributed mainly to evaporation from excited projectile- and target-like fragment, an intermediate-velocity source appears responsible for large amount of the observed intermediate-mass fragment yields for high excitation energies. Fragments emitted from the intermediate-velocity source appear to be produced dynamically in the overlap zone of the projectile and target nuclei.
I. INTRODUCTION

It is well known that in heavy-ion reactions at low bombarding energies (below 10 MeV/nucleon), the excited primary projectile- (PLF) and target-like (TLF) fragments deexcite primarily via emission of neutrons and light-charged particles (LCP)[1]. With increasing bombarding energy one begins to observe additionally emission of intermediate-mass fragments (IMFs)[2], which are seen to account for a considerable fraction of the system mass/energy for central collisions[2–5]. It has been subsequently found that many salient features of low-energy reactions are still present in the intermediate bombarding energy range of E/A= 10-100 MeV[5]. In the associated reaction scenario, the reaction cross-section is dominated by dissipative binary collisions, with well defined projectile- and target-like fragments (PLF and TLF) [1]. The observed particle yield is then to a large extent associated with statistical emission from these two fragments, excited in the course of their mutual interaction. However, with increasing collision energy, a new source of fragments is expected to enter the play that moves with a velocity intermediate between the velocities of PLF and TLF[3–7]. This intermediate-velocity source (IVS) can be conceptually associated with the overlap region of PLF and TLF[3], and is similar to the mid-rapidity source, observed in collisions at much higher energies[8]. As this source appears responsible for much of the IMF yield, it is the latter yield and its characteristics that provides an experimental handle on the formation and properties of IVS.

Creation of the intermediate velocity sources in heavy ion collisions is suggested by: (i) the semi-classical microscopic dynamical calculations - BUU [9], BNV [10], VUU [11], LV [12], (ii) molecular dynamics calculations - QMD [13–16], QPD [17], FMD [18], AMD [19], and (iii) stochastic models [20]. Existence of the IVS was experimentally verified by inspecting invariant velocity plots [3, 21, 22], rapidity and transversal energy distributions [23], or by a reconstruction-subtraction procedure [15].

In the present, relatively brief and cursory outline of a long paper in preparation, results are reported for the measurement of an excitation function of the $^{136}\text{Xe} + ^{209}\text{Bi}$ reaction. This system was studied at E/A = 28, 40, and 62 MeV, using two 4π detector systems, one for charged particles and one for neutrons. A description of experimen-
tal procedure is given in Section II. Main features of fragment emissions are presented in Section III. The PLF yield is analyzed in Section IV. In Section V the total excitation energy is reconstructed. Section VI gives a characteristic of different emission sources in four bins of total excitation energy. The final section contains a summary and conclusions.

II. EXPERIMENTAL SET-UP AND PROCEDURE

The experiment was performed at the National Superconducting Cyclotron Laboratory of Michigan State University. Beams of $^{136}$Xe ions with energies of E/A=28, 40, and 62 MeV were focused on self-supporting 3.5 mg/cm$^2$ thick $^{209}$Bi targets.

The experimental setups included in every case two 4π detector systems:
(i) the Washington University charged-particle detector array - Dwarf Ball/Wall [24],
(ii) the University of Rochester neutron calorimeter RedBall for 28 MeV/nucleon study, and the SuperBall neutron multiplicity meter [25] for 40 and 62 MeV/nucleon studies.

Projectile-like fragments were detected using silicon-detector telescopes placed at very forward angles, including the angular region around the grazing angle. In the 28 and 62 MeV/nucleon studies, the PLF telescopes were position sensitive, allowing for an accurate measurement of the PLF emission angle. Additionally, in the 40 and 62 MeV/nucleon experiments four of the Dwarf Ball phoswitch detectors were substituted by two Si-Si and two Si-CsI detectors allowing mass resolution up to $Z = 8$.

The used setups were highly efficient and allowed one to collect information on the multiplicities of emitted neutrons and charged-particles and on the energies and Z-identity of charged reaction products. The Dwarf Ball/Wall array provided for a good mass resolution for $Z = 1$ and 2. Good charge resolution was achieved up to $Z = 35$, and approximate good resolution up to $Z = 40$. The details of charge and energy calibration for the Dwarf Ball/Wall array are published in paper[26]. The 40 and 62 MeV/nucleon experiments were calibrated using a new method based on fitting observed light output in actual data from the physics or “production” runs, to energy deposits calculated for detector elements [26]. The method was verified using 28 MeV/nucleon experiment data where calibration beams were used along with standard calibration method [24].
The RedBall and SuperBall neutron multiplicity meters were using Gd-doped liquid scintillator (National Diagnostic ND-309), allowing to capture moderated neutrons. The light pulses from neutron capture were counted, allowing multiplicity measurement. Recorded capture times allowed to eliminate “cross-talk” from different scintillation detectors. Additionally, the Superball neutron multiplicity meter was able to measure prompt light output produced by neutrons and gamma rays entering the liquid scintillator. An example of correlation between prompt light output and measured neutron multiplicity is shown in Fig. 1, for $^{136}\text{Xe} + ^{209}\text{Bi}$ reaction at 40 MeV/nucleon. As one can see, the correlation is almost linear, except for central collision region (high neutron multiplicities). Generally, the amount of emitted neutrons is proportional to the centrality of collision. Because of the dispersion in emitted fragments for given impact parameter, as well as, due to detection system efficiency, a broad range of “central” collisions form a “bump” for high neutron multiplicities.

The RedBall and Superball neutron multiplicity meters were operating using 128µs time gate for collecting light output from moderated (captured) neutrons. Such a time gate is very long as compared to $2 \div 3\mu s$ time gates used by the Dwarf Wall/Ball array. Therefore, the combined RedBall or Superball and Dwarf Ball/Wall $4\pi$ detector systems were working using low beam intensities (few hundred counts per second maximum). The low beam intensities have, on the other hand, an advantage in very low event pile-ups, allowing one to collect “pure” event samples. In the “off-line” analysis background and random counts for RedBall/Superball device were eliminated. For the Dwarf Ball/Wall array all pedestal counts were removed, and only identified particles were accepted. The greater then zero neutron multiplicity and/or greater then zero charged particle multiplicity condition was used to select “well defined” events. Finally, the collected statistics in the $^{136}\text{Xe} + ^{209}\text{Bi}$ reaction at $E/A = 28, 40,$ and 62 MeV was $16 \times 10^6, 10 \times 10^6,$ and $6 \times 10^6$ “well defined” events, respectively.

The average efficiency for single neutron detection for SuperBall neutron multiplicity meter is presented in Fig. 2. Effective neutron multiplicity for a given event was determined by simulation, including geometry of detection system, moderator material, as well as light output collection threshold and efficiency. Similar technique was applied for charged particles, where each of Dwarf Ball/Wall or other detector is simulated, taking
into account its geometry and detection threshold for a given species. The detection thresholds of Dwarf Ball/Wall are shown in Fig. 3. The four most forward Dwarf Wall detectors were additionally shielded with \( \approx 200\text{mg/cm}^2 \) Pb absorbers. Table I summarizes average efficiencies for neutron, LCP, and IMF detection, for given bombarding energies.

<table>
<thead>
<tr>
<th>SPECIES</th>
<th>28 MeV/nucleon</th>
<th>40 MeV/nucleon</th>
<th>62 MeV/nucleon</th>
</tr>
</thead>
<tbody>
<tr>
<td>neutron</td>
<td>0.52</td>
<td>0.65</td>
<td>0.57</td>
</tr>
<tr>
<td>LCP</td>
<td>0.42</td>
<td>0.38</td>
<td>0.32</td>
</tr>
<tr>
<td>IMF</td>
<td>0.38</td>
<td>0.32</td>
<td>0.25</td>
</tr>
</tbody>
</table>

**TABLE I**: Average efficiency for neutron, LCP, and IMF\((3 \leq Z_{\text{IMF}} \leq 16)\) detection for \(^{136}\text{Xe} + ^{209}\text{Bi}\) reactions at \(E/A= 28, 40\) and 62 MeV, respectively.

### III. CHARACTERISTICS OF PARTICLE EMISSION

The main features of particle emission is well represented by a joint distribution of neutron and light-charged particles (LCP) multiplicities, presented in Fig. 4. The contour plot was made for 62 MeV/nucleon. Profiles for 28, 40, and 62 MeV/nucleon data are presented as squares, triangles, and circles, respectively. The white stars illustrates model calculations, where the production of primary fragments was modelled by classical dynamical code CLAT [27], based on the stochastic nucleon exchange model NEM [28]. Subsequently, the decay of the primary fragments was modelled by the equilibrium-statistical deexcitation model as implemented in the code GEMINI[29].

The joint multiplicity distribution provides a good information about the reaction evolution with decreasing impact parameter (increasing excitation energy). Its characteristic shape reflects basic features of particle emission. Thus, for very peripheral collisions one observes only neutron emission - the ridge parallel to the \(m_n\) axis \((0 \leq m_n \leq 28)\) in Fig. 4. Since neutrons do not have a Coulomb barrier for emission, they can be emitted even for very low excitation energies. When the excitation energy is high enough to let charged-particles to pass the barrier, the light-charged particles (LCP) emission starts, and one observes a bend of emission pattern in Fig. 4.
As one can see, the model calculations represents quite good the experimental data. This indicates that the subset of experimental observations included in this figure is consistent with a scenario of predominantly binary dissipative collisions followed by statistical decay of the primary PLF and TLF. However, one should notice that the evaporation stage has a dominant influence on the shape of these fragment distributions. Therefore, this agreement is not sufficient to exclude other reaction modes as the participant-spectator model [30].

The profiles of joint multiplicities of neutron and LCPs shown in Fig. 4, exhibit nearly an invariance with respect to bombarding energy, starting with the same offset for LCP emission. Such invariance is an indication of thermal scaling. It can be understood in terms of thermal (statistical) emission from equilibrated PLF and TLF sources [31].

With further increase of excitation energy the emission of intermediate-mass fragments (IMF) becomes possible, competing with light-charged particle emission. This fact is illustrated in Fig. 5, were the joint distribution of neutron and LCP multiplicities was “gated” with multiplicity of the detected IMFs. For \( m_{IMF} = 0 \) one observes mostly neutron and LCP emission for peripheral and midperipheral collisions. Generally, the emission of IMFs starts for higher excitation energies, as compared to LCP emission, and is characteristic for central collisions. On the other hand, even for peripheral collisions there is a very small, but not negligible, probability for emitting IMF.

The emission of IMFs is strongly correlated with the thermal excitation energy. This fact is well illustrated in Fig. 6, where the average atomic number of IMFs, \( < Z_{IMF} > \), was plotted as a function of associated neutron and LCP multiplicities. As seen in Fig. 6, the \( < Z_{IMF} > \) increases noticeably and systematically toward higher neutron and LCP multiplicities as the excitation energy increases. Assuming thermal emission, one would expect increase of IMF emission with excitation energy. Also, one would expect smaller sizes of IMFs when more LCPs is emitted, according to \( Z \) conservation. The observed shift of equi-\( < Z_{IMF} > \) lines is opposite. This fact can be explained assuming a prevail of the dynamical emission of IMFs over the statistical one. In the dynamical emission the observed correlation pattern would show “secondary” character, reflecting “primary” dynamical and/or geometrical correlation between IMF \( Z \) distribution and violence of collision and/or the size of the overlap (interaction) zone, produced in course of the
collision between the projectile and target nuclei. In such a scenario, it is natural to expect a positive correlation of IMF sizes with size of the interaction zone. Further, for equal sizes of the overlap zones (equal $< Z_{IMF} >$ in hypothesis under consideration), the degree of achieved energy dumping would increase with increase of bombarding energy. This would give a rise to more neutrons and LCPs emission at higher bombarding energy for a fixed $< Z_{IMF} >$. As one can see, the letter effect is consistent with experimental patterns in Fig. 6.

IV. THE PLF YIELD

The patterns presented in this section were observed using correlations with charge detected by the PLF telescope, $Z_{PLF}$, placed close to the grazing angle.

Fig. 7 illustrates experimental correlations between the atomic number $Z$ and the energy $E$ for charged fragments detected near the grazing angle as observed for $^{136}Xe + ^{209}Bi$ reactions at bombarding energies of E/A=28, 40, and 62 MeV. The fragment yield is seen to be concentrated along well-defined “ridges”, running from regions of elastic and quasielastic collisions down to the regions of intermediate-mass fragments near the origins of the plots. The continuity of the yield ridges in these plots and the absence of any discernible bimodal pattern in the yield distributions, which would be characteristic for PLF fission [32], indicates that the observed yield is associated mainly with PLF residues and is not contaminated by PLF fission fragments to any significant degree. The observed product yield is also free from contamination by the residues of target-like fragments (TLF), as the latter do not have energy sufficient to overcome the identification threshold. The white boxes seen in Fig. 7 for E/A=40 MeV illustrates model calculations by CLAT+GEMINI codes. The model calculations reproduce the location of crest line of the yield ridge quite accurately. However, one should note that the evaporation stage has a dominant influence on the shape of this correlations.

The charge of the detected PLF residuum is correlated with the excitation energy of the primary PLF. The higher is the excitation energy, the more particles is emitted, and consequently, the smaller is the detected PLF charge. In order to illustrate this relation Fig. 8 presents correlation between the average atomic number of the detected
PLF charge, $<Z_{PLF}>$, and the associated neutron and LCP multiplicities. As one can see, with increase of excitation energy, i.e. increase of neutron and LCP multiplicities, the average charge of the PLF residuum decrease. This correlation is, in fact, opposite to that observed for average charge of emitted IMFs in Fig. 6. Thus, with increase of excitation energy the average charge of PLF decrease and the average charge of emitted IMFs increase. To some extend this behavior can be interpreted simply in terms of charge conservation rule. However, one should take into account also the reaction mechanism, where important role is played by the overlap zone of colliding heavy-ions.

The predominantly binary dissipative collisions scenario is confirmed by the observed “deflection-function plots” of the fragment yield as a function of the energy and the emission angle of PLFs. Such plots, analogous to Wilczynski plots[33], are displayed in the two top leftmost panels of Fig. 9 for bombarding energies of E/A=28 and 62 MeV. Additionally, the middle and rightmost panels of this figure illustrate the PLF deflection functions separately, for peripheral ($M_{IMF} = 0$), and for mid-central/peripheral collisions ($M_{IMF} > 0$). The bottom panel of Fig. 9 presents a schematic view of various classes of system trajectories, representing elastic (1), grazing (2), moderately-damped (3), and negative-deflection (4) collisions. The regions of the deflection-function plots corresponding to these four schematic classes of trajectories, are labelled by the respective numerals 1 – 4 in the two top leftmost panels of Fig. 9. As seen in these panels, sections of the yield ridges associated with elastic scattering connect areas labelled as 1 and 2. For midperipheral collisions, the two colliding heavy-ions form a transient dinuclear system and orbit around each other for a fraction of a revolution while dissipating some of their relative kinetic energy. This process is reflected in segments 2-3 and 3-4 of the yield ridges. For the segment connecting areas 2 and 3, the PLF deflection angle is seen to decrease with increasing energy dissipation, reflecting the fact that both, the energy dissipation and the deflection angle are functions of the impact parameter, and that with decreasing impact parameter, the former increases while the latter decreases. For the segment connecting areas 3 and 4, on the other hand, the deflection angle is seen to increase with increasing energy dissipation and, hence, decreasing impact parameter. This could simply be a result of an experimental inability to distinguish negative-angle deflection (as in the case of the class 4 of trajectories in the bottom panel of Fig. 9) from
the positive-angle scattering. It is worth noting that there is no conceptual difference between dissipative orbiting leading to either positive or negative deflection angles, since the angle of zero degrees plays no special role.

It appears from Fig. 9 that the general collision scenario at $E/A=62$ MeV, is identical as dissipative orbiting with a subsequent statistical decay of the primary PLF and TLF. It is essentially the same as it was earlier found at lower bombarding energies [1]. As for lower bombarding energies, at $E/A=62$ MeV too, most of the reaction cross section is associated with binary collisions, where the term “binary” refers to the primary collision phenomenology of only two massive fragments, and not to the number of reaction products actually observed. In view of the above, the transition to a high-energy scenario dominated by two-body interactions and two-body dissipation [34] must occur at higher bombarding energies than $E/A=62$ MeV [35].

Figure 10 presents a comparison between experimental Wilczynski plots and model calculations (CLAT+GEMINI). As seen in this figure, the stochastic nucleon exchange model explains the general “topography” of the yield ridges qualitatively, such that each experimentally observed section of the ridge finds its equivalent in the model predictions. The model calculations are in good agreement with experimental data for “grazing” angle, as well for peripheral collisions. Such an agreement is expected since for very peripheral collisions the one-body dissipation is dominant. It fails, however, to describe the lower ridge of observed pattern quantitatively. In particular, it is due to the fact that the one-body dumping of the relative kinetic energy is too strong for high energy dissipations, where two-body dissipation becomes to play important role. It might indicate also, a change of reaction mechanism for high energy dissipation, where the scenario of binary dissipative collisions followed by statistical decay of the primary PLF and TLF is no longer dominant.

V. THE EXCITATION ENERGY

The system behavior during the fragmentation process depends on the excitation energy [1, 2]. As it was shown here, the joint neutron-LCP multiplicity, correlated with IMF and PLF emission allows one study the reaction properties for different ranges of
Generally, the total excitation energy can be calculated from the following formula:

\[ E^* = E_{CM}^{P+T} + Q_{gg} - \sum_{i=1}^{M} E_{cm}^i, \]  

where \( E_{CM}^{P+T} \) is the collision energy in the center-of-mass system, \( Q_{gg} \) is the difference between the mass excess of final and initial fragments, and \( E_{cm}^i \) is the center-of-mass energy of the \( i^{th} \) fragment. Here, \( M \) is the multiplicity of all final fragments in a given event.

Formula 1 is exact only if one measures all final fragments. In the real experiments, there is no full coverage of the solid angle, and detectors are working with certain threshold for particle detection (see Fig. 3). Therefore, instead of Formula 1 one should use an approximation:

\[ E^* = E_{CM}^{P+T} + < Q_{gg}(Z_{PLF}) > - E_{cm}^n(Z_{PLF}) - E_{cm}^{TLF}(Z_{PLF}) > \]

\[-m_n < E_{cm}^n(Z_{PLF}) > - \frac{1}{P_n} < P_n > < m_n(Z_{PLF}) > < E_{cm}^n(Z_{PLF}) > \]

\[-\sum E_{cm}^{LCP} - \frac{1}{P_{LCP}} < m_{LCP}(Z_{PLF}) > < E_{cm}^{LCP}(Z_{PLF}) > f_{LCP}^{R} \]

\[-\sum E_{cm}^{IMF} - \frac{1}{P_{IMF}} < m_{IMF}(Z_{PLF}) > < E_{cm}^{IMF}(Z_{PLF}) > f_{IMF}^{R} \]

The summation in Formula 3 is made over detected particles, while the corrections for not detected particles are made using the average values of particle multiplicity and energy in different bins of the detected PLF charge (see Fig. 8). The \( P_n, P_{LCP}, \) and \( P_{IMF} \) coefficients denotes the average probability of detecting single neutron (n), light-charge particle (LCP), and intermediate-mass fragment (IMF), respectively - see Table I. The Dwarf Ball/Wall detector array used for charged particles detection is a relatively high detection threshold device - see Fig. 3. Therefore, the average values of particles energy taken from experimental data will be overestimated. The correction factors \( f_{LCP}^{R} \) and \( f_{IMF}^{R} \) - see Table II, obtained from simulation, correct this overestimation.

Figure 11 presents experimental and model (CLAT+GEMINI) dependence of Formula 3 parameters as a function of the detected PLF charge, \( Z_{PLF} \). In the top row the model estimation of \( Q_{gg} \) and TLF center-of-mass energy is shown. The experimental
TABLE II: Correction factors for LCP, and IMF(3 \leq Z_{IMF} \leq 16) average energy for $^{136}Xe + ^{209}Bi$ reactions at E/A = 28, 40 and 62 MeV, respectively.

<table>
<thead>
<tr>
<th>FACTOR</th>
<th>28 MeV/nucleon</th>
<th>40 MeV/nucleon</th>
<th>62 MeV/nucleon</th>
</tr>
</thead>
<tbody>
<tr>
<td>$f^R_{LCP}$</td>
<td>0.92</td>
<td>0.80</td>
<td>0.80</td>
</tr>
<tr>
<td>$f^R_{IMF}$</td>
<td>0.85</td>
<td>0.75</td>
<td>0.75</td>
</tr>
</tbody>
</table>

and model (average neutrons energy) dependence of average multiplicities and average center-of-mass energies for different species are shown in the subsequent rows. Fig. 11, together with Tables I and II allows one to reconstruct the total excitation energy for different bombarding energies using Formula 3. One should note, that such a reconstruction is model dependent. However, the model dependence can be reduced using a low threshold $4\pi$ array with the possibility of TLF detection, such as the CHIMERA multidetector[36].

The excitation energy reconstruction based on correlations with detected PLF charge is limited to $Z_{PLF} = 18$ (see Figs ?? and ??). Lighter PLFs will be not distinguishable from the IMFs. It means, that one cannot use the approximation based on Formula 3 to reconstruct the excitation energy for very central collision, where the system is highly fragmented. Of course, the very central collisions represent only a very small amount of the entire reaction cross-section. It does not affect the analysis of reaction gross-features presented in this contribution.

The total excitation energy per nucleon obtained by applying Formula 3 to the experimental data is presented in Fig. 12. The left column shows the total excitation energy per nucleon as the function of the associated neutron and LCP multiplicities. The borders of the equi-$E^*/A$ lines were used to divide the entire reaction region into four regions, representing peripheral, midperipheral, midcentral, and central collisions, respectively. The distributions of reconstructed total excitation energy per nucleon within the chosen bins are shown in the right column. The average values of the total excitation energy per nucleon for different reaction regions are shown in Table III. As one can see from Fig. 3 and Table III, the average value of excitation energy increase for subsequent selection regions associated with increasing joint neutron-LCP multiplicity.
Table III: Average total excitation energy per nucleon in specified reaction regions for $^{136}Xe + ^{209}Bi$ reactions at E/A = 28, 40 and 62 MeV, respectively.

<table>
<thead>
<tr>
<th>REGION</th>
<th>28 MeV/nucleon</th>
<th>40 MeV/nucleon</th>
<th>62 MeV/nucleon</th>
</tr>
</thead>
<tbody>
<tr>
<td>peripheral</td>
<td>0.61</td>
<td>0.68</td>
<td>0.95</td>
</tr>
<tr>
<td>midperipheral</td>
<td>1.43</td>
<td>1.43</td>
<td>1.50</td>
</tr>
<tr>
<td>midcentral</td>
<td>1.82</td>
<td>1.95</td>
<td>2.19</td>
</tr>
<tr>
<td>central</td>
<td>2.18</td>
<td>2.56</td>
<td>2.62</td>
</tr>
</tbody>
</table>

Figure 12 shows the increase of excitation energy with increase of neutron and LCP multiplicities. As one can see, the central collisions region, where the excitation energy is highest, is very broad. This fact is rather due to an uncertainty caused by the reconstruction procedure, than to dispersion of the real excitation energy for given class of events. An eventual use of lower detection threshold $4\pi$ device should reduce the uncertainty of total excitation reconstruction.

The effects of change of IMF emission with bombarding and excitation energy is presented in Fig. 13, for charge distributions seen by the Dwarf Ball/Wall multi-detector array. The top left figure shows inclusive charge distributions for 28, 40, and 62 MeV/nucleon, respectively. The inclusive spectra show a maximum for the LCP’s and then decreases rapidly with the Z value of emitted fragments. No increase is observed for the largest Z values which could indicate some PLF associated fragments. This effect is due to Dwarf Ball/Wall high detection thresholds and mainly due to the detector geometry, which allows one to detect charge particles in the range from $\theta = 4.6^\circ$ to $\theta = 167.1^\circ$. Thus, Dwarf Ball/Wall allows one to measure mainly LCPs and IMFs with a very small amount of PLF residues. Figures presenting charge distributions for corresponding impact parameter (excitation energy) ranges are made using the selection of Fig. 12. It can be clearly seen that for peripheral collisions LCP emission is dominant, while for central ones IMF emission begins to compete, playing an important role in a system deexcitation. Comparing the relative yields of IMF production for different impact parameter range one can see that most of the IMF production, as compared to the total reaction cross section, is for central collisions. As one can see, the observed
trend for increasing bombarding energy is opposite to that with increasing excitation energy. Higher is the bombarding energy, smaller IMFs are produced. In opposite, higher is the excitation energy, emission of heavier IMFs is preferred. This picture is coherent with that shown in Fig. 6, and as Fig. 6, would suggest a strong competition between thermal and dynamical production of IMFs.

VI. THE SOURCES OF PARTICLE EMISSION

A common technique of identifying sources of particles emitted in low- and intermediate-energy heavy-ion reactions involves measurement of particle velocities and the subsequent construction of Galilei-invariant distributions of these velocities. Such distributions are conveniently visualized in the form of contour plots of the invariant (renormalized) cross section in a coordinate system of the velocity components parallel and perpendicular to the beam axis. A total of 24 such plots for α-particles and IMFs (here, $3 \leq Z_{IMF} \leq 10$) emitted in $^{136}Xe + ^{209}Bi$ reactions at $E/A = 28, 40, \text{ and } 62 \text{ MeV}$ is shown in Figs. 14 and 15. The plots were “gated” with different bins in the associated impact parameter (excitation energy) range (see Fig. 12).

In “landscapes” of the plots shown in Figs. 14 and 15, three distinct components can be discerned that can be attributed to three emitting effective sources. As seen in Fig. 14, for peripheral and mid-peripheral collisions there are two sources of emitted particles discernible, that can be identified with fully accelerated PLF and TLF. The particle yield is concentrated in this case along two well-defined semi-circular “Coulomb ridges”, centered around the projectile and target velocity, respectively. This picture is largely the same as observed for low energy collisions [1]. The source characteristics changes gradually with increasing excitation energy (decreasing of impact parameter). As seen in Fig. 14, a third source of emitted particles, of a velocity intermediate between the PLF and TLF velocities, first appears and then becomes more and more pronounced as the associated excitation energy increases. This intermediate-velocity source (IVS) is especially prominent at $E/A= 40 \text{ and } 62 \text{ MeV}$. For high bombarding energies, the high relative TLF-PLF velocity allows for a good separation of sources of emitted fragments.

Similar emission patterns to those seen for α-particles in Fig. 14 are observed in
Fig. 15 for intermediate-mass fragments. The intermediate velocity region is seen to be populated much more strongly for central collisions than for peripheral ones. Comparing the relative intensities in the IVS region for $E/A = 40$ MeV and $E/A = 62$ MeV, one finds there similarities in increase of IMF emission for central collisions. The increase in the IMF production rate in the IVS region is seen also for the $E/A = 28$ MeV case. However, due to smaller PLF-TLF relative velocity, one observes large overlap of IMF emission patterns associated with PLF, TLF and IVS. It is worth to notice, that even for peripheral collision one still observe non-negligible IMF emission rate.

The observed fragment yield, associated with various sources, can be presented using Galilei-invariant velocity plots to achieve kinematical separation of different sources. Results of such a separation are illustrated in Fig. 16 for the case of intermediate-mass fragments from the $^{136}$Xe+$^{209}$Bi reaction at $E/A = 40$ and 62 MeV. The $E/A = 28$ MeV reaction was here excluded, since the PLF-TLF relative velocity is in this case too small to allow for a good kinematical separation of the particle yields associated with PLF and TLF sources. The IMF multiplicity distributions presented in the bottom panels of Fig. 16 were obtained by segregating events according to the location of their image on the $v_r$ vs. $v_\parallel$ plane relative to the “operational” boundaries for the three sources. Such boundaries are denoted by solid lines in the top panels of Fig. 16.

The probabilities of IMF emission from different sources identified by Fig. 16 selection, are listed in Table IV. It is clear that such a “raw” selection does not prevent certain degree of mis-identification of the particle emission source. Effects of such mis-identification decrease with increasing relative TLF-PLF velocity. The Table IV shows, that in central collisions, the IMF emission from the IVS dominates, being nearly as twice large as emission from TLF or PLF sources.

As seen in Fig. 16 and Table IV, for events with high IMF multiplicities for central collisions, large amount of the observed IMFs are originating from IVS. The increase of IMF emission in the IVS zone for central collisions can be explained, to some extent, by the geometry of the transient dinuclear system formed in a heavy-ion reaction, where the effective volume of the matter-overlap zone of the two colliding heavy ions is increasing with decreasing impact parameter. An increased rate of IMF production may be then a simple reflection of an increase in the volume of the overlap region. Assuming that the
IVS source represents the nuclear matter in the overlap region[3, 37], one would expect to observe increased fragment emission from this region for central collisions. Such a dynamical emission scenario is consistent with the trends in relative contributions from IVS zone seen in bottom Fig. 16.

It appears, that for low impact parameters, large amount of $\alpha$ particles and IMFs are produced \textit{dynamically} in the PLF and TLF overlap region, rather than being emitted \textit{statistically} from equilibrated hot PLF and TLF fragments. A support for this claim comes from Fig. 17, where energy distributions of $\alpha$ particles and IMFs, (here $Z=3$), are presented in the emission source velocity frame of PLF, TLF, and reaction center-of-mass (IVS), respectively. Here, bins 1 and 2 for peripheral collisions (see Fig. 12), were added in order to increase statistics. In order to decrease mis-identification of emission sources, the fragments were selected in forward and backward hemispheres of source velocity for the PLF and TLF, respectively. Fragments from the IVS zone were selected between the end of “Coulomb ring” of TLF (see Figs. 14 and 15 and the center-of-mass velocity.

As one can see, the energy distributions of fragments emitted from PLF and TLF have the same slopes(the energy spectra of fragments emitted from PLF for 62 MeV/nucleon are truncated because of “punch-trough” effect), while fragments produced in the IVS zone exhibit more “hard” energy spectra. It is so, also for peripheral collisions, where the IVS component is clearly visible for high energies. The “hard” - \textit{dynamical emission of IMFs}, from the IVS becomes more competitive with \textit{statistical emission} from PLF

<table>
<thead>
<tr>
<th>REGION</th>
<th>40 MeV/nucleon</th>
<th>62 MeV/nucleon</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$P_{IMF}^{TLF}$</td>
<td>$P_{IMF}^{IVS}$</td>
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<tr>
<td>peripheral</td>
<td>0.41±0.12</td>
<td>0.44±0.10</td>
</tr>
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<td>midperipheral</td>
<td>0.30±0.08</td>
<td>0.50±0.06</td>
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<tr>
<td>midcentral</td>
<td>0.27±0.08</td>
<td>0.50±0.05</td>
</tr>
<tr>
<td>central</td>
<td>0.26±0.08</td>
<td>0.49±0.05</td>
</tr>
</tbody>
</table>

TABLE IV: Probabilities of IMF emission from different sources in selected reaction regions (see Fig. 16) for $^{136}Xe + ^{209}Bi$ reactions at E/A= 40 and 62 MeV, respectively.
and TLF, for central collisions, where one observes an increase of fragment emission from IVS (see Fig. 16).

<table>
<thead>
<tr>
<th>CHARGE</th>
<th>COLLISIONS</th>
<th>28 MeV/nucleon</th>
<th>40 MeV/nucleon</th>
<th>62 MeV/nucleon</th>
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<tbody>
<tr>
<td></td>
<td></td>
<td>$T_{PLF}$</td>
<td>$T_{IVS}$</td>
<td>$T_{PLF}$</td>
</tr>
<tr>
<td>Z=1</td>
<td>PERIPHERAL</td>
<td>4.8</td>
<td>7.8</td>
<td>5.6</td>
</tr>
<tr>
<td></td>
<td>CENTRAL</td>
<td>5.6</td>
<td>8.6</td>
<td>6.7</td>
</tr>
<tr>
<td>Z=2</td>
<td>PERIPHERAL</td>
<td>5.0</td>
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<td>5.5</td>
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<tr>
<td></td>
<td>CENTRAL</td>
<td>5.8</td>
<td>14.4</td>
<td>6.7</td>
</tr>
<tr>
<td>Z=3</td>
<td>PERIPHERAL</td>
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<td>20.0</td>
<td>9.1</td>
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<tr>
<td></td>
<td>CENTRAL</td>
<td>9.1</td>
<td>26.4</td>
<td>10.9</td>
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</tbody>
</table>

TABLE V: Apparent temperatures in $MeV$ for PLF and IVS sources, respectively. Temperatures were extracted from slopes of energy distributions of protons ($Z=1$), alphas ($Z=2$) and Li ($Z=3$) fragments, for $^{136}Xe + ^{209}Bi$ reactions at $E/A = 28, 40$ and $62$ MeV, respectively.

The energy spectra are commonly used for evaluation of source temperatures. Figure 18 presents the apparent source temperatures for PLF and IVS sources, for peripheral and central collisions. Selection of sources is the same as in Fig. 17. The temperature parameter, $T_s$, was extracted from slopes of energy spectra assuming Maxwellian-Bolzmann distribution

$$P(E)dE \propto \sqrt{E} \exp \left( -\frac{E}{T_s} \right) dE,$$

where $E$ is the energy of specified particle. The values of apparent temperatures obtained from fitting energy spectra with formula 3 are specified in Table V. As one can see, the apparent source temperatures are much higher for clusters ($\alpha$-particles and IMFs) emitted from the IVS source, as compared to the PLF source. In fact, the IVS clusters apparent temperatures are much too high for statistical emission. Such a “hard” (high temperature) spectra can be explained only assuming dynamical emission from the IVS source. While the temperatures of protons and $\alpha$-particles (LCPs) emitted from the PLF are the same, the temperature of IMFs (here $Z=3$) is much higher. This effect can be explained by admixture of IMFs from the IVS source. It would suggest a prevail of
dynamical IMF production. One can notice a small increase of source temperatures with increase of bombarding energy, except for the IVS temperatures for 28 MeV/nucleon. As was already mentioned, the relative PLF-TLF velocity for 28 MeV/nucleon is too small for good separation of the IVS zone from the PLF and TLF emission regions (see Figs. ??, and ??).

VII. SUMMARY AND CONCLUSIONS

The observed correlations between energies and deflection angles of projectile-like fragments reveal that, for intermediate energies as high as E/A=62 MeV, the general reaction scenario is similar to that observed for low energies and understood in terms of dissipative binary collisions. Thus, at E/A=62 MeV, the reaction cross section appears still dominated by dissipative binary reactions involving the survival of well-defined projectile- and target-like fragments. The emission from the PLF and TLF sources observed in joint distribution of neutron and LCP multiplicities shows a thermal scaling with bombarding energy, indicating statistical emission from equilibrated sources.

On the other hand, the Galilei-invariant velocity plots for alpha particles and intermediate-mass fragments show clearly the existence of a third intermediate-velocity source in addition to the PLF and TLF sources. In contrast to the dominantly statistical emission from the PLF and TLF, fragments emitted from the IVS are likely to be produced dynamically in the overlap zone of the projectile and target nuclei. For central collisions and high IMF multiplicities, the IVS component becomes dominant, representing the overlap region of PLF and TLF. Although, for peripheral collision still one observes non-negligible emission rate, with clear IVS component.

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FIG. 1: Logarithmic contour plot of measured joint distribution of neutron ($m_n$) and prompt light output for $^{136}\text{Xe} + ^{209}\text{Bi}$ reaction at E/A=40 MeV.
FIG. 2: SuperBall average efficiency of detecting single neutron having energy $E_n$.

