STUDIES OF HEAVY-ION REACTIONS AND TRANSURANIC NUCLEI

W. Udo Schröder
Principal Investigator

September 2009
Prepared for

The US Department of Energy
Agreement No. DE-FG02-88ER40414
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Progress Report for the Period
September 1, 2007 – August 31, 2009

W. Udo Schröder
Principal Investigator

University of Rochester, Department of Chemistry
Rochester, New York 14627-0216

September 2009

Prepared for
THE UNITED STATES DEPARTMENT OF ENERGY
UNDER GRANT NO. DE-FG02-88ER40414

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I. Abstract

The nuclear chemistry group at the University of Rochester studies the interactions between atomic nuclei and their behavior in highly excited, meta-stable states close to equilibrium. Ongoing investigations of the flow of energy and entropy from internal degrees of freedom into the fragments emerging from the fission-like decay of such nuclei have implications for our understanding of the microscopic, quantal structure of nuclei, as well as for applications of basic energy science. The group’s experimental and theoretical research program provides scientific training for students at different stages of their education.

Specifically, progress has been made in modeling the expansion of a nucleus growing in size with increasing excitation until it softens and loses cohesion, first at its surface. While phenomena associated with nuclear disintegration appear like indications of a liquid-gas transition of phase, the process is understood more naturally in terms of dramatic changes in the shape of nuclei in extreme states. The theoretical understanding gained by the group also explains how the quantal isotopic (isospin) symmetry energy reveals itself in experiments, mainly as a function of nuclear matter density.

Quantum mechanical isospin effects have also been studied in collisions between heavy ions. Here, the effects are more difficult to interpret in terms of the underlying structure and/or dynamics, since particle emission can occur on a fast time scale, before the colliding heavy-ion system can equilibrate, as well as during later stages. In any case, extensive simulations of potential underlying reaction scenarios have been found necessary to arrive at a sound appreciation of observed reaction phenomena. Comparative studies of correlated emissions of light and intermediate-mass nuclear fragments performed by the group have demonstrated deficiencies in current reaction theory in predicting the experimental observations. It appears that processes leading to copious emission of nuclear clusters are strongly influenced by the dynamics of violent nuclear collisions and not easily interpreted in terms of statistical models of disintegration.

In addition to theoretical and experimental projects in low-energy nuclear science, the nuclear chemistry group has also developed new instruments and methods for such research, for example several electronic modules have been built. A new type of neutron detector has been designed, a prototype module of it has been built and tested to satisfaction. A set of radio-chemical experiments on tritium transport has been designed and operated in collaboration with other university departments. These experiments have relevance to basic nuclear energy development and provide unique training opportunities for students at the University of Rochester.

The group has contributed greatly to successful efforts to improve the academic nuclear science infrastructure at this university, which is now looking forward to the commissioning of an advanced nuclear science education laboratory, the first in the undergraduate and graduate curriculum.
# TABLE OF CONTENTS

I. Abstract .......................................................................................................................... 5  
   Table of Contents ........................................................................................................... 7  
II. Introduction ................................................................................................................... 9  
III. Research Program ......................................................................................................... 19  
   A. Common signatures of statistical Coulomb fragmentation of highly excited nuclei and phase transitions in confined microcanonical systems .......... 21  
   B. Isoscaling in statistical fragment emission in an extended compound nucleus model ........................................................................................................ 50  
   C. Surface entropy and density dependence of the symmetry energy in a harmonic interaction Fermi gas model ................................................................. 66  
   D. Isospin effects in heavy-ion collisions: Results from CHIMERA experiments and prospects with radioactive beams .......................................................... 82  
   E. Evidence for an impact parameter dependence of dynamical intermediate-mass fragment formation in Ca+Sn reactions at 45 A MeV ...................... 88  
   F. Correlations between reaction product yields as a tool for probing heavy-ion reaction scenarios .............................................................................................. 110  
   G. Use of fission fragment kinetic energies and mass asymmetry in the analysis of the dynamical fission of fast projectile-like fragments ......................... 128  
   H. A simple method for rise-time discrimination of slow pulses from charge-sensitive preamplifiers ...................................................................................... 137  
   J. Phaser 8-A phase-locked RF prescaler for CHIMERA experiments ................. 145  
   K. Development of a novel neutron detector and performance of its prototype .......................................................................................................................... 148  
   L. Dependence of desorption rate of HTO from metal samples on temperature and carrier gas humidity ........................................................................... 180  
   M. Tritium removal from stainless steel samples by plasma induced sputtering .......................................................... 206  
   N. Ambient and neutron induced gamma background in the UR nuclear science laboratory .......................................................................................... 212  
   O. Development of the University of Rochester Advanced Nuclear Science Education Laboratory ANSEL ........................................................................... 227
IV. Publications and Activities ...............................................................235
   A. Articles ..........................................................................................235
   B. Invited Lectures and Presentations ...............................................236
   C. Contributed Papers/Abstracts .......................................................237
   D. Professional Activities .................................................................238

V. Personnel .......................................................................................238
   Visitors ..............................................................................................239
II. Introduction

This report describes the research activities and results obtained by the University of Rochester (UR) Nuclear Chemistry group over the period from September 2007 through August/September 2009. The research program of the group is supported by the United States Department of Energy under Grant DE-FG02-88ER40414. The group has an active experimental program at the Laboratorio Nazionali del Sud in Catania/Italy which is supported by the Italian INFN.

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), Indiana University (Bloomington), Oregon State University (Corvallis), as well as with nuclear physics groups at the Universities of Warsaw and Cracow, Poland. We have also continued a technical collaboration on transport phenomena of radioactive species in materials and on neutron diagnostics in fusion with the University of Rochester Laboratory for Laser Energetics (LLE), which is also supported by the U.S. DOE. These projects are important for nuclear fusion and fission basic energy science research and applications. In addition, they provide educational opportunities for our undergraduate and graduate students, as well as for summer interns, who get introduced to radio-chemical experimentation.

Moreover, in collaboration with a nuclear physics research team of the UR Department of Physics and Astronomy, we are building a set of experiments for a new Advanced Nuclear Science Education Laboratory (ANSEL), which for the first time in the history of this university introduces a dedicated nuclear science course into the University of Rochester undergraduate science curriculum. In helping to set up and run ANSEL experiments of varied complexity, our students acquire hands-on experience with nuclear instruments and methods for radiation detection and applications. The broad interest expressed by the University community (Chemistry, Physics, Geology, Engineering, Medical Physics and Medical Imaging, Health Physics) in the ANSEL laboratory, as well as our group’s technical collaboration with the LLE provide strong and objective arguments supporting longer-term prospects of nuclear chemistry, science and technology at the University of Rochester.

The main thrust of the group’s activities is directed onto basic nuclear research and has focused on the exploration of nuclear reaction and decay mechanisms induced by
intermediate-energy heavy ions. The most important results obtained during the past grant period include

- A further theoretical development of our ideas on the importance of an expansion of hot nuclei for their statistical decay and the role played by the surface entropy in the process;
- A better understanding of the influence of the density and temperature dependent nuclear symmetry energy on mass-to-charge division in nuclear decay ("iso-scaling");
- The realization that features often attributed to a nuclear liquid-gas phase transition are more easily and naturally understood as realizations of a different transition, one between different nuclear configurations, similar to shape transitions known from nuclear structure studies;
- 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 the Coulomb barrier for emission, the statistical independence of clusters emitted in heavy-ion reactions, the Arrhenius-like behavior of cluster multiplicities and, most recently the phenomenon of bimodality.

The above theoretical ideas have now cumulated in the formulation of a comprehensive view of the nuclear decay phenomenology. It has been shown that a consistent account of the expansion of a hot nucleus and an incorporation of thermal surface fluctuations lead to a unified understanding of the “classical” compound nucleus decay, of fission-like processes, of intermediate-mass fragment production, and of multifragmentation. Furthermore, it has been shown that such a unified compound nucleus phenomenology including surface fluctuations and 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. It is also now understood why and how other types of ad hoc model calculations arrive at predictions of unusual observations relating to, e.g., negative nuclear heat capacities or to apparent bimodality of experimental product distributions.

We are currently in the course of applying the HIFGM to the interpretation of isotopic ("isospin") dependencies of experimental observables in terms of the underlying symme-
try energy, i.e., one represented by the liquid-drop model symmetry energy coefficient $C_{\text{sym}}$. The underlying symmetry effect has a quantal origin in the Pauli Exclusion Principle. From this principle one expects that a diluted equilibrium matter density, which is reached upon expansion by a highly excited nucleus, can greatly decrease the efficacy of the Pauli Principle. On the other hand, one expects a relatively small effect of a dilution of just the momentum space density, when the spatial matter density distribution is kept constant. Even at the highest temperatures sustainable by a realistic meta-stable nucleus, the momentum distribution is well approximated by that of a degenerate Fermi gas. Therefore, the effective symmetry energy should depend dominantly on the diluted nuclear matter density but less so on its temperature. These considerations suggest that the nuclear liquid-drop symmetry energy reveals itself in experiments involving nuclei even at high excitations. However, a realistic interpretation of experimental data in terms of the symmetry energy has to resort to a model for the matter density distribution of such highly excited nuclei. The HIFGM description of hot nuclei advanced by our group is well suited to provide a translation between experimental observables and desired fundamental nuclear properties as demonstrated in several reports included below.

In the context of a broader view of nuclear decay, particle evaporation rates from excited nuclear systems at (diluted) equilibrium matter densities have been studied within the Harmonic-Interaction Fermi Gas Model (HIFGM) combined with Weisskopf’s detailed balance approach. Based on this extended compound nucleus model, we have investigated isospin effects in statistical fragment emission from excited nuclear systems. An experimentally observed scaling behavior of the ratio of isotope yields $Y_i(N;Z)$ from two similar emitting sources with different neutron-to-proton ratios is predicted also theoretically, i.e., the relationship of $Y_2/Y_1 \propto \exp(\alpha N + \beta Z)$ is demonstrated within the HIFGM. The symmetry energy coefficient $C_{\text{sym}}$ extracted from first simulation results is $C_{\text{sym}} = 27$ MeV which is consistent with other realistic theoretical estimates and fits recent experimental data. The influence of the surface entropy on the resulting isoscaling behavior is discussed in detail. It is found in this now published work that, although the surface entropy increases the numerical values of isoscaling parameters $\alpha$ and $\beta$, it does not affect qualitatively the isoscaling behavior and has only a minor effect on the extracted symmetry energy coefficient.

In a related article, readied for submission, our model is shown to predict the experimentally observed decrease of the isoscaling parameter $\alpha$ and the symmetry energy
with increasing excitation energy. To predict a reduced symmetry energy for diluted matter is the essence of the HIFGM which models the expansion of a hot nucleus in a manner consistent with overall liquid drop energetics. On this basis, a density dependence of the symmetry energy of \( C_{\text{sym}} = 25.2(\rho/\rho_0)^{0.46} \) or \( C_{\text{sym}} = 22.0(\rho/\rho_0)^{0.71} \) is derived from our simulation for the cases in which the surface entropy effect on the fragment emission is either considered or not. It is found that surface entropy has a noticeable effect on the observable symmetry energy coefficient.

The above theoretical calculations, carried out with a very robust interacting-Fermi gas model, provide strong motivation for future experimental studies of the isospin dependence of the nuclear symmetry energy (or the iso-equation of state). The most promising domain that can be explored is that of densities below the saturation density, \( \rho < \rho_0 \). Here, the complicating factors associated with finite nuclear temperatures can be treated well in low-order approximation. In comparison, phenomena observed in the domain of high densities, \( \rho > \rho_0 \), is more difficult to investigate because of very strong disequilibrium effects and alterations in the consistency of the matter studied. Already at bombarding energies of \( E/A > 50 \) MeV, the onset of pion production softens the hadronic equation of state in comparison to the nuclear EoS.

Considerations of an isotopic dependence of reaction phenomena have influenced the strategy we have followed in our experimental research. The UR nuclear chemistry group has been involved in several experimental projects in which comparisons can be made between reactions induced by projectile/target systems with different isospin asymmetries. As constituent of the CHIMERA collaboration at LNS Catania, our group has derived important information on the time scales and the mode of intermediate mass fragment emission. A survey over some recent results is given, together with an outlook on very interesting opportunities available at LNS for isospin studies using secondary, rare isotope beams (RIB) in conjunction with the multi-detector array CHIMERA. Several of our younger group members have been associated with tuning of the new CHIMERA beam line and transmission measurements for RIBs generated by in-flight fragmentation. Their experience with RIB facilities abroad will be useful for future experiments at the new F-RIB facility under construction.

The high efficiency and granularity of detector arrays such as CHIMERA come at the expense of great efforts that have to be devoted to the analysis of data. Analysis of our
comparative experiments on the reactions $^{40}$Ca + $^{112}$Sn and $^{48}$Ca + $^{112,124}$Sn at 45 A MeV have now progressed far enough to be able to report evidence for an impact parameter dependence of dynamical intermediate-mass fragment formation for several of the projectile/target pairs. In the exclusive $4\pi$ CHIMERA experiment (PhD project of MQ), projectile-like fragments (PLF) were measured in coincidence with IMFs and light-charged particles (LCP). A significant fraction of the measured events show production characteristics consistent with a dynamical formation mechanism. This assignment is made on the basis of the measured relative velocity between the fragments as well as the asymmetry of their decay patterns. The decay patterns are shown to be sensitive to variables canonically associated with the collision centrality. They appear to exhibit non-equilibrium production of IMFs with an interesting forward/backward alignment. While such emission mechanisms are interesting in their own right, non-equilibrium modes have to be identified and separated from statistical processes that carry information on equilibrium phases.

It is worth pointing out that current reaction theory has not been able yet to provide a consistent, quantitative account of the reaction phenomena experimentally observed in with heavy ions at intermediate bombarding energies. The anecdotal successes of models in “adaptive” reproductions of select inclusive data sets should not be taken as consistent evidence for a given reaction scenario. In a paper that has already been submitted for publication, the performance of several reaction models is demonstrated in terms of the respective explanations offered for correlations between reaction product yields such as we have measured for $^{136}$Xe+$^{209}$Bi reactions at E/A= 28, 40 and 62 MeV. This work also illustrated the strong influence an experimental “filter” can have on the appearance of correlations, even if all reaction products including neutrons are measured in coincidence and in $4\pi$. Rare data sets such as obtained by collaborations led by our group are instrumental in demonstrating the remaining significant discrepancies between theory and data. Similar data sets for several heavy-ion systems have already been measured and analyzed by our group and will be published in the near future.

It is perhaps already surprising that heavy-ion reactions at intermediate bombarding energies, where most primary reaction products are expected to emerge in highly excited states, presumably to disintegrate into a potentially complex mix of secondary fragments and light particles, are at all amenable to a detailed analysis. An even greater surprise is that the final product distributions do exhibit characteristic structure, often deceptively simple ones. An edifying example of the importance of detailed model simu-
lations for the verification of presumptive reaction scenarios is discussed in a contribution to this report, which will soon be made available in the open literature. The article investigates possible dangers of using PLF fission fragment data alone for the evaluation of PLF excitation energies. It is pointed out that for mass-asymmetric fission, the systematic errors of such a method depend noticeably on the orientation of the fission axis, which may result in a faux forward-backward asymmetry of the reconstructed angular distribution of PLF fission fragments. Accordingly, purely statistical mass-asymmetrical fission of the PLF may incorrectly appear as a fast dynamical process taking place in the immediate vicinity of the TLF. Our study also suggests that such instrumental biases can be minimized by data sorting with respect to light-particle multiplicities. Detailed simulations of this kind are underway for the reactions $^{40}$Ca + $^{112}$Sn and $^{48}$Ca + $^{112,124}$Sn at 45 A MeV discussed previously, before final conclusions can be drawn as to the dominant mechanism of IMF production.

In addition to theoretical work, execution and analysis of experiments, our group has also developed new and/or more efficient nuclear detection and particle identification methods for a variety of applications, including use in heavy-ion reaction studies. For example, in a recent publication we demonstrate the performance of a simple method of particle identification via pulse rise time discrimination. The identification method is applicable in particle experiments involving large-area silicon detectors but is easily adaptable to other detectors with a response corresponding to pulse rise times that are significantly different for different particle species. The method is based on a comparison of the amplitudes of two pulses derived from charge sensitive preamplifier. It competes with more elaborate and expensive digitization methods. The method could see application in future fusion reactions with light RIBs.

In the operation of time-of-flight experiments with pulsed-beam accelerators, experimenters often encounter “wrap-around” timing ambiguities, which complicate the data analysis. This has been an issue for such work at the LNS Catania superconducting cyclotron since some time. To avoid this problem in future without the necessity for experimenters to monitor and manually adjust timing relative to observed beam pulsing, a programmable electronic module, the “Phaser 8,” has been designed and built. The module automates the synchronization of a pre-scaled RF timing signal with beam monitor signals. The module can be operated as an autonomous phase-locked prescaler or as a phase controller for an external prescaler.
In experiments studying energetic heavy ion reactions, spallation reactions induced by relativistic light particles and even in laser-induced nuclear fusion, often the most important signal is missed, the one corresponding to the emission of large numbers of neutrons. In addition, neutron beams in a wide range of energies are used in imaging and test applications. Because of the deep penetration of neutrons into materials, efficient neutron detectors are large and susceptible to background radiation of various types. Exploitation of neutron/gamma discrimination based on different decay times of, typically liquid, scintillator components is feasible and economic only for small detectors with low efficiencies. Cost and environmental hazards prohibit wider applications of large neutron detectors based on liquid organic scintillators.

The group has developed a new type of solid scintillator neutron detector (N*) that circumvents a number of the problems faced by other detector designs. As described in a report further below (Sect. III), following extensive simulation calculations a prototype N* module has been built and tested with encouraging results. The module consists of a stack of plastic scintillation slabs alternating with thin Gd-loaded transmuter films. The plastic scintillator functions both as a neutron moderator, slowing neutrons down to thermal velocities, and as scintillator sensing thermal-neutron capture γ-rays produced in the transmuter films. This detector has a high efficiency in a broad dynamic range of neutron energies (from thermal up to several MeV, depending on volume), an essentially zero energy threshold for neutrons and multi-hit capability. Design and development of the prototype detector module are described in the report which will be submitted shortly to a technical journal. Simulation calculations have relied on a newly extended simulation code (DENIS(E)).

Several of our R&D projects have important educational and student training components, as discussed in two contributions to this report. Absent a radio-chemical curriculum at the UR, we have started a collaboration with the University of Rochester Laboratory for Laser Energetics (LLE), in which we have set up and run several experiments addressing the transport of tritium compounds. The experiments are of a scope and complexity found in the Nuclear Chemistry Summer Schools organized yearly by the ACS on two sites in the U.S. In addition, the experiments have real-life importance since tritium and its compounds are radioactive components used or produced in nuclear energy technology both using regular nuclear fission or inertial DT fusion. The first such experiments tested different methods to decontaminate tritium exposed metallic surfaces by
thermal desorption. A second method employed plasma induced sputtering. In both cases it was found that the surface tritium contamination was removed efficiently and quickly from metallic samples, while longer-term tritium transport occurs through diffusion from the interior of the solid lattices to the surface. These radio-chemical experiments have been of a scope and character that attracts students of different stages of their training, from high school to graduate programs. Several summer interns and beginning graduate students have worked on, and have benefited from these experiments.

Student training in the Rochester nuclear chemistry group has also profited from the development of experiments for the new UR nuclear education laboratory course ANSEL. The development uses mostly new equipment funded (by NRC) separately from this grant but utilizes some obsolete group equipment and other resources. Perhaps most importantly, the ANSEL buildup provides training opportunities for graduate students in basic technical skills related to nuclear science experimentation. It exposes them to a number of modern experimental methods used in applied fields of imaging and nuclear forensics but also in scattering and detection of charged particles and neutrons. A newly acquired pulsed-beam neutron generator has already helped in the debugging of our new neutron N* detector. The experiments required information of the ambient and induced background radiation in the nuclear chemistry laboratory, which is now available through a special report.

During the Academic Years 2007/08 and 2008/2009, the Principal Investigator has spent approximately 20% of his time to research and student training supported by this grant, increasing to 80% during summers. The Senior Scientist (J.T.) and the Research Associates (W.Y., H.S.) spent 100% of their efforts on these research projects. During the past two academic years the resident graduate PhD students (I.P., M.Q.) have each worked 100% on grant research, several graduate MS students (Y.T., E.P.) have performed grant related research only part time, each approximately one semester equivalent. Incoming graduate PhD students (E.H. in 2008, M.S. in 2009) work several summer months on grant research, including preparatory training. One student (M.S.) was supported by an LLE fellowship. During the academic year, the junior students take
courses and have, at least partial, teaching assistantships. During these periods, such students work part time on grant research.

Within the reporting period, Yun-Tse Tsai, Elizabeth Pollock, and Eric Henry have earned their Master of Science degrees in physics and chemistry, respectively. Mike Quinlan has received a travel grant from the Italian INFN for a one-month research stay at Catania. He has also received a prestigious Weissberger University fellowship. Eric Henry was honored with a Departmental Sherman Clarke Fellowship. Congratulations are due to all of them.

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 and training program by the United States Department of Energy. We also acknowledge indirect (equipment and training) support by the U.S. Nuclear Regulatory Committee.

In addition, we are grateful to the Italian research community for giving us support and access to their LNS laboratory and accelerator facility.

W. Udo Schröder
Rochester, September 2009
III. Research Program
Common Signatures of Statistical Coulomb Fragmentation of Highly Excited Nuclei and Phase Transitions in Confined Microcanonical Systems

J. Töke and W. U. Schröder

Departments of Chemistry and Physics

University of Rochester, Rochester, New York 14627

ABSTRACT

Characteristic signatures of statistical Coulomb fragmentation of highly excited nuclear systems are analyzed. It is found that in many important aspects, they coincide with perceived signatures of phase transitions in confined hypothetical pseudo-microcanonical systems and, therefore, may give rise to an incorrect interpretation of certain experimental observations in terms of phase transitions occurring in nuclear matter. It is demonstrated that domains of negative heat capacity predicted by certain classes of pseudo-microcanonical model calculations for the immediate vicinity of phase transitions are artifacts of an un-physical truncation of the model phase space and that such domains disappear already with a very rudimentary enhancement of this phase space. Appearance of bimodality and of signatures of critical phenomena in Coulomb fragmentation is discussed.
I. INTRODUCTION

For over a quarter of century now, the observed process of disintegration of highly excited nuclear systems into multiple intermediate-mass fragments (IMF) has provided a strong driving force for research in the field of intermediate-energy heavy-ion reactions. This process, commonly called multifragmentation, has inspired both, theoretical speculations regarding its mechanism and experimental effort to map out its characteristics. Experimental observations point often to dynamical IMF production mechanisms, but they also reveal “robust” patterns commonly attributed to statistical production mechanisms [1]. While there appears to be a general consensus regarding the presence of a statistical component in the observed IMF yield, the standard equilibrium-statistical decay codes [2, 3] with their gentle Boltzmann-like scaling of yields are not able to predict meaningful yields of intermediate-mass fragments (IMFs) heavier than lithium. The difficulty arises here from the fact that experimental yields show a significant departure from Boltzmann-like scaling with a fixed transition-state energy. Rather, single and multiple IMF production are seen to set in rapidly with excitation energy, in a fashion reminiscent of phase transitions, raising tempting prospects for experimental studies of phase transitions in microscopically small (nuclear) systems.

As a result of the difficulties encountered by standard equilibrium-statistical models to explain abundant statistical IMF production and to effectively parameterize some of the salient trends in the observed IMF yields, a number of models [4–9] have been proposed which supplement the traditional framework of statistical theory by incorporating either explicitly or implicitly ancillary (ad hoc) assumptions. The nature of these ad hoc assumptions is such that there appears to be no obvious way to justify them in the framework of known physical theory and that their only “validation” appears to be a satisfactory fit of resulting “predictions” to select experimental observations. Furthermore, these models describe mostly stationary equilibrium states, typically of spatially confined systems, without addressing the paramount issue of stability with respect to actual decay modes. The relevance of such model calculations for nuclear matter is sometimes asserted by invoking the principle of universality, which is merely a heuristic notion. Similarly, although IMF yields predicted in these models [5, 6, 8] appear to exhibit pat-
terns reminiscent of phase transitions, conclusions that nuclear multifragmentation is a manifestation of a nuclear liquid-gas phase transition are in fact unfounded hypotheticals. Nevertheless, in spite of an obvious lack of a sound theoretical foundation, over the years a paradigm or dogma has appeared in linking nuclear multifragmentation with nuclear phase transitions and “collateral” phenomena such as negative heat capacity, bi-modality, or criticality.

To illustrate in more detail the point regarding the ad hoc nature of “mainstream” multifragmentation models, one notes that in the Copenhagen [5] and Berlin [6] multifragmentation codes, treated here as benchmarks, the model “nuclear” matter is tacitly taken to be infinitely incompressible in the liquid phase and infinitely compressible in the gaseous phase. Such a peculiar model equation of state (EOS) is not stated explicitly, but can be inferred from the fact that these models do not allow matter to undergo thermal expansion and set interaction energy to zero for the gas phase, regardless of matter density (including the ground-state density of nuclear matter). The equilibrium state assumed in these models is that of an ensemble of spherical objects which are continually scattering off each other and off the walls of a hypothetical spatial containment, the so-called “freezeout” volume, while at the same time evaporating, re-absorbing, and exchanging nucleons and excitation energy among themselves. An important, but little noticed fact is that, contrary to common beliefs, these models do not predict statistical breakup of the system at all in the sense the term “breakup” appears to suggest, i.e., as a fast single act. What they actually do predict is only that highly excited matter with the particular model EOS would time-asymptotically end up fragmented. It may do so plausibly by evaporating nucleons into the free space of the “freezeout” volume, where these nucleons subsequently synthesize into intermediate-mass clusters or fragments. Obviously, the nature of these models is such that they cannot distinguish between a physically single-act breakup and the more lengthy process of evaporation and nucleo-synthesis.

The present work is part of a continued effort [10–18] to model the behavior of finite nuclear systems with diffuse surface domains within the framework of liquid drop and Fermi gas models and to construct a thermodynamical framework for understanding nuclear multifragmentation as a natural, fission-like decay mode of highly excited com-
pound nuclei, all while avoiding *ad hoc* assumptions of the kind discussed above. It is worth noting in this context that the standard statistical decay models [2, 3] were designed for compound nuclei at low to moderate excitations, where particle evaporation and fission rates obey quite well Boltzmann scaling. At higher excitation energies, of the order of $E^*/A > 2–3$ MeV, physical phenomena of thermal expansion, reduction in surface tension, and growth of shape fluctuations can become important and alter nuclear decay patterns qualitatively.

Recently [12, 15], it was shown that, when even a very rudimentary allowance is made for the surface entropy and thermal expansion of nuclear matter, the basic statistical scenario of asymmetric fission is quite sufficient to explain the large observed IMF yields and also to explain multifragmentation as a form of generalized fission associated with multifragment saddle shapes. In this scenario, an excited nucleus approaches a state of local thermodynamical equilibrium that is conceptually identical to that of a compound nucleus. The term “local” refers here to a finite volume in the phase space delimited by the hypersurface of transition states, i.e. states connecting to the open decay channels. In contrast to the “classical” low-energy compound nucleus, however, the system is now allowed to maximize its entropy by expanding thermally to corresponding equilibrium density and by exercising substantial shape fluctuations.

Obviously, equilibration of shape degrees of freedom is part of the overall equilibration process, where the system is “racing” for its survival against particle evaporation and other decay modes and where its “life expectancy” diminishes with increasing excitation energy as the decay time scales shorten. How far the system is able to advance on its path toward equilibrium depends critically on the latter time scales. The general assumption here, as in “classical” compound nucleus model, is that the degree of equilibration actually reached is sufficient to justify application of equilibrium statistical thermodynamics as a meaningful approximation. It is also worth noting in this context that the equilibration process sets in already as first amounts of excitation energy are supplied to the system by the collision dynamics and not, as some models (e.g. Ref. [4]) assume, after all of the excitation energy had been supplied. “Fortunately”, in the (doomed) race for survival mentioned above, the system is assisted by a peculiar feedback such that thermal expansion and excitation of shape degrees of freedom cause
nuclear temperature to decrease (expansion cooling), thus extending particle evaporation time-scales [13]. It is worth noting that such a feedback mechanism is an expected manifestation of Le Chatelier’s Principle, where the system responds to the supplied excitation energy in a way (expansion plus fluctuations) that opposes the direct result of this supply – increase in temperature. As the system, on its way toward equilibrium, reaches randomly a particular binary or more complex saddle configuration, it is driven toward scission by Coulomb and/or centrifugal forces resulting in observed fragment yields.

For the purpose of the following discussion, the decay scenario described above is named Coulomb fragmentation, to reflect the crucial role of Coulomb forces in the ultimate dynamical breakup of the system into two or more individual fragments. In fact, this is the same scenario that is considered, e.g., by the statistical decay code Gemini [3]. This code succeeds in describing quasi-symmetric but not highly asymmetric binary fission, where the latter failure is attributable to an inadequate accounting, of thermal expansion of nuclei and of the role which the surface entropy plays in the process.

As was shown recently, [15] asymmetric binary Coulomb fragmentation is described by equations that are largely equivalent to the parameterization used with the Nuclear Fisher Droplet Model [19] to fit a large volume of experimental data. In addition, multiple Coulomb fragmentation is consistent with the numerical procedures used in the statistical multifragmentation codes SMM [5] and MMMC [6], providing an explanation for why these codes appear to agree with selected sets of experimental observations.

The present study shows for the first time that statistical Coulomb fragmentation of finite nuclei shares some prominent signatures with first- and second-order phase-transitions in spatially confined pseudo-microcanonical systems. Most notably, such a process sets in rather suddenly with excitation energy and in a “non-Boltzmannian” fashion and may also exhibit apparent negative heat capacity and bimodality. Furthermore, at sufficiently high excitation energies, where the surface tension vanishes, statistical Coulomb fragmentation may also be expected to exhibit signatures of criticality, where the outcome is decided by combinatorial factors only. In other words, a rapid onset of single and multiple IMF production, the bimodality of select distributions, an apparent negative heat capacity and the “rule” of power law, are all consistent with and occur
naturally in Coulomb fragmentation of hot nuclei. These findings challenge the concept of multifragmentation as a manifestation of a liquid-gas phase transitions in finite nuclear systems and favor the simpler, more plausible Coulomb fragmentation scenario.

The following Section II briefly reintroduces the schematic model of Ref. [11] and demonstrates that the two phenomena, second-order phase transitions in confined pseudo-microcanonical systems and statistical Coulomb fragmentation, are described by a common mathematical formalism. Both are manifestations of the crossing of relevant partition functions on the excitation energy scale.

In Section III, the significance of experimental trends in fragment productions is discussed in terms of Coulomb fragmentation. Here, the possibilities for observing signatures of first and second order phase transitions, as well as criticality are assessed.

Section IV offers an extended discussion and a summary. This section stresses the importance of the diffuse surface domain for nuclear stability and provides a tentative road-map for a further exploration of Coulomb fragmentation.

II. MODELING OF PHASE TRANSITIONS IN CONFINED PSEUDO-MICROCANONICAL SYSTEMS AND OF STATISTICAL COULOMB FRAGMENTATION

For the purpose of this study, a schematic model [11] is considered that emulates essentials of the (benchmark) pseudo-microcanonical models SMM [5] and MMMC [6], as far as phase transitions are concerned, but at the same time allows one to model Coulomb fragmentation [15]. The model considers a finite amount of iso-neutral Fermi matter that is allowed to assume two spatial configurations, a spherical mono-nucleus and a symmetric di-nuclear configuration of two equal touching spheres. These two configurations are taken to represent either two distinct phases of a confined nuclear system or two spatial configurations of an open system. In the latter context, the di-nuclear configuration represents a transition state (saddle) connecting to an open binary fragmentation channel. Importantly, this model accounts for the diffuse nuclear surface domain, including in the calculations both surface energy and entropy [12]. As demonstrated in Ref. [12], the diffuse surface domain has profound, qualitative effects
on the evolution of the system with increasing excitation energy.

In the qualification of SMM [5], MMMC [6] and other models, the prefix “pseudo” is used to stress the important fact that only a truncated phase space is numerically manageable in calculations for systems at the high excitation energies of interest here. Quite naturally, the same prefix applies to the model used in the present study. As shown further below, an incomplete accounting of phase space may have non-trivial, qualitative consequences specifically in domains of phase transitions. In fact, the omission of certain parts of phase space may well be responsible for apparent but false signatures of such transitions, such as the negative heat capacity and some forms of bimodality reported in the literature [1].

In what follows, a “phase” is defined as a macroscopically distinct state of the system. Accordingly, a phase transition is defined here as an event in which the most likely phase of the system changes, as the value of the control parameter changes. Of course, small systems fluctuate strongly between different phases, where the relative dwelling times are given by phase partition functions. Since the fundamental difference between the conventional first- and second-order phase transitions is that the former involve transfer of latent heat while the latter do not, the phase transitions considered here are “nominally” of second order.

The above definitions of phases and phase transitions are fully consistent with conventional thermodynamics, a fact that is reiterated here to enhance the clarity of a chain of arguments made further below. While for the sake of specificity, in what follows, reference is mostly made to phases and phase transitions, one should keep in mind that the same applies always to macroscopic configurations of an open system, as well.

Formally, the probability \( w_i \) of finding the system in a particular phase is given by the associated partition function,

\[
    w_i = \frac{Z_i}{\sum_k Z_k}.
\]

For a microcanonical statistical ensemble, which is the ensemble most suitable for the description of isolated systems, partition functions for individual phases (phase partition functions) are expressed via the associated entropies \( S_i \) (phase entropies). Therefore,
\[ w_i = \frac{e^{S_i}}{\sum_k e^{S_k}}. \]  

(2)

With the above conventional definition of material phases, a phase transition occurs when the partition functions for different phases intersect, as the controlling parameter (such as the total energy or the temperature) is varied. This point is illustrated with the schematic pseudo-microcanonical model of Ref. [11], which permits just two distinct macroscopic states of an excited nuclear system, the mononuclear and the symmetric di-nuclear configuration. In this case the pseudo-microcanonical weight functions \( w_i(E^*) \) for the two configurations \( i \) are functions of the total excitation energy \( E^* \) of the system. They are related to the phase entropies \( S_m \) and \( S_d \) associated with mono- and di-nuclear configurations, respectively. These are the formal entropies calculated for the system in either a pure mono-nuclear (subscript \( m \)) or a pure di-nuclear (subscript \( d \) ) configuration,

\[ w_{m/d}(N, E, V) = e^{S_{m/d}(N,E,V)}. \]  

(3)

Here, \( N \) is the number of particles in the system and \( V \) is the system volume, here taken to be constant. Because of the above simple microcanonical relationship between a weight function and the corresponding configurational entropy, a (second-order) phase transition will occur at a system energy of \( E = E_{P.T.} \), where the entropy functions for the two configurations or phases cross, i.e., where \( S_m(N, E_{P.T.}, V) = S_d(N, E_{P.T.}, V) \).

Note that for canonical ensembles (constant particle number \( N \), constant temperature \( T \) and fixed volume \( V \)), the corresponding weight functions can be expressed in terms of the Helmholtz free energies \( A_i \),

\[ w_{m/d}(N, T, V) = e^{-A_{m/d}(N,T,V)/T}. \]  

(4)

Accordingly, for an isothermal-isobaric ensemble (constant values of \( N \), temperature \( T \) and pressure \( p \)) the weight functions are properly expressed in terms of the Gibbs free energies \( G_i \),

\[ w_{m/d}(N, T, p) = e^{-G_{m/d}(N,T,p)/T}. \]  

(5)
Obviously, for all three kinds of ensembles considered above, any crossing of the configurational weight functions for any two macroscopically distinct configurations (phases) occurs at that value of the control parameter \( E \) for microcanonical ensembles, otherwise \( T \) at which the corresponding pairs of thermodynamic state functions \( (S, A, \text{or} G) \) intersect. In the thermodynamical limit, such crossing of the relevant special thermodynamic state functions results in a "logarithmic singularity" in the relevant partition function such that the respective first derivatives of logarithms of these partition functions are discontinuous. This singularity is reflected in corresponding singularities of the various thermodynamical functions.

Using the Fermi gas model to calculate the level densities of the constituent spherical fragments, the conditional entropies for the pseudo-microcanonical mono- and symmetric di-nuclear configurations considered here can be written as

\[
S_m(A, E^*) = 2\sqrt{a_mE^*}
\]

and

\[
S_d = 2\sqrt{a_d[E^* - (E_{pot}^d - E_{pot}^m)]}.
\]

In Eqs. 6 and 7, \( a_m \) and \( a_d \) are the level density parameters (\textit{little}-a) for mono- and di-nuclear configurations, respectively, and \( E_{pot}^m \) and \( E_{pot}^d \) are the potential energies of these configurations. The level density parameters for a realistic nuclear matter distribution with diffuse surface domain can be calculated using the Thomas-Fermi approximation. They can be expressed approximately [10] in terms of volume and surface contributions,

\[
a_m = \frac{A}{14}(1 + 4A^{-1/3})MeV^{-1}
\]

and

\[
a_d = 2\frac{A}{28}[1 + (\frac{A}{2})^{-1/3}]MeV^{-1}.
\]

The potential energies \( E_d \) and \( E_m \) for di- and mono-nuclear configurations can be calculated from the liquid drop model [20] such that their difference is equal to the difference in the sums of surface and Coulomb energies for the two configurations.
Here \( c_{\text{Surf}} \) is the surface energy coefficient, and \( F_d^d = 2^{1/3} \) is the ratio of the surface area of the symmetric di-nucleus to that of a single sphere of the same volume. The Coulomb term is left out for the purpose of this study.

Accounting for a surface specific contribution to the level density parameter is essential for the present study. In particular, it is crucial for developing a quantitative understanding of Coulomb fragmentation, both binary and multiple. This surface level density term approximately accounts for the excess entropy per nucleon (surface entropy) contributed by the dilute matter in the diffuse surface domain, relative to that of the denser bulk matter. At elevated excitation energies this extra surface entropy becomes large enough to significantly enhance the chances for a system to populate configurations with large surface areas such as represented by the di-nuclear phase of a confined system and the saddle configuration of an open system. The surface entropy is also directly responsible for an intersecting of the weight functions for different configurations at characteristic excitations [11] signaling a phase transition as defined above. This feature is illustrated in figures presented further below.

In Figs. 1 - 2 results are shown of model calculations for a two-configuration system allowing mono-and symmetric di-nuclear configurations within the schematic formalism [11] discussed above. For the sake of an easier demonstration of the effects of the size of the system the Coulomb interaction was left out in these particular calculations.

The top panel of Figure 1 illustrates the intersecting of the entropy functions for the two \((\text{mono} \text{ and } \text{di})\) model phases at a “cross-over” excitation energy of \( E/A \approx 7\text{MeV} \), which also signifies the crossing of the corresponding weight functions for the two configurations. The calculations are for a system of \( A = 300 \) nucleons.

The bottom panel of Figure 1 illustrates the way in which the relative population probabilities of the two phases of interest evolve with increasing excitation energy. As seen in this panel, at low excitations, where \( S_m > S_d \), the system dwells dominantly in the mono-nuclear configuration or phase (solid line). As the excitation energy increases and approaches the crossing point, the system fluctuates more frequently away from the mono-nuclear configuration into the di-nuclear domain (dashed line). In the vicinity
FIG. 1: Functional dependence of entropies (top panel) and relative population probabilities (bottom panel) for mono- and di-nuclear phases, solid and dashed lines, respectively) on excitation energy per nucleon exhibiting a crossing at approx. $E/A = 7$ MeV. The dotted line in bottom panel illustrates Boltzmann-like behavior for a hypothetical di-nuclear phase with 4 MeV of potential energy and with no gain in surface entropy.

of the “cross-over” energy, the two phases coexist, but not in the sense of conventional first-order phase transitions for macroscopic systems, where, e.g., finite amounts of liquid and gas are simultaneously present in a given volume. Rather, the system continually jumps from one phase to the other (at no cost in latent heat) but at any instant in time is either in one or in the other pure macroscopic state or phase. At higher excitation energies, beyond the cross-over energy, the system is found dominantly in the di-nuclear state or phase. Notable in Fig. 1, bottom panel is the rapidity with which the di-nuclear
phase sets in, as excitation energy is increased. It should be kept in mind that the same “phase-transition-like” scaling with $E/A$ applies to the population probability of the fragmentation saddle configuration as compared, e.g., with the population of (compact) particle emission transition states. Such a “phase-transition-like” scaling is in stark contrast with the Boltzmann-like scaling illustrated in the bottom panel of Fig. 1 by a dotted line. The latter is calculated for a hypothetical configuration with just 4 MeV of potential energy and with no gain in surface entropy with respect to the mononuclear configuration. Note that the population probability of this hypothetical configuration approaches asymptotically the probability of the mono-nuclear configuration, but never “overtakes” it.

