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Boiling Patterns of Iso-Asymmetric Nuclear Matter

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Abstract. Limits of thermodynamic metastability of self-bound neutron-rich nuclear matter are explored within the framework of microcanonical thermodynamics of interacting Fermi Gas model in Thomas-Fermi approximation. It is found that as the excitation energy per nucleon of the system is increased beyond a certain limiting value, the system loses metastability and becomes unstable with respect to joint fluctuations in excitation energy per nucleon and in isospin per nucleon. As a result, part of the system is forced to boil off in a form of iso-rich non-equilibrated vapors. Left behind in such a process, identifiable with distillation, is a more iso-symmetric metastable residue at a temperature characteristic of its residual isospin content. With a progressing increase in the initial excitation energy per nucleon, more neutron-rich matter is boiled off and a more iso-symmetric residue is left behind with progressively increasing characteristic temperature. Eventually, when all excess neutrons are shed, the system boils uniformly with a further supply of excitation energy, leaving behind a smaller and smaller residue at a characteristic boiling-point temperature of iso-symmetric matter.

1. Introduction

One of the issues of central interest to nuclear science is the issue of thermodynamic stability of nuclear systems both, infinite and finite and search for the signatures of the loss of stability.[1, 3, 2, 4] The issue is usually framed in terms of spinodal instability of a mechanical kind associated with the appearance of negative compressibility and the associated loss of restoring forces to the matter density fluctuations.[5] In the confined thermodynamical ensembles commonly considered, this kind of instability leads always to phase separation, with a stable final configuration of coexisting liquid and gas phases. In systems endowed with isospin, the spinodal instability that appears in certain conditions is somewhat more complex and involves joint fluctuations in neutron and proton matter densities separately. The loss of stability against such joint fluctuations leads also to phase separation but with liquid and gas phases characterized by different neutron-to-proton ratios, commonly referred to as isospin fractionation.

The studies of spinodal instabilities in confined thermodynamical ensembles of choice are undoubtedly of substantial academic interest as they, e.g., help understanding what would happen to the matter under different conditions. However, their practical utility for understanding the actual behavior of realistic excited nuclei is far from obvious. This is so, because there is no practical way of confining nuclear matter in a fixed volume and there is no real rationale for expecting thermodynamic equilibrium in any fixed volume but without a confinement vessel present.

In contrast to idealized thermodynamical ensembles, excited atomic nuclei as studied in heavy ion collisions never are stable. They are at most metastable and it is this metastability that is at the crux of the sound nuclear thermodynamics embodied in the time-proven concept of

compound nucleus. This concept views an excited nucleus as an open microcanonical system that is confined only by a thought hypersurface of transition states, a collection of particle emission barriers and fragmentation saddle points, which lead to open decay channels. The latter hypersurface is thought to exist in the full $6N$ -dimensional coordinate and momentum phase space of an N -particle system, and not solely in the “primitive” 3-dimensional coordinate space as is the case with the famed but *ad hoc* freezeout volume. Within the confines of the said hypersurface, the excited compound nucleus is thought to be fully equilibrated in the microcanonical sense and free to undergo all possible fluctuations and, among other, those in energies of individual particles, those in global shape, those in local neutron and proton matter densities, and local energy densities. Such a compound system is thought to decay via statistical evaporation whenever finite fluctuations in particle energies bring it to a continuum state at the boundary of the confining hypersurface and it is thought to fragment, whenever finite fluctuations in shape bring it to a fragmentation saddle configuration at the confining hypersurface. One notes, that the notion of the hypersurface of the transition states, even if usually not spelled out explicitly, is implied by the very construction of the compound nucleus model, allowing the system to occupy different volumes for different decay modes and not forbidding thermal expansion in principle.

While it was generally accepted that at elevated excitations the concept of a quasi-equilibrated compound nucleus may lose validity, until very recently [6, 7] missing were theoretical studies of thermodynamic limits of metastability of compound nuclei, i.e., the limits of applicability of the very concept of compound nucleus. These limits are intimately linked to thermal expansion and the fluctuations in local matter and energy densities, both of which are generally deemed irrelevant in the practical model implementations of the concept of a compound nucleus. In a sense, they may be indeed considered irrelevant up to the point when they become ultimately relevant to the extent of invalidating the very concept of a quasi-equilibrated compound nucleus.