$^{136}\text{Xe} + ^{209}\text{Bi}$ reactions at $E/A = 28, 40, \text{ and } 62 \text{ MeV}$
FIG. 3: Average detection thresholds for DwarfWall (solid line) and DwarfBall (broken line) detector arrays.
FIG. 4: Logarithmic contour plot of joint distribution of neutron ($m_n$) and light-charged particle ($m_{LCP}$) for $^{136}$Xe + $^{209}$Bi reactions at E/A=62 MeV. Symbols represent centroids of distributions for 28, 40, and 62 MeV/nucleon, and model calculations, as indicated by labels. All distributions were corrected for detection efficiency. See Table I.
FIG. 5: Logarithmic contour plot of joint distribution of neutron ($m_n$) and light-charged particle ($m_{LCP}$) for $^{136}$Xe + $^{209}$Bi reactions at $E/A = 28, 40$ and $62$ MeV, respectively. Bins in the associated multiplicities of detected intermediate-mass fragments are shown. All distributions were corrected for detection efficiency. See Table I.
FIG. 6: Logarithmic contour plot of average atomic number of IMFs, $\langle Z_{IMF}\rangle$, as a function of associated neutron and LCP multiplicities as observed in $^{136}$Xe + $^{209}$Bi reactions at E/A = 28, 40, and 62 MeV, respectively. Neutron and LCP distributions were corrected for detection efficiency. See Table I.
FIG. 7: Logarithmic contour plots of the PLF yield in an atomic number, $Z_{PLF}$, vs. energy, $E_{PLF}$ representation, for $^{136}\text{Xe} + ^{209}\text{Bi}$ reactions at E/A=28, 40, and 62 MeV, respectively. Calculations by CLAT+GEMINI codes for E/A=40 MeV are indicated by rectangles.
FIG. 8: Logarithmic contour plot of average atomic number of detected PLF charge, \( \langle Z_{PLF} \rangle \), as a function of associated neutron and LCP multiplicities as observed in \(^{136}\text{Xe} + ^{209}\text{Bi} \) reactions at \( E/A = 28, 40 \), and 62 MeV, respectively. Neutron and LCP distributions were corrected for detection efficiency. See Table I.
FIG. 9: Logarithmic contour plots of the PLF yield in a “deflection-function” representation of the fragment energy, $E_{\text{plf}}$, vs. the fragment emission angle, $\theta_{\text{plf}}$, for $^{136}\text{Xe} + ^{209}\text{Bi}$ reactions at $E/A=28$ and 62 MeV, respectively. Middle and right panels present $E_{\text{plf}}$ - $\theta_{\text{plf}}$ correlations as gated with different multiplicities of detected intermediate-mass fragments $M_{\text{imf}}$. Bottom panel illustrates schematically four classes of collision trajectories (see text).
FIG. 10: Comparison of logarithmic contour plots of the PLF yield in a “deflection-function” representation with the results of model calculations, using the stochastic nucleon exchange model CLAT and the equilibrium-statistical decay model GEMINI.
FIG. 11: Experimental and model (CLAT+GEMINI) dependence of Eq. 3 parameters as a function of associated PLF charge, $Z_{PLF}$, for $^{136}Xe + ^{209}Bi$ reactions at $E/A = 28, 40$ and $62$ MeV, respectively.
FIG. 12: Left column - Logarithmic contour plot of average total excitation energy per nucleon, $<E^*/A>$, as a function of associated neutron and LCP multiplicities for $^{136}$Xe + $^{209}$Bi reactions at $E/A = 28, 40$ and $62$ MeV, respectively. Right column - Distributions of total excitation energy per nucleon for different bins indicated in the left column. Elastic events excluded.
FIG. 13: Ratio of TLF excitation energy per nucleon, $\varepsilon_{\text{TLF}}^*$, to PLF excitation energy per nucleon, $\varepsilon_{\text{PLF}}^*$, for given bins of the total excitation energy per nucleon, $<E^*/A>$, for $^{136}\text{Xe} + ^{209}\text{Bi}$ reactions at $E/A = 28, 40$ and $62$ MeV, respectively.
FIG. 14: Charge distributions for $^{136}$Xe + $^{209}$Bi reactions at E/A = 28, 40, and 62 MeV. Top left picture shows inclusive spectra. Rest of pictures show charge distributions for bins in the associated total excitation energy (see Fig.12).
FIG. 15: IMF charge distributions for $^{136}$Xe + $^{209}$Bi reactions at E/A = 28, 40 and 62 MeV (blow-up of Fig.14), respectively, for bins in the associated total excitation energy (see Fig.12).
FIG. 16: Logarithmic contour plots of Galilei-invariant velocity distributions of $\alpha$-particles, for different bins in the associated total excitation energy (see Fig.12).
FIG. 17: Logarithmic contour plots of Galilei-invariant velocity distributions of intermediate-mass fragments, for different bins in the associated total excitation energy (see Fig.12).
FIG. 18: Multiplicity distributions of IMFs (bottom panels) associated with different decay sources as determined by the source selection criteria depicted by solid lines in the associated Galilei-invariant velocity distributions of the top panels, and "gated" by different bins in the associated total excitation energy (see Fig.12).
FIG. 19: Energy distributions of α particles and Li fragments in the emission source velocity frame of PLF, TLF, and reaction center-of-mass (IVS), respectively (see text). Distributions were gated by different bins in the associated total excitation energy: PERIPHERAL (BINS 1 and 2), and CENTRAL (BIN 4) - see Fig.12.
FIG. 20: Apparent temperatures for $^{136}$Xe + $^{209}$Bi reactions at E/A = 28 (green), 40 (red) and 62 MeV (black), respectively. Temperature parameters were extracted from slopes of energy distribution of protons ($Z = 1$), alphas ($Z = 2$) and Li ($Z = 3$) fragments in the emission source velocity frame of PLF (circles) and reaction center-of-mass (IVS)- triangles, respectively (see text). Events were selected for different bins in the associated excitation energy: PERIPHERAL (BINS 1 and 2), and CENTRAL (BIN 4) - see Fig. 12.
Data Analysis for CHIMERA Detector: CECIL Experiment

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Abstract

In order to test a proposed theoretical mechanism for statistical multi-fragmentation, the CECIL experiment has been undertaken using the CHIMERA detector at LNS in Catania, Sicily. The properties of the detector and electronics set-up allow for the detection of a wide range of reaction products. In recent years, the field of multi-fragmentation has come to rely increasingly on complex $4\pi$ detectors that provide a great amount of information but require a lengthy analysis process. Before the copious amounts of data that were produced can be used to study the physics of the reaction, the individual detectors must be calibrated and the recorded events must be identified. To aid in this effort, software is being developed for the purpose of automating the necessary tasks as much as possible. The resulting programs should be useful for other researchers in the process of analyzing data from CHIMERA.

1 Introduction

In recent years, the field of intermediate-energy heavy-ion reactions has devoted significant time to the study of the emission of Intermediate Mass Fragments (IMF’s) [1, 2, 3]. Scenarios for IMF emission have been posited for both dynamical emission and statistical emission from an equilibrated system. In particular, statistical emission has proven difficult to model by means of standard evaporation codes such as GEMINI or PACE [4, 5]. Theoretical approaches such as the Expanding Emitting Source Model (EESM) [20], the Microcanonical Metropolis Monte Carlo (MMMC) model [21], and the Berkeley approach (BM) [22] have been proposed in order to improve the simulation of cluster emission. However, the physical justifications for some of the assumptions made by these models have come into question [6]. A promising alternative has recently been proposed, which suggests that increased emission rates could be accounted for by surface effects and the expansion of the excited nucleus [7, 8, 9]. In order to test the hypotheses pro-
posed by this alternative model, a collaboration was established between the Nuclear Physics group at the University of Rochester and the CHIMERA collaboration based at Laboratorio Nazionale del Sud(LNS) in Catania, Italy. The first in a series of experiments in the CECIL project was conducted using the CHIMERA 4π multi-detector. The study of multi-fragmentation has come to rely on data from 4π detectors in recent years, and CHIMERA is ideally suited to meet the needs of the CECIL experiments due to the device’s ability to detect a wide range of fragments, from Light Charged Particles(LCP’s) to Projectile and Target-Like Fragments(PLF’s and TLF’s).

When a reaction product is collected by the device, information is recorded that is sufficient to determine the particle’s direction, identity, and energy. However, this information is contained in the raw electrical signals produced by the detectors and processed by the CHIMERA electronics system. Before the collected data can be effectively analyzed and used to reconstruct the nuclear reactions of interest, the device must be properly calibrated and the data points that were recorded must be identified as particles of a known species and energy. The task of calibrating and analyzing the data from the 1192 CHIMERA detectors requires the careful development of methods and tools to handle the large volume of data in an efficient manner. To this end, efforts have been made to adapt and extend the techniques already used by the CHIMERA collaboration to improve the efficiency of the data analysis process.

The current work will first present a description of the detector array, followed by a detailed account of the chains of electronics modules used to process and record the analog signals from the individual detectors as digital information. This will be followed by a discussion of the opening stages of the analysis procedure and the efforts that have been made to streamline and improve the efficiency of the current methods of calibration. The focus throughout will be on the conversion of the information about a detected particle’s direction, identity and energy into raw digital data and the subsequent extraction of useful, calibrated information from that data.

2 Detector Specifications

The current section is devoted to a description of the structure of CHIMERA and the composition of the individual detectors that make up the array. Special attention will be payed to the methods used to not only measure an incident particle’s kinetic energy, but also determine its charge and mass.

---

2 The Rochester group has recently made the decision to halt use of the FORTRAN-based PAW data analysis platform and switch to ROOT, a data analysis environment based on C++ and developed by CERN [10]. ROOT utilizes Object Oriented programming and shows great promise for the development of software to automate the analysis of large amounts of data. All scripts and programs currently in development by this research group for the analysis of CHIMERA data were written in C++ for use with the ROOT environment.
2.1 Array Description

The CHIMERA multi-detector (pictured in Figure 1) is an array of 1192 detectors placed around the target in two main structures: the wheels, which detect reaction products in the most forward angles ($1^\circ – 30^\circ$); and the sphere, which covers most of the remainder of the $4\pi$ solid angle ($30^\circ – 176^\circ$) [12].

![Figure 1: A schematic of the CHIMERA multi-detector [12]. The beam enters the detector from the upper right of the figure and strikes a target film placed at the center of the sphere. When fully assembled, the structure is about 4m in length and is enclosed within the CICLOPE vacuum chamber at LNS.](image)

The individual detectors, or “telescopes”, consist of a thin (250$\mu$m) Si detector coupled to a CsI scintillation crystal. A detailed description of the telescopes is found in Section 2.2.

Most reaction products are focused forward due to the reaction kinematics. For this reason, the forward wheels have a high detector granularity, which gives high angular resolution at forward angles. The sphere has lower resolution, but surrounds the target in order to detect particles in the full solid angle and allow full reconstruction of the reaction. The large size of the detector, which is over 4m long when fully assembled, creates a long flight path for particles leaving the target film at the center of the sphere. Coupled with the time resolution of the Si detectors, the detector size allows the identification of particle mass based on the Time-Of-Flight technique [12].

The large number of telescopes provides excellent angular resolution and covers 94% of the $4\pi$ scattering angle. This allows measurement of a large number of emitted particles and provides good event reconstruction, as well as information on the angular distribution of the reaction products. However, while good detection coverage is provided by the geometry of the array, the quality of measurements about each fragment’s mass, charge and energy is determined by the properties of individual telescopes. The properties of the CHIMERA detectors are discussed in detail in the following section.
2.2 Telescope Description and Physics

A particle detector should ideally be capable of not only measuring the kinetic energy carried by an incident particle, but allowing for the identification of its charge and mass. Such a full identification is not always possible, particularly in regards to mass, but the detectors that make up CHIMERA are capable of identifying a large range of particle charge, from light charged particles to heavy ions, over a wide energy range [12]. There are studies underway to determine if at least some information about neutrons produced in the reaction may also be extracted, though at this time the detector is sensitive only to charged particles [14]. When an incident particle interacts with the matter in the detector, some or all of its energy is deposited in the detector material. That energy is converted into an electrical pulse which is amplified and recorded by the electronics and Data Acquisition system (DAQ) associated with the detector. The height and shape of the pulse depend on the energy, charge and mass of the reaction fragment. The signal can then be recorded and analyzed to recover the necessary information about the original particle. This section will give a description of a typical telescope that makes up CHIMERA and the mechanism by which it produces a useful signal.

The exact dimensions of a particular telescope depend on its location within the array, but the overall structure of the detectors is displayed in Figure 2.

A nuclear fragment that impinges on the thin (≈ 250μm) Si detector will deposit at least some of its kinetic energy in the semiconductor material. The ionization process by which the particle primarily interacts with the detector is described by the Bethe-Bloch formula. A voltage bias is applied to the detector in order to collect the particle-hole pairs created within the Si and to produce an electronic signal. The electrical pulse produced in this manner is proportional to the energy deposited by the particle and depends on the charge — and to a lesser extent, the mass — of the incident particle. The signal is subsequently processed and recorded by the DAQ system as described in Section 3.1. The electrical response of the Si detector is sufficiently fast to determine the time of arrival of the incident particle, which is also analyzed and recorded by the DAQ. The time information, together with the known flight path from target foil to detector and the energy deposited in the detector, determines the velocity of the particle; if it is stopped in
the Si layer, the total energy measurement can be used to identify the mass of the incident particle via the Time-of-Flight (TOF) technique.

A particle that is not stopped in the Si layer will deposit its remaining energy in the scintillation crystal, where light is produced in the process of stopping. The light output of the crystal depends on the energy deposited by the original particle, but that dependence is not a simple proportionality. The nonlinear response of the crystal must be compensated for in the analysis phase. The scintillation light is collected by a photodiode with a $1.8 \times 1.8 \text{ cm}^2$ active area (compared with the approximately $5 \times 5 \text{ cm}^2$ area of the crystal at its base) and converted to an electrical signal which is subsequently analyzed and recorded by the DAQ. The pulse may be analyzed using the Pulse Shape Discrimination (PSD) technique, which relies on the internal structure of the light pulse to identify the incident particle. This process will be described in detail in Section 3.2.

After the particle has been fully stopped in the detector, the information about its original energy and identity is stored in the electrical pulses that are amplified, processed and recorded by a system of electronics modules. This system will be discussed in the following section, which will describe the treatment of the detector signals leading to digital data that is recorded for future analysis.

3 Electronics

The previous section was devoted to the conversion of the information about a given particle’s charge, mass and kinetic energy into electrical pulses by the two components of the CHIMERA telescopes. This section will cover the processing and conversion of these analog signals to digital data that are recorded in the experimental data set.

3.1 Si Electronic Chain

Figure 3 displays a schematic view of the electronics chain used for the processing of signals from the CHIMERA Si detectors [15]. The small analog pulse from the Si detector is first boosted by a pre-amplifier mounted on the array structure. In addition to pulses from real events, the pre-amplifier also receives periodic pulses from an automatic pulse generator (pulser) outside the vacuum chamber. These pulses, which range from $10 \text{ mV}$ to $1000\text{'s of mV}$ in regular intervals, are used to calibrate the digitized data in the analysis phase; this process will be described in more detail in Section 4.1. Data collected from the pulser are tagged and may be removed from the data set when no longer required for calibration purposes.

The pre-amplifier output is a negative, mono-polar signal with a pulse height ranging
from a few mV to a few V. An example pulse, pictured in Figure 4, has a rise time of 30 - 200ns and a 200\(\mu\)s decay time. Raw signals from the pre-amplifiers are passed outside the vacuum chamber to banks of electronics modules for further processing, beginning with an amplifier. Two signals are produced by the amplifier: an analog energy signal integrated by a QDC and recorded for use in TOF and \(\Delta E/E\) analysis; and an analog fast timing signal which is processed by a discriminator to obtain a logical timing signal giving the arrival time of the incident particle in the detector.

The logic signal is sent to both the ‘stop’ input of a TDC to be used for TOF measurements (the ‘start’ is provided by the cyclotron RF frequency) and the MUSE trigger system to register the detection of a particle by the telescope. It also serves the start signal for the gate generator used by the QDC.

In order to produce the analog energy signal from the raw pre-amplifier signal, a complicated series of shaping operations are first applied to the raw pre-amplifier signal by the amplifier. The primary effects are the differentiation and integration of the pulse into a negative front bipolar pulse, shown in Figure 5. The positive component of the pulse — shown in a zoomed view in Figure 6(a) — is small compared to the full pulse height, but must still be cut as shown in Figure 6(b). Some pulses produced by the pre-amplifier have significant pulse-heights on the order of a few V. Such pulses are due mostly to high-energy elastic scattering at forward angles, and are only detected by the most forward wheels. These pulses require no amplification, so the appropriately shaped signals from rings 1 – 4 are simply passed to the remainder of the electronics chain with a gain.
Figure 4: A typical output pulse from the pre-amplifier of a CHIMERA Si detector. The pulse height is displayed in arbitrary units on the abscissa, while time is plotted in $\mu s$ on the ordinate. The pulse is a negative, mono-polar signal with a representative rise time of 50 ns and a 200 $\mu s$ decay time.

Figure 5: A typical pulse from the amplifier following the differentiation of the input signal from Figure 4. The pulse height is displayed in arbitrary units on the abscissa, while time is plotted in $\mu s$ on the ordinate. The pulse is of the order of a few $\mu s$ in duration.
Figure 6: In (a), the pulse from Figure 5 is displayed in a zoomed scale. It shows that the pulse is actually a negative front bipolar pulse with a positive component. In (b), the same pulse is shown following a cut of the positive component. The pulse height is displayed in arbitrary units on the abscissa, while time is plotted in µs on the ordinate. The pulse is of the order of a few µs in duration.
factor of 1. Gain factors of 2 and 4 are applied to rings 5 – 11 and 12 – 24 respectively because particles impinging on these rings have reduced kinetic energies due to the reaction kinematics.

As shown in Figure 7, the energy signal is integrated by a QDC within a time gate generated using the logical timing pulse and set to encompass the full analog energy pulse. Before the resulting analog charge value can be digitized, the range of the QDC

![Figure 7: An illustration of the integration of the Si pulse by the QDC using a time gate of about 5μs generated using the logical timing pulse. The pulse height is displayed in arbitrary units on the abscissa, while time is plotted in μs on the ordinate. The pulse is of the order of a few μs in duration.](image)

must be adjusted. The default range setting on the 12 bit (4096 channel) QDC accepts the full range of pulses produced by the amplifier: 0 – 8V. However, some of the pulses come from low energy particles and, if digitized with the default settings, would only be analyzed at low resolution by a relatively small number of channels. For this reason, the QDC’s used in CHIMERA are “Two-Range” QDC’s [16]. If the integrated charge is less than about 1/8 of the default 4096 channel range of the QDC(≤channel 500) the QDC range is reduced by a gain factor of about 8.35. This amounts to a linear re-scaling of the range in order to improve the resolution for low energy pulses. The resulting digital data is marked as “High Gain”(HG) and recorded by the CHIMERA DAQ system. If the integrated charge exceeds this threshold, it is digitized at the default gain setting, marked as “Low Gain”(LG) and also recorded by the CHIMERA DAQ system.

At this point the electrical pulse from the Si detector, with information about the particle’s energy and identity, has been converted to digital data by the QDC.
### 3.2 CsI Electronic Chain

As discussed in Section 2.2, the CsI crystal produces a pulse of light in response to an impinging particle. The CsI light output signal, which is pictured in Figure 8, has a rapid rise time with an exponential tail. However, the tail decays with two time constants, indicating a mixture of two signals: the “fast” and “slow” components. The amplitude of the pulse can be described by the following equation [17]:

\[
A = \text{Constant} \left[ 1 - \exp\left( -\frac{t}{\tau_0} \right) \right] \left[ \frac{1}{\tau_1} \exp\left( -\frac{t}{\tau_1} \right) + \frac{r}{\tau_2} \exp\left( -\frac{t}{\tau_2} \right) \right]
\]  

(1)

The time constant \( \tau_0 \) defines the rise time of the pulse, while \( \tau_1 \) and \( \tau_2 \) correspond to the fall times for the fast and slow components respectively. The parameter ‘r’ is the ratio between the slow and fast components\(^3\). The relative strength of these two components is nonlinear, and depends on the charge and mass of the impinging particle. The technique of separating the two components in order to retain the information about the particle identity is called Pulse Shape Discrimination (PSD). To implement this method, the pulse is integrated twice by the CHIMERA electronics chain; each time in such a way as to emphasize either the fast or slow components.

Figure 9 displays a schematic view of the electronics chain used for the processing of

\(^3\)The values chosen for Figure 8 are .28\(\mu s\), .54\(\mu s\), 2.0\(\mu s\) and .29 for \( \tau_0 \), \( \tau_1 \), \( \tau_2 \) and \( 'r' \) respectively [17].

Figure 8: A typical output pulse from a CHIMERA CsI detector. Time is represented on the ordinate axis in \( \mu s \), with pulse amplitude on the abscissa in arbitrary units on a log scale.
signals from the CHIMERA CsI detectors [15]. The initial set of components in the CsI electronics chain are similar to those in the Si sequence. However, the amplifier has two energy outputs. One signal, from which the Slow signal is obtained, is multiplied by a gain factor of about 10 and passed to a Two-Range QDC; the other signal is first passed through a stretcher which extends the original pulse in time at the maximum pulse height. The output of the stretcher is, in turn, fed into a separate Two-Range QDC.

The time output can be used in the trigger system to register the detection of a particle in the crystal. This is an optional setting, and was not used in the CECIL experiments. The logical signal produced by the trigger is also used to generate the gates employed by the QDC’s.

The pulse that is output by the pre-amplifier is subjected to similar shaping operations as the Si signals discussed in Section 3.1. The result is a pulse for which the pulse height is closely tied to the decay of the fast signal, and the decay tail to the decay of the slow signal. For this reason, two energy pulses are output by the amplifier and analyzed separately by two QDC’s. The Slow component is isolated by directly integrating the pulse as shown in Figure 8(a). Using a time gate generated in reference to the cyclotron RF signal, a portion of the decay tail of the pulse is integrated which corresponds to the Slow component of the CsI signal. Because the pulse has decayed significantly before the onset of the gate, the signal is multiplied by a factor of about 10 by the amplifier in order to utilize the full range of the QDC. In Figure 8(b) the Fast component is found by first
passing the signal through a stretcher, which draws the pulse out in time at the level of its maximum pulse height. This ensures that the same time gate will effectively integrate a portion of the pulse primarily depending on the Fast component of the CsI signal. Both branches are digitized by separate Two-Range QDC’s (identical to the modules used in the Si electronics chain) and the values are recorded by the DAQ. At this point the electrical pulse from the photodiode of the CsI detector, with information about the particle’s energy and identity, has been converted to digital data by the QDC’s.

This section has chronicled the processing and digitization of raw electrical signals from CHIMERA telescopes. The final data stream was recorded by the DAQ system on tape and subsequently backed up on DVD in order to ease analysis. The data must now be properly calibrated and analyzed to reconstruct the reaction events and extract interesting physical information. The calibration and data analysis procedure will be discussed in the next section.

4 Data Analysis

The raw data produced by CHIMERA for the CECIL experiments totaled over 300 GB on over 70 DVD’s. Before this information can be rendered into useful physical information it must be calibrated and information about the reaction products must be extracted. This section is devoted to the calibration process that will be used to deduce the energy and identity of the particles that were detected in the course of the CECIL experiments.

4.1 Pulser Analysis

As discussed in Section 3.1, the QDC’s used to digitize the signals produced by CHIMERA do so at two separate range settings: HG and LG. The HG and LG settings differ only by a linear re-scaling depending on two parameters: a gain factor and a small offset value. The relationship between the channel numbers for HG ($X_{HG}$) and LG ($X_{LG}$) graphs is:

$$X_{HG} = X_{LG} \times gain + offset$$

(2)

In order to generate a unified data set, the linear re-scaling operation applied to the QDC range must be identified and corrected for. The required parameters can be found by analyzing the data points produced by the pulser during the course of the experiment.

Twice during the experiment, data was taken for a brief period while the beam was not present. This ensured that the only counts accumulated during this time were due
Figure 10: The pulse from Figure 8 is shown in (a) and (b) following processing by the pre-amplifier and amplifier in the CHIMERA electronics chain. Note that the rise time of the pulse is now on the order of about 2-3\(\mu\)s due to the shaping time of the pre-amplifier. Time is represented on the ordinate axis in \(\mu\)s, with pulse amplitude on the abscissa in arbitrary units on a log scale. In (a) the pulse is integrated directly by a QDC within the shaded ‘Gate’ region, where the Slow component is dominant. In (b) the pulse has first passed through a time stretcher which maintains the signal at the maximum pulse height; as a result, the same time gate integrates a region of the pulse where the Fast component is dominant. In each case the gate is about 2\(\mu\)s in duration and begins about 9\(\mu\)s after the onset of the pulse.
to the pulser and electronic noise. The pulser was also set to run during the experiment as a check against electronic drift, but at a much lower rate and only at two voltage levels. Typical spectra generated by the pulser are displayed in Figure 11 for both HG(a) and LG(b). Each peak represents a pulse with a fixed pulse height that was transmitted sequentially to the pre-amplifiers of all detectors. Pulses at 100 and 1000 mV were produced about three times as often as the rest, and so the corresponding peaks contain more counts. This was done in order to simplify the correct identification of the peaks.

The task of identifying the pulser peaks typically requires the use of several programs created for this purpose by the CHIMERA collaboration. While serviceable, the procedure is inherently tedious and a more efficient means of performing the analysis would be appreciated not only by those associated with the analysis of CECIL data, but potentially by other groups involved in calibrating data from CHIMERA experiments. To this end, a program has been written making use of the ROOT data analysis package that will perform the tasks of the proprietary CHIMERA software.

The “Pillbug” program makes use of a package of tools contained in ROOT for the identification of peaks in 1D spectra. The results of an attempted search are evident in Figures 11(a) and (b), where small, red triangles have been automatically placed to mark the calculated locations of the peaks. The order of the peaks in HG and LG are consistent between detectors. Consequently, the mV value corresponding to each peak is easily determined.

While most detectors respond well to the pulser and produce simple, easily analyzed spectra, some spectra defy quick analysis by an automatic algorithm. For these exceptions, a simple GUI interface has been developed for Pillbug that allows the user to

Figure 11: Typical spectra produced by the pulser for both HG(a) and LG(b). The number of counts is given on the abscissa while the QDC channel number is on the ordinate. Each peak in the spectrum corresponds to signals with the pulse height indicated. The red triangles were placed by the automatic peak-finding subroutine and indicate the mean value of each correctly-identified peak. The 100 mV and 1000 mV peaks contain an increased number of counts because the pulser was set to produce pulses at these voltages at a higher rate than pulses of other voltages.
quickly change parameters and visually check the spectra and correct for mistakenly-identified peaks and other problems that can arise. A screen shot of the Control Window is displayed in Figure 12. Graphs of the current histograms and fits are displayed in other windows and appear similar to the graphs in Figures 11 and 13.

Once the pulses are correctly identified, the voltage values may be plotted versus channel number. Linear fits, pictured in Figure 13(a), are sufficient to determine the correspondence of channel number to mV for both HG and LG. In Figure 13(a) the HG and LG pulser data are plotted separately with mV on the abscissa and channel number on the ordinate. The gain and offset are easily obtained from the fit values for the separate HG and LG lines. Using the simple relationship from Equation 2, the HG spectrum may be re-scaled and combined with the LG spectrum in a single plot, shown in Figure 13(b). The data spectra may be similarly combined using the same gain and offset parameters. This process is illustrated by the sequence of plots in Figure 14(a)-(d).

Figure 14(a) shows a typical HG Fast signal spectrum from a detector in the wheels section. Note that the spectrum covers nearly all the available 4096 channels. Figure 14(b) shows the same spectrum following a correction for the gain factor (~8.35) that was used by the Two-Range QDC when the signal was first processed (See Section 3.1). It is displayed opposite Figure 14(c), which displays the LG Fast spectrum from the same detector. The HG spectrum must also be corrected for the offset value (~100 channels), at which point it may be combined with the LG spectrum to form a unified 1D spectrum of data as shown in Figure 14(d).

The values for the gain and offset are constant for most detectors throughout the experiment. A few detectors, however, experienced significant electronic drift during the course of the experiment; these detectors must be carefully re-scaled using the pulser...
peaks contained in the data runs, or removed altogether from consideration.

The advantage of applying different range settings for low energy and high energy particles in the Two-Range QDC can now be better appreciated. While the full range of potential particle energies has been covered, the interesting features at low energies have been captured at a higher resolution.

The unified 1D spectra for Si energy, Fast, and Slow signals may be combined in 2D plots to facilitate particle identification, which will be discussed in Section 4.2.

4.2 $\Delta E/E$

The unified spectra from the Si and CsI Fast signals for a particular telescope may be combined in 2D graphs to highlight the dependence of the telescope response to the charge — and to a lesser extent, the mass — of the incident particle. Plotting the energy deposited in the Si detector ($E_1$) versus the energy deposited in the CsI stopping detector ($E_2$), will result in particles of the same species grouped into ridges on the 2D surface, as shown in Figure 15.

The ridges provide the opportunity to reconstruct both the identity and energy of the particle hits recorded in the data set. The ridges must first be defined for the computer, because although the differences in the ridges are obvious to the human observer, such distinctions represent a difficult image processing problem for a computer. In the standard CHIMERA analysis software used for $\Delta E/E$ data [19], each ridge is described by manually placing a series of points along the maximum of the ridge. This applies to both ridges defined by charge and, where distinguishable, ridges of different isotopes. For most detectors this is necessary only up to $Z=10$. The points can then be fit to a 9 parameter phenomenological function that describes the behavior of the overall $\Delta E/E$ curves for all $Z$ and $A$, including ridges not directly identified by the user.
Figure 14: This figure illustrates the procedure for matching the 1d HG and LG spectra for each detector, using the Fast HG and LG spectra for a detector in Ring 6 as an example. (a) shows a HG spectrum using the full 4096 channel range of the QDC on the HG setting, while (b) displays the same spectrum following the application of the gain factor. (c) shows the corresponding LG spectrum, with a noticeable gap in the data set from channel number 0 to 577. (d) shows the combined HG and LG spectrum, corrected for an offset value of 101 and a gain factor of 8.35.
The data points are identified by comparing the location of the point to the nearest curve defined by the previously discovered fit function. Charge identification is usually good for fragments with $Z \geq 3$. Depending on the quality of the telescope, isotope identification can be achieved up to $Z=8$.

While this method has many advantages, especially compared to the classical method of manually placing graphical cuts around those ridges that can be identified by eye, it remains time consuming. The user’s time is mostly occupied by manually clicking with the mouse on the ridges, so a suitable method of removing the necessity of this phase of the procedure would greatly improve the efficiency of the process. To this end, an effort has begun to write software to automate the identification of the ridges.

The next section will describe the particle identification software currently in production.

4.2.1 Particle ID

The method currently used for identification of the $\Delta E/E$ ridges depends on carefully selecting a small region or “chunk” of the main 2D histogram containing a small part of a single ridge. The chunk is analyzed to determine the maximum of the ridge at the location of the chunk and the process is repeated.

This approach\(^4\) is depicted in Figure 16. A starting point is first selected along a given ridge. A box-like “chunk” of the histogram is selected just to the left of the starting point, and the histogram in this region is projected along the $y$-axis. The result should be a

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\(^4\)The basic method was originally suggested by Prof. Harvey Rhody, a collaborator at the Center for Electronic Imaging Systems at the Rochester Institute of Technology.
peak that is approximately Gaussian in shape, and the mean and standard deviation of the peak can be easily determined. The mean of the peak corresponds to the $y$-value of the point defining the ridge, while the $x$-value of this point is chosen to be the left edge of the selected chunk region. This approach is used in series, tracking along each ridge independently.

The algorithm works, in principle: the results of tracking a few well-defined ridges with little curvature are displayed in Figure 17. However, a fundamental problem must be addressed before the algorithm can be generally useful. The size and shape of the histogram chunk must be dynamically adjusted, due to both the curvature of the ridges
and the fact that the ridges have some natural variation in width.

The chunk cannot be set to be arbitrarily large as it will encroach on neighboring ridges in the histogram, which will distort the analysis of the chunk and the algorithm will fail. Conversely, if the chunk is too small, it may not include the full ridge which will also lead to an incorrect calculation. A simple solution to the width problem is to use the standard deviation of the projection as a measure of the width of the ridge and dynamically adjust the size of the chunk as it is necessary.

The curvature of some ridges is such that it can distort the projection of the chunk and result in an incorrect calculation for the mean, as shown in Figure 18. If the ridge curvature within the chunk is significant, the projection will be distorted and the mean will not correspond to the maximum of the ridge. This problem could potentially be solved by choosing a chunk region with a width sufficiently small such that the ridge curvature within the chunk boundaries is negligible. Unfortunately, such an approach would also reduce the number of data points within the chunk. If the curvature is strong, the chunk may be too small to accumulate meaningful statistics.

A promising alternative is to adjust the shape of the chunk region to approximate the curvature of the ridge as shown in Figure 19. The chunk region is shifted linearly to approximate the curvature of the ridge. This operation amounts to a linearization of the curve and the resulting projection should be approximately Gaussian.

The results of an algorithm making use of this alternative are shown in Figure 20. The slope of the chunk region must be determined by the slope of the ridge at the location where the chunk is placed. This is found by performing a fit of the points already determined to a phenomenological function\(^5\). The fit is then projected to the chunk region.

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\(^5\)The function currently used in the program was developed by W. Gawlikowicz for the purpose of calculating the energy loss in target films employed by the CHIMERA detector [23], which is a similar, but not identical system. The development plan for this project calls for a switch to the function used by Le Neindre, et al. [19], which has been developed specifically for CHIMERA \(\Delta E/E\)
Figure 19: In this version of the scenario presented in Figure 16, the chunk region has been linearly shifted to follow the curvature of the ridge. This approach ensures that the projection is approximately Gaussian and the mean correctly corresponds to the maximum of the curve.

Figure 20: Shown are results of the algorithm on a single ridge for a telescope from ring 4. Points previously set by the program are marked with stars. The colored line represents a fit by a phenomenological function. The chunk region is bordered on the top and bottom by parallel lines of stars. The newly calculated point is marked with a filled circle.
to determine the likely path the ridge will follow. The function is differentiated at the midpoint of the chunk and the result serves as the slope of the linear correction employed by the chunk.

At the termination of the ridge, the primary loop is halted and the process is repeated on a different ridge. When all discernible ridges have been identified in this manner, the function may be fit to all ridges and the charge identification is complete. The function parameters are the only outputs needed by the data processing program that will ultimately be used to read the raw data files and identify the experimental events.

It should be noted at this point that while the program by Le Neindre, et al. includes mass as well as charge identification for those elements possessing discernible mass ridges, the current program does not include such a provision. Mass identification may be included in future versions of this program, however it may not be possible to automate isotope identification in the same manner.

Some obstacles remain and the program must be made more robust before it can be considered a useful tool. The principle upon which it operates, however, is sound and should result in software that will be useful to other research groups engaged in the analysis of CHIMERA data.

Once the particle identification procedure is applied, the charge (and for some elements, the mass) of each data point can be determined. In this manner, the identity of the reaction products may be extracted from the raw data produced by CHIMERA.

4.2.2 Energy Calibration

Producing an energy distribution of detected particles requires the proper calibration of the Si detector. This is accomplished by analyzing the results of elastic scattering experiments for which the energies of the reaction products are precisely determined by the reaction kinematics. A typical Si spectrum resulting from such an experiment is shown in Figure 21. The peak in the graph at channel 100 is noise from the detector electronics. The second peak, at channel 220, is due to elastically scattered projectiles at a known energy. Since the conversion from channel number to mV is known from the pulser calibration discussed in Section 4.1, the only necessary conversion is from mV to MeV. This conversion factor is provided by the calibration spectra shown here. Calibration points from several different projectiles may be averaged to obtain a more reliable calibration, but in principle only one is required.

The energy deposited by the particle in the Si layer is obtained by applying the cone-
version factor. Using the particle identification information obtained by the method in Section 4.2.1, the energy of the incident fragment can be calculated from the energy deposited in the Si layer using standard energy loss calculations based on the Bethe-Bloch formula. In this way, the energy calibration of the detector can be used to recover the information concerning the incident energy of the particle.

Most of the analysis of the 2004 calibration runs has been analyzed by other research groups. While the results should be mostly usable, a few Si detectors were replaced between the calibration runs and the CECIL experiment runs, and other methods of calibrating these detectors must be explored.

5 Conclusion

While $4\pi$ detectors remain a standard tool for the study of intermediate-energy nuclear physics, the increasing complexity of such devices demands more sophisticated methods of analysis. As an example, the CHIMERA detector provides excellent granularity and detection of a wide range of fragments, but produces copious amounts of data which must be calibrated and analyzed in order to be useful. The data acquired in the CECIL experiment from CHIMERA contain information on the reaction products’ identity, energy, and direction; this information must be extracted by careful calibration and analysis.
The automation of the calibration/identification process, whenever possible, is essential to the timely completion of the data analysis phase of this experiment. It is hoped that the techniques and software developed by this group will be useful to the community of researchers using the CHIMERA detector and possibly the larger field of nuclear physics as well. The data, when fully analyzed, will provide insights into the nuclear reactions under scrutiny and help provide evidence to confirm or refute the various competing theories currently proposed to account for the phenomenon of multi-fragmentation.

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Light Charged Particle Production in 1.2 GeV-Proton Induced Spallation Reactions on Al - Th.¹

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Abstract

Using the 4π detector NESSI, absolute cross sections, energy spectra, and angular distributions have been measured for ¹,²H, ³,⁴He, ⁶,⁷,⁸Li and ⁷,⁹,¹⁰Be isotopes produced in 1.2 GeV proton-induced spallation reactions on 13 different target nuclei between Al and Th. Calculations with the intra-nuclear cascade code INCL2.0 and the statistical model code GEMINI give, all in all, an adequate description of the experimental proton energy spectra, cross sections of evaporated charged particles, mean excitation energies, and linear momentum transfer. From the measured energy spectra of composite particles the cross section of pre-equilibrium emission is deduced. Relative to the total production cross section this cross section amounts to 40-60% for ²H and ³He, 20-40% for ³H and 5-20% for ⁴He. The experimental data indicate that the parameters for a coalescence model for calculating the pre-equilibrium cross sections have to be readjusted as a function of target mass. Compared to the thermal excitation energy, which is deduced from the mean multiplicities of evaporated light charged particles and neutrons, the mean energy carried away by pre-equilibrium composite particles amounts to about 30%.

Key words: spallation reactions, absolute production cross sections of H, He, Li, and Be, energy spectra, angular distributions, intra nuclear cascade, statistical evaporation, pre-equilibrium emission

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1 Introduction

Medium-energy proton-induced spallation reactions present significant interest for a wide range of applications and fundamental research fields. They are at the crux of spallation neutron sources [1], transmutation of nuclear waste in accelerator driven systems [2], cosmology and astrophysics [3], cosmic ray physics [4,5], planetary and geochemical science [6], and last but not least, nuclear physics. These considerations have motivated the NESSI collaboration to investigate systematically spallation reactions induced by relativistic protons [7–12]. The motivation for this program is, in fact, twofold. First, the applications mentioned above call for an experimental validation of available spallation reaction models, before costly and ambitious engineering endeavors can rely on simulations and extrapolations based on such models. For instance, structural materials of the target station of a spallation neutron source are exposed to intense GeV proton beams and must thus withstand the internal production and retention of large amounts of hydrogen and helium. Since the amounts of the latter gas are of critical significance to the life span of a target station, reliable production estimates are needed for design and engineering processes. Second, spallation reactions are interesting to fundamental nuclear physics because proton as well as antiproton induced reactions produce nuclei at high thermal excitation energies [13,14] but with presumably little compression, deformations, or spins. These characteristics allow one to study the decay of relatively well characterized, thermally highly excited nuclei, in particular nuclear fission at high excitation energies [15,16].

To illustrate the above opportunity, thermal excitation energies of up to about 800 MeV can be obtained in 2.5-GeV proton induced reactions on Au [11]. This excitation is only 100-200 MeV lower than generated in reactions induced by a more exotic beam of 1.2-GeV antiprotons [13]. An important and interesting role in this energy balance is played by particle emission from the various stages of the intra-nuclear cascade of nucleon-nucleon collisions by which the initial projectile kinetic energy is eventually equilibrated and spread

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over the target nucleus. Branching ratios, spectra, and angular distributions of such pre-equilibrium particles signal the evolving properties of the matter of the target nucleus. As will be discussed further below, contemporary nuclear reaction theory is challenged by the task to describe these pre-equilibrium processes.

The first specific objective of the present work concerns the measurement of total cross sections for hydrogen and helium isotopes produced in 1.2-GeV proton-induced reactions on target nuclei ranging from Al to Th. Previous measurements of such cross sections have employed essentially three methods. (i) Volumetric gas measurements combined with mass spectrometry [17–26] and, in the case of tritium, with activation methods; (ii) Measurements using detector telescopes or time-of-flight methods [9,11,27–32]; and (iii) Projectile fragmentation in inverse reactions, employing magnetic spectrometers [33–35]. To some extent, these methods complement one another. Experiments with detector telescopes provide also particle energy spectra, which are useful to distinguish between evaporative (equilibrium) and pre-equilibrium particle emission.

Systematic deviations between results obtained with methods (i) and (ii) have been reported in Ref. [9] for targets in the iron/nickel region, while for lead targets the results of the two methods agree reasonably well [26]. To resolve existing discrepancies, the present work has remeasured the production cross sections for $E_p = 1.2 GeV$.

