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Abstract

A novel prompt decay mode of highly excited nuclear systems is shown to set in with necessity as a certain critical excitation energy is reached. It is driven by a peculiar, open-ended type of spinodal instability, unique to self-bound open systems, and consists in parts of the system undergoing spontaneous indefinite thermal expansion ending in vaporization into the surrounding open space. The mode, named here spinodal vaporization, is distinctly different from all known decay modes of excited nuclei and faces no competition from the latter. It sets a natural upper limit for the excitation energy that can be thermalized by compound nuclear systems, while setting also a limit to the applicability of thermodynamics to the description of highly excited nuclear systems.

Keywords: Thermodynamics, Spinodal instability, Prompt decay, Microcanonical, Boiling

1. Introduction

Understanding the limits of thermodynamical stability of excited nuclear systems has been a focus of numerous theoretical and experimental studies [1, 2, 3, 4, 5] from the dawn of nuclear science. Theoretical studies were so far overwhelmingly concentrated on instabilities in bulk nuclear matter kept under controlled conditions and, notably, at a fixed volume. The model calculations show that under such condition, owing to the Van der Waals - like equation of state for uniform matter, three kinds of domains exist in the space of controlling parameters for such matter. Firstly, there are domains where uniform nuclear systems would be stable. Then, there are domains where the uniform matter would be metastable and where, given time, it would end up as a two-phase system as a result of evaporation or condensation - aided, perhaps, by fluctuative nucleation. And, finally, there is a domain commonly named spinodal, where no uniform matter could ever be formed. Unlike in the previous two domains, a system brought to this spinodal domain would not become uniform even transiently, but would rather undergo rapid spontaneous phase separation until all of its constituent parts are at least metastable. The above domains are trivially identifiable on the standard plots of Van der Waals'ian isotherms as functions of specific volume and pressure.

In contrast to the above theoretical view, experimental studies necessarily involve finite nuclear systems formed in the course of nuclear reactions. Such systems are not subject to external confinement and are thus free to evaporate particles, and to undergo shape fluctuations leading to fragmentation - the two basic statistical decay modes of metastable excited compound nuclei. It is worth noting here that the metastability is at the very crux of sound thermodynamical models of nuclear decay, such as Weisscopf's evaporation model and various models of compound-nuclear fission. The lack of external confinement has, however, one other unavoidable consequence which is thermal expansion. The paramount importance of the latter at elevated excitation energies has been largely overlooked in theoretical modeling of the equilibrium-statistical decay of excited nuclear systems - other than these reported in precursor conference proceedings [6] of the present study.

The present work is part of a continued effort [7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17] to construct an open microcanonical framework for understanding decay phenomena of highly excited metastable nuclear systems produced in the course of heavy-ion collisions and the limits of compound-nuclear metastability of such systems. It reveals the crucial and hitherto unappreciated role of thermal expansion in a peculiar spinodal destabilization of highly excited nuclear systems. The latter gives rise to a hitherto unknown prompt decay mode of such systems, named here spinodal vaporization, which then imposes a natural upper limit on excitation energy, a compound-nuclear system is capable of thermalizing.

A term "open microcanonical" is used in this study to stress the fact that, unlike conventional microcanonical models of statistical nuclear decay, the formalism used here allows for both, large-scale shape fluctuations and thermal expansion, and that it is the latter phenomenon that is at the crux of the spinodal destabilization of highly excited nuclear systems, that has been overlooked in conventional models.

2. Outline of the Theoretical Framework

studies of the properties of highly excited nuclear systems, both, infinite and finite. [7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17] The model is based on the fundamental thermodynamical principle that an isolated system will assume macroscopic configurations fluctuating around the one that shows highest Boltzmann entropy. These maximum-entropy configurations are assumed to be homogeneous for infinite systems and isotropic with a smooth density profile, for finite systems. By infinite systems are generally understood systems that are large enough to warrant the neglect of the inhomogeneous surface domain. In contrast, the latter domain is of essence in modeling the behavior of finite systems. The above assumptions of uniformity and isotropicity are well justified given the nature of the formalism used.