The thermodynamics of phase transitions in the present schematic model can be further explored by studying the evolution of the system entropy (as opposed to phase entropies) with increasing excitation energy. This system entropy is given by the logarithm of the microcanonical partition sum

$$S_{[m,d]}(E) = \ln[e^{S_m(E)} + e^{S_d(E)}], \quad (11)$$

where $S_m(E)$ and $S_d(E)$ are phase entropies for the mono- and di-nuclear configurations. For a better demonstration of important mathematical properties of the system entropy $S_{[m,d]}$, a reduced entropy is defined such that a linear function is subtracted from the former which makes the latter zero at the boundaries of the energy region of interest. Importantly, the subtraction of a linear function preserves the second derivative and thus does not affect properties of interest here, such as convexity or concavity.

A reduced entropy function per nucleon (multiplied by 1000) for the ensemble considered here and the corresponding caloric curves are illustrated in Fig. 2 for three different sizes of the system, infinite ($TL: A > \infty$), large ($A = 300$) and intermediate ($A = 200$). The calculations were performed according to Eqs. 6, 7, and 11 for the case of $A = 300$. To preserve the location of the crossing point of the entropy functions and to isolate the effect of the overall system size from that of the relative size of the surface domain, the entropy for the case of $A = 200$ was calculated using Eq. 11, with $S_m$ and $S_d$ obtained by renormalizing by a factor of 200/300 the entropies calculated for the system of $A = 300$. In the same spirit, the data for the thermodynamic limit were obtained
FIG. 2: Reduced entropy (top panel) and system temperature (bottom panel) vs. excitation energy per nucleon for the two-phase system of mono- and di-nuclear configurations and three sizes of the system - thermodynamic limit (TL), “large” with A=300, and “intermediate” with A=200. (See text)

from calculations for the A = 300 system, by taking the mononuclear entropy function for excitation energies below the cross-over point and the di-nuclear entropy function above this point. Obviously, in the true thermodynamic limit, not only do fluctuations vanish but also any surface effects must vanish.

As seen in the top panel of Fig. 2, in the thermodynamic limit the reduced entropy (and hence the model entropy) features a singularity at the crossing of entropy functions around E/A=7 MeV but remains always a concave function of the energy. Correspondingly, caloric equation of state features a singularity in the form of a jump (bottom
panel). Fluctuations present in finite systems make the entropy differentiable at all energies, although larger systems with smaller fluctuations exhibit a qualitatively different behavior. As seen also in the top panel of Fig. 2, in the “large” system ($A = 300$), a convex domain or “convex intruder” persists as a telltale remnant of the kink present in the case of the thermodynamic limit. This convex intruder maps then onto the caloric equation of state as a domain of negative heat capacity (bottom panel). The more “robust” fluctuations present in the “intermediate-size” system ($A = 200$) further “heal” the entropy function, which is now strictly concave. Accordingly, the heat capacity is always positive (bottom panel. The thermodynamic temperatures plotted in the bottom panel of Fig. 2 are calculated according to the standard microcanonical expression for the average temperature

$$T^{-1} = \beta = \frac{dS}{dE} \quad (12)$$

A. Origins of an Apparent Negative Heat Capacity

In principle, irregularities or non-monotonicities in the caloric curve $T(E)$ appear possible in the phase transition domain of physical systems. However, to prove in numerical modeling the presence of a negative heat capacity in such a domain is exceedingly difficult, if not impossible. This is so because it is obviously technically impossible to include in the model calculations explicitly all energetically allowed microstates of a physical system. Neither is it feasible to include all possible macroscopic configurations. Therefore, it is impossible to deduce the true entropy function and the corresponding true microcanonical temperature. Actual numerical simulations can only address finite subspaces of an untractably large true microcanonical phase space. Therefore, in such simulation calculations only apparent or pseudo thermodynamic potentials and functions are evaluated and not the true functions. It is therefore impossible to tell whether or not any particular fine (as opposed to gross or average) trend in an apparent thermodynamical quantity deduced for a numerical model system reflects its true thermodynamical counterpart in a physical system. Specifically, it is impossible to predict fine trends in thermodynamical functions for physical systems with phase space truncated numerical
calculations on which models such as SMM [5], MMMC [6], and lattice gas models [8], are based. Concerning reports of apparent negative heat capacities for physical systems, for example, it is demonstrated further below that the exclusion of certain classes of microstates from experimental observation may result in a corresponding artifact.

In evaluating such apparent caloric phenomena, one notes first that it is physically impossible for a nuclear system to populate just two distinctly different macroscopic spatial configurations, such as the ones \( m \) and \( d \) used in the schematic model introduced previously, without also populating the entire continuum of intermediate macrostates, those “connecting” the two limiting spatial distributions. In fact, physical systems must be able to follow continuous pathways connecting any number of macroscopic spatial configurations. In the present context, it is impossible for a spherical mono-nucleus to jump to a symmetric di-nuclear configuration without passing through a continuous sequence of intermediate states of various intermediate deformations. Similar considerations apply to the configuration spaces considered by SMM [5], MMMC [6], and lattice gas [8] models. Importantly, the intermediate macroscopic configurations have a noticeable influence on the apparent trends of thermodynamic functions exactly in the domain of phase transitions, i.e., where the weight functions for the phases (configurations) of interest cross. This is so, because the weight functions of the intermediate configurations must intersect the weight functions for the phases of interest in the immediate vicinity of their crossing. Therefore, the intermediate configurations make a noticeable contribution to the overall partition function selectively only in this domain. Consequently, exclusion of any of these intermediate configurations depletes the model entropy locally (on the energy scale) and may give rise to a false convex intruder in the entropy function \( S(E) \). Effects of such false convex intruder propagate to other thermodynamic functions and representations of thermodynamic observables. In particular, a false intruder results in a caloric curve featuring a domain of apparent negative heat capacities. This rather trivial mechanism of generating \textit{unphysical} negative heat capacity in truncated-space microcanonical calculations is illustrated in more detail in Fig. 3.

Figure 3 illustrates effects of an exclusion of intermediate macrostates by considering the effect associated with just one extra deformed configuration intermediate between the two limits of spherical mono- and deformed di-nuclear ("base") configurations. The
FIG. 3: The “healing” effect of an inter-phase configuration on the apparent caloric curve for the two-phase system of mono- and di-nuclear configurations for a $A=200$ system.

The assumed inter-phase configuration corresponds to a deformation parameter of $F_2 = 1.13$, where $F_2$ is the ratio of the surface area of a configuration to the surface area of a sphere of equal volume. This parameter value is half-way between the respective values for spherical mono-nuclear ($F_2 = 1$) and di-nuclear ($F_2 = 2^{1/3}$) configurations. As seen already in Fig. 2, the apparent entropy $S_{[m,d]}(E)$ for a system including only these two base configurations (m and d) exhibits a convex intruder in the vicinity of the crossing point of the phase entropy functions $S_m(E)$ and $S_d(E)$. The caloric curve $T_{[m,d]}(E)$ deduced for a hypothetical system with such a drastically truncated phase space features a negative heat capacity in the vicinity of this crossing (cf. Fig. 2). As seen in Fig. 3, restoring the previously neglected additional intermediate inter-phase (i) configuration with a deformation parameter of $F_i^2 = 0.5(F_m^2 + F_d^2)$ changes the apparent caloric curve $T_{[m,d,i]}(E)$ qualitatively, causing the domain of apparent negative heat capacity to disappear. The mechanism of elimination of this domain can be understood in more detail from Fig. 4.

The top panel of Fig. 4 illustrates the sequential crossing of entropy functions for mono-nuclear (solid line), intermediate (dashes), and dinuclear (dotted line) phases in
FIG. 4: Crossing of relative phase entropy functions (top panel) and phase weight functions (middle panel) for three macroscopic configurations, mono- and di-nuclear (subscripts $m$ and $d$) and intermediate ($i$) as functions of energy. The bottom panel illustrates the extra system entropy resulting from the presence of the intermediate phase $i$.

A three-phase system. The entropy functions are plotted relative to that for the mono-nuclear configuration to enhance the view. At low excitations, mono-nuclear configuration dominates. As the excitation energy increases, first the intermediate configuration “catches up” with the mono-nuclear phase, becoming the dominant phase around $E/A \approx 7$ MeV. Subsequently, the di-nuclear phase “catches up” with the intermediate one, becoming dominant around $E/A \approx 7.3$ MeV. As seen in the middle panel of
Fig. 4, the “inter-phase” configuration (i) plays a noticeable role selectively only in the vicinity of the cross-over energy of the weight functions \( w_m \) and \( w_d \) for the two phases considered. Accordingly, as seen in the bottom panel of Fig. 4, it contributes to the overall apparent entropy only in the energy region of the phase transition, but not much beyond it. It is this additional entropy component, which is not accounted for in the truncated two-phase phase space, that restores the overall concavity of the entropy as a function of energy. This ”healing” is achieved by a slight reduction in the system temperature caused by the ascending slope and a slight increase caused by the descending slope of the intermediate entropy function, consistent with the definition of temperature as \( T^{-1} = \partial S/\partial E \).

The present schematic calculation demonstrates that, in the vicinity of an anticipated phase transition (crossing of weight functions), apparent thermodynamic quantities behave qualitatively differently when certain classes of valid macrostates are excluded from the numerical calculations or from observation. As it is practically impossible to guarantee that all relevant states are included in any realistic model description, model calculations do not provide a sound foundation for conclusions regarding fine trends in thermodynamical functions. This is especially true in the vicinity of phase transitions.

**B. Apparent Bimodality**

Recently, suggestions have been made [21, 22] that the bimodality of certain distributions or of thermodynamic functions represents a robust signature of a nuclear liquid-gas phase transitions. In this context, one is immediately reminded of examples of bimodal distributions that are associated with other phenomena. For example, the statistical competition of Coulomb fragmentation or fission with particle evaporation results naturally in bimodal distributions of various observables. In the schematic model discussed previously, post-evaporation fragment mass distribution are bimodal for excitation energies close to the cross-over energy, where the entropy functions for mononuclear configuration \( (S_m(E^*)) \) and di-nuclear saddle configuration \( (S_d(E^*)) \) intersect. In this case, one peak in the mass distribution represents the massive evaporation residues of the primary mononuclear product, while the second peak just below one-half
of the total system mass represents the evaporation residues of the two, nearly symmetric fragments produced in the primary Coulomb fragmentation process. One may also expect a bimodal temperature distribution, with a lower average temperature for the fragmentation channel. This kind of bimodality is in fact well known from fission studies. Evidently, bimodality is not an exclusive signature of phase transitions and its experimental observation can therefore not be taken as proof that such a transition actually occurs in excited nuclei.

C. First Order Phase Transitions in Small Systems with Truncated Phase Space

Quite generally, first-order phase transitions in small confined microcanonical systems can be viewed as a rapid succession along the energy axis of crossings of weight or entropy functions for a series of macroscopic configurations or phases, e.g., those associated with a successively increasing number of gas particles. In the aggregate, a succession of such second order phase transitions has the appearance of a conventional first order phase transition. In this case, the latent heat conventionally associated with first-order transitions is represented here by the difference in excitation energies between adjacent crossing points of phase entropy functions. As discussed further above, at crossings, phase transitions occur without infusion of energy from an external source, i.e., with zero latent heat. To illustrate the above point, a sequence of crossings of entropy functions for 20 configurations, including the mono-nuclear, di-nuclear, and 18 intermediate with uniformly spaced deformation (surface area) parameters $F_2$ is illustrated in Fig. 5.

As discussed further below, the sequence seen in Fig. 5 may be viewed as representing a first-order phase transition of matter from the dense bulk to the less dense surface domain.

In actual (small) nuclear systems, the energy intervals between the consecutive crossings (the apparent latent heat) may fluctuate significantly, for example, due to pairing or isospin effects. A scenario of successive second order phase transitions accommodates such variations naturally and offers therefore a more direct access to the underlying transitional phenomena than one based on the corresponding picture of conventional
FIG. 5: Crossing of relative phase entropy functions for 20 macroconfigurations (phases) with surface area parameters $F_2$ increasing from 1 (mono) to $2^{1/3}$ (di) in 20 equal steps.

first order transition.

It is worth noting that in the SMM and MMMC models an otherwise dominant population of the gas phase is suppressed by an ad hoc resetting of the gas interaction energy to zero at any density. At the same time, these calculations artificially enhance the relative weights of multi-fragment configurations by an ad hoc setting of the nuclear incompressibility modulus to infinity. As a result, in these models, the crossings of the overestimated weight functions for many multi-fragment configurations occur in the same excitation energy domain where crossings would occur between entropy functions for different liquid–gas compositions, obscuring possibly a true first-order liquid–gas transition. It is of interest to have SMM or MMMC type calculations performed for “numerical” matter with a more realistic effective EOS to see whether or not such more realistic model matter would still be predicted to “clusterize” (via evaporation and nucleo-synthesis) into various sets of IMFs plus relatively few free nucleons floating in
a “freeze-out” volume. Alternatively and more likely, such more realistic matter would end up as a large, thermally expanded liquid residue surrounded by a small amount of gas of interacting nucleons. However, such interest is purely academic, as it is not possible to confine a highly excited nuclear system to a fixed spatial volume, as would be required to confirm or falsify improved model predictions.

III. EXPERIMENTAL PHASE-TRANSITION-LIKE SIGNATURES IN COULOMB FRAGMENTATION

As discussed briefly further above, Coulomb fragmentation is a process similar to binary fission but generalized to arbitrary saddle shapes, including multifragment shapes [15]. The process can be described approximately in terms of microcanonical thermodynamics of an excited nuclear system, which surface tension confines transiently to a finite domain of 6A-dimensional \((6A)\) phase space or the system phase space. This domain is delimited by a hypersurface in phase space that includes all possible transition states, i.e., those defined by particle evaporation barriers or thresholds, as well as the complete set of (multi-)fragmentation saddle configurations. To achieve maximum entropy, microcanonical equilibrium thermodynamics requires the system to explore all energetically allowed microstates with equal \(a\ priori\) probability. In the course of such an “exploration” and before it is completed, the system reaches eventually a microstate on the hypersurface of transition states that is connected to an open decay channel. If this happens to be a particle emission channel, a particle escapes while the residual system evolves toward a new equilibrium characterized by accordingly reduced energy and particle numbers. Depending on the degree of the equilibration reached, the just emitted particle is viewed as an evaporated or as a preequilibrium ejectile. On the other hand, if the transition state represents a fragmentation saddle configuration, the system is driven toward scission, all the while continuing its exploration of the accessible phase space. The basic macroscopic phenomena associated with equilibration are thermal expansion and thermal shape fluctuations. The latter are responsible for Coulomb fragmentation, i.e., for bringing the system occasionally to a particular saddle configuration from which it is dynamically driven apart by Coulomb forces. The above
picture is essentially identical to a compound nuclear fission scenario and to the IMF production scenario adopted in statistical decay models. A novel observation made only recently [12] is that extra entropy associated with the diffuse surface domain plays a critical role in facilitating large and complex shape fluctuations.

A. Signatures of Second-order Phase Transitions

The probability for an excited nuclear system to arrive at a particular transition-state configuration is given by a respective weight function identical to the one discussed further above in the context of phase transitions of confined systems. The weight functions for various transition-state configurations, given by Eq. 3, intersect at different, characteristic excitation energies. Since compact transition-states configurations are characterized by low potential energies, their entropy functions “take off” already at low excitation energies which, on the other hand, are insufficient for supporting higher potential energies associated with more deformed transition-state configurations. However, when with increasing excitation energy the entropy functions for deformed configurations eventually do “take off”, they grow at a rate superior to that for compact configurations. As a result, the entropy (weight) functions for different fragmentation channels cross and leave signatures reminiscent of second-order phase transitions.

It is worth noting that the observation of Coulomb fragmentation phenomena is subject to an inherent experimental filter which allows to pass only the saddle configurations, but not all those configurations that did not “quite make it” to the saddle, i.e., configurations intermediate between the deformed saddle and the spherical reference configurations. However, to extract the true, and not just some apparent, values of the interesting thermodynamical parameters from experimental Coulomb fragmentation data from comparisons with theoretical simulations, such simulations need to include also the experimentally “invisible” configurations in the model space. To reiterate a statement made previously, it is virtually impossible to guarantee that deduced fine trends such as an apparent negative heat capacity are not simply artifacts resulting from the neglect of relevant configurations in the model phase space.

In view of the fact that reasonably-sized systems have a large number of possible
saddle shapes and corresponding crossings of the respective weight functions, one may wonder if, and under what circumstances, irregularities show up in the apparent trends in thermodynamic quantities inferred from a cursory sampling via Coulomb fragmentation channels. Obviously, crossing points of the configurational weight functions are not distributed uniformly in energy. Rather, their distribution is expected to exhibit statistical fluctuations including some local purely statistical bunchings. Sufficiently strong bunchings of crossing points along the excitation energy may show up as irregularities in an apparent caloric curve and perhaps even simulate a negative heat capacity. Such irregularities, however, should not be taken to signal phase transitions or transitions from one preferred individual fragmentation channel to another. Rather, such phenomena reflect statistical fluctuations in the distribution of crossing points in a fashion reminiscent of Ericson fluctuations [23] in compound nucleus decay at low excitations.

B. Signatures of First-order Phase Transitions and Criticality

An instructive view on first-order phase transitions in finite nuclei can be obtained with the conventional definition of phases as referring to parts of the system, which allows one to consider one part of the system being in one phase (liquid) and the other part in a different phase (gas). In this approach, as was pointed out in Ref. [15], an excited nucleus is inherently a two-phase system, consisting of liquid quasi-uniform bulk matter and diluted surface-domain matter. Obviously, these two phases are characterized by different densities, energies per nucleon, heat capacities and even pressures.

With increasing excitation energy, in a quest for maximum entropy, the system seeks out an optimum macroscopic configuration with increased effective level density parameter, but at the cost of also increased potential energy. Note that quite generally, an increase in level density parameter signifies a corresponding reduction in correlations - spatial, but also pairing and shell correlations. An obvious way to achieve maximum entropy leads through thermal expansion [12]. The other way leads through a transfer of matter from the spatially more correlated bulk to the less correlated surface domain, in what may be viewed as a true first-order liquid-gas phase transition. Such transfer is effected at the cost of increased potential energy in two distinctly different ways. Firstly,
the surface area and diffuseness will increase with increasing excitation energy. Secondly, and more importantly from the experimental point of view, thermal shape fluctuations grow in size with increasing excitation energy allowing one to experimentally probe their reaching of transition state deformations.

The presence of such a first-order, bulk-to-surface phase transition manifests itself in the Coulomb fragmentation excitation function as a fast succession of crossings of weight functions for fragmentation saddle configurations with successively increasing surface areas (see Fig. 5). This is a true conventional first-order liquid (bulk) – gas (surface) phase transition with latent heat represented by the amount of energy needed to cause transfer of a certain amount of nuclear matter from bulk to surface domain. With increasing excitation energy, the individual configuration entropy functions eventually converge at a critical excitation energy, where the surface tension vanishes. As a result of this convergence, the probabilities of observing experimentally various multi-fragment configurations will be governed by combinatorial principles only, resulting in a power-law distribution of fragment masses and, consequently, in an appearance of signatures of criticality.

While it is not obvious what can be learned from discussing Coulomb fragmentation in the framework of first-order phase transitions, it appears likely that experimental trends and patterns of this phenomenon reflect primarily properties of the diluted, non-uniform nuclear surface matter. Therefore, a better understanding of fragmentation properties may help to detail the mapping of the EOS of moderately dilute nuclear matter, including its isospin dependence.

IV. DISCUSSION AND SUMMARY

It appears rather obvious that trends in apparent thermodynamical quantities modeled in numerical simulations of nuclear systems that sample only a fraction of the microcanonical or canonical phase space do not accurately represent corresponding trends in their actual physical counterparts. This observation poses the question of whether or not such model trends reflect at least qualitatively the correct underlying physics and, conversely, under what circumstances predicted model trends in apparent quantities may
even be qualitatively misleading. The present study demonstrates that qualitative discrepancies between physical processes and model interpretations are likely to occur for phenomena involving phase transitions, the very focus of many numerical modeling attempts. The numerical examples discussed above show that an omission of macroscopic configurations intermediate between those associated with different phases depletes the partition function selectively exactly in the excitation energy domain of interest but not much beyond it. As a result of such a depletion, one observes a deficit in apparent entropy (so called “convex intruder”) and an apparent or false negative heat capacity in the model calculations. No such deficit would be present in a calculation performed with a more complete phase space and a proper modeling of the containment vessel consistent with the 2nd Law of Thermodynamics.

While modeling of a confined system is of purely academic interest, modeling the decay modes of excited nuclear systems is of practical importance. This latter venture aims at a deeper understanding of experimental observations, rather than just parameterizing them. For the first time, the present study demonstrates that Coulomb fragmentation exhibits signatures deceptively similar to those of phase transitions in confined systems. Both sets of signatures are understood to reflect crossings of respective weight functions with increasing system excitation. Hence, this work offers a plausible explanation why Coulomb fragmentation can easily be mistaken for a nuclear phase transition in a confined system, a transition of either first or second order. It is also important to note that, not only in theoretical modeling but also in measurements of binary or multi-fragment decay, only a fraction of the relevant underlying phase space is sampled. Sampled are only saddle-point configurations but none of the “intermediate” pre-saddle configurations. Therefore, also experimentally, one can determine directly only apparent thermodynamic quantities and their trends, but not the trends in the true thermodynamic quantities describing the highly excited nuclear systems. A more accurate determination of trends has to rely on a comparison with simulations admitting a representative, sufficiently large subspace of the full configuration space.

Because of the large number of possible saddle configurations in reasonably sized systems and because of thermal fluctuations, it is unlikely for any single second-order “phase” transition between two distinct saddle configurations to leave a distinct and
unambiguous experimental “fingerprint”. More likely, phase-transition-like signatures are observable in Coulomb fragmentation when random, statistical “bunching” of the crossing points of weight functions for different saddle configurations occurs at certain excitation energies. Such statistical fluctuations could result in observable irregularities, e.g., on caloric curves, somewhat reminiscent of Ericson fluctuations [23] in compound nucleus decay.

While it is not yet clear, what there is to be learned from the irregularities of apparent thermodynamic quantities extracted from nuclear fragmentation data, it is clear that an analysis of experimental data in terms of Coulomb fragmentation (generalized fission) has important implications for an understanding of properties and behavior of the surface domain of excited nuclei. In particular, such data may allow one to probe specifically the nuclear EOS of the diluted surface domain matter and, separately, also that of bulk nuclear matter diluted through thermal expansion. It is rather obvious from the lessons of compound nuclear fission that the stability of nuclear systems against fragmentation depends critically on the presence and the properties of a diffuse surface domain. As has been pointed out in Ref. [15] and further above, because of the differences in average density, binding energy, and level density parameter between bulk and surface matter, finite nuclei are inherently two-phase systems. As the excitation energy of such a two-phase system is raised, matter is transferred from the bulk to the surface domain, which can be viewed as a true first-order liquid-gas phase transition. It is also natural to expect that, at high excitation energies, the surface tension generated by the density gradient in the surface domain will vanish and give rise to critical phenomena.

The signatures of Coulomb fragmentation can be probed experimentally in greater detail by studying excitation functions for individual fragmentation channels. Since the process appears to be controlled by the manner in which weight functions for various multi-fragment saddle shapes cross in succession, the yield for individual fragmentation channels is expected to exhibit a rise and fall with increasing excitation energy. (See Fig. 4, middle, Gaussian-like curve.) This may potentially open a completely new venue of experimental exploration of Coulomb fragmentation - spectroscopy of multifragment saddle configuration, where the excitation functions for various channels are studied, e.g., as functions of fragment sizes and isospins. Furthermore, it may be possible to
use the location of the peaks in various yields on the energy scale as a measure of excitation energy, i.e., as a tool of calorimetry of highly excited nuclei. Also, there must be situations, where two or more weight functions run quasi-parallel avoiding each other, but crossing some other weight functions. This effect will give rise to gentle Boltzmann-like scalings for some relative yields embedded in phase-transition like scalings of some other yields. This situation may arise when two saddle shapes differ in isospin but not so much in surface area, leading to iso-scaling.

With the above experimental opportunities in mind, a more coordinated theoretical effort appears warranted aiming to achieve a better quantitative understanding of the evolution of the properties of the surface domain with excitation energy [17, 18] and other variables. To understand nuclear dynamics on a deeper level, it is important to assess the morphing of quasi-symmetric binary fission first into asymmetric binary Coulomb fragmentation, and then into multiple fragmentation. Such an effort would also advance thermodynamic theory of small open quantal systems inherently endowed with diffuse surface domains - a valuable goal in its own merits.

Finally, it is worth noting that, while the physics underlying the present schematic model differs conceptually from that reflected implicitly in the “benchmark” multifragmentation models SMM [5] and MMMC [6], the formal basis of all three models is the same [15]. It is expressed in the fact that more fragmented (i.e., less correlated) spatial configurations show higher entropy production rates (larger effective level density parameter \(a\)) and thus are able to “catch” up in terms of entropy (as excitation energy is increased) with more correlated but less fragmented configurations. In the present model, the extra catch-up entropy is produced for fragmented configurations due to their larger total surface domain. In SMM [5] and MMMC [6] models, on the other hand, extra entropy comes from the implicit thermal motion of spherical constituents of fragmented state within a hypothetical oversize confinement vessel - named as “freezeout” volume. By adjusting the volume of this latter invisible vessel, one fine-tunes the magnitude of the extra entropy, such that these models may ultimately appear to provide fits to selected experimental data. It is interesting to note that when moderately time-averaged, such thermal motion of the spherical fragments produces an effective matter distribution that emulates the diffuseness of the surface domain of the actual matter distribution.
and emulates also complex multi-fragment saddle geometries. Consequently, to a good extent, the extra entropy generated by the fragment thermal motion is equivalent to the entropy due to the diffuse surface domain of the matter distribution. In fact, it is the latter observation that has inspired the present effort of exploring the role of the surface diffuseness in facilitating fragmentation of highly excited nuclear systems.

**Acknowledgments**

This work was supported by the U.S. Department of Energy grant No. DE-FG02-88ER40414.


Isoscaling in statistical fragment emission in an extended compound nucleus model

W. Ye *, J. Tőke and W. U. Schröder

Departments of Chemistry and Physics, University of Rochester, Rochester, New York 14627, USA

Abstract

Based on an extended compound nucleus model, isospin effects in statistical fragment emission from excited nuclear systems are investigated. An experimentally observed scaling behavior of the ratio of isotope yields $Y_i(N, Z)$ from two similar emitting sources with different neutron-to-proton ratios is predicted theoretically, i.e., the relationship of $Y_2/Y_1 \propto \exp(\alpha N + \beta Z)$ is demonstrated. The symmetry energy coefficient $C_{\text{sym}}$ extracted from the simulation results is $\sim 27$ MeV which is consistent with realistic theoretical estimates and recent experimental data. The influence of the surface entropy on the isoscaling behavior is discussed in detail. It is found that although the surface entropy increases the numerical values of isoscaling parameters $\alpha$ and $\beta$, it does not affect the isoscaling behavior qualitatively and has only a minor effect on the extracted symmetry energy coefficient.

Key words: Isoscaling, Compound nucleus, Fragment emission

PACS: 25.70.Pq, 24.10.Pa, 21.65.+f, 21.60.Ev

1 INTRODUCTION

The search for a nuclear equation of state (EOS) has been one of the dominant factors driving interest in heavy-ion collisions at intermediate and high energies, ever since such beams have become available. Exploration of the isospin dependence of the EOS is an inseparable part of the task. Originally, the latter studies were confined to use stable projectile beams with only a moderately wide range of isotopes between neutron-poor and neutron-rich nuclei. However,

* Present address: Department of Physics, Southeast University, Nanjing 210096, People’s Republic of China
exotic secondary beams of projectile nuclei with extreme neutron-to-proton ratios that have become available recently offer new and intriguing opportunities to study isospin physics in heavy-ion collisions [1–3]. These studies have already resulted in the discovery of several interesting isospin-related systematics [4–7], prompting further efforts both, in experiments and in theoretical modeling. In particular, a so-called isoscaling analysis of light fragment emission in the decay of very hot nuclear systems produced in heavy-ion collisions has attracted attention as a possible tool for deducing the nuclear symmetry energy from the relative fragment yields [7–9,11,10].

For strongly damped collisions fragment isotopic yields were found to follow a “Qgg systematics” [12–14]. Isotopic scaling, termed “isoscaling” [7–9], is observed for other types of reactions such as evaporation [7,15], fission [16,17], projectile fragmentation [18,19], and multifragmentation [7,5,6]. This scaling law refers to a general exponential relation between the ratios of yields $Y_i(N, Z)$ for fragments $(N, Z)$ emitted from systems which differ only in their isospin or $(N_s, Z_s)$, In particular, if two reactions lead to primary systems $i = 1$ and 2 having approximately the same temperature but different isospin, the ratio $R_{21}(N, Z)$ of the experimental yields of a given fragment $(N, Z)$ emitted from these systems exhibits an exponential dependence on the fragment neutron number $N$ and atomic number $Z$ of the form,

$$R_{21}(N, Z) = \frac{Y_2(N, Z)}{Y_1(N, Z)} = Cexp(\alpha N + \beta Z).$$

(1)

Here, $\alpha$ and $\beta$ are the isoscaling parameters and $C$ is an overall normalization constant.

On the theoretical side, isoscaling has been extensively examined in the antisymmetrical molecular dynamics model [20], a Boltzmann-Uehling-Uhlenbeck model [5], a lattice gas model [21,22], the expanding evaporating source model [23] and in statistical multifragmentation models [8,9,11,24]. In the present paper it will be shown how the essential features of isoscaling observed in experiment are related to the nuclear symmetry energy in an extended compound nucleus (ECN) model. This model is closely related to that known from fission studies, and its central notion is a relatively high entropy (particularly at moderately high excitation energies) associated with the diffuse nuclear surface region (as opposed to bulk matter). Application of this ECN model reveals a new mechanism of nuclear fragmentation caused by the softening of the diffuse nuclear surface at high excitations [25]. The ECN model is equivalent to conventional compound nucleus models at low excitation energy as represented, e.g., by GEMINI [26], but greatly extends the validity of the compound nucleus concept towards high excitations [27]. Therefore, the ECN model provides a unified description of statistical emission of light particles and fragments in a wide range of excitation energies. While the present model
is still somewhat schematic, it is based on a physically transparent picture and thus provides direct insight into the phenomena of interest.

2 THEORETICAL FRAMEWORK

The probability \( p \) of emitting a fragment from an equilibrated compound nucleus (CN) is evaluated using the Weisskopf formalism [28]:

\[
    p \propto e^{\Delta S} = e^{S_{\text{saddle}} - S_{\text{eq}}},
\]

where \( S_{\text{eq}} \) and \( S_{\text{saddle}} \) are the entropies for the equilibrated CN and a saddle-point configuration of touching spheres, respectively. Within the Fermi gas model, the entropies for the two configurations can be calculated as

\[
    S_{\text{eq}} = 2\sqrt{a_A E^*_{\text{tot}}},
\]

and

\[
    S_{\text{saddle}} = S_{\text{res}} + S_{\text{frag}} = 2\sqrt{(a_{\text{res}} + a_{\text{frag}}) E^*_{\text{th saddle}}}.
\]

In Eqs. (3) and (4), \( a_A, a_{\text{res}}, \) and \( a_{\text{frag}} \) are the level density parameters of the CN system at equilibrium, of the residue, and of the fragment, respectively. The part \( S_S \) of the entropy \( S \) of the system, associated with the diffuse surface domain, has been found [29] to have a pronounced effect on the fragment emission probability. One has

\[
    S = S_V + S_S,
\]

where \( S_V \) is the entropy of bulk matter. Consequently, the level density parameter \( a \) includes volume and surface terms [30],

\[
    a = a_V + a_S = \alpha_V A + \alpha_S A^{2/3} F_2,
\]

Here \( A \) is the atomic number, \( F_2 \) is the surface area relative to a spherical shape. \( E^*_{\text{th saddle}} \) in Eq.(4) is the thermal excitation energy of the system in the saddle-point configuration. The latter quantity is calculated as

\[
    E^*_{\text{th saddle}} = E^*_\text{tot} - V_{\text{saddle}},
\]
where $V_{saddle}$ is the collective saddle-point energy.

For the level density parameter $a$, the parametrization of Tőke and Swiatecki [30] was employed with $\alpha_v = 1/14.6$ MeV$^{-1}$ and $\alpha_s = 4/14.6$ MeV$^{-1}$. The calculations assume saddle-point shapes to be represented by two touching spheres. $V_{saddle}$ is the difference in deformation energies for the saddle-point shape and equilibrium-state shape. It contains contributions from Coulomb, volume, and surface energies. Volume and surface energies depend on the system isospin $I$ (defined as $(N - Z)/A$) in a functional form $-\alpha_v(1 - \kappa_v I^2)A$ and $\alpha_s(1 - \kappa_s I^2)A^{2/3}$, respectively. The parameters are taken from [31] as $\alpha_s = 21.13, \kappa_s = 2.3, \alpha_v = 15.9937, \kappa_v = 1.927$. The nuclear temperature can be obtained from the commonly used Fermi-gas model relationship between the temperature $T$ and the excitation energy of the system $E^*_\text{tot}$:

$$T = \sqrt{\frac{E^*_\text{tot}}{a}}. \quad (8)$$

The present calculations do not account for the effects of an expansion of the CN prior to its decay, so the nuclear density is that of the ground state. Therefore the additional influence of the nuclear matter density on the extraction of symmetry energy coefficient is not studied here. The present work concentrates on the temperature dependence of the relation between symmetry energy and isoscaling parameters.

3 RESULTS AND DISCUSSION

In this work several pairs of equilibrated CN sources with proton number $Z_s = 75$ and mass numbers $A_s = 165, 175, 185, \text{ and } 195$, are considered at initial excitation energies of $E^*_\text{tot}/A = 2, 3, 4, 5, 6, \text{ and } 7 \text{ MeV/nucleon}$. As the ratio $R_{21}(N, Z)$ is insensitive to sequential decay, it carries information on the original excited fragments prior to their decay [18]. In particular, it has been found that the values of $\alpha$ and $\beta$ are not much affected by sequential decay of the primary fragments [9]. These observations justify the neglect of sequential decay in the following analysis.

The yield ratio $R_{21}(N, Z)$ is constructed using the convention that index 2 refers to the neutron-rich system and index 1 to the neutron-poor one. The observation summarized in Eq. (1) shows that experimental yield values of $\ln(R_{21}(N, Z))$ plotted vs. $N$ ($Z$ fixed) or $Z$ ($N$ fixed), produce straight lines. This feature also emerges from the present MCN model. As a demonstration, Fig. 1 shows the theoretical yield ratios $\ln(R_{21}(N, Z))$ plotted vs. fragment
neutron number $N$, for individually values of $Z = 6 − 9$ (top panel), and vs. proton number $Z$, for individually values of $N = 6 − 9$ (bottom panel). The ratios have been calculated for the two source pairs ($A_2 = 175, A_1 = 165$) and ($A_2 = 185, A_1 = 165$) at an excitation energy of $E^*_{tot}/A = 5$ MeV/nucleon. One can see from Fig.1 that isotope ratios of the same element $Z$, or isotone ratios of the same $N$, tend to lie on a logarithmic straight line. The solid lines represent linear fits to each series of $Z$ isotopes and each series of $N$ isotones, respectively. It is obvious that these fit lines are nearly parallel to each other. The positive slope in the upper panel of Fig.1 illustrates that neutron-rich fragments are more easily produced from the more neutron-rich sources, as can be expected. Analogously, the negative slope in the bottom panel of Fig.1 illustrates that proton-rich fragments are more readily produced from the more proton-rich emitting sources. These are the typical features associated with “isoscaling” behavior showing that the scaling behavior of the isotope and isotone yield ratios appears naturally within the present CN model framework.

It was reported [29] that the ratio of a certain fragment $(N, Z)$ emission probability from two equilibrated systems can approximately be written as

$$P_2(N, Z)/P_1(N, Z) \propto \exp[(V_1(N, Z) - V_2(N, Z))/T],$$

where $V_i (i = 1, 2)$ is of the same meaning as $V_{saddle}$, namely the interaction energy at the saddle point. An analysis indicates that in the difference $(V_1 - V_2)$, most terms cancel except for terms that are directly related to the isospin of the CN system. These isospin-related terms can be combined as $(\alpha_v \kappa_v - \alpha_s \kappa_s / A^{1/3})(N - Z)^2/A = C_{sym} (N - Z)^2/A = E_{sym}$. Here $C_{sym} (= \alpha_v \kappa_v - \alpha_s \kappa_s / A^{1/3})$ is symmetry energy coefficient [32] incorporating both the volume and surface contributions to the symmetry energy $E_{sym}$ [9]. Hence the value of $(V_1 - V_2)$ is dominantly determined by the difference in symmetry energy between the two emitting sources and their residues. This demonstrates that the origin of the isoscaling phenomenon found in the present frame can be traced to the symmetry energy term in the nuclear binding energy, as also suggested in other work (e.g., [7]).

The isoscaling parameters $\alpha$ and $\beta$ can be extracted from fits of model predictions to the data points shown in Fig.1. An average value of $\alpha$ is calculated over the range $5 \leq Z \leq 9$, an average $\beta$ is calculated over the range $5 \leq N \leq 9$. Figure 2 depicts the dependence of the average coefficients $\alpha$ and $\beta$ vs. excitation energy (left panel) and vs. inverse of the temperature (right panel). One notices that the absolute values of $\alpha$ and $|\beta|$ decrease monotonically with excitation energy, implying that isospin effects decrease with increasing excitation energy. There is a significant sensitivity to the excitation energy at low energy, but both the sensitivity to excitation energy and the overall isospin effect are weakened at very high excitation. In addition, it is rather evident that both $\alpha$ and $\beta$ show a linear dependence on $1/T$.

The surface contribution $a_S$ to the level density parameter $a$ appreciably enhances the fragment emission probability via significant surface entropy effects.
It is therefore interesting to examine the effects of surface entropy on the isoscaling phenomenon in the present model. Results are displayed in Figs. 3 and 4, to be compared to Figs. 1 and 2 which have been calculated without surface contribution to the entropy. Evidently, familiar isoscaling behavior is predicted well in either case. The linear relationship between isoscaling parameters and inverse temperature is not affected by the surface contribution either, as seen from the right panel of Fig. 4. However, an account of the surface term predicts larger values of $\alpha$ and $\beta$ (comparing Fig. 4 and Fig. 2), implying that the surface entropy effect on fragment emission becomes stronger with increasing isospin of the emitting sources, also as expected.

Since the scaling behavior of ratios of fragment isotopic yields measured in separate nuclear reactions has been utilized to probe the symmetry energy [7,10,33,32], in the following a symmetry energy coefficient is extracted from the theoretical isoscaling systematically analytical expressions for $\alpha$ and $\beta$. It is worth noting that, for a given $E_{\text{tot}}/A$ these four sources have the same temperature when the surface contribution to the level density parameter is neglected.

It has been shown [9,11] that the isoscaling parameters $\alpha$ and $\beta$ are related to the symmetry energy coefficient $C_{\text{sym}}$ as

$$\alpha = 4\frac{C_{\text{sym}}}{T} \left[ (\frac{Z_s}{A_s})_1^2 - (\frac{Z_s}{A_s})_2^2 \right]$$ (9)

and

$$\beta = 4\frac{C_{\text{sym}}}{T} \left[ (\frac{N_s}{A_s})_1^2 - (\frac{N_s}{A_s})_2^2 \right].$$ (10)

Equations (9) and (10) have been used to constrain the symmetry energy coefficient $C_{\text{sym}}$ based on experimental data (e.g. Ref. [10]). Figure 5 depicts the product $\alpha \cdot T$ as a function of $(Z_s/A_s)_1^2 - (Z_s/A_s)_2^2$ and $\beta \cdot T$ as a function of $(N_s/A_s)_1^2 - (N_s/A_s)_2^2$ for the initial four source pairs at various excitation energies. All these systems with different source sizes and isospin asymmetries lie along one single straight line, which illustrates that the isoscaling parameters $\alpha$ and $\beta$ are not sensitive to the system size. By fitting the theoretical data in Fig. 5 with Eqs. (9) and (10), a symmetry energy coefficient $C_{\text{sym}}$ can be obtained to be $27.3 \pm 0.1$ MeV, which is close to the standard liquid drop model value $C_{\text{sym}} = 25$ MeV and also in reasonable agreement with that obtained in Ref. [18]. In that experimental work [18], a value of $C_{\text{sym}} = (27.2 \pm 2.2)$ MeV was deduced based on an experimental analysis of 25-MeV/nucleon $^{86}$Kr + $^{124}$Sn reactions. The present analysis demonstrates consistency with other realistic theoretical estimates [34,3] and indicates the validity of an interpretation of isoscaling data in terms of the symmetry energy.
Although the inclusion of the surface term \(a_s\) in the level density parameter leads to a difference in the temperatures for the four systems, calculations indicate that for each excitation energy \(E_{\text{tot}}^*/A\), the temperature differences among the four sources are less than 0.1 MeV, so the temperature value of four systems can be regarded as approximately constant when the excitation energy per nucleon is fixed. Figure 6 displays the effect of the surface entropy on the extracted symmetry energy coefficient. The value deduced from the theoretical data in Fig. 6 is \(C_{\text{sym}} = (27.6 \pm 0.1)\) MeV, which is comparable with that predicted without considering the surface entropy effects. The reason is that although the surface entropy increases the values of \(\alpha\) and \(\beta\), it also decreases the value of temperature due to an increased level density parameter \(a\) [see Eq. (6)]. Consequently, \(\alpha \cdot T\) and \(\beta \cdot T\) are little affected by the surface entropy. This result indicates that surface entropy probably has a minor influence on the extraction of the symmetry energy coefficient. Moreover, it also implies that the isoscaling observable is a robust probe of the symmetry energy.