The second objective of the present study is to determine the branching ratios for pre-equilibrium (PE) composite-particle emission in reactions induced by relativistic protons of various energies in a wide range of target nuclei. According to current views, spallation reactions are seen as three-step reactions. The first step is best illustrated by an intra-nuclear cascade (INC) of successive nucleon-nucleon collisions. During this stage, a large fraction of the initial kinetic energy of the projectile is carried away by energetic cascade nucleons, including PE nucleons, which manage to escape from the target. Depending on the impact parameter, a larger or smaller fraction of the involved nucleons remain in the residual nucleus, carrying the thermalized excitation energy. In a second reaction step, almost concurrently with the INC and prior to the attainment of statistical equilibrium, composite particles are emitted from the target nucleus. Subsequently, at the third reaction step, the equilibrated residual nucleus deexcites via particle evaporation. The first and the last reaction steps are presumably relatively well understood and described in the framework of various models. However, the second step, the stage at which intermediate pre-equilibrium emission of composite particles takes place, is not well understood. This PE process is thus generally not well described by customary models.
Often pre-equilibrium, direct reaction-type complex-particle emission is treated in coalescence [11,36] or exciton [37] models. While the latter model gives a good account of PE nucleon emission, it fails to reproduce the observed branching ratios of composite PE particles. The coalescence model, on the other hand, was quite successfully employed in simulations of pre-equilibrium emission of deuterons, tritons, and $^3$He in 2.5 GeV proton induced spallation reactions on Au [11,39] and 300, 480 MeV p+Ag [38]. But it was also reported [11] that, assuming energetic nucleons from the INC stage to coalesce in momentum and phase space to form complex PE particles, reduces the theoretical PE nucleon yields predicted for energies of a few tens of MeV. This destroys partially the previously obtained agreement between model and the experimental high-energy nucleon spectra. Corresponding observations were made by Boudard et al. [40] who implemented a percolation procedure into the Liège intra nuclear cascade code INCL4 [41]. While they achieved good agreement with experimental spectra of energetic clusters, the yield of promptly emitted nucleons was underestimated above about 30 MeV.

The present work investigates systematically equilibrium (evaporative) and pre-equilibrium (PE) emission of p, d, t, $^3$He, and $^4$He, including particle energy spectra up to at least 100 MeV. This investigation was performed for target nuclei between Al and Th and an incident proton energy of 1.2 GeV. Experimental results are compared with the predictions of the intra nuclear cascade code INCL2.0 [42] modelling the prompt emission of highly energetic, i.e., pre-equilibrium, nucleons. At this stage of the calculations, the PE emission of composite particles is not considered. The output event data of the INCL2.0 Monte Carlo code for the intermediate target-like remnant, consisting of remnant mass and atomic numbers ($A_R^*,Z_R^*$), its excitation energy ($E_R^*$) and linear momentum ($p_R$) in the lab, is then used as input to the statistical model code GEMINI [43] simulating the statistical decay of the excited target remnant via the evaporation of neutrons and light charged particles (LCP) and intermediate mass fragments (IMF).

The following Section 2 is meant to describe the experiment, with special emphasis on the corrections applied in the determination of absolute cross sections and the associated uncertainties. In Section 3, experimental results are presented for total production cross sections, which are compared to both, previous measurements and model calculations. In addition, experimental results are given for pre-equilibrium emission of protons and composite particles. Section 4 presents a summary and conclusions.
2 Experimental Method and Data Analysis

The measurements discussed in this paper have been performed with the external proton beam from the Cooler Synchrotron Facility COSY of the Forschungszentrum Jülich. The experiments utilized the $4\pi$-detector NESSI (NEutron Scintillator tank and SIlicon ball) for the efficient, simultaneous detection of neutrons and charged particles. In addition to the Berlin Neutron Ball (BNB), and the Berlin Silicon Ball (BSiB), the setup used six ancillary four-element detector-telescopes and a set of plastic scintillation detectors for beam diagnostics purposes. A schematic drawing of the experimental setup is shown in Fig. 1. Detailed descriptions of the NESSI detector are given elsewhere [44,45]. In the following, those parts of the detector are described again briefly that are of special importance for the measurement of the absolute production cross sections of light charged particles (LCP) and intermediate-mass fragments.

2.1 Detectors

The outer spherical shell of the NESSI detector contains the scintillator liquid of the Berlin Neutron Ball (BNB) which measures with high efficiency the multiplicity and the total kinetic energy of neutrons evaporated from excited target remnants. The inner vacuum chamber of the BNB houses the Berlin Silicon Ball (BSiB) consisting of 151, 500-μm thick, silicon detectors. The solid angles covered by the individual BSiB detectors have been determined experimentally to $(66.9 \pm 0.8)\text{msr}$ [45]. The angular acceptance of each of these detector is about $\pm10^6$. Together, the Si detectors form the self-supporting BSiB sphere of $20 -\text{cm}$ diameter. For the present experiment, six positions in the BSiB detector array, selected at polar angles of 30°, 2×75°, 2×105°, and 150°, have been equipped with 4-element Si/CsI(Tl) detector telescopes, each covering a solid angle of about 21 msr. Each telescope consists of a stack of three $\Delta E$ Si-detectors with thicknesses of 25–27μm, 80–90μm, and approximately 1000μm, backed by a 7 – cm thick, and 4 – cm dia. CsI(Tl) crystal. The solid angles of the telescopes have been determined from a comparison to regular BSiB detectors at neighboring polar angles, using the respective He yields measured with heavy targets.

2.2 Beam Definition and Monitoring

As illustrated in Fig. 1, about 11m upstream from the target, the proton beam passes through a 0.3 – mm thick and 20 – mm dia. plastic scintillator (S1) read out by a fast photo multiplier. This fast scintillator detector provides
for a start signal for the time-of-flight (TOF) measurement (see Sect. 2.4); it is also used to count the number of incident beam protons. Additional scintillator detectors \(S_3-S_8\), installed about 100 cm upstream from the target, are employed to identify and reject off-axis beam protons. These detectors are arranged so as to form a circular "active" collimator of 20 cm inner diameter. This collimator is followed by a second, rectangular "active" collimator with an adjustable aperture of typically 10 x 10 mm\(^2\). This configuration of collimators defines an accepted proton beam converging to a focus of about 2 - 3 mm diameter at the target position in the center of BSIB. Ancillary downstream scintillators \(S_{10}-S_{14}\) are utilized to align and monitor effective beam direction and focal point.

Using an extraction RF signal from the COSY storage ring, the electronic data acquisition is inhibited during the first few seconds of extracted beam, in order to avoid overloading the electronics with excessively high instantaneous beam intensities. After this initial off-period, the event acquisition was enabled and triggered whenever a beam proton was detected by \(S_1\) in coincidence with other detector signals. Consequently, protons that did not pass through \(S_1\) do not disturb the cross section measurements, even if they induce a reaction in the target. "Off-axis" protons that traverse the \(S_1\)-detector at an angle to the beam axis large enough to miss the target, are identified by the active collimator system. The yield of such beam protons was determined from "inclusive" data, which were taken periodically during the experiment. For example, every 10\(^5\)-th proton counted by the \(S_1\)-detector was used to start the acquisition without any further condition.

The accuracy in counting the number of valid incident protons largely contributes to the error in the experimental cross section. In the present measurements, scintillation detector \(S_1\) provides the primary proton count. This detector was inhibited during the busy time of the data acquisition. The proton beam intensity admissible for a precise determination of the absolute production cross sections was limited to relatively moderate intensities of < 1.5 x 10\(^5\) pps. On the other hand, achieving meaningful statistics for LCPs and clusters (IMFs) for the 6 detector telescope requires measurements to be taken also at much higher intensities, e.g., at about 4-7 x 10\(^6\) pps. For these measurements the data acquisition was started by the detection of at least one charged particle by the BSIB or the telescopes independently of \(S_1\). This data was normalized with the help of the He yields measured with the BSIB as will be described below in Sec. 2.4.2.
2.3 Targets

Targets used in the present experiments were self-supporting metallic films of (1-10)-mg/cm\(^2\) thickness, prepared by mechanical rolling. They were glued onto 0.2-\(mm\) thin, 25 \(\times\) 27mm\(^2\) rectangular aluminum frames with a circular aperture of 20-\(mm\) diameter. The targets were positioned in the center of BSIB by means of a movable target ladder. The ladder carried up to four target frames glued to a 14-\(cm\) long Al “flag-pole” with a 0.5\(\times\)5-\(mm\) cross section. Most of the telescope data were taken with the target surface oriented at an angle of approximately 45\(^\circ\) with respect to the beam axis. The uncertainty of \(\pm 5^\circ\) in the manual angular target alignment corresponds to an uncertainty of \(\pm 8\%\) in the effective target thickness. To avoid such a systematic uncertainty, the data were ultimately normalized to runs taken with the target surface oriented perpendicular to the beam axis, where the uncertainty in effective target thickness is minimal, and at low beam intensities. This normalization procedure used the respective \(He\) yields measured with BSIB.

A precise determination of absolute cross sections depends crucially on how well the actual target thickness is known. In the present work, three independent methods have been applied to determine target thicknesses and associated uncertainties. Initially, the thicknesses of the 23 \(\times\) 24mm\(^2\) target films were determined by just weighing. In this method, only the film thicknesses averaged over the whole target surface are determined. More precise, differential information on target thickness and uniformity was obtained with a second method relying on energy loss measurements with alpha particles. In these measurements, a collimated ThC source was employed, emitting 6.06-MeV and 8.76-MeV alpha-particles in the decays of \(^{212}\)Bi and \(^{212}\)Po, respectively. The source collimator had a diameter of 4-\(mm\) defining a surface area approximating the proton beam spot on the target. The target uniformity was investigated by scanning the target surface. The thicknesses were then deduced from the measured energy losses and the stopping powers of various target materials for alpha-particles. These calculations were performed with the computer code TRIM [46]. In most cases, discrepancies between target thicknesses determined by this latter method and simple weighing were found to be smaller than 3\%. The results are listed in Table 1.

In a third method, the total neutron yields measured with BNB for the employed ”mg targets” was compared with similar measurements for much thicker (0.5-10 g/cm\(^2\)) and more uniform (2-3\%) targets. However, such a comparison can be performed with sufficient accuracy only for targets heavier than about Ag. For such heavy targets, the beam related neutron multiplicity is high enough even for mg-thin targets, such that errors due to the background contribution are tolerable. For Zr, Ag, W, Au, and Pb, the results obtained with this third method confirm, within 1.5\%, values for target thick-
nesses obtained with the above energy loss method. The \( H_0 \) target was an exception, with a discrepancy of \( 6.3 \pm 0.6\% \) between the two methods. This behavior is ascribed to the large nonuniformity of \( 6\% \) of the \( H_0 \) target.

For the determination of the cross section of the \( H_0 \) target, a \( 6\% \) increased thickness relative to the energy loss measurement is employed. For all other targets, the thicknesses derived from the energy loss measurements were used, as given in the third column of Table 1, with an estimated overall systematic error of \( 5\% \). Generally, for energetic \( He \) ions (3-9 MeV), the systematic uncertainty of the used stopping power data is found to be 3-5% \([47,48]\). This error essentially limits the precision of the absolute cross section data presented in this paper.

2.4 Particle Identification and Energy Thresholds.

This paper reports on cross sections measurements performed with both, the \( 4\pi \) Silicon Ball BSiB and the set of 6 \( Si/CsI(Tl) \) detector telescopes. The following discussion relates to methods of particle identification, various associated sources of systematic uncertainties, thresholds, as well as to corrections to the measured data. A more detailed description is given in Ref. \([45]\). Particle identification with BSiB detectors was obtained by means of energy and time-of-flight measurements; for the \( Si/CsI(Tl) \) telescopes, the \( \Delta E-E \) method provided this information. One advantage of the TOF method consists in an energy threshold of only about 2 MeV, considerably lower than those of the telescopes (4-7 MeV, see Table 2). On the other hand, identification of charge and mass with the telescopes is much superior to the TOF method.

2.4.1 Measurements with BSiB without isotopic resolution.

Due to the short TOF pathlength of 10 cm and the 500 \( \mu m \) thickness of the silicon detectors, BSiB allows one to discriminate only between hydrogen and helium particles. Additional information on the particle species was obtained by utilizing the pulse shape discrimination (PSD) of the detector signals \([45]\).

Since the TOF method measures the mass, it appears impossible to separate \( ^3He \) from \( ^3H \) at energies below the punch through energy of about 12 MeV for tritium in a 500\( \mu m \) thick Si-detector. This energy can be further reduced to about 9 MeV by utilizing the PSD signal (see Fig. 7 in Ref. \([45]\)). Furthermore, the difficulty is mitigated by the plasma delay which increases the apparent TOF of \( ^3He \) by about 1 ns compared to \( ^3H \). With the help of simulation calculations, the loss of \( ^3He \) from the He gate can be estimated. The thus obtained corrections amount to an 7.5, 5.0, 2.4\% increase of the measured helium (\( ^3He + ^4He \)) cross section for Al, Fe, and Ag targets, respectively.
Correspondingly, the measured hydrogen (p + ^2\text{H} + ^3\text{H}) production cross sections were decreased by 3.2, 1.5, and 0.8% of the measured value. These corrections are decreasing with target mass thanks to the increasing Coulomb barrier, which tends to move increasing fraction of the ^3\text{He} spectrum beyond the tritium punch-through energy.

The low Coulomb barrier of light nuclei, such as Al, results in a considerable cut-off of the hydrogen and helium yield by the lower detection thresholds. This is demonstrated in Fig. 2 where the measured angle-integrated helium energy spectra (dots) are compared with simulation calculations (gray histograms) performed using the codes INCL2.0 and GEMINI. The data show counts also below 2 MeV since the hardware thresholds of individual detectors are smaller than the off line threshold. In order to have a well defined lower threshold, an off-line cut-off at 2 MeV is employed for all detectors. The simulated data are filtered with the acceptance of the active BSiB detectors and include also the energy loss in the target. The calculated spectra have been normalized to the experimental spectra in the energy region indicated by the two vertical dashed lines. As seen in Fig. 2, considerable yield is found below 2 MeV, especially, for the lighter targets. The loss of yield is particularly important at backward detection angles due to the additional reduction of the CM energy in the lab system. The calculated yield at zero energy, as for instance for Al in Fig. 2, is due to particles which are stopped in the target. For an off-line threshold of 2 MeV, the simulation calculations result in a correction of 17% and 3% of the measured helium cross section of Al and Fe, respectively. For hydrogen, the corresponding corrections amounts to 9 and 4%. In addition to these corrections, the production cross sections as measured with BSiB were corrected for double hits, geometrical efficiencies, background as measured with empty target frames, and dead time (see Ref. [45]).

The lower energy threshold defines an upper energy cut-off for punched-through particles, which depends on the effective depletion depth of each Si-detector. Since these thicknesses varied between 440 and 500\mu m, it was necessary to set the off-line cut-off energy for punched through protons in order to have a well defined upper cut-off energy (here, 26 MeV) for protons. In Fig. 3, the energy and energy-loss spectra are displayed for stopped (H_{slow}) and punched-through (H_{fast}) hydrogen ions. Since a large spectral range of punched-through protons is compressed into a small energy loss bin, any uncertainty in the cut-off energy results in a considerable error in the derived integral yield. For hydrogen, the measured yield \sigma_H, extrapolated to zero energy, represents the sum of the cross sections of protons, deuterons, and tritons (\sigma_H = \sigma_p(0 - 26 MeV) + \sigma_d(0 - 51 MeV) + \sigma_t(0 - 77 MeV)). For helium ions, the spectrum is measured up to about 100 MeV, representing almost the full range (see also Tab. 2).

In Figs. 4, experimental angular distributions are shown for He ions, as mea-
sured with the BSib detectors with the target foil at $\Theta_T = 90^\circ$ (top panel) and $\Theta_T = 45^\circ$ (lower panel). In the case of a perpendicular orientation to the beam axis, detectors near $90 \pm 10^\circ$ are “shadowed” due to considerable energy loss in the target film. Their yield has therefore been omitted in the fit of the angular distributions with a $\cos(\theta)$ function

$$Y(\theta) = P_0 \cdot (1 + P_1 \cdot \cos(\theta) + P_2 \cdot \cos^2(\theta)). \quad (1)$$

Angle-integrated cross sections were then determined from the analytical integral of the above expression over 4$\pi$ sr, $Y_{4\pi} = 4\pi P_0 \cdot (1 + P_2/3)$.

2.4.2 Telescope isotopic-yield measurements

As mentioned previously, measurements employing the Si/CsI(Tl) detector telescopes suffered from energy thresholds that are considerably higher than those of the BSib silicon detectors used with a TOF-method. The actual lower energy thresholds and upper cutoff energies are given in Table 2 for the different measured hydrogen and helium isotopes as well as for Li and Be. In order to correct the measured yields for the corresponding low-energy thresholds, the experimental spectra were extrapolated to zero energies based on theoretical spectra calculated with the INCL2.0/GEMINI combination of codes discussed earlier. These corrections are largest for light target nuclei with low Coulomb barriers, such as Al. Here, the corrections amount to 15-50%, for H, and 29-100%, for He. In the case of Fe, the corresponding values are 14-26% and 8-31%, respectively. For Ho and heavier targets with high Coulomb barriers, the corrections are less significant and amount to only 1-2%. The corresponding corrections for Li and Be are given below in Sec. 3.5.

The mass spectra of hydrogen, helium, lithium, and beryllium isotopes are shown in Fig. 5 as obtained with the three different combinations of $\Delta E$ and E detectors: for the lowest particle energies $25 \mu\text{m}-80 \mu\text{m}$ ($\Delta E25$-E80), and for higher energies $80 \mu\text{m}-1000 \mu\text{m}$ ($\Delta E80$-E1000) as well as 1000$\mu\text{m}$-7cm ($\Delta E1000$-ECsI). The mass resolution of the last two combinations is sufficient to separate uniquely all isotopes while in the case of $\Delta E25$-E80 the separation is marginal in particular for Li and Be. The systematic error for the separation of $^6$Li, $^8$Li, $^9$Li, and $^{9,10}$Be in the mass spectrum of $\Delta E25$-E80 was estimated to be 10%, 20%, 30%, and 15%, respectively.

Additional effort was related to the contamination of $^3$He by the intensive yield of $^4$He in the mass spectra as obtained from the Si-detectors. The contribution of $^4$He in the peak range of $^3$He has been determined from the summed data of all target runs in order to achieve sufficient statistics to fit the data by two Gaussians and an additional component for the low-mass tail of $^4$He. This procedure resulted in a correction of 30-60% and 12-20% of the $^3$He yield for
the combination $\Delta E25-E80$ and $\Delta E80-E1000$, respectively. The uncertainty of this correction has been estimated to be about 50% of the reduction and constitutes the largest error contribution for the $^3$He cross sections.

The background in the mass spectrum as measured by runs with an empty target frame is indicated by the dark histograms in Fig. 5. The corrections of the data obtained with the telescope detectors $\Delta E25-E80$ and $\Delta E80-E1000$ are typically <1% for H and <0.5% for He while maximum values of about 2-3% for protons and 5-6% for deuterons and tritons are observed in the measurement with the thinnest targets Al, Fe and Cu. The background for the detection of the Li and Be isotopes is negligible.

Considerably larger background contributions are observed for energetic particles reaching the thick CsI detectors which are also sensitive to neutrons and gamma rays. As seen in Fig. 5 the empty target-frame measurement cannot describe completely the intensity below the prominent peaks and thus indicates a certain background contribution which is correlated with the reactions in the target layer. Therefore, the background was estimated as indicated by the dashed lines, resulting in values of 5-33% for protons, 7-40% for deuterons, 8-53% for tritons, 10% for $^3$He and 5% for $^4$He. In the case of the hydrogen isotopes, the largest corrections again correspond to the measurements on Al, Fe, and Cu, and the smallest to the heavy targets between Ag and Th. An uncertainty of 30% of the correction has been taken into account in the error calculation for the cross section.

As mentioned in Sec. 2.2 the telescope data was measured at high proton beam intensities between 4-7×10$^6$ pps. Since at these intensive beams the counting of the incident protons is affected by considerable dead-time losses, the telescope yields had to be normalized to get absolute cross sections. The normalization procedure was accomplished by employing the yields of He with kinetic energies between 11 and 58 MeV, i.e., the energy range where He could be identified with the BSiB detectors by exploiting the pulse-shape signal PSD instead of the time-of-flight since TOF is difficult to provide in the high-intensity measurements [45]. The normalization factor was derived from the integrated angular distributions of the BSiB data at low and at high beam intensity by the irradiation of identical targets. This procedure takes into account both the different number of incident beam protons as well as effective different target thicknesses at different orientations (45° and 90°) of the target surface with respect to the beam axis. The error of the beam normalization procedure was estimated to about 2%.

The nominal solid angle of the telescopes ($\Delta \Omega=20.97\text{msr}$) has been also verified by employing the He yield identified by E-PSD. The solid angles of the first telescope detectors 25μm-80μm, derived from the experimental He yield ratios and the precisely known solid angles of the BSiB-detectors [45], are found to
deviate only slightly from the nominal value by a few percent. However, a reduction of the solid angle was observed for the second and third detector combinations $\Delta E_{80}$-$E_{1000}$ and $\Delta E_{1000}$-ECsL. The resulting efficiency losses are furthermore increased by double hits which are - in case of the CsI-detectors - generated also by neutrons and gamma-rays. The reduced efficiencies for the detector pairs $\Delta E_{80}$-$E_{1000}$ and $\Delta E_{1000}$-ECsL have been taken into account in the data analysis with typical corrections of about 10% and 25%, respectively, and an estimated error of 30% of this correction. As shown in Fig. 4 for the reaction at W, the differential cross sections of the telescope data are finally in good agreement with the fit of the angular distribution measured by the BSiB detectors.

A linear and particle-independent energy calibration has been obtained for the Si-detectors by measurements with $^4$He sources [45] and by the analysis of the punch-through points for the different particle branches. The energy calibration of the CsI-detectors has been determined separately for p, d, t, $^3$He, and $^4$He with reference to the energy losses in the 1000 $\mu$m Si detector in front. The relation has been fitted by a polynomial function. However, the energy calibration of the CsI-detectors turns out to be not very different from linear approximations, and the differences between the various particles are pronounced only in the low-energy part. In the high-energy part of the hydrogen isotopes, where the energy losses in the 1000 $\mu$m Si detector are small and their relative errors large, the position of the punch-through points have been exploited for the energy calibration. The used punch-through energies have been calculated for the 7-cm thick CsI crystals resulting in values of 160 MeV for protons, 210 MeV for deuterons and 250 MeV for tritons.

The statistical errors of the production cross sections as measured with the telescopes amount typically to about 0.5-1.5% for p, d, and $^4$He, to 1-3% for t and $^3$He, 5-8% for $^{6,7}$Li, 10-25% for $^6$He, $^8$Li, and $^{7,9,10}$Be and to 20-50% for $^9$Li. The total errors are calculated by quadratic summation of the various uncertainties.

3 Results

3.1 Data from 4$\pi$ measurements with BSiB

Experimental results obtained in this work for hydrogen and helium production cross sections at $E_p = 1.2$ GeV are presented in Table 3, together with previously published data [9,24,26]. The present data were taken with the BSiB silicon detectors covering essentially the entire solid angle, except for domains occupied by the 6 detector telescopes.
Since the measured cross sections have been extrapolated to zero particle energy, as described in Sect. 2.4.1, the present cross section figures can be compared directly with earlier results, which were based on volumetric gas measurements which have no lower energy cut-off. In Fig. 6, the measured helium production cross sections are compared to such previous data [24,26]. As seen in this figure, while the present Pb data agree within errors bars with the earlier results [26], for Al, Fe, and Ni targets discrepancies by about 4 standard deviations exist between new and previous [24] results. This latter disagreement might be related to the fact that the earlier Al, Fe, and Ni data were obtained in measurements [24, 26] utilizing stacks of targets, in which secondary particles can induce additional reactions. This effect was avoided in more recent measurements for a Pb target [26], where a new "Mini-Stack" target configuration was employed.

In order to compare the present data with the numerical production cross sections measured previously with NESSI [9] the latter data were also extrapolated to zero particle energies (see Sect. 2.4.1). For this comparison, the data originally reported on in Ref. [9], have been re-analyzed assuming a more correct, lower (-7.7%) geometrical efficiency [45] and applying a 1-3% double-hit correction that had originally been neglected. The corrected data of Ref. [9] are included in columns 3 and 5 of Table 3 and Fig. 6. Although the corrected earlier helium data agree within error bars with the present results, all of the former data are systematically lower by 6% - 14%. Furthermore, the hydrogen data are even 20% to 31% smaller than the present data given in column 2. This additional deviation by about 15% from the present helium data can be ascribed to the lower effective thresholds for \( H_{fast} \) realized in the present experiment, as indicated in Fig. 3. In Ref. [9], all detector thicknesses were assumed, but not measured, to be uniformly equal to 500\( \mu \)m. In contrast, in the present work, these thicknesses were derived, individually for each BSIB detector, from the corresponding punch-through energies of \( \alpha \)-particles.

In Fig. 6, the total production cross sections of He (\(^3\)He + \(^4\)He) with energies between 0 and 100 MeV are furthermore compared with results of different model calculations. Theoretical results obtained with a combination of the intranuclear cascade code INCL2.0 [42] and the evaporation code GEMINI [43] are represented by the solid curve (INCL2+GEMINI). As seen in this figure, the experimental data are systematically underestimated by the model calculations. This finding can be ascribed to the additional pre-equilibrium emission, a process which is not taken into account by the INCL2/GEMINI simulation calculations. The amount of pre-equilibrium contribution to the total production cross section of about 5-20% as will be discussed below in Sec. 3.3. In Fig. 6, the data are also compared to predictions by the codes LAHET [49] and HERMES [50], which have been employed extensively in design studies of target stations for spallation neutron sources. Although these latter two codes describe reasonably well the neutron production in thick tar-
gets [10,50], they largely fail to describe the helium production, corroborating previous findings [9,51].

3.2 Results obtained based on telescope data

As mentioned in Sect. 1, it is of particular interest for application purposes to know precisely the absolute integral production cross sections. In addition, the tritium production cross sections are relevant. The isotope-resolved production cross sections of hydrogen and helium induced by 1.2 GeV protons on targets between Al and Th are given in Tab. 4. The cross sections were integrated between 0 and 100 MeV. Except for protons, it is a good approximation to identify these cross sections with the total production cross section, as seen from the energy spectra discussed further below. The summed cross sections of all three hydrogen isotopes differ considerably from the values given in Tab. 3 due to differences in the integration intervals 0-26, 0-51, and 0-77 MeV for p, d, and tritons, respectively, compared to 0-100 MeV for all isotopes in Tab. 4 while for helium, the cross sections agree within the experimental uncertainties.

Similarly to the He cross sections deduced from the BSiB data, the comparison with the volumetric gas measurements of Ref. [24] indicates that the present cross sections are about 20% smaller for both He isotopes produced in Al, Fe, and Ni targets. At the same time, for the Pb target, the present $^4\text{He}$ cross section is about 20% larger than the value measured in Ref. [26] (1113±150 mb) at a proton energy of 1.163 GeV. In this reference the $^3\text{He}$ cross section was corrected for the decay of tritium by assuming $\sigma_t/\sigma_{^3\text{He}} = 3$ while we find $\sigma_t/\sigma_{^3\text{He}} = 7.2 \pm 2.0$ for a Pb target (see Tab. 4). Using our $t/^3\text{He}$ ratio and assuming a decay time of 5 years the $^3\text{He}$ cross section would decrease from 91 ± 13 mb to 55 ± 18 mb, which would also be consistent with our value of 78 ± 13 mb.

3.3 Pre-equilibrium emission versus evaporation of LCPs

For comparison with model calculations, it is of particular interest to separate experimentally the contributions from PE emission and evaporation to the total production cross section. In this section, we discuss the decomposition of the total cross sections into evaporation and pre-equilibrium emission components based on the shapes of the measured kinetic energy spectra. As an example, the measured spectra (dots) of the various hydrogen ($^{1,2,3}\text{H}$) and helium isotopes ($^{3,4}\text{He}$) produced in reactions on the Ta target are shown in the left panel of Fig. 7 at forward angles of 30° and 75° and at one backward angle of 150°. The spectra clearly feature two components - a low-energy evaporation
peak and a high-energy tail due to particle emission prior to the attainment of thermal equilibrium. The latter high-energy tail is weaker at backward angles. The spectral shape of the low-energy evaporative component is well described by the combination of INC and evaporation code INCL2+GEMINI, as indicated by the shaded histograms.

The high-energy proton component is also reasonably well described by the INC code. A closer inspection of the proton spectra reveals, however, that the calculated yield of protons with energies between 10 and 20 MeV strongly overestimates the experimental data. This becomes more obvious in a linear plot in which we confront the calculated INC proton spectrum with the difference spectrum of the measured proton energy distribution and the calculated evaporative protons: \( d^2 \sigma_{\text{exp}}/(d\Omega dE) - d^2 \sigma_{\text{EV}}/(d\Omega dE) \). This spectrum corresponds to protons which are not evaporated and thus emitted during the INC and/or PE phase. In the right panels of Fig. 7 this experimental INC spectrum is compared with the calculated one indicating a large excess of calculated yield at proton energies smaller than 20 MeV. This finding is observed for heavy (Ta) as well as for light (Ni) targets and the discrepancy seems to be largest at small emission angles. The observed discrepancy indicates that the cut-off energy or the reaction time after which the cascade is stopped should be larger or smaller, respectively. In variance to this finding, however, the neutron energy spectra from the reaction 1.2 GeV p+Pb are very well described in the low energy region by calculations with the INCL4 code plus evaporation [40]. Neutron spectra from heavy targets are, however, less sensitive to the contribution from the INC part near the energy regime of evaporation since their evaporative multiplicity is about ten times larger than for protons and thus dominates the yield below 20 MeV for .

As mentioned above, the angular asymmetry of EV- and PE-particles is quite different - the EV component is more isotropic than the PE part. To demonstrate this more clearly the angular distributions of all LCPs for both components are shown in Fig. 8. In the left panel of Fig. 8 the EV component of is confronted with the results of INCL2+GEMINI calculations (dashed lines). The observed small asymmetry is due to a small recoil velocity of the emitting residual nuclei after the prompt intra nuclear cascade. The good agreement between experiment and calculation indicates that this recoil velocity is well predicted by the INCL2 code as will be discussed in more detail in Sec. 3.4. The angular distribution of pre-equilibrium component displays a much larger forward backward asymmetry as one would expect for these more directly emitted particles. In the case of protons, which are treated in an intra nuclear cascade code, the angular distribution is well describe by the INCL2 code for proton energies between 16 and 100 MeV.

The results for the total production cross sections \( \sigma_{\text{tot}} \) with energies between 0 and 100 MeV are listed for all hydrogen and helium isotopes and targets
in Tab. 4. The evaporative part \( \sigma_{EV} \) of these cross sections was obtained by normalizing the evaporation spectrum, as calculated with INCL2+GEMINI, to the measured evaporation spectra and then by integrating over energy. The PE cross section \( \sigma_{PE} = \sigma_{tot} - \sigma_{EV} \) is listed in Tab. 4 as well. Instead of plotting the production cross sections, we show in Fig. 9 the mean particle multiplicities \( \bar{M}_{EV,PE} = \sigma_{EV,PE}/\sigma_{inel} \) where \( \sigma_{inel} \) is the total inelastic cross section [54]. The latter is within a few percent identical to the geometrical cross section \( \sigma_{geo} = \pi r_0^2 \cdot A_T^{2/3} \) with \( r_0 = 1.26 \text{ fm} \). The thus obtained multiplicities reflect the probability to emit an EV or PE particle in one inelastic reaction. The mean evaporative multiplicities of tritium, deuterons, and \( \alpha \)-particles amount to about 0.15, 0.31, and 0.72 per inelastic reaction, respectively, and are almost independent of the target mass. In particular, these multiplicities are well described by the INCL2 + GEMINI calculations (lines) indicating once again that the mean excitation energy after the prompt INC is correctly calculated with INCL2. For the \( ^3 \text{He} \)-multiplicity, the agreement is somewhat worse, but the strong decrease with target mass, quite different to the other isotopes, is correctly described.

In the case of the halo nucleus \(^6\text{He}\) it was not possible to separate the PE and EV components due to low statistics. That is why we compare in Fig. 10 the total \(^6\text{He}\) multiplicity with the results of the INCL2 plus GEMINI calculations. Obviously the calculation overestimates the production of this neutron rich \( \text{He} \) isotope. It is interesting to note that the multiplicities are strongly increasing with increasing \( Z_T \) which is very similar to the behaviour of the neutron rich tritium as shown in Fig. 9. We associate this finding with the larger neutron excess \( N/Z \) of heavier targets.

The pre-equilibrium multiplicities up to maximum energies of 100 MeV are shown in the right panel of Fig. 9. First of all we observe that the magnitude of PE emission of composite particles is very similar to the evaporation except for \(^4\text{He}\). The relative contribution of PE emission amounts to 40-60\% for \( \text{d} \) and \(^3\text{He}\), 20-40\% for tritium and only 5-20\% for \(^4\text{He}\), where the lower and upper value applies to lighter and heavier targets, respectively (see also Tab. 4).

Whereas the multiplicity of evaporated \( \alpha \) particles is more than 4 times larger than that of tritium, the probability for PE emission for these two particle species is almost identical. This finding is somewhat difficult to understand within the framework of a successive coalescence model \([11,40]\), since for the formation of \(^4\text{He}\) three steps are required, while the formation of \(^3\text{H}\) takes only two steps. Furthermore, the coalescence of fast, that is INC nucleons, to form composite particles, removes nucleons from the calculated INC yield in the few tens of MeV energy region. This results in a deterioration of the good agreement obtained for fast protons, as shown in the right panel of Fig. 9, similar to the findings of \([11,40]\) for the \( p+\text{Au} \) reaction at 2.5 GeV.
By adjusting the phase space parameters used in the coalescence model for the production of different composite particles, it is possible to achieve satisfactory agreement with experimental data [11,40]. However, these parameters would have to be readjusted as a function of target mass. This can be seen from Fig. 11, where the ratio of the experimental PE composite particle yield $\sigma_{PE}$ to the calculated yield of fast protons $\sigma_{PE,p}$ with energies below 100 MeV is plotted. Within the coalescence model, one would expect this ratio for fast deuterons to be proportional to $N_d/N_p$ or to the multiplicity of fast neutrons $M_{n,INC}$. In fact, this multiplicity can be shown to be proportional to $R_T \cdot N_T/A_T$, where $N_T$ are the number of neutrons in the target nucleus with $A_T$ nucleons and radius $R_T$. In Fig. 11 the $Z_T$ dependence of this simple relation is shown by the upper dotted line after being normalized to $\sigma_{PE}/\sigma_{PE,p}$ for the Au-target. For light nuclei, the experimental data for the PE emission of deuterons deviates considerably from the coalescence expectation $N_d/N_p$. On the other hand, the $A$ dependence of the data is well described by the $N/A$ ratio of the target nuclei as shown by the dashed line which is also normalized to $\sigma_{PE}/\sigma_{PE,p}$ at Au. For the emission of PE tritons, one would expect similarly $N_t/N_p \sim (N_d/N_p)^2$ which is almost fulfilled near the Au-target but deviates again for lighter targets. This qualitative discussion shows that for a given nucleus, it might be possible to reproduce to some extent the PE emission of composite particles but the parameters would have to be adjusted separately for different mass regions of the target.

3.4 Mean excitation energies and linear momentum transfer

For calculation of the cross sections discussed further above, it is essential that the energy dissipation in the initial intra nuclear cascade is modeled correctly. Sensitive observables for the deposition of excitation energy are the excitation energy distribution [9] and the linear momentum transfer (LMT). In [9,11,12], it was shown that the experimentally deduced excitation energy distributions agree quite well with the INCL2 calculations, whereas they largely differ from the results of calculation performed with the LAHET code [49].

The average of the nuclear excitation energy distribution represent the mean energy $<E^*_p>$ dissipated and transferred to the nucleus in an inelastic $p$-induced reaction. This mean energy was deduced experimentally as described in [16] from the mean multiplicities of evaporated light charged particles (Fig. 9) and neutrons. The latter were measured separately with BNB [44,55] on somewhat thicker targets ranging between 0.1 and 1.0 g/cm$^2$. The thus obtained mean excitation energies (squares) are confronted in Fig. 12 as a function of the atomic number $Z_T$ of the target together with the predictions (solid line) of the INCL2 model. The calculation agree well with the experimental values between $Al$ and $Pb$ though for heavier target nuclei the
calculated mean excitation energy slightly overestimates the experimental results. It is worth noting that the "efficiency" \( <E^*/E_p> \) of converting the kinetic energy \( E_p \) of the incident proton into excitation is only 6% for a nucleus as light as Al, increasing to 16% for a heavy nucleus such as \( Pb \) while the balance is carried away by INC nucleons. This value for \( Pb \) is even higher than achieved in antiprotonic heating with 1.22 GeV antiprotons on \( U \), where only 11% [16] of the available energy (including annihilation energy) is converted.

While the intra nuclear cascade model accounts for the energy carried away by energetic nucleons, it does not account for the energy carried away by PE emission of composite particles. Since in the present work we have measured the energy spectra, we can calculate the mean energy \( <E_{PE}^i> \) per PE composite particle of species \( i \). The mean excitation energy carried away by PE composite particles is then given by the sum of \( <M_{PE}> \times (E_{PE} + B_i) \) over \( i = ^{2,3}H \) and \( ^{3,4}He \) by employing the mean multiplicities \( <M_{PE}> \) of PE composite particles from Fig. 9. Here, \( B_i \) is the binding energy of composite particle \( i \). The largest contribution to this sum is due to deuterons, for which it is about 80% . The result of this analysis is shown in Fig. 12 by the filled circles. The open circles in this figure display the sum of both components of mean energies carried off by LCP evaporation and PE-emission of composite particles out of which about 20-25% is due to PE composite particle emission.