Boltzmann entropy is evaluated in the framework of zero-temperature Fermi gas model and Thomas Fermi approximation. The zero-temperature formalism was utilized for the sake of simplicity, but its is also a reasonably good approximation in the range of excitation energies considered. Thus, Boltzmann entropy is expressed as:

$$S_{config} = 2\sqrt{a_{config}(E - E_{config})}, \quad (1)$$

with E, a_{config}, and E_{config} denoting the total energy, the level density parameter and the interaction energy of the configuration considered, respectively. The latter represents the zero-temperature energy of the configuration considered and includes potential energy and the Pauli kinetic energy of the Fermi matter. Equation 1 is the base equation of the model, allowing one to evaluate S_{config} for any configuration of interest characterized solely by the matter density distribution $\rho_{config}(\vec{r})$.

The level density parameter a_{config} was calculated using the formalism proposed in Ref. [7]:

$$a_{config} = \alpha_o \rho_o^{2/3} \int \rho(\vec{r})^{1/3}(\vec{r}) d\vec{r}, \quad (2)$$

where α_o expresses the value of the level density parameter per nucleon at normal matter density ρ_o.

The interaction energy is here calculated by folding a standard Skyrme-type EOS interaction energy density $\epsilon_{int}^{EOS}(\rho)$ with a Gaussian folding function and the folding length, adjusted so as to approximately reproduce the experimental surface diffuseness of finite droplets of nuclear matter.

$$E_{int}^{EOS} = R_{Gauss} \int \epsilon_{int}^{EOS}(\rho(\vec{r} - \vec{r'})) e^{-\frac{(\vec{r}-\vec{r'})^2}{2\lambda^2}} d\vec{r} d\vec{r'}, \quad (3)$$

For the equation of state, the present study adopted a standard form consistent with Skyrme-type nucleon-nucleon interaction, which implies the interaction energy density (appearing in Eq. 3) in the form of

$$\epsilon_{int}^{EOS}(\rho) = \rho[a(\frac{\rho}{\rho_o}) + \frac{b}{\sigma + 1}(\frac{\rho}{\rho_o})^{\sigma}] \quad (4)$$

The values of the parameters a, b and σ in Eq. 4 are determined by the requirements for the binding energy, matter density, and the incompressibility modulus to have prescribed values. The values chosen in this study of a=-62.43 MeV, b=70.75MeV, and $\sigma = 2.0$ imply a normal density of $\rho_o = 0.168 fm^{-3}$, binding energy per nucleon at normal density of ϵ_{EOS}/ρ_o=-16MeV, the incompressibility modulus of $K = 220 MeV$, and Fermi energy at normal density of E_o^{Fermi}=38.11MeV.

One notes that in the model calculations for uniformly distributed matter, the finite range of interaction is of no consequence and the configuration energy can be written simply as $V\epsilon_{EOS}(\rho)$, where V is the system volume.

3. Results

The general behavior of uniform Fermi matter can be well understood from the appearance of isotherms in the familiar Van der Waals type plots. These are illustrated in Fig. 1 for the bulk model matter with Skyrme-type EOS with a compressibility constant of K=220 MeV. The isotherms were constructed using microcanonical approach, with T and p representing microcanonical temperature and pressure, and are identical to those computed in a canonical approach, in virtue of ensemble equivalence for uniform matter. The isotherms feature prominently the familiar finite length (measured by specific volume) spinodal domains of negative compressibility for canonical ensemble, but importantly, they feature also, albeit less conspicuously, the crucial open-ended spinodal domain for an open microcanonical ensemble of interest in the present study. The presence of