4 SUMMARY AND CONCLUSIONS

In conclusion, in the framework of an extended compound nucleus model, isoscaling behavior and its relation to the nuclear symmetry energy is revealed. The symmetry energy coefficient is found to be \(C_{\text{sym}} \sim 27\) MeV from an analysis of theoretical “data”, suggesting that isoscaling data can be interpreted in terms of the symmetry energy. In addition, in this work surface entropy influences on the isoscaling phenomenon have been studied. The present ECN model approach leaves sufficient room for further improved treatment of interesting physical effects [29], such as nuclear expansion. Such a more detailed analysis is required to study the evolution of the symmetry energy with the excitation energy, as has been explored recently also by Shetty et al. [35]. The model will be utilized to deduce the effects of nuclear expansion on the isoscaling parameters and will correspondingly illustrate how to deduce the density dependence of the symmetry energy in such more general and realistic scenarios. Work along this direction is in progress.

ACKNOWLEDGMENTS

This work is supported by the U.S. Department of Energy Grant No. DE-FG02-88ER40414. The work of W.Y is also partially supported by the NSFC
under Grant No. 10405007 and China scholarship council. W.Y is also grateful to Rochester University for support and hospitality extended to him.
References


Fig. 1. (Color online) The logarithm of the ratio of elements $Z = 5-9$ isotopes yields (top panel) and of $N = 6-9$ isotones yields (bottom panel) from pairing source $A_s = 175$ and $A_s = 165$ (left column) as well as from pairing source $A_s = 185$ and $A_s = 165$ (right column) at excitation energy $E_{\text{tot}}^\ast/A = 5$ MeV/nucleon. Here the calculations do not consider the surface entropy, i.e., not including $a_S$ in the expression for the level density parameter $a$ [see Eq. (6)]. Solid lines are the linear fitting to the data points.
Fig. 2. (Color online) Dependence of isoscaling parameters $\alpha$ and $|\beta|$ on excitation energy (left panel) and the inverse temperature (right panel) for various source pairs. Symbols in the figures are $\alpha$ (solid symbols) or $|\beta|$ (open symbols) from four source pairs $Y_{A_x}=175/Y_{A_s}=165$ (squares), $Y_{A_x}=185/Y_{A_s}=165$ (circles), $Y_{A_x}=195/Y_{A_s}=175$ (up-triangles), and $Y_{A_x}=195/Y_{A_s}=185$ (down-triangles). Here the calculations do not consider the surface entropy, i.e., not including $a_S$ in the expression for the level density parameter $a$ [see Eq. (6)].
Fig. 3. (Color online) Same as Fig 1 but consider the surface entropy effect via inclusion of $a_S$ in the expression for the level density parameter $a$ [see Eq. (6)].
Fig. 4. (Color online) Same as Fig.2 but consider the surface entropy effect via inclusion of $a_s$ in the expression for the level density parameter $a$ [see Eq. (6)].
Fig. 5. (Color online) $\alpha \cdot T$ (positive parts) and $\beta \cdot T$ (negative parts) as a function of $(Z_s/A_s)_{1}^{2} - (Z_s/A_s)_{2}^{2}$ or as a function of $(N_s/A_s)_{1}^{2} - (N_s/A_s)_{2}^{2}$ of the sources for four source pairs, i.e., $Y_{A_s=175}/Y_{A_s=165}$, $Y_{A_s=185}/Y_{A_s=165}$, $Y_{A_s=195}/Y_{A_s=175}$, and $Y_{A_s=195}/Y_{A_s=185}$, at excitation energies of $E_{tot}/A = 2, 3, 4, 5, 6$, and $7$ MeV/nucleon. Here the calculations do not consider the surface entropy, i.e., not including $a_S$ in the expression for the level density parameter $a$ [see Eq. (6)].
Fig. 6. (Color online) Same as Fig. 5 but consider the surface entropy effect via inclusion of $a_S$ in the expression for the level density parameter $a$ [see Eq. (6)].
Surface entropy and density dependence of the symmetry energy in a harmonic interaction Fermi gas model

W. Ye,1,2 J. Tőke,1 and W. U. Schröder1

1Department of Chemistry and Physics,
University of Rochester, Rochester, New York 14627, USA
2Department of Physics, Southeast University,
Nanjing 210096, People’s Republic of China

Abstract

An experimentally observed decreasing function of the isoscaling parameter, $\alpha$, and symmetry energy with excitation energy is predicted theoretically in the framework of a harmonic interaction Fermi gas model which takes the nuclear expansion into account. On this basis, the density dependence of the symmetry energy is extracted from our simulation, namely $C_{sym} = 25.2(\rho/\rho_0)^{0.46}$ and $22.0(\rho/\rho_0)^{0.71}$ for the cases that the surface entropy effect on the fragment emission is considered or not. It is found that surface entropy has an effect on the density density and increases the magnitude of the symmetry energy.

PACS numbers: 25.70.Pq, 24.10.Pa, 21.65.+f, 21.60.Ev
I. INTRODUCTION

Isospin dependent phenomena have attracted considerable interest in recent years since it can help reveal the asymmetry term of the nuclear equation of state (EOS). The key unknown in the EOS of asymmetric nuclear matter is the symmetry energy, particularly its density dependence. The latter is important for understanding heavy ion reactions [1–4], the structure of neutron-rich nuclei [5] as well as many interesting astrophysical problems [6, 7]. The availability of beams with large neutron-to-proton ratios provides an opportunity to explore the symmetry energy term in nuclear equation of state in very asymmetry nuclear systems. Experimental information on the symmetry energy can be obtained from nucleus-nucleus collisions. To study EOS at subnormal densities, experimentally one possible means is through the intermediate energy heavy-ion reactions. For example, the disassembly of a hot expanded nucleus offers the best tool to explore the characteristics of the symmetry energy [8, 9].

Up to now impressive results about the determination the symmetry energy have been obtained by means of various theoretical approaches, ranging from dynamical models [10–15], a lattice gas model [16, 17], to statistical models [18–27]. Also, it was suggested recently that by analyzing the the scaling behavior of isotopic yields measured in two different reactions, one can extract important information about the symmetry energy and its density dependence [8, 9, 18, 28, 29]. Despite these progresses, the density dependence of symmetry energy has remained largely unconstrained and been the most uncertain part of the EOS of neutron-rich matter [3]. In this context, more experimental and theoretical works are therefore needed to probe the density dependence of symmetry energy. In a recent work, experimentally observed isoscaling phenomenon has naturally appeared in a harmonic interaction Fermi gas (HIFG) model [30, 31], and moreover the theoretically predicted symmetry energy at normal nuclear density has been found to be consistent with realistic theoretical estimates and experimental data [32]. Therefore, this HIFG model will be employed here to deduce the density dependence of symmetry energy. Since an expansion process of a compound nucleus prior to its decay can alter the compound nucleus decay pattern appreciably [33], in particular when the surface entropy effect on the fragment emission is considered [31], the present work will account for the expansion effect in the framework of the HIFG
model. On this basis, the role of surface entropy effect in deducing the density dependence of the symmetry energy will be investigated.

The paper is organized as follows. Section II presents a brief introduction to our model. Results and discussion are given in Sec. III. The nuclear expansion effects on the isoscaling behavior are first studied. Then the evolution of isoscaling parameters and the symmetry energy with excitation energy is analyzed. After that the density dependence of the symmetry energy is deduced, and the effect of the surface entropy on the density dependence is investigated in detail. Conclusions and further work are given in Sec. IV.

II. THEORETICAL MODEL

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.,

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

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

$$
S = 2\sqrt{aE^*_{\text{th}}} = 2\sqrt{a(E^*_{\text{tot}} - E_{\text{compr}})}, \tag{2}
$$

where $a$ is the level density parameter, $E^*_{\text{th}}$ is the thermal excitation energy, and $E_{\text{compr}}$ is the collective compressional energy. It has been pointed out [31] that the presence of a surface contribution to the level density parameter is of crucial importance as it describes that part $S_S$ of the entropy $S$ of the system, which is associated with the diffuse surface domain. This surface contribution has been found to have a pronounced effect on the fragment emission probability. One has

$$
S = S_V + S_S, \tag{3}
$$

where $S_V$ is the entropy of the bulk matter. Because of this reason, the level density parameter $a_0$ at ground-state matter density $\rho_0$ includes volume and surface terms [34]

$$
a_0 = a_V + a_S = \alpha_V A + \alpha_S A^{2/3} F_2, \tag{4}
$$
where $A$ is the atomic number, $F_2$ is the surface area relative to a spherical shape. The dependence of the level density parameter on the nuclear matter density for finite nuclear matter is given by the Fermi-gas model:

$$a(\rho) = a_0 \left(\frac{\rho}{\rho_0}\right)^{-2/3}, \quad (5)$$

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

$$f_\rho(r) = c^3 f_0(cr), \quad (6)$$

where $f_0(r)$ is the ground-state density profile function and $c$ is a scaling constant.

The compressional energy in Eq. (2) is approximately in the present study following prescription proposed in the expanding emitting model [35], i.e.,

$$E_{compr} = E_b \left(1 - \frac{\rho}{\rho_0}\right)^2, \quad (7)$$

where $E_{compr}$ and $E_b$ are the compressional and the ground-state binding energies, respectively. This equation ensures that the compressional energy varies with $\rho$, from zero at ground-state density $\rho_0$ to $E_b$ at zero density.

Equations (1)–(3) and (7) allow one to obtain an analytical expression for the equilibrium density $\rho_{eq}/\rho_0$ of nuclear matter as a function of the excitation energy:

$$\frac{\rho_{eq}}{\rho_0} = \frac{1}{4} \left(1 + \sqrt{\frac{9}{8} - \frac{E_\text{tot}}{E_b}}\right). \quad (8)$$

Equation (8) reflects the fact that for $E_\text{tot} < E_b$, 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 $1/2 \leq \rho_{eq}/\rho_0 \leq 1$.

The probability $p$ of emitting a fragment from an equilibrated compound nucleus (CN) is evaluated using the Weisskopf formalism [36]:

$$p \propto e^{\Delta S} = e^{S_{\text{saddle}} - S_{\text{eq}}}, \quad (9)$$

where $S_{\text{eq}}$ and $S_{\text{saddle}}$ are the entropies for the equilibrated CN and the saddle-point configuration of touching, respectively. Within the Fermi gas model, the entropy for the two configurations can be calculated as

$$S_{\text{eq}} = 2 \sqrt{a_A[E_\text{tot} - E_b(1 - \frac{\rho_{eq}}{\rho_0})^2]}, \quad (10)$$
and

\[ S_{\text{saddle}} = S_{\text{res}} + S_{\text{frag}} = 2\sqrt{(a_{\text{res}} + a_{\text{frag}})E_{\text{saddle}}^*}. \]  

(11)

In Eqs. (10) and (11), \( a_A \), \( a_{\text{res}} \) and \( a_{\text{frag}} \) are the level density parameters of the system at equilibrium, the residue, and the fragment, respectively. \( E_{\text{saddle}}^* \) in Eq.(11) is the thermal excitation energy of the system in the saddle-point configuration. The latter quantity is calculated as

\[ E_{\text{saddle}}^* = E_{\text{tot}}^* - E_b(1 - \frac{\rho_{\text{eq}}}{\rho_0})^2 - V_{\text{saddle}}, \]  

(12)

where \( V_{\text{saddle}} \) is the collective saddle-point energy.

For the level density parameter \( a \), the parametrization of Töke and Swiatecki [34] was employed with \( \alpha_V = 1/14.6 \text{ MeV}^{-1} \) and \( \alpha_S = 4/14.6 \text{ MeV}^{-1} \). In the calculations, assuming the saddle-point shape is represented by two touching spheres. \( V_{\text{saddle}} \) is the difference of deformation energy between the saddle-point shape and equilibrium-state shape, which is composed of the differences of Coulomb energy, volume energy, and surface energy between these two shape configurations. The volume energy and surface energy are related to the system isospin \( I \) (defined as \((N-Z)/A\)) and denoted by \(-\alpha_v(1-\kappa_v I^2)A \) and \( \alpha_s(1-\kappa_s I^2)A^{2/3} \), respectively. The parameters are taken from [38] as \( \alpha_s = 21.13, \kappa_s = 2.3, \alpha_v = 15.9937, \kappa_v = 1.927 \). The nuclear temperature can be obtained from the commonly used Fermi-gas model relationship between the temperature \( T \) and the thermal excitation energy the system \( E_{\text{th}}^* \):

\[ T = \sqrt{\frac{E_{\text{th}}^*}{a(\rho)}} = \left(\frac{\rho_{\text{eq}}}{\rho_0}\right)^{1/3} a_0^{-1/2} \sqrt{E_{\text{tot}}^* - E_b(1 - \frac{\rho_{\text{eq}}}{\rho_0})^2}. \]  

(13)

Note that temperature varies with the density \( \rho_{\text{eq}}/\rho_0 \).

III. RESULTS AND DISCUSSION

It has been shown that the ratio of the fragment isotopic yields in two different nuclear reactions, 1 and 2, \( R_{21} = Y_2(N, Z)/Y_1(N, Z) \), obey an exponential dependence on the fragment neutron number \( N \) and the proton number \( Z \) of the isotopes, an observation known as isoscaling [18]. This dependence is characterized by three parameters \( \alpha, \beta \) and \( C \):

\[ R_{21}(N, Z) = C \cdot \exp(\alpha N + \beta Z), \]  

(14)
where $C$ is an overall normalization constant. It was indicated [18, 21] that the isoscaling parameter $\alpha$ is related to the symmetry energy through a relation

$$\alpha = \frac{4C_{sym}}{T}[(\frac{Z}{A})_1^2 - (\frac{Z}{A})_2^2], \quad (15)$$

Here $Z_1, A_1$ and $Z_2, A_2$ are the charge and the mass numbers of the source 1 and 2, respectively. $T$ is the temperature of the system in MeV, $C_{sym}$ is the symmetry energy.

In this work four equilibrated sources with proton number $Z_s = 76$ and mass numbers $A_s = 165, 175, 185,$ and $195,$ are chosen at several initial excitation energies $E_{tot}^*/A.$ Since most models show that the effect from sequential decays on isoscaling is negligible [39], in this study we will pay attention to the primary fragments and neglect their sequential decay.

Due to the nuclear expansion, the density of the system changes. This possibly affects the symmetry energy [37]. In our calculations its effect on Coulomb energy is estimated through radius parameter

$$r_{coul} = r_0(\rho/\rho_0)^{-1/3} \quad (31)$$

with $r_0$ being 1.16 fm [38]. Besides, the variation of the density changes the temperature of the system [Eq.(13)], and hence affects the volume energy and surface energy coefficients. This effect can be estimated via the free energy formula suggested in [21], and the resulting expressions for volume energy and surface energy used here are the following:

$$E_v(T) = (\epsilon_0 - \alpha_v T^2/\epsilon_0)((1 - \kappa_v T^2)A, \quad (16)$$

and

$$E_s(T) = (\alpha_s(T) - T\frac{d\alpha_s(T)}{dT})(1 - \kappa_s T^2)A^{2/3}. \quad (17)$$

where $\epsilon_0 = 16$ MeV, and $\alpha_s(T) = \alpha_s(T_c^2 - T^2)^{5/4}$ with $T_c$ being the critical temperature of infinite nuclear matter, whose value is set as 18 MeV. Other symbols in formulas (16) and (17) are of the same meaning as those mentioned before. Because the magnitude of the compressional energy plays a role in the fragment emission, to better survey the isospin effect of the system on the emission of the fragments, the values of the ground-state binding energy, $E_b,$ appearing in the compressional energy formula [Eq.(7)] for different isospin systems are directly taken from the experimental data [40].

Figure 1 displays the effect of nuclear expansion on the isoscaling parameter $\alpha.$ Solid points are theoretical values, solid lines are the linear fits to the data points. One can see that these fitting lines are nearly parallel to each other. Since the distances between elements are related to the proton isoscaling parameter $\beta,$ Fig.1 shows the almost equivalent distances.
between two successive elements for $Z = 5$ to 9. These observations indicate that isoscaling is insensitive to the nuclear expansion. In addition, given the nuclear expansion can alter the compound nucleus decay pattern significantly when surface entropy contribution to the level density is included [31], we depict in the bottom panel of Fig.1 the surface entropy effect on the isoscaling behavior. A comparison with the top panel of Fig.1 exhibits that although the surface entropy has an influence on the magnitude of the $R_{21}$, the isoscaling still holds.

A nucleus expands with increasing excitation energy, leading to a change of density. This implies a density dependence of the symmetry energy. To deduce the density dependence by means of the isoscaling technique, we study the evolution of isosacling parameters $\alpha$ with excitation energy. The observable $\alpha$ can be constructed from the yield ratio of isotope fragments (see Fig.1), and be extracted by reproducing the data points shown in Fig.1. Here an average value of $\alpha$ is evaluated over the range $5 \leq Z \leq 9$. The calculations are performed for two pair sources ($A_2 = 175, A_1 = 165$) and ($A_2 = 185, A_1 = 165$) at different excitation energies. Results are shown in Fig.2. Two evident features are observed from this figure. One feature is that $\alpha$ is a decreasing function of excitation energy, a phenomenon that has been reported in some experiments [9, 41, 42]. It indicates that the experimentally observed changing trend of $\alpha$ with excitation energy is predicted theoretically in the HIFG model. As for the symmetry energy ($C_{sym}$) (calculated with Eqs. (15) and (13)) vs. excitation energy, one can notice that the numerical values of the symmetry energy become smaller with increasing excitation energy. This is consistent with the experimental observation by Fevre et al. [42], where a smaller symmetry energy at high excitations has been suggested by comparing the $C_{sym}$ values deduced from the data of the peripheral and central collisions in the reaction of $^{12}$C on $^{112,124}$Sn.

The second feature is concerning the surface entropy effect in deducing $\alpha$ and $C_{sym}$. To this end, in Fig.2 we demonstrate the calculations for two cases: neglecting the surface entropy effect (denoted by open symbols) and including the surface entropy effect (represented by solid symbols). As can be seen, the inclusion of the surface entropy effect increase the $\alpha$ value, and the increase becomes larger with increasing excitation energy. Take the situation computed for the source pair ($A_2 = 185, A_1 = 165$) as an example. At $E_{tot}^*/A = 2.5$ MeV/nucleon the gap between solid squares and open ones, i.e. the difference of $C_{sym}$ caused by the surface entropy effect, is 3.14 MeV, and the gap rises up to 4.33 MeV at
\( E_{\text{tot}}^*/A = 4.0 \text{ MeV/nucleon} \) and it further amounts to 4.68 MeV at excitation energy of 7.0 MeV/nucleon. These numerical values clearly illustrate that the surface entropy effects on the symmetry energy become stronger at higher energy, a conclusion that is unrelated to the choice of the source pair (see left panel in Fig.2). Because the nuclear density is lowered at large excitation energy, the results derived above actually show that a smaller nuclear density can enhance the effect of the surface entropy on the symmetry energy. A physical understanding for this point is that at high energy the surface of a hot nucleus softens, and this leads to a larger role of the nuclear surface entropy in the fragment emission. The second feature is not varied for another pair source \((A_2 = 175, A_1 = 165)\).

In a recent work, it is found that the differences of the symmetry energy extracted at a fixed normal density but at different excitation energies (i.e. at different temperatures) are extremely small \([32]\). It implies that the symmetry energy has no dependence on the temperature. This conclusion has also been reached in several works which use different models, e.g. \([26, 43]\). Therefore, the significant variation of the symmetry energy with excitation energy revealed in Fig.2 is due to the variation of the density, although the variation of excitation energy can change the temperature and the density of the system simultaneously. Because of this reason, we depict in Fig.3 the symmetry energy as a function of density computed for four source pairs \((A_2 = 175, A_1 = 165)\), \((A_2 = 185, A_1 = 165)\), \((A_2 = 195, A_1 = 185)\) and \((A_2 = 195, A_1 = 175)\). The density is calculated by Eq.(8).

Considering the generally used form for density-dependent symmetry energy, namely

\[
C_{\text{sym}}(\rho) = C_0 (\rho/\rho_0)^\gamma. \tag{18}
\]

We use Eq.(18) to fit \(C_{\text{sym}}(\rho) \) vs. \(\rho/\rho_0\) as demonstrated in Fig.3. The lines in the Fig.3 are the fit. They give \(C_0 \sim 22.0, \gamma \sim 0.71\) (for the case neglecting the surface entropy effect) and \(C_0 \sim 25.2, \gamma \sim 0.46\) (including the surface entropy effect). A direct comparison for the two specific density-dependent form, 22.0 \((\rho/\rho_0)^{0.71}\) and 25.2 \((\rho/\rho_0)^{0.46}\), points out that the magnitudes of the latter form at various densities are always greater than the former form. In other words, the surface entropy effects make the nuclear equation of state become stiff. Furthermore, it is interesting to compare the present results with other independent work where various models and different experimental observables were utilized to deduce the density dependence. Such a comparison is plotted in Fig.4. One can see that a general trend of the symmetry energy with density is similar, that is, it is a rising function with
the increment of the density. However, the predictions for the magnitude of the symmetry energy at various densities have evident differences. We notice that compared to other studies the shape of the density dependence of the symmetry energy and its magnitude in the HIFG model are more close to that reported by Tsang et al. [18], who derived the density dependence by analyzing the data involving deep inelastic collision, evaporation and multifragmentation reactions, and by performing a simulation with the expanding evaporation source model [35]. Interestingly, Tsang’s result [18] is just between our ones (see lines tagged as numbers 7, 8, 9 in Fig.4). Although the mechanism for nuclear fragmentation assumed in this study [30, 31] and that in [35] is completely different, the similarities of the function form $C_{sym}(\rho)$ obtained in these two works could come from two aspects. One reason is that the observable analyzed and the technique used to gain the information of the symmetry energy are basically the same. Another reason is that a similar nuclear expansion model [35] was employed to simulate the expansion process of a highly excited nucleus.

IV. CONCLUSIONS AND OUTLOOK

In conclusion, based on a harmonic interaction Fermi gas model which includes the nuclear expansion, we study the effect of a hot nucleus expansion on the symmetry energy. Calculations show that the isoscaling behavior is not affected qualitatively by the nuclear expansion. In addition, the experimentally observed decreases of both the isoscaling parameter $\alpha$ and the symmetry energy at high excitation energies are predicted theoretically in the HIFG model. Furthermore, we deduce the specific form of the density dependence of the symmetry energy under the condition that whether the surface entropy effect on the fragment emission is included or not. It is demonstrated that the surface entropy influences the density dependence of the symmetry energy and increase the magnitude of the symmetry energy.

It should be noted that within an approach, suggested by Sobotka et al. [49], that combines the physical transparent picture of maximal-entropy mononuclear configurations found in the work by Töke et al. [31] with the effective-mass change with excitation energy found by Natowitz et al. [50], it was shown that the combined influence of expansion and in-medium effects flattens the curve of temperature verse excitation energy. This finding
indicates that besides nuclear expansion, the in-medium effect also influences the relationship between temperature and excitation energy of a hot nucleus system. Therefore, it is valuable in the future to survey the effect arising from the effective-mass change with excitation energy on the density dependence of the symmetry energy in the framework of the HIFG model.

ACKNOWLEDGMENTS

This work is supported by the U.S. Department of Energy Grant No. DE-FG02-88ER40414. The work of W.Y is also partially supported by the NSFC under Grant No. 10405007 and China scholarship council. W.Y is also grateful to Rochester University for support and hospitality extended to him.


FIG. 1: (Color online) The logarithm of the yield ratios of elements $Z = 5$-
9 isotopes for the case of not including surface entropy effect (top panel),
and for the case of including surface entropy effect via inclusion of $a_S$ in the
expression for the level density parameter $a$ [see Eq.(4)] (bottom panel) from
pairing source $A_s = 175$ and $A_s = 165$ (left column) as well as from pairing
source $A_s = 185$ and $A_s = 165$ (right column) at excitation energy $E_{tot}^* / A =
5$ MeV/nucleon. Solid lines are the linear fitting to the data points.
FIG. 2: Dependence of isoscaling parameters $\alpha$ and symmetry energy $C_{\text{sym}}$ on the excitation energy for source pair $A_s = 175$ and $A_s = 165$ (left column) as well as for pairing source $A_s = 185$ and $A_s = 165$ (right column). Solid and open symbols correspond to the case of including and not including surface entropy effect, respectively.
FIG. 3: (Color online) Density dependence of the symmetry energy calculated for four source pairs for (a) not including the surface entropy effect, and (b) including the surface entropy effect via inclusion of $a_S$ in the expression for the level density parameter $a$ [see Eq.(4)]. Solid lines denote the fit. See text for details.
FIG. 4: (Color online) Comparison of the density dependence of the symmetry energy obtained in this study and other independent work. Here tags [1]-[6], [8] and [10] correspond to the results reported in Ref.11, Ref.44, Ref.9, Ref.45, Ref.46, Ref.47, Ref.18, and Ref.48, respectively. Tags [7] and [9] represent our results.
Isospin Effects in Heavy-Ion Collisions: Some Results From CHIMERA Experiments At LNS And Prospects With Radioactive Beams

G.Cardella\textsuperscript{a}, F.Amorini\textsuperscript{b,c}, A.Anzalone\textsuperscript{b}, L.Auditore\textsuperscript{d}, R.Barn\textsuperscript{a},
A.Benisz\textsuperscript{c}, I.Berceanu\textsuperscript{g}, M.B.Chatterjee\textsuperscript{e}, S.Cavallaro\textsuperscript{b,c}, E.De Filippo\textsuperscript{a}, U.Emanuele\textsuperscript{d}, W.Gawlikowicz\textsuperscript{h}, L.Grassi\textsuperscript{a,c}, G.Giuliani\textsuperscript{c},
F.Giustolisi\textsuperscript{b,c}, A.Grzeszcuk\textsuperscript{e}, P.Guazzoni\textsuperscript{i}, S.Kowalski\textsuperscript{i}, E.La Guidara\textsuperscript{a,c},
I.Lombardo\textsuperscript{b,c}, G.Lanzalone\textsuperscript{a,c}, D.Loria\textsuperscript{d}, C.Maiolino\textsuperscript{b}, N.G.Nicolis\textsuperscript{l},
A.Pagano\textsuperscript{a}, M.Papa\textsuperscript{a}, I.Pawelczak\textsuperscript{m}, M.Petrovici\textsuperscript{g}, E.Piasecki\textsuperscript{h,q},
S.Pirrone\textsuperscript{a}, R.Planeta\textsuperscript{b}, G.Poli\textsuperscript{a,c}, A.Pop\textsuperscript{g}, F.Porto\textsuperscript{b,c}, F.Previdi\textsuperscript{i},
M.Quinlan\textsuperscript{m}, F.Rizzo\textsuperscript{b,c}, E.Rosato\textsuperscript{n}, P.Russotto\textsuperscript{b,c}, W.U.Schröder\textsuperscript{m},
I.Skwira-Chalot\textsuperscript{o}, K.Siemenska\textsuperscript{a}, K.Schmidt\textsuperscript{t}, A.Sochocka\textsuperscript{p},
L.Swiderski\textsuperscript{q}, A.Trifir\textsuperscript{d}, M.Trimarchi\textsuperscript{d}, J.Töke\textsuperscript{m}, G.Verde\textsuperscript{a}, M.Vigilante\textsuperscript{n},
J.Wilczynski\textsuperscript{q}, L.Zetta\textsuperscript{i} and W.Zipper\textsuperscript{e}

\textsuperscript{a} INFN, Sez di Catania, Via S.Sofia 64 - 95123 Catania Italy
\textsuperscript{b} INFN Lab. Naz. del Sud, Via S.Sofia 44 - 95123 Catania Italy
\textsuperscript{c} Dep. of Phys. and Astr. Univ. di Catania Via S.Sofia 64 - 95123 Catania Italy
\textsuperscript{d} INFN & Dep. of Phys. Univ. di Messina, Italy
\textsuperscript{e} Inst. of Phys., Univ. of Silesia, Katowice, Poland,
\textsuperscript{f} Saha Inst. of Nucl. Phys., Kolkata, India
\textsuperscript{g} Inst. for Phys. and Nucl. Eng., Bucharest, Romania
\textsuperscript{h} Heavy Ion Lab. Warsaw, Poland
\textsuperscript{i} INFN & Dip.Fis.Milano, Italy
\textsuperscript{j} Dep. Phys. Univ. Ioannina Grece
\textsuperscript{k} Dep.of Chem. Univ.Rochester USA
\textsuperscript{l} INFN & Dep. of Phys. Univ. Napoli, Italy
\textsuperscript{m} Inst.Exp. Phys. Warsaw Poland
\textsuperscript{n} Inst.of Phys. Jagiellonian Univ. Krakow Poland
\textsuperscript{o} A.Soltan Inst. Nucl.Phys. Swierk/Warsaw Poland

Abstract. CHIMERA is a 4\pi multi-detector array for charged particles operating at LNS Catania. A new method to measure the timescale for the fragment emission is described, together with some observations on the isospin of fragments emitted at different stages of a reaction. Competition between fusion-like and binary reactions near the threshold for nuclear multifragmentation is also discussed. Opportunities are pointed out to use the detector at low and intermediate energies using the kinematical-coincidence method.

Keywords: time scale, isospin, kinematical coincidence, radioactive beams

PACS: 21.65.Ef, 25.70.-z
INTRODUCTION AND TIME SCALE MEASUREMENTS

The CHIMERA $4\pi$ multi-detector has been operational at LNS Catania since 2000[1]. It consists of 1196 two-stage detector telescopes, each with a front 300-$\mu$m thick silicon detector followed by a CsI(Tl) scintillation detector with photodiode readout. Figure 1 is a photograph of CHIMERA in its new scattering chamber.

![Figure 1](image1.png)

**Figure 1:** The CHIMERA multi-detector array in its new scattering chamber.

The very low identification thresholds obtained allow one to perform accurate measurements also of target-like fragments emitted in heavy ion intermediate energy reactions. Thanks to this feature one was able to utilize [6] a new method to measure the emission time of intermediate mass fragments (IMF) produced in heavy-ion reactions. This method consists in the measurement of the correlation between relative velocities of IMFs with respect to coincident target (TLF) and projectile (PLF) remnants.

These IMF-fragment velocities, each normalized to the respective Viola fission-like velocity ($V_{VIOLA}$), are plotted in Fig. 2 as contour diagrams, for different relative IMF-fragment emission angles (proximity angles). An IMF that is sequentially emitted by the projectile (PLF) or the target (TLF) remnant is expected to have approximately the Viola velocity and produce an event near the $\frac{V_{REL}}{V_{VIOLA}}=1$ velocity guide lines.

![Figure 2](image2.png)

**Figure 2a, c, d:** Contour plots of IMF-TLF vs. IMF-PLF fragment velocities emission time scale plots with different conditions (a, c, d) on the proximity angle. **2 b)** Plot of count rate vs. proximity angle (ref.7).
shown also in Fig. 2. On the other hand, if IMFs are emitted early, when projectile and target nuclei are still in close proximity, they are influenced by the combined Coulomb field of both PLF and TLF reaction partners. Such events should appear in a region far from these velocity guide lines.

Experimental examples of this kind of velocity correlation plot are displayed in Fig. 2 a, c, and d for the reaction $^{124}$Sn+$^{64}$Ni at 35 MeV/A. The corresponding proximity angles are depicted in panel b) of the figure. This observable is defined [7] as the angle between the PLF velocity vector and the IMF-TLF separation axis. As can be inferred from Fig. 2 (c and d), IMFs emitted early in the reaction are very well aligned ($\cos \theta_{\text{prox}}>0.8$) with PLF and TLF, while a broader angular distribution ($\cos \theta_{\text{prox}}<0.8$) is obtained for IMF fragments sequentially emitted by PLF or TLF (Fig. 2d).

Using this information on the emission time scale one can now study the isospin equilibration of reaction partners during the various stages of the reaction. It is very interesting to observe that fragments emitted at the very beginning of the reaction, i.e., fragments along the diagonal of the “time scale” plot of Fig. 2, are much also more neutron rich than IMFs emitted at later stages of the reaction. This is apparently true not only for neutron rich systems but also for the neutron poor reactions, as demonstrated in Fig. 3 displaying results for the $^{112}$Sn+$^{58}$Ni reaction at $E = 35$ MeV/A.

**COMPETITION BETWEEN REACTION MECHANISMS**

From the width and curvature of the beta-stable valley one notices immediately that the stability of a nuclear system depends on its isospin and mass, i.e., very heavy systems must be neutron rich to attain stability. It is very interesting to see whether this mass/isospin dependence changes with system excitation energy. This has motivated a search for heavy residues produced in the reactions $^{40}$Ca+$^{30,48}$Ca and $^{40}$Ca+$^{46}$Ti at

![Figure 3: Average N/Z values vs. IMF-fragment velocities measured for IMFs emitted from the Sn+Ni reaction as deduced from an analysis of IMF-fragment velocity correlations.](image-url)
E=25 MeV/A. In fact, the experiments revealed an influence of the isospin on the competition between reaction mechanisms producing at least one large fragment in the final stage of the reaction.

These two mechanisms lead to distinct differences in the experimental distributions of normalized mass differences, \( \Delta M_{\text{nor}} = (m_1 - m_2)/m_{\text{tot}} \), between the two largest detected fragments with masses \( m_1 \) and \( m_2 \), respectively. These \( \Delta M_{\text{nor}} \) distributions are compared in Figs. 4a and 4b for the above three reactions. Obviously, events with large values of \( \Delta M_{\text{nor}} \) are most likely produced with the neutron rich \( ^{48}\text{Ca} \) target, while smaller differences between the two largest fragments are observed for the \( N \approx Z \) targets \( ^{46}\text{Ti} \) and \( ^{40}\text{Ca} \). Such results are predicted by CoMDII [8] calculations assuming a stiff dependence of the symmetry term of the nuclear matter equation of state [9].

**PROSPECTS FOR RADIOACTIVE BEAM EXPERIMENTS**

At LNS Catania low-energy radioactive ion beams (RIBs) are produced by the EXCYT facility [10], while the in-flight fragmentation technique [11] is used to provide intermediate-energy RIBs. For the near future extension of experimental investigations are planned for both types of RIBs. During the past two years transmission test expe-
Experiments were performed for fragmentation beams. Positive transmission test results for the beam line in the new CHIMERA hall provided motivation for a relocation of the CHIMERA detector from the old CICLOPE chamber to its intended place in the new hall. In Fig. 5 an example is shown of an identified fragmentation cocktail beams transported up to the new CHIMERA chamber. The beam was produced using a primary $^{20}$Ne beam of 45 MeV/A impinging on a $^9$Be production target. The best transmission results were obtained by configuring the standard beam line magnets to simulate a fragment separator. In the case illustrated in Fig. 5 the fields were set to maximize the transmission for $^{18}$Ne.

A proposal to use this new $^{18}$Ne beam has been submitted to the LNS PAC, with the intent to investigate the isospin dependent production of heavy residues mentioned previously. In the experiment different beam-target combinations will be studied, including neutron rich and neutron poor systems with equal total mass. Another proposal to use fragmentation RIBs produced with a primary $^{13}$C beam has also been submitted, whose goal is a study of nuclear structure in the $^{11}$Be region.

This latter proposal emphasizes the structure of light nuclei, a theme that will be pursued also with EXCYT beams. In particular, a proposal has been submitted to use a $^9$Li beam for the study of the $^{10}$Li resonance excited in the reaction $^9$Li(d, p). These nuclear structure measurements will benefit from the $4\pi$ angular acceptance of the CHIMERA detector providing a 100% efficiency for kinematical-coincidence measurements. Using such technique provides tools for efficient rejection of background processes. Furthermore, by requirements imposed on energy and on the detection angle of the second reaction partners, the device provides an improved effective angular resolution. Of course, the high detector efficiency also aides in obtaining sufficient statistics in reactions induced by faint RIBs.

Figure 6 demonstrates the cleaning efficiency of the kinematical coincidence method is shown. The intense and broad energy spectrum of $^7$Li particles scattered off H (CH$_n$, polyethylene target) shown in this figure has been collected at $\theta = (4.1\pm0.5)\degree$ (CHIMERA Ring 2E). Two other spectra are plotted for comparison as dashed histograms, illustrating the rejection of a large background produced by the requirement of a kinematical $^7$Li coincidence with a proton detected at a mean angles of $<\theta$> = 10.8° or 13.8° (CHIMERA Rings 5I and 6I, resp.).

Figure 6: Singles and coincidence spectra of particles produced in the reaction $^7$Li (CH$_n$, p).
OUTLOOK AND CONCLUSIONS

For the near future it is planned to move CHIMERA to other laboratories for certain experimental campaigns that utilize regular and secondary beams of very different energies. For example, a combination of CHIMERA sub-components with the LAND neutron detector [12] at GSI could be used to measure the differential neutron-proton flow. Such measurements have been predicted to be very sensitive to the symmetry energy term of the nuclear matter EOS at high density and can help to complete the exploration of the density dependence of this term, which this group has already pursued at lower beam energies. Other studies are underway exploring the use of CHIMERA at SPES, the new radioactive beam facility that is planned for LNL[13]. Also in this case the nearly complete angular coverage and the high efficiency of the CHIMERA detector should provide the high quality data needed to extend current knowledge of nuclear reactions and nuclear structure into new domains.

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Abstract: A qualitative study of intermediate-mass fragment (IMF) production was performed using asymmetric heavy-ion reactions of $^{40}\text{Ca} + ^{112}\text{Sn}$ and $^{48}\text{Ca} + ^{112,124}\text{Sn}$ at 45 A MeV. Projectile-like fragments (PLF) were measured in coincidence with IMFs and light-charged particles (LCP) in an exclusive study with the 4π multi-detector array CHIMERA. A significant fraction of the measured events show production characteristics consistent with a dynamical formation mechanism. This assignment is made on the basis of the measured relative velocity between the fragments as well as the asymmetry of their decay patterns. The decay pattern is shown to be sensitive to variables canonically associated with the collision centrality.

Introduction:
At intermediate bombarding energies (25-100 AMeV) collisions of heavy ions have been found to produce an unexpectedly high yield of fragments with atomic numbers (3 ≤ Z ≤ 10) termed intermediate-mass fragments (IMFs). These IMFs join other reaction products, principally the projectile-like (PLF) and target-like fragments (TLF), so named for their similarity to the original beam and target nuclei. Neutrons and light-charged particles (LCP) make up the remainder of the reaction products.

The high probability of producing these IMFs has been difficult to explain by extrapolating predictions of models originally developed for a lower bombarding energy range extending from near the Coulomb barrier up to 10AMeV [1]. These models rely on the assumption that nuclei are equilibrated and decay with little regard for their history of formation; rather the probability of decay is dependent, in a statistical fashion on the total available phase space. Diversions from equilibrium have been noted in heavy-ion collision dynamics already at bombarding energies as low as 7 AMeV [7]. At intermediate bombarding energies one might expect more substantial diversions, due to the decreased interaction time between projectile and target, more substantial deformations and compression, and the increased time necessary for thermal relaxation. In addition, the higher beam energies may open previously inaccessible phase space. The interacting projectile and target nuclei could form a transient three body system comprised of an excited projectile-like fragment (PLF*) connected to an excited target-like fragment (TLF*) by a neck. The properties of this neck region, as well as the overall dynamics of the interacting system, are the subject of active interest in the field. If IMFs are produced in the disassembly of

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1 One must point out that lower energy reaction models do not assume equilibration of all degrees of freedom, but often suppose that the un-equilibrated, collective modes are weakly coupled to those in equilibrium. In the case of weak coupling to a heat bath the dynamics of these non-equilibrium modes can be modeled by a transport process.
such a composite system, IMF kinematics may maintain a memory of the reaction entrance channel and thus be characterized as dependent on the collision dynamics.

With regard to this dynamic IMF production, mechanisms have been proposed for peripheral and mid-peripheral collisions. Two of the proposed models are neck emission and multiple neck rupture[8]. The neck emission model emphasizes the strong Coulomb forces acting on particles’ emitted from a low density neck connecting PLF* and TLF*. The strong forces focus the emitted fragments (LCP and IMF) into the space perpendicular to the neck axis akin to observations of ternary fission[9,10]. The multiple neck rupture model, sometimes termed dynamical fission, emphasizes the importance of a dynamic nuclear surface instability to explain the aligned break-up of a highly deformed di-nuclear system[11-14].

A complete description of IMF formation in heavy-ion collisions requires an understanding of the relative importance of all production mechanisms, both dynamical and statistical. A consistent picture will elucidate simultaneously many properties of nuclear matter in extreme conditions. Properties such as the nuclear level densities and isospin dependent nuclear equation of state (EOS) are likely responsible for the particulars of the IMF production mechanisms and the overall yield produced in nuclear reactions[15]. A characterization and estimation of the relative importance of different IMF production mechanisms can be accomplished by exploiting the kinematical relationships unique to different formation modes and the particulars of heavy-ion collision.