It is amusing and worth noting the important and complex but little known or appreciated “behind the scene” role of thermal expansion and fluctuations in matter and energy density distribution in the fight of a compound nucleus for survival. This role comes first as a blessing when such fluctuations along with thermal expansion reduce the temperature of the compound nucleus and retard statistical decay processes.[8] Then, upon reaching the boiling-point excitation energy, this role comes as a curse as it tends to invalidate the very *raison d’être* of the compound nucleus as a metastable object. And then, the same expansion and fluctuations come again as a blessing in that they help the compound system to promptly shed excess energy along with just a part of the system so the rest can survive as a metastable residue. When viewed through the lens of the Le Chatelier’s principle, such a picture adds certain, perhaps unexpected and discreet, charm to the apparently strict and “dry” thermodynamic theory, dominated at low excitation energies by the “boring” Boltzmann scaling. It is the thermal expansion that makes interesting thing to happen much differently than in accordance with the latter scaling and more like according to phase-transition like scaling, [16] rapidly on the excitation energy scale. Boiling is one of such interesting and “sudden” phenomena, which owes it to both, thermal expansion and unconstrained fluctuations.

As was revealed recently,[6, 7] when the excitation energy of an open nuclear system is increased beyond a certain value, the system becomes unstable with respect to boiling, a special case of spinodal instability occurring solely in open microcanonical systems. Under the term “boiling” it is here understood the phenomenon known from the everyday life when upon supplying the heat energy to a liquid (water), the liquid is first seen to “gently” evaporate its parts into the open space, while undergoing thermal expansion. Subsequently, upon reaching the boiling point, the liquid is seen to cease to “behave”, such that various parts of it are seen to expand spontaneously in the form of bubbles and these bubbles are seen to leave the system and disappearing into the open space. The same is bound to happen to nuclear matter brought to

boiling-point excitation as this matter is also characterized by the van der Waals type equation of state. In fact, given what is known about the nuclear EOS, it appears absolutely inconceivable for the highly excited nuclear systems being immune to boiling.

One is tempted to posit that boiling phenomenon in unconfined bulk Fermi liquid and the surface boiling in finite nuclei, while obvious, is the most overlooked and under-appreciated phenomenon in nuclear thermodynamics in recent memory. As discussed in greater detail below, it is also a phenomenon with an obvious importance to understanding a host of known experimental observations [9, 10] and one with “inviting” characteristics that can be potentially verified experimentally, all tied tightly to the specifics of nuclear equation of state.

In the present study we discuss first in some detail the essentials of spinodal instabilities in general (Section 2). Then in Sections 3 and 4 the boiling variant of such an instability is discussed for iso-neutral open microcanonical ensembles along with its hallmark signature, the presence of an apparent limiting temperature. In Section 5 characteristics of boiling instabilities are studied for iso-asymmetric open microcanonical systems and Section 6 presents a summary.

2. Spinodal instabilities and trivial ensemble inequivalence

Generally, the term “spinodal instability” is used to refer to a particular state of uniform matter in which any infinitesimally small local fluctuation in uniformity fails to give rise to restoring forces that would return the matter to uniformity but, instead, results in forces driving the system even further away from uniformity. It is commonly being discussed in terms of the character of the formal dependence of the characteristic function of the thermodynamical ensemble considered on its extensive parameters, volume, numbers of neutrons and protons, and energy - whichever applies.[5] For a uniform system to be stable, its characteristic thermodynamic function must be either concave or convex with respect to its extensive arguments, depending on the type of thermodynamic ensemble considered and, in particular, on whether this function is expected to be at maximum (concavity required for entropy) or minimum (convexity required for free energy) in the state of thermodynamic equilibrium. If the opposite is true, i.e., if the entropy function develops convexity or the free energy function develops concavity, the system will increase its entropy or decrease its free energy, respectively, when “going” non-uniform. In other words, when formally in spinodal domain, the system will prefer non-uniformity over uniformity, a preference that in confined systems results in phase separation and in open systems results in parts of the system vaporizing into the surrounding open space. Mathematically, the concavity or convexity of a function is determined by the eigenvalues of its Hessian, a symmetric matrix known also as the curvature matrix and built of second derivatives of this function with respect to its arguments. Specifically, for a function to be concave (entropy), all eigenvalues of its Hessian must be negative, which means that Hessian must be negative definite. On the other hand, for a function to be convex, all its eigenvalues must be positive, which means that Hessian must be positive definite. These mathematical criteria for the stability are more stringent than those requiring simply second derivative with respect to individual arguments to be of a particular sign and they account for the fact that the convexity or concavity is not expected to be aligned with the argument axes but rather with the eigenvectors of Hessian.

