

SURFACE BOILING – AN OBVIOUS BUT LIKE NO OTHER DECAY MODE OF HIGHLY EXCITED ATOMIC NUCLEI

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Essentials of a generalized compound nucleus model are introduced based on a concept of an open microcanonical ensemble which considers explicitly the role of the diffuse surface domain and of the thermal expansion of nuclear systems in the quest for maximum entropy. This obvious generalization offers a unique and universal thermodynamic framework for understanding the changes in the gross behavior of excited nuclear systems with increasing excitation energy and, specifically, the competition between different statistical decay modes, including classical evaporation and binary fission, but also the Coulomb fragmentation of excited systems into multiple fragments – the famed multifragmentation. Importantly, the formalism offers a natural explanation, in terms of boiling or *spinodal vaporization*, for the experimentally observed appearance of limiting excitation energy that can be thermalized by an excited nuclear system and the associated limiting temperature. It is shown that it is the thermal expansion that leads to volume boiling in an infinite matter and surface boiling in finite nuclei. The latter constitutes an important and universal, but hitherto unappreciated decay mode of highly excited nuclei, a mode here named *surface spinodal vaporization*. It is also shown that in iso-asymmetric systems, thermal expansion leads to what constitutes distillation – a decay mode here named *distillative spinodal vaporization*.

1. Introduction

The concept of a compound nucleus [1, 2] is one of the most fundamental concepts in nuclear theory, which has not faced serious challenges in its over 70 years of existence. Implemented in numerous computer codes, such as e.g., PACE[3] and GEMINI [4, 5] this concept has consistently provided a sound framework for interpreting a whole host of experimental observations, but has also provided a sound basis for nuclear microcanonical thermodynamics.[1] The concept of a compound nucleus rests on the assumption that an excited nucleus is a metastable object that is able to reach approximate microcanonical equilibrium, before finite fluctuations in particle energies and in the global shape bring it to one of the possible transition states for particle emission or binary Coulomb fragmentation, i.e., fission. One may view the collection of all possible transition states as a hypersurface in the whole $6N$ -dimensional phase space of particle coordinates and particle momenta, and the microcanonical equilibrium refers then to a part of the total phase space confined by the said hypersurface [6]. Obviously, no true microcanonical equilibrium is possible for nuclei excited in excess of particle separation energies, while such is possible for confined systems. We call here the system confined only by the hypersurface of transition states an “open microcanonical” system, acknowledging the fact that such a system is allowed to decay into the continuum whenever it reaches any (microscopic) state on this hypersurface. While not spelled out explicitly in the theory of compound nucleus, the existence of such a hypersurface is implied by the very use of Boltzmann’s entropy in the quantification of decay rates. The reason it is spelled out here is to contrast this kind of “liberal” self-confinement with the purely hypothetical “rigid” confinement in the global 3-dimensional coordinate space at the crux of most mainstream models and, more importantly, to stress the fact that the said “liberalism” of such a (thought) confinement has consequences far beyond those contemplated by standard implementations of the concept of a compound nucleus.

In its classical form, the theory of compound-nucleus recognizes two fundamental modes of statistical decay – particle evaporation and fission. In terms of the hypersurface of transition states, the former happens whenever statistical fluctuations bring the system to a point on the hypersurface which is associated with any particular particle continuum state. The latter happens when the point in question is a fragmentation saddle-point configuration. In its classical form, the model predicts Boltzmann-like scaling for the yields of decay products both, evaporative and fragmentation-like, with quantitative trends depending on transition-state energy for any particular decay channel.

While it is generally understood that the metastability at the crux of this model becomes not quite so well justifiable at elevated excitations, there has been no concerted theoretical effort undertaken to actually study quantitative criteria for the absolute loss of validity of this concept, should such criteria exist. Rather, to account for some important and intriguing experimental observations, vague and relative narratives have been proposed [7 - 9] for why the classical compound nucleus picture should be replaced with one that would apparently explain not only the appearance of certain modes of decay (such as , e.g., nuclear multifragmentation) but also their non-Boltzmannian, phase-transition-like scaling. These narratives fail to identify any particular “cross-over” point on the excitation energy or temperature scale, even as some experimental observations appear to be identifying such candidate points as, e.g., the point of the (rapid on the energy scale) onset of multifragmentation and the point where the limiting temperature would be reached.[7, 10, 11].