To this end a recent study of asymmetric heavy-ion collisions of $^{40}$Ca + $^{112}$Sn and $^{48}$Ca + $^{112,124}$Sn at 45 AMeV started with the characterization of binary collision events, followed by a study of the disintegration of the PLF*. The chosen reactions maximize the difference in neutron to proton asymmetry in the entrance channel and thus provide the highest sensitivity to effects dependent on the (EOS). Analysis of results of this dependence is ongoing and will be published later. In the present work, certain classes of events were identified. These events show that kinematical symmetries, expected from equilibrium decay of a PLF* (Ca like primary reaction product) into PLF and IMF, are absent. Specifically the IMF-PLF relative velocity is observed to be larger than that expected from statistical fission-like binary decay. In addition, the orientation of the PLF* break-up is found to be correlated with properties of to the reaction entrance channel.

The following experimental section reviews important aspects of the CHIMERA multi-detector used in the present experiment. Subsequent sections report the results of the aforementioned analysis and their tentative interpretation. In all cases, the assumptions used to identify the sources of IMFs are presented in the context of global reaction characteristics.

**Experimental:**

Heavy-ion beams of $^{40,48}$Ca (45 AMeV) were provided by the superconducting K800 cyclotron of the Laboratori Nazionali del Sud (LNS), Catania. Beam currents of $10^8$pps for $^{40}$Ca and $10^7$pps for $^{48}$Ca were bunched with a repetition rate of 120ns. The projectiles impinged on self supporting targets of $^{112,124}$Sn with thicknesses 627 and 689 $\mu$g/cm$^2$, respectively. The targets were placed in the CHIMERA multi-detector array in
the Ciclope scattering chamber at the LNS. A schematic diagram of the detector array is shown in Fig. 1 [16].

![Schematic illustration of the CHIMERA multi-detector](image)

**Fig. 1: Schematic illustration of the CHIMERA multi-detector [16].**

The array consists of 1192 Si-CsI(Tl) telescope detectors. The silicon energy response, CsI(Tl) light output and particle time-of-flight were measured. The array has a high atomic number resolution ΔZ=1 for charged particles (1≤Z≤21) coupled with 4π coverage and high granularity in forward laboratory angles (Θ<30°). A representative sample of the raw data showing the atomic number resolution is shown in Fig. 2.

The aim of the experiment was to perform an exclusive study of IMF production in an intermediate-energy heavy-ion collision. Therefore, the CHIMERA array used a minimum-bias trigger condition requiring the detection of at least two charged particles. Specifically, the acquisition electronics were triggered when two silicon detectors measured a charged particle within a coincidence time of 250ns.

All analog detector signals were digitized with multi-channel charge-to-digital converters (QDC) which were enabled with a common gate signal. The time-of-flight of charged particles was also measured and digitally recorded using time-to-digital converters (TDC). Of note in the analysis is the possibility that reaction/elastic-scattering event(s) from a subsequent beam burst are included by the large (>3μs) QDC gate. These events are the main source of background. However, in the data presented here, these pile-up events are easily recognized and rejected by the reader.
Identification of charged particle atomic and mass numbers was accomplished in the off-line analysis. Regions corresponding to different elements (cf. Fig. 2) were identified and stored digitally offline for all working detectors. Similar 2-dimensional correlations of CsI(Tl) detector signals allowed for the atomic and mass number identification of LCPs[17]. The measured multiplicity of light-charged-particles (MLCP) for all events using the aforementioned identification technique is shown in Fig. 3. In the present experiment the CHIMERA detector was 70% efficient in identifying LCPs with energy greater than 5 AMeV. The energy response of the silicon and CsI(Tl) detectors were calibrated using charged ions of known kinetic energy in dedicated experiments. The kinetic energy of ions with atomic number (Z>2) were inferred using the measured energy deposited in silicon, the detector thickness, and standard energy-loss tables[18]. The detector array and acquisition electronics are also described in more detail elsewhere[19].
General Reaction Characteristics:

A broad overview of the general reaction mechanism is important to any analysis attempting to establish the relative importance of different IMF formation mechanisms. A general binary-dissipative signature is established for peripheral to mid-peripheral collisions by the data presented in Fig. 4. The events presented were selected and defined as PLFs on the basis of their atomic number ($Z \geq 10$).

The data in the figure show a high intensity grouping at small angles and approximately 2200 MeV kinetic energy which corresponds to elastic scattering. These events are due to event pile-up explained in the previous section. The elastic intensity significantly decreases as one approaches the quarter point angle. The probability for such event pile-up is low, but sufficiently high to produce a “ghost image” of elastic scattering events. For events with higher inelasticity the pile-up is, of course, less noticeable and within experimental errors. Therefore, considering a background level given by the elastic pile-up one can safely extract characteristics of inelastic reaction events.

A second band with lower intensity but larger width extends from small angles at 1500 MeV to larger angles and lower energies. The events from this second band are expected to have suffered mid-peripheral to mid-central collisions in which the beam-like fragments’ trajectory has significantly deviated from that expected from only Coulomb forces acting between the projectile and target.
Fig. 4: Logarithmic contour diagram of the double-differential cross section of PLFs (Z≥10) plotted vs. their laboratory kinetic energy and laboratory scattering angle. The data were measured in the reaction 48Ca+124Sn. Symbols connected by curves represent the mean predictions of simulations. Open squares represent the results of CLAT simulation (see text). Solid circles represent the results of the CLAT simulation modified by sequential statistical decay modeled by GEMINI (see text).

The data are compared to semi-classical simulations of the initial projectile and target interaction. These simulations approximate the adiabatic response of projectile and target along their classical trajectory (CLAT)[20]. The kinetic energy of relative motion is transferred to internal (thermal) degrees of freedom through the mechanism of nucleon exchange (NEM)[21]. The rate of kinetic energy dissipation is given by the one-body window and wall formulas[17,18]. The latter is parameterized in terms the size of a neck connecting projectile and target primaries. In the simulation, larger necks are formed in more central collisions for geometrical reasons. Larger necks allow more nucleon exchange. Thus the integration of the equations of motion for more central collisions will result in trajectories which deviate from Rutherford scattering due to a stronger nuclear attraction, and more dissipation of the relative kinetic energy. The simulations make predictions for the properties of the binary reaction products, namely their mean kinetic and excitation energies, their mean elemental and isotopic identities, as well as their asymptotic scattering angles and internal angular momenta.

The results of the dynamical simulations for PLFs were used as input for the statistical decay code GEMINI++[2]. The code simulates the evaporation of light-charged particles (LCPs) according to the Hauser- Feshbach formalism[24]. IMF formation is modeled in the statistical formalism by weighting with the density of states at the conditional asymmetric-fission saddle-point for each of the energetically open decay channels. The spin-dependent conditional asymmetric-fission barriers are those calculated in GEMINI++ by modifying the Sierk barriers [25] to allow an additional
isotopic degree of freedom and assuming a saddle shape corresponding to the nascent fragments separated by distance of \((d=2 \text{ fm})\) [26].

The two-step simulation combining the projectile-target interaction with the sequential decay is capable of approximating the general trend of the data seen in Fig. 4. Comparison of these model predictions with the data suggests that the CHIMERA multi-detector triggers efficiently on mid-peripheral collisions. A calculation using the two-step reaction model described above and a software filter designed to mimic the CHIMERA thresholds and detector response, suggests a triggering efficiency greater than 50\% for collisions with impact parameter of at least \((b=9 \text{ fm})\). The choice of projectile sizes \((Z_{\text{PLF}} \geq 10)\), although informed by expectations for a PLF fission-fragment mass distribution, is arbitrary and necessary to the stated degree of agreement.

A discrepancy between predictions of the two-step sequential reaction model and the data occurs for the joint probability distribution of PLF laboratory velocity and scattering angle. Such a comparison is made in Fig. 5. The two-step reaction model clearly overestimates the degree to which the kinetic energy of relative motion is transferred into internal degrees of freedom. The observed agreement between the model and data in Fig. 4 can be explained in an examination of the complete derivative of kinetic energy with respect to mass and velocity (see Eq. 1),

\[
dT = m\partial v + \frac{v^2}{2}\partial m
\]

Fig. 5: Logarithmic contour of the double-differential cross section of PLF laboratory velocity and scattering angle. The data were measured in the reaction of \(^{48}\text{Ca} + ^{124}\text{Sn}\). Closed circles illustrate the predictions of the two-step reaction model.

Apparently, the model overestimates the strength of nuclear friction and therefore the excitation energy of the reaction primaries, PLF* and TLF*, a discrepancy obscured by
statistical emission. These observations are consistent with a reaction model in which dynamical emission of massive particles is responsible for the observed change in PLF kinetic energy. Such a dynamical model skirts the requirement for high nuclear temperatures (and large changes in PLF velocity) by exciting the IMF decay mode directly rather than through a statistical process. Incomplete kinetic energy damping has been observed earlier in reactions of $^{136}$Xe+$^{209}$Bi at 28 AMeV[27] and $^{86}$Kr+$^{197}$Au at 35 AMeV[28]. Dynamical IMF production was previously suggested as a new mode of kinetic energy dissipation by Tőke, et.al.[29].

**IMF Emission - General Characteristics:**

To ascertain and de-convolute multiple sources of IMF emission, a general study of PLF emission characteristics was undertaken. The vector components of charged particles detected by the CHIMERA array are plotted in Figs. 6-12 for different ranges in fragment atomic number. These figures represent the Galilei-invariant cross section

$$\frac{d^2\sigma}{d^3v}$$

in velocity (v) space plotted with respect to the velocity components parallel ($v_\parallel$) and perpendicular ($v_\perp$) to the beam axis. In some of the figures, the effects of the identification thresholds for low energy IMFs are visible.

![Invariant cross section for charged particles](image)

**Fig. 6** Invariant cross section for charged particles with atomic numbers ($18 \leq Z \leq 20$) plotted in the overall reaction center-of-mass reference frame. Data are taken from the reaction of $^{48}$Ca+$^{124}$Sn. The two vertical lines denote the velocity of the beam and the nucleon-nucleon center-of-mass.
Fig. 7: Same as Fig. 6 but for $(15 \leq Z \leq 17)$.

Fig. 8: Same as Fig. 6 but for $(12 \leq Z \leq 14)$. The cutoff at lower $v_{||}$ is due to the identification thresholds for these charged particles.
Fig. 9: Same as Fig. 6 but for $(9 \leq Z \leq 11)$. The cutoff at lower $v_\parallel$ is due to the identification thresholds for these charged particles.

Fig. 10: Same as Fig. 6 but for $(6 \leq Z \leq 8)$. The cutoff at lower $v_\parallel$ is due to the identification thresholds for these charged particles.
Fig. 11: Same as Fig. 6 but for (3≤Z≤5). The cutoff at lower $v_\parallel$ is due to the identification thresholds for these charged particles.

Through examination of the plots it is clear that the criterion (see discussion of the general reaction characteristics) of $Z_{plf} \geq 10$ should be relaxed. Fragments with atomic number $Z=6$ are prominently found with velocities close to the beam velocity $v_{cm}=6.5\text{cm/\text{ns}}$ (cf. Fig. 10). In addition, as the charge of the particles decreases (cf. Fig. 11) a second broader distribution of parallel velocities is visible. While the existence of fast fragments with $Z=6$ could result from statistical emission from a PLF* we provisionally assume in the following analysis, that the efficiency of the CHIMERA multi-detector is sufficient to effect a proper identification of PLFs based on their relative size alone. Therefore, the relationship between these two components is explored in further detail below.

**IMF PLF Atomic Number, and Angular Correlations**

To further explore the nature of IMF production in the reactions studied here, events were explored in which both an IMF and PLF were detected in coincidence. The general nature of two-body events, defined as the coincident detection of two charged particles with atomic numbers $Z \geq 3$, is illustrated in Fig. 12. Apparent from examination of the figure is the fact that the two-body events detected by CHIMERA are consistent with the break-up of the PLF into roughly two large pieces under the assumption that LCP emission is responsible for the lowering of the sum $Z_1+Z_2$. Clearly, a significant yield of larger fragments falls below the previous PLF assignment of $\geq 10$. In the following, this assignment is dropped and only the relative size of the two fragments $(Z_1,Z_2)$ is used to differentiate a PLF from an IMF. Considerable numbers of random
coincidences arising from the high beam current are also visible. They constitute the maximum background of the present experiment.

Fig. 12: Logarithmic contour plot of the coincident yield $\left( \frac{d^2\sigma}{dz_1 dz_2} \right)$ (arb. units) of two-body events plotted with respect to the atomic numbers of the two fragments. Data are taken from the reaction of $^{48}$Ca+$^{124}$Sn. The solid line denotes the expected region populated by the projectile splitting into only two pieces.

Fig. 13: Invariant cross-section for charged particles with the larger atomic number detected in two-body events plotted in the center-of-mass reference frame. Data are taken from the reaction of $^{48}$Ca+$^{124}$Sn.
Fig. 14: Invariant cross-section for charged particles with the smaller atomic number detected in two-body events plotted in the center-of-mass reference frame. Data are taken from the reaction of $^{48}\text{Ca} + ^{124}\text{Sn}$.

The invariant cross section for the larger fragments detected in these two-body events is shown in Fig. 13. A similar plot for the smaller fragments is shown in Fig. 14. Comparison of the two figures shows large relative (IMF-PLF) velocities are observed. This observation is difficult to reconcile with a reaction model suggesting all IMFs are produced in a purely sequential statistical decay of a well-defined PLF*. Therefore, further characterization of these events is explored below.

Statistical emission of light charged particles is isotropic in the non-rotating rest-frame of the emitter. The fission of an equilibrated, but rotating nucleus is isotropic in the plane perpendicular to the axis of rotation. Thus in principle, one can differentiate between the different IMF formation mechanisms by comparing the observed IMF-PLF correlations to expectations based on binary decay patterns. The two-body events described above were analyzed on an event-by-event basis to determine the symmetries present in the decay using the vector diagram depicted in Fig. 15. The results of this analysis are presented in Fig. 16. The distribution is clearly anisotropic and favors backward emission of IMFs in the PLF* frame. Possible sources of this anisotropy include a component of IMFs emitted from the TLF* of from a fusion-like compound system (CS*) but incorrectly attributed to PLF* emission, and the dynamical mechanisms described above.
Fig. 15: Definition of decay angle ($\Theta_{\text{decay}}$) from two-body event components.

Fig. 16: Angular distribution of the smaller fragment in the decaying PLF* reference frame defined by the 2-body event kinematics. Data are taken from the reaction of $^{48}$Ca+$^{124}$Sn and presented in the histogram. The smooth curve schematically illustrates expectations for sequential statistical decay of a PLF* with fixed laboratory velocity and excitation energy.

Without a detailed characterization of the TLF* or CS* temperature, quantitative subtraction of the contributions from the sequential statistical decay of such sources is not possible. One expects contributions of sequential emission from the TLF* to dominate other modes at laboratory angles in excess of 140° [30]. The current sample of detectors calibrated by the CHIMERA collaboration does not extend to these backward angles. At angles currently accessible, emission from the neck and dynamical fission are expected to effectively compete with statistical sources. Therefore, an arbitrarily defined angular
region is used to qualitatively explore the nature of the multiple neck rupture mechanism described above.

**Qualitative Nature of Aligned Fragmentation**

Absent quantitative predictions for both the quantity and quality of the possible sources for IMF production the authors have chosen to study the qualitative nature of the most prominent emission patterns clearly visible in Fig. 14. This emission shows a broad distribution of velocities parallel to the beam direction and narrow transversal components with slightly less than 2 cm/ns. The distribution appears to lie predominantly at velocities lower than that of the PLF and at small angles (cf. Fig. 13), suggesting that the IMFs are emitted behind the PLF, as the latter moves away from the target. Other authors have also reported such an aligned emission with a variety of systems, reaction kinematics, and detector geometries [8,11,31-34].

The dominant IMF component was isolated by considering 2-body events where both particles have laboratory scattering angles ($\Theta_{\text{lab}} < 22^\circ$). The results presented below subject to this constraint are qualitatively insensitive to a 20% change in the laboratory angle cutoff. The invariant cross section for the larger of the two-body event fragments subject to the constraint is plotted in Fig. 17, while the same observable for the smaller fragment is shown in Fig. 18. Comparison of Fig. 13 with Fig. 17 indicates that the constraint excludes events with small PLF atomic numbers, but includes the vast majority of measured events. Comparison of Fig. 14 with Fig. 18 indicates that a large number of IMFs detected at larger angles are excluded by the constraint, leaving those which are qualitatively consistent with aligned emission. Events which originate from IMFs that are emitted behind the PLF* are excluded more than those emitted in the forward direction (see discussion below for this distinction). This rather arbitrary constraint is designed to maximize the inclusion of events which show aligned emission, while minimizing contributions from sources consistent with a TLF*, CS* or neck emission.

The angular distribution subject to the constraint is plotted in Fig. 19. In comparison with Fig. 16 which showed a structured angular distribution the data now appear to have not more than two components. The first has symmetry consistent with statistical emission, while the second shows an anisotropy which favors backward (aligned) emission. Traditional approaches of deducing the relative yield of statistical and dynamical IMF emission assume that IMFs emitted towards forward angles in the PLF* rest frame result from sequential statistical decay. The relative yield is then deduced by extrapolating the yield in this region to more backward angles [32].

Further comparison of the IMF-PLF angular correlation with expectations based on sequential statistical decay is shown in Fig. 20. The data show IMF-PLF relative velocities significantly larger than expected from binary decay of a PLF* for backward emission angles. This observation is consistent with the dynamical production mechanism discussed above. This has been observed by others in other reactions [8,11,31-34]. However in our data, similarly large relative velocities are also observed at forward angles (cf. Fig. 21); though the yield for forward emission of IMFs is significantly less than that for backward emission. At a minimum this observation indicates that estimates of IMF production yields from equilibrated PLF*s based simply on cross sections at forward angles, and then extrapolated using symmetry arguments (cf.
Fig. 19) are an overestimate. These estimates include events which have unreasonably high decay energies.

Fig. 17: Invariant cross-section for charged particles with the larger atomic number detected in two-body events in which both fragments have $\Theta_{\text{lab}} < 22^\circ$ and plotted in the center-of-mass reference frame. Data are taken from the reaction of $^{48}\text{Ca} + ^{124}\text{Sn}$.

Fig. 18: Invariant cross-section for charged particles with the smaller atomic number detected in two-body events in which both fragments have $\Theta_{\text{lab}} < 22^\circ$ and plotted in the center-of-mass reference frame. Data are taken from the reaction of $^{48}\text{Ca} + ^{124}\text{Sn}$. 
Fig. 19: Angular distribution of the smaller fragment in the decaying PLF* reference frame defined by the 2-body event kinematics subject to a selection described in the text. Data are taken from the reaction of $^{48}$Ca+$^{124}$Sn and presented in the histogram. The smooth curve schematically illustrates our expectations of sequential statistical decay of a PLF* with fixed laboratory velocity and excitation energy.

Fig. 20: Cosine of the decay angle defined above versus the IMF-PLF relative velocity subtracting that expected from asymmetric binary decay. Events are defined by the 2-body event kinematics subject to the selection (see text). Data are taken from the reaction of $^{48}$Ca+$^{124}$Sn.
Evidence for Impact Parameter Dependence of Aligned Emission

This observation suggests that clearer model descriptions of the projectile-target interaction are necessary to disentangle the complex IMF emission patterns into components consistent with the mechanisms proposed in the literature. In support of this goal, characteristics of the dynamical forward emission of IMFs were compared to the properties of IMFs dynamically emitted in the backward direction. Specifically IMFs with \( \cos(\Theta_{\text{decay}}) > 0.6 \) are characterized as dynamically forward, while IMFs with \( \cos(\Theta_{\text{decay}}) < -0.6 \) are characterized as dynamically backward. This assignment assumes that the contributions of sequential statistical decay of a PLF* is negligible in these two regions.

Three experimental observables canonically considered sensitive to the projectile and target collision centrality are the \( M_{\text{LCP}} \), PLF atomic number and the PLF laboratory scattering angle[7]. Comparison of the average behavior of these observables for events differentiated between forward and backward emission as defined above is presented in Figs. 22-24. The measured average multiplicity of LCPs for events in which the IMF is emitted dynamically backward is \( \langle M_{\text{LCP}} \rangle \approx 5 \). The measured average multiplicity of LCPs for events in which the IMF is emitted dynamically forward is \( \langle M_{\text{LCP}} \rangle \approx 6 \). The data presented in Fig. 22 are consistent with the numbers reported above. Further simulation is required to ensure that the noted difference independent of any difference in the LCP detection efficiency for the two classes of events. The magnitude associated with the observed difference in \( M_{\text{LCP}} \) for the two groups of PLF* decays (forward vs. backward) is consistent with the incomplete identification of all LCPs produced in the reaction. Further study of other experimental observables proportional to the \( M_{\text{LCP}} \), but independent of the limitations of the identification procedure is ongoing. In summary,
the multiplicity of LCPs is seen to increase on average when the IMF is emitted dynamically forward which implies that these events could originate from more central collisions.

Fig. 22: LCP multiplicity distribution for events categorized as dynamically backward emission (black) and dynamically forward (red). Data are sampled from the reaction of $^{48}$Ca+$^{124}$Sn.

Fig. 23: PLF atomic number distribution for events categorized as dynamically backward emission (black) and dynamically forward (red). Data are sampled from the reaction of $^{48}$Ca+$^{124}$Sn.
Fig. 24: PLF angular distribution for events categorized as dynamically backward emission (black) and dynamically forward (red). Data are sampled from the reaction of $^{48}\text{Ca} + {^{124}\text{Sn}}$.

Likewise the decreased size and increasing deflection angle of the PLFs observed in events with fast, forward emitted IMFs suggest that these events could also result from more central collisions. In both distributions the second moments are also observed to depend on the orientation of the PLF* decay. The PLF atomic number distributions appear to have underlying structure. This effect is not well understood and is still under study. The broader angular distribution of PLFs for events with $\cos(\Theta_{\text{decay}}) > 0.6$ could result from increased evaporation of LCPs, due to the presumably higher excitations produced in more central collisions.

**Conclusions:**

The observation of high IMF-PLF relative velocities in presumed binary decays aligned with the center-of-mass motion (cf. Fig. 20) is difficult to explain with dynamics dependent on Coulomb repulsion alone. Under the assumption that the measured events correspond to the binary decay of an excited PLF, even the inclusion of the Coulomb forces produced by a TLF in close proximity to the decaying system fails to explain the majority of the data. This refinement would instead serve to decrease predictions of the IMF-PLF relative velocity for IMFs emitted behind the PLF.

More sophisticated modeling of the statistical decay pattern of excited PLF*'s produced in a heavy-ion collision is justified to ensure that the expected forward-backward symmetry remains after accounting for detector thresholds and the kinematical selections described herein. However, the large IMF-PLF relative velocities observed for IMFs emitted in both the backward and forward directions demand physical
interpretation and the observation of asymmetric decay patterns offer an important, if yet only tentative, clue.

A qualitative interpretation of the data is possible upon inclusion of strong damping (stopping) of the relative motion of only the overlapping surface matter of both projectile and target. Under this assumption the largely undamped regions not associated with projectile and target maintain their velocities (*participant-spectator model*). The resulting difference in damping, if sufficiently large could deform the projectile well past the fission saddle.

In such a model one expects the relative size, velocity and spatial orientation of the PLF and IMF to depend on the degree of initial overlap between projectile and target surface matter. The observed correlations between variables relating to degree of initial projectile/target overlap and the orientation of the break-up axis of the PLF* adds credence to such a qualitative hypothesis. The large IMF-PLF relative velocities suggest that the non-uniform damping of the projectile and target leads to significant deformation of the initially spherical nuclei. Highly deformed nuclei are significantly unstable to binary division. Thus this qualitative description of the reaction dynamics could offer a natural explanation of the large cross sections for IMF production cited in other studies.

However, a quantitative comparison of the yields for statistical, sequential emission and the mechanism(s) for dynamical production requires significant modeling of the complex reaction dynamics. This is because the decay patterns from multiple sources produced in the reaction are difficult to quantitatively disentangle experimentally. However, the conclusions of this study illustrate important qualitative features which should be reproduced by future reaction models.

These future reaction models should investigate the mechanisms for non-uniform damping self-consistently with other nuclear properties. State-of-the-art reaction models currently employ theoretical predictions for nucleon-nucleon collision cross sections derived independently, but used concurrently with mean field effective interactions[35]. While these reaction models have had moderate success in producing IMFs with intermediate velocities, they have had limited success in reproducing the commonly observed yields cited in the literature [36]. The models should also be tested with respect to the spatial and size dependencies reported herein. Also on the horizon is a proper investigation of the N/Z dependence of the fragments produced by the discussed mechanism(s).

In summary, the angular distributions of IMFs dynamically emitted from a PLF*, either as the latter re-separates from the target or soon afterwards, show a dependence on impact parameter. This has been established using three independent experimental observables. This result has implications for understanding the phase-space available to nuclei both during an intermediate-energy heavy-ion collision and once equilibrium has been established.

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Correlations between Reaction Product Yields as a Tool for Probing Heavy Ion Reaction Scenarios

W. Gawlikowicz¹,², D.K. Agnihotri¹, S.A. Baldwin¹, W.U. Schröder¹, J. Tóke¹, R.J. Charity³, D.G. Sarantites³, L.G. Sobotka³, R.T. deSouza⁴, T. Barczyk⁵, K. Grotowski⁵, S. Micek⁵, R. Planeta⁵, and Z. Sosin⁵

¹Departments of Chemistry and Physics, University of Rochester, Rochester, NY 14627, USA
²Heavy-Ion Laboratory, Warsaw University, Warsaw, Poland
³Department of Chemistry, Washington University, St.Louis, MO 63130, USA
⁴Departments of Chemistry, Indiana University, Bloomington, IN 47405
⁵Institute of Physics, Jagellonian University, 30-059 Kraków, Poland

Abstract

Experimental multidimensional joint distributions of neutrons and charged reaction products were analyzed for \(^{136}Xe + ^{209}Bi\) reactions at \(E/A = 28, 40,\) and \(62\) MeV, and were found to exhibit several different types of prominent correlation patterns. Some of these correlations have a simple explanation in terms of the system excitation energy and pose little challenge to most statistical decay theories. However, several other types of correlation patterns are difficult to reconcile with some, but not other possible reaction scenarios. In this respect notable are correlations between the average atomic number of intermediate-mass fragments, on the one hand, and light particle multiplicities, on the other hand. This kind of multi-particle correlations provides a useful tool for probing reactions scenarios, which is different from the traditional approach of interpreting inclusive yields of individual reaction products.

PACS numbers: 25.70.Pq, 25.70.Mn
I. INTRODUCTION

Over the last 20 years, or so, considerable effort has been made both, experimentally and theoretically, to explore 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 prevailing 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 convert an ever increasing (with time passing) part of the kinetic energy of relative motion into intrinsic thermal and rotational energies. Phenomena of energy and angular momentum dissipation and mass transfer are thought to be effected mainly by means of stochastic nucleon exchange between the 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, to decay statistically. Superimposed 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 the 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. For example, a notorious pattern is that seen in typical Wilczyński plots [1] of PLF yield as a function of PLF kinetic energy and deflection angle.

At higher bombarding energies, up to $E/A = 62$ MeV, the underlying general dissipative collision scenario still appears to be consistent with a variety of experimental observations made at energies close to the interaction barrier [2]. However, here a consensus is still lacking as to the dominant production mechanisms of the observed light and intermediate-mass reaction products. To some extent, this is so because of expectations of an increased role of two-body interactions and of increased preequilibrium emission,
possibly including emission from the dynamically unstable interfragment “neck-like” structure [3–5]. But more importantly, it is so because of the observation of copious production of intermediate-mass-fragments (IMFs), which exhibits certain aspects of statistical independence [6, 7] but appears difficult to reconcile with classical scenarios of a statistical decay of an equilibrated nuclear system. A number of unconventional statistical approaches have been developed [9, 10, 27] to address the shortcomings of traditional models of statistical particle emission when applied to IMFs. On the other hand, several experimental studies [11–15] have concluded a dominantly non-equilibrium mode of IMF production.

The failure to reach a consensus at these higher energies may well point to inherent limitations 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, a 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 $4\pi$ angular coverage using the Rochester Superball or RedBall neutron calorimeter/multiplicity meters in combination with one of the available $4\pi$ 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 $^{209}$Bi + $^{139}$Xe reactions at $E/A$=28, 40, and 62 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 PLFs 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 beams of $^{136}$Xe ions from the K1200 cyclotron, with energies of $E/A = 28$, 40, and 62 MeV were focussed on a self-supporting 3.5-$mg/cm^2$ thick $^{209}$Bi target placed in the operational center of the detector setup. The latter consisted of two $4\pi$ detector systems, the Washington University charged-particle detector array, DwarfBall/Wall [16] and one of the University of Rochester neutron calorimeter/multiplicity meters, SuperBall [17] or RedBall. Additionally, two position-sensitive silicon-detector telescopes were placed at forward angles, so as to cover an angular range encompassing the anticipated grazing angle ($\theta_{\text{Grazing}} = 4.48^\circ$, 3.90$^\circ$, and 2.91$^\circ$, for 28, 40, and 62 MeV/nucleon reactions, respectively). The Dwarf array provided for a reliable $Z$ identification for atomic numbers up to $Z=35$, but also for the energy and emission angle measurement [18]. 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. Since the mean detection efficiencies for the relatively weak components of high-energy preequilibrium neutrons and protons are much smaller [SuperBall,Djerroud] than for the less energetic, statistically emitted particles, the multiplicities measured for neutrons and LCPs reflect mainly the thermal excitation energies of the emitter nuclei.

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. In all cases, the "minimum bias" trigger for the data acquisition was provided by one charged particle registered by any of the detectors. While even elastic scattering of projectiles was measured in the experiments, a PLF coincidence was not required in the definition of the minimum bias trigger. Thus the experimental setup allowed one to obtain an almost complete characterization of individual reaction events in terms of product identification, their yields, and corresponding kinematical parameters.
III. THEORETICAL MODELING OF VARIOUS REACTION SCENARIOS

The ultimate goal of modeling of any particular reaction scenario is to obtain model predictions for experimentally measured patterns in reaction product yields, so the latter can be used to verify the consistency of, or to falsify the scenario under scrutiny. Given the complex nature of the processes leading from the heavy-ion collision to the detection of products, the modeling is in practice always a multi-step endeavor involving both, “fundamental” modeling of physical phenomena of interest and a more technical in nature modeling of the detection processes. Quite generally, the latter is free of hypotheses and includes modeling of the geometrical and electronic acceptance (thresholds, dynamical ranges) of the detector setup to reaction products, but may also include calculations of multi-particle Coulomb trajectories from the moment in time when the particles are set free in the fundamental theoretical models to the moment they reach their respective asymptotic trajectories.

In heavy-ion reactions at low and intermediate bombarding energies, the fundamental theoretical modeling itself is typically a two-step process, with the first step aiming at describing the collision dynamics and the second one aiming at describing the decay of the primary reaction products. Generally, the outcome of such simulation calculations depends on assumptions made in both steps, which tends to obscure an interpretation of possible discrepancies between the predictions and the experimental observations. Therefore, of special value are predictions for patterns that are uniquely sensitive to one of the two steps but not to the other.

In the present study, two theoretical codes were used alternately to model the dynamical, interaction stage of the collision, while two other codes were used alternately to model the subsequent statistical decay of the primary products emerging from the interaction stage with the dynamical model predictions for product mass and atomic numbers, excitation energies and spins. And so, the interaction stage was modeled using either the classical transport code CLAT [19], based on a stochastic nucleon exchange model NEM [22] or the Quantum Molecular Dynamics (QMD) code CHIMERA [23], accounting in a better detail for the two-body interactions. The version of the CHIMERA code used [23] included isospin dependent nuclear interactions, and calculations were performed
for the time interval from 0 to up to 300 fm/c assuming a soft EOS \((K \approx 200\text{MeV})\) with symmetry energy strength coefficient corresponding to an ASY-STIFF EOS \((C = 31.4\ \text{MeV})\) [24].

The statistical decay of the primary fragments predicted by the dynamical models of the interaction stage was then modeled using either the equilibrium-statistical, sequential decay code Gemini [25] or the (pseudo-microcanonical) statistical multifragmentation code SMM [27].

Note, that the physical scenario nominally implemented in the code CLAT may be not too an accurate representation of the interaction at higher bombarding energies. What matters in the present study, however, is that it provides still a surprisingly good effective parametrization of the interaction, and in particular of the dissipation function. The code SMM, on the other hand, as a matter of principle does not represent any realistic physical scenario, that can be supported by generally accepted theories of nuclear matter. Therefore, also this code is here viewed merely as providing a reasonably good effective parametrization of the statistical decay of excited nuclear systems.

A multi-particle Coulomb trajectory calculation routine [28] was applied to the end products predicted by the “standard” GEMINI code [25], making use of the (statistical) timing, spatial locations, decay axes, and relative energies of all (binary) decays along the deexcitation cascade. This kinematic “after-burner” allowed one to calculate energies and asymptotic emission angles of all reaction products.

The results of model calculations were subsequently passed through a routine calculating the response of the DwarfBall/Wall and SuperBall \(4\pi\) detector systems to each and every reaction product predicted by model calculations. The routine accounted for the geometrical acceptance and the detection efficiency of all detectors for various particle species. The efficiency of the SuperBall was calculated using a version of the well-known code DENIS [26]. This procedure of converting “generic” theoretical predictions into custom predictions for a given detection setup is often called “filtering”. The “filtered” model predictions, is what can be meaningfully compared to the experimental data.
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 [29] that the joint multiplicity distributions of neutrons and light charged 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 form of a (logarithmic) contour plot, while the respective distributions expected for different interaction/decay scenarios are shown in the other panels. As seen in the first panel in Fig. 1, the experimental 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 $m_n \approx 20$, 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 statistical 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 the nuclear 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.

A somewhat less conspicuous feature of the joint neutron-LCP multiplicity distribution is the presence of a “pass” in the ridge at around $(m_{LCP}, m_n) \approx (5, 28)$. Note that both terms, the “pass” and the “ridge” refer to a hypersurface in the three-dimensional space spanned on particle multiplicities and on the number of events. Mathematically, the “pass” is a saddle point on this hypersurface. As the distance along the ridge from the origin of the plot represents reasonably well excitation energy in the system, the presence and the location of the “pass” are reflections of a particular mix of dissipative and conservative forces acting between the collision partners. More specifically, they are
reflections of the form of dissipation function, i.e., of the impact-parameter dependence of the dissipated energy. Because of this, the location of the "pass" can be used to validate effective interactions used in collision codes.

Correspondingly, the ridge features a peak at high particle multiplicities around \((m_{LCP}, m_n) \approx (10, 35)\), the "central collision bump". The latter feature is common to particle multiplicity data measured in a variety of different reaction types \([20, 21]\) and interpreted in terms of the geometrical overlap of projectile and target nuclei or collision centrality.

The experimental crest line is shown in all panels of Fig. 1, liberally extrapolated beyond the high-multiplicity peak of the ridge. Note, that the ridge cannot be uniquely defined mathematically beyond that peak.

As seen in Fig. 1, all theoretical scenarios, represented here by combinations of model

![Logarithmic (base 2) contour plots of experimental (top left panel) and various model (remaining panels) joint distributions of neutron \((m_n)\) and light-charged particle \((m_{LCP})\) for \(^{136}Xe + ^{209}Bi\) reaction at \(E/A = 40\) MeV.](image)
codes “CLAT + GEMINI” (one-body interaction and sequential decay), “CLAT+SMM” (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 [19] and Gemini [25] provide for the best agreement with experimental observations, as far as the location of the crest line of the yield ridge and the location of the “pass” in the ridge are concerned. However, it still fails to reproduce the location of the peak at high particle multiplicities. In model calculations, this latter location depends on “dissipation function” describing the way in which degree of effected kinetic energy dissipation depends on the impact parameter. However, it is not clear whether the observed discrepancy between the actual and model locations of the high-multiplicity peak is due partially or wholly to a flawed modeling of the dissipation function by the code CLAT with its particular implementation of the stochastic nucleon exchange model, NEM. This is so because the CLAT + GEMINI combination neglects entirely the role of an effective intermediate-velocity source (IVS) which emits particles, and especially IMFs with higher average kinetic energies than the purely thermal energies characteristic of equilibrated PLF and TLF sources. Inclusion of preequilibrium emission and production of IMFs would result in shifting the peak in theoretical (CLAT + GEMINI) yield ridge in Fig. 1 towards lower particle multiplicities, bringing it to a better agreement with the experimental one.

As seen in the corresponding panel in Fig. 1, a combination of the codes CLAT and SMM predicts a saturation in neutron multiplicity not observed experimentally, such that the model calculations “misplace” the crest line itself. Here, not only the location of the high-multiplicity peak in the joint neutron and LCP multiplicity distribution, but also the location of the “pass” in the ridge disagree markedly with the experimental ones. Although these facts do not necessarily disqualify the SMM as a viable model, they point to specific model deficiencies in the description of relative neutron and LCP
yields and should help in devising corrections to the model that remedy this deficiency.

As seen in the bottom right panel in Fig. 1, the QMD + GEMINI calculations largely fail to reproduce the “topography” of the yield ridge such that not only they misplace the crest line, but also miss the “pass” in the ridge and the high-multiplicity peak altogether. Clearly, these model calculations over-predict the emission of neutrons compared with LCPs, which may be indicative of the neutron yield being strongly affected by dynamical emission during the interaction stage of the collision. If so, it would be a deficiency that, apparently, cannot be remedied by simple *ad hoc* corrections to the QMD code CHIMERA [23]. Therefore, one may conclude tentatively that at $E/A = 40$ MeV, the collision dynamics is still largely governed by one-body dynamics and that the effects of direct nucleon-nucleon interactions are still too weak 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.

In conclusion of this section, one must note that the “topography” of the yield distribution as a function of neutron and LCP multiplicities, which features prominently a “ridge”, a “pass” and a “peak” at high particle multiplicities may serve not only as a primitive tool of validating theoretical models but also as a more nuanced tool of actually revising and up-grading these models.

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

In recent years, prominent correlations between the average sizes of IMFs and the joint multiplicity of neutrons and protons have been discovered [2], which were found also to exhibit non-thermal scaling, such that they depend on the bombarding energy.

Such correlations are illustrated for three bombarding energies in Fig. 2, each in form of a contour diagram of the average atomic number $< Z_{IMF} >$ of IMFs plotted *versus* the associated neutron and LCP multiplicities. No bias has been imposed on the distribution of IMFs admitted in these plots other than $3 \leq Z_{IMF} \leq 10$. This segment of the product Z distribution is well separated from the domains of PLFs, their evaporation
residues and their fission fragments. As seen in these plots, the average size of the IMFs produced is correlated prominently with the joint multiplicity of neutrons and LCPs, which reflects the dissipated and "thermalized" energy in heavy-ion collisions [20]. Hence, larger IMF sizes are associated with higher excitation energies. Interestingly, higher excitation energies also lead to higher multiplicities $M_{IMF}$ of IMFs, which are mostly emitted [2] at excitation energies associated with the "central collision bump" in the joint neutron/LCP multiplicity distribution mentioned previously. The clear "equi-size" contour lines running almost perfectly parallel to each other appear to coincide with lines of constant excitation energy. Note that this kind of multi-particle correlation involves three independently measured quantities, neutron and LCP multiplicities and

FIG. 2: (Color online) Logarithmic contour plot of average atomic number of IMFs, $< Z_{IMF} >$, as a function of associated neutron and LCP multiplicities as observed in $^{136}Xe + ^{209}Bi$ reactions at $E/A = 28$ MeV (top panel), $E/A = 40$ MeV (middle panel), and $E/A = 62$ MeV (bottom panel). Here, $3 \leq Z_{IMF} \leq 16$. 
the atomic numbers of IMFs.

Importantly, as is clear from the comparison of the three panels in Fig. 2, the contour lines of equal $<Z_{IMF}>$ shift systematically toward higher neutron and LCP multiplicities as bombarding energy increases, which means that they do not scale directly with thermal excitation energy. This observation suggests that a significant portion of IMFs may be produced in dynamical processes. Indeed, this kind of scaling of the average size of IMFs is consistent with scaling according to the size of the overlap region between projectile and target or possibly the size of the neck-like structure formed transiently between the interacting PLF and TLF. Such a conclusion is based on the observation that for higher bombarding energies a comparatively smaller overlap region leads to the same given excitation energy than at lower bombarding energies, hence the shift of contour lines toward higher excitation energies but same overlap.

Results of attempts to reproduce the observed correlations at $E/A=40$ MeV by three reaction scenarios are illustrated in Fig. 3 along with the experimental distribution (left top panel). Model calculations included always the “experimental filter” accounting for the response of the detector setup. As seen in the second panel in the first row, the combined “CLAT + GEMINI” results only resemble the data in so far as the $m_n$m_{LCP} correlation is concerned, but misses the size correlation pattern, consistent with the presence of the dynamical component in IMF yield as discussed 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 left panel in the bottom row of Fig. 3 illustrates the failure of the statistical multifragmentation model SMM [27] to account for a prominent experimental correlation pattern. While based on the argument regarding the presence of a dynamical component in the IMF yield one would not expect a particularly good agreement with experiment in this case, the correlations predicted for the CLAT + SMM scenario are to a good extent “orthogonal” to those actually observed. This may be taken as indicative of a simultaneous statistical multi-particle breakup playing little, if any role in IMF production in the bombarding energy range considered and suggesting that the IMF production occurs, perhaps, with no meaningful competition from particle decay channels.