Note that the spinodal instability is of differential nature in a sense that it applies uniformly to any infinitesimally small portion of the system independently of what happens to the rest of the system. This is in contrast to a possible global instability such as, e.g., with respect to uniform expansion or with respect to spheroidal or quadrupole deformation.

As discussed further below, the detailed nature of a spinodal instability depends on the thermodynamic ensemble considered to the extent that in some ensembles such as grand canonical and iso-neutral isothermal-isobaric spinodal instability does not occur at all. In fact, the very presence of such trivial ensemble inequivalence stems from the fact that the spinodal

instabilities are all about the spontaneous growth of fluctuations, whether mechanical, thermal, chemical, or of mixed nature and that the idealized thermodynamical ensembles differ exactly in the way they suppress, only theoretically, fluctuations. It is also this ensemble inequivalence that helps to understand why the spinodal instability of boiling type could have gone unnoticed for all this time and namely the fact that such instability is expected to occur only in an unconventional open microcanonical ensemble that has attracted almost no serious theoretical attention.

2.1. Spinodal instabilities in canonical ensembles

The state of an iso-neutral canonical ensemble is fully determined by the volume V , the number of particles N , and the canonical temperature T and is encoded in the Helmholtz free energy A as a function of these three arguments:

$$A = A(N, V, T) \quad (1)$$

From the three arguments, only two, N and V are extensive. However, for the uniform matter of interest, these two are not independent arguments as the free energy depends non-trivially only on their ratio, the matter density ρ . Therefore, the curvatures of Helmholtz free energy function with respect to both of these arguments are strictly correlated and for the purpose of the stability consideration it is sufficient to consider either the second partial derivative of $A(N, V, T)$ with respect to volume V (at constant T and N) or the second partial derivative of $A(N, V, T)$ with respect N , at constant T and V . The first expresses the bulk isothermal incompressibility modulus and the second one expresses the bulk isothermal chemical susceptibility. Their signs are always equal and must be positive for the system to be stable against the density fluctuations. Which means that to study the onset of instability it is sufficient to check at which point either the incompressibility modulus or the chemical susceptibility turns negative and the ensuing instability with respect to density fluctuations is of mechanical type. For iso-asymmetric canonical ensemble, there is an additional extensive argument of iso-asymmetry $I = (N - Z)/(N + Z)$ and, accordingly, the relevant Hessian is a non-diagonal 2x2 matrix and the nature of spinodal instabilities is here of chemo-mechanical type.

2.2. Spinodal instabilities in confined microcanonical ensembles

The state of an iso-neutral microcanonical ensemble is fully determined by the volume V , the number of particles N , and the energy E and is encoded in the Boltzmann entropy S as a function of these three extensive arguments:

$$S = S(N, V, E) \quad (2)$$

Here, all three arguments are extensive but similarly to what was discussed in the previous section for canonical ensembles, curvatures with respect to N and V are strictly correlated and one needs to consider only 2x2 Hessians either in the space of (N, E) or space of (V, E) . The ensuing instabilities are here of thermo-mechanical kind. In the case of iso-asymmetric matter there is again the additional extensive argument of iso-asymmetry I and, accordingly, the spinodal instabilities are here of thermo-chemo-mechanical type, following from the fact that Hessian is a non-diagonal 3x3 matrix.