The deposited excitation energy is correlated with the linear momentum transfer, which then provides an independent cross check of the INC calculation. In the following, we compare the experimentally deduced mean LMT with INCL2 predictions. The linear momentum transfer to the heavy residues can be deduced based on the asymmetry of the angular distribution of evaporated particles. And so, the forward-backward asymmetry \( Y(0^\circ)/Y(180^\circ) = (1+P_1+P_2)/(1-P_1+P_2) \) with \( P_1 \) and \( P_2 \) from Eq. 1 can be shown to be given by \( (\langle v_p + <v_{cm} > \rangle/\langle v_p - <v_{cm} > \rangle)^2 \), where \( <v_{cm} > \) and \( v_p \) are the mean center of mass and particle emission velocities, respectively, with the latter obtained from simulation calculations. This relation holds, however, only for isotropically emitted, that is evaporated particles. It should be noted that this method tends to favor the higher velocities since the excitation energy and thus also the \( \alpha \) multiplicity is larger for higher linear momentum transfers. In Fig. 13, the center of mass velocities deduced from evaporated \( \alpha \)-particles are shown by the filled circles as a function of the target \( Z_T \). The experimental results are intermediate between the INC calculations with (solid line) and without (dashed line) multiplicity weighting. Due to the pre-equilibrium emission of composite particles one would expect that the residue velocities are somewhat smaller than the INC predictions, where PE emission of composite particles is not taken into account.
3.5 Production cross sections for Li and Be

In the present NESSI experiment, also the production cross sections of Li and Be were measured. However, for these intermediate-mass fragments, the experimental lower energy cut-off is considerably larger than for LCPs, and a larger fraction of the IMF spectrum is missed in the measurements. In the cases of Li and Be the thresholds were equal to 10.5 and 16 MeV, respectively. To demonstrate the effect of this low energy cut-off, in Fig. 14 are shown the kinetic-energy spectra for Li and Be fragments produced in a light (Fe), a medium-weight (Ag), and a heavy (Au) target. The shaded histograms in this figure are results of INCL2+GEMINI simulation calculations. The shapes of the theoretical spectra have been used to calculate the reduction in the experimental particle yields caused by the significant energy thresholds. For light and medium-weight targets, the deduced corrections to the measured yields are substantial. For Li (Be) they amount to 140% (320%), 44% (98%), and 1% (8%) for targets of Al, Fe, and Ag. In contrast, for heavier targets the corrections are smaller than 1%. The deduced total production cross sections for Li- and Be-isotopes are listed in Table 5. The quoted errors include 30% of the extrapolated yields.

The production cross sections of Li- and Be-isotopes are shown in Figs. 15 and 16 as a function of target Z_f and compared with previous results. The present \(^9\)Li production cross sections agree within the experimental errors with the results obtained in Ref. [56] at a somewhat lower bombarding energy of 1.0 GeV on targets between Al and U (see Fig. 15). The present total production cross sections for the long lived radionuclides \(^7\)Be and \(^{10}\)Be agree quite well (Fig. 16) with the cross sections obtained[24] in activation measurements, for 1.2 GeV proton induced reactions on targets between Al and Cu.

In Figs. 15 and 16, the total production cross sections for Li- and Be-isotopes are compared with the results of the calculations with the same INCL2 + GEMINI code as employed for the LCPs. Since generally the GEMINI-code is not expected to describe the emission of IMFs correctly [43], the observed agreement between data and calculations is surprisingly good for the lighter Li and Be isotopes and the lighter targets. The calculations underestimate the experimental cross sections more significantly for heavier IMFs and heavier target nuclei. Discrepancies are larger for Be than for Li, and even some trends are incorrectly predicted for the former IMFs.

These findings can be attributed to the fact that the physics of statistical emission complex IMFs from a heavy nucleus is not well represented by a scenario of volume emission from the interior of such a nucleus at normal density. The corresponding Coulomb barriers for such processes are estimated to be by factors of 5-20 larger than prevailing nuclear temperatures. Both the relatively
large IMF yields and the measured energy spectra (see Fig. 14) are in qualitative support of a new statistical model[57] describing the emission of complex clusters from hot, expanded nuclei. In this model, the entropy associated with the enlarged surface of an expanded nucleus plays an important role in driving its binary decay. Emission barriers in this model are naturally lower than calculated for nuclei at their normal matter densities.

For lighter targets such as Al and Ti, it becomes conceptually more difficult to distinguish between evaporation IMFs and target residues. Here, the detected lithium and, in particular, beryllium fragments represent probably to an increasing extent evaporation residues of the original target nucleus, rather than having been evaporated from such nuclei. Converting, as described in Sect. 3.3, the cross section into multiplicity, one observes that the multiplicity of Be from reactions on Al is about 3 times larger than for heavy targets. Nevertheless, absolute IMF multiplicities even for these lighter targets remain small; the mean multiplicity is of the order of 0.03 for Li and 0.01 for Be. Consequently, the mean overall energy carried off per reaction by evaporated IMFs is also small, on average less than 1-2 MeV.

At high particle kinetic energies, the energy spectra of IMFs (cf. Fig. 14) and LCPs (cf. Fig. 7) show very similar exponential behavior. In this domain, the experimental yields are considerably larger than the calculated evaporative yields. Employing the same method as in the extraction of the PE component for the LCPs discussed in Sect. 3.3, one obtains PE contributions to the respective total yields of about 25% for Li and about 15% for Be. These values are relatively independent of the target Z value, except for the Al-target, where these contributions are smaller than 5%. In this case, one may expect such low yields, if the IMFs are actually target-like residues. For heavy targets, however, the relative yield of the highly energetic component IMF is very similar in magnitude to that obtained in Sect. 3.3 for the PE emission of α-particles.

In the case of LCPs, a strong argument in favor of an identification of this component with PE emission was its larger forward backward asymmetry compared to the low-energy evaporative component (see Fig. 8). In order to illustrate the presence or absence of this characteristic signal for Li and Be fragments, in the left panels of Fig. 17 the energy spectra of these IMFs measured at a forward angle (30°) are compared with those detected at a more backward angle (150°). Despite the low statistical accuracy, a significant forward/backward yield asymmetry is observed. The angular distributions of the low- and high-energy components, shown in the right panels of Fig. 17, demonstrate more quantitatively the difference between evaporative and PE emission. The observed high forward/backward asymmetry for highly energetic IMFs rules out the hypothesis that these IMFs are evaporated from extremely highly excited nuclei produced in processes in which a much larger
fraction of the initial proton energy than predicted by the INCL2 code is converted to thermal excitation of the target nucleus. In summary, one concludes that the highly energetic \( Li \) and \( Be \) fragments originate from a PE-process that is somewhat similar to that leading to the fast emission of energetic LCPs. However, the precise nature of such a PE emission process remains unclear. A stochastic coalescence model of formation and early emission of relatively heavy \( IMFs \) does not present a plausible scenario.

4 Conclusions

In the present work, systematics of total production cross sections are reported for hydrogen, helium, lithium, and beryllium isotopes emitted in 1.2-GeV proton-induced reactions on 13 targets between \( Al \) and \( Th \). These cross sections have been measured with typical total uncertainties between 5 and 10\%, for LCPs, or 10-30\%, for IMFs. The cross sections were derived from particle energy spectra extrapolated to energies below the experimental detection thresholds. This procedure results in uncertainties depending on the target Coulomb barriers. Associated errors are therefore larger for the lighter target elements with lower barriers, noticeable in particular for \( Al \).

Precisely known shapes of particle energy spectra, measured here between app. 0 and 100 MeV, allows one to decompose the \( ^1,^2,^3H \) and \( ^3,^4He \) production cross sections into evaporative and pre-equilibrium components. The deduced contributions of PE-emission amount to 40-60\% for \( ^2H \) and \( ^3He \), to 20-40\% for \( ^3H \), and to only 5-20\% for \( ^4He \). Very similar relative yields (15-25\%) have been deduced for the PE-emission of Li- and Be-fragments. The cross sections of evaporated particles are quantitatively reproduced by calculations based on a combination of INCL2.0 and GEMINI codes. On the other hand, the PE-emission process is not modelled in any realistic detail. Nevertheless, semi-quantitative arguments suggest that the simple statistical coalescence model is unable to reproduce the observed PE emission probabilities of complex particles with model parameters independent of the target mass. It is therefore of considerable interest to investigate other formation and emission models for complex particles. The present systematic data, collected over a wide range of target nuclei, provide excellent testing grounds for such new models, e.g., a combination of an intranuclear cascade mechanism with a surface cluster emission model [?] or a recently proposed percolation model[40].

The angular and energy distributions of non evaporative protons is well described by the INCL2 calculations. At energies below 20 MeV the yield of protons is, however, strongly overestimated by the INC calculations whereas for neutrons no such discrepancy was observed [40]. It is argued that protons are more sensitive to test the PE nucleon contribution in the near evaporative
energy regime due to a smaller multiplicity of evaporated protons compared to neutrons.

The calculated mean excitation energy which is dissipated in the initial intra nuclear cascade, a prerequisite for any further statistical decay calculation, agrees very well for a large range of target nuclei between Al and Pb with the mean excitation energy deduced from the measured mean multiplicities of light charged particles and neutrons. This finding corroborates a previously observed agreement between calculated and experimentally deduced excitation energy distributions [9,11,12]. Consequently, the PE composite particle emission does not significantly reduce the mean excitation energy of the target remnant. Rather, these PE emission processes induce a redistribution of energies between all energetic PE particles, nucleons as well as intermediate-mass clusters.

The present experiment provides independent information on the relation between mean excitation energy of the target remnant and the mean joint multiplicities of light particles, i.e., of neutrons and light charged particles. However, it is important to realize that the corresponding correlation is in general non-linear and not sharp. The mean linear-momentum transfer to the heavy residues during the prompt intranuclear cascade represents an independent observable characterizing nuclear excitation. In the present work, the transfer of linear momentum was deduced from the anisotropy of the angular distributions of the evaporated $\alpha$ particles and found to agree quite well with the INC predictions corroborating the above results.

5 Acknowledgments

We are indebted to the COSY staff for the good beam quality, J. Cugnon for providing us with the INCL2.0 code. This work was partially supported by the EU-TMR project ERBFMRXCT980244 and by US-DOE Grant No. DE-FG02-88ER40414.

References


[55] C.-M. Herbach et al., to be published.


Table 1
Targets used in the measurements of the absolute production cross-sections of H and He at an incident proton energy of 1.2 GeV. The layer thicknesses $\Delta x_w$ and $\Delta x_o$ have been determined by weighing and by energy loss measurements. The deviation 

$\frac{(\Delta x_o - \Delta x_w)}{\Delta x_w}$

is given. The nonuniformity has been investigated by energy loss measurements afar from the target center.

<table>
<thead>
<tr>
<th>Target</th>
<th>$\Delta x_w$ (mg/cm$^2$)</th>
<th>$\Delta x_o$ (mg/cm$^2$)</th>
<th>$\frac{\Delta x_o - \Delta x_w}{\Delta x_w}$ (%)</th>
<th>Nonuniformity (%)</th>
</tr>
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<td>&lt;7</td>
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<td>Ti</td>
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<td>2.42</td>
<td>-2</td>
<td>1</td>
</tr>
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<td>Fe</td>
<td>1.75</td>
<td>1.74</td>
<td>-1</td>
<td>4</td>
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<td>Ni</td>
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<td>1.73</td>
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<td>Cu</td>
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<td>1.46</td>
<td>-1</td>
<td>2</td>
</tr>
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<td>Zr</td>
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<td>2.10</td>
<td>-1</td>
<td>3</td>
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<td>Ag</td>
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<tr>
<td>Ho</td>
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<td>3</td>
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<td>Th</td>
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<td>6.17</td>
<td>5</td>
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Table 2
Experimental lower and upper energy thresholds for the measurements of the cross sections with BSiB and telescopes. The given lower thresholds for the measurements with the telescopes refer to the particle identification with $\Delta E^{80}$-E1000.

<table>
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<th>particle</th>
<th>BSiB</th>
<th>Telescopes</th>
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<td>$E_{low}$ (MeV)</td>
<td>$E_{upper}$ (MeV)</td>
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<td>$^1$H</td>
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<td>26±4</td>
</tr>
<tr>
<td>$^2$H</td>
<td>2.0</td>
<td>51±6</td>
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<td>$^3$H</td>
<td>2.0</td>
<td>77±7</td>
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<td>$^3$He</td>
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<td>100</td>
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<td>$^4$He</td>
<td>2.0</td>
<td>100</td>
</tr>
<tr>
<td>$^6$He</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$^6,7,8,9$Li</td>
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<td></td>
</tr>
<tr>
<td>$7,8,9$Be</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
Table 3
Production cross-sections of hydrogen and helium as measured with BSiB at an incident proton energy of 1.2 GeV. The cross sections are corrected for the counting losses below the lower energy threshold of 2 MeV, the upper cut-off energies correspond to values of 26 MeV, 51 MeV and 77 MeV for protons, deuterons and tritons, respectively, as well as to about 100 MeV for helium. The results of Enke et al. [9] have been corrected for a reduced geometrical efficiency of -7.7% [45] and counting losses due to double hits and due to lower energy cut offs. The Pb value of Ref. [26] for the He production cross section $\sigma_{He}$ in column 6 is obtained by $\sigma_{He} = \sigma_{4He} + \sigma_{c}/4$ with $\sigma_{c}$ the cumulative $^3$He cross sections quoted in Ref. [26].

<table>
<thead>
<tr>
<th>Target</th>
<th>H-production cross-section</th>
<th>He-production cross-section</th>
</tr>
</thead>
<tbody>
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<td></td>
<td>$0 \leq E_p \leq 26 \pm 4$ MeV</td>
<td>$0 \leq E_{He} \leq 100$ MeV</td>
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<tr>
<td></td>
<td>$0 \leq E_d \leq 51 \pm 6$ MeV</td>
<td>$0 \leq E_{He} \leq 77 \pm 7$ MeV</td>
</tr>
<tr>
<td>$\sigma_{H}$</td>
<td>$\sigma_{H}$</td>
<td>$\sigma_{He}$</td>
</tr>
<tr>
<td>(b)</td>
<td>(b) [9]</td>
<td>(b)</td>
</tr>
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<td>0.70±0.07</td>
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<td>Ti</td>
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<td>0.55±0.04</td>
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<tr>
<td>Fe</td>
<td>1.75±0.13</td>
<td>1.44±0.15</td>
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<td>Ni</td>
<td>2.29±0.15</td>
<td>0.68±0.05</td>
</tr>
<tr>
<td>Cu</td>
<td>2.03±0.13</td>
<td>0.70±0.05</td>
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<td>Zr</td>
<td>2.60±0.17</td>
<td>0.86±0.06</td>
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<td>Ag</td>
<td>3.11±0.21</td>
<td>1.05±0.07</td>
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<tr>
<td>Ho</td>
<td>3.13±0.26</td>
<td>1.25±0.10</td>
</tr>
<tr>
<td>Ta</td>
<td>3.44±0.20</td>
<td>2.44±0.20</td>
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<tr>
<td>W</td>
<td>3.39±0.23</td>
<td>2.45±0.20</td>
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<tr>
<td>Au</td>
<td>3.55±0.22</td>
<td>2.66±0.20</td>
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<tr>
<td>Pb</td>
<td>3.23±0.30</td>
<td>2.52±0.20</td>
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<td>Th</td>
<td>3.35±0.21</td>
<td>2.55±0.10</td>
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<td>U</td>
<td>2.38±0.20</td>
<td>1.27±0.10</td>
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Table 4
Total production cross-sections $\sigma_{tot}$ of hydrogen and helium isotopes with kinetic energies $0 \leq E_{He} \leq 100$ MeV for 1.2 GeV proton induced spallation reactions on targets between Al and Th. The pre-equilibrium cross sections $\sigma_{PE}$ are also listed while the evaporative cross section $\sigma_{EV}$ can be obtained from $\sigma_{EV} = \sigma_{tot} - \sigma_{PE}$.

<table>
<thead>
<tr>
<th>Target</th>
<th>$^1H$</th>
<th>$^2H$</th>
<th>$^3H$</th>
<th>$^4H$</th>
<th>$^6Li$</th>
<th>$^7Li$</th>
<th>$^8Li$</th>
<th>$^9Li$</th>
<th>$^{10}Be$</th>
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<tr>
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<td>762±198</td>
<td>390±20</td>
<td>189±50</td>
<td>189±50</td>
<td>72±14</td>
<td>62±14</td>
<td>12±3</td>
<td>42±7</td>
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<tr>
<td>Ti</td>
<td>198±505</td>
<td>833±126</td>
<td>307±55</td>
<td>165±28</td>
<td>10±17</td>
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<td>66±8</td>
<td>35±4</td>
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<tr>
<td>Fe</td>
<td>379±301</td>
<td>1166±139</td>
<td>462±24</td>
<td>217±29</td>
<td>116±15</td>
<td>46±7</td>
<td>76±8</td>
<td>35±4</td>
<td>57±7</td>
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<tr>
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<td>466±63</td>
<td>220±37</td>
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<td>47±9</td>
<td>72±8</td>
<td>31±4</td>
<td>59±9</td>
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<tr>
<td>Cu</td>
<td>330±308</td>
<td>156±196</td>
<td>69±73</td>
<td>347±46</td>
<td>77±4</td>
<td>9±14</td>
<td>73±5</td>
<td>42±3</td>
<td>77±67</td>
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<td>174±220</td>
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<td>114±16</td>
<td>85±11</td>
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<td>74±12</td>
<td>40±6</td>
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<td>621±73</td>
<td>516±46</td>
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<td>137±104</td>
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<td>1009±111</td>
<td>607±74</td>
<td>49±48</td>
<td>214±30</td>
<td>78±12</td>
<td>39±5</td>
<td>132±114</td>
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<tr>
<td>Au</td>
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<td>2937±438</td>
<td>1282±131</td>
<td>696±82</td>
<td>55±50</td>
<td>209±37</td>
<td>80±12</td>
<td>41±5</td>
<td>149±113</td>
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<tr>
<td>Pb</td>
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<td>78±13</td>
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<tr>
<td>Th</td>
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<td>779±530</td>
<td>72±68</td>
<td>290±44</td>
<td>82±12</td>
<td>49±6</td>
<td>147±120</td>
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Table 5
Total production cross-sections $\sigma_{tot}$ in mb for Li and Be isotopes with kinetic energies $0 \leq E_{Li,Be} \leq 100$ MeV for 1.2 GeV proton induced spallation reactions on targets between Al and Th.

<table>
<thead>
<tr>
<th>Target</th>
<th>$^6Li$</th>
<th>$^7Li$</th>
<th>$^8Li$</th>
<th>$^9Li$</th>
<th>$^7Be$</th>
<th>$^9Be$</th>
<th>$^{10}Be$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>11.5±3.8</td>
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<td>0.26±0.17</td>
<td>7.8±3.2</td>
<td>3.0±1.4</td>
<td>2.8±1.3</td>
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<tr>
<td>Ti</td>
<td>12.2±2.6</td>
<td>1.8±0.5</td>
<td>0.24±0.11</td>
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<td>2.3±0.8</td>
<td>1.8±0.7</td>
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<tr>
<td>Fe</td>
<td>12.8±2.5</td>
<td>1.6±0.5</td>
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<tr>
<td>Ni</td>
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<td>Zr</td>
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<td>Ho</td>
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<tr>
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<tr>
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<tr>
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<td>10.4±1.7</td>
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Fig. 1. Experimental setup of the NESSI-detector mounted at the COSY beam-line (from Ref. [45]). The two 4π-detectors BNB for neutrons and BSiB for charged particles are supplemented by six fourfold detector-telescopes. A thin plastic scintillator $S_1$ is used to count the incident beam protons and to deliver start signals for time-of-flight measurements. Further scintillation detectors are exploited as active beam collimators $S_7$-$S_8$ and for beam diagnostics $S_{10}$-$S_{14}$.

Fig. 2. Comparison of the angle integrated helium energy spectra as measured with the BSiB detectors (dots) and as calculated with the codes INCL2.0 and GEMINI (gray histograms). The dashed lines define the energy ranges which were employed to normalize the simulated spectra to the measured yields. The helium yield below the lower threshold has been determined from the simulated spectra. Particle absorption within the target layer contribute to the yield in the first energy bin (see histograms of Al and Ti).
Fig. 3. Spectra of stopped ($H_{\text{slow}}$) and punched-through ($H_{\text{fast}}$) hydrogen ions accumulated with BSiB in the reaction $p(1.2 \text{ GeV}) + Cu$. The lower off-line thresholds are indicated by the dashed lines. For punched through hydrogens ($H_{\text{fast}}$) the lower threshold corresponds to the upper cut-off energy for protons at about 26 MeV.

Fig. 4. Angular distribution of helium ions from the reaction 1.2 GeV $p + W$ as measured with the BSiB detectors (circles) and telescopes (stars). The data points in the top panel were obtained with the target foil perpendicular to the beam direction and low proton intensity of about $10^5$ pps while in the lower panel the target was rotated to $45^0$ and the employed beam intensity was about $5 \times 10^6$ pps. The lines are the result of a fit with the function of Equ. 1 to the BSiB data points (filled circles).
Fig. 5. Mass spectra of H, He, Li, and Be ions as measured with the $\Delta E$-E combinations of the telescope detectors 25$\mu$m-80$\mu$m, ($\Delta E$25-E80), 80$\mu$m-1000$\mu$m ($\Delta E$80-E1000) and 1000$\mu$m-7cm ($\Delta E$1000-ECsI).

Fig. 6. Experimental (symbols) and calculated (lines) total He ($^3$He+$^4$He) production cross sections as a function of the atomic number of the target $Z_T$. The cross section data obtained with particle detectors have been corrected for lower detection thresholds. In addition to the present BSiB data (filled circles) previous data have been included from Enke [9], Michel [24], and Leya [26].
Fig. 7. Left panels: Kinetic energy spectra of $^1,^2,^3 H$, and $^3,^4 He$ at 300, 750, and 1500 for the reaction $p(1.2 \text{ GeV}) + \text{Ta}$. The shaded histograms display the calculated evaporation spectra normalized to the experimental data. The calculated (INCL2) yield of promptly emitted protons is plotted by the dashed histogram. Right panels: Calculated INC proton spectrum (shaded histogram) and experimental difference spectrum $d^2\sigma_{\text{exp}}/(d\Omega dE) - d^2\sigma_{\text{EV}}/(d\Omega dE)$ for two targets, Ta and Ni.

Fig. 8. Experimental (symbols) and calculated (dashed lines) angular distributions of evaporative (left panel) and pre-equilibrium (right panel) LCPs emitted in the reaction 1.2 GeV p+Au.
Fig. 9. Left panel: experimental (symbols) and calculated (solid and dashed lines) mean multiplicities of protons and composite particles as a function of the target atomic number $Z_T$. Right panel: experimentally deduced mean multiplicities of PE particles (symbols) with energies below 100 MeV. The solid line is the result of INCL2 calculations for protons with kinetic energies smaller than 100 MeV.

Fig. 10. Experimental (symbols) and calculated (solid line) mean $^6$He multiplicities as a function of the target atomic number $Z_T$. 

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Fig. 11. Pre-equilibrium emission cross sections for $d$, tritons (t), $\alpha$, and $^3$He divided by the cross section of INC protons with kinetic energies below 100 MeV are displayed by different symbols as labelled in the figure; lines see text.

Fig. 12. Calculated (solid line) and experimental (■) mean dissipated thermal excitation energy deduced from evaporated neutrons and light charged particles. The energy carried off by PE emission of composite particles is shown by filled circles. The open circles display the sum of both components: evaporation plus PE emission.
Fig. 13. Mean recoil velocities (filled circles) of heavy residues as extracted from the forward backward emission asymmetry of evaporated alpha particles as a function of the atomic number of the target $Z_T$. Calculated recoil velocities with and without weighting with the alpha multiplicity are shown by the solid and dashed lines, respectively.

Fig. 14. Experimental (symbols) and calculated (shaded histograms) spectra of Li (top panels) and Be (lower panels) produced in the reactions 1.2 GeV p+Ti (left panels), p+Ag (middle panels) and p+W (right panels).
Fig. 15. Experimental (symbols) and calculated (lines) production cross sections of $^6$, $^7$, $^8$, $^9$Li with energies smaller than 100 MeV as a function of the atomic number of the target $Z_T$. The summed cross sections of all isotopes is also shown (Li). For $^9$Li the data of Dostrovsky et al. [56] are shown as well.

Fig. 16. Experimental (filled circles) and calculated (lines) production cross sections of $^7$, $^9$, $^{10}$Be with energies smaller than 100 MeV as a function of the atomic number of the target $Z_T$. The summed cross sections of all isotopes is also shown (Be). For comparison the results of Michel et al. [24] for $^7$Be and $^{10}$Be are shown by the stars.
Fig. 17. Left panels: Experimental energy spectra of Li (top panel) and Be (lower panel) at 30° (●) and 150° (○) from the reaction 1.2 GeV p+Au. Right panels: angular distributions of Li and Be with energies larger (○) and smaller (●) than the energy defined by the dashed line in the left panels at 42 MeV for Li and at 52 MeV for Be.
Fast decision in favor of the slow fission process

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The fission probability $P_f$ of highly excited target-like nuclei produced in reactions of 2.5 GeV protons on Au, Bi, and U was studied as a function of excitation energy $E^*$ whereby $E^*$ is deduced event-wise from the multiplicity of evaporated light particles. At the highest $E^*$ of 800 - 1000 MeV $P_f$ amounts to $\approx 50\%$ with all 3 target nuclei irrespective of the initial fissility. Statistical-model calculations satisfactorily reproduce the observed evolution of $P_f$ with $E^*$ - provided that no extra transient delay is introduced. Fission thus is decided upon very fast and early in the long deexcitation chain towards scission which comprises as much as $\approx 80\%$ of all evaporated alpha particles.

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At low excitation energy nuclear fission is strongly influenced by nuclear shell effects, resulting for instance in the well known binary split into two mass asymmetric fission fragments. With increasing excitation energy these nuclear structure effects are washed out and macroscopic properties such as nuclear dissipation dominate the collective flow of nuclear matter from the equilibrium deformation via the saddle point towards scission into two fragments of about equal size. The overall time elapsed from the equilibrium deformation up to scission is found to be in the order of a few times $10^{-20}$ seconds. This time is quite long compared to other nuclear processes such as the emission of neutrons which is typically 10 to 100 times faster. This observation has led to the notion of fission being a slow process [1].

Since the magnitude of nuclear dissipation is most likely deformation dependent [2] the characteristic time governing the flow over the saddle point, where the decision about fission is made, might be considerably shorter than the one close to the scission point. In particular at high excitation energies, where the emission times of light charged particles are very short, a long transient delay for fission at the saddle point would strongly favor the emission of charged particles thus reducing the fissility and, consequently, first- and higher-chance fission probabilities. Conversely, low dissipation at the saddle or a minimum transient delay tends to keep fission competitive with particle evaporation even at high excitation. In other words, although the entire fission process is slow the decision to fission can be fast.

The maximum excitation energy, $E^*$, reached in the present investigation is only about 4-5 MeV/nucleon or $\approx 1000$ MeV, as will be shown below. This excitation is well below that reported for the onset of multi-fragmentation [3], the decay of a hot nucleus into multiple clusters and nucleons. Thus the two main reaction channels expected in the present experiment are spallation induced fission and evaporation, the latter process leading to the survival of a heavy target-like residue plus many evaporated nucleons and light charged particles (LCPs).

We have chosen to exploit 2.5 GeV proton-induced reactions to excite three target nuclei, Au, Bi, and U with different fissilities $Z^2/A = 31.7, 33.0, \text{ and } 35.6$, respectively, in order to study the evolution of the fission probability as a function of excitation energy and fissility. These reactions, similar to antiproton- [4, 5] or peripheral relativistic heavy-ion reactions [6], are thought to deposit high thermal excitation energies with minimum ballast from collective excitations (angular momentum, shape distortions, or density compression) which could have a detrimental influence on the nuclear decay.

The amount of energy dissipated into excitation is deduced in the present experiment event by event from the total number of evaporated light particles (n, H, He), a well proven technique, the details of which are described in [7, 8]. It presupposes a 4\% coverage and high acceptance at high granularity of the detectors, requirements fulfilled by the combination of our two detectors, the Berlin Neutron Ball (BNB) and the Berlin Silicon Ball (BSB). BNB is a large spherical tank with 1300 l of Gl-loaded scintillator liquid which in its central reaction chamber houses the BSB, a self-supporting shell of 151 individual Si-detectors. These Si-detectors provide a total geometrical acceptance of about 80% with their energy thresholds as low as 2 MeV. Below are briefly summarized some details of the present experiment.

The experiment was performed at the COSY accelerator in Jülich with 2.5 GeV protons impinging on Au (2 $\times$ 324 $\mu$g/cm$^2$), Bi (629 $\mu$g/cm$^2$), and UF$_4$ (725$\mu$g/cm$^2$) + C (36$\mu$g/cm$^2$) targets. The hardware threshold of BNB was adjusted to detect all reactions with an inelasticity larger than 3 MeV. The high ($\approx 85\%$) detection efficiency of BNB for slow neutrons and the low ($\approx 15\%$) efficiency for energetic ($\geq 100$ MeV) ones makes BNB sensitive rather to evaporative neutrons than to fast ones from the direct interaction of the incident proton with the target nucleus. In BSB the identification of evaporative LCPs and the reconstruction of the mass of heavier fragments, i.e., of intermediate-mass fragments (IMFs), fission fragments (FFs), and heavy evaporation residues (HRs), are based on the measurement of time of flight and energy.

While the dissipated thermal excitation energy, $E^*$, can be deduced from the experiment for each event, the $A$- and $Z$-content of the excited nucleus after the fast intra-
nuclear cascade (INC) is accessible from INC calculations only. Since the knowledge of $A$ and $Z$ is a prerequisite for the calculation of the fission probability, the reliability of the employed INC model needs to be ascertained by comparing calculated inclusive excitation energy distributions with the experimental data. Such a comparison is made in Fig. 1 exhibiting indeed a good agreement for the three target nuclei between experiment (open circles) and the simulation with the INCL 2.0 model from Cugnon [9] (solid histograms). We conclude that also the $A,Z$-population is correctly described by the model.

![Fig. 1](image1.png)

**Fig. 1:** Measured inclusive ($\circ$) and fission (△) differential cross sections $d\sigma/dE^*$ as a function of excitation energy $E^*$ for reactions induced by 2.5 GeV protons on U (left), Bi (middle) and Au (right) nuclei. The solid histograms are the result of INCL 2.0 [9] calculations of the inclusive cross sections.

The mass resolution in BSIB is relatively poor, in particular for the heavier fragments, chiefly due to the short flight path of only 10 cm. Setting limits on the total mass of all detected reaction products, $A_{\text{tot}}$, and on the mass of the heaviest, $A_1$, and second heaviest particle, $A_2$, allows one to separate the three major fragmentation products HRs, FFs, and IMFs, as is illustrated in the left panel of Fig. 2. It exhibits, for the case of Bi with a mass intermediate between those of Au and U, the correlation between $A_1$ and $A_2$. For the purpose of this plot, the experimental $A_1-A_2$ data have been symmetrized. Here in Fig. 2, the events with both FFs detected are centered in the middle at $A_1, A_2 \approx 90$. Events with a HR detected together with some lighter masses appear close to the $x$- and $y$-axis near $A_1, A_2 \approx 130$, while most of the events with only one FF and some lighter masses detected are suppressed by the additional requirement $A_{\text{tot}} > 70\%$ of the mass of the target nucleus $A_{\text{targ}}$. Also, as expected, there are hardly any multi-fragmentation events, which would populate the lower left corner of the plot with $A_1, A_2 \leq 30$. In order to separate events with two FFs detected from HR events we use the further condition $A_1, A_2 > 32$ indicated by the dashed lines in the left panel.

The two other panels in Fig. 2 are meant to demonstrate that fission events selected according to the above criteria also fulfill the conventional conditions known from low-energy fission studies on the total kinetic energy TKE and the folding angle between the FFs. Indeed, the middle panel shows that these events are grouped around the solid line calculated with the Viola formula $<\text{TKE}> = 0.1189 Z^2/A^{1/3} + 7.3$ MeV [10] with $Z = 0.45 (A_1 + A_2)$, whereas they lie somewhat above the dashed line with $Z$ values chosen according to the $\beta$-stability - in agreement with other investigations [11]. As to the folding angle distribution of the FFs plotted in the right panel, we witness a growing broadening and deviation from $180^\circ$ with increasing $E^*$ as the result of more and more numerous emission of lighter particles and increasing momentum transfer. Because of this strong evolution with $E^*$, we refrained from requiring more or less back-to-back emission for the FFs by choosing a limiting folding angle.

Once the FFs have been selected with the two conditions (which, by the way, have been used before [4, 5]) namely $A_{\text{tot}} > 70\%$ of $A_{\text{targ}}$ for complete events and by $A_1, A_2 > 30, 32$ and 35, for Au, Bi, and U, respectively, their yield is corrected for detection efficiency as is described in detail in Ref. [8] to give finally the fission cross sections shown in Fig. 1 as a function of $E^*$. The total fission cross sections deduced amount to $\sigma_f = 200\pm 60$ mb, $320\pm 50$ mb, and $1350\pm 120$ mb, for the Au, Bi, and U targets, respectively. These values are in reasonable agreement with the respective fission cross sections of $71\pm 7$ mb, $180\pm 9$ mb, and $1480\pm 60$ mb obtained by Vaishnave et al. [12] at a lower proton energy of 1 GeV, however.

The fission probability $P_f$ is now obtained directly as the ratio of the thus determined fission cross section and the inclusive cross section. The resulting evolution of $P_f(E^*)$ with the excitation energy $E^*$ is shown in Fig. 3 by symbols for 2.5 GeV p+Au (a), Bi (b), and U (c).

The figure shows that the evolution of $P_f(E^*)$ with excitation energy is quite different for the three targets. At the low excitation of about 150 MeV, the fission probability of an U-like nucleus is close to 90% while for the less fission Bi-like nucleus it is 23% and only 5% for Au-like nuclei. At such low excitation energy, the excited nuclei are still similar in $A$ and $Z$ to the target nucleus since only a few nucleons are removed during the INC stage, and the different behavior of $P_f(E^*)$ is still dominated by the diverse initial fissility. At higher excitation energy considerable more nucleons are emitted during the

![Fig. 2](image2.png)

**Fig. 2:** Logarithmic contour plots used for the selection of fission fragments from the reaction $p+Bi$. (a): Correlation between the two heaviest fragments $A_1$ and $A_2$. The lines for $A_1, A_2=32$ indicate the border between “two-FFs-detected” and “HRs-detected”; (b): TKE of the two FFs as function of their sum mass $A_1+A_2$. The expected relation between $<\text{TKE}>$ and $A_1+A_2$ for $Z$-values according to the $\beta$-stability or for $Z = 0.45 (A_1 + A_2)$ is indicated by the dashed or full line, respectively; (c): Distributions of the folding angle $\theta_{\text{fold}}$ between the FFs (in arb. units/rad) as function of $E^*$. 

The figure shows that the evolution of $P_f(E^*)$ with excitation energy is quite different for the three targets. At the low excitation of about 150 MeV, the fission probability of an U-like nucleus is close to 90% while for the less fission Bi-like nucleus it is 23% and only 5% for Au-like nuclei. At such low excitation energy, the excited nuclei are still similar in $A$ and $Z$ to the target nucleus since only a few nucleons are removed during the INC stage, and the different behavior of $P_f(E^*)$ is still dominated by the diverse initial fissility. At higher excitation energy considerable more nucleons are emitted during the...
INC (at $E^*=1000$ MeV about $\Delta A=12$ mass and $\Delta Z=4-5$ charge units according to INCL2.0 [9], very similar for Au, Bi, and U). Consequently, the fissility of the thus produced nuclei is considerably lowered. This results in a strong decline of the fission probability in the case of U from the high value of about 90% at 150 MeV down to 35% at about 900 MeV, while for the Au target a continuous increase of $P_f(E^*)$ with $E^*$ is observed. For the Bi-target one notes an intermediate behavior, with an almost constant value of about 25% between 200 and 600 MeV, followed by a further increase to about 30%. Obviously, in the flat region of $P_f(E^*)$ for Bi the loss in fissility from the A,Z-depopulation is compensated by an enhancement of fission due to the increase of $E^*$, while from then on the effect of an increasing $E^*$ dominates.

Despite the large differences in fissility and fission barriers $B_f \approx 5, 12$, and 21 MeV (finite-range fission barriers [14] used in the calculations discussed below) of the initial nuclei U, Bi, and Au, respectively, and thus also of the excited nuclei one observes that at high excitation energy the fission probability assumes the same value of about 30% for all three target nuclei U, Bi, and Au.

After this empirical discussion we confront now the observed evolution of $P_f$ with $E^*$ with Statistical-model calculations applying the Transition-State model to fission. To this purpose we have used the population of nuclei as a function of $E^*$ predicted by the INCL2.0 code [9] as input to the Statistical-model code GEMINI by Charity et al. [15]. The angular momentum imparted during the INC (up to 20 units of $h$ at the highest $E^*$) is likewise taken into account, though it is here of minor importance for $P_f(E^*)$. For the level density parameters we assumed $\alpha_n = (A/10) \text{ MeV}^{-1}$ and for their ratio at the transition state and at ground state deformation $\alpha_f/\alpha_n = 1.000, 1.017$, and 1.022 for U, Bi, and Au respectively. For U and Au these values are the same as in [4], while for Bi we have chosen an intermediate value. No extra transient time for fission is applied. The result of these calculations is shown in Fig. 3 for the three targets by the dashed (U), dotted (Bi), and solid (Au) histograms. It is obvious that the calculations reproduce the characteristics different trends in $P_f$ at lower $E^*$, as well as the almost equal $P_f$ at the highest $E^*$ nearly quantitatively, and, most important, they do so without the use of any additional transient time.

What can be learned from this agreement for the understanding of fission? From the calculations we can identify the range of multiple-chance fission steps or the range in excitation energy $E_{\text{ex}}^*$ when fission is decided upon. At $E^* = 800$ MeV of initial excitation, for example, fission occurs at a mean excitation energy of $<E_{\text{ex}}^*>=700$ MeV for Au and Bi with a width of $\sigma_{E_{\text{ex}}} = 110$ MeV, while for U the corresponding values are 600 and 180 MeV. This implies that fission occurs rather early in the long deexcitation chain at these high initial excitation energies. For U, due to its lower fission barriers, fission is spread over a larger range of deexcitation steps and thus comes on average on a lower energy $<E_{\text{ex}}^*>$.

These conclusions are at variance with the findings of Jurado et al. [16] reporting a maximum excitation energy for the decision to fission of less than 300 MeV in the reaction $^{238}$U (1 A GeV) on $(CH_2)_n$. Also, Benlliure et al. [13] concluded from a similar inverse-kinematics experiment ($^{197}$Au (0.8 A GeV) + p) on a mean energy for fission of $<E_{\text{ex}}^*> = 128 \pm 20$ MeV for Au-like nuclei.

Certainly, the quality of the reproduction of the $P_f(E^*)$ data by the model calculation depends critically on the ratio of the level density parameters. Benlliure et al. [13] used $\alpha_f/\alpha_n = 1.05$, as suggested by Ignatyuk et al. [17], in the analysis of their fission cross section. Since, however, such an enhancement of the level densities at the saddle boosts fission to very large values at high $E^*$, the authors had to introduce a transient delay as long as $2 \times 10^{-21}$ s with the intention to reduce the theoretical $\sigma_f$ to their experimental value of $33 \pm 10$ mb. In order to confront our Au data with these parameters, we show the thus calculated fission probability $P_f(E^*)$ by the dot-dashed line in Fig. 3. Clearly, this calculation is in complete disagreement with our experimental outcome at low as well at high initial excitation energy, and the same holds for similar calculations with $\tau_f = 2 \times 10^{-21}$ s for Bi and U. On the other hand, the calculated cross section of $\sigma_f = 170$ mb for Au is in reasonable agreement with our measured value of $\sigma_f = 200$ mb. But the mean excitation energy at saddle, $<E_{\text{ex}}^*>$, in this calculation is only about 150 MeV, which is very similar to the one deduced in Ref. [13], but much lower than that obtained with the ratio $\alpha_f/\alpha_n = 1.022$ and no transient delay time.