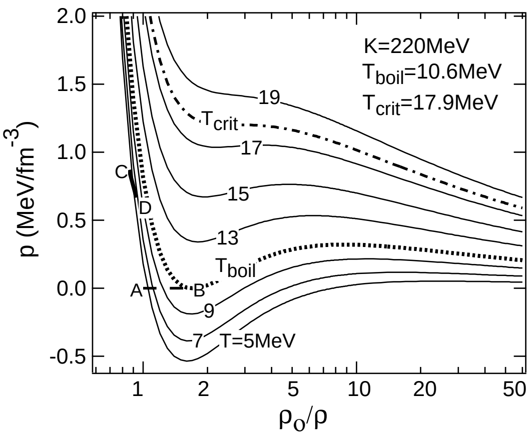

Figure 1: Isotherms for the model matter. The isotherm corresponding to zero-pressure boiling temperature is shown in dotted line and the critical isotherm is shown in dash-dotted line. The adiabatic trajectory for a hypothetical infinite system is shown in dashes (line AB), while such for the bulk of a finite (A=100) system is shown in bold solid line (line CD).

the latter becomes evident when one follows the evolution of a self-bound system at zero pressure with increasing excitation energy per nucleon, starting at point A towards point B, and beyond. The excitation is measured here (see also Fig. 2) by the specific volume displayed on the abscissa and the trajectory of such an evolution is illustrated by the dashed line AB. One notes readily, that first, the temperature rises with increasing specific excitation energy, while the system undergoes thermal expansion. However, beginning at point B, the trend reverses, with the heat capacity becoming formally negative. This indicates that at point B the system enters spinodal domain of convexity of Boltzmann entropy as a function of excitation energy and, thus becomes unstable. Not only cannot it exist in this domain as a uniform matter, neither can it exist here as a two-phase system. This is so, because the spinodal domain is here open-ended, with one end at point B and the other at infinity. Which means, that upon acquiring excitation energy in excess of what is needed to reach the "boiling" point B, the system will promptly shed a portion of itself via a boiling-like process named here spinodal vaporization, leaving behind a "leaner" metastable residue at point B. The latter will subsequently decay statistically via well-known decay modes of evaporation and fragmentation showing the source temperature as that at point B, regardless of the initially acquired excitation energy per nucleon. One may posit that the spinodal vaporization is akin to boiling, even as in practical realizations the latter may to some extent rely on nucleation

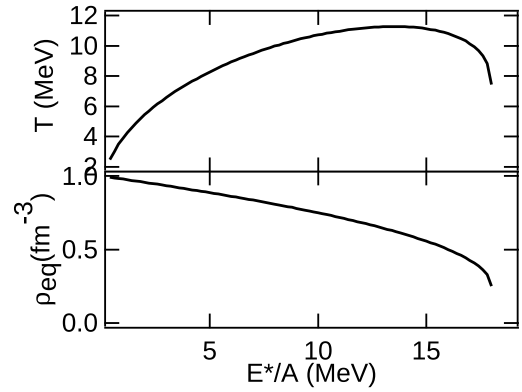

Figure 2: Caloric curve and equilibrium density as functions of excitation energy for a self-bound Fermi liquid under zero pressure.

phenomena. Whether the nucleation is essential or not, it is an unverified contention or speculation of this study, that the spontaneous growth of bubbles in commonly observed boiling is due to the spinodality of the bubble matter.

It is worth noting that the system enters here the spinodal domain with necessity, and not by a *fiat* of modeling, as soon as its initial energy is raised in excess of that at point B, and that the vaporization must take place regardless of any open statistical decay channel. In that latter sense, it is an imposing mechanism. Note also that the nature of spinodal vaporization is such that portions of the system would expand indefinitely, while harvesting energy from neighboring parts, whether the latter are, or are not, spinodal themselves. While doing so, these portions cool down and, ideally, do not increase in mass number. Obviously, the presented picture is that of a prompt decay, different from all decay modes so far considered. Specifically, it is substantially different from commonly known pre-equilibrium emission as the latter lacks the positive feedback mechanism characteristic to spinodal instabilities. It is the positive feedback that makes the process prompt and it appears generally as a result of the "wrong" curvature of the characteristic thermodynamic function - here, the convexity of Bolzmann entropy as a function of excitation energy.