2.3. Spinodal instabilities in isothermal-isobaric ensembles

The state of an iso-neutral isothermal-isobaric ensemble is fully determined by two intensive arguments of temperature T and pressure p and one extensive argument of volume V and is encoded in the Gibbs free energy as a function of these arguments.

$$G = G(N, T, p) \quad (3)$$

Here, only one argument N is extensive and the dependence of G on N is trivial linear. Because of the latter, no spinodal instability can be present in iso-neutral isothermal-isobaric ensembles. In the case of iso-asymmetric ensemble, the additional extensive argument of iso-asymmetry I allows for spinodal instabilities of purely chemical type to appear.

2.4. Spinodal instabilities in grand canonical ensembles

The state of a grand canonical ensemble, whether iso-neutral or iso-asymmetric, is fully determined by the intensive arguments of chemical potentials for neutrons and protons μ_n and μ_p and temperature T and the sole extensive argument of volume V and is encoded in the Landau potential:

$$L = L(V, \mu_n, \mu_p, T) \quad (4)$$

Since the dependence of L on V is here trivial linear, no spinodal instabilities will ever appear in grand canonical ensembles.

3. Boiling phenomenon in open microcanonical self-confined nuclear matter

As was pointed out in the Introduction, realistic excited nuclear systems are at best metastable. Importantly, they are also self-bound and isolated, and can thus be considered microcanonical thermodynamic objects as long as the metastability holds. Here we consider microcanonical thermodynamics of such systems within the framework of Fermi gas model in Thomas-Fermi approximation. The system is presumed to adapt its matter distribution by the way of thermal expansion so as to arrive at maximum entropy and, accordingly, is characterized by an entropy function that for iso-neutral systems is a function of only one extensive argument, the energy E and for iso-asymmetric systems is a function of two extensive arguments, the energy E and the iso-asymmetry parameter I , i.e., $S = S(E, I)$. Since the system by the nature of “liberal” microcanonical constraints is allowed to undergo fluctuations in both the local energy density, and in isospin density, such will eventually at elevated energies destabilize the system in a spinodal sense. That arriving at the spinodal instability of thermal kind is unavoidable for open iso-neutral systems at zero pressure is obvious already from the known appearance of the van der Waals’ian isotherms as illustrated in Fig. 1.

The isotherms were obtained by using a general microcanonical formalism as used earlier in references [11] - [16] and assuming a Skyrme-based EOS with a compressibility constant of $K=220$ MeV . In this formalism, the Boltzmann entropy for the (uniform) configuration considered is expressed generally as:

$$S_{config} = \sqrt{(E - E_{config})a_{config}}, \quad (5)$$

where E_{config} is the configuration energy and a_{config} is the level density parameter. The latter can be expressed as [11]

$$a_{config} = \alpha_o \rho_o^{2/3} \int \int \int \rho^{1/3}(\vec{r}) d\vec{r}, \quad (6)$$

where α_o expresses the value of the level density parameter per nucleon at normal matter density ρ_o . The configuration energy E_{config} was calculated by folding a Skyrme-type interaction energy density with a Gaussian folding length emulating finite range of nuclear interaction. The folding is essential only in the case of finite nuclei considered further below and is not consequential for bulk matter. Details of the formalism employed are given in Ref. [7]. As seen in Fig. 1, as the excitation energy of bulk matter at zero pressure (maximum entropy) is increased starting at point A , the system expands and progresses toward point B . The system first “visits” isotherms with increasing temperatures but beginning at point B , it begins “visiting” isotherms with

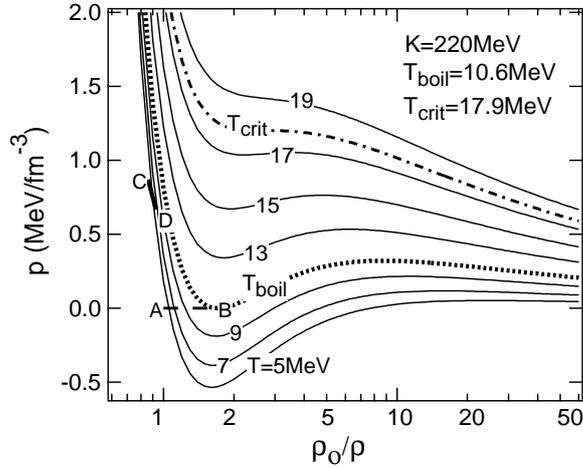


Figure 1. Isotherms for the model matter. The isotherm corresponding to zero-pressure boiling temperature is shown in dotted line and the critical isotherm is shown in dash-dotted line. The adiabatic trajectory for a hypothetical infinite system is shown in dashes (line AB), while such for the bulk of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