The above comparison of measured and calculated fission probabilities as function of $E^*$ suggests that the mean excitation energy at which fission is decided on is relatively high and thus fission seems to be initiated on a fast time scale. If these conclusions are correct, the high remaining excitation energy must be carried off, either during the descent from saddle to scission or after scission. Indeed, according to the calculation at $E^*=800$ MeV only between 11% (Au) and 23% (U) of all light particles are emitted before reaching the saddle, or, more directly in terms of pre- and post-saddle LCP multiplicities,
$M_{\text{pre-saddle}}^{LCP} = 1.9, 1.9, 3.3$ and $M_{\text{post-saddle}}^{LPC} = 11.0, 10.4$, and 7.6 for Au, Bi, and U, respectively.

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{energy_spectra.png}
\caption{Energy spectra of $\alpha$-particles which are emitted into 3 angular domains relative to the motion of the light fission fragment: $\Theta_{\alpha-LF} = 10^\circ$ to $40^\circ$, $70^\circ$ to $100^\circ$, and $140^\circ$ to $170^\circ$. Three calculated components are fitted to the experimental distribution (circles): the contribution from the compound nucleus prior to scission (thick continuous lines) and from the light (dashed) and the heavy (dotted) fission fragment. At $70^\circ$ to $100^\circ$ a further component for the neck emission is added (thin line). The total calculated spectrum is shown by the histogram. Example from 2.5 GeV p+Au at $E^* = 600-900$ MeV.}
\end{figure}

While from the present experiment the pre-saddle LCP emission cannot be disentangled from the one occurring during the transition from saddle to scission, the amount of the post-scission LCP emission can be deduced from their angular correlation with the fission axis. To this end we show in Fig. 4 the energy spectra of alpha particles (with a 10% admixture of $^3\text{He})$ for three ranges of emission angles $\Theta_{\alpha-LF}$ relative to the motion direction of the light fission fragment.

The spectra are plotted in the center-of-mass system of the fission fragments from the reaction p + Au at $E^* = 600-900$ MeV, as an example. The spectra in the three angular zones $\Theta_{\alpha-LF} = 10-40^\circ$, 70-100$^\circ$, and 140-170$^\circ$ are fitted with the predicted contributions from three relevant emission sources: from the compound nucleus prior to scission which is unrelated to the scission axis and from the two fission fragments which vary in shape and intensity with $\Theta_{\alpha-LF}$. At 70-100$^\circ$, i.e. perpendicular to the scission axis, it turned out to be necessary in agreement with previous observations [18] - to add a small ($\approx 5\%$ of the total $\alpha$-evaporation) component for neck emission which peaks at 15 MeV. The sum of all components shown by the histograms provides a 24-level approximation at least to the lower-energy ($E_\alpha \lesssim 35$ MeV) part of the experimental spectra, in particular to the low-energy shoulders visible near $E_\alpha = 5$ MeV due to the emission from the heavy (10-40$^\circ$) or the light (140-170$^\circ$) FF. The pronounced high-energy tails in the measured spectra, instead, are mostly due to pre-equilibrium $\alpha$-emission which is not taken into consideration in the evaporation calculation with the code GEMINI, but is irrelevant for the present purpose. Since the spectra below $E_\alpha \lesssim 35$ MeV are dominated by evaporation, one concludes from the good fit that as much as about 80% of the total evaporation yield stems from the compound nucleus prior to scission, while only (20$\pm$10$\%$) originates from the separated fragments. Hence, for the reactions at hand this observation corroborates the general findings that the fission process is relatively slow [1], which so far are based mostly on heavy-ion reactions and much lower excitation energy.

In summary, we have studied the excitation energy dependence of the fission probability in 2.5 GeV proton-induced reactions on Au, Bi, and U. Simulations with the combined Intra-Nuclear-Cascade-/Statistical-model hybrid provide a very satisfying reproduction of the observed evolution of the fission probability $P_f$ with excitation energy $E^*$. Even the surprising observation of similar fission probabilities, $P_f \approx 30\%$, for all three target nuclei at the highest $E^* = 800-1000$ MeV is reproduced. Prerequisite for this good agreement is, that no additional transient delay time is allowed for the fission dynamics at the saddle. Fission thus seems to be determined upon very fast and early in the long deexcitation chain from the highest initial excitations - a conclusion which is, however, at variance with recent conclusions from other provenience [13, 16]. The finding of a fast decision to fission is supplemented by the observation that the major part (about 80\% at $E^* = 600-900$ MeV) of all evaporated alpha particles is emitted prior to scission, or, that the entire fission process is indeed relatively slow - in agreement with the previous evidence for this process.

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\end{itemize}
A Simple Method for Rise-Time Discrimination of Slow Pulses from Charge-Sensitive Preamplifiers

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Abstract

Qualities of a simple method of discriminating slow pulses from charge-sensitive preamplifiers according to their rise times have been analyzed for the range of rise times from 10 ns - 500 ns. The method is based on measuring, using fast peak-sensing ADCs, peak values of two pulses derived from the raw preamplifier pulses by the way of using two amplifiers with largely differing shaping times. It is is found that for the charges injected corresponding to energy deposits in silicon detectors of a few tens of MeV, the resolution in rise time can be achieved that is of the order of 1 ns. The method is expected to be useful in experiments involving large-area silicon detectors, whenever the mechanics of charge collection leads to a desired spread in pulse rise times for different species of impinging particles, such as is the case with $\Delta E$ detector constituents of the CHIMERA multidetector array [1].
I. INTRODUCTION

Pulse-shape discrimination is a commonly used technique of identifying particles, based on the time-dependence of the electrical response they generate in the detectors. There are different physical reasons for the response of a particular detector being dependent on the specie of an impinging particle and, accordingly, many different pulse-shape discrimination methods have been devised.

The present study is inspired by the actual demand of measuring rise times of pulses originated in large-area silicon $\Delta E$ detector constituents of the CHIMERA telescope array [1] and aims at investigating an alternative to measuring rise time by means of two constant fraction discriminators, operated with different fraction settings. The study focuses on an idea of measuring rise times of slow pulses from charge-sensitive preamplifiers, by shaping (filtering) these pulses with two different timing constants, one of which is much longer than the relevant maximum rise time and the other is of the order of the shortest rise time expected in the particular application. With proper shaping, the height of the “slow” pulse (representing the total charge injected into the preamplifier) will then be virtually independent of the rise time of the raw preamplifier pulse, while the height of the “fast” pulse (representing the fraction of charge injected on a short timescale) will show a distinct dependence on the rise time of the preamplifier pulse. Subsequently, the heights of both pulses can be digitized by fast peak-sensing Analog to Digital Converters (ADCs), such as, e.g., Phillips 7164 [2] or Silena [3]. Alternatively, depending on the data acquisition setup on hand, the shaped pulses can be stretched and their height measured by means of current-integrating ADCs (QDC). Ultimately, the rise time is determined event-by-event based on the ratio of “fast” to “slow” pulse heights.

II. THE EVALUATION SETUP

A block diagram of the evaluation setup is shown in Fig. 1. As depicted in this diagram, calibrated charge was injected into an ORTEC 142A charge-sensitive preamplifier, with three different rates, resulting in preamplifier output signals that had nominally rise times 50, 100, and 200 ns. Charge pulses were obtained by charge-terminating pulses...
FIG. 1: Block diagram of the electronic setup used to evaluate the quality of the pulse-shape discrimination.

from a precision ORTEC 448 pulser. The rise time was controlled by selecting different rise time settings of the pulser, while the decay time was fixed at 50 µs. A typical pulse with a nominal (set on the pulser module) rise time of 100 ns is illustrated in Fig. 2 and is seen to have an actual rise time that is close to the nominal one.

The output signal from the preamplifier was fanned out into two spectroscopic channels characterized with different shaping times. The “slow” channel had the shaping time fixed at 1 µs, while for the “fast” channel, three different differentiation times of 10, 20, and 50 ns were explored. The integration time of the ORTEC 454 timing filter amplifier (TFA) used in the “fast” channel was fixed at 50 ns.

To be able to use a common ADC gate for both, fast and slow pulses, a TC215 delay amplifier was used in the fast channel, between the output from the ORTEC 454 TFA and the input to the ADC. The resulting timing of the pulses arriving at the peak sensing Phillips 7164 ADC is illustrated in FIG. 3. Note that a Phillips 7164 ADC was here
FIG. 2: Appearance of the leading edge of a typical pulse from the charge-sensitive preamplifier with nominally 100 ns rise time.

FIG. 3: The timing diagram of the “fast” and “slow” spectroscopic pulses and of the ADC gating pulse.

chosen because its capability of digitizing pulses with short rise times, as low as 50 ns.
FIG. 4: The dependence of the average height of the “fast” pulse on the average height of the “slow” pulse for different amounts of injected charge, different rise times of the raw preamplifier pulse, and different shaping times, as indicated in the panels.

III. RESULTS AND ANALYSIS

The measurements were performed for series of pulses with three different rise times, 50 ns, 100 ns, and 200 ns and for three different shaping times for the “fast” response, 10 ns, 20 ns, and 50 ns. To assess the effect of inherent fluctuations in the amount of charge injected, the latter amount was varied in the range of .222 pC to 3.55 pC, corresponding to energy deposits in a silicon detector in the range of 5 MeV to 80 MeV.

Results of the measurements are illustrated in Figs. 4, 5, 6, and 7. Fig. 4 illustrates the dependence of the amplitude of the “fast” pulse on the height of the injected charge as measured for different shaping times and different rise times. As seen in this figure, shorter shaping times provide for a better separation of faster pulses, a trend that agrees
FIG. 5: Spectra of the ratios of heights of “fast” versus “slow” pulses, measured with various shaping times and for two representative amounts of injected charge. The three peaks seen in every panel correspond to three different rise times, as indicated by labels.

qualitatively with what is expected based on the principles of filtering.

A more quantitative measure of the pulse-shape discrimination power of the method over a range of injected charges is seen in Fig. 5, where spectra of the ratios of the peak values of “fast” pulses to peak values of “slow” pulses are displayed for various shaping times. As expected, the resolution of the method deteriorates with decreasing amount of injected charge. Again, short shaping times are seen to provide for a larger spread between pulses of short rise times, at the expense of a loss in amplitude.

Relative width of peaks seen in Fig. 5 are illustrated in Fig. 6 as functions of injected charge for different shaping times and different rise times. As seen in this figure, the relative resolution is almost equally good for 10-ns and 20-ns shaping times and is well below 1% over most of the range of injected charges, except for the smallest one.
FIG. 6: Relative resolution in the height ratio of “fast” and “slow” pulses for different amounts of injected charge, different shaping times, and different rise times, as indicated by labels.

Fig. 7 illustrates absolute resolution in rise-time in units of nanoseconds, achieved for different amounts of injected charge and for different shaping times and different rise times. As seen in this figure, for the range of rise times explored, the 10 ns and 20 ns shaping times offer almost equally good resolution of a few ns over much of the range in charges explored. The weak but steady loss of the resolution with decreasing amount of charge injected (expressed in units of MeV equivalent energy deposit for silicon), comes from the increase in statistical fluctuation in charge. It is encouraging that except for the lowest charge injected, the resolution is of the order of a few ns, and is approximately 1 ns for the range of energies involved in CHIMERA experiments.
A method of discriminating pulses from charge sensitive preamplifiers based on measurements of heights of pulses derived by shaping the raw preamplifier pulses with different time constants was evaluated and found to provide for a very good resolution in rise times at a low cost and high degree of simplicity. The method, in fact, requires no tuning of electronics circuitry involved and should, therefore, be well suited for multidetector systems. By design, it considers pulses produced by charge-sensitive preamplifiers, such as are already in place in the complex CHIMERA setup. This makes it easy to implement as an addition to the existing setup rather than a redesign. In the present study,
a timing filter amplifier was used to produce the “fast” signal. In fact, a most primitive RC differentiator should do as good a job, provided its output amplitude is matched to the range of the available ADC. At any rate, it appears well justified to carry out test experiments with real signals from the CHIMERA detectors and to compare the merits of this method to the merits of the “two-discriminator” method based on direct measurement of the rise time.

Acknowledgments

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Abstract—Mass and atomic-number identification (ID) of reaction products is a fundamental requirement of any nuclear reaction study. An effective particle-ID method is demonstrated, based on pulse shape analysis/discrimination (PSD) applied to large-area, single-element silicon detectors. This technique uses commercial electronic modules and achieves atomic number resolution rivaling that typically obtained with multi-element (AE–E) detector telescopes. The method is applied to the CHIMERA detector system without compromising its time-of-flight (TOF) resolution. In-beam tests of the PSD method have been performed with large-area, 300-μm thick CHIMERA silicon detectors, measuring particles from the $^19F + ^{12}C$ reaction at Tandem energies. Performance of a simple PSD set up is discussed, for front and rear particle injection.

This work was supported in part by the Italian Ministry for University and Research (MIUR) under contract COFIN2002.

The CHIMERA array consists of two large-area silicon detector banks, each of which is a 2-D grid of silicon detectors. Each detector element is a 200×200 mm$^2$, 1.0 mm thick XEOL detector CHIMERA [10-12]. The front bank is the 17×50 detector grid of the CHIMERA array.

I. - INTRODUCTION

In heavy-ion reaction studies, it is important to characterize reaction products according to mass (A), charge (Z), and energy (E). Precise mass and charge determinations of particles from compound-nucleus or direct reactions achieved in early nuclear heavy-ion reaction studies at low bombarding energies (E/A<10 MeV) have been instrumental in the elucidation of the underlying reaction mechanism [1-2]. In the more recently explored domain of heavy-ion reactions at Fermi or intermediate bombarding energies (10 MeV< E/A< 100 MeV), an accurate characterization of reaction fragments according to their A and Z (or their N/Z ratio) has become perhaps even more important. Here, one is interested in comparing the isospins $I_x = (A-2Z)/2A$ of fragments formed at lower or higher matter densities to those of particles produced at the normal nuclear matter density [3-4]. Emphasis on isospin transport in much of contemporary and planned nuclear research has therefore motivated efforts to improve the experimental resolution in particle energy, mass, and charge currently obtained with existing 4π detector systems [4-7].

Improvements in the resolution of the above physical properties with large detector arrays depend crucially on the technology employed for the front-end electronics of these arrays. Crucial components are, in particular, large-bandwidth preamplifiers, as well as fast-timing and efficient (fast) pulse shape analysis methods. The present work discusses new developments of high-quality current-sensitive preamplifiers with large bandwidths that allow one to study the time dependence of intrinsic detector currents [8-9]. In addition, results are presented of studies of particle discrimination with large-area silicon elements of the 4π detector CHIMERA [10-11] based on simple pulse shape analysis.

It is well known that the timing characteristics of the current pulse produced by electron and hole collection in silicon detectors do not only depend on the intensity of the electric field along the particle track. In fact the detailed shape of a current pulse results from the interplay of rather complex physical phenomena arising along in the path of the ionizing radiation [12, 13]. Ionization density, electron/hole recombination, and effective charge separation vary along the particle...
track and are influenced by the magnitude of the intrinsic electric field. Of most practical consequence is the realization that the shape of the current signal generated in the active region of a silicon detector depends strongly on which side the ionizing particle enters the detector. In front-side injection into a totally depleted n-type silicon detector of low or medium resistivity (~5000 \( \Omega \text{cm} \)), a particle experiences a relatively high electric field only during the initial portion of its track, where the ionization density is small, while much of the ionization processes occur at a relatively low field strength. In contrast, in rear-side injection, most of the ionization induced at the later parts of the trajectory occurs at high field strength. The higher electric field strengths in the p-type region reduce recombination and produce high drift velocities of the holes, which are mainly responsible for the signal formation. This is why rear (n-side) particle injection into such an n-type silicon detector results in a greater variation of the pulse shape with particle species and particle injection into such an n-type silicon detector used in these studies were manufactured (by Eurisys Mesures, France or by Micron, England) in float zone technique from n-type silicon. The elements have large (~5 cm\(^2\)) sensitive areas and high resistivity (~8000 \( \Omega \text{cm} \)). Since good time resolution with constant-fraction discrimination (CFD) is prerequisite for this time-of-flight (TOF) application, the CHIMERA Si detectors were configured for front-side injection [16]. As discussed above, such front-side injection of the particles is not ideal for optimum particle ID. Nevertheless, because of the potentially significant gains in utility of the CHIMERA silicon detectors for heavy-ion induced reaction studies, the present work was undertaken to study feasibility and optimum performance of simultaneous applications of PSD and TOF techniques.

II. EXPERIMENTAL METHOD

A. Off-line PSD simulation

Off-line PSD simulation and rise time calibration have been carried out in the Detector Test laboratory of INFN in Catania using commercially available NIM electronics, including a precision pulser generator (BNC model PB-4). A block diagram of the electronics is shown in Fig.1.

![Fig.1- Electronic setup used for both, off–line pulse rise time calibration and in-beam pulse shape discrimination (see text).](image)

Using a passive splitter, the negative input signal from the pulser is divided into two identical pulses. One of them is shaped and amplified by a spectroscopy amplifier (ORTEC 572) and serves as energy signal. The other pulse is fed into a timing filter amplifier TFA (ORTEC 474). The TFA output pulse is split again with a passive splitter. One of these signals is fed into a constant-fraction discriminator CFD (ORTEC 934) set for 20% of the rise time. This CFD is used to generate the start signal of a time-to-amplitude converter TAC (ORTEC 567). Therefore, the internal delay time for this CFD was fixed at 20 ns, as optimized in CHIMERA TOF measurements. The other CFD output pulse is split again into two pulses, which are further transformed, added, and fed into a trigger, essentially simulating a CFD with a 90% threshold. Specifically, one of the pulses is delayed by a variable amount, ranging between 20 ns and 140 ns, depending upon the pulse generator setting. The other pulse is inverted and attenuated by 10% (CAEN mod. N109). The resulting pulses are then added to-
gether, generating a crossover signal which is amplified using a fast timing amplifier (EG&G-ESN FTA410). The negative leading edge (LE) of the output signal triggers a discriminator (LeCroy model 620CL) providing the stop pulse for the TAC. This latter logic signal simulates an effective 90% CFD output signal. According to the scheme illustrated in Fig. 1, energy and time signals from the outputs of the spectroscopy amplifier and the TAC, respectively, are converted by two analogue-to-digital converters ADC (SILENA model 7423 UHS) read out by a PC based data acquisition system.

The response of the PSD setup (Fig. 1) to negative input pulses of 40-mV and 400-mV amplitude and the effect of changes in various parameters of the setup have been studied, and a calibration was obtained for the TAC in terms of the 20%-90% rise time of the primary input pulses. Pulses from the precision pulse generator had rise times between 50 ns and 500 ns, but a fixed decay time of 200 μs. With the TFA parameters set to 20 ns integration time (INT) and 20 ns differentiation time (DIFF), the TAC output showed a remarkably linear response to the input pulse rise times (50 ns - 500 ns). The TAC output was also carefully measured for the same range of primary input pulse rise times and TFA differentiation times varied from 50 ns - 500 ns, while the TFA integration time was held constant at 20 ns.

These simulation experiments demonstrate that the setup shown in Fig. 1 permits one to measure the rise time of primary input pulses in a wide dynamic range, where the TAC output pulse amplitude is a calibrated linear function of primary input rise time. For example, for the settings INT=20 ns, DIFF=50 ns, and “internal delay” of the “90% CFD” fixed at 20 ns, one obtains the relation

\[
\text{Time}(\tau) \approx a \times \tau + b
\]

between TAC output amplitude Time and primary rise time \(\tau\) of a pulse produced by the FTA module. Here, the calibration parameters turned out to be \(a = 1.2\) and \(b = 35.0\) ns. Plausibly, changing the internal delay of the “90% CFD” affects the calibration parameters \(a\) and \(b\). However, for purposes of the present analysis, knowledge about absolute rise times are not needed. The parameter Time is only used as useful scale parameter for particle identification.

**B. In-beam PSD measurements**

The set up illustrated in Fig. 1 was also used to study PSD in in-beam experiments. These measurements were carried out with \(^{19}\text{F}\) beams of 85.75 and 95 MeV from the Tandem Accelerator of the LNS Catania and a \(^{12}\text{C}\) target. Two large area (~20 cm\(^2\)) “two-Pad” silicon detectors of the CHIMERA array [16] were tested, coupled to low-noise, charge-sensitive preamplifiers especially developed for this purpose [8]. The detectors were placed inside the LNS 2000-mm diameter scattering chamber aligned at angles of 10° and 8° with respect to the beam direction. Detector configurations with front-side and rear-side injection were explored. The experimental set up was similar of the one described elsewhere [4].

The measurements included a study of the influence of the detector bias on PSD performance. In a typical run, one of the employed detectors (D1) at room temperature was biased to the full depletion value of ~38 V (nominal) resulting in a leakage current of about 0.6 μA. PSD spectra were then taken for different bias voltages. Within a range of ± 15% of the full depletion voltage, no appreciable change resulted in the particle-ID energy threshold demarking the domain of satisfactory Z-resolution. Therefore, all other studies of PSD with this detector (D1) were carried out with the bias set at the nominal voltage of ~38 V.

The set up of the timing circuitry, including timing amplifiers and constant fraction discriminators, were kept within the experimental target area, at a distance of about 2 m from the detector. The split preamplifier and CFD digital signals were transmitted via 30-m long 50-Ω cables to the data acquisition site, where they were used for energy and time measurements, respectively.

In-beam studies performed at 85.75 MeV detected particles produced in \(^{19}\text{F} + ^{12}\text{C}\) reactions. Detector D1 was placed at an angle of 10° with respect to the beam direction fully exposed to the scattered beam from the target. The stability of the beam current around 500 pA was monitored by a Faraday cup placed at about 2m from the target. Typical signals generated from elastically scattered \(^{19}\text{F}\) beam particles had rise times of about 40 ns, as observed with a 400-MHz oscilloscope (TEKTRONIX-2467B). In these measurements, the TFA integration time was kept constant at 20 ns, while other parameters were varied. For example, the TFA differentiation time was changed to 50 ns, and the effective internal delay of the “90% CFD” was varied by 20 ns. The TAC full range was kept at 200 ns in all measurements illustrated in the figures shown below.

A PSD spectrum (energy vs. time scatter plot) obtained in front-side injection mode is shown in Fig. 2. Placed at a dis-
tance of 40 cm from the target, the detector covers an angular range of ±3.5°, which corresponds to a wide acceptance for this inverse-kinematics scattering. The main feature of Fig. 2 is an intense ridge of yield associated with elastically and inelastically scattered \(^{19}\text{F}\) projectiles. In addition, ridges due to reaction products (O, N, C, B, Li, and \(\alpha\)) are clearly discernible in this plot. At low energies, events distorted by edge effects and background are visible in this figure as well.

Energy calibrations for this and other plots (33 - 41 keV/ch) shown below were done using the maximum energy deposit in the detector by “punch-through” alpha particles and the energy of the elastic \(^{19}\text{F}\) peak at the mean detector angle, corrected for the pulse height defect. Time calibrations of the TAC (54 -105 ps/ch) were carried out with cable delays.

To reduce changes in kinematics over the detector acceptance and variations in detector thickness, as well as to eliminate edge effects, a 1-cm diameter aperture was placed in front of the detector. Evidently, effects of the electric capacitance on spectral characteristics of noise and signal shape are not altered by an aperture.

Improved performance of the detector D1 with an acceptance limited by the above aperture is illustrated by the PSD plots of Figs. 3a and 3b. The data shown in these plots correspond to particles from the same reaction \((^{19}\text{F} + ^{12}\text{C})\), but at a slightly higher beam energy (95 MeV) and at an angle of 8° relative to the beam direction. As before, detector D1 was operated with front-side injection. PSD spectra were recorded with a TFA integration time of 20 ns and a differentiation time of 50 ns. The internal delay of the “90% CFD” was varied from 20 ns to 100 ns. Data of Fig.3a and Fig.3b were obtained with this internal delay set to 20ns and 60ns, respectively. The 60-ns internal delay corresponds to the best defined identification pattern. Figure 3a differs from Fig. 2 essentially unchanged. Therefore, one concludes that the size of the sensitive detector area does largely not influence the identification pattern. However, for consistency in the following only measurements are shown featuring apertures in front of the detectors.

Another CHIMERA detector (D2) of approximately the same size as D1 was tested for PSD performance. D2 was operated at room temperature with bias voltage of – 40V resulting in a leakage current of ~ 0.16 μA. The detector was placed at an angle of 8° with respect to the beam direction and with its rear side facing the target, i.e., in a geometry for rear-side injection. Detector acceptance was again defined by an aperture of 1-cm diameter. Particles from the \(^{19}\text{F} + ^{12}\text{C}\) reaction at 95 MeV were measured again in this setup. PSD spectra were recorded with a TFA differentiation time of 100 ns and two settings (125 ns and 147 ns) of the internal delay of the “90% CFD”. The TAC full range was increased to 500 ns.

A PSD spectrum taken with the above (rear-side injection) configuration and a 147-ns internal delay is depicted in Fig. 4. Comparison of the patterns of yield ridges corresponding to different particles injected in the front (Figs. 3) with that of rear-side injection (Fig. 4) suggests slightly better resolution and more linear particle-ID response for the latter. Such behavior is expected from the different balance between ionization density and recombination discussed previously. However, the quality of particle identification achieved with these detectors is surprisingly similar in both cases.
III. RESULTS AND DISCUSSIONS

In spite of the high background contribution in the matrix of energy vs. time shown in Fig. 2, the different particles produced in the $^{19}$F + $^{12}$C reaction are well identified, even when no apertures are used and the detector is fully exposed to the scattered beam. Energy thresholds for particle-ID for the well identified particle species, such as nitrogen and carbon, were determined from the intersection of different ridges in the 2-D spectra (Fig. 2). The determination of the particle-ID energy threshold for oxygen is complicated because the corresponding yield is heavily contaminated by tails of the intense ridge of elastically scattered fluorine ions. Contributions from background and edge events were reduced considerably with the use of apertures in front of the detector. Overall quality and separation of the identification patterns are enhanced by such a measure, as demonstrated by Figs. 3a and 3b relative to Fig.2.

Earlier, Mutterer et al. [15] reported on the particular efficacy of a PSD technique using rear-side injection of particles into n-TD and other silicon detectors. These measurements used particles from the $^{14}$N + $^{12}$C reaction at 150 MeV, which had somewhat higher energies and detectors of smaller dimensions than those of the CHIMERA array considered in the present work. It is therefore difficult to compare the performance of the present PSD method directly with that employed by Mutterer et al. [15].

Notice that, in the present study of the $^{19}$F + $^{12}$C reaction at 95 MeV, the energies of oxygen, nitrogen and carbon ions determined from different sets of data agree with each other to within ± 3 MeV. Average energies of the oxygen, nitrogen and carbon ions determined from the various sets of data are 71, 65, and 59 MeV respectively. There are some differences in the PSD response between rear-side and front-side injection. For example, the two injection methods lead to differences in the full range of pulse rise times, and relative rise times for detected particles of about a factor three were measured in the two configurations. However, no significant difference in the quality of the PSD identification was observed, as measurements yielded comparable particle-ID energy thresholds (cf. Table I and Table II). For front-side particle injection into the present large-area detectors, one observes an average particle-ID energy threshold of ~3.44 ± 0.05 MeV/nucleon. Best results were obtained with an internal delay of 60 ns for the “90% CFD.” This optimum internal delay coincides with that determined for rear-side particle injection.

Average particle-ID energy thresholds of the order of ~3.44 MeV/nucleon determined in this work for both injection types are not unusual for low-energy reactions. Mutterer et al. [15] observed a somewhat lower particle-ID threshold of 2.5 MeV/nucleon with a smaller area (5 cm$^2$) n-TD silicon detector. It is noteworthy however, that in the present work the above figure of merit has been achieved for 5-times larger, CHIMERA-type detectors manufactured from normal silicon, and without compromising good timing resolution (front-side entry).

<table>
<thead>
<tr>
<th>Atomic Number</th>
<th>Threshold (MeV/nucleon)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>90%-CFD delay =20ns</td>
</tr>
<tr>
<td>6</td>
<td>3.75</td>
</tr>
<tr>
<td>7</td>
<td>3.70</td>
</tr>
<tr>
<td>8</td>
<td>3.75</td>
</tr>
</tbody>
</table>

TABLE II: Results for particle-ID energy thresholds: Rear side injection
TFA settings: INT=20 ns, DIFF=100 ns (see text)

<table>
<thead>
<tr>
<th>Atomic Number</th>
<th>Threshold (MeV/nucleon)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>90%-CFD delay=125ns</td>
</tr>
<tr>
<td>6</td>
<td>3.44</td>
</tr>
<tr>
<td>7</td>
<td>3.49</td>
</tr>
<tr>
<td>8</td>
<td>3.51</td>
</tr>
</tbody>
</table>

Fortunately therefore a detector set up with front side injection is suggested that allows for an accurate measurement of the time of flight (TOF) with particle identification, based on simultaneous measurement of the pulse rise time. This finding opens up the possibility for an upgrade of just the CHIMERA electronics to achieve an efficient simultaneous charge and mass identification of low-energy particles with this 4π detector. Evidently, it is necessary to develop compact electronics circuitry incorporating the essential features of the set up used in the present measurements (cf. Fig.1) for the almost 1200 CHIMERA preamplifier output lines [11].
The present work has studied application of a PSD particle-ID method for heavy-ion reactions at relatively low bombarding energy. In fact, the particle-ID energy threshold is the most crucial parameter to be taken into account in future applications. However, additional studies with this PSD set up are needed of reactions at higher bombarding energies, in order to determine the performance of the method also for higher-energy particles in a broader range of atomic numbers. In particular, it will be very important to identify particle mass by a TOF method using pulsed beams with good timing properties. The aim of such experiments is to achieve low energy thresholds for both atomic and mass number identification, especially in the region of intermediate masses (10 < A < 50).[17]

IV. CONCLUSION

In this work, an efficient PSD method has been developed for large-area, n-type (implanted) silicon detectors, which are especially configured for fast-timing applications. The method is based on the rise time measurement of charge signals obtained from fast preamplifiers with broad dynamic range. Rise time information was obtained from the difference in response times of two constant-fraction discriminators triggering at different pulse levels (fractions). Rear-side and front-side particle injection have produced particle ID of similar quality.

V. ACKNOWLEDGMENT

The authors would like to thank the operations crew of the LNS Tandem accelerator for the efficient and smooth operation of the accelerator, and to Drs. V. Campagna, A. Di Stefano, and S. Salomone for their assistance during the experiment. One of us (MBC) is grateful to the INFN, Sezione di Catania and the Physics Department of the University of Catania, for the financial support, and to the Laboratori Nazionali del Sud for the hospitality extended to him. In addition, two of us (WUS and JT) acknowledge support by the US DOE Grant No DE-FG02-88ER40414.

VI. REFERENCES

Decontamination of Tritiated Steel Samples

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Abstract

Tritiated stainless steel samples were decontaminated using several techniques. The tritium content was measured prior to inert gas purging with a surface activity monitor. The samples were purged in a linear desorption furnace under He gas flow. The tritiated compounds removed from the stainless steel were trapped in bubblers and the corresponding activity was measured. The properties of these compounds and their binding to the stainless steel were explored.

I. Introduction

Although decontamination of contaminated metal samples has been studied before [1, 2], a systematic study of tritium decontamination of such samples is still lacking. This is an important problem because the release of tritiated compounds from contaminated steel poses a health and safety problem for decommissioning fusion research facilities. The JET facility in Culham, UK is one such institution. This work will set the basis for a series of decontamination experiments conducted on behalf of the decommissioning project for this facility.

In addition to providing experimental basis for testing decontamination procedures this work has sought to explore the physical and chemical processes involved in the physic/chemi-sorption and desorption of tritiated species. The use of a new automated tritium detector, has opened the possibility of conducting thermal
spectroscopy of the desorption process. It is hoped that this spectroscopy will provide clues to the nature of binding of these absorbed species, be it chemisorption or physisorption.

The experiments conducted here rely on two apparatus to verify and dynamically measure the tritium content and character of specially contaminated stainless steel samples. A tritium surface activity monitor (SAM) is employed to make a rough estimate of the surface activity [3]. A linear desorption apparatus similar to those described by: Antoniazzi and Shmayda [1] and Shmayda, Antoniazzi and Surette [2]. These previous linear desorption experiments demonstrated the presence of organic species after the stainless steel samples were contaminated with pure T2. The use of a new device made by IN/US Systems has allowed dynamic measurements never previously possible. This has contributed to the possibility of further understanding the properties of the desorbed species, and possibly measuring their individual binding affinities for the steel.

II. Experimental

Tritiated 303 stainless steel samples, of dimensions 2cm x 5cm x 0.5cm and commonly referred herein as coupons, were removed from packaging under an inert He atmosphere. Prior to thermal desorption, the tritium content of the coupons was measured using the tritium surface activity monitor.

The desorption experiment apparatus consists of four bubblers and two furnaces. A He carrier gas flows into the first furnace, through two bubblers(#1-#2) connected to its outlet, and into the second furnace. Upon exiting the second furnace the gas flows through two more bubblers (#3-#4) and then out a fume hood stack.

The first furnace is a quartz tube wrapped in a heating coil. Controlled current is supplied to the coil and it resistively heats the quartz linearly from room temperature to 950 ºC. Bubbler #1, the first bubbler after the linear desorption furnace, was filled with IN/US scintillation cocktail and equipped with an IN/US β-RAM detector. This detector allows constant, automated sampling of the activity in bubbler #1. Bubblers#2-4 were filled with DI H2O. Bubbler #2 was placed in series after bubbler #1, such that the gas overflow from bubbler #1 flows through bubbler #2 and then subsequently to the Cu/CuO
oxidation furnace. The Cu/CuO furnace is similar in construction to the linear desorption furnace except that the quartz tube is filled with Cu/CuO ribbon. The oxidation furnace is kept at a constant 750 °C during all experiments. The carrier gas and any compounds that flow through this furnace are oxidized by the CuO. Bubbler #3 was placed immediately after the Cu/CuO oxidation furnace. Bubbler #4 was placed in series after bubbler #3 to catch the overflow of tritiated species in a manner similar to bubbler #2. The outlet from bubbler #4 was directed up a fume hood stack.

Samples from bubblers #2-4 consist of 0.5 mL aliquots. Unlike the samples taken from bubbler #1, these aliquots are never returned to their respective bubblers.

A series of background readings from each of the bubblers were taken prior to inserting the sample into the desorption furnace, starting gas flow, and beginning a linear temperature ramp. The background samples taken from bubblers #2-4, were extracted every five minutes adhering to the following regimen. The β-RAM detector started to pump and measure the instantaneous activity of bubbler #1. Five minutes after this the tritiated stainless steel coupon was inserted into the center of the linear desorption furnace. Five minutes after insertion of the coupon sample #1 from the H₂O bubblers was taken. Samples #2-4 were subsequently taken every five minutes. Sample #2 was taken concurrently with the start of carrier gas flow. Sample #4 was taken concurrent with the start of the linear temperature ramp of the linear desorption furnace. These four samples from bubblers #2-4 constitute the background readings of these bubblers, while the accumulation of real time data from bubbler #1 over the initial span of 25 minutes serves as the background readings for this bubbler.

After starting the linear temperature ramp of the linear desorption furnace, the intervals with which samples from bubblers #2-4 are taken depends on the ramp rate. Table 1 below notes which intervals correspond to which ramp rates.

<table>
<thead>
<tr>
<th>Ramp Rate (°C/min)</th>
<th>Sample Interval (min)</th>
</tr>
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<tbody>
<tr>
<td>10</td>
<td>5</td>
</tr>
<tr>
<td>5</td>
<td>10</td>
</tr>
<tr>
<td>2.5</td>
<td>15</td>
</tr>
<tr>
<td>1.5</td>
<td>20</td>
</tr>
</tbody>
</table>
Samples were taken according to this scheme until the ramp had reached its maximum temperature. The samples taken by hand were counted in a Wallac scintillation counter. After the coupon had cooled its tritium content was measured with the surface activity monitor.

### III. Results

The raw data taken from 12 coupons is included in Appendix 1. The raw data is presented in plots of both the total activity of bubblers #1-4 as a function of temperature in the linear desorption furnace, and the trapping efficiency of bubblers #1 and #3, calculated from the measured overflow of activity to the respective secondary bubblers. The data from coupons #1-4 was taken by hand. Data from coupons #5-12 was in part by hand, but involved the use of an IN/US β-ramp scintillation counter. The measurable increase in the quality of data taken with this device is easily observed in Appendix 1.

Appendix 2 contains notes to the figures of Appendix 1. In some cases they remark on sampling errors. In other cases, they mention the exact experimental conditions that lead to characteristic observations.

Comprehensive data collected from coupons 1-12 are presented in Table 2 below. The coupons are presented in the order in which they were desorbed. This table notes the total amount of water soluble and water insoluble tritiated compounds desorbed from each compound. It also notes the amount of tritium measured by the surface activity monitor prior to desorption.

<table>
<thead>
<tr>
<th>Run</th>
<th>ºC/min</th>
<th>HTO (µCi)</th>
<th>HT (µCi)</th>
<th>Total (µCi)</th>
<th>SAM (µCi)</th>
<th>HTO/Total</th>
<th>HT/Total</th>
<th>Comments:</th>
</tr>
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<tr>
<td>1</td>
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<td>665.4417</td>
<td>175.8867</td>
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<td>632.4</td>
<td>0.790942</td>
<td>0.209058</td>
<td>Inert Purge, 50 SCCM He</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
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<td>112.7086</td>
<td>802.614</td>
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<td>3</td>
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<td>92.49429</td>
<td>1423.775</td>
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<td>0.064964</td>
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<tr>
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<td>137.1982</td>
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<tr>
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<td>5</td>
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<td>69.54361</td>
<td>1149.456</td>
<td>763.98</td>
<td>0.939499</td>
<td>0.060501</td>
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<tr>
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<td>2.5</td>
<td>1017.621</td>
<td>102.8001</td>
<td>1120.421</td>
<td>812.94</td>
<td>0.908249</td>
<td>0.091751</td>
<td>Inert Purge, 50 SCCM He</td>
</tr>
<tr>
<td>7</td>
<td>2.5</td>
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<td>52.92961</td>
<td>997.9626</td>
<td>661.98</td>
<td>0.946962</td>
<td>0.053038</td>
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<td>8</td>
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<td>1085.894</td>
<td>33.67022</td>
<td>1119.564</td>
<td>841.5</td>
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<td>34.64767</td>
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Outright comparison of coupons is difficult because of the variance in total activity. This is believed to have been the result of packaging prior to shipment. One can note the relatively constant ratio of water soluble species, believed to primarily be HTO, to water insoluble species, believed to be either HT or some light organics.