As revealed in a series of studies [6, 12 - 17], thermal expansion has profound qualitative effects on the behavior of excited nuclei some of which are only indirectly reflected in experimental observations but some other are directly observable in the decay modes and their interplay. At low excitations, the effects of thermal expansion appear insignificant, and it appears well justified to neglect them. However, as the excitation energy is raised, the compound system expands more and more and, as a result, lowers somewhat its temperature with respect to the non-expanded configuration. As shown in Ref. [15], this reduction in temperature leads to a reduction in Weisskopf’s [1] (evaporative)

decay rates and helps the system to maintain metastability. Additionally, thermal expansion allows for stronger local matter density fluctuations and global shape fluctuations which tend to further lower the system temperature. This kind of action by thermal expansion is consistent with Le Chatelier's principle requiring the system to respond to a stimulus (here, excitation energy) in a way that minimizes the effects of this stimulus (here, the increase in temperature). Further, the fact that the surface domain also expands thermally helps in reducing the surface tension beyond what results from a simple increase in surface entropy.[13] The latter then results in an increase in the magnitude of the global shape fluctuations and the relative enhancement of Coulomb fragmentation, [12, 13] as compared with particle evaporation. Such global shape fluctuations further contribute to the reduction of temperature and the retardation of statistical evaporation. In the light of the Le Chatelier's principle, one may then view at moderate excitation energies thermal expansion and fluctuations as a blessing for the concept of compound nucleus in that they appear to extend the time scales of statistical decay and, thus, allow for a more profound thermalization of the excitation energy.

Interestingly, thermal expansion of the surface domain and the resulting reduction in surface tension and, thus in fragmentation saddle energies, affect the fragmentation decay rates in a way that makes these rates to deviate substantially from the Boltzmann scaling and to resemble more phase-transition like scaling.[6] The latter scaling is characterized by a rapid onset on the energy scale, as it has been observed with respect to the onset of Coulomb fragmentation, usually called multifragmentation. One may posit that it is this action of thermal expansion on the Coulomb fragmentation rates that has given rise to numerous speculations linking nuclear multifragmentation to phase transitions.

The present study concentrates on the boiling phenomenon which appears tightly associated with thermal and not purely mechanical expansion.[16, 17] Boiling reflects the system becoming spinodally unstable with respect to local thermal fluctuation as the heat capacity of the system turns formally negative (because of expansion cooling). By boiling here is meant a phenomenon similar to that known from everyday life to occur to water heated in an open kettle. In a "run-up" to boiling, water is seen first to evaporate while remaining metastable, with the evaporation rate increasing with increasing temperature. What is less conspicuous but equally well known is that in the course of heating water expands. And then, upon reaching the boiling point specific energy, bubbles are seen to form chaotically throughout the volume, expand, and separate from water and finally, are seen to disappear into the surrounding open space. Also, temperature is seen to stay constant until the last drop has vaporized.

The present paper is constructed as follows: in Section 2, the employed theoretical formalism is discussed in terms of Fermi gas model in Thomas Fermi approximation followed by a revisiting of the essentials of spinodal instabilities in uniform matter. Then, results of calculations are presented as pertaining to boiling instabilities in infinite (Section 3, Subsection 1) and finite (Section 3, Subsection 2) iso-neutral matter, and in iso-asymmetric infinite matter (Section 3, Subsection 3). Then, Section 4 presents a discussion and summary.