The right panel in the bottom row of Fig. 3 illustrates predictions by the QMD code
CHIMERA [23] complemented by code GEMINI and the Coulomb trajectory “afterburner” [28]. 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 to a large extent produced in dominantly dynamical processes, expected to be well described by QMD type of codes. Indeed, as seen in the respective panel in Fig. 3, CHIMERA is capable to correctly render the trends observed experimentally and, most notably, the

FIG. 3: (Color online) Logarithmic contour plots of average atomic number of IMFs, \(< Z_{IMF} >\) as a function of associated neutron and LCP multiplicities \(m_n\) and \(m_{LCP}\), as predicted by three sets of model calculations for \(E/A=40\) MeV (see text).
increase of average IMF size with increasing neutron and LCP multiplicities. In this respect, it would be highly desirable to compare CHIMERA predictions to correlations experimentally observed for a range of bombarding energies.

FIG. 4: (Color online) 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 \( E/A = 28 \text{ MeV} \) (top), 40 MeV (middle), and 62 MeV (bottom).
C. Correlations between the Size of PLF and the Joint Multiplicity of Neutrons and LCP

Figure 4 illustrates correlations between the average size of the PLF and the associated joint multiplicity of neutrons and LCPs, as observed at three bombarding energies of $E/A = 28$ MeV (top panel), 40 MeV (middle panel), and 62 MeV (bottom panel). As seen in Fig. 5, 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 modeled by a two-step $CLAT + GEMINI$ calculation. However, they can also be 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 is concerned. Yet the fact that they do not contradict conclusions reached further above is in itself encouraging.

V. SUMMARY

The present work 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 even at $E/A = 62MeV$ the reaction dynamics is still largely dominated by one-body interactions with lesser propensity to a preequilibrium release of particles and fragments than exhibited by a two-body interaction dominated scenario.

Concerning IMF production, a spontaneous nuclear break-up as modeled in an SMM-like freezeout scenario appears to be strongly contradicted by the prominent experimental trends, and at this time there seems to be no obvious remedy for this deficiency of SMM. The latter observation is perhaps related to the recently reported [30] inconsistency of SMM predictions with the IMF production observed in a spallation reaction, whereas the same data were well explained by GEMINI calculations. In that work [30],
an intranuclear cascade code (INC) was used to simulate the interaction stage of the process.

In contrast, but in agreement with other recent works [11–14], the present study suggests that IMF production in heavy-ion reactions is dominated by dynamical processes resembling QMD-like scenarios. This conclusion is largely based on the bombarding-energy dependence of the experimental correlations between the average size of IMFs and the joint neutron and LCP multiplicity reported so far in only one, very heavy ion

FIG. 5: (Color online) Logarithmic contour plot of average atomic number of the PLF, $< Z_{PLF} >$, as a function of associated neutron and LCP multiplicities, as predicted for the $^{136}Xe + ^{209}Bi$ reaction at E/A = 40 MeV by multi-step simulation calculations using codes CLAT and GEMINI (top row), CLAT and SMM (middle row), and QMD and GEMINI (bottom row).
This work was supported by the U.S. Dept. of Energy - Grants No. DE-FG02-88ER40414, DE-FG02-87ER-40316, and DE-FG02-88ER-40406, the Polish Ministry Of Science grant No. N N202 035636, and the M. Skłodowska-Curie Fund MEN/DOE-97-318.

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Use of Fission-fragment Kinetic Energies and Mass Asymmetry in the Analysis of the Dynamical Fission of Fast Projectile-like Fragments

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

Departments of Chemistry and Physics, University of Rochester

Rochester, New York 14627

Abstract

Simulation calculations are presented that demonstrate dangers of using PLF fission fragment data alone for the evaluation of the PLF excitation energy. Notably, for mass-asymmetric fission, the systematic errors of such evaluation depend noticeably on the orientation of the fission axis, which may result in a faux forward-backward asymmetry of the reconstructed angular distribution of PLF fission. Accordingly, purely statistical mass-asymmetrical fission of the PLF may incorrectly appear as a fast dynamical process taking place in the immediate vicinity of the TLF. It is suggested that sorting of data with respect to light-particle multiplicities avoids such bias in the interpretation of the data on the fission of projectile-like fragments.

I. Introduction

Dynamical fragmentation or multifragmentation of nuclear system in the course of heavy-ion collisions at intermediate bombarding energies has been the subject of many experimental studies in the last two decades [1-8], offering insight into the mechanical or hydrodynamical properties of heated nuclear matter. Of equal interest has been a statistical fragmentation of equilibrated systems formed in such collisions [9-14], offering potentially an experimental handle on a different set of properties of nuclear matter at elevated excitations in general and of the surface domain, in particular [14]. Accordingly, developing means of a reliable experimental discrimination between dynamical and statistical fragmentation is of paramount importance. Such a discrimination appears facilitated for situations where there are only three sizeable fragments in the exit channel, such as is the case where the projectile-like fragment (PLF) emerging from a dissipative collision undergoes binary fragmentation or fission. In such a case, the time-scales of the fragmentation can be inferred from the reconstructed angular distribution of the two fission fragments, provided the sorting scheme is neutral with respect to the orientation of the PLF fission axis. The present study alerts on a possible appearance of a false-positive signal of fast dynamical fission in a particular analysis scheme utilizing kinetic energies of (post-evaporation) fission fragments alone for the evaluation of the excitation energy of the fissioning PLF.
II. Statistical PLF fission observables

When events of statistical PLF fission are sorted into bins according to the sum of kinetic energies of the (evaporation residues of the two primary) fission fragments, $E_{H}+E_{L}$, and the fragment residue mass asymmetry, $A_{H}/A_{L}$, at least two distinct kinds of biases are introduced that create an illusion of a peculiar forward-backward asymmetry of statistical mass-asymmetric fission.

One bias arises from the fact that, for a mass-asymmetric statistical fission, any given value (or bin) of $E_{H}+E_{L}$ samples different regions of the PLF excitation energy for different orientations of the fission axis. Importantly, different excitation energy regions are sampled for the events where the lighter fragment is emitted in backward direction and different for the events where such fragments are emitted in forward direction. Since the statistical fission probability depends strongly on the excitation energy, one is to expect forward-backward asymmetry of the mass-asymmetric fission yield, when this yield is sorted into bins according to $E_{H}+E_{L}$.

A different bias arises from an additional binning of events according to the mass asymmetry $A_{H}/A_{L}$ of the remnants of the two fission fragments. This is so, because for a fixed PLF excitation energy, this mass asymmetry is strongly correlated with $E_{H}+E_{L}$. A “cut” on $A_{H}/A_{L}$ imposes, hence, an unintended “cut” on $E_{H}+E_{L}$, on top of the nominal cut due to the intended binning according to $E_{H}+E_{L}$.

Quite generally, the above biases result from the fact the same fragments are used to construct the angular emission patterns (invariant velocity plots) on the one hand, and to establish the parts of the yield to be included in individual patterns, on the other hand. Here, they arise specifically from the fact that the light primary fission fragment of a given temperature loses a different fraction of its mass to evaporation than does the heavy primary fragment of the same temperature, giving rise to what may be called a for/aft excitation energy defect within bins of constant $E_{H}+E_{L}$ and $A_{H}/A_{L}$.

II. Formalism and simulation calculations

Energy balance for a dissipative collision can be written as:

$$E_{beam} = E_{PLF}^{kin} + E_{TLF}^{kin} + E_{PLF}^{*} + E_{TLF}^{*} - Q$$

(1)

Assuming for the sake of simplicity that (i) the PLF and TLF are isotopically identical to the beam and target nuclei, respectively, (ii) that the excitation energy is divided between the PLF and TLF proportionally to their masses, and (iii) that PLF moves in the beam direction, one writes
$$E_{\text{beam}} = E_{\text{PLF}}^{\text{kin}} (1 + \frac{A_{\text{PLF}}}{A_{\text{TLF}}}) + E_{\text{PLF}}^* (1 + \frac{A_{\text{TLF}}}{A_{\text{PLF}}})$$

Eq. 2 expresses a simple relationship between the PLF kinetic energy and excitation energy, inviting one to use the former as a measure of the latter.

When subsequently PLF undergoes binary fission, its kinetic energy is split between the two fission fragments as is the gained Viola (kinetic) energy:

$$E_{H'}^{\text{kin}} + E_{L'}^{\text{kin}} = E_{\text{PLF}}^{\text{kin}} + E_{\text{Viola}}(A_{H'}, A_{L'})$$

Eq. 3, $A_{H'}$ and $A_{L'}$ denote mass numbers of the two primary fission fragments, such that $A_{H'} \geq A_{L}$ and $A_{H'} + A_{L} = A_{\text{PLF}}$. Eqs. 2 and 3 reveal a relationship between the sum of kinetic energies of the primary fission fragments and the excitation energy of the fissioning PLF, inviting one to use the former as the measure of the latter. Unfortunately, however, one does not detect the primary fission fragments but only what is left of them after they have de-excited mainly via particle evaporation.

In a tempting and perhaps intuitively reasonable first approximation, one could assume that the primary fission fragments acquire excitation energy proportionally to their mass numbers and that the amounts of evaporated masses are then also proportional to the primary fragment mass numbers. Under such an assumption, the sum of kinetic energies of the fragment remnants (evaporation residues) would be proportional to the sum of kinetic energies of primary fragments making the former a reasonably good measure of the excitation energy, as well. Unfortunately, such approximation is not accurate enough for the studies of angular patterns of PLF fission. This is demonstrated in series of figures reflecting results of simulation calculations for asymmetric fission of $^{124}$Sn as a PLF.

All figures shown are for a case where $^{124}$Sn fissions into primary fragments with $(A, \ Z)_{H'}=(100,40)$ and $(A, \ Z)_{L'}=(24,10)$ and $E^*_{H'}=320 \ \text{MeV}$ and $E^*_{L'}=80 \ \text{MeV}$. Similar effects were observed for different mass asymmetries and excitation energies. It was additionally assumed that the heavy fragment has a spin of 15 units, while the light one has 2 units of spin. The considered spins have little effect on the decay patterns. Their values were obtained by assuming sticking limit for the decay of a PLF with spin =39 hbar (taken from classical trajectory calculations). The statistical decay of primary fragments was simulated using the code Gemini.

Figs. 1 and 2 illustrate the distributions in mass numbers of heavy and light residues. Notably, the light fragments are seen to evaporate on the average a significantly larger fraction of their initial mass than are their heavy counterpart. Notably also, the distribution is quite broad for the lighter residues.
Accordingly, distributions of other derived quantities are quite broad, as well. This is seen in Figs. 3 – 6.

Figs. 3 illustrates the distribution of the (secondary) mass asymmetry $\Delta H/\Delta L$, while Figs. 4 – 5 illustrate the distributions of kinetic energies carried away by evaporated particles for events where the light fragment is emitted in forward direction (Fig. 4) and where it is emitted in backward direction (Fig. 5).
As seen in Fig. 3, the distribution of the apparent mass asymmetry is very broad where the primary mass asymmetry was fixed. This fact plays a critical role in biasing data when binning events according to this asymmetry.

Fig. 3. Distribution of apparent mass asymmetry of PLF fission.

Fig. 4. Distribution of total kinetic energy of all evaporated particles for events in which the light fragment is emitted in forward direction.
Fig. 5. Distribution of total kinetic energy of all evaporated particles for events in which the light fragment is emitted in backward direction.

![Heavy fragment, forward](image)

Fig. 6. Forward/backward kinetic energy defect.

![Forward/backward E defect](image)

The forward/backward kinetic energy defect represents the difference in total kinetic energies of evaporated particles for events in which the light fragments are emitted in forward direction and in which they are emitted in backward direction. This defect is a direct measure of the difference in excitation energies of PLF contributing to a given bin in the sum of kinetic energies of secondary fission fragments for different fragment alignments (light forward vs. light backward). The large width of this distribution virtually guarantees that a binning according to $E_{H} + E_{L}$ will result in an apparent forward-backward asymmetry of fission, where the true angular pattern is symmetric. This is seen
perhaps even more clearly in Fig. 7 illustrating correlations between \((E_L + E_H)\) and its forward/backward defect on the one hand and the mass asymmetry of secondary fragments on the other hand.

![Graph showing correlations between \(E^{\text{kin}}\) and mass asymmetry for light and heavy forward fragments](image)

**Fig. 7.** Correlations between the sum of kinetic energies of secondary fragments and the mass asymmetry of the secondary fragments for events in which light fragments are emitted in forward direction (top panel) and in which heavy fragments are emitted in forward direction (middle panel). The bottom panel illustrates the forward/backward kinetic energy defect as a function of mass asymmetry.
The bottom panel of Fig. 7 demonstrates the “biasing power” of binning according to $A_{1H}/A_L$, such that bins in lower values of this parameter show negative defect, while bins in higher asymmetries show positive defect. Note that a negative defect means that events in which light fragment is emitted in backward direction originate from PLF’s that have more excitation energy than events in which light fragment is emitted forward. Assuming that fission probability increases with excitation energy, this asymmetry results in the favoring of backward emission of lighter fragments. Conversely, for positive defects characteristic of higher mass symmetries, the particular binning would favor light fragments being emitted in forward direction.

III. Conclusions and suggestions

It appears impossible to prove that an observed forward-backward asymmetry of PLF fission is not an effect of biases such as demonstrated above. The analysis method guarantees distortion of the true angular emission pattern. To claim that what one observes is stronger than this “instrumental” asymmetry, one would need to carry out complex simulations for which there simply is no reliable input data.

There is a way to bin data according to the PLF excitation energy neutral with respect to forward-backward asymmetry of asymmetric fission. The traditional way consists in binning according to particle multiplicities. One could also bin alternatively with the kinetic energy of the TLF residue. While the latter is expected to offer rather poor resolution in the excitation energy, it is virtually 100% neutral with respect to the effect studied, i.e., the forward-backward asymmetry of the PLF fission.

The present simulations suggest that for higher mass asymmetries, the trend may be reversed, i.e., the light fragment will be emitted (apparently) preferentially forward. The emission patterns for these higher asymmetries should be inspected in spite of suspicions one might have regarding the origin of lighter fragments.

Acknowledgments

This work was supported by the U.S. Dept. of Energy - Grant No. DE-FG02-88ER40414

References


A Simple Method for Rise-Time Discrimination of Slow Pulses from Charge-Sensitive Preamplifiers

Jan Tőke, Michael J. Quinlan, Wojtek Gawlikowicz *, W. Udo Schröder

Departments of Chemistry and Physics, University of Rochester, Rochester, New York 14627

Abstract

Performance of a simple method of particle identification via pulse rise time discrimination is demonstrated for slow pulses from charge-sensitive preamplifiers with rise times ranging from 10 ns to 500 ns. The method is based on a comparison of the amplitudes of two pulses, derived from each raw preamplifier pulse with two amplifiers with largely differing shaping times, using a fast peak-sensing ADC. For the injected charges corresponding to energy deposits in silicon detectors of a few tens of MeV, a rise time resolution of the order of 1 ns can be achieved. The identification method is applicable in particle experiments involving large-area silicon detectors, but is easily adaptable to other detectors with a response corresponding to significantly different pulse rise times for different particle species.

Key words: Electronics, Pulse-shape discrimination, Particle ID
PACS: 21.65+f, 21.50.Ev, 23.70.Pq

1 Introduction

Pulse-shape discrimination is a well known technique often used to identify particles according to their atomic and mass numbers. The method is based on the characteristic time-dependence of the electrical or optical response they generate in the detectors. There are different physical effects responsible for the sensitivity of different detectors on the species of impinging particles. Accordingly, many different pulse-shape discrimination methods have been

* Present address: Heavy-Ion Laboratory, Warsaw University, Warsaw, Poland

Preprint submitted to Elsevier Science 15 May 2008
devised. In particular, the sensitivity of semiconductor detectors to different particles is due to the variation of the ionization density along the particle track in the detector material and the resulting temporal variation of the charge collection rate in the applied electric field.

The present study is inspired by the actual demand of measuring rise times of pulses produced by charged particles in large-area silicon detectors now used in multidetector arrays such as the CHIMERA telescope array [1]. 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, while the other is of the order of the shortest rise time expected in the particular application. With proper shaping, the amplitude of the “slow” pulse (representing the total charge injected into the preamplifier) will be virtually independent of the rise time of the raw preamplifier pulse, while the amplitude 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 amplitudes of both shaped pulses can be digitized by fast peak-sensing Analog to Digital Converters (ADCs), such as, e.g., the Phillips 7164 [2] or Silena [3] modules. Alternatively, depending on the available data acquisition, both shaped pulses can be stretched and their amplitudes be measured using current-integrating ADCs (QDC). Ultimately, the rise time is determined, event-by-event, from the ratio of “fast” to “slow” pulse amplitudes.

2 The Test Setup

A block diagram of the test 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 nominal rise times of 50, 100, and 200 ns, respectively. Charge pulses were obtained by charge-terminating pulses from a precision ORTEC 448 pulse generator. Different pulse rise times were employed as defined by the rise time settings of the pulse generator, while the decay time was fixed at 50 µs. A typical pulse with a nominal (set on the pulse generator module) rise time of 100 ns is illustrated in Fig. 2. It is seen to have an actual rise time close to nominal.

The output signal from the preamplifier was routed into two spectroscopic channels characterized by different shaping times. The “slow” channel had a shaping time fixed at 1 µs, while for the “fast” channel 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.
Fig. 1. Block diagram of the electronic setup used to determine the performance of the pulse-shape discrimination method.

Fig. 2. Appearance of the leading edge of a typical pulse from the charge-sensitive preamplifier with nominally 100 ns rise time.
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 to delay the output pulse from the ORTEC 454 TFA. The resulting timing of fast and slow pulses at the input of the peak sensing Phillips 7164 ADC is illustrated in FIG. 3. The present setup requires a peak sensing ADC capable of digitizing pulses with 50-ns short rise times, hence the choice of the Phillips 7164 ADC module.

3 Results and Analysis

The measurements were performed for series of pulses with rise times of 50 ns, 100 ns, and 200 ns and for shaping times for the “fast” response of 10 ns, 20 ns, and 50 ns. To assess the effect of inherent fluctuations in the amount of charge ($Q$) injected into the preamplifier, the latter charge was varied in the range from 0.222 pC to 3.55 pC, corresponding to energy deposits in a silicon detector in the range from 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 ($Q_F$) of the “fast” pulse on the injected charge ($Q$), as measured for different shaping and rise times denoted by time constants $RC$ and $RT$, respectively. As seen in this figure, shorter $RC$ shaping times provide for a better separation of faster pulses, a trend that agrees qualitatively with what is expected based on the principles of filtering.
Fig. 4. The dependence of the average amplitude ($Q_F$) of the “fast” pulse on the average height ($Q$) 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. The right column displays the corresponding ratios $Q_f/Q$.

A more quantitative measure of the pulse-shape discrimination performance of the method over a range of injected charges is seen in Fig. 5, where spectra of the ratios of the amplitudes ($Q_F$) of “fast” pulses to the amplitudes ($Q$) of the corresponding “slow” pulses are displayed for various $RC$ 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 the amplitude ($Q_F$) of “fast” pulses.

The relative widths $\Gamma_R/R$ of the peaks seen in Fig. 5 are plotted in Fig. 6 vs. injected charge for different shaping and rise times. The charge is expressed in units of MeV of equivalent energy deposit for silicon. As seen in this figure, the relative resolution is almost equally good for 10-ns and 20-ns shaping times. The resolution is better than 1% over most of the range of injected charges, except for the smallest one.

Fig. 7 illustrates the absolute resolution $\Gamma_{RT}$ in rise time in units of nanoseconds, achieved for different amounts of injected charge ($E$) and for different
Fig. 5. Spectra of the amplitude ratios ($Q_F/Q$) of “fast” ($Q_F$) and “slow” ($Q$) pulses, measured with various $RC$ shaping times and two representative injected charges. The three peaks seen in every panel correspond to the different rise times indicated by labels.

shaping and rise times. As seen in this figure, for the range of rise times explored, the 10-ns and 20-ns shaping times offer an almost equally good resolution of a few ns, over much of the range in charges explored. The weak but steady deterioration of the resolution with decreasing amount of charge injected, comes from the relative increase in statistical fluctuation in charge. It is encouraging that, except for the lowest charges injected, the resolution is of the order of a few ns, and is approximately 1 ns for the range of energies involved in typical experiments.

4 Conclusions

A simple and economical method has been demonstrated for rise time discrimination of pulses from charge sensitive preamplifiers. The method is based on measurements of the amplitudes of pulses derived from the raw preamplifier pulses by shaping with different time constants. It is shown to provide for a very good resolution in rise times. The method requires no fine tuning of the
Fig. 6. Relative resolution obtained for amplitude ratios of “fast” and “slow” pulses as functions of injected charge for different $RC$ shaping times and rise times ($RT$), as indicated by labels. The injected charge is represented by the corresponding energy deposit $E$ in a silicon detector, plotted on the abscissa.

electronics circuitry involved and should, therefore, be well suited for multi-detector systems. In the present study, a timing filter amplifier was used to produce the “fast” signal. In fact, a most primitive RC differentiator should perform sufficiently well, provided its output amplitude is matched to the range of the available ADC. The present design processes pulses from charge-sensitive preamplifiers used with solid-state detectors, but the method can easily be adapted to other systems such as scintillation detectors providing pulses with significantly different rise times.
Fig. 7. Absolute resolution (FWHM) in rise time $RT$, as extracted from data for different amounts of injected charge and different shaping and rise times, as indicated by labels.

5 acknowledgments

This work was supported by the U.S. Department of Energy grant No. DE-FG02-88ER40414.

References

PHASER 8 – A PHASE-LOCKED RF PRESCALER FOR CHIMERA EXPERIMENTS

J. Tőke a), I. Pawelczak a), M. J. Quinlan a), W. U. Schröder a), G. Cardella b), E. De Filippo b), A. Pagano b), and S. Pirrone b)

a) Departments of Chemistry and Physics, University of Rochester, Rochester, NY, USA
b) INFN Sezione di Catania and Dipartimento di Fisica e Astronomia Università di Catania

Abstract

An electronic module has been designed and built that automates the synchronization of prescaled RF timing signal to the beam monitor signals. The module, named Phaser 8, can be operated as an autonomous phase-locked prescaler or as a phase controller for an external prescaler. Its mode of operation and the desired RF phase with respect to the beam bursts is programmable via jumpers and two front-panel rotary switches. The logic of Phaser 8 is implemented in an XC95144XL CPLD (Complex Programmable Logic Device) by Xilinx and can thus be readily updated by an (in-system) reprogramming of the CPLD.

In experiments at cyclotron facilities with well defined beam microstructure, the RF signal is often used in data acquisition as a timing reference signal. When the facility is set up to generate a beam burst at the target site not at every RF cycle, but at a definite “sub-harmonic” of the RF frequency, need arises to extract a subset of RF signals that have a fixed timing with respect to the beam bursts. A simple way of generating such a subset is to use a prescaler operating at the same prescaling factor as is established between the RF pulses and the actual beam bursts. Unfortunately, because of the operating instabilities of the cyclotron, the timing relationship or the phase between the RF prescaler pulses and the beam bursts will occasionally change causing inconvenience in the subsequent interpretation of the data acquisition timing information. To minimize such an inconvenience, it is desirable to constantly monitor the relative phase between the (timing) prescaled RF signal and the beam bursts and to take appropriate corrective measures whenever this relative phase changes.

To automate and facilitate the control of the phase of the prescaled RF signal, a versatile NIM module, named Phaser 8, was designed that synchronizes the 1-of-n (n=2-8) prescaler output signal to a dominant component in the time spectrum of a reference signal. In its main design function, Phaser 8, called from here on simply Phaser, generates a prescaled cyclotron RF signal with a phase locked to a quasi-periodic and noisy reference signal – the dominantly elastic scattering events contaminated by some inelastic events. Thus, in this main mode of operation, it serves as a phase-locked RF prescaler, with the output signal useful as a time reference (stop signal) for TOF measurements. All that is in this case required from the reference signal is that a dominant component be present in its time spectrum that is at least 17/3 or 17/7 (jumper-selected) times more intense than the second strongest component – a condition that is easily fulfilled by the timing signal from almost any detector responding to beam-produced radiation.

Apart from the above phase-locked prescaler (Presc) mode, the Phaser can be used as an external prescaler phase controller (Ext) for an independent RF prescaler, such that it will generate RF Veto signals of a duration needed to shift the phase of the prescaler output to a desired value with respect to the reference signal.

In the absence of a suitable reference signal, Phaser can be also operated in an interactive-only phase-control mode, both, in Presc and the Ext settings. In this interactive mode, the phase of the prescaler output shifts in response to an external trigger (when in Presc mode) or a single-period RF Veto signal is generated in response to such a trigger (when in Ext mode).

A photograph of the completed Phaser is shown in Fig. 1, featuring prominently eight diagnostic LEDs and a number of LEMO jacks and control switches.

In spite of its many capabilities and an apparently complex look of the front panel, the actual use of Phaser is quite simple and may entail only one-time hardware setup consisting of

1) Setting the prescale factor consistent with the beam microstructure (front-panel rotary selector).
2) Supplying cyclotron RF and beam monitor timing pulses (NIM) to dedicated NIM inputs.
3) Selecting the desired phase of the prescaler output relative to the elastic scattering signal (rotary selector).
4) Selecting automatic operation (three-position switch).
5) Routing the RF/n phase-locked signal from the dedicated NIM output to the data acquisition setup.

Phaser is in fact an enhancement to the existing setup of the phase control of the cyclotron RF prescaler at the Chimera location, which makes its setup even more obvious to those familiar with the existing setup.
The operation of the Phaser can be understood from the timing diagram of the relevant signals shown in Fig. 2. In this diagram, it is assumed that the beam bursts arrive at the target every 4-th RF cycle and, for the sake of simplicity, any delays between the various waveforms are neglected. Accordingly, elastic scattering signals arrive sparsely simultaneously with beam bursts and, occasionally, inelastic events occur at “random” times. The presence of the RF signal is essential for the functioning of Phaser. An RF failure is indicated by the red LED labeled as “RF Fail”. The latter comes on whenever the duration of the RF pulse is shorter than approximately 1/8 of the RF period.

Phaser sorts monitor events into \( n = n_{\text{presc}} \) (prescale factor) time bins of width equal to the RF period, representing different phases of the monitor signals with respect to the prescaler output. Then, it identifies the dominant component in the monitor signal (elastic events) as the one that has an intensity of at least 17/3 (optionally, 17/7) of the next strongest component. When such a component exists and the module is set to operate in automatic phase-locking mode, Phaser compares the phase (bin number) of this component to the desired phase selected on the rotary selector. When these phases do not match, Phaser generates an RF veto signal of duration \( T_{\text{Veto}} = T_{\text{RF}} \cdot \Delta \text{Phase} \),

where \( T_{\text{RF}} \) is the RF period and \( \Delta \text{Phase} \) is the difference between the measured and preset phases. This veto applied to the RF prescaler input causes the phase of the prescaler output to shift so as to match the dominant phase of the monitor signal relative to prescaler output to the desired preset value. The attainment of the latter match is indicated by the green Lock LED lighting up, and the measured phase displayed in binary fashion on four yellow LEDs marked Phase. The generation of the Veto signal is indicated by blinking of the red Veto LED.

When operated in phase-locked prescaler mode \( (\text{Presc}) \), Phaser applies the above veto signal to its internal prescaler input resulting in the \( \text{RF}/n \) (NIM) signal that is phase-locked to the dominant component in the monitor signal timing spectrum. Whenever a dephasing occurs (see the bottom two traces in Fig. 2, Phaser establishes the new phase of the dominant monitor signal and generates again a suitable veto signal. Note that because of how the algorithm defines the dominant phase, it takes 17 dominant events to determine a new phase and to initiate the corrective action.

When operated in external phase-controller mode, Phaser routes the veto signal to the Veto I/O port for use in conjunction with an external prescaler.

A completed Phaser underwent successful tests both, on bench and in experiments.

This work was supported by the U.S. Department of Energy grant No.DE-FG02-88ER40414.
Fig. 2. Timing diagram of Phaser. Traces illustrate (from top to bottom) the cyclotron raw RF, the beam microstructure obtained by suppressing 3 out of 4 original beam bursts, a sample (exaggerated) monitor signal, the four possible phases of an RF/4 prescaler, the output of a phase-locked 1/4 prescaler locked to phase 0, and the RF veto generated by Phaser to restore phase 0 (after a "nuisance" dephasing to phase 1). Note that in a Presc operation, the RF Veto is generated only internally, while in Ext mode it is available at the Veto I/O port for the use in conjunction with an independent prescaler.
Development of a Novel Neutron Detector and Performance of its Prototype

I. A. Pawelczak\textsuperscript{a}, J. Tõkea, W. U. Schrödera,b

\textsuperscript{a}Department of Chemistry, University of Rochester, Rochester, NY 14627
\textsuperscript{b}Department of Physics and Astronomy, University of Rochester, Rochester, NY 14627

Abstract

Development and performance of a prototype module of N\textsuperscript{*} (NSTAR: “Neutron Sandwich Transmuter/Activation-\(\gamma\) Radiator”) are described. N\textsuperscript{*} is a large-volume modular neutron detector utilizing Gd-doped plastic scintillators. It realizes a principle of operation previously reserved to organic liquid scintillators or small gel-type detectors. The N\textsuperscript{*} design provides high transparency and light collection while avoiding environmental hazards characteristic of liquid scintillators. The N\textsuperscript{*} design uses the plastic scintillator both as a neutron moderator and as scintillator sensing neutron capture \(\gamma\)-rays. This detector has a high efficiency in a broad dynamic range of neutron energies (from thermal up to several MeV), an essentially zero energy threshold for neutrons and multi-hit capability. Design and development of a prototype detector module are described. They are based on simulation calculations using a newly extended simulation code (DENIS(E)). The prototype module consists of a stack of plastic scintillation slabs alternating with thin Gd-loaded transmuter films. The stack is viewed on both ends by photomultipliers attached via light guides. The response of the prototype module to cosmic-ray muons, gamma-rays (\textsuperscript{22}Na, \textsuperscript{137}Cs) and neutrons (\textsuperscript{252}Cf and D(d,n)\textsuperscript{3}He) is described. The experimental average neutron capture time \(<t_c> = (16 \pm 3)\, \mu s\), a characteristic property of the prototype detector determining its “speed.” The neutron detection efficiency of \(\epsilon = 26\%\) measured for the prototype module at an electronic threshold of 1 MeV\textsubscript{pe} is in good agreement with simulation predictions.

1. Introduction

Gd-loaded liquid scintillation detectors have been used in number of nuclear physics experiments from solar neutrino detection to light and heavy ion reactions as neutron multiplicity meters [1-8]. High efficiency measurement of the number of particles emitted from both reaction fragments reveals information on reaction excitation energy. In neutron rich systems, multiple neutron emission carries away a significant fraction of the excitation energy. However, in most reaction experiments the neutron multiplicity, number of neutrons produced/emitted per reaction event, is not measured. In laser induced fusion experiments, neutron spectrometry carries information of plasma temperature while neutron multiplicity would have would provide information on core density. However, neutron diagnostics [9-12] used in
these measurements still presents deficiencies and faces major challenges associated with application of detectors in harsh environment. Therefore, there is a high demand for high efficiency detectors, particularly of low energy neutrons. In dark matter experiments, such as the LUX and ZEPLIN projects [13-15], neutrons produce undesirable background events that need to be identified and rejected. There are numerous technical applications of strongly penetrating neutron radiation and its detection, ranging from nuclear energy and waste management applications [16-18], to reactor material testing, neutron radiography [19-20] and nuclear forensics, i.e. the search for illicit weapon and nuclear materials at borders.

Different experimental techniques are used in neutron detection, mainly classified according to neutron energy [21] and desired information. Thermal neutrons are measured indirectly by counting products of nuclear reactions they induce [22-27], e.g. charged particles or gamma rays. For slow neutrons only the neutron multiplicity can be obtained since their kinetic energy is negligible in comparison to reaction Q value. Fast neutrons, on the other hand, are measured by detecting proton recoil in hydrogen rich materials [28-34]. Neutron multiplicity for multi-MeV neutrons is primarily obtained by employing a pulse shape discrimination method available for certain liquid scintillation detectors [35], while energy can be measured with a time-of-flight technique. All of the mentioned above neutron detection methods are characterized by low detection efficiency, unless the detectors are voluminous.

In the next section, the design of a new type of neutron detector will be discussed together with the operating principles. It is followed by a description of the optimization process based on the Monte Carlo code DENIS(E) in Section 3 and detector development in Section 4. The response of the NSTAR module to cosmic ray muons, gamma-rays and neutrons will be discussed in Section 5 based on experimental data.

2. Concept of NSTAR

We propose a Gd-loaded plastic scintillation detector that provides excellent light collection and avoids environmental hazards associated with conventional detectors based on liquid organic scintillator. This detector, if configured in appropriate dimensions and geometry, can detect both, fast and slow neutrons with relatively high efficiency. More specifically, the proposed design of NSTAR needs to satisfy the following goals:

- detects neutrons in a wide range of energies (thermal to multi-MeV),
- has a zero energy threshold for n detection,
provides time-of-flight, energy deposit, and multi-hit information,
detects and tags muons,
is modular and can be configured in different geometries,
is made of inert, environmentally neutral materials,
is economical in construction, operation, and lifetime use.

The NSTAR detector module shown schematically in Figs.1-2, is a stack consisting of plastic scintillation slabs alternating with thin Gd-loaded films. The stack is viewed on both ends by a photomultiplier with a light guide. The main processes that are involved in neutron detection by NSTAR include neutron energy moderation, diffusion and neutron capture by Gd, which is an additive to the scintillator detector. Absorption of a thermal neutron is followed by γ-ray release, scintillation light emission and collection.

The organic scintillator serves dual purpose, as a scintillator and a neutron moderator due to its high content of hydrogen nuclei and density close to 1 g/cm³. Proton recoils from incoming scattered neutrons produce “prompt” fluorescence light which can provide information of integrated neutron energy. Moderated to thermal energies, a neutron diffuses for a few micro-seconds in the detector until it is captured by gadolinium nuclei in the radiator film between two adjacent scintillation slabs. Upon this delayed neutron capture, a cascade of prompt gamma rays with respect to absorption, is emitted of about of 8 MeV total energy. γ-rays interact with electrons in the scintillator mainly through Compton scattering producing fluorescence light. The delayed light output signal corresponds to the energy deposited by the γ-rays.

While neutrons are slowed down to thermal energies within the first 100 ns after entering a detector, it takes times of the orders of μs on average for a neutron to encounter and be absorbed by a Gd nucleus. Since the process depends somewhat on the Gd concentration and detector geometry, the mean capture time can be tuned by converter concentration and detector geometry. The associated delayed captured-γ signals are spread statistically in time, can be counted individually and provide neutron multiplicity information. Intuitively, one would incorporate gadolinium directly into plastic scintillator. However, any additive to the scintillator significantly lowers light attenuation length, making the detectors applicable only in small sizes [36-37]. Therefore, we separated scintillator from Gd-radiator and built a detector that consists of scintillator layers alternating with Gd-loaded films. This design satisfies the requirements mentioned above; high light output for the considered type of radiation, a large light attenuation length, fast timing, chemical and thermal stability. As a neutron converter, Gd was selected due to its excellent absorption properties for thermal neutrons. Capture thermal neutron cross-sections for natural composition of
gadolinium is of order of $5 \times 10^4$ barns. In the present design neutrons are identified by recoil protons and gamma rays following neutron capture.

3. Simulation Calculations

Design and performance of a prototype NSTAR detector module have been optimized through theoretical simulations combined with detector development and experimental testing. Monte Carlo calculations were performed for different detector architectures. The optimum thickness of scintillation slabs was determined based on numerical predictions of the neutron diffusion length. Some of these results will be discussed in this section.

An existing neutron transport computer code [40-41] was extended to include the NSTAR geometry resulting in a version DENIS(E). It is a Monte Carlo realization of neutron transport and capture followed by gamma interaction with matter. The geometry of detector, individual scintillator and radiator sheets in vacuum, is defined in terms of pixels. To determine an optimum spatial arrangement of the radiator films, various thicknesses (5cm; 2.5cm; 2cm) of scintillation slabs alternating with thin Gd-loaded films were considered for a fixed total volume/thickness of the scintillator. In the following, three different configurations, all with 20 cm width, 100 cm length, total scintillator thickness of 10 cm and 0.5 wt% of Gd, have been compared to the case of a homogenous distribution of Gd within the scintillator. The fact that a homogenous distribution already of small amounts of Gd makes the scintillator opaque and thus dysfunctional was neglected in the calculations. The theoretical light collection efficiency was set to 1 for all considered detector geometries. Monoenergetic beam of neutrons of 1 MeV was injected in the center of the bottom plane with direction perpendicular to the detector stack. Results of the simulations are presented in Figs. 3-4. Figure 3 illustrates the distributions of distances of capture sites from the neutron injection point, calculated for the geometries listed above. As capture distance one considers a path length that a neutron travels from the point at which its energy is at least equal to 50 meV until it is captured by a Gd or H nucleus. The diffusion length, plotted in Fig.4, was obtained from a Gaussian fit to the capture distance distributions as $\sigma$, $\lambda_{\text{diff}} = \sigma$.

With increasing number of Gd films, i.e., decreasing thickness of the individual scintillation sheets, the capture distance distribution approaches the characteristic shape for gadolinium spread homogenously within the detector. The goal of this exercise was to find geometry with a diffusion length close to that characteristic of a homogenous Gd distribution. Based on the results shown in Figs 3-4, a thickness of 2 cm for each scintillator sheet was considered to produce a good approximation and was therefore used for the prototype detector. Additional simulations were performed
to investigate the capture time distribution and to predict the detection efficiency as a function of Gd concentration. A reasonable compromise between efficiency and mean capture time was found to be given by a Gd concentration of 0.5% per weight.

The above calculations suggest the following optimum detector geometry: 2-cm thick 20x100 cm² scintillation slabs alternating with 0.1-cm thick radiator foils representing an overall Gd concentration of 0.5 wt. % per detector.

4. Development

Detector development was focused on fabrication of Gd-loaded radiator films, experimental tests of detector components, particularly light collection properties of scintillator and design of detector support structure.

Among the desired properties of the film are chemical and thermal stability, flexibility, integrity of large pieces and preservation of their shape in polymerization process. Poly–dimethylsiloxane, PDMS, commonly known as silicon rubber, satisfies all the requirements and was selected as gadolinium ‘holding glue’ for the radiator sheets. PDMS consists of two components, the base component (dimethyl siloxane, dimethylvinyl-terminated) and the curing agent (dimethylvinylated and trimethylated silica). Polymerization is achieved upon combining the two reagents which brake one of the double bonds between carbon atoms of terminal vinyl groups and attaches different parts of the curing agent components. The polymeric base was mixed with gadolinium oxide (III) and cyclohexane. The later solvent was added to dissolve and to obtain an even distribution of Gd₂O₃ within the film. Curing agent in a mass ratio of 1:10 was added to the base. The mixture was stirred for a few minutes and poured into a mold of the desired film area dimensions 20 x 100 cm². Addition of gadolinium oxide noticeably slowed down the polymerization process. The film was cured for 48 hours, the first 12 hours at room temperature, in order to allow air bubbles, which have been entrained during the mixing process to escape. For the final 36 hours the mold was placed under heat lamps. Prepared according to this prescription, the film, turned out to be flexible and easy to remove in one piece from the mold. The film was subjected to further tests to determine gadolinium homogeneity and film reflectivity.

Estimates of the gadolinium homogeneity within the film were obtained by exposing randomly chosen film samples to a flux of thermal neutrons and determining number of absorbed events. For that purpose, a ²⁵²Cf source was positioned behind a 6 cm thick block of paraffin resulting in production of flux of quasi-thermal neutrons measured with ³He proportional counter. The number of captured neutrons by gadolinium nuclei within the tested samples was established from a difference measurement of thermal neutron flux with and without a sample of Gd-loaded PDMS
film between the moderator and the detector. Results illustrated in Fig. 5 show a discrepancy of ±2% among the samples. The expected theoretical thermal neutron absorption systematically exceeds measured values by about 20%. This discrepancy is attributed to an overestimation of the thermal neutron flux. Only neutrons of fixed thermal energy (25 meV) were considered in the calculations, neglecting the finite width of the experimental distribution.

Information on film reflectivity was obtained by scanning randomly selected film patches with UV-Vis light. At wavelength of $\lambda=425$ nm, corresponding to maximum light emission of the BC-408 scintillator, reflectivity fluctuates at 40±6%, as seen in Fig. 6. In order to maximize the reflectivity and therefore the light collection efficiency resulting in better detector resolution, various reflective materials were tested for reflectivity. To ensure good light collection efficiency of the prototype detector, comparison of light output measurements was performed with a sample of BC-408 scintillator covered in two different reflector materials, specular reflecting aluminized Mylar foil and diffusive reflecting nitrocellulose paper. The scintillator was polished on all sides and wrapped in reflective material, except for the faces attached to photomultipliers. Coupling to the PMTs was direct, using optical grease. Medium-fast 5” 9390B photomultiplier tubes were used for the tests. The whole system was wrapped in black vinyl electrical tape to block ambient light.