The experimental procedures and apparatus were tested with respect to ramp rate, time out of controlled atmosphere, and consistency. Figures 1, 2 and 3 below demonstrate the dependence of these variables on the total activity measured.

![Figure 1. Total Coupon Activity vs. Ramp Rate](Image)

The data in Figure 1 indicate that there is no dependence on the ramp rate. Thus one can assume that the quality of the measurement of the total activity is independent of the ramp rate.

![Figure 2. Total Coupon Activity vs. Time Outside Controlled Atmosphere](Image)
Figure 2 demonstrates the lack of correlation between the total activity of the coupons and the time they spent out of a controlled He atmosphere. This indicates that the coupons were shipped already having varying amounts of absorbed tritium.

![SAM, Thermal Desorption Correlation](image)

Figure 3. Surface Activity Monitor Measurements vs. Thermal Desorption Measurements

Figure 3 illustrates the correlation of the measurements made by the tritium surface activity monitor and the thermal desorption apparatus. The relationship should be linear and the correlation coefficient $R^2=0.86$ indicates a reasonable fit given the first order approximation used to calculate the activity measured with the surface activity monitor. This calculation is included in Appendix 3. Thus one can conclude that the data presented in Table 2 is free of systematic experimental error.

Normalizing the data with respect to the total activity allows one to compare the data taken from bubbler #1. Figure 4 is a plot of the normalized activity of bubbler #1. It contains data from all twelve coupons.
Figure 4 illustrates the characteristic increase in activity as tritiated compounds are released from heated coupon surface. Data from bubblers #2 and #4 represent the trapping efficiency of bubblers #1 and #3 respectively and cannot be normalized without loss of any systematic trends arising from the change in solvents. Composite data from these two bubblers are presented in Figures 5 and 6.

Figure 5 demonstrates the change in trapping efficiency that occurs when the solvent in bubbler #1 is varied. The trends are not normalized, but one can clearly see the low amount of activity carried over from the desorption of the first four coupons. Bubbler #1 contained 185 mL H$_2$O for the first four desorption experiments but was then switched to scintillation cocktail to allow use of the β-ram device.
Figure 6. Total Activity of Bubbler #4 as a Function of Temperature, inset key refers to the coupon number

Figure 6 shows the very small amount of spillover from bubbler #3. Coupon #12 shows an extremely large amount of spillover relative to the previous runs. This may have resulted from inadequate preparation of the Cu/CuO oxidation furnace. The data from bubbler #3 cannot be normalized because of sampling errors.

A more important form of the data collected is the relationship between change in total activity and the temperature of the linear desorption furnace. By plotting this relationship one hopes to resolve peaks specific to the desorption of individual compounds from the coupon. Figure 7 below illustrates one example curve.

Figure 7. Normalized, Smoothed Change in Total Activity as a Function of Temperature, Bubbler #1, Coupon #10, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 7 indicates that the integration curves presented in Appendix 1 have structure evident in the plateaus spanning approximately 100 °C. This structure is related to the different compounds and different rates of desorption at a given temperature.

IV. Discussion

From the results presented above one can make two conclusions. The first is that the coupons were not prepared uniformly. The coupons were shipped in a sealed container that maintained an inert He atmosphere. However, they were individually wrapped in aluminum foil. Human error contributed during the process of individually wrapping the coupons is currently suspected as the reason for the observed variance in surface activity. This is suspected because of the experimental setup was tested for systematic error and found to be free of significant inaccuracies. Two independent measurements confirm the variance in total coupon activity.

The second conclusion is that the current experimental method can be used to study the out-gassing of tritiated compounds from stainless steel with resolution sufficient enough to resolve the differences in binding affinity the compounds. The structure observed in the differentiated integration curves indicates differences in out-gassing rates at different temperatures. When combined with the constantly increasing temperature ramp one can conclude that different processes are occurring. Further study is underway in this area and the hope remains to associate the temperature of the peaks seen in Figure 7 with the binding energy of the species they represent.

As the investigation into the binding affinity of these tritiated compounds to stainless steel continues, the experimental method has also proven sufficient enough to begin work on testing specific decontamination procedures. The hope is that by combining the knowledge gained from the thermal desorption spectroscopy of the tritiated contaminants and the initial tests of decontamination procedures will suggest an effective procedure to be used in the decommissioning of future fusion facilities.
V. Conclusion

Effective experimental methods have been developed to test the tritium content of stainless steel and perform thermal desorption spectroscopy on the compounds contained therein. This sets the groundwork for testing and developing better decontamination procedures needed for the future decommissioning of fusion technology facilities.

References:


Appendix 1: Raw Data

Figure 1a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #1, He gas, 50 SCCM, 10 °C/min Ramp Rate

Figure 1b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #1, He gas, 50 SCCM, 10 °C/min Ramp Rate
Figure 1c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #1, He gas, 50 SCCM 10 °C/min Ramp Rate

Figure 1d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #1, He gas, 50 SCCM 10 °C/min Ramp Rate
Figure 1e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #1, He gas, 50 SCCM, 10 °C/min Ramp Rate

Figure 1f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #1, He gas, 50 SCCM, 10 °C/min Ramp Rate
Figure 2a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #2, He gas, 50 SCCM, 10 °C/min Ramp Rate

Figure 2b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #3, He gas, 50 SCCM, 10 °C/min Ramp Rate
Figure 2c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #2, He gas, 50 SCCM, 10 °C/min Ramp Rate

Figure 2d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #2, He gas, 50 SCCM, 10 °C/min Ramp Rate
Figure 2e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #2, He gas, 50 SCCM, 10 °C/min Ramp Rate

Figure 2f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #2, He gas, 50 SCCM, 10 °C/min Ramp Rate
Figure 3a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #3, He gas, 50 SCCM, 5 °C/min Ramp Rate

Figure 3b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #4, He gas, 50 SCCM, 5 °C/min Ramp Rate
Figure 3c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #3, He gas, 50 SCCM, 5 °C/min Ramp Rate

Figure 3d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #3, He gas, 50 SCCM, 5 °C/min Ramp Rate
Figure 2e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #3, He gas, 50 SCCM, 5 ºC/min Ramp Rate

Figure 2f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #3, He gas, 50 SCCM, 5 ºC/min Ramp Rate
Figure 4a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #4, He gas, 50 SCCM, 5 °C/min Ramp Rate

Figure 4b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #4, He gas, 50 SCCM, 5 °C/min Ramp Rate
Figure 4c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #4, He gas, 50 SCCM, 5 °C/min Ramp Rate

Figure 4d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #4, He gas, 50 SCCM, 5 °C/min Ramp Rate
Figure 4e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #4, He gas, 50 SCCM, 5 ºC/min Ramp Rate

Figure 4f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #4, He gas, 50 SCCM, 5 ºC/min Ramp Rate
Figure 5a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #5, He gas, 50 SCCM, 5 °C/min Ramp Rate

Figure 5b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #5, He gas, 50 SCCM, 5 °C/min Ramp Rate
Figure 5c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #5, He gas, 50 SCCM, 5 °C/min Ramp Rate

Figure 5d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #5, He gas, 50 SCCM, 5 °C/min Ramp Rate
Figure 5e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #5, He gas, 50 SCCM, 5 °C/min Ramp Rate

Figure 5f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #5, He gas, 50 SCCM, 5 °C/min Ramp Rate
Figure 6a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #6, He gas, 50 SCCM, 2.5 °C/min Ramp Rate

Figure 6b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #6, He gas, 50 SCCM, 2.5 °C/min Ramp Rate
Figure 6c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #6, He gas, 50 SCCM, 2.5 °C/min Ramp Rate

Figure 6d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #6, He gas, 50 SCCM, 2.5 °C/min Ramp Rate
Figure 6e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #6, He gas, 50 SCCM, 2.5 °C/min Ramp Rate

Figure 6f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #6, He gas, 50 SCCM, 2.5 °C/min Ramp Rate
Figure 7a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #7, He gas, 50 SCCM, 2.5 °C/min Ramp Rate

Figure 7b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #7, He gas, 50 SCCM, 2.5 °C/min Ramp Rate
Figure 7c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #7, He gas, 50 SCCM, 2.5 °C/min Ramp Rate

Figure 7d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #7, He gas, 50 SCCM, 2.5 °C/min Ramp Rate
Figure 7e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #7, He gas, 50 SCCM, 2.5 °C/min Ramp Rate

Figure 7f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #7, He gas, 50 SCCM, 2.5 °C/min Ramp Rate
Figure 7g. Fit to Asymptotic Behavior of Activity as a Function of Temperature, Coupon #7, He gas, 50 SCCM, 2.5°C/min Ramp Rate

Figure 7h. Fit to Behavior of Activity as a Function of Temperature, Coupon #7, He gas, 50 SCCM, 2.5°C/min Ramp Rate
Figure 8a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #8, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 8b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #8, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 8c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #8, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 8d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #8, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 8e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #8, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 8f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #8, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 8g. Total Activity of Bubbler #1 as a Function of Time, Coupon #8, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Total Activity in Bubbler #1 as a Function of Time
Coupon #8, 50 SCCM He, 1.5 deg C/min
(Ramp Breaks Down at ~400 Deg C; ~300 min)
Figure 9a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #9, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 9b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #9, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 9c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #9, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 9d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #9, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 9e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #9, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 9f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #9, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 10a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #10, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 10b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #10, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 10c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #10, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 10d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #10, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 10e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #10, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 10f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #10, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 11a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #11, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 11b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #11, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 11c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #11, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 11d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #11, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 11e. Trapping Efficiency of Bubbler #1 as a Function of Temperature, Coupon #11, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 11f. Trapping Efficiency of Bubbler #3 as a Function of Temperature, Coupon #11, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 12a. Total Activity of Bubbler #1 as a Function of Temperature, Coupon #12, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 12b. Total Activity of Bubbler #2 as a Function of Temperature, Coupon #12, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 12c. Total Activity of Bubbler #3 as a Function of Temperature, Coupon #12, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 12d. Total Activity of Bubbler #4 as a Function of Temperature, Coupon #12, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Figure 12e. Trapping Efficiency of Bubbl er #1 as a Function of Temperature, Coupon #12, He gas, 50 SCCM, 1.5 °C/min Ramp Rate

Figure 12f. Trapping Efficiency of Bubbl er #3 as a Function of Temperature, Coupon #12, He gas, 50 SCCM, 1.5 °C/min Ramp Rate
Appendix 2: Notes on Raw Data

Figure 1a: This was the first experiment performed with the setup. Here bubbler #1 is filled with 185 mL distilled H₂O. The samples were taken every 5 min by hand and later matched to the desorption oven temperature.

Figure 1c: The point around 750 ºC is a sampling error. The curve also shows that the copper, copper oxide oxidation furnace is contaminated with tritium. In order to fix the data one could subtract the activity collected before the plateau at 300 ºC as the background.

Figure 1d: This curve also shows the contamination of the copper, copper oxide furnace via the trapping inefficiency of bubbler #3.

Figure 1e: The trapping efficiency levels off at 99% which means that almost all of the water soluble activity that passes through bubbler #1 remains there.

Figure 1f: Again the trapping efficiency levels off at 99%. We expect this because the bubblers have the same construction and are filled with the same material (185 mL distilled H₂O).

Figure 2a: This was the second desorption measurement. There were less sampling errors. These measurements were taken by hand and bubbler #1 was filled with 185 mL distilled H₂O.

Figure 2c: The curve still shows that the copper, copper oxide furnace is contaminated, but to a much lesser extent. This is because furnace was flushed with compressed air overnight between the desorption of coupon #1 and coupon #2. The error could be fixed by subtracting the background accumulated up to 500ºC.

Figure 2d: The activity of bubbler #4 is much less due to the relative decontamination of the oxidation furnace.

Figure 2e: These are the expected values for bubbler #1 filled with 185 mL distilled H₂O.

Figure 3a: Bubbler #1 was filled with 185 mL distilled H₂O and sampled by hand.

Figure 3c: The data point at 870 ºC is a sampling error and can be excluded.

Figure 4a: Bubbler #1 was filled with 185 mL distilled H₂O and sampled by hand.
Figure 4c: The data point at 540 ºC is a sampling error and could be deleted.

Figure 5a: Bubbler #1 filled with 185 mL IN/US systems scintillation cocktail. Measurements made with IN/US β-ram.

Figure 5e: The trapping efficiency looks very similar to the cases where distilled H₂O is in bubbler #1, but the loss of activity from bubbler #1 to bubbler #2 can be clearly seen in Figure 5b.

Figure 6a: Bubbler #1 was filled with 160 mL IN/US systems scintillation cocktail and measurements were taken with an IN/US systems β-ram.

Figure 6d: The data point at 877 ºC is a sampling error and can be ignored. Data points from 22-428 ºC are negative because of poorly sampled background readings.

Figure 6e: The reason this measurement recorded the lowest trapping efficiency of 96% was because bubbler #1 was only filled with 160 mL of scintillation cocktail.

Figure 7a: Bubbler #1 was filled with 185 mL IN/US systems scintillation cocktail and sampled with an IN/US systems β-ram.

Figures 7g-h: The fit to the asymptotic behavior of Bubbler #1 shows a steady loss of activity. A fit to the data for the same temperature range from Bubbler #2 shows a constant gain of activity. Equation (1.1) below compares the amount of activity lost from Bubbler #1 to the amount gained by Bubbler #2.

\[
\begin{align*}
\Delta A_1 &= -0.05 \times \Delta T = -0.05 \times (950 - 600) = -17 \\
\Delta A_2 &= 0.04 \times \Delta T = 0.04 \times (950 - 600) = 14
\end{align*}
\]

(1.1)

Based on this crude approximation we can say that all the activity leaving Bubbler #1 is measurably trapped by Bubbler #2.

Figure 8a: Bubbler #1 was filled with 185 mL IN/US systems scintillation cocktail and sampled with an IN/US systems β-ram.

Figure 9a: Bubbler #1 was filled with 185 mL IN/US systems scintillation cocktail and sampled with an IN/US systems β-ram.

Figure 9c: The data point at 857 ºC is a sampling error and can be ignored.

Figure 10a: Bubbler #1 was filled with 185 mL IN/US systems scintillation cocktail and sampled with an IN/US systems β-ram.
**Figure 11a:** Bubbler #1 was filled with 185 mL IN/US systems scintillation cocktail and sampled with an IN/US systems β-ram. The linear thermal desorption furnace for this coupon was ramped from 20 ºC to 600 ºC.

**Figure 11d:** The data point at 108 ºC is a sampling error and can be ignored.

**Figure 12a:** Bubbler #1 was filled with 185 mL IN/US systems scintillation cocktail and sampled with an IN/US systems β-ram. The linear thermal desorption furnace for this coupon was ramped from 20 ºC to 600 ºC.

**Appendix 3:**

The following equation describes the approximation used to calculate the activity measured by the tritium surface activity monitor.

\[
    n \approx \frac{2\omega I}{\epsilon}
\]

- \(I\) - current generated by decay
- \(\omega\) - energy required to form an ion pair in air (33.7 eV / ion pair)
- \(\epsilon\) - mean β particle energy (5.7 keV)
- \(n\) - number of atoms in near surface

The factor of two accounts for ½ of the electrons being lost in the surface
IV. Facsimiles of Selected Papers in Print
Morphing of the Dissipative Reaction Mechanism

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Abstract

Important trends in the evolution of heavy-ion reaction mechanisms with bombarding energy and impact parameter are reviewed. Essential features of dissipative reactions appear preserved at E/A = 50-62 MeV, such as dissipative orbiting and multi-nucleon exchange. The relaxation of the A/Z asymmetry with impact parameter is slow. Non-equilibrium emission of light particles and clusters is an important process accompanying the evolution of the mechanism. Evidence is presented for a new mechanism of statistical cluster emission from hot, metastable primary reaction products, driven by surface entropy. These results suggest a plausible reinterpretation of multi-fragmentation.

1 INTRODUCTION

The evolution of the heavy-ion reaction mechanisms with bombarding energy from the dissipative regime [1] towards higher energies (E/A ∼ 20 MeV → 100 MeV) is characterized by the phenomenon of copious emission [2] of intermediate-mass nuclear cluster fragments (IMF). Such clusters are defined by atomic numbers approximately in the range 4 = Z ≤ 20 for heavy-ion reactions such as 197Au+36Ar, 197Au+36Kr, or 209Bi+136Xe. Conventional statistical models of nuclear decay predict [3] negligible probabilities for the evaporation of IMF clusters from projectile-like (PLF), target-like (TLF), or composite-nucleus (CN) type reaction products. The absence of this decay channel is understood to be due to the high Coulomb barriers (Vc ~ 50-60 MeV) for emission of clusters from a heavy nucleus, estimated to exceed even the maximum observed nuclear temperatures (T ≤ 6 MeV) by an order of magnitude. So-called multi-fragmentation, central-collision events with several substantial clusters in the exit channel (up to 12 observed) seem therefore far beyond the realm of traditional statistical nuclear decay models.

As an interesting alternative, the above cluster emission process is often attributed to a hypothetical nuclear phase transformation occurring in a new domain of mechanical instability of hot nuclear matter. Observations of limits to nuclear temperatures [4-6] have been taken as supporting evidence for such a nuclear (liquid-gas) phase transition. Its demonstration would be of interest far beyond nuclear science, because nuclei are finite quantal systems produced in heavy-ion reactions in metastable nonequilibrium states [1] decaying into vacuum.

Attempting to justify equilibrium-statistical treatment of cluster decay, theories [7-9] modeling multi-fragmentation in terms of a phase transition typically concentrate on a small cross section associated with the most central collision events, potentially leading to a hot CN. Unfortunately, such events belong to those experimentally most difficult to reconstruct. Circumventing difficulties associated with high cluster emission barriers mentioned previously, these models ascribe the observed product distributions to statistical population of hypothetical “freeze-out” volumes, in which particles and clusters interact only via Coulomb forces. However, since actual transition states for cluster emission are strongly influenced by the nuclear interaction [10, 11], the implications of data comparisons with models neglecting it are not obvious.

In view of the difficulties faced by experimental and theoretical research of multi-fragmentation, now since some two decades, it appears necessary to take a fresh look at the entire heavy-ion reaction environment and its evolution with bombarding energy and impact parameter. For some heavy systems, data are now available for a dynamic range of >50:1 in bombarding energies over the interaction barrier. As will be illustrated below, this evolution turns out to be smooth, resembling a morphing of the
well-known dissipative mechanism. Previously enigmatic phenomena such as cluster emission and limitations of thermal energy sustained by a nucleus emerge as natural expressions of nuclear response at higher energies.

2 EXPERIMENTAL SYSTEMATICS

2.1 Cross Sections

It is interesting to note that, for heavy reaction systems and the bombarding energy range of interest here (E/A=30-60 MeV), the reaction cross section remains almost constant at $\sigma_r = (5-6)b$ [12-14], tracing experimental systematics [15], even though free NN scattering cross sections decrease by factors 2-3, leading to corresponding increases in surface transparency. These figures are obtained from an analysis of the Coulomb dominated elastic-scattering angular distributions, which for reactions such as $^{197}$Au+$^{86}$Kr and $^{209}$Bi+$^{136}$Xe are of the Fresnel type still at E/A = 50-60 MeV. Within their (8-10)% experimental uncertainties, these cross sections are found consistent with direct integration of reaction events, with a PLF or its remnant as a distinctive leading particle at forward angles.

2.2 Experimental Methods

Because of the potentially high excitations, reconstruction of the main experimental observables requires an efficient measurement of the emission patterns of secondary decay products.

In the experiments reported here, neutrons, light charged particles (LCP), and IMF clusters have been measured with $4\pi$ coverage, using Rochester SuperBall [16] and St. Louis Dwarf [17] or MicroBall detector arrays. More massive PLF and/or TLF remnants are sampled with position-sensitive Si-strip detector telescopes. More details on experimental setup and performance can be found in Refs. [12-14].

Under certain conditions, an efficient $4\pi$ measurement of neutrons and LCPs allows one to reconstruct the massive primary reaction products, even if they disintegrate completely in the exit channel. Crucial is a (meta-) stability of the primary fragments that is sufficiently high, allowing acceleration to approximately asymptotic velocities to occur prior to disintegration.

Of particular interest is of course the total excitation energy $E^*$ generated in a reaction, one of the fundamental observables. As long as particle kinetic energies are relatively well known, or small compared to their binding energies in the parent nuclei, this information can be obtained already from an analysis of the particle multiplicities.

Experimental joint multiplicity distributions $P(m_{\nu}, m_{\text{LCP}})$ of neutrons and LCPs, resp., are illustrated in Fig. 1, for the $^{209}$Bi+$^{136}$Xe reaction and energies between E/A = 28 and 62 MeV. These distributions virtually overlap, hence only the 62 MeV data are depicted in detail. The characteristic shape is due to the Coulomb barriers for LCP evaporation from PLF and TLF, which introduce strong non-linearities in all LCP-$E^*$ correlations, e.g., the popular $m_{\text{LCP}}(E^*)$ relations. Here, neutron measurements resolve ambiguities.

![Figure 1](image.png)

**Figure 1:** Contour diagram (log) of the measured joint multiplicity distributions of neutrons and LCPs, for the $^{209}$Bi+$^{136}$Xe reaction at E/A=62 MeV. Average correlations for E/A=28 and 40 MeV are indicated by dotted curves.
Detailed modeling [18], verified by calibration measurements, shows that the joint multiplicity distribution $P(m_e, m_{\text{LCF}})$ is a functional of the thermal excitation energy distribution $P(E^*)$,

$$P(m_e, m_{\text{LCF}}) = \tilde{P}[P(E^*)]$$

(1)

The direction of increasing average excitation energy is indicated by the solid curve $(E^*)$ in Fig. 1. Regions in $\{m_e, m_{\text{LCF}}\}$ space correspond

in non-linear fashion to intervals in total excitation of primary massive fragments, rather independently of their multiplicity and mass splits, as long as cluster emission is not significant. Qualitatively, a cluster emission degree of freedom adds a third dimension [19] to the plot of Fig. 1, but the functional equivalent to Equ. 1 has not yet been obtained quantitatively.

The data shown in Fig. 1, together with the experimental efficiency filter, can already be used for benchmark testing of statistical decay models. As shown elsewhere [18], the statistical model MMMC [7] fails this test, due to its significant neglect of neutron phase space, while the competing SMM [8] is consistent with data.

The emission patterns of light particles emitted in a heavy-ion reaction provide important clues on the reaction mechanism. In Fig. 2 [13], simulated events are shown where $\alpha$ particles are evaporated from an accelerated TLF or PLF "source", produced in a dissipative $^{197}\text{Au} + ^{86}\text{Kr}$ reaction at $E/A = 39$ MeV. The simulation used a dissipative reaction event generator [20] modeling the NEM one-body exchange mechanism [21], followed by statistical decay model GEMINI [22]. An emitter with established kinematics is recognizable by a "Coulomb ring" pattern centered at the emitter velocity, ideally like the distribution marked "TLF" in Fig. 2. The strong distortions of the expected circular patterns seen for PLF emission is due to limited granularity and stopping power of the detectors at forward angles sensitive to this latter source.

2.3 Dissipative Reaction Dynamics

Emission patterns of projectile-like fragments in the reaction $^{209}\text{Bi} + ^{136}\text{Xe}$ at $E/A = 28$ and 62 MeV are shown in Fig. 3 as logarithmic contour plots of lab PLF kinetic energy vs. angle (top row) or fragment atomic number (bottom row). The solid and dotted lines in these plots represent simulation calculations [14] with the NEM [20, 21] correcting for sequential decay of the

*Figure 2: Invariant cross sections for sequential evaporation of $\alpha$ particles from TLF, PLF (top and middle), and the combined distribution (bottom) vs. parallel and perpendicular $\alpha$ velocity.*

*Figure 3: The $^{209}\text{Bi} + ^{136}\text{Xe}$ reaction at 2 energies. Energy-angle correlations (top row), energy-Z correlations (bottom). Solid lines indicate average predictions by the NEM, arrows the direction favored by sequential decay. [14]*
primary fragments.

The fragment energy-angle correlations depicted in the top panels of Fig. 3 illustrate the presence of 3 cross section ridges. The ridge of elastically scattered events is visible as intense horizontal pattern, while a ridge of partially damped events outlines a correlation between dissipation and forward scattering. Finally, the distribution at lowest energies is attributed to negative-angle scattering.

The top panels of Fig. 3 demonstrate a well-known dissipative-orbiting phenomenon, a hallmark of the dissipative reaction mechanism. Clearly, dissipative forces are strong still at $E/A = 62$ MeV, and there is no direct evidence that the net conservative force has become repulsive. In fact, as illustrated by the solid lines superimposed on the cross section features, NEM simulation calculations provide a good representation of the data with the set of forces and adiabatic prescriptions that reproduce trends at much lower bombarding energies.

The same reaction model, based on a diffusion-like multi-nucleon exchange process, predicts average primary PLF charges to be close to that of the projectile, $<Z_{\text{PLF}}>$ ~ $Z_{\text{proj}} = 54$ (vertical lines “NEM” in bottom panels of Fig. 3). The characteristically narrow diagonal ridges in these $E_{\text{PLF}}-Z_{\text{PLF}}$ plots are dominantly the result of evaporative decay of the primary reaction fragments proceeding in the general direction indicated by arrows in Fig. 3. This fact has been demonstrated for $^{208}\text{Bi}+^{136}\text{Xe}$, as well as for the $^{197}\text{Au}+^{86}\text{Kr}$ reaction, already at several bombarding energies, by direct reconstruction of the primary PLF-Z distributions. Unfortunately, due to significant uncertainties in the reconstruction procedure, no quantitative studies of the fluctuations in the fragment distributions are available as yet.

The reconstruction of the primary reaction fragments makes use of the fact that most particles are evaporated from these fragments in flight, revealing their origin in the corresponding invariant emission patterns (Fig. 2). In Fig. 4, some sample spectra are shown of protons (top row) and $\alpha$ particles (bottom) emitted at the indicated angles from the $^{197}\text{Au}+^{86}\text{Kr}$ reaction at $E/A = 38.7$ MeV. Solid dots represent data, while curves indicate contributions calculated with “moving-source” models assuming random evaporation from average PLF and TLF emitters, as well as app. isotropic emission of high-energy particles from a virtual intermediate-velocity source (“IVS”). Corresponding particle velocity ($v_p$) spectra can be written in invariant form as,

$$ \frac{d\sigma}{d^2v_p} \approx \frac{M_p}{4\pi} \frac{m_p}{2} \sqrt{2\gamma p - V_{\text{Coul}}} \cdot \exp \left\{ -\frac{m_p (\bar{v}_p - \bar{v})^2}{2T_s} \right\} $$

(2)

where $\bar{v}_p$ is the emitter velocity. Fit parameters include the emitter A, Z, Coulomb barrier $V_{\text{Coul}}$, and average velocity, $v_e$. For statistical emission, multiplicity $M_p$ and spectral slope parameter $T_s$ are related to emitter excitation energy $E^*$ ($T_s \sim T$). Branching ratios ($M_p$) and spectra of these particles provide good constraints on the properties of the respective primary fragments. It is interesting to observe that, like at near-barrier bombarding energies, also at the upper boundary of the Fermi energy domain, reaction partners do not have sufficient contact time to relax to equilibrium, as far as excitation energy division [12, 13, 23] or mass-density (A/Z) equilibration are concerned.

The hypothetical IVS emitter is used to represent non-equilibrium particle emission, a process that is expected [1, 24, 25] to contribute significantly at the present bombarding energies.

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**Figure 4**: Experimental (dots) proton and $\alpha$ spectra measured for the reaction $^{197}\text{Au}+^{86}\text{Kr}$ at 38.7 MeV at the lab angles indicated. The various curves represent moving-source fits to the data. [13]
The corresponding particle energy distribution can typically be described in terms of a random spectrum of higher-energy particles emerging from a kinematical center moving with an “intermediate velocity” \( v_e \) of app. 50%-70% of the beam velocity [24]. The exponential energy spectra of these latter, non-equilibrium particles correspond to logarithmic slope parameters of the order of \( E_0 = 15-20 \text{ MeV} \). Presumably, these emission patterns are caused by couplings with the intrinsic Fermi motion of the nucleons.

In any case, these \( E_0 \) parameters are so large that the associated emission process is well distinguished from slow, thermal evaporation \( E_0 = T = 6 \text{ MeV} \). The non-equilibrium particle distributions contain important information on the dynamical evolution of multi-scattering cascades within the nuclear medium, which in turn is related to the equation of state of nuclear matter and its dissipative properties.

The comparison between data and model fits illustrated in Fig. 4 demonstrates that thermal evaporation of protons and \( \alpha \) particles from PLF or TLF dominates at far forward or far backward angles, respectively. The non-equilibrium component is best visible at intermediate angles, side-ways to the beam, in regions kinematically inaccessible to PLF or TLF evaporation. However, it is interesting to observe in Fig. 4 the non-equilibrium proton component exceeding the thermal spectrum also at angles as large as \( \theta = 138^\circ \).

The appearance of significant non-equilibrium emission complicates considerably the primary fragment reconstruction, unless the relative contributions from projectile and target to this process are well known. As illustrated below, utilizing projectile/target combinations with different A/Z asymmetries allows one to model the primary distributions in the presence of this complication.

As stated previously, a persistent disequilibrium of most degrees of freedom is a marked characteristic of the dissipative mechanism. This includes the relaxation of the A/Z (or N/Z) asymmetry brought in by projectile and target nuclei. There are very few detailed data yet on A/Z relaxation for the intermediate energy domain, owing to difficulties associated with event reconstruction, but this topic is gaining increased attention by the field [26 - 28].

In Fig. 5, such A/Z relaxation is viewed through the ratio \( M_1/M_0 \) of the multiplicities of neutrons and protons, respectively, evaporated from PLFs produced in peripheral reactions \(^{112}\text{Sn} + ^{40}\text{Ca}\) and \(^{112}\text{Sn} + ^{40}\text{Ca}\) at E/A = 35 MeV [23].

\[ \text{Figure 5: Multiplicity ratio of neutrons and protons evaporated from PLFs from the } \]

\[ ^{112}\text{Sn} + ^{40}\text{Ca} \text{ reactions vs. total excitation energy. [23]} \]

Here, the multiplicity ratio is plotted vs. the total reconstructed excitation energy, which is a measure of impact parameter. Indicated by dashed lines in this figure are the multiplicity ratios expected for “global” N/Z equilibration, defined in each case by the bulk N/Z of the combined system. The solid curves represent expectations based on an unchanged N/Z ratio of the PLF, given by the projectile N/Z.

Clearly, one observes a relaxation of the N/Z asymmetry with increasing \( E^* \), or decreasing impact parameter, which depends on the initial conditions, the projectile-target N/Z asymmetry. Although the equilibrium N/Z ratios for the two systems are not dramatically different, the evolution is strikingly opposite for the two systems. While the n-poor \(^{40}\text{Ca}\) projectile tends to pick up a net number of neutrons, \(^{40}\text{Ca}\) does not appear to donate any net number of neutrons to the n-poorer \(^{112}\text{Sn}\) target nucleus.

From the evolution of the experimental multiplicity ratios seen in Fig. 5, one concludes that global equilibrium is not reached, except perhaps for the highest excitation energies measured in the experiment. An opposite behavior of the multiplicity ratio \( M_1/M_0 \) with \( E^* \) in the two reactions can be understood as a consequence of
the locations of the injection points relative to the local structure of potential energy surface (PES) driving nucleon exchange between the interacting nuclei. The $^{112}$Sn+$^{40}$Ca injection point is located on the steep slope of the PES, whereas this point is located in the PES minimum for $^{112}$Sn+$^{48}$Ca trapping this latter system, for all measured impact parameters ($E^*_{\text{cm}}$). This feature is completely equivalent to that observed [1] at low energies and demonstrates similarity to the dissipative reaction mechanism. It should be mentioned that, even at low bombarding energies, the detailed evolution of mass and charge exchange with impact parameter has remained largely unexplained by theory.

While the above data illustrate the fragment N/Z ratios at late times, derived from their slow statistical decay, it has recently become possible to study the dynamics of N/Z (“isospin”) relaxation at very early times, when presumably fast, non-equilibrium particles are emitted from the colliding system. For peripheral collisions and bombarding energies of E/A ~ 30 - 40 MeV, non-equilibrium particles are emitted still with relatively small multiplicities. These particles are identified by their characteristic energy and angular distributions, consistent with the hypothetical IVS emission patterns introduced earlier.

In Fig. 6, the evolution with total excitation is displayed for the ratios $M_{\text{n}}/M_{\text{p}}$ of the multiplicities of non-statistical neutrons and protons emitted in the two $^{112}$Sn+$^{40,48}$Ca reactions discussed already above [23]. In either case, significantly more neutrons are emitted than protons. In view of the bulk ratios of only $N/Z_{\text{cs}}$ ~ 1.2 and 1.3 for $^{112}$Sn+$^{40}$Ca and $^{112}$Sn+$^{48}$Ca, respectively, the relative magnitude of excess non-equilibrium neutron emission is surprising. Comparison between the two systems suggests that this discrepancy is probably not caused by the Coulomb barrier, which is not significantly different for these two systems.

In addition, one again observes a dramatically different behavior of the (non-equilibrium) $M_{\text{n}}/M_{\text{p}}$ ratio with excitation energy for the two reactions. However now, the ratio is relatively stable for the $^{112}$Sn+$^{40}$Ca system ($M_{\text{n}}/M_{\text{p}}$ ~ 1.6), while it decreases from a large value of 7.6 down to a still significant $M_{\text{n}}/M_{\text{p}}$ ~ 3, for the more n-rich system. The bulk N/Z ratios are not reached by either system.

This observation demonstrates that non-equilibrium neutron and proton emission occurs mainly at times when projectile and target nucleons have not mixed. There is a clear memory of the entrance channel visible in the non-statistical $M_{\text{n}}/M_{\text{p}}$ ratio, specifically of the projectile N/Z. In fact, this mixing process, the relaxation of the mass-to-charge density (“isospin”) occurs significantly after the impact phase, which is thought responsible for the emission of fast nucleons.

In terms of macroscopic nuclear dynamics, the above process of non-equilibrium nucleon emission and its changes with impact parameter have to be attributed to the presence of a strong isospin dependence of the macroscopic nuclear equation of state (EOS). Overall matter compressibility must be significantly different for neutrons and protons. Qualitatively, such behavior is expected from theoretical models of isospin dynamics in heavy-ion reactions [26-28].

Reactions at intermediate energies are quite complicated to analyze in terms of isospin (N/Z asymmetry) relaxation effects or an “iso-EOS.” However, the fact that several reaction phenomena depend simultaneously, but in different
ways, on isospin removes ambiguity and provides constraints on theory.

Models of dissipative reaction dynamics have been developed [1] for heavy-ion reactions at near-barrier energies leading to small density overlap, where assumptions of perturbation theory are relatively well fulfilled. The success of these models also in domains, where their physical foundations are questionable, implies that these models have successfully captured the trends governing the dissipation of energy and angular momentum, the exchange of nucleons between the interaction partners and the evolution of their approximately distinct existence and incomplete communication. In particular, it is worthwhile to remember that the reaction systems are essentially in disequilibrium with respect to all degrees of freedom. The intrinsic transport mechanisms lead only slowly to a relaxation towards equilibrium. However, even highly excited, primary massive reaction products are stable enough to achieve internal equilibration, before decaying statistically on their asymptotic trajectories.

To the extent that heavy-ion reaction mechanisms at Fermi and intermediate bombarding energies follow these same trends, one recognizes the same underlying physical mechanism, a smooth transition to a new regime, and a gradual development of new phenomena. These latter phenomena concern mainly the mechanisms of cluster emission, which becomes important for E/A > 20 MeV, as discussed next.

2.4 Cluster Emission in Heavy-Ion Reactions

Emission of particles heavier than α-particles or other He isotopes is rare in nuclear processes at low excitations, although spontaneous cluster radioactivity has been observed for actinides [29] generating keen interest in the structure of such nuclei. Challenges to the understanding the emission of massive clusters (carbon or oxygen, say) from a heavy nucleus relate to the difficulty to model multi-particle in-medium correlations associated with the “preformation” of such clusters, as well as the inhibition by 50-60-MeV high Coulomb barriers. Interesting new attempts at understanding the phenomenon, both in nuclear structure and dynamics, involve therefore the nuclear surface.

Experimentally, there are at least two different and rather distinct mechanisms of cluster emission observed in reactions such as $^{197}$Au+$^{40}$Kr and $^{209}$Bi+$^{136}$Xe at bombarding energies between E/A = 20 and 60 MeV. In peripheral reactions leading to PLF and TLF products that are essentially cold, one observes [30-32] Be, B, and C clusters with IVS-type kinematical patterns and multiplicities of the order of $M_{\text{MF}} \sim 10^3$. Both Z and energy distributions are exponential in character. The slope parameters $T_s$ of the cluster energy spectra are significantly larger than emission temperatures (T ≤ 6 MeV) of PLF and TLF, measured via evaporated light particles.

The second observed type of cluster emission is sequential, statistical emission from primary PLF (or TLF). It is quite remarkable $a)$ that sequential cluster emission is observed at all and $b)$ that cluster decay is slow enough to allow the hot primary fragments to undergo Coulomb acceleration, requiring at least some $\sim 10^{20}$ s to complete. Most data sets to date do not distinguish between these 2 cluster emission mechanisms [3].

A set of typical data is illustrated by Fig. 7 [14] for the dissipative $^{209}$Bi+$^{136}$Xe reaction at three bombarding energies. Shown are invariant
velocity contour diagrams for IMF clusters sorted according to centrality, as defined by the joined multiplicity of neutrons and LCPs. Events are plotted vs. velocity components parallel and perpendicular to the beam direction. Clusters are defined as having $Z_{\text{IMF}} = 3$, with oxygen serving as an average, typical cluster.

It is obvious from Fig. 7 that the cluster emission patterns follow to a large extent the essentially binary kinematics of PLF and TLF sources, as the bombarding energy is raised from 28 MeV to 62 MeV. The dashed Coulomb semi-circles, indicating qualitative expectations based on similar patterns for LCP emission, emphasize this trend.

Detailed simulations of sequential cluster emission from accelerated PLF and TLF are not able to reproduce the experimental patterns with satisfactory accuracy. As is necessary in the description of LCP emission patterns, an intermediate IVS source is also required for an adequate description of experimental cluster emission patterns. The presence of the IVS source is particularly obvious in the distributions at lower bombarding energies (Fig. 7, left column) and at angles sideways to the beam. Here, the contours change less rapidly than at forward angles, indicating a harder energy spectrum for IVS clusters.