While Fig. 1 proves the inevitability of spinodal vaporization or boiling for any Van der Waals - like matter at elevated excitations and demonstrates that the phenomenon in question has been in "plain sight" for all these years, a more conventional depiction of the instability is offered in Fig. 2 in the form of a caloric curve for the open-microcanonical system considered. As seen in this figure, the caloric curve for such a

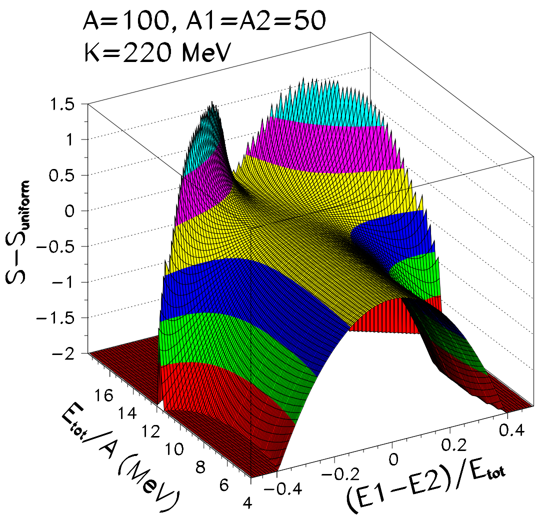

Figure 3: (Color online) Reduced two-phase configuration entropy surface for a configuration of two equal-A subsystems with differing split of the available excitation energy E^* between these phases.

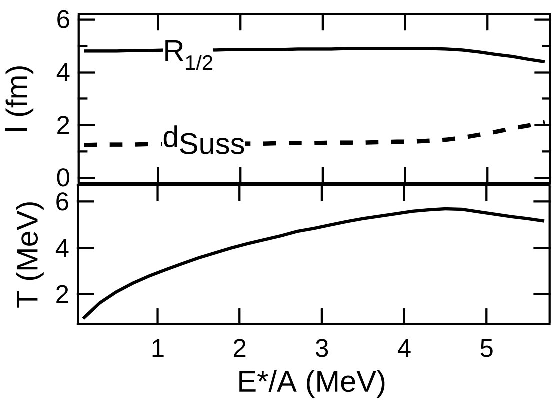

Figure 4: Evolution of the mass density distribution parameters and the microcanonical temperature with excitation energy per nucleon (See text).

system features prominently a domain of formal negative heat capacity, i.e., spinodal instability. This domain is open-ended on the side of high excitations, but in the modeling it ends at the point where the system loses stability with respect to uniform expansion. It is worth noting that the matter density behaves here monotonically in the domain of interest, even though it is thermal expansion that is at the crux of the, here thermal and not mechanical, spinodal instability. Note also that thermal spinodal instability is unique to open iso-neutral microcanonical systems, the only ones where the Boltzmann entropy depends solely on total energy and the only ones where the convexity of entropy function translates directly into the formally negative heat capacity. In contrast, in conventional spatially confined microcanonical systems this entropy depends further on volume and, possibly iso-asymmetry and, hence, the possible spinodal instability will be of thermo-mechanical (iso-neutral system) or iso-thermo-mechanical (iso-asymmetric system) nature. The instability here will be directly associated neither with the formally negative heat capacity, nor the formally negative compressibility, nor the formally negative chemical susceptibility, but associated rather with the loss of the negative definite character of the now two- or three-dimensional Hessian (curvature) matrix of the underlying two- or three-extensive-argument entropy function.