Cosmic-ray muons were utilized to produce light flashes at the injection point defined via a set of 2 thin 2cm-wide scintillation detectors placed across the tested scintillator, operated in triple coincidence with the scintillator. It was found that the light output doubled when a diffuse reflector material was used as a wrapping. Because of superior performance, nitrocellulose paper was chosen as a reflector wrapping material in NSTAR detector. The above set of measurements also tested the sensitivities of the 5” Thorm-EMV 9390B and Philips XP2041 photomultiplier tubes, for BC-408 scintillation light. Better sensitivity and larger gain was achieved with XP2041 ensuring greater light collection efficiency and resolution.

Additionally, the light attenuation length $\lambda$ was determined for BC-408. Tests of light output as a function of scintillator length were conducted for an individual scintillation slab, connected through a light-guide to a XP2041 photomultiplier. Scintillator and light-guide were wrapped in nitrocellulose paper and covered with black tape. Cosmic-ray muons utilized in these measurements were defined as a triple coincidence of the scintillator with two 2cm-wide scintillator detectors placed across the sheet identifying injection points at distances of $25\pm1$ cm, $50\pm1$ cm and $99\pm1$ cm from the light-guide. Corresponding pulse height spectra are plotted in Fig.7. The light attenuation length for plastic scintillator BC-408 obtained from these measurements was $\lambda = 650 \pm 56$ cm.
Based on simulations and above tests, finally an NSTAR prototype detector was constructed out of six 2cm-thick BC-408 scintillator slabs with area of 20 x 100 cm², individually wrapped in nitrocellulose paper, which has excellent light reflection properties. Gd-loaded PDMS films of 0.1 cm thickness, with total amount of natural gadolinium equal to 0.5 wt.% were placed between the scintillator layers. The detector stack was viewed by a fast 5” XP2041 photomultiplier tube on either end, 20 cm long lucite light-guides were used for the PMT attachement to obtain better light collection efficiency. The light-guides, covered with nitrocellulose paper, and the detector stack module itself were wrapped in black paper and black vinyl electrical tape to create light tight system. Optical coupling between the stack module and the light-guides, as well as, interfaces between the light-guides and glass windows of the photomultipliers, was achieved by optical grease. The photomultiplier tubes were shielded from magnetic field by mu-metal.

The prototype detector was subjected to extensive series of tests conducted with cosmic ray muons, gamma and neutron sources. The detector response will be discussed in the next section.

5. Performance

5.1 Technical evaluation

The performance of the prototype detector module was evaluated based on its response to cosmic-ray muons and standard laboratory gamma sources, e.g. ²²Na, ¹³⁷Cs. In the following, studies with respect to time, position resolution and light output will be presented in the following section. The detector response to ²⁵²Cf and D(d,n)³He neutrons will be discussed in section 5.2.

Cosmic-ray muons are commonly used as a radiation source for calibration of scintillation detectors. Considering that vertical flux of relativistic muons deposits on average 24 MeV of energy per muon to the NSTAR prototype module, calibration can be provided in an energy domain characteristic of neutrons emitted during nuclear reactions. The entry point of muons impinging on the detector was defined by applying a triple coincidence requirement of two 2cm-wide scintillation strip detectors placed across the NSTAR, requiring a signal from either of the two attached photomultipliers. In the measurement, the relative time between signals from the photomultipliers was recorded together with the two pulse height spectra. The anode signal from either photomultiplier of the NSTAR module was attenuated and split, as seen in Fig. 8. One signal was used to trigger a constant fraction discriminator while the second one was delayed and sent to a QDC for light output information. Logical signals from the two CFDs were sent to a TAC whose output was digitized by an ADC.
for the measurement of relative time. Both, ADC and QDC were triggered by either
signal of NSTAR in coincidence with the two position defining scintillation detectors.
The difference in transit time of scintillation light generated along the path of muon, at
two photomultipliers was measured to obtain the position of the muon track along the
detector.

Relative time (position) distributions recorded at different positions of the
strip detectors along the NSTAR module are presented in Fig. 9. Here, the origin 0
(both in cm and ns) corresponds to the middle of the detector. The obtained time
resolution of $\Delta t_{FWHM} = (550 \pm 16)$ ps corresponds to a position resolution of $\Delta x_{FWHM} = (8.2 \pm 0.25)$ cm. Small deviations from linearity are due to a TAC instability during the
24h-long measurements. The effective in-medium velocity of scintillation light was
found to be $c_{eff} = 15$ cm/ns. The measured light attenuation length is $\lambda = (230 \pm 5)$ cm. A
resolution of $\Delta x_{FWHM} = 20\%$ was measured for the summed light output from the two
photomultipliers.

Light output calibration in a domain characteristic for neutrons from
radioactive sources, e.g. $^{241}$Am/$^9$Be, was performed with the gamma-ray sources $^{22}$Na,
and $^{137}$Cs. Gamma-rays interact with scintillator mainly through Compton scattering.
Experimentally, the summed light output for the $^{22}$Na ‘Compton edge’ at 1.06 MeV
indicates a resolution of $\Delta x_{FWHM} = 38\%$. Relative time distributions were measured
by requesting a coincidence of collimated beam of 0.511 MeV annihilation photons
from $^{22}$Na source. Data collected at various positions along NSTAR are plotted in Fig.
10. Time resolution is $\Delta t_{FWHM} = (3.4 \pm 0.24)$ ns resulting in a position resolution of
$\Delta x_{FWHM} = (47 \pm 3.2)$ cm. The time resolution obtained with annihilation gamma-rays of
0.511 MeV was worse than for the 24 MeV light generated by cosmic-ray muons, due
to the much smaller light output. Considering a light output distribution produced by
neutrons from $^{252}$Cf source used for testing purposes of the prototype NSTAR, this
module is essentially position insensitive for such neutrons. Firstly, a neutron
interacting with a proton in a collision can deposit from zero to its total energy,
therefore light output can vary from collision to collision decreasing time resolution.
Moreover, the scintillation light output associated with such proton recoils is
significantly smaller than that produced by electrons of the same energy (quenching).

5.2 Response to neutrons

5.2.1 Measurements with a $^{252}$Cf source

The prototype detector was subjected to a series of tests with $^{252}$Cf and 2.54
MeV D(d,n)$^3$He neutrons produced by a ThermoFisher Scientific MP 320 neutron
generator. Tests have been conducted using several neutron sources with different
activity, neutron multiplicity and associated gamma-ray background with different intensities and energy distributions. In the following, the prototype response will be discussed with respect to $^{252}$Cf and DD neutrons together with comparison to simulation predictions by DENIS-E.

The response of the prototype NSTAR module to neutrons was tested with a 0.189 µCi $^{252}$Cf source, providing 860 neutrons per second in 4π. Both, the prompt and the delayed detector response to neutrons were investigated in these measurements. Coincidence $\gamma$-rays [44] where used to “tag” neutrons. A time-of-flight experiment was performed which included a measurement of the prompt light output. The later delayed light output results from the gamma-rays associated with thermal neutron capture in gadolinium. Experiments were conducted with a $^{252}$Cf source placed at two distances, 60 and 160 cm from the center of the detector stack module, as seen in Fig. 11. The START signal for the time measurement was generated in the NaI detector by gamma rays from fission fragments emitted together with neutrons. A block diagram of electronics used in the measurement is displayed in Fig. 12. A signal from the NaI detector was amplified and split to feed a QDC and to produce a logical signal used as a START trigger for a TDC. Additional START signals appearing during the processing time of the data acquisition system of 140µs were vetoed, as explained in Fig. 13. Signals from both of the photomultiplier tubes of NSTAR were amplified and split. One pulse was used to trigger a constant fraction discriminator, the other one was used to measure a pulse height spectra. Signals from the discriminators were delayed and connected to the STOP inputs of a LeCroy 4802 23-bit TDC, operated in multi-hit mode with common START and 3 ns dead time and binning of one ns per channel. Information on prompt and delayed time distributions was obtained in these measurements.

Pulse height spectra of prompt and delayed light output summed over the signals from photomultipliers on both ends, were also recorded providing information on the integrated neutron energy deposit in units of electron energy equivalent, MeVee, and integrated capture gamma ray energy deposit, respectively. In elastic collisions with hydrogen nuclei in the scintillator neutrons are moderated to thermal energies promptly, i.e. within a few tens of ns. The trigger for the measurement of the prompt light output generated in the neutron slowing down process was constructed from either of the two STOP signals and vetoed at 100 ns with respect to a common START. That allowed one to measure the prompt of pulse height spectra and to disentangle this fast light output component from the delayed light output following neutron diffusion and capture generated at µs scale. The trigger of a LeCroy 2249W 12-bit QDC dedicated to the measurement of the delayed light output generated by
delayed gamma rays was produced in a very similar fashion but with a complement anti-veto gate instead of the veto, allowing for neutron capture events to be detected in the time interval from 100 ns to 100 µs.

Time of flight experiment was performed in order to test the detector’s fast and characteristic delayed response to neutrons. Time of flight and capture time were generated offline on an event-by-event basis as average of anode signals from photomultipliers on both ends of the detector module, according to eq.1 and eq.2,

$$tof = \frac{t_{p1} + t_{p2}}{2} + t_0$$  \hspace{0.5cm} (1)

$$t_c = \frac{t_{d1} + t_{d2}}{2} + t_0$$  \hspace{0.5cm} (2)

where $t_{p1}$ and $t_{p2}$ are prompt times measured by photomultiplier 1 and 2 respectively, $t_{d1}$ and $t_{d2}$ correspond to capture time alternatively, $t_0$ is a time offset. Two measurements were made for different flight distances. The corresponding time-of-flight spectra are plotted in Fig. 14, with time resolution of 3.4 ns. Calibration of time-of-flight spectra was performed using the $^{22}$Na coincident 0.511 MeV annihilation gamma rays triggering the NaI and NSTAR detectors. Due to the limited time resolution at the shorter distance, neutrons of energies above 8 MeV could not be discriminated against gamma rays by time of flight. In principle, neutron energy deposit information can be obtained from the “prompt” light output generated during a first few tens of ns after neutron entry to the detector. However, in practice this information is very difficult to disentangle from background radiation light output generated by gamma rays emitted from neutron capture, as can be seen in Fig. 15. Neutrons were discriminated against gamma rays by setting a gate on the neutron component in the time-of-flight spectrum and requiring a delayed event in the interval from 1-20 µs. The conditional experimental spectrum is plotted in Fig 16. For comparison, information on the expected prompt light output spectrum was extracted from DENIS-E simulations, folded with the detector resolution. The results are plotted in Fig. 17. The theoretically generated spectrum takes into account attenuation of light, assuming that on average scintillation light is generated in the middle of detector and its path to a photomultiplier’s photocathode is 50 cm long. Even though the conditional spectrum is quite similar to the one obtained in simulations, the origin of gap below 1 MeVee and peak about 1.2 MeVee is not understood.

According to the operational principle of Gd-loaded scintillation detectors, neutrons emitted simultaneously are captured by Gd (or H) nuclei at instances distributed over a relatively large time, due to stochastic nature of diffusion process. The capture time distribution can be described [45] by a two-parameter exponential
The experimentally obtained normalized neutron capture time distribution is shown in Fig. 18. For a comparison, capture time distribution obtained with the same conditions from DENIS-E is plotted in Fig. 19. As can be seen, the most probable capture time is 7 µs for a detector loaded with 0.5% Gd per weight, while the average capture time is $16.3 \pm 0.23$ µs, compared to $17.8 \pm 0.20$ µs obtained in simulations with DENIS(E). Hence, the experimental distribution is in a good agreement with theoretical predictions by DENIS-E simulations.

Considering light output spectrum of delayed gamma rays, as one can see in Fig. 20, while the low-energy capture gamma spectrum is dominated by background, the energy spectrum between 2.5 and 5 MeV is about 2 times higher in intensity than the background. Since scintillator is composed of light elements, H and C, one would expect a Compton continuum characteristic for a cascade of gamma rays directly from $^{158}$Gd and $^{156}$Gd. The probability of full absorption of the 2-3 capture gamma rays is close to zero for the prototype module. The origin of the Compton edges observed at about 1.3 MeV and 2.3 MeV is purely environmental. One can speculate that the lower energy Compton edge corresponds mainly to 1.48 gamma–rays from $^{40}$K, contamination by fission gamma rays and neutrons and gamma-rays emitted upon thermal neutron capture. On the other hand, the Compton edge at 2.3 MeV could be associated with background Th and gamma-rays emitted upon neutron capture by H. For the measurements performed with the NSTAR detector, one can only make a point that these features originate from background radiation. However, the resolution of the NSTAR (nor NaI) does not allow one to identify radiation sources and any assignment is only speculative. Since one expects the highest probability of neutrons being captured in time interval between 1 to 20 µs, one could use that information to reject background events from delayed gamma-rays. Selection of events was obtained by the requirement that a neutron be detected in the time-of-flight measurement and then captured within 20 µs. The shape of the resulting gated spectrum, which is displayed in Fig. 21, is obviously different from data previously shown. For comparison reasons,
theoretical spectrum obtained with DENIS(E) and folded with detector resolution is illustrated in Fig. 22.

From measurements of neutron time of flight and capture time, one can estimate the relative neutron detection efficiency, integrated over neutron energy distribution characteristic of a $^{252}$Cf fission-neutron source. Experimentally determined neutron detection efficiency by means of neutrons identified by time of flight and then captured, with threshold of proton recoil energy set to $E = 1$ MeVpe is $\varepsilon = (26 \pm 1)\%$ while a value predicted by DENIS-E is 25%. Obtained by Monte Carlo calculations performed with DENIS-E code, neutron detection efficiency as a function of neutron energy, characteristic for the prototype detector geometry is presented in Fig. 23.

From the above described data one can conclude that NSTAR detector can discriminate efficiently against gamma-rays in measurements of time but not in light output.

5.2.2 Measurements with a neutron generator, MP 320

Neutron generators provide a neutron flux in a well controlled fashion. A ThermoFisher Scientific MP 320 neutron generator can produce a pulsed neutron beam with a frequency ranging from 250 Hz to 20 kHz and minimum duty cycle of 5%. Alternatively, the generator can also be operated in a continuous mode. Neutron intensity can be controlled by timing settings of the accelerator and also by beam current - linear fashion, and accelerator voltage - exponentially. TTL pulses can be drawn from the MP 320 unit, which width and delays can be easily adjusted by LabView GUI used to run the generator.

Neutron generators have several advantages over alpha induced or fission sources. First of all, the intense gamma background associated with the de-excitation of an excited complex nucleus is absent from experiments with the neutron generator. That is a very significant improvement in the specificity of test measurements with a detector that is sensitive to gamma rays. Secondly, neutrons from the generator are approximately mono-energetic, emitted isotropically and can be produced with varying intensity and in a pulsed beam. Unfortunately, the beam pulsing operates on a microsecond scale and therefore, the beam pulses are not sharp enough in time to employ a time-of-flight technique. However, the generator provides the experimenter with a choice of different neutron multiplicity and can therefore be utilized to test multi-hit capability of the prototype.

The neutron multiplicity dependence on accelerator high voltage and beam current was studied with NSTAR prototype. The accelerator was run with frequency
of 10 kHz and duty cycle of 5%, providing time resolution of (0.937±0.01) μs. The electronics setup used in that measurement was very simple – consisting only of high voltage power supply, discriminator and CAMAC-USB controller with an incorporated internal scaler. The anode signal from one of the photomultiplier tubes was converted to a logical pulse by an ORTEC 934 quad constant fraction discriminator and counted by the CAMAC controller with an internal 11-bit scaler, in increments of 20 ns. The collected multiplicity events were used to fill in time spectrum with resolution of 100 ns per channel. The time window for the scaler was set to 400 μs with respect to the signal provided by the generator which actually triggered data acquisition, ensuring at the same time a coincidence requirement between the generator and the detector. The width of the pulses was set to 30 ns in order to introduce minimum dead time. The data were collected for beam current of 30 μA and high voltage of 45, 60, 75, 80, 85 and 90 kV, and 60 μA and 90 kV. Each measurement was carried for 5 minutes. The sequence of the measurements was later followed by 1-h long background measurement triggered with a pulser of the same settings as the signal provided by the generator; 5 μs width and 95 μs delay. One of the most important features of that measurement is that it provides info on the shape of the capture time distribution. This distribution is specifically characteristic of the geometry of the NSTAR prototype detector and the amount of gadolinium it contains. The other one is the flat time spectrum for background events which can be easily scaled to match the background intensity generated by neutrons in each measurement, extrapolated and subtracted from the measured distribution. Multiplicities recorded over 400 μs are plotted in Fig. 24. Also an example of time spectrum, generated with accelerator settings of high voltage of 75 kV and beam current of 30 μA is illustrated in Fig. 25 together with background represented by a red line. Binning is performed in 0.1 μs per channel intervals. There are two characteristic components present in the spectrum. Sharp peak corresponding to fast neutrons detected by proton recoil and a gradually decaying distribution over 100 μs for delayed gamma rays. For comparison, time spectra zoomed into time length of one cycle, measured with different neutron intensities varied by accelerator high voltage and beam current are displayed in Fig. 26 together with associated backgrounds. All of the above mentioned spectra result from combining signals generated by prompt neutrons, delayed gamma rays emitted during capture of thermalized neutrons, natural background radiation and neutron induced background, e.g. activation of 27Al. Therefore, to obtain neutron multiplicity, gate was set on time spectra in range from 60-1000 channels (for all for cycles), background was subtracted and number of neutrons per a burst was calculated. The results are plotted in Figs. 27-28 as a function of beam current and accelerator high voltage, respectively. According to manufacturer, MP 320 should produce 1x10^6 n/s in 4π at
accelerator settings of 60 μA and 90 kV. That means that 100 n/burst are generated in
4π at these settings. The solid angle coverage of the prototype detector was dΩ=13%,
resulting in flux of 13 n/burst. Experimentally obtained neutron multiplicity was 3.12,
therefore the prototype efficiency is ε=24% at threshold of 0.1 MeVee. These way
obtain efficiency estimates are relatively high in comparison to simulation predictions
with DENIS(E), according to whose, the prototype efficiency for 2.54 MeV neutrons
is ε=14% at 0.1 MeV threshold. That would suggest that the generator produces more
than 1x10^6 n/s in 4π. In order to estimate the prototype efficiency more accurately one
needs to calibrate neutron flux from the MP 320 unit. The calibration will be
performed with liquid scintillation detector, BC-501 A. Pulse shape discrimination
method will be used to identify neutrons.

6. Conclusions

The goal of this work was to develop and test the prototype of a new neutron
detector module. The new NSTAR module fulfils the main design goals with respect to
light output, time and energy. It has excellent light collection properties and represents
an environmentally much safer alternative to Gd-loaded liquid scintillation detectors
and could be used in confined spaces, e.g., in underground labs. The design of the
detector stack module was optimized as suggested by a set of Monte Carlo simulations
performed with an extended DENIS-E code and experimental tests. The following properties
of the NSTAR determined experimentally:

1) Scintillator light attenuation length
   a. including reflection on one end of the stack, \( \lambda = (650 \pm 56) \text{ cm} \)
   b. without reflection on one end, \( \lambda = (230 \pm 5) \text{ cm} \).

2) light output resolution of the detector
   a. 20% at 24 MeV (cosmic muons)
   b. 38% at 1.06 MeV (\(^{22}\)Na Compton edge)

3) effective speed of light,
   a. measured \( c_{\text{eff}} = 15 \text{ cm/ns} \)
   b. calculated from BC-408 refractive index, \( n_{\text{BC-408}}=1.58 \), \( c_{\text{eff}} = 17 \text{ cm/ns} \)

4) time resolution, \( \Gamma_{\text{FWHM}} \)
   a. (550 ± 16) ps at 24 MeV
   b. (3.4 ± 0.24) ns at 0.511 MeV

5) position resolution
   a. (8.2 ± 0.25) cm at 24 MeV
   b. (47 ± 3.2) cm at 0.511 MeV

6) average capture time
   a. experimentally determined \( <t_c> = (16.3 \pm 0.23) \mu s \)
b. from DENIS(E) \( \langle t_e \rangle = (17.8 \pm 0.20) \mu s \)

7) neutron detection efficiency of fast neutrons, integrated over energy distribution, with detection threshold of 1 MeV
   a. experimentally determined \( \varepsilon = (26 \pm 1)\% \)
   b. from DENIS(E) \( \varepsilon = 25\% \)

To summarize, the main design goals; no energy threshold for neutron energy, high detection efficiency for neutrons of energies ranging from thermal to 2 MeV, multi-hit capability and excellent light output efficiency, light attenuation length and fast timing are satisfied by the prototype detector. Average neutron capture time and the detection efficiency of fast neutrons are in good agreement with values obtained with a modified version of the DENIS(E) code. The prototype efficiency for D(d,n)3He will be obtained after calibration of neutron yield of MP 320 with liquid scintillator, BC-501 A.

Acknowledgments

This work has been supported by U.S. Department of Energy Grant No. DE-FG02-88ER40414.

References

Fig. 1 Schematic view of an NSTAR detector stack module.

Fig. 2 NSTAR prototype detector.
Fig. 3 Capture distance distributions for considered geometries of the NSTAR prototype.

Fig. 4 Diffusion length for corresponding detector architectures.
Fig. 5 Thermal neutron absorption by Gd-loaded PDMS samples of area of 6.5 x 6.5 cm$^2$, selected randomly through film, measured with $^3$He proportional counter.

Fig. 6 Light reflectivity of Gd-loaded PDMS samples at $\lambda = 425$ nm wavelength of maximum light emission of BC-408.
Fig. 7 Pulse height spectra recorded with individual BC-408 scintillation sheet coupled to XP2041 photomultiplier tube via a lucite light-guide. Measurement performed utilizing cosmic-ray muon radiation with injection points at 25±1, 50±1 and 99±1 cm from the light-guide.
Fig. 8 a) Schematic view of NSTAR cross-section and implementation of principle of position measurement by means of relative time, described in Section 5.1. Vertical arrow indicates flux of incoming muons, $d_s$ – position difference from muon interaction to the center of the detector. b) Electronics diagram used in the measurements of the position resolution.
Fig. 9 Relative time distributions of cosmic-ray muons at several positions across the detector, where 0, (both in cm and ns), corresponds to the middle of the detector.

Fig. 10 Relative time distributions of 0.511 MeV gamma rays at several positions across the detector, where 0, (both in cm and ns), corresponds to the middle of the detector.
Fig. 11 Experimental setup used in time of flight and capture time measurements. Distance between neutron source and detector is not sketched up to scale.

Fig. 12 Block diagram of the electronics for time of flight measurement together with neutron capture time, prompt and delayed pulse height spectra.
Fig. 13  Timing diagram of a measurement of time of flight, capture time and corresponding light output spectra.

Fig. 14  Time of flight spectra detected by NSTAR relative to START signal from NaI detector. $^{252}\text{Cf}$ placed at distance of a) 60 cm, b) 160 cm from the NSTAR module.
Fig. 15 Net light output spectrum of fast neutrons together with separately measured background.

Fig. 16 Light output spectrum of fast neutrons. Gating condition was defined by neutrons detected in time-of-flight and captured within 1-20 μs.
Fig. 17 Prompt light output spectrum simulated with DENIS-E code for Maxwell-Boltzman neutron energy distribution with temperature, $T = 1.5$ MeV, characteristic to $^{252}\text{Cf}$ source. Triangles indicate DENIS-E spectrum while histogram represents theoretical spectrum folded with detector resolution.
Fig. 18 Experimental capture time distribution obtained with $^{252}$Cf neutron source.

\[ \lambda = 0.6631 \pm 0.0183 \text{ ms}^{-1} \]
\[ \beta = 0.0757 \pm 0.0009 \text{ ms}^{-1} \]
\[ A = 1942.2 \pm 40.1 \]

Fig. 19 Capture time distribution simulated with DENIS-E.

\[ \lambda = 1.0052 \pm 0.0430 \text{ ms}^{-1} \]
\[ \beta = 0.0631 \pm 0.0007 \text{ ms}^{-1} \]
\[ A = 722.019 \pm 13.653 \]
Fig. 20 Net pulse height spectra of delayed gamma rays together with separately measured background.

Fig. 21 Pulse height spectra of delayed gamma rays with conditions set on neutron time of flight and first 20 µs of capture time distribution. Corresponding background is illustrated by open triangles.
Fig. 22 Energy spectrum of gamma-rays emitted upon neutron capture, simulated with DENIS-E (triangles) and folded with detector resolution (histogram).

Fig. 23 Theoretical neutron detection efficiency as a function of neutron energy simulated for time of flight experiment with DENIS-E code.
Fig. 24 Neutron multiplicity recorded over 400 μs. Pulse width was set to 30 ns. Accelerator was run with frequency of 10 kHz and duty cycle 5% while neutron yield dependence on high voltage and beam current has been tested.

Fig. 25 Neutron time spectra recorded with NSTAR in coincidence with MP 320, plotted with resolution of 0.1 μs per channel. Full scale corresponds to 400 μs.
Fig. 26 Time spectra of DD neutrons recorded with NSTAR in coincidence with MP 320, plotted with resolution of 0.1 µs per a bin, providing the full range of 100 µs.
Fig. 27 Neutron multiplicity determined from capture time distribution per a burst as a function of accelerator beam current at high voltage of 90 kV.

Fig. 28 Neutron multiplicity determined from capture time distribution per a burst as a function of accelerator high voltage for beam current of 30 μA.
Dependence of Desorption Rate of HTO from Metal Samples on Temperature and Carrier Gas Humidity*

A. Brown, M. J. Quinlan, W.T. Shmayda, and W.U. Schröder

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

Laboratory for Laser Energetics, University of Rochester, Rochester, NY, 14623

Abstract
Thermal desorption of tritium from exposed mill finished coupons was studied at various temperatures and carrier gas humidity. The coupons were prepared in three different ways and at different times before measurements. The effects of the variables were studied by examining the initial amounts of tritium activity on each coupon, the amount removed and the time dependence of desorption. In the experiments it was found that the tritium desorption rate increases with temperature, although the magnitude of the effect depends on coupon type and preparation. High humidity of the carrier gas also increases tritium desorption, but at lower humidity this trend was inconclusive.

I. Introduction
At the University of Rochester Laboratory for Laser Energetics (LLE), the upcoming National Ignition Facility (NIF), as well as at other such research facilities around the world, tritium is utilized as fusion fuel. Tritium is also used in radio-luminescent and pharmaceutical industrial applications. In addition, tritiated water has become an issue of growing importance in the nuclear power sector where, in so far relatively small amounts, this radioactive compound is routinely produced in the operation of light-water reactors and typically discharged untreated into the environment. When metals are exposed to tritium or tritiated compounds, these compounds not only adsorb easily onto the metal surfaces but in time can penetrate into the bulk, contaminating the material throughout. Before these parts can be serviced or discarded, the activity must be removed from the metal. However, at present no efficient decontamination or recycling methods are generally available to research and industry.

The nuclear chemistry (Nuchem) group follows several different radio-chemical approaches to discover an efficient method to recycle tritium and tritiated compounds from metallic samples. The present article gives an account of progress made in the thermal desorption of tritium from exposed stainless steel samples. A related goal of this activity by the Nuchem group has been to provide training grounds for students and other personnel in radio-chemical methods.

*Undergraduate Senior Research Project in nuclear chemistry
II. Experimental

For the purpose of the present study, three different batches of various metal samples (“coupons”) were loaded with tritium by exposing them to a tritium gas atmosphere. By measuring the release of activity from these coupons in different environments, one hopes to gain a better understanding of the adsorption of tritium to stainless steel and its desorption from the metal, leading eventually to the development of efficient tritium removal methods.

Consequences of differences in the amount of activity loaded onto a coupon or in the loading method have been examined, both by monitoring loss of activity over time and total activity removed by thermal desorption. Total activity removed and associated rates were investigated for each coupon, under various conditions of temperature and water vapor pressure in the desorption chamber. In previous research conducted by the group², it was found at all temperatures that an increased humidity of the carrier gas led to greater tritium desorption. It has been the specific goal of the present study to investigate trends in the temperature and carrier gas humidity dependencies, as well as to gain insight into the chemistry of tritium compounds adsorbed at stainless steel surfaces.

IIA. Preparation of Samples

Over the past five years, three different tritium loading methods have been implemented to adsorb tritium to stainless steel type SS316 coupons, designated here as Types I, II and III, respectively, with the conditions noted in Table 1. The set of experiments reported here tested all three types.

<table>
<thead>
<tr>
<th>Table 1. Tritium Loading Conditions</th>
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<td>Exposure Date</td>
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Type I Coupons: These coupons, 2 cm x 5 cm x 5 mm thick low carbon 316 stainless steel samples, were loaded with tritium at the British UKAEA/JET facility. The metal samples had not been forged or polished but were cleaned of oils and fingerprints, in accordance with JET in-vessel procedures. No heat treatment was applied to the samples before their >105-hour exposure to HT gas at 375.03 Torr and 26.8 °C. The samples were charged with tritium in sets of two batches. The first exposure of 36 samples was completed on February 26, 2004, while the remaining samples were removed from the dosing facility on March 5, 2004. These two batches of differing exposure times were not differentiated within this set of experiments. Following both loadings, the HT was evacuated and replaced with inert gas. The samples were individually wrapped in paper and an aluminum over-wrap and fixed in
slots in the shipping container such that they could not touch each other or the container walls. All coupon handling was carried out in an inert gas environment. The shipping container was filled with argon gas, was sealed and received at LLE on March 15, 2004.

At the LLE, the shipping container was opened in a helium glove box operated at a moisture level below a –60 °C dew point. Twenty coupons were unwrapped and sealed in a separate container that was backfilled with air for later decontamination tests. The remaining coupons were transferred from the glove box to the experiment, as the need arose, in groups of 2 to 4 in a sealed container backfilled with air.1

Type II Coupons: The next set of coupons were prepared at LLE on September 25, 2007. The batch consisted of 30 coupons, with five each of the following: 316 stainless steel, mill finish, 316 stainless steel satin finish, 110 copper alloy mirror finish, 110 copper alloy mill finish, 110 copper alloy OFHC mill finish, and 6061 aluminum mill finish. They were degreased in an ultrasonic bath in a solution of surfactant and water, then dried in alcohol. Loading was carried out for 24 hours in an atmosphere of 50% DT at 748.8 Torr and 29 °C. Following exposure, the coupons were stored in a helium gas atmosphere. Eight coupons were initially transferred in a He atmosphere into a container and delivered on February 4, 2008, to the thermal desorption facility. Five more 316 stainless steel mill finish coupons, were transferred and delivered on June 6, 2008.

Type III coupons: The final batch of coupons was also prepared at LLE under conditions similar to those used for the previous set. The coupons were loaded on September 11, 2008. The batch consisted of 21 pieces of 316 stainless steel mill finish and 9 pieces of 316 stainless steel mirror finish. They were exposed for 96 hours to 729.4 Torr of DT at 28.5 °C. Again, the coupons were exposed only to a helium cover gas when transferred. Initially, 10 mirror finish coupons were delivered on October 23, 2008, followed by a delivery of 10 mill finish coupons on October 28, 2008. Unlike previous samples, these coupons were kept in a He environment from initial loading to desorption, with new precautionary measures being taken to minimize exposure to air.

IIB. Experimental Setup and Procedures

Each individual coupon was analyzed with a variety of different instruments and in a variety of ways, leading to several different sets of data. A discussion of the instruments and experimental conditions follows.

Surface Activity Monitor (SAM): The Surface Activity Monitor shown in Figure 1 is a non-invasive tool for measuring the amount of tritium on the surface of a sample. A contaminated coupon placed in the monitor was surrounded by a Faraday cage. The cage is closed when a measurement is taken. Since the electronics took time to adjust, each coupon was left in the monitor for about 20 minutes, in order to obtain an accurate reading. Under helium this process took even longer; the coupon

Figure 1. Surface Activity Monitor
had to be left in the monitor for between one and six hours in order for the reading to steady. The SAM displays the β activity measured from the coupon in units of mV. This provides as accurate measurement of the amount of tritium from the surface to a depth of .8 µm into the metal. At the end of a desorption experiment, the coupon was again measured with the SAM to obtain the amount of activity remaining on the (near) surface.

The SAM principle of operation utilizes a correlation between the number of tritium decays and the amount of current. When a beta particle, a low energy electron, is released by a decaying triton, it ionizes other atoms inducing a stream of electrons, i.e., an electric current. By measuring this electronic current, the activity on the sample can be estimated using the formula,

$$I_{sat} = \frac{eE_m}{2W} \lambda n_s$$  \hspace{1cm} (1)

Here, \(I_{sat}\) is the saturation current, \(\lambda\) is the decay rate, and \(n_s\) is the number of tritons. The mean energy of the betas is denoted as \(E_m (=5700\text{ eV})\), and \(e\) is the elementary electric charge \((1.602 \times 10^{-19}\text{ C})\) carried by each electron. \(W\) denotes the amount of energy needed to generate an electron pair, which has a value of \(W=33.7\text{ eV/ion pair in air and } W=41.9\text{ eV/ion pair in helium. In the SAM, a coupon also acts as an anode on which the electron current induces a signal. This signal is then boosted by a trans-impedance amplifier, to be finally displayed as a voltage. Voltage and the current are related by the equation}

$$V(mV) = 0.196 \times I(pA)$$  \hspace{1cm} (2)

Combining Equations 1 and 2 yields the conversion:

$$\lambda n_s = 0.01020 \times V(mV) \text{ mCi}$$  \hspace{1cm} (3)

for measurements carried out in air, where \(\lambda n_s\) is the number of tritons decaying on the surface. This equation changes to

$$\lambda n_s = 0.01276 \times V(mV) \text{ mCi}$$  \hspace{1cm} (4)

for measurements conducted in a helium environment.

**Thermal Desorption Facility:** The Thermal Desorption Facility shown in Figure 2 contains a number of different components to remove, segregate and collect tritium released from the contaminated coupon. Ultra High Purity Helium was passed through a dryer and then split into two streams. These streams were separately controlled and measured by a flow meter, although the total flow was always equal to 100 standard cm$^3$ per minute (sccm). A portable flow meter was utilized at different points in the system to ensure that there were no leaks and that the flow was steady. Stream 2 was diverted through Bubblor 0 (B0), which served as a humidifier for this helium carrier stream. The streams were then reunited and sent through the desorption furnace.
Figure 2. Thermal Desorption Setup

The furnace comprised a quartz tube that was enclosed in an insulated oven. This furnace was heated to desired temperatures using a resistive heating element, whose temperature was controlled by a LabView program and monitored using a K-type OMEGA thermocouple. The temperature of the coupon was also measured directly using a second, similar thermocouple. While inside the quartz desorption chamber, a coupon was exposed to a carrier gas stream, which exited the oven on the opposite side. The gas then traveled through a series of bubblers which removed water soluble species (HTO) from the stream. Bubbler 1 (B1) was filled with a water/liquid scintillator mix and connected to an in-line Beta Radiation Activity Monitor (β-RAM). A syringe was also inserted into the bubbler allowing one to withdraw aliquots that could be analyzed using a separate external Liquid Scintillation Counter (LSC). After passing through B1, the stream moved through Bubbler (B2), which was filled with water. The contents of B2 were analyzed before and after each experiment to determine a typically minute remnant activity. B2 provided a measure of the collection efficiency of B1 for water soluble components. The gas was then oxidized using a Cu/CuO filled oven set to a temperature of 700 °C. At this temperature, water insoluble HT molecules are converted to water. Emerging from the oxidation furnace, the gas passed through Bubbler 3 (B3), which was connected to a second β-RAM. Like B2, Bubbler 4 (B4), which contained only water, was sampled prior to and following each experiment.

Humidifying the He Carrier Stream. To humidify the stream, the carrier gas was passed through B0, where it picked up moisture at equilibrium vapor pressure. It was then combined with the dry helium stream to obtain the desired humidity. At room temperature, 22 °C, the equilibrium vapor pressure of water is 19.8 Torr. This value can be converted to a concentration by dividing by the standard atmospheric pressure, 760 Torr, to give an expected value of 26,053 ppm when all of the gas flows through B0. Multiplying by the
fraction of total gas stream passed through BO, theoretical values for the amount of Table 2. humidity in the carrier stream can be calculated, as shown in Table 2.

**β Radiation Activity Monitor:** B1 is monitored by one β-RAM, while B3 is hooked up to the other. The β-RAM consists of a detector connected to one of the collectors (B1 or B3), through which scintillation fluid is continuously circulated. The detector is a photo-multiplier tube (PMT), which detects light flashes caused by β particles in the scintillation fluid. Thus, the number of photons that a PMT “sees” is approximately proportional to the number of β decays. The β-RAM outputs a measure of the activity in the sample drawn from the bubbler in units of counts per minute. The instrument is highly useful because it constantly measures the activity coming off a coupon in real time, allowing for a small delay, which is discussed in the next section.

**Delays for Bubblers 1 and 3** There is a delay associated with the time it takes the carrier gas to flow over the coupon, where it picks up activity and to reach the bubbler, where it deposits activity into the scintillation fluid. From there, the scintillation fluid must travel through the β-RAM tubing to reach the cell where it is counted, adding to this delay. The response time can be calculated using the following formula for the gas volume per time:

\[ V = \pi r^2 L \]  

(5)

where \( r \) is radius of the tube and \( L \) the length of the tube, both of known values. Dividing this volume by the flow rate \( \frac{dV}{dt} \), a value for the delay time can be calculated. Length and radius measurements were made for each component of the system, including the oxidation furnace. The delays for both bubblers were then calculated using these values, as explained below.

The inner radius of the tube going from the oven to B1 was measured to be 0.4 cm, while its length was 53 cm; the volume traversed by the gas was 26.63 cm\(^3\). Dividing this value by the flow rate, 100 cm\(^3\) per min, we find that the system response time was 0.27 min. From there, the activity must travel to the β-RAM, where it is measured. Formula (7) can be used to calculate this second delay, carried by a 200-cm long tubing of 0.1 cm radius.

**Table 2:** Expected Water Vapor Pressures based on flow through BO

<table>
<thead>
<tr>
<th>Percent Flowing through BO</th>
<th>Mole fraction of H(_2)O (ppm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>26,053</td>
</tr>
<tr>
<td>75</td>
<td>19,533</td>
</tr>
<tr>
<td>50</td>
<td>13,026</td>
</tr>
<tr>
<td>25</td>
<td>6,513</td>
</tr>
</tbody>
</table>

**Table 3.** Parameters of setup relevant for transport of tritium activity from a sample to the β-RAM detectors. The last column lists estimated total delays from sample to detector.

<table>
<thead>
<tr>
<th>Bl</th>
<th>Radius to Bubbler (cm)</th>
<th>Length to Bubbler (cm)</th>
<th>Tubing Volume (cm(^3))</th>
<th>B1 +B3 Volume (cm(^3))</th>
<th>Flow Rate (cm(^3)/min)</th>
<th>Delay Time (min)</th>
<th>Radius to β-RAM (cm)</th>
<th>Length to β-RAM (cm)</th>
<th>Tubing Volume (cm(^3))</th>
<th>Flow Rate mL/min</th>
<th>Delay Time (min)</th>
<th>Total Delay Time (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.4</td>
<td>53</td>
<td>26.6</td>
<td>n/a</td>
<td>100</td>
<td>0.3</td>
<td>200</td>
<td>6.3</td>
<td>1</td>
<td>6.3</td>
<td>6.5</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>0.4</td>
<td>212</td>
<td>106.5</td>
<td>1613.8</td>
<td>100</td>
<td>17.2</td>
<td>0.1</td>
<td>200</td>
<td>6.3</td>
<td>1</td>
<td>6.3</td>
<td>23.5</td>
</tr>
</tbody>
</table>
Dividing this length by the $\beta$-RAM pump rate of 1 mL per minute, one finds that a delay of 6.28 minutes, giving a total delay of 6.5 minutes for the activity to travel from the oven containing the coupon to the $\beta$-RAM. These data, along with the corresponding values for B3, are summarized in Table 3.

Liquid Scintillation Counter: The LSC is a LKB-Wallac 1209 RackBeta. Small aliquots, between 0.1mL and 0.5mL, were withdrawn from each bubbler before and after thermal desorption. After thermal desorption, typically most of the original activity on the coupon has been removed and deposited in the bubblers. The aliquots from each bubbler can therefore be used to calculate the total activity removed from the coupon in the following manner.

The aliquots were added to $(15.29 \pm 0.01)$ mL of pure scintillation fluid and loaded into the LSC. The instrument operates in a manner similar to the $\beta$-RAM, counting photons produced by $\beta$ decays. In order to convert the output value from counts per minute (cpm) to decays per minute (dpm), the machine efficiency, a measure of how many decays are actually represented each count, must be taken into account. For the machine used in the measurements, the efficiency was $\varepsilon = 0.5$. Once the dpm value has been calculated, it can easily be converted into units of Becquerel or Curie. In this study mCi are used to represent activities, mainly because the amounts of activity on the coupons fell within this range.

In order to calculate the total amount removed from the coupon, the cpm value was divided by the efficiency, to give dpm. This was then converted to Becquerel and mCi. This value was divided by the total volume of fluid in the bubbler to determine the total amount of activity in that bubbler.

Experimental Runs: Over the past year, a total of 18 experiments were run following the procedure detailed above. Each run varied in the type of coupon used, the amount of activity on that coupon, and the vapor pressure of the carrier gas. All of the experiments and the various conditions under which they were run are outlined in the directory shown below (Table 4).