Results of a preliminary analysis of LCP and Li-like cluster spectra are shown in Fig. 8 for the reaction $^{209}$Bi+$^{136}$Xe at E/A = 28 MeV (green), 40 MeV (red), and 62 MeV (black).

Spectral slope parameters for H and He particles emitted sequentially from the PLF have values of the order of $T_s = 5 - 7.5$ MeV. Such events are collected at forward angles. As expected, the highest $T_s$ values appear for central collisions (Fig. 8, right panel) and for the highest bombarding energy. Such particles emitted from the IVS source, presumably non-statistical particles, have energy spectra with slope parameters of the order of 12-14 MeV. For these particles, the deduced $T_s$ values are lower for the higher bombarding energies, a fact that has yet to be understood.

Finally, spectra of Li clusters evaporated from PLFs show slightly higher slope parameters than the corresponding H and He distributions. Most remarkable, however, are the high slope parameters obtained for the non-statistical Li clusters associated with the IVS source. These latter values range from $T_s = 15$ MeV for peripheral collisions to $T_s = 25$ MeV, for more central events.

For the heavy systems studied here both average cluster multiplicity ($\langle M_{\text{IMF}} \rangle$) and width of the multiplicity distribution $P(M_{\text{IMF}})$ increase with increasing dissipation (decreasing impact parameter). At the same time, the balance between statistical and non-statistical cluster probability shifts somewhat in favor of the latter. For example, in mid peripheral $^{209}$Bi+$^{136}$Xe collisions, the 3 sources, PLF, TLF, and IVS, each contribute approximately similarly to the total cluster multiplicity $M_{\text{IMF}}$. At central collisions, on the other hand, non-statistical cluster emission clearly dominates.

This competition between production mechanisms has an unexpected effect on the correlation between the cluster multiplicity $M_{\text{IMF}}$ and the joint neutron–LCP multiplicity distribution. As explained earlier, the latter signifies a range in thermal excitation energy of the entire system.

The above correlation effect is demonstrated in Fig. 9 for the $^{197}$Au+$^{86}$Kr reaction at 35 MeV,
which is representative for other reactions studied in this bombarding energy regime. As expected, one observes that a region of small multiplicities or small excitations is associated with the absence of clusters \((m_{\text{IMF}}=0)\) and that the emission of one cluster requires already a significant amount of excitation acquired by the system in semi-peripheral collisions. Less expected is the fact that always the same \((m_a, m_{LCP})\) region appears “illuminated,” regardless of how many clusters are measured in coincidence. This may indicate an irregular pattern of competition between the emission of clusters and that of light particles. However, at present, correlations of the type shown in Fig. 9 are not available separately for the sequential, statistical cluster component.

In characterizing the cluster production mechanism, it is interesting to explore its dependence on bombarding energy and excitation. As it turns out, the statistical cluster multiplicity distribution for fixed thermal excitation changes very little with bombarding energy, while the non-statistical process becomes more dominant at higher bombarding energies.

Clearly, statistical and non-statistical cluster components (PLF/TLF vs. IVS) are produced by different mechanisms. Experimental data have already generated constraints on possible candidates for theoretical models of dynamic, non-statistical cluster production, which is observed to occur at all impact parameters. Elastic or inelastic projectile/target breakup on impact is an obvious candidate for the production of the latter, highly energetic IVS-type clusters. establishing their origin unambiguously requires highly specific cluster correlation measurements, which are not yet available. Using projectile/target combinations with significantly different N/Z asymmetries would facilitate identification of the non-equilibrium mechanism. In addition, experimental data of cluster emission in hadron-induced reactions [34, 35] could be used to complement observations in heavy-ion reactions.

3 MODEL OF STATISTICAL CLUSTER EMISSION

In the following, a new model [36] is briefly described for previously unexplained sequential cluster emission from excited primary fragments from dissipative heavy-ion collisions. This model hence concerns just one of the two observed cluster processes.

It is based on the well known principle of reaction kinetics that a reaction that is energetically disfavored can nevertheless take place, if the associated entropy gain is sufficiently high. The model further utilizes the fact that the density of nucleonic states in dilute nuclear matter such as found in nuclear surface is greater than in dense bulk matter. This property is illustrated by the level density parameter systematics [37] with mass number \(A\),

\[
a(\rho) = a_v + a_s = (A\alpha_v + A^{2/3}\alpha_s) (\rho/\rho_0)^{-2/3} \tag{3}
\]

The level density parameter \(a\) in Eqn. 3 contains a volume and a surface term, \(a_v\) and \(a_s\), respectively. In order to be able to treat nuclei that have densities \(\rho\) different from the normal nuclear matter density \(\rho_0\), an overall scaling factor \((\rho/\rho_0)^{-2/3}\) has been incorporated in the definition of \(a\), as suggested by the Fermi gas model for self-similar expansion or contraction.
The level density parameter determines the magnitude of the nuclear entropy, $S$, for a given thermal excitation energy $E_{\text{therm}}^*$:

$$S = 2 \sqrt{a \cdot E_{\text{therm}}^*}$$  \hspace{2cm} (4)

Nuclei with temperatures $T$ of the order of several MeV do not retain normal matter density but expand to an equilibrium density, $\rho_{\text{eq}} < \rho_0$, defined by maximum entropy. Using a harmonic approximation of the mean field [38], the density can be expressed analytically

$$\frac{\rho_{\text{eq}}}{\rho_0} = \frac{1}{4} \left( 1 + \sqrt{9 - 8 \frac{E_{\text{total}}^*}{E_{\text{binding}}} } \right)$$ \hspace{2cm} (5)

in terms of the total excitation energy

$$E_{\text{total}}^* = E_{\text{therm}}^* + E_{\text{comp}}^*$$ \hspace{2cm} (6)

The total excitation energy is a sum of thermal and compression energies. Since the nuclear compressibility is a function of the nuclear asymmetry energy, the second term in Eqn. 6 establishes interesting contact to the isospin dependent nuclear equation of state.

As a consequence of the energy consumed for expansion of the nucleus, to densities of $\rho_{\text{eq}} / \rho_0 \sim 1/3$, for $E_{\text{total}}^* / A > 8\,\text{MeV}$, the nucleus cools. The excitation energy dependence of the nuclear temperature becomes nonlinear, as illustrated in Fig. 10 for an A=208 nucleus. The temperature rises up to a maximum of $T \sim 6\,\text{MeV}$, in this example, and decreases at higher excitations leading to negative nuclear heat capacities. The maximum temperature shown in Fig. 10 is the limit an A=208 nucleus can sustain, before disintegrating into nucleons. Entropy driven nuclear expansion is hence seen as the ultimate reason for experimentally observed [4-6] limits of thermal stability. Due to the reduction in nuclear temperature caused by expansion, nuclear stability is actually increased, as compared to the same nucleus at normal density. In addition, negative heat capacities have also been reported in the literature [39, 40]. For the first time, the present harmonic interaction Fermi gas model (HIFGM) gives a consistent explanation for both these effects, suggesting a host of interesting studies, e.g., of the isospin dependence of thermal stability and heat capacity.

Furthermore, it is a natural consequence of the HIFGM that a hot, expanded nucleus tends to produce dinuclear transition states for the fission-like evaporation of a complex nuclear cluster. For example, one calculates that for $E_{\text{total}}^* / A > 4.5\,\text{MeV}$ the system of the two daughter nuclei $^{102}\text{W}$ and $^{16}\text{O}$, touching at their nuclear surface, has a higher entropy than the A=208 parent nucleus $^{208}\text{Pb}$. Hence, $^{16}\text{O}$ cluster emission is likely to occur, even though the nuclear temperature is one order of magnitude lower than the emission barrier.

Numerical HIFGM predictions of emission

![Equilibrium Temperature vs. Total Excitation Energy](image1)

**Figure 10:** Dependence of temperature on total excitation per nucleon, predicted by the harmonic approximation Fermi gas model [36].

![Cluster Emission Probability vs. Z_cluster](image2)

**Figure 11:** HIFGM cluster emission probability for a Au nucleus, normalized by proton emission probability, vs. $Z_{\text{cluster}}$ and excitation energy per nucleon. [36]
probabilities $p_{\text{cluster}}$ are shown in Fig. 11 for clusters with atomic number $Z_{\text{cluster}}$, relative to the emission probability of a proton, $p_p$. The different logarithmic lines are calculated for different excitation energies $E^* = E_{\text{total}}/A$ of an A=197 (Au) nucleus. As can be seen from this figure, cluster emission is predicted to be an extremely likely process for excitations exceeding 4-5 MeV per nucleon. One calculates, for example, that $^{18}$O emission from a hot Au nucleus is more likely even than the emission of a proton.

The main reason for this effect is the complexity and high intrinsic level density of massive clusters. Strongly bound clusters or clusters with low densities of states are not so favored, according to the HIFGM. Such unusual schemes of competition between the evaporation of clusters and simpler particles could be responsible for the correlations between $m_{\text{bar}}$ and the joint neutron/LCP multiplicity discussed in the context of Fig. 9.

A more meaningful comparison of HIFGM predictions with experiment requires modeling of the entire evaporation cascade of a hot nucleus, e.g., with a code like GEMINI [22]. Work is underway to modify and expand this code accordingly.

4 CONCLUSIONS

In summary, it has been demonstrated that many of the salient features of dissipative heavy-ion reactions persist at bombarding energies at the upper boundary of the Fermi domain. Even though reconstruction of reaction events is often experimentally problematic, due to high excitations of reaction primaries, $4\pi$ measurements of all decay products have provided examples where reconstruction of these primaries has been possible to a large extent. As a result, one observes dissipative orbiting, for heavy reaction systems at least up to $E/A = 62$ MeV. With these features, one recognizes multi-nucleon exchange as the underlying mechanism, recovering a dissipative reaction environment largely familiar from lower bombarding energies, including incomplete relaxation of mass-to-charge (isospin) asymmetry and excitation energy division.

As a new, transitional feature of the reaction mechanism at the higher energies of interest, emission of complex, intermediate-mass clusters is observed on different time scales. A fast cluster emission mechanism is probably associated with projectile and target breakup in the approach phase. This mechanism is strongly dependent on nuclear overlap and bombarding energy. This mechanism has not yet been studied in any detail and requires dedicated experimentation of non-equilibrium phenomena in heavy-ion reactions at Fermi energies.

In addition, hot primary PLF (TLF) reaction fragments are observed to emit clusters in a delayed, statistical emission process and at times close to reaching asymptotic velocities. The relative stability of hot reaction primaries is understood to be due to nuclear expansion, lowering temperature and evaporation widths of simple particles. A plausible micro-canonical model for hot, expanding nuclei, based on the interacting Fermi gas, is able to account qualitatively for the observed cluster decay of such nuclei. In this model, the nuclear surface entropy plays an important role. Thermal stability limits and negative heat capacities are simple, natural consequences of such a model view. A host of new physical phenomena, including interesting isospin dependent effects, have been predicted by the model. The model decay widths are worked into the scheme of a conventional statistical model computer code to accommodate cluster evaporation. With these new tools, the long-standing puzzle of nuclear multi-fragmentation is finally being solved. In addition, statistical cluster emission promises to define a new research direction exploring a new mode of nuclear decay.

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Surface entropy in statistical emission of massive fragments from equilibrated nuclear systems

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Statistical fragment emission from excited nuclear systems is studied within the framework of a schematic Fermi-gas model combined with Weisskopf’s detailed balance approach. The model considers thermal expansion of finite nuclear systems and pays special attention to the role of the diffuse surface region in the decay of hot equilibrated systems. It is found that with increasing excitation energy, effects of surface entropy lead to a systematic and significant reduction of effective emission barriers for fragments and, eventually, to the vanishing of these barriers. The model predicts a maximum (effective) nuclear temperature and the occurrence of negative nuclear heat capacities, effects that have been reported in the literature. It also accounts for the observed linearity of pseudo-Arrhenius plots of the logarithm of the fragment emission probability versus the inverse square root of the excitation energy, but does not predict true Arrhenius behavior of these emission probabilities.

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I. INTRODUCTION

Over the last decade, considerable efforts have been made to understand the phenomenon of the production of multiple intermediate-mass fragments (IMF) in individual nuclear reactions. As a contribution to these continuing efforts, the present study demonstrates that a simple scenario, closely related to that known from fission studies, offers an explanation of how, at moderately high excitation energies, IMF emission can compete effectively with nucleon evaporation. It represents an extension of ideas and formalism presented in a recent publication [1]. Central to this formalism is the notion of a relatively high entropy associated with the diffuse nuclear surface region (as opposed to bulk matter).

The model formalism adopted in the present study is described in detail in Section II. While this formalism is somewhat schematic, it is believed to capture the essential physics underlying the processes involved. One benefit of such a schematic treatment is that it provides direct insight into the phenomena of interest, disregarding a multitude of secondary details demanded by a more rigorous approach. Results of calculations are presented in Sec. III, while the conclusions are presented in Sec. IV.

II. THEORETICAL FORMALISM

The present study assumes that an excited nuclear system expands in a self-similar fashion so as to reach a state of approximate thermodynamic equilibrium, where the entropy $S$ is maximal for the given total excitation energy $E_{\text{tot}}$, i.e.,

$$\frac{\partial S(E_{\text{tot}}, \rho)}{\partial \rho} \bigg|_{E_{\text{tot}}^*} = 0. \quad (1)$$

The functional dependence of the entropy on $E_{\text{tot}}$ and bulk nuclear matter density $\rho$ is evaluated using the Fermi-gas model relationship

$$S = 2\sqrt{a E_{\text{th}}} = 2\sqrt{a(E_{\text{tot}} - E_{\text{compr}})}, \quad (2)$$

where $a$ is the level density parameter, $E_{\text{th}}^*$ is the thermal excitation energy, and $E_{\text{compr}}$ is the collective compressional energy. The dependence of $S$ on bulk nuclear matter density $\rho$ arises in Eq. (2) through the dependence of both, the level density parameter $a$ (“little-a”) and the compressional energy $E_{\text{compr}}$, on the matter density.

The dependence of little $a$ on the nuclear matter density for infinite nuclear matter is given by the Fermi-gas model:

$$a = a_0 \left(\frac{\rho}{\rho_0}\right)^{-2/3}, \quad (3)$$

where $a_0$ is the level density parameter for the nuclear matter at ground-state matter density $\rho_0$.

The above equation holds approximately also for finite nuclei if the expansion or compression of these nuclei is assumed to occur in a self-similar fashion. This is so because for finite nuclei, the little-$a$ parameter consists of volume and surface terms, $a_V$ and $a_\sigma$, respectively, both of which are proportional to $\rho^{-2/3}$ under the assumption of self-similar expansion:

$$a = a_V + a_\sigma = A \left(\frac{\rho}{\rho_0}\right)^{-2/3} \alpha_V + A^2 \left(\frac{\rho}{\rho_0}\right)^{-2/3} \alpha_\sigma, \quad (4)$$

where $\alpha_V$ and $\alpha_\sigma$ are volume and surface coefficients, respectively, independent of bulk nuclear matter density.

The term “self-similar expansion” is used here to describe a type of expansion in which any change in the matter density profile is reducible to a simple rescaling of the radial coordinate, such that

$$f_\rho(r) = c^3 f_\sigma(r), \quad (5)$$

where $f_\rho(r)$ is the ground-state density profile function and $c$ is a scaling constant.

The presence of a surface contribution to the level density parameter is of crucial importance in the present study as it describes that part $S_\sigma$ of the entropy $S$ of the system, which
is associated with the diffuse surface domain and is seen to have significant effect on the fragment emission probability. One has

$$S = S_V + S_\sigma,$$

(6)

where $S_V$ is the entropy of the bulk matter.

The compressional energy in Eq. (2) is approximated in the present study following the schematic prescription proposed in the expanding emitting source model EESM [4], i.e.,

$$\varepsilon_{compr} = \varepsilon_b \left( 1 - \frac{\rho}{\rho_o} \right)^2,$$

(7)

where $\varepsilon_{compr}$ and $\varepsilon_b$ are the compressional and the ground-state binding energies per nucleon of the system, respectively. Note that Eq. (7) ensures that the compressional energy varies parabolically with $\rho$, from zero at ground-state density $\rho_o$ to $\varepsilon_b$ at zero density.

Equations (1)–(3) and (7) allow one to obtain an analytical expression for the equilibrium density $\rho_{eq}/\rho_o$ of nuclear matter as a function of the excitation energy per nucleon, $\varepsilon_{tot}^* = E_{tot}^*/A$, where $A$ is the mass number of the system:

$$\rho_{eq} = \frac{1}{4} \left( 1 + \sqrt{9 - \frac{8\varepsilon_{tot}^*}{\varepsilon_b}} \right).$$

(8)

Equation (8) reflects the fact that for $\varepsilon_{tot}^* < \varepsilon_b$, the system is bound, as far as the self-similar expansion mode is concerned, and features a single maximum entropy for the range of matter densities $1/2 \leq \rho_{eq}/\rho_o \leq 1$. For $\varepsilon_{tot}^* > \varepsilon_b$, the system still has a local, metastable maximum at a finite density given by Eq. (8), i.e., in the range of matter densities $1/4 \leq \rho_{eq}/\rho_o \leq 1/2$. The latter metastable (with respect to self-similar expansion mode) maximum in entropy is separated from the divergence at zero density by a minimum at $\rho_{saddle}$, where

$$\rho_{saddle} = \frac{1}{4} \left( 1 - \sqrt{9 - \frac{8\varepsilon_{tot}^*}{\varepsilon_b}} \right).$$

(9)

Here, $E_b = A \varepsilon_b$.

The probability $p$ of emitting a fragment from an equilibrated excited system, as defined above, can be evaluated using the Weisskopf formalism [5]:

$$p = e^{-S} = e^{-S_{saddle} - S_{eq}},$$

(10)

where $S_{saddle}$ and $S_{eq}$ are saddle-point and equilibrium-state entropies, respectively. The latter two entropies can be calculated using Eq. (2):}

$$S_{eq} = 2 \sqrt{a_A \left( E_{tot}^* - E_b \right) \left( 1 - \frac{\rho_{eq}}{\rho_o} \right)^2},$$

(11)

In Eqs. (11) and (12), $a_A$, $a_{res}$ and $a_{frag}$ are the level density parameters of the system at equilibrium, the residue, and the fragment, respectively, while $E_{saddle}^*$ is the thermal excitation energy of the system in the saddle-point configuration. The latter quantity is calculated as

$$E_{saddle}^* = E_{tot}^* - E_b \left( 1 - \frac{\rho_{eq}}{\rho_o} \right)^2 - V_{saddle},$$

(13)

where $V_{saddle}$ is the (collective) saddle-point energy.

Note that the present formalism of statistical decay of excited nuclear systems, albeit schematic, makes no use of intensive parameters, such as temperature or pressure. As evident from Eqs. (1)–(13), the present model builds entirely on extensive variables, both thermostatic (total excitation energy, compressional energy, and entropy) and geometrical (density parameter $\rho$, used here to describe the volume of the system). This is in stark contrast to many approaches commonly used to describe statistical decay of nonextensive systems, i.e., systems for which the thermodynamic limit cannot be reached. These latter approaches include the equilibrium statistical model Gemini [6], the expanding emitting source model [4], Fisher’s model [7], and the Arrhenius-type Berkeley approach [8], all of which rely inherently on the notion of a temperature, an intensive parameter. While the above statements should not be construed as a criticism of the cited models, it had been argued [9] that, thermodynamical models based on extensive observables, such as entropy, total excitation energy, and geometry, are better suited for the purpose of nonextensive thermostatics of “small” systems than models built on intensive parameters.

While not constituting an inherent element of the present model, the notion of a (microcanonical or effective) nuclear temperature is used in the following section to discuss a caoric curve—an entity that has attracted much attention in the course of multifragmentation studies [10,11]. The notion of an (effective) temperature has been used also in constructing an Arrhenius plot for fragment emission probabilities, another entity that has attracted much attention in recent years [8]. While using the notion of an effective temperature, however, one has to keep in mind its possible limitations as far as the description of nonextensive systems is concerned. For the above specific application, an effective nuclear temperature can be obtained from the commonly used Fermi-gas model relationship between the temperature $T$ and thermal excitation energy $E_{th}^*$:

$$T = \sqrt{\frac{E_{th}^*}{a}} = \left( \frac{\rho_{eq}}{\rho_o} \right)^{1/3} a_{res}^{1/2} \left[ E_{tot}^* - E_b \left( 1 - \frac{\rho_{eq}}{\rho_o} \right)^2 \right].$$

(14)

To allow one to quickly evaluate the magnitude of the effects due to surface entropy, the notion of an effective barrier $B_{eff}$ determining the emission probability $p$ is used in
the following section. This notion is introduced so as to formally reduce the emission probability $p$ given by Eqs. (10)–(12) of the present formalism to a more intuitive, effective Boltzmann factor. The effective barrier is defined by the equation

$$p \propto e^{-B_{\text{eff}}/T},$$

and hence,

$$B_{\text{eff}} = -T \Delta S.$$

Note again that Eq. (16) does not include any intensive parameters, the parameter $T$ being constructed from purely extensive observables [see Eq. (14)].

A selection of results of calculations performed using the above formalism is presented in Sec. III below.

**III. RESULTS OF MODEL CALCULATIONS**

Results of the calculations performed in the framework of the formalism presented in Sec. II are presented in Figs. 1–7. In these calculations, values of $\alpha_V = 1/14.6$ MeV$^{-1}$ and $\alpha_s = 4/14.6$ MeV$^{-1}$ have been assumed for the coefficients $\alpha_V$ and $\alpha_s$, as suggested in the literature [2]. Further, $\epsilon_b = 8$ MeV was assumed [4] for the ground-state binding energy per nucleon, while the saddle-point collective energy was approximated by the Coulomb energy of the residue and fragment represented by two touching spheres of radius parameter $r_{\text{Coul}} = 1.3(p/p_0)^{-1/3}$ fm. The calculations were made for excited $^{197}$Au nuclei.

Figure 1 illustrates the dependence of the equilibrium density $\rho_{eq}$ of bulk matter on the excitation energy per nucleon (solid curve). As seen in this figure, for the range of excitation energies readily accessible in experiments, the bulk matter density in a state of maximum entropy differs substantially from that of the nuclear ground state. This affects both, the caloric equation of state and the fragment emission probability, two entities of considerable interest.

The dashed curve seen in Fig. 1 illustrates the net gain in entropy resulting from the relaxation of the self-similar expansion mode in an excited $^{197}$Au system. Large gains in entropy associated with the relaxation of this mode emphasize the importance of this mode for a statistical description of excited nuclear systems, notably for models based on the concept of microcanonical [12] or pseudomicrocanonical [13] equilibrium.

The caloric curve calculated for states of maximum entropy, i.e., for the states of equilibrium density $\rho_{eq}$, is depicted in Fig. 2. Not surprisingly, this curve shows considerable deviation from the simple Fermi-gas form of $T \propto \sqrt{E_{tot}}$. Note that in experiments such as the recently reported ISIS experiment [14], it is $E_{tot}$ and not the purely...
thermal contribution to it, \( E_{th}^* \), that is in fact measured. This is so, because the static compressional energy \( E_{compr} \) is experimentally undistinguishable from thermal excitation \( E_{th}^* \).

Most notably, the caloric curve predicted in Fig. 2 features a maximum temperature of approximately \( T_{\text{max}} = 6 \text{ MeV} \). This is an indication that, for higher temperatures, the system is inherently unstable and does not find an equilibrium density. In other words, under the assumption that its matter distribution is homogeneous, a nuclear system placed in a heat bath of \( T = 6 \text{ MeV} \) would expand indefinitely, for which it would derive increasing amounts of energy from the heat bath. In a more realistic case, which is beyond the present consideration, before reaching thermal equilibrium, the system would likely decay dynamically into a “gas” of fragments and free nucleons that would continue its indefinite expansion. It is worth noting that, in the present formalism, a limiting, maximum nuclear temperature arises naturally from the requirement of the dynamical equilibrium implied by Eq. (1). This is in contrast to many other statistical-decay models for excited nuclear systems, notably to all models relying on the concept of the existence of a freezeout volume [12,13] or a breakup configuration [15]. It is also in contrast to Fisher’s model [7], which has recently been reconsidered in the context of intermediate-mass fragment production [16]. None of the above model approaches [7,12,13,15,16] predicts a maximum temperature. However, the existence of such a limiting temperature appears to be supported by a series of experimental observations [10].

The particular form of the caloric curve seen in Fig. 2 can, perhaps, be better understood when inspecting the dependence of the free energy \( F \) on matter density \( \rho \) and temperature \( T \), which is illustrated in Fig. 3. The calculations show

![Figure 4](image4.png)

**FIG. 4.** Effective barriers for the emission of \(^{12}\text{C}\) and \(^{16}\text{O}\) fragments from equilibrated excited \(^{197}\text{Au}\) systems as functions of excitation energy per nucleon.

![Figure 5](image5.png)

**FIG. 5.** Distributions of relative emission probabilities of various IMFs from an excited, equilibrated \(^{197}\text{Au}\) system as functions of the total excitation energy (solid curves). The dashed line represents the boundary of the domain of dynamical instability of the system (see text).

![Figure 6](image6.png)

**FIG. 6.** Arrhenius plots for emission of \(^{12}\text{C}\) and \(^{16}\text{O}\) fragments from excited \(^{197}\text{Au}\) systems.

![Figure 7](image7.png)

**FIG. 7.** Pseudo-Arrhenius plots for emission of \(^{12}\text{C}\) and \(^{16}\text{O}\) fragments from excited \(^{197}\text{Au}\) systems.
that, at temperatures below $T \approx 6$ MeV, the free energy as a function of $\rho$ features two equilibrium points, a local minimum and a local maximum, where the latter reflects an unstable equilibrium. These two points, predicted for any temperature $T$ of less than approximately 6 MeV, correspond to different excitation energies per nucleon, $E_{\text{tot}}^*$. Note that at both these extremal points, the pressure of the system at its surface is zero. This is so, because the pressure is proportional to the partial derivative of the free energy with respect to density at constant entropy, and since both, the free energy and the entropy, are stationary with respect to the matter density at the extremal points in question. At $T \approx 6$ MeV, the minimum and the maximum in $F(\rho)$ merge into an inflection point. At even higher temperatures, the free energy features only a monotonic decrease with decreasing $\rho$. The inflection point in $F$ for $(T \approx 6$ MeV) corresponds to a maximum of $T_{\text{stable}}$ equilibrium. These two points, predicted for any temperature $T$, and since $T$ is a nonmonotonic function of $E_{\text{tot}}^*$, one would expect the fragment emission probabilities $p$ to deviate significantly from a simple Arrhenius law with an exponential functional dependence on the inverse temperature $1/T$. This expectation is confirmed by Fig. 6, where logarithmic plots of $p$ versus $1/T$ are seen to feature clear deviations from linearity, including a prominent “back-bending.”

The latter backbending is obviously expected, in view of the maximum in the caloric curve seen in Fig. 2. Such a behavior is also suggested by the general deviation of the caloric equation of state from the one for a low-temperature Fermi gas.

On the other hand, pseudo-Arrhenius plots depicted in Fig. 7, where the logarithm of the emission probability, $\ln(p)$, is plotted versus the inverse square root of the total excitation energy, $1/\sqrt{E_{\text{tot}}^*}$, are to a good approximation straight lines. This observation comes as a surprise, as it cannot readily be expected, based on the details of the theoretical formalism employed. Therefore, no simple explanation for such a linearity can be offered at this time, the very rationale behind the construction of such pseudo-Arrhenius plots being numerous experimental findings [8,17–20]. While it may be purely fortuitous, the linear character of pseudo-Arrhenius plots predicted by the present model seems to be in agreement with experimental observations.

IV. CONCLUSIONS

A model has been developed to describe quantitatively, albeit in a schematic fashion, a scenario of purely statistical emission of massive fragments from finite equilibrated systems. The formalism is based on the use of extensive observables only, complemented by the geometry of the used phase space. It is, hence, free of the possible limitations of models relying on intensive variables. The presented formalism recognizes the importance of thermal expansion of hot matter and considers the stability of such systems at equilibrium nuclear densities. Unlike many other models and approaches, the present formalism predicts in a natural fashion a limiting maximum temperature bound nuclear system can sustain, an effect supported by numerous experimental observations. The important role of surface entropy consists in an effective “softening’’ of the nuclear surface, resulting in enhanced fragment emission probabilities. The decay scenario underlying this formalism is that of a thermally expanded system with developed thermal fluctuations of the diffuse surface.

Note added in proof. It came to our attention that the curve $\rho_{eq}/\rho_0$ (cf. Fig. 1) obtained from the single-parameter function in Eq. (8) essentially coincides with a similar curve resulting from a finite-temperature Hartree-Fock calculation, as reported in Ref. [21].

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[6] R.J. Charity, computer code GEMINI, see wunmr.wustl.edu/pub/gemini
Liquid-gas coexistence and critical behavior in boxed neutral, isosymmetric pseudo-Fermi matter

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A schematic model is presented that allows one to study the behavior of interacting neutral isosymmetric pseudo-Fermi matter, locked in a thermostatic box. As a function of the box volume and temperature, the matter is seen to show all of the familiar characteristics of a van der Waals gas, which include the coexistence of two phases under certain circumstances and the presence of a critical point.

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I. INTRODUCTION

The possibility that a liquid-gas phase transition in finite nuclei may manifest itself via copious production of intermediate-mass fragments in energetic heavy-ion reactions has driven both, theoretical and experimental studies of nuclear multifragmentation over more than a decade. A prominent role in theoretical considerations is played by the concept of a freezeout configuration [1,2]. This concept implies the existence of a definite volume, within which the system reaches a state close to thermal equilibrium. While the existence of an effective freezeout volume may be debatable, the concept itself is useful for understanding the possible behavior of nuclear matter under various conditions. With such a didactic strategy in mind, and as an extension of earlier studies modeling the behavior of finite Fermi systems [3–5], the present study considers neutral, isosymmetric pseudo-Fermi matter confined to a box of definite volume. It evaluates the isothermal behavior of such matter and its dependence on box volume and temperature. The utmost simplicity of the formalism allows one to gain insight into physical phenomena that may be obscured in more rigorous approaches.

While the present formalism can be readily extended to include isosymmetric matter, inclusion of a Coulomb interaction would require a number of qualitative changes, all resulting from the long range of the Coulomb interaction. Notably, the presence of a Coulomb interaction renders crucial extensive thermodynamic quantities nonadditive. For example, the Helmholtz free energy of a charged two-phase system is no longer equal to the sum of free energies of its separated gaseous and liquid parts. Furthermore, neither the pressure nor the density of charged matter is constant over the volume of any single phase, depending not only on the geometrical configuration assumed by this phase, but also on that of the coexisting phase. The above, highly nonlinear features make it very difficult in the present formalism to include the Coulomb interaction in a satisfactory manner. In common practice, this interaction is also disregarded in more rigorous approaches to the liquid-gas coexistence (see, e.g., Ref. [6], and references therein).

II. THEORETICAL FORMALISM

The present study considers a scenario of neutral, isosymmetric pseudo-Fermi matter of mass number A, locked in a spherical box of a volume V, and kept at constant temperature $T$. In the proposed formalism, thermostatic properties of the matter are modeled by two equations, the isochoric caloric equation of state and the zero-temperature equation of state. The equilibrium state of the system is then found as a function of $V$ and $T$, based on the requirement that the free energy of the system is minimal.

First, we consider homogeneous systems with constant density throughout their volume, i.e., systems in states where only one phase is present. The isochoric caloric equation of state for such a system is taken in a simple form adequate for low-temperature Fermi gases

$$E_{\text{therm}}^* = aT^2,$$

(1)

where $E_{\text{therm}}^*$ is the thermal energy and $a$ is the level density parameter. The latter parameter is assumed to depend on matter density as

$$a = a_o \left( \frac{\rho}{\rho_o} \right)^{-2/3},$$

(2)
in accordance with the low-temperature Fermi gas model.

It is worthwhile keeping in mind that, for Fermi gases, Eq. (1) is a good approximation only for temperatures that are small compared to the Fermi energy, i.e., for $T < E_{\text{Fermi}}^c$. Notably, for diluted or very hot Fermi systems, with a matter density approaching zero, $\rho \to 0$, or for high temperatures, $T \to \infty$, the isochoric caloric equation of state approaches asymptotically that of a classical gas:

$$E_{\text{therm}}^* \to \frac{3}{2} a_o T^2.$$

(3)

For the sake of simplicity and without loss of generality, the present study uses Eq. (1) over the full range of matter densities and temperatures considered. The term “pseudo-Fermi” matter is used to distinguish the matter considered here from true Fermi matter. Note that for a noninteracting Fermi gas, the Fermi energy scales with the density as $E_{\text{Fermi}}^c \approx 39(\rho/\rho_o)^{2/3}$ MeV. For interacting Fermi gases, this energy is even higher, as it scales with the inverse of the effective nucleonic mass. Accordingly, the present concept of pseudo-Fermi matter provides a reasonably good approximation of true Fermi matter, in the most interesting domains of the plots discussed in Sec. III, notably, in the vicinity of the critical point ($T_c \approx 10$ MeV $< E_{\text{Fermi},c} \approx 22$ MeV).

The second defining equation, the zero-temperature equation of state, expresses the compressional (potential) energy of the system as a function of the system volume or as a function of matter density. The present study adopts a har-
monic approximation in the form used in the expanding emitting source model [7]. Here, the in-medium nucleonic (potential) energy changes quadratically with the relative deviation of the actual matter density from the ground-state density by an amount

$$\epsilon_{\text{compr}} = \epsilon_B \left( 1 - \frac{\rho}{\rho_o} \right)^2. \quad (4)$$

In Eq. (4), $\epsilon_{\text{compr}}$ and $\epsilon_B$ are compressional and ground-state binding energies per nucleon, respectively, and $\rho$ and $\rho_o$ are the actual and the ground-state matter densities, respectively. Equation (4) implies an effective ground-state incompressibility constant of $K_o = 18 \epsilon_B$. Assuming $\epsilon_B = 8$ MeV, the effective incompressibility, including the effects of surface tension, equals $K_o = 144$ MeV. Note that, for infinite nuclear matter characterized by $\epsilon_B = 16$ MeV, the incompressibility constant turns out to be equal to $K_o = 288$ MeV, in this harmonic approximation. The latter value places the present harmonic approximation “neutrally” between the currently considered limits of “soft” and “hard” equations of state for nuclear matter.

While the two defining equations (1) and (4) may be considered rather crude approximations, they do contain the essential physics responsible for first-order phase transitions and critical phenomena. Given these two equations, one can write expressions for all thermodynamic quantities characterizing the system, including the Helmholtz free energy $F$. The state of the system can then be found by minimizing the free energy, for any box volume $V$ and temperature $T$.

Based on Eq. (1), one can write for the entropy $S$ of a homogeneous system

$$S = \int_0^{E^*} \frac{d\epsilon}{T} = 2 \sqrt{a E_{\text{therm}}^*}. \quad (5)$$

The free energy $F$ for a homogeneous, single-phase system is given by

$$F = E_{\text{total}}^* - ST = E_{\text{compr}}^* + E_{\text{therm}}^* - 2aT^2 = E_{\text{compr}}^* - aT^2. \quad (6)$$

For the sake of simplicity, the free energy is expressed in Eq. (6) relative to a constant ground-state energy of the system. Furthermore, using Eqs. (2) and (4), the free energy [Eq. (6)] can be rewritten in a form revealing explicitly the important dependence on the matter density $\rho$, i.e.,

$$F = E_B \left( 1 - \frac{\rho}{\rho_o} \right)^2 - a_o \left( \frac{\rho}{\rho_o} \right)^{-2/3} T^2. \quad (7)$$

Equation (7) allows one to write expressions for the system pressure $p$ and the chemical potential $\mu$ as functions of volume (matter density) and temperature.

The pressure $p$ can be expressed generally as the negative partial derivative of the free energy $F$ with respect to volume $V$, at fixed number of nucleons $A$ and fixed temperature $T$, i.e.,

$$p = -\frac{\partial F}{\partial V}_{A,T} = \frac{1}{A} \rho^2 \frac{\partial F}{\partial \rho}_{A,T}. \quad (8)$$

Thus, for the case of pseudo-Fermi matter considered here, one obtains, based on Eqs. (7) and (8),

$$p = -2 \epsilon_B \rho_o \left( 1 - \frac{\rho}{\rho_o} \right)^2 + \frac{2}{3} \alpha_o \rho_o \left( \frac{\rho}{\rho_o} \right)^{1/3} T^2, \quad (9)$$

where $\epsilon_B$ and $\alpha_o$ are the binding energy per nucleon and the level-density parameter per nucleon, respectively.

The chemical potential $\mu$ can be expressed generally as the partial derivative of the free energy $F$ with respect to the number of nucleons $A$, taken at fixed volume $V$ and fixed temperature $T$, i.e.,

$$\mu = \frac{\partial F}{\partial A}_{V,T}. \quad (10)$$

Using Eqs. (7) and (10) and noting further that $\rho = A/V$, $E_B = A \epsilon_B$ and $a_o = A \alpha_o$, one obtains for the chemical potential

$$\mu = \epsilon_B \left[ 1 - 4 \frac{\rho}{\rho_o} + 3 \left( \frac{\rho}{\rho_o} \right)^2 \right] - \frac{1}{3} \frac{\alpha_o}{\rho_o} \left( \frac{\rho}{\rho_o} \right)^{-2/3} T^2. \quad (11)$$

It is worth noting in Eq. (11) that, as a result of the requirement that $V$ is a constant, the magnitude of the chemical potential differs significantly from the value of the free energy per nucleon. For example, the contribution of thermal excitation to the chemical potential is only one-third of what constitutes the thermal part of the free energy per nucleon.

### III. LIQUID-GAS COEXISTENCE

Confined to a thermostatic box, matter will eventually fill the available volume $V$ entirely such that the free energy of the system is minimized. For noninteracting matter, the minimum free energy always corresponds to a uniform density distribution. This is generally not true for an interacting system. In particular, for interacting pseudo-Fermi matter at the minimum free energy, low-density and high-density phases coexist.

