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ROCHESTER, NEW YORK 14267-0216, USA

Compound Nucleus with Surface Entropy; A Unified Phenomenology of Particle Evaporation, Fission, Fragment Production, and Multifragmentation

Jan Tõke and W. Udo Schröder

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J. Tõke and W. U. Schröder

Department of Chemistry, University of Rochester
Rochester, New York 14627, USA

Abstract

It is shown that a consistent incorporation of thermal excitation of surface oscillation modes into a general scenario of the compound nucleus decay provides a unified description of the "classical" compound nucleus decay, fission-like processes, intermediate-mass fragment production, and multifragmentation. Further, it is shown that such a unified phenomenology of a compound nucleus with shape fluctuations leads to the same approximate mathematical equations or numerical codes that are used by some other models (albeit on different grounds) and that appear to be successful in describing quantitatively a host of experimental observations on intermediate-mass fragment (IMF) production.

A strong interest over the last two decades or so, in the phenomena related to intermediate-mass fragment (IMF) production and to multifragmentation, in particular, has been largely driven by a belief or hope that such phenomena may reflect liquid-gas phase transition (LGPT) in finite nuclear systems and, thus, provide a valuable insight into the workings of nuclear matter at elevated excitation energies. Indeed, many patterns observed in the fragment yield distributions appear to be coinciding with anticipated signatures of LGPT, resulting too often in claims that such a transition

has been observed. Yet, such claims lack solid justification in terms of a plausible "geometrical" scenario under which liquid and gas could coexist in small systems confined by the attractive nuclear forces only. By the term "geometrical" it is here understood the actual evolution of the mass and charge distributions. The justification for the claims of LGPR is in fact lacking, because either explicitly or implicitly, all of the proposed models make use of an artificial transient confinement vessel to allow the gaseous phase to equilibrate, with then a possible condensation (e.g., via a spinodal decomposition mechanism) into sizeable fragments. While such models are often capable of replicating many salient features observed in the IMF yield distribution, it has not been shown that the results would be substantially similar in the absence of the confinement vessel, i.e., in a realistic scenario. Quite to the contrary [1], for non-confined (other than by the nuclear attraction) excited systems, it appears rather certain that any particle that succeeds in (statistically) overcoming this attraction, is bound to leave the system "for good", practically, without any chance of ever interacting with any part of the system again.

At the same time, there is a true first-order phase transition taking place in finite nuclei - a transition between the bulk matter and the surface-domain matter that is most likely responsible for the observed signatures of phase transitions. It is important to appreciate the fact that a finite nuclear system is inherently a two-phase system, consisting of bulk matter and of the surface-domain matter, where the whole diffuse surface-term is approximated by just one phase. The latter coexist within a volume defined by the action of nuclear forces - the nuclear potential well, but have distinctly different thermodynamical properties, such as matter density, specific heat, average binding energy per nucleon, etc. A (first order) transition between the bulk phase and the surface-domain phase occurs as thermal shape fluctuations develop in the course of the system evolution toward microcanonical equilibrium. What is essential for such phase transition actually taking place is the often overlooked or *"lost-in-approximations"* fact that the bulk and surface-domain matters are characterized by different heat capacities or level density parameters and that, notably, the low-density surface-domain matter is characterized by a larger level density parameter (per nucleon) than the high-density bulk matter. The latter fact is reflected in the presence of a positive surface term in the level density parameter *little-a* [2], thought to explain the enhanced compound nucleus fission rates. Because of this excess entropy per surface-domain nucleon, the system will maximize its entropy by effectively moving part of its constituent nucleons from the dense bulk to the low-density surface domain in what is a classical first-order phase

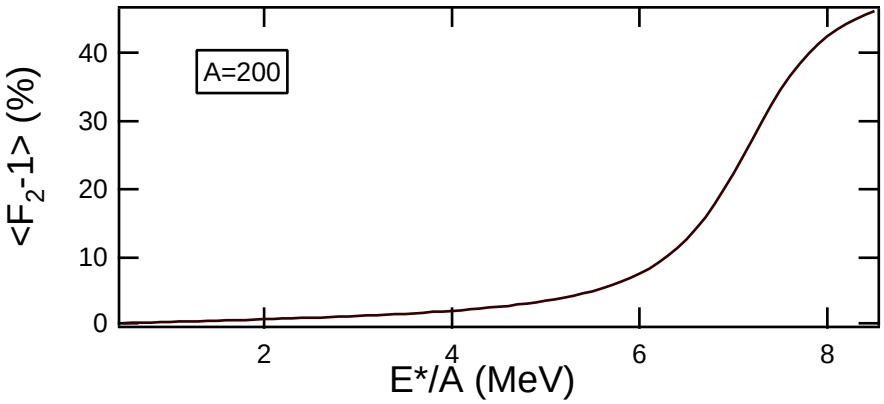

Figure 1: Evolution of the average surface area of excited nucleus with excitation energy as predicted by CNSEM.