2. Theoretical Formalism

The formalism employed in this study is described in detail in series of papers, [12 - 17, 6] all based on an expression for the level density parameter of little a as proposed in Ref. [18]. It aims at identifying the configuration of maximum entropy for any given excitation energy and then inspecting characteristics of interest for such configuration. By a configuration here is understood a particular macroscopic distribution of nuclear matter that is parameterized in terms of one or more parameters. The configuration entropy is written as:

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

where E is the system energy, E_{config} is the zero-temperature configuration energy and a_{config} is the level density parameter for the configuration of interest. Equation 1 is the base equation of the formalism, allowing one to evaluate entropy for any spatial matter density distribution $\rho_{config}(\mathbf{r})$. For a_{config} one writes: [18]

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

where α_o expresses the value of the level density parameter per nucleon at normal matter density ρ_o , I is the iso-asymmetry factor $I = (N - Z)/(N + Z)$, and $R(I)$ is the asymmetry-dependent factor[18] equal to $(1 - 1/9I^2)$.

The zero-temperature energy of a given configuration E_{config} was calculated by folding a Skyrme-type interaction energy density with a Gaussian folding function emulating the finite range of nuclear interaction [16]. Note that the folding is essential only in the case of finite nuclei and is not consequential for the bulk infinite matter.

The characteristic property of central interest in the present study is the curvature of the entropy function with respect to its arguments E and I . It is this curvature that decides whether the system can persist as metastable or will be forced to promptly decay via *spinodal vaporization* of a part of itself into the surrounding open space. Mathematically, this curvature is determined by the eigenvalues of the Hessian (a symmetric non-diagonal matrix built of second derivatives) of the entropy function, which is here defined as:

$$H(S) = \begin{bmatrix} \frac{\partial^2 S}{\partial E^2} & \frac{\partial^2 S}{\partial E \partial I} \\ \frac{\partial^2 S}{\partial I \partial E} & \frac{\partial^2 S}{\partial I^2} \end{bmatrix}. \quad (3)$$

For the uniform configuration of interest to be stable, the entropy function must be a concave function of its arguments and this means that both eigenvalues of Hessian must be negative and, thus, Hessian must be a negative definite matrix. Note that for symmetric or one-component matter, Hessian degenerates into a trivial 1x1 matrix with the second derivative of entropy with respect to energy as the only element and also the sole eigenvalue. Here, a practical condition for the entropy being a concave function of energy is the heat capacity to be positive and this condition is usually visualized in the form of a caloric curve of the temperature T plotted as a function of the excitation energy E .

Note that were the entropy a convex function of its arguments, the (uniform) configuration would be thermally unstable such that local fluctuations in the excitation energy would fail to generate restoring forces returning the system to the uniformity presumed for the configuration of maximum entropy. Rather, a driving force would be generated that would drive the system even further away from uniformity. Note also that the kind of instability associated with the “wrong” curvature of the characteristic thermodynamic function for a given type of ensemble is termed “spinodal instability” [19 - 21] and that its character depends on the type of (idealized) thermodynamic ensemble considered. For example, the most commonly discussed spinodal instability in a canonical ensemble is of mechanical type, associated in a synchronous manner with both, the negative compressibility and the negative chemical susceptibility (first derivative of the chemical potential with respect to concentration). The boiling phenomenon appears only in open microcanonical systems, is of thermal type and associated with negative heat capacity. It shows also negative chemical susceptibility as the entropy function depends in a nontrivial linear manner only on the energy per nucleon and not on the energy and the number of nucleons taken separately.

3. Results of calculations

3.1. Thermal instabilities in infinite systems

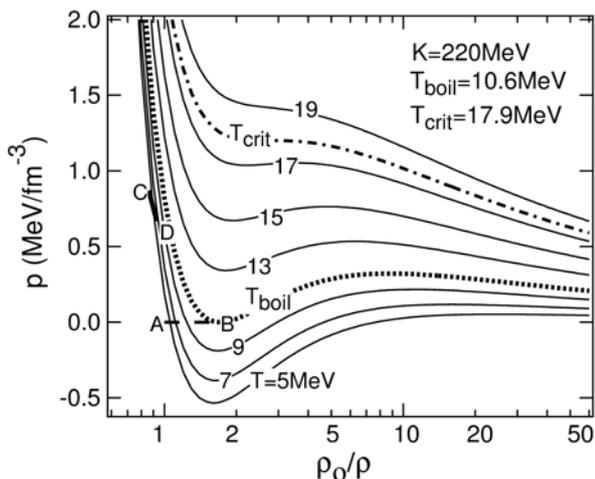


Fig. 1. Isotherms for the model matter. The isotherm corresponding to zero-pressure boiling-point temperature is shown in dotted line and the critical isotherm is shown in dash-dotted line. The adiabatic thermal expansion trajectory for the system at zero-pressure is shown in dashes as line AB, with B being the boiling point. The bulk interior expansion trajectory for finite system is shown in bold solid line CD.