For a two-phase system of $A$ nucleons at temperature $T$ confined to a volume $V$, the free energy can be written as a function of two variables, e.g., in terms of the volume of the gaseous phase $V_{\text{gas}}$ and the number of nucleons contained in this phase, $A_{\text{gas}}$,

$$F = F_{\text{gas}} + F_{\text{liquid}}. \quad (12)$$

Inserting for $F_{\text{gas}}$ and $F_{\text{liquid}}$ the expressions given by Eq. (7) for the corresponding numbers of nucleons, $A_{\text{gas}}$ and $A_{\text{liquid}}$, respectively, one obtains

$$F_{\text{gas}} = A_{\text{gas}} \epsilon_B \left( 1 - \frac{A_{\text{gas}}}{V_{\text{gas}} \rho_o} \right)^2 - A_{\text{gas}} \alpha_o \left( \frac{A_{\text{gas}}}{V_{\text{gas}} \rho_o} \right)^{-2/3} T^2 \quad (13)$$

and

$$F_{\text{liquid}} = (A - A_{\text{gas}}) \epsilon_B \left[ 1 - \frac{A - A_{\text{gas}}}{(V - V_{\text{gas}}) \rho_o} \right]^2 - (A - A_{\text{gas}}) \alpha_o \left( \frac{A - A_{\text{gas}}}{(V - V_{\text{gas}}) \rho_o} \right)^{-2/3} T^2. \quad (14)$$
The condition for the minimum free energy can be expressed in a form of two equations, reflecting requirements of dynamical and chemical equilibrium, respectively,

\[
\frac{\partial F}{\partial V_{\text{gas}}} = \frac{\partial F}{\partial V_{\text{gas}}} + \frac{\partial F}{\partial V_{\text{liquid}}} = p_{\text{gas}} - p_{\text{liquid}} = 0 \quad (15)
\]

and

\[
\frac{\partial F}{\partial V_{\text{gas}}} = \frac{\partial F}{\partial V_{\text{gas}}} + \frac{\partial F}{\partial V_{\text{liquid}}} = \mu_{\text{gas}} - \mu_{\text{liquid}} = 0. \quad (16)
\]

Results of a numerical minimization of the free energy of a two-phase system are shown in Figs. 1–4. Figure 1 presents system isotherms, as predicted by the present formalism, in a representation of system pressure \( p \) versus system volume \( V \). Note that Eqs. (13) and (14) represent single-phase states as special cases. A pure liquid/gas state is thus among the possible outcomes of the calculations with \( A_{\text{gas/liquid}} = 0 \). As seen in Fig. 1, the isotherms feature segments representing pure liquid, pure gas, or, notably, a liquid-gas coexistence “plateau.” The presence of such coexistence plateaus does not come as a surprise, since the harmonic interaction term of Eq. (4) has the salient characteristics of a van der Waals interaction.

It is worth noting that the coexistence plateaus in this calculation result naturally from the actual minimization of the free energy and are not obtained via the well-known phenomenological Maxwell construct. Isotherms for hypothetical single-phase states are shown as dotted lines. These latter isotherms feature domains of spinodal instability characterized by negative compressibility. The dashed curve in Fig. 1 illustrates the boundary of the liquid-gas coexistence domain. The “summit” point of this curve represents the critical point for the system and corresponds to a critical temperature of \( T_c \approx 10.0 \text{ MeV} \). At temperatures higher than \( T_c \), the system can reside only in a single-phase, vapor state.

A different representation of the liquid-gas coexistence line is illustrated in Fig. 2. In this case, the temperature \( T \) is plotted versus the matter density, for points along the boundary of the liquid-gas coexistence line (dashed line in Fig. 1). In the domain below this curve, the system is in a two-phase state. In this coexistence domain, the densities of gaseous and liquid phases at a given temperature are defined by the intersection points of the coexistence curve with the horizontal line for \( T = \text{const} \). On the left shoulder of the curve and in the domain further left to it, the system is in a pure gaseous state, while on the right shoulder and in the domain further right to it, the system is in a pure liquid state. The difference

![FIG. 1. Isotherms calculated for an \(^{197}\text{Au}\)-like system. Dotted lines illustrate isotherms for hypothetical single-phase matter, while the dashed line visualizes the boundary of the liquid-gas coexistence domain.](image1)

![FIG. 2. Liquid-gas coexistence line in the temperature versus matter density representation, as predicted by the present formalism.](image2)

![FIG. 3. Isochoric caloric curves for boxed pseudo-Fermi matter, calculated for different volumes of the confining box, as indicated by labels.](image3)
The search for the minimum free energy for a given temperature \( T \) and a given volume \( V \) yields the total excitation energy \( E^* \) of the boxed matter, along with the corresponding volumes \( V_{\text{gasliquid}} \) and densities \( \rho_{\text{gasliquid}} \) of gaseous and liquid phases. This allows one to construct isochoric caloric curves for the modeled system at different volumes of the confining box. A set of such caloric curves is illustrated in Fig. 3. As expected, these curves do not feature plateaus of the kind reported in various experimental studies [8], but show rather inconspicuous kinks at the locations on the boundary of the coexistence domain.

**IV. CRITICAL BEHAVIOR**

One of the salient features of critical behavior in van der Waals systems is the presence of a singularity at the critical point. When the system temperature \( T \) approaches the critical temperature \( T_c \), the difference between the densities of coexisting liquid and gaseous phases is predicted to vanish according to a power law

\[
\rho_{\text{liquid}} - \rho_{\text{gas}} = C(T_c - T)\beta.
\]

In Eq. (17), \( C \) is a constant and \( \beta \) is the critical exponent. The magnitude of the critical exponent can be extracted conveniently by fitting a straight line to a double-logarithmic plot of \((\rho_{\text{liquid}} - \rho_{\text{gas}})\) versus \((T_c - T)\). Such a plot, obtained in the present calculations, is shown in Fig. 4. The plot features, indeed, an approximately 2-MeV-wide linear domain extending from the critical temperature \( T_c \), down to lower temperatures. A linear fit to this domain allows one to extract the "coordinates" of the critical point of \( T_c = 10.0 \) MeV, and \( \rho_{\text{liquid}} / \rho_{\text{gas}} = 0.42 \). Furthermore, it yields a value of \( \beta = 0.51 \) for the critical exponent.

Given the schematic nature of the present formalism, the predicted characteristics of the critical point are well within a reasonable domain that can be inferred from more sophisticated, but less transparent calculations.

**V. DISCUSSION**

A simple formalism has been presented that allows one to model the behavior of interacting neutral, isosymmetric pseudo-Fermi matter under the conditions of controlled volume and temperature. The formalism is shown to capture the essential physics of a first-order liquid-gas phase transition. The calculation demonstrates the characteristics of a nuclear liquid-gas coexistence in a certain domain of system parameters and the presence of a critical point. While such characteristics are well expected, based on the similarity of the utilized form of the nuclear interaction to that of the van der Waals interaction, the present formalism offers many didactic benefits. For example, due to its simplicity, it offers clear insight into physical phenomena it purports to describe and a relatively strict test bench for possible qualitative or "handwaving" arguments. The results obtained, while almost trivial, may alert one to possible challenges found by more complete models.

The formalism presented here leaves ample room for further refinements, such as a more strict modeling of dilute Fermi matter, or the inclusion of isotopic and, perhaps, Coulomb effects. At any rate, it offers a convenient didactic tool to achieve a better understanding of nuclear thermodynamics.

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Caloric Curve for Mononuclear Configurations

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The caloric curve for mononuclear configurations is studied with a schematic model. We investigate the dependence of the entropy on the density and effective-mass profiles. In finite nuclei, a plateau in the caloric curve is a consequence of decreasing density and the destruction of correlations rather than an indication of phase coexistence. The mononuclear regime is metastable with respect to binary fission at low excitation energy and with respect to multifragmentation at high excitation. The statistical framework presented here is suitable to treat scenarios where experimental conditions are set to favor a population of highly excited mononuclei.

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Heavy compound nuclei (CN) are metastable objects, i.e., local mononuclear entropy maxima that are separated from the more stable dinuclear states by transition regions of significantly lower entropy. Standard statistical-model treatments of CN decay are predicated on a time-scale separation between the CN formation time and the time scales for simple (mostly single-particle) decay modes as well as the massively collective decay processes we call fission. The former are usually treated using the prescriptions offered by Weisskopf [1] or Hauser and Feshbach [2], while the latter are usually treated by a transition-state formalism initially developed for chemical reactions by Erying [3]. The distinction between decay modes can be bridged in concept [4] and practice [5]; however, the massively collective decay channels can be retarded by transient delays [6].

It has been known for almost four decades that, below approximately 1/3 of the saturation density ρ∗, α matter has a lower free energy than uniform nuclear matter [7,8]. However, mononuclear configurations at reduced density can be reasonable subjects for statistical decay treatments as long as they are either metastable or protected from massively collective decays modes by transient delays. Specifically with increasing excitation energy, an equilibrium (i.e., local maximal entropy) mononuclear density profile can be reached, the decay of which can be treated with only minor modifications to the well known formalisms, as long as a time-scale separation exists between formation and all conceivable decay modes.

This work does not deal with the important issue of time-scale separation [9], only with the gross statistical properties of mononuclei at high excitation energy. In particular, we show that the relaxation of the density profile of mononuclei, in pursuit of maximal-entropy, causes the caloric curve T(ε) (where T is the statistical nuclear temperature and ε is the excitation per nucleon) to flatten out and exhibit a quasiplateau. We believe this explains the nature of the caloric curve first studied by Wada et al. [10] and later by Pochodzalla et al. [11], and for which systematics have recently been analyzed in detail by Natowitz et al. [12].

The approximate saturation of the statistical temperature is primarily due to density reduction, but it is also influenced by the evolution of the effective mass of nucleons in the nuclear medium. The first effect is just the sequestration of energy in the potential energy of nuclear expansion. The energy spent on expansion reduces the thermal part of the total excitation in much the same way as the collective rotational energy does in the case of high angular momenta CN.

The ratio of the effective mass to the bare nucleon mass m∗/m differs from one due to the finite range of the nuclear force and the time nonlocality of the interaction. The former effect, which is responsible for making the optical model potential energy dependent, reduces m∗/m by a density dependent factor m∗(ρ) which must return to one at low density. The time nonlocality can be thought of as the coupling of low-lying surface modes to single-particle degrees of freedom [13,14]. This collective effect brings states down from high energy, increasing the many-body density of states at low excitation energy. The effective-mass factor, m∗(ρ′, T), accounting for this relocation of levels, while greater than one at low energy and localized on the surface of the quantum drop, must return to one in the limit of high excitation or low density gradient.

We confine our analysis to a one parameter description of expansion and the literature descriptions of how the effective-mass terms evolve with density and excitation. Our approach combines the physically transparent picture of maximal-entropy mononuclear configurations found in the recent work by Töke et al. [15], with the effective-mass change with excitation energy found in the works of Natowitz, Shlomo, and collaborators [16].

The dominant term in the expression for the entropy of a quantum drop of degenerate Fermi liquid can be written...
as [17]

\[ S = 2\sqrt{a U} = 2\sqrt{\alpha (E_T - E_K)} = 2\sqrt{\alpha A (\varepsilon - \varepsilon_E)}, \tag{1} \]

where \( a \) is the level-density parameter and the thermal, total, and expansion energies are \( U, E_T, \) and \( E_K, \) respectively. With total particle number \( A, \varepsilon \) and \( \varepsilon_E \) are the total and expansion energies per particle. In the local density approximation (LDA) [18], the level density depends on the nuclear profile, the local Fermi momentum \( k, \) and the effective-mass [19,20],

\[ a = \frac{\pi^2}{4} \sum_{\tau} \int \frac{\rho_\tau(r)}{\hbar^2 k_{\tau}^2(r)/2m^*} \, dr. \tag{2} \]

The density profiles \( \rho_\tau(r) \) of the two isospin partners (with index \( \tau \)) are taken to be the same functional form, scaled in proportion to the number of nucleons. The native \( (\varepsilon = 0 \text{ MeV}) \) radial profiles are of the “standard” type with a Gaussian derivative,

\[ \rho_n(r) = \frac{\rho_\tau}{2} \left[ 1 - \text{erf} \left( \frac{r - R_o}{\sqrt{2} b} \right) \right], \tag{3} \]

with effective sharp radius \( R_o = r_o A^{1/3} \) \((r_o = 1.16 \text{ fm})\) and surface width \( b = 1.0 \text{ fm}. \) The expansion is limited to the one-dimensional self-similar family, i.e., \( \rho(r, c) = c^3 \rho_n(cr) \). The expansion parameter \( c \) is found by maximizing the entropy,

\[ \left( \frac{\partial S}{\partial c} \right)_\varepsilon = 0. \tag{4} \]

The collective energy involved in expansion is taken as the simple upside down bell shaped form, involving only the central density, suggested by Friedman [21], \( \varepsilon_E(c) = \varepsilon_E(1 - \frac{(c0.5)^2}{2}). \) We have used \( \varepsilon_E = 8 \text{ and } 6 \text{ MeV} \) in the present calculations. The energy required for expansion using \( \varepsilon_E = 8 \) is almost identical to that calculated (with Coulomb) using the logic of Myers and Swiatecki [22] and a nuclear matter compressibility coefficient of \( K_n = 234. \) In this schematic model, \( \varepsilon_E = 6 \text{ MeV} \) simply implies a 25% reduction in the energy cost for expansion.

Execution of Eq. (4) not only finds the metastable mononuclear expansion but also ensures that the surface pressure is zero. This procedure is therefore logically different from the physically unreal but true equilibrium condition found by placing a drop in a box and having a surrounding vapor supply a pressure.

We choose the phenomenological form for the effective mass suggested by Prakash et al. [20] and used by De et al. [23]:

\[ \frac{m^*}{m} = (m_i)[m_w] = \left( 1 - \alpha \frac{\rho(r,c)}{\rho_o} \right) \left[ 1 - \beta(T) \frac{\rho'(r,c)}{\rho_o} \right], \tag{5} \]

with \( \alpha = 0.3, \) \( \beta(T) = 0.4 A^{1/3} \exp[-(TA^{1/3}/21)^2]. \) (The \( T \) dependence requires knowing the caloric curve \( T(\varepsilon) \).) We solve this problem by iteratively starting with the \( m^*/m = 1 \) caloric curve. This iteration ensures that the \( T \) is uniquely determined by \( \varepsilon \) and satisfies the stationary condition.) The effective-mass factor is suppressed in the bulk, peaks at the surface [24], and degrades to one with decreasing density and increasing thermal energy. These two many-body effects, to a large extent, offset one another in near ground-state nuclei, yielding \( a = A/8 \) for unexpanded \(^{199}\text{Au}, \) the nucleus considered here. However, the destruction of the cooperativity encoded in these two effective-mass terms does not occur on identical energy scales. While the detailed density and the excitation energy dependence of these terms are unknown, the present work shows how the gross effects captured by these terms couple with expansion to dictate the form of the caloric curve.

The excess entropies above the unexpanded \((\varepsilon = 1)\) native shape are shown in Fig. 1. The maximum entropy determines the equilibrium expansion and mononuclear entropy \( S_M(\varepsilon). \)

The reduction of the equilibrium central density with excitation \( \rho_x(\varepsilon) \) (at the extremum in entropy) is shown in Fig. 2(a) for these cases: \( m^*/m = 1 \) with \( \varepsilon_E = 8 \) \text{ MeV} and \( m^*/m = m_i(\rho)m_w(\rho', T) \) for both \( \varepsilon_E = 8 \) and 6 \text{ MeV}. Consideration of \( m_i(\rho) \) alone exhibits a reduction in the central density similar to that with \( m_i(\rho)m_w(\rho', T), \) while consideration of \( m_i(\rho)m_w(\rho') \) leads to approximately the same \( \rho_x(\varepsilon) \) as with \( m^*/m = 1. \) Without the effective-mass terms, the \( \rho_x(\varepsilon) \) dependence is almost identical with the (extended) finite-temperature Hartree-Fock calculation reported in [16]. As one should expect, the decrease in density is more substantial with the reduced energy cost of expansion. The central densities implied by the maximal-entropy procedure used here with \( m^*/m = m_i(\rho)m_w(\rho', T) \) and \( \varepsilon_E = 6 \) \text{ MeV} are similar to those extracted from caloric curve data (diamonds) reported by Natowitz et al. [16]. On the other hand, \( \rho_x(\varepsilon) \) does not

![Figure 1](https://example.com/fig1.png)

**FIG. 1.** Representative calculation of total excess entropy as a function of expansion.
As a result of the extremum condition used to determine the entropies used in Eq. (7), the expansion parameter $c$ is uniquely determined by the excitation energy and the condition of zero external pressure. The latter condition implies that no thermodynamic work is done by nuclear expansion.

As shown in [15] and implied in [16], the relaxation of the density profile substantially flattens the temperature rise with $\varepsilon$ (compare black dotted and solid curves). The inclusion of the effective-mass evolution increases $T$ for $\varepsilon < 3$ MeV and decreases it for $\varepsilon > 5$ MeV, changes that give the appearance of a plateau. Decreasing the compressional energy constant reduces the value of the temperature of the pseudoplateau.

Figure 3 shows how the level-density parameter $a$ evolves along the metastability ridge. Without effective-mass considerations ($m^*/m = 1$, dotted line) $a$ just increases uniformly as the density drops with increasing $\varepsilon$. If only the $m_k$ factor is included (dashed line), $a$ is initially suppressed but grows faster with $\varepsilon$. As the density of states grows with $m^*$, the increase in the $m_k$ factor provides positive feedback to the expansion process. However, adding the $m_q$ dependence (solid line) provides a surface enhancement at low excitation, an enhancement which dies by $\varepsilon \sim 2$ MeV. We can therefore make the following three statements. First, consideration of a momentum dependent interaction ($m_k$ factor) is required to predict the expansion rate with $\varepsilon$. Second, as the change in the surface enhancement to $a$ is essential for producing the pseudoplateau, the plateau is a finite-size effect rather than an indication of phase coexistence. Third, the height of the pseudoplateau has to do with the energy cost of expanding the finite system, and thus the surface and the Coulomb energies are both important [26].

What relevance could these metastable mononuclei have to reaction observables? At low energy, the mononuclear density of states of $^{197}$Au is a reasonable subject for study because the fission barrier is sufficiently large that the mononuclear lifetime $\tau_M$ is longer than the characteristic time for equilibration within the mononuclear region of phase space $\tau_{eq}$. At higher energies,
the time required to thermally populate the extended fission shapes can transiently suppress the fission decay width, increasing the energy region where decay within the mononuclear family dominates the decay [6].

Because of the time required for shape equilibration, the condition $\tau_{eq} < \tau_M$ will be satisfied to higher excitation energies in light ion, $\pi$, and $\bar{p}$ induced reactions than for heavy-ion reactions. The decay width of equilibrated mononuclei will be the sum of the widths for decay within the mononuclear family and outside of this family into the multifragmentation channel. However, as is the case with fission, a transient delay (this time associated with the amplification of density fluctuations [9]) is likely to initially suppress the multifragment decay and increase the energy region over which the mononuclear entropy controls the decay process. After the transient delay, the multifragment decay will contribute in proportion to the multifragment density of states. To determine the latter, one needs $s_{MF}$, the entropy per particle for multifragmentation, which depends on the prescription used for the free volume [27,28]. A comparison of $s_M$ (for the case $m^*/m = 1$, at extracted temperatures) to $s_{MF}$ as a function of volume (taken from the ideal phase space model of Das Gupta and collaborators [29]) indicates the following: (i) The volume capturing 99.75% of the mononuclear matter is much smaller than reasonable volumes for multifragmentation. (ii) $s_{MF}$ will exceed $s_M$ somewhere above $\varepsilon > 3$ MeV if a freeze-out volume of 3 times the unexpanded volume is used. As the freeze-out volume is much larger than the mononuclear volume, mononuclei can still be considered metastable.

Finally, if an excited nucleus expands within the mononuclear family, it can undergo an irreversible transition to the higher entropy multifragmented state. One should expect event averages sampling such a process to yield fluctuations in excess of those for a reversible process. It is therefore reasonable to suspect that the observed excessive fluctuations of the kinetic energy [30] result from an irreversible transition from a lower entropy (larger free energy in a canonical treatment) but kinetically trapped, mononuclear phase to the multifragmented phase. This is equivalent to arguing that the ensemble (the event sample) is nonergodic [31]. However, as discussed above, this transition is not required to explain a plateau in a caloric curve.

This work makes use of several simplifying assumptions. These include the LDA [18], self-similar expansion and the expression for the entropy itself (which ignores the continuum and all detailed quantum structure), all subjects that deserve further study. Nevertheless, this work shows that a near plateau in the caloric curve should be expected for a finite nuclear system due to the evolving influence of the finite range of the interaction and collective effects as the system expands.

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[18] The major problem with the LDA is its sensitivity to the tails of the density profile. The profile chosen should be less sensitive than the standard Woods-Saxon. Our choice of a heavy nucleus also reduces this problem. See S. Shlomo, Nucl. Phys. A539, 17 (1992).
[28] As no stationary condition is used to determine $s_{MF}$, the thermodynamics problem is not well posed.
Status of the ZEPLIN II experiment

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Presented by J.T. White

Abstract

ZEPLIN II is a 30-kg two-phase xenon detector designed for direct detection of cold dark matter in the form of WIMPs. Currently in the commissioning phase, it will begin operation in the Boulby Mine, UK later this year. ZEPLIN II is capable of discriminating between nuclear recoils and background events and has a design reach up to two orders of magnitude beyond current limits. It will also serve as a step in the development program for a next-generation ton-scale detector. © 2005 Elsevier B.V. All rights reserved.

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1. Introduction

A 70-year series of astronomical observations beginning with Zwicky’s (1937) are best explained by the hypothesis that most of the matter in the universe, is in the form of some new type of non-relativistic (cold), non-luminescent (dark), weakly interacting, massive, elementary particle (WIMP). One of these observations is that the galactic halo appears to be composed predominantly of cold dark matter, which leads to the possibility of direct detection with a sufficiently sensitive underground detector. A number of experiments (Abrams et al., 2002; Benoit et al., 2002; Alner et al., 2005) have now achieved cross section limits for WIMP dark matter approaching the $10^{-6}$ pb/nucleon level, but the most promising theoretical candidate, the lightest neutralino in supersymmetric extensions of the Standard Model (Jungmann et al., 1996), might require a sensitivity three to four orders of magnitude greater before detection is possible.

A detector concept based on liquid xenon that appears to have the properties needed to achieve the required sensitivity has been developed by the ZEPLIN Collaboration. Based on studies carried out by UCLA/Torino/ICARUS (Cline et al., 2000) and low-background techniques and facilities developed by UKDMC, a staged series of experiments is now underway with the ultimate goal to reach at least four orders of magnitude beyond currently achieved limits. One that is currently in the commissioning stage is the ZEPLIN II experiment. It is designed to have a reach up to two orders of magnitude beyond current limits, and will also provide critical information needed for the design of the next-generation detector.

2. Detector concept and design

The ZEPLIN II experiment is based on the two-phase xenon concept (Dolgoshein et al., 1973). Particle interactions in the liquid target will produce both prompt scintillation (primary scintillation) and ionization. The ionization electrons are drifted to the surface where they are extracted from the liquid into the gas phase. In the gas, the electric field is sufficiently strong to cause electroluminescence (secondary scintillation). The key concept is that measurement of both the primary and secondary scintillation provides a method to discriminate between the electron recoils from gamma and beta interactions and the nuclear recoils from neutron and WIMP interactions.

In liquid xenon, this feature was demonstrated as shown in Fig. 1. (Cline et al., 2000). In this study, with a drift field less than 1 kV/cm, nuclear recoils were observed to produce an extremely low ionization signal as expected from considering recombination along the track of a heavy ionizing particle. The discrimination level was estimated to be $\sim 10^{-3}$ at low recoil energies.

Based on these results, the ZEPLIN II detector was designed to operate without requiring a secondary signal from nuclear recoils. As illustrated in Fig. 2, the 30-kg liquid xenon target is contained in a tapered PTFE basin with minimal dead space.
The relatively large secondary signal from electron recoils is efficiently detected and achieves the required discrimination.

The target is viewed from above by seven 5" low-activity VUV PMTs designed for ZEPLIN II (Electron Tubes 9372KFLQ). The drift field is set up between two fine stainless steel meshes, one on the bottom of the basin and one just below the surface of the liquid. A series of 10 OFHC copper rings outside the basin provide field uniformity. A third mesh just above the surface will be used to provide the extraction and electroluminescence field.

To achieve 100% extraction (Dolgoshein et al., 1973), the field in the liquid will be set to \( \sim 5 \) kV/cm, which implies an electroluminescence field in the gas of 10 kV/cm. The drift field will be of the order 1 kV/cm. The large ratio of the extraction to drift field, combined with the dimensions of the mesh, also assures 100% transmission of electrons from the drift region to the extraction region.

The spectra of both the primary and secondary scintillation light in xenon are centered at 175 nm. Simulations of the light collection efficiency, assuming a diffusive light reflectivity of 90% for the PTFE, indicate a light collection efficiency of \( \geq 3 \) photoelectrons per keV for the primary scintillation. This response is very uniform throughout the entire 30 kg volume. The electroluminescence yield in xenon is given by \( N_{\text{ph}} = 70(E - 1.3P)d \), where \( E \) is the field in the gas in kV/cm, \( P \) is the pressure in atm, and \( d \) is the drift distance in cm. Taking \( d = 0.5 \) cm, \( P = 2 \) atm and \( E = 10 \) kV/cm, the yield will be \( \sim 250 \) photons per ionization electron. This will result in a secondary scintillation signal \( \geq 10 \) photoelectrons per electron. For events with both primary and secondary scintillation, the 3D position of an interaction will be well determined by using the drift time to measure the depth of the interaction and the amount of light in each tube to determine the transverse position.

The data acquisition system is designed to trigger on \( N \)-out-of-seven PMTs at a threshold efficient for a single photoelectron. The signal is read out using an Acqiris digitizer system that will be set to digitize the signals in 2 ns bins for the period \( \pm 60 \) s. The configuration is designed to be efficient for triggering on the primary signal but will also allow detection of events with secondary signals, but with less than \( N \) phototubes having a primary signal.

3. Physics sensitivity

In situ measurement of critical parameters such as the light collection efficiency, low energy recoil response and background rate are required before the precise sensitivity of the experiment can be determined, but it is possible to make estimates based on indirect measurements and simulations. WIMP collisions in xenon will result in nuclear recoil spectra as, for example, those shown in Fig. 3 for WIMP masses of 50 and 100 GeV, respectively. Because of the nuclear form factor, most of the interactions will have recoil energies less than \( \sim 50 \) keV. The scintillation efficiency, \( f_{S\text{N}} \), for nuclear recoils with energies greater than 40 keV was measured (Arneodo et al., 2000) to be \( f_{S\text{N}} \sim 18\% \) as
shown in Fig. 4. Good agreement with the Lindard theory (Lindhard et al., 1963) is evident, which suggests that $f_N$ will decrease only slightly at lower energies. However, it is possible that the efficiency will increase slightly at lower energies as is the case in NaI(Tl) and similar crystals (Murray and Meyer, 1961). In either case, nuclear recoils up to 50 keV will appear in the electron-equivalent (e.e.) range less than 10 keV. By triggering on two or three photoelectrons ($\sim$1 keV e.e.), the detector will be quite efficient by $\sim$3 keV e.e., which translates to about 17 keV nuclear recoil energy. Thus, taking a recoil threshold of $\sim$20 keV and estimating possible backgrounds using ZEPLIN I experience and simulations, the reach can be estimated as shown in Fig. 5.

Because of the strong background rejection in ZEPLIN II, it will also be sensitive to an annual modulation signal for lower mass WIMPs. An estimate of the reach using the cosine projection method (Freese et al., 1988) is shown in Fig. 6. The curves shown are the cross sections at which a random signal is rejected at the 90% confidence level, while the efficiency for detecting an oscillation is 90%.

![Fig. 3. Nuclear recoil spectra from WIMP collisions in xenon for WIMP masses of 50 and 100 GeV.](image3)

![Fig. 4. Scintillation efficiency in liquid xenon (from (Arneodo et al., 2000)).](image4)

![Fig. 5. Estimated reach of ZEPLIN II.](image5)
4. Conclusion

ZEPLIN II surface tests will be complete this summer and the detector will be placed underground in the fall. It has a potential reach to the $10^{-10}$ pb/nucleon level, and will help provide the critical information needed to design the next-generation two-phase xenon detector with the goal to reach the $10^{-10}$ pb/nucleon range.

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References

ZEPLIN IV: A future large-scale liquid xenon dark matter detector


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Presented by Hanguo Wang

Abstract

We present a possible design of future large-scale dark matter detector using liquid xenon. A low energy threshold is achieved by using a large CsI-coated internal photocathode for primary light collection. A focusing field, in the active volume and an electron focusing structure at the liquid surface, are used to transport free-electrons to the luminescence field. A 3D reconstruction of the events can be achieved by timing and location of the luminescent signal. A liquid xenon Compton veto and self-shield are integrated in the compact design. This detector could be used to probe most of the SUSY predicted dark matter. ZEPLIN IV can be initially tested at Boulby and then be moved to the USA to operate at a US deep underground science and engineering laboratory.

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1. Introduction

The search for dark matter particles is among the most fundamental of all astroparticle physics goals. We know that at least 30% of the matter in the universe is due to this source. An excellent guide to the search is given by the SUSY–WIMP model. As new collider physics results have appeared the calculations for the rate of such WIMPs in dark matter detectors has gone down. Currently the expected value is less than $10^{-2}$ events/kg/day.
As current experiments Edelweiss, CDMS and ZEPLIN I Schnee, 2004; Stefano, 2004; Smith, 2004 nearing these sensitivity, new experiments ZEPLIN II, III, and CDMS II online soon will be able to probe even further. To explore most of the SUSY–WIMPs well below this level will require a ton-scale powerful discriminating detector.

The key issues of future large-scale detectors are background neutrons from muons, neutrons and gammas from U, Th in the rock and in the detector construction materials. Muon induced neutron can be reduced to acceptable level by going deep underground such as the SNOLAB or future US deep underground science and engineering laboratory (DUSEL) or by using muon veto system if operate at shallower site. Lab gammas and neutrons from U and Th in the rock can be shielded by lead or ion and modulated by low atomic number materials. U and Th contamination in detector construction materials can only be reduced by careful selection of materials and is limited by availability. To further reduce such background, the detector itself must be capable of discriminating background.

The goal of the ZEPLIN collaboration is to fully utilize the gained experience on xenon R&D and design an ultimate detector with very low energy threshold and powerful background rejection and operate it in a suitable site (Boulby, SNOLAB, or US DUSEL) to probe most (if not all) of SUSY predicted region.

2. ZEPLIN IV concept design

The key properties of liquid Xe and its principle use in discriminating detector design have been discussed extensively in literature (Cline et al., 2000; Wang, 1998; Benetti et al., 1993) and during this conference (Smith, 2004; White, 2004; Summer, 2005).

The goal of the ZEPLIN IV design is to achieve large active mass (one ton), low energy threshold (sub-keV), integrated Compton veto, integrated active neutron veto. Fig. 1 shows a cross-section view of the conceptual design.

An array of 37 2-in. PMT in the center looking down at an electroluminescent structure (described later) located at the liquid surface. Active xenon is surrounded by a xenon Compton veto and a PTFE cone. The active xenon and veto xenon are separated by a thin metal shell, which is at negative high voltage. The upper surface of the metal shell is coated with CsI, which act as an internal photocathode. The field shaping rings in the PTFE and the metal shell form a focusing field to guide ionization electrons and photoelectrons to drift upward to the electroluminescence field at the liquid surface. Sixteen 5-in. cryogenic VUV sensitive PMTs are used to measure scintillation light in the veto volume.

The key feature of this design, is that the use of large CsI-coated surface acts as an internal photocathode, significantly improves the primary light collection efficiency. Instead of measuring the primary scintillation light directly by the PMTs, the large CsI internal photocathode converts them into photoelectrons. These photoelectrons will be measured easily by PMT after they produce large amount of luminescent photons at the luminescence field. The high focusing drift field will extract ionization from background events. Due to the non-destructive nature of the liquid xenon Time-Projection-Chamber (TPC), all charges will drift...
toward the luminescent field with relative position un-changed, so a 3D event structure can be reconstructed. Notice that photoelectrons at the CsI surface will arrive at the luminescence field at different time because different location from the center has different drift length. At low energies, most electrons will arrive individually hence the PMT array will be able to measure each electron arrival time and location by weighted sum. The background discrimination is achieved by reconstructing the ionization yield at the event center and the photo-electron distribution on the CsI surface. Nuclear recoil events will have completely different charge yield at the event center compared to that of background events. Fig. 2 shows a 3D artistic view of the internal parts.

The integrated Compton veto also act as a self-shield and the whole detector is very compact (radius – 0.65 m, height – 1.3 m). This compact design allows a modest size gamma shield and neutron modulator. For shallow site test, it requires relatively small muon veto setup.

Neutron modulator can be easily fitted at the bottom of this detector and if loaded with Gadolinium, the Compton veto can be used as active neutron veto by capturing the gammas emitted from neutron capture by gadolinium. Detailed simulations are underway to optimize the geometry.

3. The luminescent structure

Large amount of secondary photons are produced when electrons arrive at the luminescence field. These photons may hit back on the CsI photocathode and hence produce more
photoelectrons. To prevent this positive feedback, a special structure can be used to stop the backwards light and allow electrons drift upwards. This structure consists of three pieces, a thin three layered plate with many small holes is placed in the liquid near the surface, a thicker plate with larger hole (half immersed in liquid) and a thin mesh above the thick plate in the xenon gas. The three layered thin plate will be used as an electron-optics to guide electrons go through the small holes. The holes have 2.5% of the total area, so most light will be blocked by this plate alone. The thicker plate will be used to further block side-moving lights while letting electron pass through the holes. The mesh in the gas combined with the thick plate provide strong field for electroluminescence to occur. The detail of this structure is shown in Fig. 3.

Fig. 4 shows the test results of the thin plate in gas argon with 10% CH₄. This gas mixture simulates the low electron diffusion in liquid xenon. The results show that the electron transfer is as predicted by the calculation. With correct field configuration, electron transparency is 100%. The backwards light blocking is simply the geometrical effect. The overall light leak is less than $1.6 \times 10^{-4}$.

Light collection efficiency is 45.0–90.0% depending on location. Assuming 30% CsI quantum efficiency and 30 eV/p.e. then the number of
photoelectrons at CsI surface for 1-keV gamma is estimated to be 5–9 photoelectrons.

This large mass design with very low energy threshold and powerful background discrimination will enable us to probe most of the SUSY predicted dark matter parameter space.

The geometrical shape of the design makes the detector construction and installation relatively simple. Detector cooling and signal feedthroughs are similar to that of ZEPLIN II.

Fig. 5 shows the expect goal of such detector. Also shown are the current ZEPLIN I limit and expected ZEPLIN II sensitivity.

4. Summary

We have shown a possible design of the future ZEPLIN IV using two-phase xenon as detector target. If correct, this would make ZEPLIN IV the most massive and sensitive WIMP detector currently being studied. The dark matter signal levels expected from most SUSY models (Nath and Arnowitt, 1995; Roszkowski; Bottino et al., 1994) could ultimately require the sensitivity in Fig. 5 and the construction of ZEPLIN IV.

Acknowledgments

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References

Roszkowski, L., 2004. This Conference.
Schnee, R., 2004. This Conference.
Smith, NJT., 2004. This Conference.
Stefano, PD., 2004. This Conference.
White, JT., 2004. This Conference.
IV. Publications and Activities

IV.A. Articles

1. “Cluster Emission in Complex Nuclear Reactions”

2. “The Nuclear Caloric Curve for Mononuclear Configurations”
   L.G. Sobotka, R.J. Charity, J. Töke, and W. U. Schröder,

3. “Retardation of Particle Evaporation from Excited Nuclear Systems Due to Thermal Expansion”
   J. Toke, L. Pienkowski, M. Houck, Jun Lu, and W.U. Schröder
   Phys. Rev. C 72, 031601(R) (2005), Rapid Communication


5. “Status of the ZEPLIN II experiment”

6. “ZEPLIN IV: A future large-scale liquid-xenon dark matter detector ”

7. “Light Charged Particle Production in 1.2~GeV-Proton Induced Spallation Reactions on Al - Th”

8. “Pulse Shape Method with Large Area Planar Silicon Detectors of the CHIMERA Array ”

9. "Systematic investigation of 1.2-GeV proton-induced spallation reactions on targets between Al and U"

IV.B. Invited Lectures and Presentations

"Nuclear Science at the University of Rochester,"
Graduate Research Seminar, Dept of Physics and Astronomy, University of Rochester, October 5, 2004
(by W. U. Schröder)

Modeling of Fragment Formation: Surface Entropy Driven Nuclear Decay
Int. Worksh. World Consensus In. 3, Texas A&M University, Feb. 12-16, 2005
(by J. Töke)

"40 Years Reports from the Scientific Front,"
Fest Colloquium in Honor of D. Hilscher, Hahn-Meitner Institut Berlin, Feb. 28, 2005
(by W. U. Schröder)

"Surface Cluster Emission Following Dissipative Reactions,"
Seaborg Award Symp. honoring L.G. Moretto, Meeting of the American Chemical Society, San Diego, March 13-16, 2005
(by W. U. Schröder)

"Isotopic Effects in Particle Emission from Complex Nuclear Reactions," Workshop "Nuclear Equation of State for Nuclei, Neutron Stars, and Supernovae", Arkansas State University, Jonesboro, April 14, 2005
(by W. U. Schröder)
IV.C Professional Activities

Member of USDOE Review Panel to evaluate nuclear physics and chemistry research program at the Texas A&M University Cyclotron Laboratory, Sept. 9-10, 2004

Fellow of the American Physical Society, Division of Nuclear Physics
Member of the American Chemical Society, Division of Nuclear Chemistry and Technology
Member of the American Association for the Advancement of Science
Member of the American Association of University Professors
Vice-President of University of Rochester Chapter of the AAUP

Co-editor of book on Isospin Effects in Heavy-Ion Reactions

Referee for Journal Articles and Proposals to Funding Agencies

Appointed Liaison Scientist for several nuclear research groups in CIS countries (Russia, Usbekhistan)

Member and Chair of ad hoc Committees, University of Rochester
Member of Radiation Safety Committee, University of Rochester
Member of Radiation Safety Advisory Committee, University of Rochester
Member of Oversight Committee for River Campus Instrument Machine Shop, University of Rochester

Chairman and/or member of Ph.D. examining committees, University of Rochester

Member of H.E. Gove Prize Award Committee, University of Rochester, Department of Physics
V. Personnel

Dr. W. Udo Schröder, Professor of Chemistry and Physics,
Dr. Jan Tõke, Senior Scientist
Dr. Wojtek Gawlikowicz, Research Associate
Mr. Mark Houck, Graduate Student, Department of Physics
Ms. Iwona Pawelczak, Graduate Student, Department of Chemistry
Mr. Zachary Chambers, Graduate Student, Department of Chemistry
Mr. Michael Quinlan, Undergraduate/Graduate Student¹, Department of Chemistry

¹Since June 1, 2005

Visitors

Dr. A. Bonasera, LNS Catania, Italy
Dr. A. Grszechuk, University of Kattowice, Poland
Dr. M. W. Johnson, Los Alamos National Laboratory