The onset of spinodal vaporization is further illustrated in Fig. 3, in a purely open microcanonical picture that allows for thermal expansion while keeping constant the total excitation energy and mass number, but not the volume. As seen in this figure, at low and moderate excitation energies, the configuration of maximum entropy is that where both equal parts have equal excitation energies, indicating uniform matter and excitation energy distribution. In this regime, the system may and will continually fluctuate away from uniformity, but the negative feedback of concave entropy will return the system back toward uniformity. The situation is dramatically different at excitation energies past the boiling point where the curvature of the entropy surface turns positive. From this point on, any acquired asymmetry would be further reinforced by the action of entropy driving now the system further away from uniformity, with one portion eventually vaporizing away into the surrounding space. It is worth noting that in the spinodal domain of excitation energy, there is no hope of ever seing the system at the state of approximate uniformity, even transiently.

For finite systems, the presence of the surface domain alters the character of the possible spinodal instability in an important way. Here, the very term can no longer be associated with a matter configuration that is uniform in space, but rather with one that is solely isotropic around the center of the system and is describable in terms of a nonuniform density profile. For iso-neutral matter considered in the present study, Boltzmann entropy is still a function of solely total excitation energy and the onset of spinodal instability can be still inferred from the appearance of the system caloric curve. The latter is illustrated in Fig. 4 along with the evolution of the two parameters, the half-density radius $R_{1/2}$ and the surface diffuseness (Süssman width [18]) parameter d, that were used to parameterize the error-function like [7] matter density profile. As seen in this figure, the spinodal instability sets now in at a significantly lower temperature than it did for bulk matter. This reflects the fact that the system has now an overall lower binding energy, but mostly the fact that the surface domain is more loosely bound than the bulk. One notes, that in the spinodal domain of a formally negative heat capacity, it is the surface domain alone that tends to expand with increasing energy, while the bulk tends actually to contract while staying at all energies safely outside of its own spinodal instability domain (*vide* Fig. 2). This is indicative of yet another new type of spinodal instability, the one where sections of the system within one fraction of the solid angle increase their surface diffuseness indefinitely while harvesting energy from the neighboring sections that undergo contraction of the surface domain, and while cooling down in the process. Since the spinodal domain has no upper limit in energy, the above scenario is bound to end with parts of the surface domain vaporizing into open space, leaving behind a metastable residue of a definite limiting temperature - a process named here surface spinodal vaporization.

4. Summary

In summary, the present study reveals the importance of thermal expansion for the fate of highly excited nuclear systems. It reveals the unavoidability of eventually entering the open-ended spinodal domain, when the excitation energy injected into the system is increased in excess of a certain critical energy per nucleon, that can be naturally associated with the boiling-point temperature. The study reveals a new, hitherto overlooked, decay mode of highly excited nuclei consisting in indefinite expansion of fragments of the surface domain ending in their vaporization - a process named surface spinodal vaporization. For the infinite matter the spinodal vaporization is a volume phenomenon entailing indefinite growth or vaporization of various parts of the system. While one can freely speculate about what is really happening in the spinodal domain, the underlying thermodynamic theory is helpless in this respect. What the latter can do, however, is to tell with certainty that when in spinodal domain, the system must promptly shead some parts of it via vaporization, before arriving at a state of compound-nuclear metastability.

The present study has numerous experimental implications, with the existence of a limiting temperature being the most prominent of them. It so happens, that the experimental verification of the existence of a limiting temperature is there since very many years [2] and was, actually awaiting a plausible theoretical explanation. Further, the model calculations imply that the vaporized matter is colder than the residue, a prediction that may possibly find an experimental confirmation. They imply also an unusually low latent heat of vaporization that is measured on the excitation energy scale from the boiling point energy per nucleon to the point of the onset of instability with respect to uniform expansion - a mere few MeV per nucleon (*vide* Fig. 2). Furthermore, the nature of the model calculations is such that one can expect effects of N-Z asymmetry. Such have been, indeed, already reported elsewhere [6].

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