b), and the appearance of convexity in the entropy (panels c and d), all as function of excitation energy per nucleon. The reduced entropy S_{red} shown in panel d is obtained by first subtracting a linear function in energy per nucleon from the entropy per nucleon shown in panel c, and then multiplying the result by a suitable normalization factor. Obviously, a subtraction of a linear function does not change the second derivative of the function, which is of interest here.

A simple explanation for the instability rests on the fact that when in the negative heat capacity domain, any fluctuation in local excitation energy per nucleon results in the recipient of the energy to cool down and the donor to heat up in terms of temperature. Subsequently, the colder recipient draws even more energy from the hotter donor in the quest for maximum entropy. This entropy driven transfer of heat and the resulting expansion of the colder, already

Fig. 1 illustrates isotherms obtained by evaluating the temperature T and pressure p by taking proper partial derivatives of the (uniform) configuration entropy given by Eq. 1, i.e.,

$$T = 1 / \left(\frac{\partial S}{\partial E} \right)_{V,N} \quad \text{and} \quad p = T \left(\frac{\partial S}{\partial V} \right)_{E,N}. \quad (4)$$

The calculations were done assuming a Skyrme-type equation of state with the incompressibility modulus of $K = 220$ MeV. As seen in Fig. 1, as the system expands with increasing excitation energy per nucleon starting from point A, its temperature first rises, what is evidenced by crossing of isotherms with progressively higher temperature labels. Then, beginning at point B, the temperature is seen to drop with a further increase of excitation energy and the induced expansion, an indication of spinodal instability of thermal type. The purpose of Fig. 1 is to demonstrate that the appearance of spinodal instability in an open microcanonical system is both, trivial and unavoidable for any system characterized by a van der Waals-type equation of state.

Fig. 2 illustrates in more detail thermal expansion (panel a), the appearance of negative heat capacity (panel

expanded part is the essence of the boiling process. The onset of thermal instability can be seen also in a purely microcanonical representation, without recourse to the notion of (microcanonical) temperature. This is seen in Fig. 3, where the entropy is shown as a function of the excitation energy per nucleon and the asymmetry in the excitation energy division between two hypothetical equal-size subsystems.

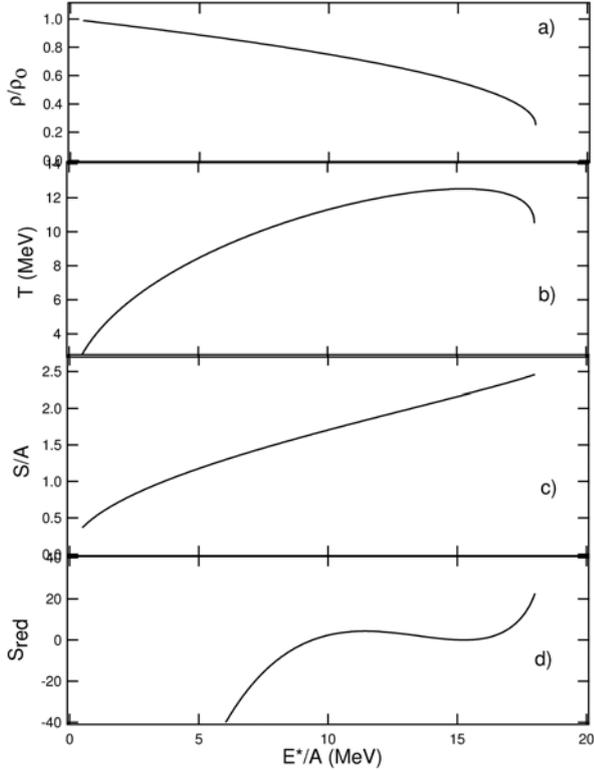


Fig. 2. Evolution of the matter density (a); the microcanonical temperature (b); the entropy (c), and the reduced entropy (d) with the excitation energy per nucleon. The appearance of convexity is made conspicuous in panel (d) through a suitable curvature-neutral linear transformation.