<table>
<thead>
<tr>
<th>Experiment/Coupon #</th>
<th>Exposure Date</th>
<th>Delivery Date</th>
<th>Experiment Date</th>
<th>Type</th>
<th>Total Flow (sccm)</th>
<th>B0 Flow (sccm)</th>
</tr>
</thead>
</table>

Table 4. Experiment conditions used with Type I (blue), Type II (green) and Type III (yellow) coupons.
III. Experimental Results

III.A. Coupon Data: The tritium activity on each coupon was measured with the SAM and independently with the external LSC before and after each run and are shown with type and date noted in Table 5. The amount of time between sample loading and the SAM measurement is noted in column 3. The activity on each coupon has been converted to mCi in column 9. The different coupon types were found to have differing amounts of activity on them. Currently, the different tritium activities are attributed to differences in the original exposure.

In Figure 3 the amount of activity detected on a coupon at first measurement is plotted against the amount of time since original loading. Here, the error bars shown are ± 10 percent of the measured activity. There is an obvious correlation between the amount of time the coupon has been left out and the amount of activity remaining on it. To determine this relationship, the data has been fit to a linear function, giving a loss of 1.7 µCi per day. This number accounts not only for radioactive decay, but also for other physical processes like adsorption into the bulk beyond the ability for a SAM measurement of desorption from
the surface. To estimate how much of the activity loss was due to the radioactive decay of tritium, the expected percent remaining was calculated using the following formula:

\[
\text{fraction remaining}(t) = e^{-0.693 \frac{t}{t_{1/2}}}
\]  
\(6\)

Here, the natural log of 2 (0.693), is multiplied by \(t\), the number of years since the sample was loaded. Because some of the coupons had been loaded more than five years ago, a non-negligible amount of the activity had decayed, as tritium has a half life of \(t_{1/2}=12.323\) a.

For the oldest sample, which was measured 1615 days after exposure, only 78.0\% of the

**Table 5.** SAM Results, with Type I (blue), Type II (green) and Type III (yellow).
initial activity would be left on the coupon. This percent was calculated for each coupon, based on the time elapsed between contamination and the date the SAM reading was taken.

![Graph showing activity on coupons vs. time](image)

**Figure 3.** Activity on Coupon vs. Time

In order to assess the amount of tritium loss that occurred due to processes other than radioactive decay, Type III coupons were analyzed. Using Equation 6, the initial measurement with the SAM was taken to be the original activity \( N_0 \), and the expected amount of activity on subsequent coupons was calculated for the time since the first coupon (number 12) was measured,

\[
N(t) = N_0 e^{(-0.693 \cdot \frac{t}{t_{1/2}})}
\]  

(7)

This information is displayed in Figure 4. The measured activities are noted for each Type III coupon. Although the standard deviation for the SAM readings is relatively large, there is an apparent decrease in the slope of this data, which is greater than the slope of the graph attributed to decay. The loss due to decay is 0.5 µCi per day, while the overall loss on the coupons was 2.7 µCi per day in helium, indicating that decay was not the only process contributing to activity loss. This data would be more reliable if the same coupon was tested over a period of time.
Table 6. Activity Deposited in Bubblers, with Type I (blue), Type II (green) and Type III (yellow)

<table>
<thead>
<tr>
<th>Exp #</th>
<th>B1 Collection (mCi)</th>
<th>B3 Collection (mCi)</th>
<th>Total in all Bubblers (mCi)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.5292</td>
<td>0.0016</td>
<td>0.5542</td>
</tr>
<tr>
<td>2</td>
<td>2.3738</td>
<td>0.0924</td>
<td>2.7016</td>
</tr>
<tr>
<td>3</td>
<td>2.5810</td>
<td>0.1392</td>
<td>3.4450</td>
</tr>
<tr>
<td>4</td>
<td>3.0566</td>
<td>0.1725</td>
<td>3.9200</td>
</tr>
<tr>
<td>5</td>
<td>3.2964</td>
<td>0.1118</td>
<td>4.1805</td>
</tr>
<tr>
<td>6</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>3.4571</td>
<td>0.1226</td>
<td>4.4290</td>
</tr>
<tr>
<td>8</td>
<td>3.4870</td>
<td>0.1110</td>
<td>4.2666</td>
</tr>
<tr>
<td>9</td>
<td>0.4551</td>
<td>0.0057</td>
<td>0.4682</td>
</tr>
<tr>
<td>10</td>
<td>0.5004</td>
<td>0.0059</td>
<td>0.5568</td>
</tr>
<tr>
<td>11</td>
<td>0.4136</td>
<td>0.0076</td>
<td>0.4942</td>
</tr>
<tr>
<td>12</td>
<td>6.0527</td>
<td>0.1921</td>
<td>8.0224</td>
</tr>
<tr>
<td>13</td>
<td>5.5605</td>
<td>0.1404</td>
<td>7.6665</td>
</tr>
<tr>
<td>14</td>
<td>7.0030</td>
<td>0.1575</td>
<td>8.5970</td>
</tr>
<tr>
<td>15</td>
<td>7.6118</td>
<td>0.1709</td>
<td>9.5731</td>
</tr>
<tr>
<td>16</td>
<td>6.1831</td>
<td>0.1121</td>
<td>6.8500</td>
</tr>
<tr>
<td>17</td>
<td>4.7647</td>
<td>0.0919</td>
<td>5.6227</td>
</tr>
<tr>
<td>18</td>
<td>5.0461</td>
<td>0.0833</td>
<td>5.7476</td>
</tr>
</tbody>
</table>
III.B. Liquid Scintillation Data: As previously mentioned, the LSC aliquots were taken before each run, to determine the background activity in the bubbler, and following, to determine the amount of activity pulled off the coupon and deposited into the bubbler. The total calculated amount, as well as the amounts in B1 and B3, are displayed in Table 5. The activities of the aliquots removed from Coupon 6 were very large and surpassed the detector’s counting capacity. Thus, the measurements are not accurate and have not been included here.

The different coupon types varied in the amount of desorbed activity that was collected in the bubblers. B1 collected far more activity than B3 in every case. For Run 9, scintillation fluid leaked out of B1 for a large part of the experiment, implying that the total activity must have been greater than was measured. The total activities varied for each coupon and are graphed in Figure 5 vs. time, differentiated according to coupon type.

From these data, it is obvious that the length of time, during which a coupon has been exposed to air before running the experiment, has an effect on the data. These results are varied by the fact that the coupons were not all prepared in the same manner.
To determine the amount of HTO removed from the surface compared to the amount of HT, one can compare the sum activity in B1 and B2 to the sum activity in B3 and B4. This information is presented in Table 7 for each run, assuming that B1 and B2 collect the water soluble HTO and B3 and B4 collect the previously insoluble HT, which has been converted to HTO in the oxidation furnace.

The majority of the activity (about 97.3 %) removed from each coupon was in the form of HTO, the component that dissolves in both B1 and B3. The Type I coupons (blue) were consistently higher than the average, while the Type II coupons (green) were consistently lower.

**III.C: Correlation between SAM and LSC measurements**

The SAM is not able to measure the total amount of activity on and in the coupon, since it has sensitivity only up to a depth of about 0.8 µm into the metal. The LSC, on the other hand, measures the total amount desorbed from the coupon. Thus, these two measurements are not identical, but should be related to each other and are expected to show a positive correlation.
Table 7. Soluble (HTO) vs. insoluble (HT) components collected from each coupon.

<table>
<thead>
<tr>
<th>Exp. #</th>
<th>HTO (mCi)</th>
<th>HT (mCi)</th>
<th>Total collected in all Bubblers (mCi)</th>
<th>% HTO/Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.552</td>
<td>0.002</td>
<td>0.554</td>
<td>99.7</td>
</tr>
<tr>
<td>2</td>
<td>2.602</td>
<td>0.100</td>
<td>2.702</td>
<td>96.3</td>
</tr>
<tr>
<td>3</td>
<td>3.289</td>
<td>0.156</td>
<td>3.445</td>
<td>95.5</td>
</tr>
<tr>
<td>4</td>
<td>3.729</td>
<td>0.191</td>
<td>3.920</td>
<td>95.1</td>
</tr>
<tr>
<td>5</td>
<td>4.033</td>
<td>0.148</td>
<td>4.180</td>
<td>96.5</td>
</tr>
<tr>
<td>6</td>
<td>4.269</td>
<td>0.160</td>
<td>4.429</td>
<td>96.4</td>
</tr>
<tr>
<td>7</td>
<td>4.126</td>
<td>0.141</td>
<td>4.267</td>
<td>96.7</td>
</tr>
<tr>
<td>8</td>
<td>0.462</td>
<td>0.006</td>
<td>0.468</td>
<td>98.8</td>
</tr>
<tr>
<td>9</td>
<td>0.549</td>
<td>0.007</td>
<td>0.557</td>
<td>98.7</td>
</tr>
<tr>
<td>10</td>
<td>0.484</td>
<td>0.010</td>
<td>0.494</td>
<td>98.0</td>
</tr>
<tr>
<td>11</td>
<td>7.800</td>
<td>0.223</td>
<td>8.022</td>
<td>97.2</td>
</tr>
<tr>
<td>12</td>
<td>7.471</td>
<td>0.196</td>
<td>7.667</td>
<td>97.4</td>
</tr>
<tr>
<td>13</td>
<td>8.395</td>
<td>0.202</td>
<td>8.597</td>
<td>97.7</td>
</tr>
<tr>
<td>14</td>
<td>9.216</td>
<td>0.357</td>
<td>9.573</td>
<td>96.3</td>
</tr>
<tr>
<td>15</td>
<td>6.699</td>
<td>0.151</td>
<td>6.850</td>
<td>97.8</td>
</tr>
<tr>
<td>16</td>
<td>5.502</td>
<td>0.121</td>
<td>5.623</td>
<td>97.9</td>
</tr>
<tr>
<td>17</td>
<td>5.637</td>
<td>0.111</td>
<td>5.748</td>
<td>98.1</td>
</tr>
<tr>
<td>Average</td>
<td></td>
<td></td>
<td></td>
<td>97.3</td>
</tr>
</tbody>
</table>

Table 8. Sam and LSC activities

<table>
<thead>
<tr>
<th>Exp. #</th>
<th>Initial (mCi)</th>
<th>Total (mCi)</th>
<th>% SAM</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.39</td>
<td>0.55</td>
<td>70.98</td>
</tr>
<tr>
<td>2</td>
<td>2.04</td>
<td>2.70</td>
<td>75.51</td>
</tr>
<tr>
<td>3</td>
<td>2.05</td>
<td>3.45</td>
<td>59.42</td>
</tr>
<tr>
<td>4</td>
<td>1.95</td>
<td>3.92</td>
<td>49.65</td>
</tr>
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<td>5</td>
<td>2.23</td>
<td>4.18</td>
<td>53.31</td>
</tr>
<tr>
<td>6</td>
<td>2.11</td>
<td></td>
<td></td>
</tr>
<tr>
<td>7</td>
<td>2.12</td>
<td>4.43</td>
<td>47.97</td>
</tr>
<tr>
<td>8</td>
<td>2.08</td>
<td>4.27</td>
<td>48.86</td>
</tr>
<tr>
<td>9</td>
<td>0.29</td>
<td>0.47</td>
<td>62.90</td>
</tr>
<tr>
<td>10</td>
<td>0.34</td>
<td>0.56</td>
<td>61.86</td>
</tr>
<tr>
<td>11</td>
<td>0.30</td>
<td>0.49</td>
<td>59.80</td>
</tr>
<tr>
<td>12</td>
<td>3.31</td>
<td>8.02</td>
<td>41.28</td>
</tr>
<tr>
<td>13</td>
<td>3.28</td>
<td>7.67</td>
<td>42.77</td>
</tr>
<tr>
<td>14</td>
<td>3.23</td>
<td>8.60</td>
<td>37.58</td>
</tr>
<tr>
<td>15</td>
<td>3.17</td>
<td>9.57</td>
<td>33.13</td>
</tr>
<tr>
<td>16</td>
<td>3.03</td>
<td>6.85</td>
<td>44.19</td>
</tr>
<tr>
<td>17</td>
<td>2.93</td>
<td>5.62</td>
<td>52.17</td>
</tr>
<tr>
<td>18</td>
<td>3.10</td>
<td>5.75</td>
<td>53.90</td>
</tr>
<tr>
<td>Average, Type I (with #1 omitted)</td>
<td>61.52</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average, Type II</td>
<td>55.79</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average, Type III</td>
<td>43.57</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average, Total</td>
<td>52.66</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
The SAM was found (cf. Table 8) to measure an average of 53% of the total activity initially on the coupon. In past research on Type I coupons, the SAM was found to measure 66.50 percent of the total activity. Taking the average of only the Type I data here, one finds that 62% was removed. Because Coupon #1 was kept in a different container and had been exposed to air much longer than the remaining Type I coupons, it was not included in this average. The average for Type I coupons was larger than those of Type II or Type III, indicating larger amounts of activity on the surface of the former. This effect can be traced to the manner in which the coupons were prepared. Type I coupons were prepared at half the pressure used for Types II and III. This would indicate that, at higher pressures more tritium travels into the bulk, while at lower pressures it would remain on the surface.

**Figure 6.** Activity on coupon as measured by SAM vs total collected in all bubblers

From Figure 6, it is obvious that the type of the coupon type had a large exposure and age effect on the activities measured with the two methods (SAM, LSC) applied. Generally, the more activity the SAM measured initially on a coupon, the more activity was released during thermal desorption. However, there were deviations from this trend, therefore error bars are shown in Figure 6. The error on SAM measurements is estimated to be ±10 percent of the indicated value. The LSC has an error of ±10 percent as well. In spite of the large
margins of error within each set of coupons, there is a noticeable trend between different coupon types.

Coupon with higher tritium loading show higher SAM readings. This trend is examined again in Figure 7, where SAM measurements are normalized to the total amount of activity observed in the desorption experiments. It is obvious from Figure 7 that, with increasing tritium activity in a coupon, the SAM measures a decreasing fraction of the total activity. This observation could imply that higher tritium loading places activity below the surface layer, deeper into the metal lattice.

![Figure 7](image_url)

**Figure 7.** Measured surface activity normalized to the total desorbed activity plotted vs. total activity

### III.D: Experimental and Theoretical Water Vapor Pressures.

The humidity of the carrier gas used in a run was calculated by measuring the mass of water lost in B0 and the time which it flowed through B0. In Table 9, these experimental values are compared to the theoretical values, calculated as described previously. Overall,
the theoretical approximation, based on the equilibrium vapor pressure of water, was a very good estimate for the amount of humidity in the carrier stream.

**Table 9: Humidity of the carrier stream for each run**

<table>
<thead>
<tr>
<th>Exp. #</th>
<th>B0 Flow (sccm)</th>
<th>Run Time (min)</th>
<th>B0 Flow, actual (ppm)</th>
<th>Theoretical Flow, theoretical (ppm)</th>
<th>Actual/Theoretical</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Dry</td>
<td>1,801</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>2</td>
<td>Dry</td>
<td>1,712</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>3</td>
<td>100</td>
<td>1,660</td>
<td>spill</td>
<td>26,053</td>
<td></td>
</tr>
<tr>
<td>4</td>
<td>75</td>
<td>1,710</td>
<td>19,633</td>
<td>19,539</td>
<td>1.00</td>
</tr>
<tr>
<td>5</td>
<td>50</td>
<td>1,653</td>
<td>13,765</td>
<td>13,026</td>
<td>1.06</td>
</tr>
<tr>
<td>6</td>
<td>25</td>
<td>1,750</td>
<td>8,668</td>
<td>6,513</td>
<td>1.33</td>
</tr>
<tr>
<td>7</td>
<td>100</td>
<td>1,673</td>
<td>24,154</td>
<td>26,053</td>
<td>0.93</td>
</tr>
<tr>
<td>8</td>
<td>50</td>
<td>1,662</td>
<td>12,643</td>
<td>13,026</td>
<td>0.97</td>
</tr>
<tr>
<td>9</td>
<td>75</td>
<td>1,663</td>
<td>18,542</td>
<td>19,539</td>
<td>0.95</td>
</tr>
<tr>
<td>10</td>
<td>75</td>
<td>1,642</td>
<td>18,704</td>
<td>19,539</td>
<td>0.96</td>
</tr>
<tr>
<td>11</td>
<td>75</td>
<td>1,698</td>
<td>18,746</td>
<td>19,539</td>
<td>0.96</td>
</tr>
<tr>
<td>12</td>
<td>Dry</td>
<td>2,003</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>13</td>
<td>100</td>
<td>2,581</td>
<td>23,654</td>
<td>26,053</td>
<td>0.91</td>
</tr>
<tr>
<td>14</td>
<td>75</td>
<td>1,995</td>
<td>18,511</td>
<td>19,539</td>
<td>0.95</td>
</tr>
<tr>
<td>15</td>
<td>Dry</td>
<td>4,708</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>16</td>
<td>Dry</td>
<td>1,962</td>
<td>0</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td>17</td>
<td>50</td>
<td>1,942</td>
<td>13,253</td>
<td>13,026</td>
<td>1.02</td>
</tr>
<tr>
<td>18</td>
<td>25</td>
<td>2,081</td>
<td>6,333</td>
<td>6,513</td>
<td>0.97</td>
</tr>
</tbody>
</table>
III.E: Coupon and Oven Temperatures.

For each run, the desorption furnace was ramped up in five temperature steps, 100 °C, 200 °C, 300 °C, 400 °C and 500 °C, with a 300-minute dwell time at each temperature. For all Type III runs, a step at room temperature was added. In order to determine the actual temperature of both a coupon and the oven throughout the process, individual thermocouples were attached to either. The oven thermocouple was in contact with the quartz oven, while the coupon thermocouple was screwed onto the coupon itself. The data collected are presented in Figure 8. To reach a step in the profile, the temperature was ramped at a rate of 10 °C per minute, causing the coupon temperature to spike slightly at first, but then to settle to a steady state value around 10 °C above that measured for the oven. Because the thermocouples were at two different locations, it is not possible to determine whether heat transfer has varied within the furnace.

![Figure 8. Coupon and Oven Temperatures vs. Time](image)

III.F. β-RAM Data: The data collected by the β-RAM, which monitored the bubbler activity continuously while the coupons experienced the temperature step profiles shown above. The
data for these runs was averaged over minutes and then analyzed as a function of time. The graphs are arranged by coupon type.

**Type 1 Data.**

![Figure 9](image-url)

**Figure 9.** Activity removed of Type I coupons vs. time, corrected for the amount of activity initially in B1, for a dry run (red line), a 18,542 ppm run (blue line), a 18,704 ppm run (green line) and a 18,746 run (yellow line), with original activities as measured by SAM noted.

In Figure 9, the dry run with Coupon #1 exhibits the least amount of activity removal at lower temperatures, as expected. In previous research, it was found that the addition of humidity to the carrier stream caused an increase in activity removal. The other three runs, all at 75% humidity flow rates, display very similar profiles. For Coupon #11, B1 began dripping about half way through the experiment, leading to the drastic dip in measured activity.
Figure 10. Normalized activity removed for Type I coupons vs time, for a dry run (red line), a 18,542 ppm run (blue line), a 18,704 ppm run (green line) and a 18,746 run (yellow line), with original activities as measured by SAM noted.

The data were normalized in order to account for differences caused by the integrated amount of total activity on the coupon released in the desorption experiment. This correction caused the 75 sccm runs to be very similar to each other during most of the respective measurements, tracking the temperature step profile in a similar fashion. The "dry run" still showed a much lower integral removal. This indicates that the addition of humidity to the carrier gas increases tritium removal rates.²
Type II Data.

Figure 11. Activity removed of Type II coupons vs. time, corrected for the amount of activity initially in B1, for a dry run (black line), a 100 sccm run (red line), an 8,668 ppm run (green line), a 12,643 ppm run (purple line), a 19,633 ppm run (yellow line), a 13,765 ppm run (blue line), with original activities as measured by SAM noted.

Seven runs were completed using coupons from the second batch of sample coupons (Type II). There were some inconsistencies seen with the data. Although the fourth run, Coupon #4, was humidified, the activity removal fell under that of the dry run, disagreeing with conclusions drawn from previous experiments performed with the same setup. This could be explained by the activity on the coupon, as its SAM reading is more than 0.1 mCi less active than all other coupons in the batch. Generally, these types show much more variation in both their original activities measured on the SAM and their desorption profiles than Type I coupons.
In Figure 12, the coupon with the highest activity measured on the SAM, Coupon #5, was seen to exhibit the greatest removal at temperatures up to 400 °C. The coupon with the second highest activity, Coupon #7, had the second largest removal amount at the lower temperature steps. This coupon also had the most humidity in the stream, so it is difficult to determine how much of an effect the humidity had, because the original activity seems to be having an effect as well. The dry run exhibited more removal at 100 °C than two very wet runs, with a similar initial activity on one of the coupons. From these data, it appears as though there are many factors effecting the removal of tritium at different temperatures, which are not yet understood.
Type III Data

Figure 13. Activity removed of Type III coupons vs. time, corrected for the amount of activity initially in B1, for three dry runs (black, yellow and green lines), a 6,333 ppm run (purple line), a 13,253 ppm run (blue line), a 18,511 ppm run (orange line), a 23,654 ppm run (red line), with original activities as measured by SAM noted.

For Runs 12 through 18 shown on the graph above, a few changes were made to the experimental procedure. The coupons were kept in a helium environment for their entire lifespan, and a room temperature step was added to the temperature profile. Comparing Coupons #12 / #13, #14 / #15, and #16 / #17 / #18, it is apparent that humidity in the carrier stream had an effect on the removal. The 1 mL cell on the β-RAM was switched for a .5 mL cell following experiment #12, so those results have been scaled accordingly. The cell was changed because it appeared as though the machine was not accurately reading the high activities coming off the new set of coupons.
Figure 14. Normalized Activity Removed of Type III coupons vs time, for three dry runs (black, yellow and green lines), a 6,333 ppm run (purple line), a 13,253 ppm run (blue line), a 18,511 ppm run (orange line), a 23,654 ppm run (red line), with original activities as measured by SAM noted.

At room temperature, the normalization of these curves caused the 23,654 ppm line to increase, moving above that of the 18,511 ppm run. At this temperature, the data show a monotonic increase in activity removal as carrier stream water vapor pressure increases. However, there is a large gap between the 18,511 ppm and the 13,253 ppm runs, indicating that at some point, perhaps when the stream becomes saturated, the effectiveness of the humidity greatly increases.

At higher temperatures, the humidity trend does not hold. At 100 °C, the amount removed is greatest for the most active coupon, #12, as measured by the SAM. Although the effect of the initial activity of the coupon was not seen at room temperatures, the data seem to begin showing a dependence as soon as the coupon is heated.
IV. Summary and Conclusions.

The present set of experiments had as their goal to determine trends associated with the addition of water vapor to the carrier gas stream. In completing this project, a wide range of activity measuring instruments were employed. This led to various sets of data which provide information about the coupons, complementing the data gained from the thermal desorption measurements.

Three different batches of coupons have been charged for use in the laboratory. All three types were used in various experiments in this study. Type I coupons, which were exposed at half the pressure for a longer time, contained more activity on the surface than other coupon types, and more activity in the form of HTO. These samples were loaded in an HT environment, which could have potentially contributed to the fact that more of the activity removed was the water soluble form of HTO. The lower pressure of this loading process would indicate that more activity was deposited onto the surface, with less traveling into the bulk. This was confirmed by the SAM readings, which were highest for the Type 1 coupons. Another factor at work here was the fact that a large amount of time passed before these coupons were tested. This could also have caused more of the activity to travel to the surface from inner layers of the material.

The analysis of the effect of radioactive decay of Type III samples seems to indicate that another process was occurring in addition to the normal radioactive decay of tritium. Such additional processes could include desorption, relating to a bonding of molecules to the surface via Van der Waal’s forces. Isotopic exchange constitutes another competing process, in which water molecules replace protons with tritons, without actually bonding to the surface of the metal sample.

Overall, this study showed some unexplained behavior in some of the runs, some of which could have been caused by the loading differences for the coupons, as differentiated by type or initial activity measured by the SAM. The SAM measurements indicated that the amounts of surface activity as a fraction of the total activity varied from coupon to coupon. As Type II thermal desorption data exhibit, the relative amount of initial activity on a coupon affects the results of all subsequent measurements.

A number of further experiments are necessary to obtain more information about the desorption of tritium from various metals. Performing SAM measurements on the same coupon would be a more accurate means of obtaining a comparison between the amount of tritium that was expected to decay, and the amount that was actually lost, similar to the analysis shown in Figure 4. In the present study, only one SAM measurement was taken per coupon, and the activity of the coupon at “time zero” could only be estimated. If only one coupon was used, the activity at time zero would be a known value. Additional thermal desorption experiments are necessary to elucidate the somewhat chaotic nature of the runs that have been performed. Finally, experiments with B0 flows in between 75 and 50 would be very interesting, in order to investigate the nature of the gap seen in Type III coupons between the two humidity levels.
Acknowledgements

This work was supported by the U.S. Department of Energy Office of Nuclear Physics (Grant No. DE-FG02-88ER40414), the University of Rochester, and the New York State Energy Research and Development Authority.

References


Tritium Removal from Stainless Steel Samples by Plasma Induced Sputtering


Departments of Chemistry and Physics, and Laboratory for Laser Energetics
University of Rochester, Rochester, NY 14627

Abstract

The efficiency of a plasma induced sputtering method has been explored in tritium decontamination of various types of stainless-steel samples previously exposed to a tritiated atmosphere. Effects of diffusion of $^3$H from the bulk lattice and subsequent surface desorption have been found to explain the long-term $^3$H removal from samples. The influence of the surface finishing of the samples was found to be small. Several thermal influences of the plasma on $^3$H removal need to be modeled quantitatively.

Introduction

Tritium ($^3$H) is used for a variety of different purposes. Some of the most interesting applications of this radioactive isotope of hydrogen occur in energy-related areas. Most prominently, $^3$H is a fuel component in nuclear fusion reactors, but is also produced in standard nuclear fission reactors. In the latter reactors, $^3$H is produced in the decay of highly excited fission fragments and, to a lesser extent, by neutron capture in the moderator. In each of these cases, the $^3$H produced is capable of attaching to the walls of the reactor vessel or to the instruments and tools used in reactor operations. The isotope is also used in the pharmaceutical and other industries. Finally, $^3$H as a “tagged” hydrogen isotope can provide a tool to study containment and transport of regular hydrogen, which is important for an investigation of the viability of a “hydrogen economy” in advanced nations. In fact, it is very difficult to contain hydrogen (or $^3$H) in a vessel due to its small atomic or molecular size enabling such gases to diffuse into the bulk of, and even penetrate the materials of containment or conduct. On a different level, since tritium is an expensive biologically hazardous, it is important to find efficient methods to decontaminate and even recover lost $^3$H. Since stainless steel is among the most popular materials used in tritium handling, the present study concentrates on a novel method to remove $^3$H from surfaces of stainless steel samples.

With this motivation, several stainless steel (SS 316) samples, initially loaded with tritium, were subjected to a low-pressure, RF-driven plasma. This process is expected to provide a potentially efficient method to remove surface layers of $^3$H based on sputtering induced by the heavy gas ions colliding with the sample surface. Direct removal by sputtering is not expected
for any $T$ components that reside in the bulk lattice of the material. However, as a by product of the sputtering process, the sample also heats up, an effect that promotes migration of $T$ from the metal lattice to the surface and subsequent thermal surface desorption. Therefore, the time dependent temperature profile of a sample exposed to the plasma has been of interest to the present study. In addition to the experimental work, some calculations were performed, in order to characterize the plasma conditions. This was done with the goal of predicting the amount of tritium removed from the surface via ion sputtering and thermal desorption.

**Experimental Setup**

Schematics of the experimental setup are shown in Fig. 1. For each exposure to the plasma, the samples were placed inside a 2.592 L quartz vacuum chamber, attaching them to a clip connected to a wire fed through the steel lid of the chamber. The clip and wire were isolated from ground and connected to a voltage and a current meter read out by a National Instruments module crate and data acquisition system. The temperature was monitored by an Omega Type-K thermocouple which was fed through the chamber lid, isolated from ground using either a Teflon© or a nylon feed-through of the upper lid of the chamber. The tip of the thermocouple was placed in tight contact with the sample and provided the temperature data.

![Figure 1: Schematics of experimental setup](image-url)
Data acquisition and monitoring was done with a PC running National Instruments LabView software written by the group.

An *Alcatel CFV 100* turbo-molecular pump backed by an *Edwards* model *ESDP 12* roughing pump was used to evacuate the chamber. The vacuum system was monitored with vacuum gauges located at the top and bottom of the chamber. In operation, the chamber was filled with 60 m torr of gas supplied from a tank and controlled using an *MKS Type 246* flow control. An *ACG-3 ENI* power supply and a *Comdel model CPM-1000M* impedance match were used to supply RF power to a copper coil around the chamber. This produced the RF power needed to ignite the gas and sustain the plasma.

<table>
<thead>
<tr>
<th>Date</th>
<th>Coupon Number</th>
<th>Finish</th>
<th>Exposed to Air</th>
<th>Peak Activity</th>
<th>Exposure Time</th>
<th>Plasma Gas</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>8/4/09</td>
<td>#36</td>
<td>Mirror</td>
<td>He, then air</td>
<td>1.72 nCi</td>
<td>420 min</td>
<td>Helium</td>
<td>10 sec RF field prior to decon.</td>
</tr>
<tr>
<td>6/3/09</td>
<td>#35</td>
<td>Mirror</td>
<td>He, then air</td>
<td>1.98 nCi</td>
<td>317.33 min</td>
<td>Argon</td>
<td></td>
</tr>
<tr>
<td>2/12/09</td>
<td>#34</td>
<td>Mill</td>
<td>He, then air</td>
<td>2.37 nCi</td>
<td>n/a</td>
<td>n/a</td>
<td>No plasma exposition</td>
</tr>
<tr>
<td>1/29/09</td>
<td>#33</td>
<td>Mill</td>
<td>He, then air</td>
<td>2.58 nCi</td>
<td>331.7 min</td>
<td>Argon</td>
<td>A ≈ const after ≈1 mo.</td>
</tr>
</tbody>
</table>

**Table 1: Summary history of sample coupons**

The above table is a summary of the properties and history of the coupons used in the study. All coupons in Table 1 were initially loaded with tritium at the same time and in the same manner. The dates listed in the first column note the first exposure to the plasma. Later samples were kept under an inert helium atmosphere until first exposure. Two of the coupons used had a different stainless steel finish. The main difference associated with different coupon finishes is the surface roughness: mirror finish has a smoother surface than mill finish.

Peak activity was the activity of a sample measured with the SAM before a first exposure to the plasma. Coupon #36 had been previously exposed for 10 seconds to an RF field in vacuum. After that, the coupon was left exposed to air for 47 days before the decontamination sequence was started. The peak activity assumed was the activity measured prior to the first exposure to the plasma. Total exposure time represents the cumulative amount of time during which the coupons were exposed to the plasma. Finally, a different gas, helium, was used for the decontamination of Coupon #36. Coupon #34 was not exposed to the plasma but was left exposed to air for test purposes.
**Experimental Results**

Experimental values obtained in the activity measurements for the samples listed in Table 1 are displayed in Figs. 2 and 3. In Fig. 2, the surface activity measured with the SAM is plotted for each sample vs. the cumulative plasma exposure time. Each point represents a SAM measurement after one in a series of exposures. These data were normalized to the initial activity recorded for each sample (Table 1) and then graphed on a double-log plot.

The plot in Fig. 2 indicates that the removal of tritium does not depend significantly on the finish on the coupon. Also, comparing the data for Coupons #35 and #36 suggests that tritium is removed at a slower rate in a helium plasma than in an argon plasma. However, this difference does not appear to be very significant.

![Figure 2: Normalized activity vs. cumulative exposure time](image)

To assess the spontaneous desorption of tritium from the samples, Coupon #34 was left exposed to air for an extended period of time and its activity measured periodically with the SAM. The test was intended to measure the tritium loss through isotropic exchange with molecular hydrogen as well as water vapor, in addition to the trivial spontaneous $T$ decay. Using the known $T$ decay rate, the latter time dependence should be given by

$$N(t) = 91.890 \times e^{-0.00159 \times t} \text{ MBq}$$

Here, $N(t)$ denotes the surface activity of the sample and $t$ the time (in days) during which the sample was left exposed to air.

Figure 3 displays the data obtained for the surface activity of the coupon left on air for a long term data. Obviously, the activity loss is relatively small. An analysis of the resulting data...
indicates that an exponential function can fit the time dependence of the surface activity relatively well. The error in the data was found experimentally to be 0.5%. However, this function shows a somewhat faster decay than is predicted by the natural radioactive decay of tritium alone. This indicates a small effect of the ambient air on the surface activity, if coupons are handled in air.

In order to determine the rates of heat transfer and heat loss, the heating and cooling curves were measured for an unloaded stainless steel “calibration coupon” (radius: 1.1cm, height: 5.1cm) which was placed in argon and helium plasmas. The coupon was left exposed to the plasma until an equilibrium temperature was reached. After allowing the coupon to remain in steady state for awhile, the RF field was turned off, and the coupon was allowed to cool under a 60 m torr flow of the gas being used for the plasma. It was found that the rate of heat transfer to the coupon under is much faster for helium than for argon. Also, for He the steady state temperature of the coupon is approximately 150°C higher than for Ar. Once the RF field was turned off, the coupon took approximately 100 minutes to cool back down to room temperature, regardless of gas specie. This behavior implies that the cooling rate is also greater for a He stream.

**Summary and Outlook**

The goal is to develop a quantitative model for $T$ removal by ion sputtering combined with and thermal desorption. There are several issues that need to be addressed in order to achieve this goal:

1. It is likely that the samples in the plasma are heated mainly through ion impact. Knowledge of the speeds and overall mean free path of the ions in the plasma will allow one determine their energy prior to entering the plasma sheath, where further acceleration occurs.
2. Results of tentative calculations, as well as experiments with helium and argon plasmas suggest that there is a characteristic dependence on the mass of the gas being used.

3. Experimental decontamination sequences for different gas species and different pressures should therefore help characterizing the plasma sputtering conditions.

4. From known amounts of energy transferred from the RF generator to carrier gas and coupon, reliable predictions of the time dependent profile and the asymptotic temperature attained by the sample should be made.

Reference

Ambient and Neutron-Induced Background Gamma Radiation
in the UR Nuclear Science Laboratory

E. Pollock, I. Pawelczak, J. Tőke, and W. U. Schröder
Departments of Chemistry and Physics, University of Rochester
Rochester, NY 14627

Abstract:

Measurements in the laboratory of the University of Rochester Nuclear Science Group typically show significant intensities of background gamma radiation. The present experiments aimed at an identification of the sources of these peaks and an understanding of the spectral shapes of this background in the energy spectra taken with different gamma detectors. So far, such background analyses have been performed with a 1.5” x 1.5" LaBr₃:Ce scintillation detector (Saint-Gobain, BrilLanCe®-380) and a 3”x3” NaI(Tl) detector. These detectors provided photo peak energy resolutions of 3% and 7% at 0.662 MeV, respectively. Both detectors recorded background gamma radiation expected from the decay of the radioisotopes ⁴⁰K, ²³⁸U and ²³²Th occurring naturally in the concrete walls of the laboratory, as well as activation radiation caused by a strong ²⁴¹Am/⁹Be neutron source in its shielding materials. In addition, with the BrilLanCe®-380 detector background gamma lines were observed in the 1.5 – 3.0 MeV range, as well as a broad structure between 0.8 and 1 MeV. Following other works these peaks are attributed to ²²⁷Ac and ¹³⁸La radioisotope contaminations of the BrilLanCe®-380 detector material.

1. Introduction:

To be able to perform more specific neutron-gamma coincidence measurements than possible with available fast NaI(Tl) detectors, our lab recently purchased a LaBr₃:Ce, 1.5” x 1.5” BrilLanCe®-380 scintillation gamma detector manufactured by Saint-Gobain Crystals. As a fast inorganic scintillator, LaBr₃:Ce crystals provide a fast response with high stopping efficiency and high light output. The fast principal decay constant (~30 ns) of this scintillator has allowed timing resolutions as good as 260 ps to be reached. The relatively good photo peak resolution of < 3% typically obtained at 0.662 MeV is a consequence of the high light output (~61,000 photons per MeV) of the LaBr₃:Ce scintillator [Loe01,Sha03, Dor04, Gon05]. For comparison, the more common NaI(Tl) scintillators have poorer resolutions, not better than 6-7% at 0.662 MeV, and hence poorer signal-to-background ratios. Nevertheless, gamma background radiation remains a potential problem also with high-resolution LaBr₃:Ce detectors and needs to be understood in detail. This knowledge is important for the preparation of a “beam” of tagged neutrons in the lab utilizing characteristic γ-rays associated with a specific neutron channel in the decay chain of a certain radio-isotope. As a practical example, one is interested in the decay $^{13}\text{C}^* \rightarrow ^{12}\text{C} + n + \gamma$. 
Thus, to study the background radiation in our lab, we chose to use the BrillanCe®-380 LaBr₃:Ce scintillator which would allow us to identify the background peaks more precisely than with the NaI(Tl) detector. Since the measurements were taken in the room housing a ²⁴¹Am-⁹Be neutron source shielded by water and paraffin, we expected to see γ-rays associated with the neutron source, such as the γ-ray from the decay $^{13}\text{C} \rightarrow ^{12}\text{C} + n + \gamma$ reaction. In addition, we suspected that the walls, specifically the concrete in the walls, were also contributing to the background radiation we observed. It was presumed that the concrete in the walls contained the radioisotopes $^{40}\text{K}$, $^{238}\text{U}$ and $^{232}\text{Th}$ and that these respective isotopes were contributing to the overall background observed in the lab.

However, due to the $^{227}\text{Ac}$ contamination and the internal radiation due to the $^{138}\text{La}$ isotope, we were unable to take full advantage of the good resolution LaBr₃:Ce offers in measurements of the wall background. A measurement with a Harshaw 3” x 3” NaI(Tl) scintillator was then performed to identify the peaks observed in LaBr₃:Ce due to internal contamination by radioisotopes. The measurement of the wall taken with the NaI(Tl) detector also offers a spectrum of γ-rays emitted from the wall that was analyzed to verify that the observed peaks originating from the $^{40}\text{K}$, $^{238}\text{U}$, and $^{232}\text{Th}$. A comparison of the resolution was made between the LaBr₃:Ce and NaI(Tl) detectors and was found to be in agreement with other works.

2. Properties of BrillanCe®-380 Detector

A. LaBr₃:Ce comparison with the NaI(Tl) detector and other available detectors

As previously mentioned, the superior resolution of the LaBr₃:Ce detector compared to the NaI(Tl) detector is a result of LaBr₃:Ce’s high light output (~61,000 photons per MeV), high stopping efficiency, fast principal decay constant (~30 ns), and timing resolution as good as 260 ps [Loe01,Sha03, Dor04, Gon05]. As a result of its fast response time, studies [Mil05c] show that LaBr₃:Ce should enable the experimenter to find peaks up to three times faster than NaI(Tl) while also resolving peaks that a NaI(Tl) detector is unable to separate. In addition, while the LaBr₃:Ce crystal retains its linearity at low

Figure 1 – (a) Saint-Gobain Crystals BrillanCe®-380 detector coupled to Hamamatsu R6251 PMT and (b) NaI(Tl) detector. (Detectors not shown to same scale.)
electron-equivalent energies (~100 – 200 keV) and shows very good light-yield proportionality, NaI(Tl) produces more light at low energies than LaBr$_3$:Ce, due to a non-proportionality in NaI(Tl)’s response [Mil05c, Sai07].

Another difference between the LaBr$_3$:Ce and NaI(Tl) detectors worth addressing is the difference in cost and availability. NaI(Tl) detectors are available in a large array of crystal sizes, while LaBr$_3$:Ce is limited to 1.5” x 1.5” up to 3” x 3” crystals due to the difficulty of growing the crystals and maintaining the desirable resolution with larger crystals [Mil05c, Sai07]. In addition, the cost of BrillLanCe®-380 is much greater than the cost of a NaI(Tl) crystal.

Thus far the comparison of LaBr$_3$:Ce has primarily been made with the NaI(Tl) detector. The superior properties LaBr$_3$:Ce offers compared to NaI(Tl) detectors suggest that experiments previously made with NaI(Tl) detectors may be replaced with the higher-resolution LaBr$_3$:Ce detectors. However, one area of caution in making this switch of detectors, aside from the cost, is the internal radiation of $^{138}$La and contamination of $^{227}$Ac in LaBr$_3$:Ce . Nonetheless, it is important to note that this internal radiation and contamination observed in BrillLanCe®-380 is not limited to the LaBr$_3$:Ce crystal.

Many detectors on the market have internal radioisotopes and contaminations [Mil05a]. For example, lutetium is another lanthanide that shows promising scintillation properties, however, like lanthanum, lutetium also has a radioisotope, $^{176}$Lu, that would cause internal peaks. Furthermore, while $^{176}$Lu has a shorter half-life ($3.78 \times 10^{10}$ years) than $^{138}$La, $^{176}$Lu represents 2.5% of the natural abundance of lutetium where as $^{138}$La only represent 0.09% of the natural abundance of lanthanum. Thus $^{176}$Lu may pose more of a problem than the $^{138}$La isotope. Gadollinium is another lanthanide with a naturally-occurring radioisotope, $^{152}$Gd, however, while the half-life is $1.08 \times 10^{14}$ years its natural abundance is only 0.02%.