transition. With increasing excitation energy, the surface-domain entropy per surface-domain nucleon increases, resulting in ever larger thermal shape fluctuations (more matter transferred from bulk to surface-domain) with an obvious consequence of ever increasing likelihood of arriving in the course of these fluctuations to any of the numerous fragment separation saddles and thus to fragment emission. At any rate, unlike the liquid-gas PT, this latter bulk - surface PT *must* occur and, indeed, is known to occur in compound fission. Recently, its signatures have been modeled in theoretical calculations using Harmonic Interaction Fermi Gas Model and have been found to exhibit phase-transition-like behavior,[3] as well as enhanced IMF production rates [4]. The latter model is referred to as a Compound Nucleus with Surface Entropy Model (CNSEM), to stress the importance of the surface domain to its workings.

The presence of a phase transition from bulk to surface domain is illustrated in Fig. 1 where the average increase in the relative surface area (with respect to a sphere of equal volume) due to thermal shape fluctuations is plotted versus excitation energy per nucleon. The calculations were performed within the CNSEM formalism described in Refs. [4] and [5]. Note a rapid increase in the magnitude of the fluctuations around E/A=7 MeV. Coulomb interaction was disregarded in the present calculations, but obviously, it will help develop even stronger fluctuations.

The above picture of a compound nucleus as a system of coexisting

bulk and surface-domain phases, with a first order phase transition taking place with increasing excitation energy, offers a unified description of particle evaporation (classical Weisskopf's process), fission (transition-state phenomenology with $a_f/a_n > 0$ (ratio of level density parameters taken at fission saddle-point and at spherical-shape), and fragment production (asymmetric fission phenomenology) [4], all of these processes being triggered by the system's evolution toward microcanonical equilibrium.

The linking of various statistical decay processes of the compound nucleus to a phase transition is more than a semantical or symbolic exercise. One has in this case, indeed, a true first-order phase transition, a one that is of significance only in small systems. Thus, according to the principle of universality, such a transition is expected to share signatures with other known phase transitions. And it is seen to do so, e.g., when one considers nuclear multifragmentation. This is so, because within the present scenario of a compound nucleus with fluctuating shape, multifragmentation is a reflection of the system arrival (with increasing excitation energy) at the point where entropy is insensitive to any increase in surface area, i.e., at a point where surface tension vanishes. At this point, any multi-fragment saddle shape is almost equally probable, resulting in a separation of the system into fragments under the action of Coulomb forces.

In view of the above, it is important to assess, how well the above "natural" scenario agrees with experimental observations. The term "natural" is used here to stress the fact that the model does not include in fact any new assumptions, not already present in time-proven models of compound nuclear decay (including fission), simply bringing the known facts to their logical consequences. This is in a stark contrast to models relying, e.g., on an extra-physical concept of a freezeout volume [8, 9] (where the size of the volume cannot be expressed in terms of known physical quantities) or purely mathematical models of percolation and lattice gas. It has been shown earlier, that accounting for the excess entropy per nucleon of the surface domain leads to a significant enhancement of fragment production at elevated excitation energies, allowing it to compete successfully with particle evaporation [4]. It has been also shown that because of thermal expansion, the highly excited system may have sufficient time to equilibrate on its own (without requiring a transient external confining box), so that the surface fluctuations can, indeed, develop [6]. While a true comparison to experimental observations would require writing of complex numerical codes and would require a detailed modeling of the behavior of hot nuclear surface domain, a more limited modeling, on par with existing models as far as the use of free parameters is concerned, appears possible. This is done here through a logical

process, where it is shown that the present model of a compound nucleus with surface entropy (CNSEM) is consistent with parameterizations of fragment yields reported to provide good fits to experimental data over a wide range of reaction parameters. Specifically, it shown that CNSEM is consistent with the parameterization of fragment yields in the nuclear Fisher's droplet model (NFDM) [10, 11] and that of the freezeout-based models of nuclear multifragmentation [8, 9].