As is seen in Fig 3, at lower excitations, the entropy favors a uniform distribution of the excitation energy, while allowing for finite fluctuations in excitation energy division between the arbitrarily chosen split of the entire system into two equal-size subsystems. At elevated energies, this trend is reversed and the entropy now favors unequal energy distribution – the essence of spinodal instability.

It is important to note that once the convexity sets in for the entropy function at the boiling-point excitation energy per nucleon, it stays there up to the end point of the curves shown in Fig. 2, where the system becomes unstable with respect to uniform expansion. This means that part of the system must separate physically from the surviving metastable residue with the “departing” temperature lower than that of the residue. Here, the boiling process is named also as *spinodal vaporization*, to reflect its prompt character and its roots in spinodal instability.

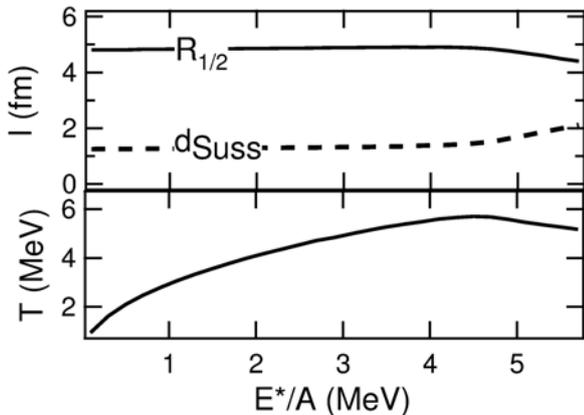


Fig. 4. Matter density distribution parameters (top panel) and the microcanonical temperature (bottom panel) as functions of the excitation energy per nucleon. (See text).

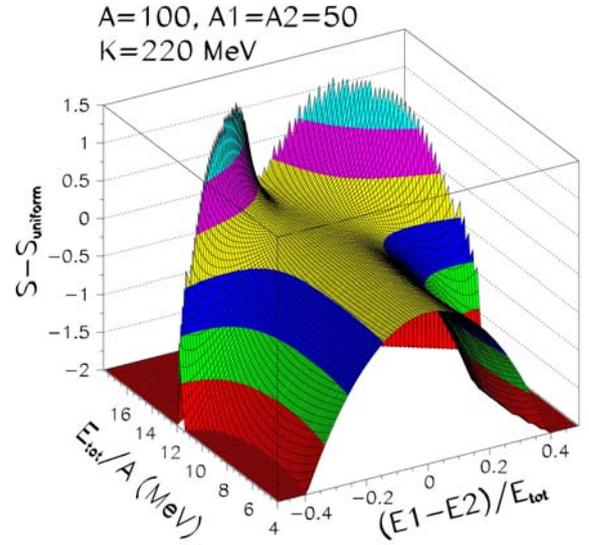


Fig. 3. (Color online) Entropy of a two-phase system with respect to that of the uniform system, plotted as a function of total excitation energy per nucleon and the asymmetry in energy distribution between the two equal-size subsystems.

3.2. Boiling in finite nuclei

Calculations for finite nuclei were performed assuming a matter density profile given by the error function, [18] with the half-density radius R_{half} and the surface width d as the two parameters:

$$\frac{\rho(r)}{\rho_0} = C(R_{half}, d) \left[1 - \operatorname{erf} \left(\frac{r - R_{half}}{\sqrt{2}d} \right) \right], \quad (5)$$

The results of maximizing entropy for the excitation energy range for which the maximum of entropy as a function of R_{half} and d exists, are shown in Fig. 4.