decreasing temperatures, all while continuing to expand thermally and increasing excitation energy per nucleon. Clearly at point B the system enters spinodal domain of negative heat capacity, i.e., convexity of the entropy function and becomes thermally unstable. This is further illustrated in Fig. 2 but here, the very purpose of Fig. 1 is to demonstrate how obvious it is that any open system (at zero pressure) with classical van der Waals'ian isotherms must become thermally unstable at some excitation energy and must begin boiling.

Fig. 2 illustrates thermal expansion and the appearance of negative heat capacity at the boiling point. Note that the expansion continues after reaching the boiling point excitation energy per nucleon and never stops before the matter reaches the end of the curve, the “point of no return” where it becomes unstable with respect to uniform expansion and able to escape the system in the form of vapor on its own supply of energy. Beginning at the boiling point, infinitesimal fluctuations in excitation energy density would be growing at the expense of energy derived from the surrounding matter and the system would not regain metastability before all excess excitation energy is shed via prompt vaporization called boiling.

Panel (c) illustrates entropy as a function of energy and the fact that this function, originally concave, turns convex at the boiling point. This is even more clear in panel (d) where a reduced entropy is displayed obtained by subtracting a suitable linear function from the entropy function of panel (c) and by multiplying the difference by a constant. Obviously, the subtraction of a linear function preserves the curvature which now is, however, more clearly visible.

Note that in such calculations, entropy is a function of energy only, as the volume is adapted so as to maximize entropy. In terms of Hessian analysis, here the Hessian reduces to a 1×1 matrix featuring only the second derivative of entropy with respect to energy, which is also the eigenvalue of this Hessian. A negative value of this eigenvalue indicates concavity of the entropy function and, thus, the stability of the system. Conversely, the positive value indicates both, the convexity and negative heat capacity - standard signals for thermal instability.

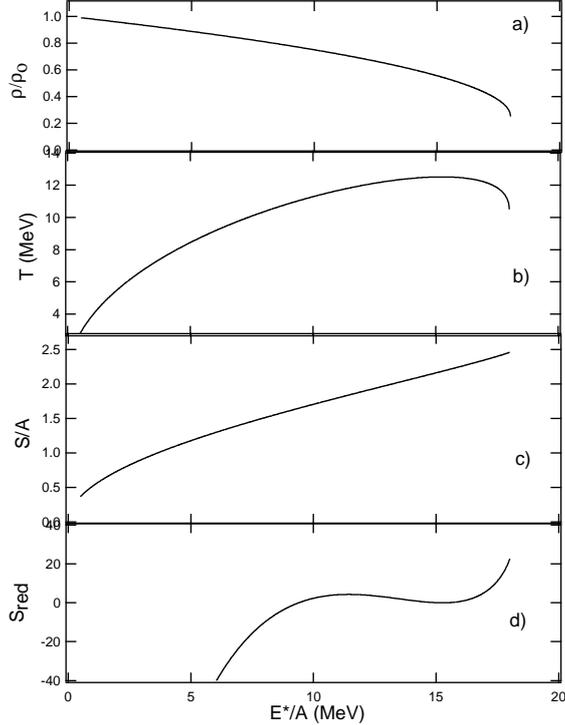


Figure 2. Destabilizing consequences of thermal expansion (a) of bulk Fermi liquid for the caloric curve (b) and the curvature of the entropy function (c,d). Panel d illustrates the reduced entropy obtained by subtracting a linear function from the entropy function and multiplying the result by a constant, such as to better visualize the convexity associated with boiling.