Similarly, the contamination of $^{227}$Ac in LaBr$_3$:Ce is not limited to this crystal. BaF$_2$ scintillators are also contaminated by $^{226}$Ra, which, similar to the actinium-lanthanum relationship, is also part of the $^{235}$U decay chain and is located below barium on the periodic table. In addition, the $^{207}$Bi contamination found in BGO high-Z scintillators produces internal x-rays and γ-rays in the spectra as well.

Thus, the internal radiation of $^{138}$La and contamination of $^{227}$Ac in LaBr$_3$:Ce present application problems that are clearly not limited to the LaBr$_3$:Ce scintillator. Yet while these issues persist, specifically, while the contamination levels of $^{227}$Ac remain as high as they currently are, certain applications may be better suited with a different detector. However, sacrificing resolution or internal radioisotopes and contamination depends upon the type of measurements desired.

B. Internal radiation of $^{138}$La and contamination of $^{227}$Ac in BrillLanCe®-380:

While LaBr$_3$:Ce detectors offer superior resolution to NaI(Tl) detectors, as previously mentioned the drawbacks include internal radiation due to the $^{138}$La isotope and contamination of $^{227}$Ac. $^{138}$La has a half-life of $1.06 \times 10^{11}$ years and constitutes 0.09% of the natural abundance of lanthanum [Led67]. As seen in Figure 2 [Nuc00], $^{138}$La produces two γ-rays following beta decay and electron capture. $^{138}$La undergoes beta decay (33.6%) to stable $^{138}$Ce, emitting a 0.779 MeV γ-ray and a beta continuum up to 0.255 MeV thus producing the broad feature from 0.789 MeV to around 1 MeV [Led67, Mil06a, Mil05b].
$^{138}$La also emits a 1.436 MeV $\gamma$-ray following electron capture (33.4%) to stable $^{138}$Ba, and $^{138}$Ba K X-rays between 31 and 38 keV are emitted from excited $^{138}$Ba atoms. In combination, the 1.436 MeV $\gamma$-rays and the 31 – 38 keV K-X-rays from excited $^{138}$Ba produce a broad peak in the energy spectrum around 1.46 MeV. This 1.46 MeV peak also makes resolving the 1.461 MeV $^{40}$K peak difficult to achieve [Mil05a, Mil05b, Mil05c, Mil06a, Mil06b].

In addition to the natural radioisotope of lanthanum, $^{227}$Ac contamination produces five broad peaks between 1.5 – 3.0 MeV. $^{227}$Ac has a half-life of approximately 22 years and is part of the $^{235}$U decay chain, producing five alpha particles in its decay to stable $^{207}$Pb [Led67]. Because alpha-particles are heavy particles which produce less scintillation light in the LaBr$_3$:Ce scintillator than gammas of the same deposited energy, alpha-particles of energies in the range of 5.0 and 7.4 MeV are observed in the region between 1.5 and 3.0 MeV of electron-equivalent energy [Mil05b]. Furthermore, since actinium is located directly below lanthanum on the periodic table, actinium has similar properties to lanthanum and is therefore difficult to separate from lanthanum.

3. Experimental Set-up

Two sets of three measurements were taken with a 1.5” x 1.5” LaBr$_3$:Ce Saint-Gobain Crystals BrillanCe®-380 scintillator coupled to a 2” diameter Hamamatsu R6231 photomultiplier tube (PMT) operated at +550 V. Each measurement was taken for six hours. The first measurement was taken right next to the neutron source, the second measurement was taken 10 feet away from the neutron source and 5 feet from the wall, and the third was taken 20 feet away from the neutron source, directly facing the wall.

For the comparison study with the NaI(Tl) spectrum, a Hanshaw 3” x 3” NaI(Tl) scintillator was used with +1985 V supplied to the photomultiplier tube. One measurement was taken for six hours, 20 feet from the neutron source, directly facing the wall.

The anode signals from the LaBr$_3$:Ce or NaI(Tl) PMT were shaped and amplified using a LeCroy 612 Amplifier, digitized by LeCroy 2249A ADC, and further processed using the lab’s EZDAQ CAMAC data acquisition system. Timing and gate signals were produced using a Philips Scientific 711 discriminator.
Light output spectra were recorded with a resolution of 2048 channels and analyzed with PAW and ROOT software packages.

For each set of measurements, the experimental set-up was calibrated in gamma (electron-equivalent) energy using three radio-active sources: $^{22}$Na (1.275 MeV full-energy peak and 0.511 MeV SE peak), $^{60}$Co (full-energy peaks of 1.173 and 1.332 MeV) and the $^{137}$Cs (full-energy peak of 0.6616 MeV) [Led67]. Calibration measurements were performed both before and after a set of experiments.

4. Results and Discussion

A. Background radiation from the paraffin and water shielding housing and the $^{241}$Am-$^9$Be neutron source

The $^{241}$Am-$^9$Be neutron source, stored in a water and paraffin housing container in the back of the UR Nuclear Science Laboratory, has a half-life of 432.7 years [Asg04]. From the $^9$Be($\alpha$,n)$^{12}$C reaction an excited $^{13}$C nucleus is created:

$$\alpha + ^9\text{Be} \rightarrow ^{13}\text{C}^*,$$

which then decays via four channels to stable $^{12}$C by emitting various gammas and neutrons [Asg04]. The $^{241}$Am-$^9$Be reaction mostly leads to the ground state and first excited state. Population of this first excited state is associated with the emission of a neutron and a 4.438 $\gamma$-ray [Asg04]. The 4.438 $\gamma$-ray leads to a single escape (SE) and double escape (DE) peak at 3.927 MeV and 3.416 MeV, respectively.

In addition, since the $^{241}$Am-$^9$Be neutron source is stored in a water and paraffin housing container, neutrons are also captured by hydrogen in the water and paraffin. The hydrogen-neutron capture reaction:

$$^1\text{H} + \text{n} \rightarrow ^2\text{H} + \gamma (2.23 \text{ MeV}),$$

results in a 2.23 MeV full energy peak and its associated SE peak at 1.72 MeV.

Figures 3 (a) and (b) show the spectra obtained from six hour measurements with the BrilLanCe®-380 detector. In Figure 3 (a) the dark spectrum ('neutron shielding') was taken right next to the neutron source, while the light spectrum ('Background') was taken near the wall. Figure 3 (b) shows the difference of the two spectra (the light spectrum subtracted from the dark spectrum). In Figure 3 (b) most of the internal peaks associated with the LaBr$_3$:Ce crystal, (see section 2 A) are eliminated by subtracting the ‘background’ measurement from the ‘neutron shielding’ measurement. This leaves a spectrum in Figure 3 (b) of the $\gamma$-peaks associated with the neutron source and shielding, which are labeled for convenience.
Thus, as seen in Figures 3 (a) and (b), the 4.438 MeV γ-ray from the decay of $^{13}$C* is clearly observed.
in the measurement taken near the $^{241}$Am-$^{9}$Be neutron source housing (the dark spectrum – Fig. 3 (a)). The associated SE and DE peaks of the 4.438 MeV γ-ray are seen at 3.927 MeV and 3.416 MeV, respectively (see Fig. 3 (b)). In addition, the 2.23 MeV full-energy peak from the hydrogen-neutron capture reaction, along with its associated SE peak at 1.72 MeV are also observed in Figure 3.

As mentioned in the Experimental Set-up, three measurements were taken with Brilliance®-380: (1) near the neutron source, (2) 10 feet from the neutron source, five feet from the wall, and (3) 20 feet from the neutron source, near the wall. When the second and third spectra were compared, the only significant difference between the spectra was a decreased count rate of the 4.438 and 2.23 MeV γ-rays and their associated SE and DE peaks (for the case of the 4.438 MeV γ-ray). The decrease in counting rate from measurement (2) to measurement (3) was consistent with the change in solid angle and distance from the neutron source.

It is important to mention that the PMT and/or counting electronics showed a time dependent gain drift. Of the two spectra displayed in Figure 3 (a), the spectrum labeled “Background” was taken with the Brilliance®-380 detector at a later time. It has clearly shifted slightly to higher energies, as compared to the earlier (“neutron shielding”) data. The effect also results in a broadening of the lines. For example, notice that the 4.438 MeV γ line due to the neutron source is actually located at ~4.5 MeV on the channel-energy calibrated graph in Figure 3 (a). A similar gain shift has been reported by others [Mil05c]. Nevertheless, the 4.438 and 2.23 MeV γ lines and associated SE and DE peaks (for the case of the 4.438 MeV γ-ray) are positively identified. As the detector is moved further away from the neutron source, the peaks associated with this source decrease in proportion with the solid angle.

### B. Background radiation from concrete

In addition to the $^{241}$Am-$^{9}$Be neutron source radiation, naturally occurring radioisotopes including $^{40}$K and the γ-rays from the $^{238}$U and $^{232}$Th decay chains found in the concrete of the walls were also contributing to the background in the lab. Literature energies of the γ-rays from these sources are listed below in Table 1 [Gup77]. These data have been obtained from measurements using both NaI(Tl) and Ge(Li) detectors. One of the NaI(Tl) measurements was taken near the wall, the other at a distance of 15 cm from it. To obtain meaningful spectra of γ-rays from the walls the NaI(Tl) detector was surrounded by lead shielding, leaving an area fit to the dimensions of the detector open to the wall. Gupta found that increasing the distance from the wall decreased the count rate by a factor consistent with the change in solid angle, identifying that the γ-rays originated from the walls. The NaI(Tl) detector measurements found peaks at 0.61 MeV, 0.92 MeV, 1.46 MeV, 1.74 MeV, 2.15 MeV, 2.25 MeV and 2.61 MeV (see Table 1 for an explanations). Since the NaI(Tl) detector cannot resolve many of the peaks due to the $^{238}$U and $^{232}$Th decay chains, Gupta performed two more 24-hour measurements with Ge(Li) detectors and was able to resolve and distinguish the many gamma lines emitted in the $^{238}$U and $^{232}$Th decay chains (See Fig. 4).
<table>
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</table>

¹From Ref. 4.

²U stands for activity in the decay chain of $^{238}\text{U}$; T stands for activity in the decay chain of $^{232}\text{Th}$.
As seen in Figure 5, the spectrum obtained with a six hour measurement next to the wall of the UR Nuclear Science Laboratory with a NaI(Tl) detector shows a broad peak between ~0.7 to 1.1 MeV, and peaks at approximately 1.46 MeV and 2.15 MeV. There may be peaks at approximately 0.6 MeV and in the region between 2.1 and 3 MeV, but longer measurement are needed to resolve these peaks and draw conclusions as to their sources. In addition to performing a longer measurement, including led shielding around the NaI(Tl) detector as done in Gupta’s measurements of the wall may also decrease any interfering background radiation and produce spectra with more defined gamma-peaks.

Figure 4 — Gamma-ray spectra taken with a 100-cm³ coaxial Ge(Li) detector, in two different rooms. The peaks at 1.17 and 1.33 MeV are from the calibration source $^{60}$Co. [Gup77]
Nevertheless, as indicated in Table 1 and the data presented by Gupta, (Gupta observed peaks in the NaI(Tl) measurement at 0.61, 0.92, 1.46, 1.74, 2.15, 2.25 and 2.61 MeV), these peaks seem to arise from

\[ ^{40}K \] and the \( \gamma \)-rays from the \( ^{238}U \) and \( ^{232}Th \) decay chains found in the walls (see Table 2 for a summary and comparison of these results).

![NaI(Tl) Background Spectrum](image)

**Figure 5** — Six hour NaI(Tl) spectrum taken next to the wall

<table>
<thead>
<tr>
<th>Table 2</th>
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<tbody>
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<td><strong>Comparison of Gupta’s Measurement [24 hours, lead shielding] vs. 6-hour no shielding measurement taken with NaI(Tl) detectors</strong></td>
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<td>peaks observed in 6-hour NaI(Tl) measurement (energy in MeV)</td>
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<td>possible peak at 0.6</td>
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<td>broad peak between 0.7 - 1.1 MeV</td>
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<td>2.61</td>
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C. Effect Internal Radiation and $^{227}$Ac in BrillLanCe®-380 has upon measurements

As seen in the light spectrum of Figure 3 (a), there are five broad peaks between 1.5 – 3.0 MeV, as well as a broad feature from 0.789 MeV to around 1 MeV. The broad feature from 0.789 MeV to around 1 MeV is a result of the internal radiation of $^{138}$La, while the peaks in the 1.5 – 3.0 MeV energy range is a result of $^{227}$Ac contamination in LaBr$_3$:Ce.

Thus, although improvements have been made to decrease the contamination levels due to $^{227}$Ac by up to two orders of magnitude ($1.3 \times 10^{-13}$ $^{227}$A atoms/La atom [Milb05b]) since BrillLanCe®-380 first came to the market, $^{227}$Ac contamination levels are still high enough to produce the five broad peaks between 1.5 and 3.0 MeV. Since the count rate from the walls is lower than the count rate of internal $^{227}$Ac contamination we were unable to identify gamma peaks associated with the $^{238}$U and $^{232}$Th decay chains in the $^{227}$Ac contaminated energy region.

Furthermore, although we did see peaks between 0.250 and 0.650 MeV (such as the 0.609 MeV and 0.352 MeV γ-rays from the $^{235}$U decay chain – see Table 1) the internal radioisotope $^{138}$La makes it a very tedious process to determine the precise intensity of 1.461 MeV γ-rays emitted from $^{40}$K. Calculations have suggested that approximately 80% of the peak at the 1.46 position are due to $^{138}$La, while 20% are due to $^{40}$K measurements [Mil05b], however, this result may not apply for all situations.

Therefore, our initial hopes to use BrillLanCe®-380 for the measurements of the gamma activity of the wall were somewhat unsuccessful. We cannot confidently conclude anything about the 1.5 – 3.0 MeV region due to the internal contamination of LaBr$_3$:Ce, and it is a tedious process to determine the intensity of 1.461 MeV γ-rays emitted from $^{40}$K versus the contribution to this peak from $^{138}$La. This is the main reason why we observed little difference between the spectra taken close to the wall and the spectra taken ten feet away from the wall. As previously mentioned, the difference observed in the spectra taken close to the wall and ten feet away was a decrease in counts due to the neutron source in order with the change in solid angle and distance from the source.

D. Comparison of BrillLanCe®-380 and NaI(Tl) Measurements:

In agreement with other findings, the best resolution obtained with the BrillLanCe®-380 detector was 2.7% at the 0.662 MeV $^{137}$Cs γ-ray, compared to 7% for NaI(Tl) detector. However, although LaBr$_3$:Ce offers a superior resolution to NaI(Tl), the internal radioactivity of $^{138}$La and the contamination of $^{227}$Ac in BrillLanCe®-380 and other lanthanum halide scintillators render lanthanum halide scintillators such as LaBr$_3$:Ce inadequate for certain measurements and experiments. As previously discussed, the $^{138}$La internal 1.436 MeV γ-ray and the 31 – 38 keV $^{138}$Ba K-x-rays produce a broad peak around 1.46 MeV which makes it difficult to determine the presence and abundance of $^{40}$K. In addition, the $^{138}$La Beta-decay produces a 0.779 MeV γ-ray and a beta continuum up to 0.255 MeV. The $^{227}$Ac contamination of LaBr$_3$:Ce produces five peaks within 1.5- 3.0 MeV from the alpha’s emitted in the $^{227}$Ac decay to stable lead. Consequently, when the count rate of interest is lower than the count rate of the $^{227}$Ac contamination it is very difficult to draw conclusions as to the sources and energies of these counts in the contaminated energy region. Thus, although NaI(Tl) detectors have a lower resolution, since NaI(Tl)
detectors do not have internal contamination or natural radioisotopes, the NaI(Tl) detector may be a preferable detector for these types of measurements.

It is also important to note that the comparison of the resolutions made between the 1.5” x 1.5” LaBr₃:Ce scintillator and the 3” x 3” NaI(Tl) scintillator in this study is not soundly justified. Although previous labs reported similar findings when comparing the LaBr₃:Ce and NaI(Tl) detectors, their comparison studies included measurements with LaBr₃:Ce and NaI(Tl) scintillators of similar sizes and masses [Mil05c, Mil06c]. In this study, the LaBr₃:Ce and NaI(Tl) detectors differ in size and mass. That is, given the densities of the LaBr₃:Ce and NaI(Tl) detectors (5.08 g/cm³ and 3.67 g/cm³, respectively) the masses of the scintillators are 221 g and 1275 g respectively, which suggests that a direct comparison between these detectors may not be completely justified since as the detector size changes, the optical path changes and thus the properties of the respective scintillators change [Ilt06, Mil05c, Sai08]. Nevertheless, for the purposes of this study, which included identifying the background radiation observed in the lab, this comparison suggests the superior resolution of LaBr₃:Ce compared to NaI(Tl) and validates the unknown peaks in BrillLanCe®-380 as internal contamination and radioisotopes found in the LaBr₃:Ce detector, but not observed in the NaI(Tl) scintillator.

6. Conclusion:

A set of experiments with the BrillLanCe®-380 and NaI(Tl) detectors were performed to compare the performance of the two detector types. A specific task was to determine the sources of the intense background gamma radiation typically observed in the UR Nuclear Science Laboratory. The radiation was found to be a combination of γ-ray products from the ²⁴¹Am-⁹Be neutron source and water shielding housing and the naturally occurring radioisotopes ⁴⁰K and the γ-rays from the ²³⁸U and ²³²Th decay chains found in concrete in the walls. Analysis of BrillLanCe®-380 in comparison to NaI(Tl) spectra verified the internal contamination and radioisotopes, ²²⁷Ac and ¹³⁸La, respectively, inherit to the BrillLanCe®-380 detector. As a result of the ²²⁷Ac contamination and ¹³⁸La natural radioisotope in the LaBr₃:Ce crystal, it is difficult to identify background radiation from the walls with the LaBr₃:Ce detector. However, the NaI(Tl) measurement taken near the wall indicates the presence of ⁴⁰K and the γ-rays from the ²³⁸U and ²³²Th decay chains. Further measurements are needed to conclusively determine that the sources of these identified peaks are the walls of the lab.

References


Development of the University of Rochester Advanced Nuclear Science Education Laboratory ANSEL

W. U. Schröder and F.L.H. Wolfs

Departments of Chemistry and of Physics and Astronomy

University of Rochester, Rochester, NY 14627

Abstract

Student training in the Rochester nuclear science research group has benefited from the development of experiments for the new UR nuclear laboratory course ANSEL. The development uses group equipment and some resources but most importantly provides training opportunities for graduate students in fundamental technical skills related to nuclear science experimentation. It exposes them to a number of modern experimental methods used in applied fields of imaging and nuclear forensics but also in scattering and detection of charged particles and neutrons.

I. Introduction

In spite of an increased societal relevance and potential of nuclear science and technology, there has been a declining efficacy in the training of U.S. personnel to acquire knowledge and skills in nuclear applications. The public has generally not been aware of the persistent lack of personnel trained in nuclear science methods. This public includes the intellectual communities of university campuses and university administrations. Moreover, scientific discipline faculty able and interested in providing both training and strategic nuclear R&D is disappearing in the U.S. at accelerating pace. On the other hand, less burdened by anti-nuclear sentiment, a younger generation now expresses interest in gaining access to skills in nuclear technology application and a basic knowledge in nuclear science. However, corresponding training opportunities are not available to students at an age where important career decisions are made. In fact, nuclear science and technology have all but disappeared from the undergraduate curriculum at U.S. institutions of higher education. With creating the ANSEL laboratory/lecture course, a collaboration between the nuclear chemistry group and a nuclear physics group has been established to help arrest and possibly reverse such detrimental trends at the University of Rochester. The project has received direct support by the U.S. Nuclear Regulatory Commission and indirectly by the U.S. Department of Energy, through providing used equipment and expertise by senior scientific personnel it supports. DOE supported students are the first ones to benefit from the buildup of a modern nuclear science education lab.
II. Specific goals of the ANSEL project

Goals of the laboratory/course experience are to develop a sophisticated understanding of our terrestrial radiation environment, as well as to acquire practical skills in the routine use of radiation detectors, monitors and electronics, ability to assess and abate radiological threats. The 4 series of in-depth ANSEL experiments will go already a long way in training well-rounded, competent nuclear personnel who are able to accomplish experimental tasks with appropriate nuclear instruments and methods. ANSEL will introduce students to the following areas:

1. Interactions of radiation with matter, radiation protection and decontamination. Electronic visualization of pulse responses and determination of detection efficiencies (Scintillation, Ge, Si, Geiger counters), basic nuclear electronics, fast and slow circuitry, digitization and pulse shape analysis. (Initial 4 weeks for all students)

2. Quantitative determinations of source activities in different lifetime regimes (ns to years) with singles and coincidence methods. Imaging using $\gamma$-$\gamma$ coincidences. (2 weeks)

3. Modern methods of forensic imaging with thermal and fast neutron activation analysis $\{(n, \gamma), (n, p), (n, \alpha)\}$, “phase-lock” coincidence method. Cosmic $\mu^+$ and $\mu^-$ decay and capture in light (plastic) and heavy materials (Pb, U). (2 weeks)

4. Recoil-less (Mössbauer) nuclear resonance absorption and emission measurements with an $^{57}$Fe source, using different samples, appropriate detectors and absorbers, to determine Fe abundance, to test local magnetic fields and chemical effects for various solid state materials, compounds and oxidation states. (2 weeks).

The curriculum is specifically designed for undergraduates at the junior level, who have just the basic background in math and science courses. However, our first-year graduate students will also benefit significantly from participating actively in a series of the more demanding nuclear experiments. As future teaching assistants in the new ANSEL they will study and apply the experimental methods in greater depth. This will better prepare them for later experimentation at national and international research centers. Fulfilling an obviously natural function, faculty, the senior scientist (JT) and the research associate (HS) will dedicate some of their time to the training of young students utilizing the ANSEL.

III. ANSEL lecture plan

The ANSEL course is offered for the first time during the Spring Term 2010, with the intention of offering the course every year thereafter. The course will be taught
by a team consisting of the authors, radiation safety professionals and guest lecturers who use nuclear radiation and methods in their professions. Initially, the course will admit undergraduate and graduate students from UR science departments but will later enroll also members of other academic or professional units of the University. It appears natural to coordinate the ANSEL with related areas of study, e.g., with a new medical physics path. The tentative lecture plan for 12 weeks of course work given below reflects the schedule of experiments to be conducted by the students.

A. **Intro to nuclear properties, radiation, detection**
   - Basic properties of nuclei, nuclear decay.
   - Principles of interactions of nuclear radiation with matter, radiation protection.
   - Response of scintillation, gas and solid-state detectors to radiation.
   - Use of oscilloscopes, basic nuclear counting electronics.
   - Signal processing, data acquisition, data analysis.

B. **Measurements of activities and lifetimes**
   - Measurements of source activities.
   - Lifetime measurements b-delayed g emission, long and short (ns/µs → min/d)
   - Lifetime of µ⁺ in weak decay (µs), µ⁻ in weak capture (50-100 ns, for heavy nuclei).

C. **Material imaging and testing**
   - Material imaging and testing with γ-rays (PET scan, γ−γ angular correlation).
   - Neutronics, n detectors, n diffusion.
   - Thermal neutron activation with a neutron source, measurement of b or g and analysis.
   - Neutron activation with a neutron generator, “phase-lock” method, fast (n, γ), (n, p), (n, α).

D. **Mössbauer measurements**
   - Principle of recoil-less nuclear resonance absorption.
   - Mössbauer instrumentation in absorption and emission, Geiger, gas, solid-state detectors.
   - Calibration of velocity scale.
   - Measurement of 57Fe X rays and 14.4-keV Mössbauer g rays with different absorbers.
   - Determination of Fe abundance in mixtures, magnetic splitting in 57Fe enriched foil, isomer shift, chemical shifts for chemical compounds in different oxidation states.

IV. **Recent experiment developments**

During the current funding period, the UR Nuclear Science Research Group has used NRC funds to acquire new or reconditioned equipment for several ANSEL experiments, including several γ-ray and neutron detectors and a thermal-neutron activation station for use with radioactive sources. Finally, after tedious explorations, evaluations and negotiations with various firms, a neutron generator has been obtained from Thermo Fisher-Scientific, an experienced US company that provides similar
equipment for field use in commercial oil exploration. Several experiment setups have been constructed and tested by undergraduate and graduate students, helping to better define realistic tasks to be achieved in the actual lab time available for the conduction of ANSEL experiments (approximately two 3-hour periods per week).

The user friendly EZDAQ remote data acquisition and analysis system has been expanded for ANSEL, specifically for the use in time dependent spectroscopy such as done in neutron activation. In addition, work on several instruction manuals for students and teaching assistants is in progress.

Progress has been made in the setup of several ANSEL experiments:

1. The ANSEL Cosmic Muon Telescope (ACMUT) has been reconfigured, and several measurements have been performed with $\gamma$-ray and $\beta$ sources, as well as with cosmic muons. Simulation calculations have been performed to help understand the performance of the telescope.
2. A preliminary ANSEL Neutron Activation and Imaging Station (ANAIL) has been assembled and used in initial activation experiments. Studies were performed on background $\gamma$-rays to help design a low-level counting station for measuring activated samples.
3. Hardware, detectors and electronics modules have been acquired and assembled for the $\alpha$ spectroscopy and Rutherford backscatter experiment.
4. A universal CAMAC data acquisition system (EZDAQ) was designed, tested and realized in several copies. It allows remote control of experiments, access to data via the University wide network.

**ANSEL Experiment** Alpha particle spectroscopy/Rutherford backscattering:

An important goal of this spectroscopy experiment is demonstrate that charged particles penetrating matter deposit a part of their kinetic energy in the stopping material. Specifically, students will test a quantitative model of the interaction of charged particles with matter (Bethe-Bloch), its dependence on the atomic number and the thickness of the material. Solid state detectors will be used to detect the kinetic energy loss of alpha particles traversing metallic foils.

**Figure 1:** Experimental setup for alpha particle spectroscopy. Left: setup with ORTEC 808 chamber, vacuum gauge and Tektronix scope. Center: Chamber rack with Si detector, target tray and $\alpha$ source. Right: Backscattering test setup (top), target container with blanks and mounted foils.
Photographs of the simple setup with vacuum chamber and accessories are shown in Fig. 1. Apart from gaining insight into the theoretical background of charged-particle interactions, in this experiment students will learn practically how to connect up and bias a silicon surface barrier detector, check out a slow (energy) electronics circuit and perform an energy calibration using α sources and a precision pulse generator. The students will also set up the electronic and software logic for the EZDAQ acquisition system with an analog-to-digital converter (ADC) for the one-parameter (alpha energy) measurement. Students will then use the setup to determine the energy resolution of the silicon detector and the thicknesses of several metallic foils.

**Figure 2:** Energy spectrum of 5.4-MeV alpha particles from a $^{241}$Am (100-nCi) alpha source. Peak on right: un-attenuated (Blank). Peak on left: transmission through a 1.3-mg/cm² Ni foil. Red curve: Gaussian fit curve.

Measurements will be performed with a blank target and various metallic (Ni, Cu, Au) foil targets. Figure 2 gives an example of a pulse height (energy) spectrum obtained with the present setup (cf. Fig. 1, center) and a (100nCi) $^{241}$Am source. The spectrum in Fig. 2 shows the unperturbed alpha peak at high energy (channel 3140) and the effect of a 1.3-mg/cm² thick Ni foil on the spectrum (channel 2760). Data analysis of the pulse height spectra include fitting of spectral lines and comparisons to the predictions by the Bethe-Bloch formula. Some of the planned future developments of this experiment, specifically depth profiling using Rutherford backscattering, will depend on the availability of sufficiently strong α sources.

**ANSEL Experiment** Gamma-gamma PET imaging:

The ANSEL will feature several experiments relevant to imaging of density depth profiles of materials including organic tissue. An early test setup of one of the experiments is pictured in Fig. 3. Two low-cost NaI(Tl) integral-line γ detectors (BICRON) and a surplus NIM electronics bin have been acquired for this setup, a movable rack for the setup and its electronic modules is on order.

The γ detectors will be used in a coincidence and angular correlation setup, which is designed to measure in coincidence the two 511-keV annihilation γ-rays emitted from a $^{22}$Na
source. The source is placed off-center somewhere on the correlation table. Maximizing the \( \gamma - \gamma \) coincidence rate with respect to relative detector angle for various fixed positions of one of the detectors allows one to determine the position of the source. The technique is the basis of diagnostic PET scans used in medicine.

In addition, a measurement of coincidence and singles \( \gamma \) rates will be used to determine the absolute activity of the \( \gamma \) source. Setup of slow, fast and DAQ electronics, as well as energy calibrations have been performed by students prior to this experiment but will have to be done here for two detectors. Test experiments are planned for this summer.

**ANSEL Experiment: Activation with thermal neutrons:**

The ANSEL will provide students with modern methods used in materials analysis, including those used in nuclear forensics, which utilize mobile thermal and fast neutron generators. Prior to the arrival of such a generator, which has been ordered from Thermo Fisher Scientific, preparatory experiments have been conducted with a medium-strong (500 mCi, 18.5 GBq) AmBe neutron source. These included a determination of the \( \gamma \) background in the laboratory, tests of the performance of several available \( \gamma \) detectors (NaI(Tl) and LaBr\(_3\):Ce, BICRON BriliLanCe®-380), theoretical simulations of radiation hazards, and the reconditioning of an available, paraffin-filled "activation drum." Figure 4 depicts a photograph and a technical drawing of the drum.

![Figure 4: Photograph of the reconditioned activation drum (Left). Technical drawing (Right). Measurements are given in units of cm.](image)

Predicted radiation levels associated with the neutron source, or the new neutron generator, suggested proper care in the design of adequate shielding and prompted the acquisition of additional shielding materials, to shield operators both against thermal neutrons and \( \gamma \)-rays. In addition, to shield the measurement of activation \( \gamma \)-rays from intense laboratory background, a low-level counting station was built from several layers of lead bricks.
Results obtained with the LaBr₃:Ce detector from the thermal-neutron activation of a thick aluminum (²⁷Al) sample are depicted in Fig. 5 on semi-log scale. A 1.78-MeV line associated with the beta-delayed γ decay of the ²⁸Si daughter isotope is clearly discernible on top of an intense background (shaded). To a large extent, the latter is due to an intrinsic detector activity. However, the better resolution of the LaBr₃:Ce detector made the measurements superior to one with a larger NaI(Tl) detector. The half-life found with the detector was $t_{1/2} = (134 \pm 6)$ seconds, in good agreement with the literature.

These measurements benefited from the new capability of the EZDAQ data acquisition software to generate time marks for every event.

The final neutron activation experiments will be very complex, requiring supervision and guidance of ANSEL students by a professional and trained faculty or laboratory associates. However, students will be exposed to techniques of identifying unknown substances by the energies and time dependent intensities of characteristic activation γ-rays. In addition to practicing the skills in detector and electronics setup acquired in previous experiments, the students will be introduced to phase locked measurements of activation γ-rays which are synchronized with the generator pulsed neutron beam.

**ANSEL Experiment** Interactions of cosmic ray muons with matter:

The mechanical and electronics parts of the setup for the ANSEL muon experiment have been improved. Plastic scintillation counters, their HV power supplies, fast and slow signal processing electronics and oscilloscope have now been incorporated in a single self-contained rack. A new channel-aluminum rack adds flexibility to the mechanical setup and allows for a continuous positioning of the targets and the various telescope and active-target counters in the muon stream.

The experiment suffers from the relatively low intensity of cosmic muons. To demonstrate the process of negative muon capture in nuclei is particularly demanding, since negative muons make only about 30% of the total muon flux. However, an effect of high-Z targets is
visible after continuous data accumulation for several days.

In Fig. 6 results are shown for such a run with a 5-cm thick Cu target. The time distribution of all events (X-rays, γ-rays, particles) measured with a thick plastic counter can be interpreted in terms of a disappearance rate, a sum of the decay and capture rates for all channels through which a muon is absorbed by the target.

**ANSEL Experiment:** Imaging with fast and slow neutrons

The MP 320 neutron generator is the newest addition to the ANSEL equipment pool. It is shown below in its temporary shielding and activation setup. Figure 7 displays the generator test stand with electronics and data acquisition. To satisfy radiation safety requirements, the generator room and the adjacent lab room had to be equipped with a set of warning lights and interlocks.

![Commissioning runs for n generator served for performance evaluation of new neutron detector N*](image-1)

![M-320 Neutron Generator Station (shielding partially removed)](image-2)

![Future application in high-energy density plasma diagnostics (LLE)](image-3)

![Test stand for n detector development](image-4)

**Figure 7:** Setup and commissioning of the ANSEL pulsed-neutron generator were performed by nuclear chemistry graduate students. The initial runs provided the opportunity to conduct important performance tests for the new neutron detector developed by the group.

To install an economic “rabbit” system for the shuttling of samples between generator and radiation detector is the next important task for the commissioning of an ANAIS station served by the generator. One can imagine that this forensic type activation experiment may be one of the favorites of the students. This may help motivating them to invest the significant efforts into acquiring nuclear forensic skills that are in demand around the country.
IV. Publications and Activities

A. Articles

"Dynamical and Statistical Fragment Production In $^{136}$Xe$^{+209}$Bi Reactions at E/A = 28, 40, and 62 MeV",
W. Gawlikowicz, J. Tõke, W.U. Schröder, R.J. Charity, and L.G. Sobotka,

“Gentle Multifragmentation – a Generalized fission”
J. Tõke and W.U. Schröder,
Report UR-NCUR 07-017
Inv. Paper, Proc. Intern. Workshop Multifragmentation and Related Topics,

“A Simple Method for Rise-Time Discrimination of Slow Pulses from Charge-Sensitive Preamplifiers,”
J. Tõke, M.J. Quinlan, W. Gawlikowicz, and W.U. Schröder,

“Effects of H2O and H2O2 on Thermal Desorption of Tritium from Stainless Steel”
M. J. Quinlan, W. T. Shmayda, S. Lim, S. Salnikov, Z. Chambers, E. Pollock, and
W. U. Schröder,
Fusion Science and Technology 54, 519 (2008)

“Common Signatures of Statistical Coulomb Fragmentation of Highly Excited Nuclei and Phase Transitions in Confined Microcanonical Systems”
J. Tõke, M.J. Quinlan, I. Pawelczak, and W.U. Schröder,
Report UR_NCUR 08-20/arXiv:0804.4698;

“Isoscaling in Statistical Fragment Emission in an Extended Compound Nucleus Model”
W. Ye, J. Tõke, and W.U. Schröder,
arXiv:0808.1917v1, and

“Correlations between Reaction Product Yields as a Tool of Probing Heavy-Ion Reaction Scenarios,”
Report UR-NCUR 08-18.2

“Energy Realpolitik: Towards a Sustainable Energy Strategy,” W.U. Schröder,
Updated version to be submitted.

“Surface Entropy and Density Dependence of the Symmetry Energy in a Harmonic Fermi Gas Model,”
W. Ye, J. Tõke, and W.U. Schröder,
To be submitted to Physical Review C (2009).
“N-Star: A Novel Neutron Detector with Broad Dynamic Range and Multi-Hit capability,”
I. Pawelczak, E. Henry, Y. T. Tsai, J. Tőke and W.U. Schröder,

"Isotopic Relaxation Effects in Nucleon Transfer and Pre-Equilibrium Emission in $^{112}\text{Sn}+^{40,48}\text{Ca}$ Reactions at E/A = 35 MeV",
D.K. Agnihotri, S.P. Baldwin, B. Djerroud, B. Lott, B.M. Quednau, W. Skulski,
J. Tőke, W.U. Schröder, and R.T. de Souza,

B. Invited Lectures and Presentations

“Nuclear Reaction Dynamics – Beyond Equilibrium and Mean Field”
Presentation at the NSAC Towne Meeting on the Long Range Plan, Chicago, Jan 21, 2007. (by W.U. Schröder)

“Gentle Multifragmentation – a Generalized Fission”
Inv. Talk Int. Workshop Multifragmentation and Related Topics,
Caen, 11/03-11/08/2007, (by J. Tőke)

“Nuclear Science at the University of Rochester”
Research Seminar, Department of Physics and Astronomy, University of Rochester,
Febr. 15, 2008, (by W.U. Schröder)

“Energy Realpolitik- Towards a Sustainable Energy Strategy”,
Inv. Talk, “Energy Perspectives” Lecture Series, University of Rochester Virtual
Institute for Energy, March 5, 2008, (by W.U. Schröder)

“Nuclear Stability and Particle Emission,”
Inv. Talk Spring Meeting of the American Chemical Society, Seaborg Symposium,
New Orleans, April 7, 2008, (by W.U. Schröder)

"Decontamination of Tritiated Stainless Steel via Thermal Desorption,"
Inv. Poster Presentation, Intern. Conf. Tritium Science and Technology, Univ. of
Rochester, 2008. (by M.J. Quinlan)

“N-STAR: Design and Testing of a Neutron Detector with Broad Dynamic Range
and Muti-Hit Capability.”
Inv. Talk, 2nd Int. Conf. Current Problems in Nucl. Physics and Atomic Energy,
Kiev, Ukraine, 2008. (by I.A. Pawelczak)

“Nuclear Chemistry Science and Education at Rochester”
Inv. Presentation, Nuclear and Radiochemistry Workshop,
Albuquerque, NM, April 17-18, 2008, (by W.U. Schröder)
"Isotopic Particle Correlations in Reactions at Intermediate Energies"
Inv. Talk, Worksh. Nuclear Symmetry Energy at Medium Energies,
Catania and Militello V.C., Catania, 29-30 May, 2008, (by W.U. Schröder)

"Energy Futures for a High-Tech Society,"
Inv. Talk Skeptical Chemyst Lecture Series, Gettysburg College, April 16, 2009.
(by W.U. Schröder)

"Cecil II: Data Calibration and Analysis Progress Report,"
Seminar LNS Catania, May 2008 (by M.J. Quinlan)

"Reality and Prospects of Research and Training in Nuclear Science at The Uni-
versity of Rochester,“
19, 2009, (by W.U. Schröder)

"Nuclear Energy and the Study of Nuclear Matter far from Stability,"
Phys. Chem. Seminar, University of Rochester Department of Chemistry,
August 2009 (by M.J. Quinlan)

C. Contributed Papers/Abstracts

in Constrained Micro-canonical Modeling of Excited Nuclear Systems,"
APS-DNP Fall Meeting, Newport News, 10/11-10/14/2007

APS-DNP Fall Meeting, Newport News, 10/11-10/14/2007

APS-DNP Fall Meeting, Newport News, 10/11-10/14/2007

Wei Ye, Jan Tőke, W. Udo Schröder, “Possible Angular momentum dependence of dissipation in nuclear fission.”
April Meeting of the APS, St. Louis, MO, 2008.

J. Tőke and W. U. Schröder, “Common signatures of Coulomb fragmentation of excited realistic nuclei and phase transitions in hypothetical confined matter,”
April Meeting of the APS, St. Louis, MO, 2008.

I. A. Pawelczak, Y. T. Tsai, “Design and performance of the N*neutron detector.”
April Meeting of the APS, St. Louis, MO, 2008.

D. Professional Activities (WUS)

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, University of Rochester Chapter of the American Association of University Professors, 1999-.
Referee for Journal Articles and Research Proposals to US (NSF, DOE, DOS (joint CIS/CRP funds)) and Canadian Funding Agencies.
Member of the Oversight Committee for the Campus Instrument Machine Shop.
Member Radiation Safety Committee, University of Rochester 1994-.
Member Radiation Safety Advisory Committee, University of Rochester 1994-.
Member of several Department of Chemistry committees.

V. Personnel

Dr. W. Udo Schröder, Professor of Chemistry and Physics, Principal Investigator
Dr. Jan Tõke, Senior Scientist
Dr. Wei Ye\(^1\), Visiting Scholar, South-Western University, Nanjing/P.R. of China
Dr. Hardev Singh\(^2\), Research Associate
Mr. Eric Henry, Graduate Student, Department of Chemistry
Ms. Iwona Pawelczak, Graduate Student, Department of Chemistry
Ms. Elizabeth Pollock\(^3\), Graduate Student, Department of Physics
Mr. Michael Quinlan, Graduate Student, Department of Chemistry
Mr. Matthew Sharpe\(^4\), Graduate Student, Department of Chemistry
Ms. Yun-Tse Tsai\(^5\), Graduate Student, Department of Physics
Ms. Ariel Brown⁶ Undergrad. Student, Department of Chemistry
Mr. C. Sowinski⁷ Summer Internship, Rochester City High School
Mr. M. Honickman⁸ Pittsford/Rochester, High School Intern

¹July 1, 2007-July 31, 2008
²From April 30, 2009
³Until May 31, 2009, graduated Master of Science in Physics, 2009
⁴From July 31, 2009
⁵Until July 31, 2008, graduated Master of Science in Physics, 2008
⁶Until May 31, 2009, graduated Bachelor of Science in Chemistry, 2009
⁷June-August, 2008
⁸Since August, 2009

Visitors

Prof. Malgorzata Pfabe, Smith College, June 12-13, 2008

Dr. Vladilen Goldberg, Cyclotron Laboratory, Texas A&M University, November 11-12, 2008