As seen in Fig. 4, with increasing excitation energy, both the half-density radius and the surface domain width

(Süssman width) first increase, as does the microcanonical temperature T . Then, beginning around 4.5 MeV/nucleon, T reverses the trend, indicating the onset of thermal spinodal instability – in this case, the surface boiling. At the same time the surface diffuseness begins increasing more rapidly. The trends seen in Fig. 4 are indicative of a thermal instability where one section of surface domain increases its diffuseness by drawing energy from the neighboring section and by cooling down as a result. It is obvious from Fig. 4 that once the system enters the spinodal domain, it cannot regain stability before the excess energy is shed along with a portion of the system itself. The boiling here constitutes a prompt decay of the excited system via diffusion of some parts of the surface away. By diffusion, it is here understood a process of a steady increase in surface diffuseness, until parts of the system separate.

Note that the surface boiling is a novel and important mode of decay of realistic nuclei, with many real and potential consequences. It is also a new kind of spinodal instability, different from the classical one where the instability applies uniformly to every infinitesimally small part of the uniform system. Here the system is uniform only in the sense of having an isotropic matter density profile and, accordingly, the instability applies here to portions of matter enclosed in infinitesimally small solid angles as viewed from the center of the system. Here, it is proposed to name this process as *surface spinodal vaporization*.

3.3. Distillative boiling in iso-asymmetric matter

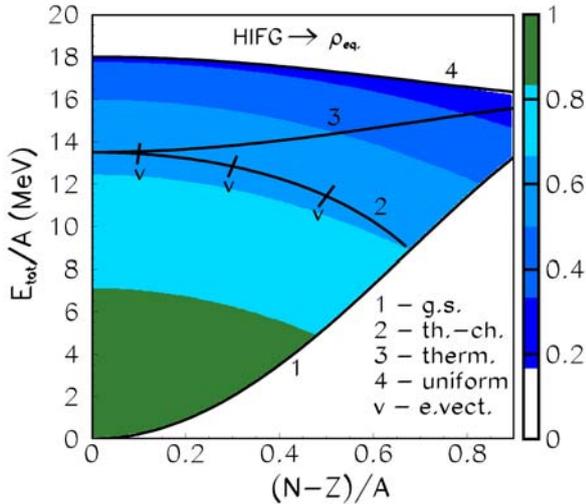


Fig. 5. (Color online) Contour plot of the equilibrium matter density as a function of iso-asymmetry, $(N - Z)/A$, and energy per nucleon E_{tot}/A . Shown are in solid lines the ground-state energy (I), the boundary of the metastability domain (2), the boundary of the domain of positive heat capacity (3), and the boundary of the domain stable against uniform expansion (4).

portion of matter that manages to reach it, such that the portion of matter in question would expand indefinitely purely on the account of its own energy. Line 2 in this Figure represents the locus of points where one of the eigenvalues of the Hessian of entropy $H(S)$ turns zero, both eigenvalues being duly negative in the domain of metastability enclosed between the line 1 illustrating the ground state energy as a function of iso-asymmetry I , and the spinodal boundary line 2. Line 3 on this plot serves illustrative purpose only and represents the locus of points where the heat capacity turns infinite, i.e. where the purely thermal instability would set in had it not been preempted by the thermo-chemical instability at line 2. The fact that the latter instability is, indeed, thermo-chemical is evidenced by the orientation of eigenvectors associated with the zero eigenvalues and represented in Fig. 5 by short bars labeled as \mathbf{v} . And, again, as in the case of iso-neutral matter, once the system enters the domain of thermo-chemical spinodal stability above line 2, it never regains stability as a whole. As indicated by the short bars, the less dense matter that eventually ends up boiling off at line 4 is more neutron-rich and colder than the residue. Note that line 2 may be considered an attractor line, i.e., a collection of points describing the metastable state of the residue left after the excess energy is boiled off, an entity that can be probed experimentally, in principle.