4. Surface boiling in a finite nuclear system

To study boiling in finite systems, one must account for the finite range of nuclear interaction and this was done via folding the interaction energy with a gaussian function with a folding width adjusted to reproduce the experimental ground-state surface diffuseness [7]. Then, the energy function was constructed for a family of matter density profiles parameterized in terms of error function:

$$\frac{\rho}{\rho_0} = C(R_{half}, d) \left[1 - \operatorname{erf} \left(\frac{r - R_{half}}{\sqrt{2}d} \right) \right], \quad (7)$$

where R_{half} and d are the half-density radius and the Süssmann surface width, respectively, and $C(R_{half}, d)$ is a normalization factor assuring the desired number of nucleons in the system (here, $A=100$). In the calculations, the half-density radius and the surface diffuseness were varied so as to obtain maximum entropy for a given excitation energy. Obviously, the entropy is here again a function of just energy and, again, Hessian analysis is just in one dimension and equivalent to inspecting the heat capacity as a function of excitation energy.

The salient results of calculations for finite nucleus with $A=100$ are displayed in Fig. 3 illustrating the trends in the matter distribution and in caloric curve as functions of excitation energy. As seen in the top panel of this figure, thermal expansion manifests itself first through a systematic increase in half-density radius $R_{1/2}$ and Süssmann width of the surface domain. However, beginning at $E \approx 4.5$ MeV/nucleon, the surface width begins to grow more rapidly, while the half-density radius begins to contract. Importantly, the heat capacity turns negative at this point, indicating onset of instability of a new kind - a surface boiling or surface spinodal.

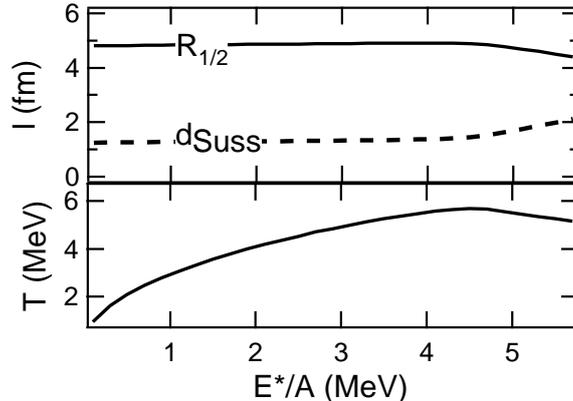


Figure 3. Evolution of finite system parameters with the excitation energy.

By the logic of the boiling discussed further above, the finite nucleus cannot be stable at energies in excess of the energy of the surface boiling point.

Note that here the spinodal instability is different from the “classical” one where the property of being unstable applies uniformly to any infinitesimally small element of volume. Here, the property applies uniformly to infinitesimally small elements of solid angle in a coordinate system that has origin in the center of the nucleus. Also the uniformity of the matter distribution refers here merely to its isotropy and not to a spatial uniformity as in the case of the bulk matter. This kind of instability indicates that matter in one element of solid angle would derive energy from the matter in the neighboring element and as a result cool down. Subsequently, it would derive even more energy in virtue of being colder. As seen in Fig. 3, surface diffuseness would increase for the “acceptor” matter while it would be decreasing for the donor matter. Physically it is indicative of parts of the surface domain “diffusing” away and ultimately escaping in the form of vapors. It is worth noting that the surface boiling sets in at a temperature much lower than that for bulk boiling and quite consistent with experimental observations. It also preempts volume boiling from showing up in the form of bubbles being formed in the bulk interior.

5. Boiling in iso-asymmetric matter

The calculations for iso-asymmetric matter were performed only for bulk matter and assuming a schematic harmonic-interaction isospin dependent configuration energy with an isospin-dependent term proportional to the matter density ρ :

$$E_{config} = c_V \left(1 - \frac{\rho}{\rho_0}\right)^2 + c_I \frac{\rho}{\rho_0} I^2, \quad (8)$$

where I is an iso-asymmetry parameter related to the numbers of neutrons and protons, N and Z , respectively via $I = (N - Z)/(N + Z)$.