CNSEM expresses fragment production rate p in terms of loss of entropy incurred to reach the fragment saddle configuration [4, 7],

$$p \propto e^{S_{Saddle} - S_{Eq}}, \tag{1}$$

where S_{Saddle} and $S_{Eq.}$ are saddle- and equilibrium-state (thermally expanded) entropy, respectively. In a (canonical) approximation, where temperature of the saddle configuration is close to the temperature of the spherical equilibrium state, the change in entropy ΔS can be replaced the change in free energy divided by temperature, $\Delta S \approx -\Delta F/T$. Then, because the nuclear part of the free energy of the bulk is preserved in shape fluctuations, the resulting change in free energy is due only to the change in Coulomb energy and to the change in surface free energy F_{surf},

$$p \propto e^{\Delta S} \approx e^{\frac{E_{Coul}}{T} - \frac{\Delta F_{surf}}{T}}. \tag{2}$$

The latter equation of CNSEM is in essence identical to the basic equation of the NFDM for the abundance n_A of fragments of mass number A,

$$n_A = q_o A^{-\tau} e^{(\frac{A\Delta\mu + E_{Coul}}{T} - \frac{\Delta F_{surf}}{T})}, \tag{3}$$

where $\Delta\mu$ represents nominally the difference in chemical potentials of the liquid and gaseous phases, τ is a topological exponent, and q_o is a normalization factor.

NFDM obtains fits to experimental yields over a broad range of excitation energies and fragment sizes, by arbitrarily parameterizing Coulomb energy and arbitrarily parameterizing surface free energy, both, without providing a meaningful justification. Taken at face value, the same parameterization can be then used in CNSEM, resulting in identically good fits of the latter to the host of experimental observations. However, while no meaningful justification for such a parameterization appears possible within NFDM, its form is quite natural in the case of CNSEM.

And so, NDFM expresses Coulomb energy in terms of the energy of two touching spheres while arbitrarily damping this energy with increasing

temperature from the full Coulomb energy to zero at a critical temperature [10]. Neither the sign of this term, nor its form, nor its vanishing with increasing temperature can be related to a scenario of droplet condensation, as the Coulomb energy can change only via rearrangement of the charge distribution. However, it is natural for a fission-like phenomenology envisioned by CNSEM, where the "mysterious" temperature dependence of the Coulomb energy reflects simply the fact of the saddle configuration becoming more and more compact as the surface tension decreases. At the point, where surface tension vanishes, any saddle configuration is spherical, reducing both the Coulomb and the surface free energy terms to zero. Note that the Coulomb term has an enormous impact on the predicted fragment yield, varying by many orders of magnitude over the range of excitation energies considered. It is, in fact responsible for the reported agreement of NFDM with experimental observations [10]. Taken at face value, the fact that the particular form of the Coulomb term "works" lends then a strong support for the CNSEM scenario, which calls for such a form of excitation energy dependence.

Regarding the surface free energy term, NFDM [10], parameterizes it as a linear function of temperature,

$$\Delta F_{surf} = c_o \frac{T_c - T}{T_c} A^{\sigma}, \tag{4}$$

where c_o is the ground-state surface energy coefficient and sigma is a dimensionality parameter. NFDM refers to Eq. 4 as the Fisher's scaling [11], yet this kind of scaling has not been justified neither in Ref. [11], nor elsewhere. It is in fact difficult to reconcile with the thermodynamical theory, which expects free energy to be a strictly concave function of temperature. On the other hand, CNSEM is consistent with Eq. 4, because CNSEM does call for the change of the free energy term with temperature from $F_{surf} = c_o$ at zero temperature to zero at a temperature where surface tension vanishes (in the range of 5 - 7 MeV), as does Eq. 4 and because the resulting free energy is, indeed, a strictly concave function of the (Fermi gas) CNSEM temperature.

At high excitation energies, where surface tension becomes negligible, multifragment saddle configurations may be populated with significant probabilities. Such configurations will then separate into individual fragments dynamically, under the action of Coulomb forces. While such a scenario is conceptually different from the one employed by freezeout-based models (FBM) [8, 9], it is similar as far as the computational apparatus is concerned. This is so, because FBM enhance entropy of multi-fragment configurations by adding to a true "freezeout" state entropy, entropy of "rattling" motion

of fragments within the "oversize" freezeout vessel. They overcount multi-fragment configurations, by incuding configurations which evolved dynamically from configurations that were already once counted. Yet, the "rattling" motion is to a good extent mocking up surface diffuseness as the associated "rattling" entropy is equivalent to the legitimate surface entropy used in CNSEM. A numerical implementation of CNSEM could then entails finding out in how many ways multi-fragment saddle configurations would fit into a volume accessible on the grounds of costs in entropy. By approximating the multi-fragment saddle shapes with sets of touching spheres with diffuse surfaces, CNSEM thus leads to the same mathematical formalism as employed successfully by FBM [8, 9].

In conclusion, CNSEM is consistent with parameterizations reported to describe a host of experimental observations, while considering fragment production and multifragmentation surface-instability rather than bulk matter related phenomena. The latter phenomena are closely related to a phase transition between bulk matter and surface-domain matter.

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