It is worth noting that the “asymmetric” (in iso-asymmetry) boiling is reminiscent of isospin fractionation in confined hypothetical systems, except that in open systems no equilibrated gas phase is ever present. Here, it is proposed to name such a process as *distillative spinodal vaporization*.

4. Summary

Boiling is an obvious and a very common phenomenon that must happen in highly excited systems featuring a van der Waals-type equation of state. Boiling is known from everyday life as a state where bubbles are appearing chaotically throughout the volume of water, growing, and then separating from water and disappearing in the

Iso-asymmetric self-confined matter is characterized by an entropy that is a function of two extensive arguments, energy E and iso-spin asymmetry I . Accordingly, the Hessian of entropy is a 2×2 non-diagonal matrix, which has two eigenvalues and two eigenvectors, with the latter not aligned with the argument axes E or I .

Results of calculations for such a model system are displayed in Fig. 5 in the form of a contour plot of the matter equilibrium density as a function of energy and iso-spin parameter. They were obtained assuming a harmonic-interaction EOS with a symmetry term linear in matter density:

$$E_{config} = c_V \left(1 - \frac{\rho}{\rho_0}\right)^2 + c_I \frac{\rho}{\rho_0} I^2, \quad (6)$$

where the parameters c_V and c_I were assumed to be -16 MeV and 23 MeV, respectively.

In Fig. 5, overlaid on the contour plot are several boundary lines of significance here.

As seen in Fig. 5, the matter density decreases with increasing energy and, eventually, at the boundary line 4 becomes unstable (globally) against uniform expansion. Line 4 in this Figure is a line of “no return” for any

surrounding open space. In self-bound infinite nuclear matter brought to boiling, also bubbles are expected to be formed and growing indefinitely and never in thermal equilibrium with the liquid phase. While such bubble formation in infinite systems may be of a true academic interest, it is the surface boiling that has implications as far as the “life” of a realistic highly excited nuclear system is concerned. As revealed in the present paper, the surface boiling occurs via spontaneous unrestrained expansion of parts of the surface domain at the expense of the energy derived from the neighboring parts. This expansion results in forced prompt *spinodal vaporization* of a part of the system into surrounding open space, clearly a decay process different from all other decay modes so far considered. The decay here is different from classical pre-equilibrium decay as it does not rely on any particular initial distribution of particle momenta and coordinates in the system. As is clear from the presented formalism, even if one could purely hypothetically manage to form a seemingly equilibrated “dream” system at maximum entropy with isotropic density profile, such a system would still decay promptly via *spinodal vaporization* - shedding of the excess excitation energy along with a definite part of itself. And it will do so in an isotropic manner (assuming that the system has zero angular momentum) with vapors being colder than the surviving metastable residue.

Decay via surface boiling, or *surface spinodal vaporization*, is intimately linked to thermal expansion and sets in rapidly on the excitation energy scale, in a stark contrast to Boltzmann-like scaling of yields in classical model implementations of the concept of a compound nucleus. It is then reasonable to assume that the decay processes commonly identified as statistical occur only after the prompt boiling process has completed and has left the metastable residue to decay via particle evaporation and Coulomb fragmentation, both, binary and multifragment.

The most prominent experimental observation that is trivially explained in terms of boiling, but has no other plausible explanation, is the appearance of limiting temperature in the measured caloric curves. Clearly, it will be of great interest to carry out experiments involving systems with different initial N/Z asymmetries to get handle on iso-spin dependence of nuclear EOS at sub-normal densities. To interpret the experimental data from such experiments, one would have to carry out very CPU-intensive calculations for finite iso-asymmetric systems, possibly including Coulomb interaction.

In summary, boiling is a prompt decay mode of highly excited nuclear systems rich in detectable, experimentally verifiable consequences. Because of its nature, it may be called *spinodal vaporization*. It is also rich in theoretical implications and offers a fertile ground for reasonable and stimulating scientific speculations. One may posit that it is one of the most overlooked phenomena in nuclear thermodynamics in recent memory and the one which, while being obvious and having a robust confirmation in a unique set of experimental observations, still awaits recognition by the nuclear science community.

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