Also, the level density parameter a_0 was corrected for the iso-asymmetry by multiplying the expression on the right-hand side of Eq. 6 by a factor of $R_I = (1 - 1/9I^2)$ [11]. In calculations of entropy, the fundamental Eq. 5 was then used and it was assumed that $c_V = -16$ MeV, $c_I = 23$ MeV, and $a_0 = A/15$ MeV⁻¹. Subsequently, to search for the onset of spinodal instabilities, Hessian of entropy function was constructed and diagonalized over the whole range in energy and iso-asymmetry, where the system was stable with respect to uniform expansion.

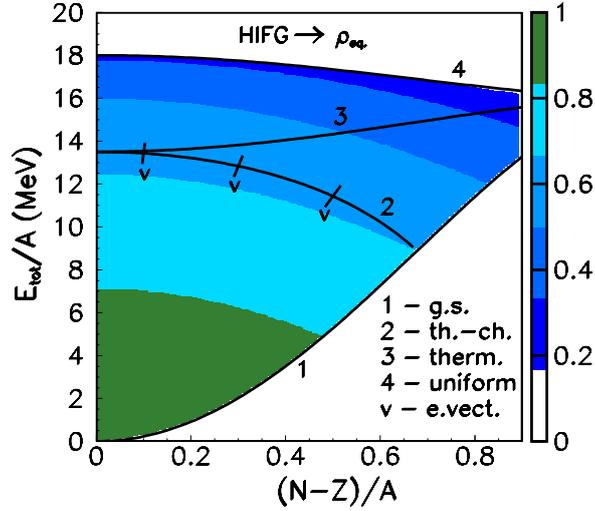


Figure 4. (Color on-line) Contour plot of the equilibrium matter density as a function of the total energy and iso-asymmetry. Lines represent the ground-state energy (1), the boundary of thermo-chemical (2) and purely thermal (3) stabilities, and the limit of stability with respect to uniform expansion (4). The short bars labeled as *v* represent the directions of the thermo-chemical instability given by eigenvectors of Hessian matrix of the entropy function associated with null eigenvalues.

The Hessian of the entropy function is in this case a 2x2 diagonal matrix:

$$H(S) = \begin{bmatrix} \frac{\partial^2 S}{\partial E^2} & \frac{\partial^2 S}{\partial E \partial I} \\ \frac{\partial^2 S}{\partial E \partial I} & \frac{\partial^2 S}{\partial I^2} \end{bmatrix}. \quad (9)$$

Because of the simplified harmonic-interaction EOS, all partial derivatives could be calculated here analytically and also, the subsequent diagonalization of the Hessian could be carried out analytically.

Results of calculations for iso-asymmetric self-contained bulk matter are illustrated in Fig. 4 in a form of a contour plot of matter equilibrium density as a function of iso-asymmetry parameter I and total energy per nucleon E_{tot}/A . It is important to note that the latter argument is different from the total excitation energy in that it includes also the iso-asymmetry-dependent ground-state binding energy. Overlaid on this contour plot are lines illustrating important boundaries - the ground state energy (1), the “ultimate” boundary of the domain where the system is stable with respect to uniform expansion (4), the boundary of the domain of positive heat capacitance (3), and the boundary of the domain of spinodal stability (2), the most demanding domain.

As seen in Fig. 4 the system becomes subject to thermo-chemical spinodal instability (line 2) on the energy scale well before it becomes purely thermally unstable (line 3) which makes the thermo-chemical instability the leading instability in this case. Mathematically, line 2 represents the locus or collection of points where the Hessian ceases to be negative definite, i.e., where one of its eigenvalues turns positive. In contrast, line 3 represent the locus of points where the second derivative of entropy with respect to energy turns positive, indicating a hypothetical onset of purely thermal instability. As expected, on the excitation energy scale, the more iso-asymmetric systems become unstable sooner such that for sufficiently large asymmetries of $I > 0.65$, or so,

the system cannot be stable even at its ground state. Note that the excitation energy scale is obtained by simply subtracting from the total energy (ordinate) value the ground-state value of line 1 for the iso-asymmetry of interest. The thermo-chemical nature of the spinodal instability at the boundary line 2, is visualized in Fig. 4 by short bars labeled \mathbf{v} . These bars indicate the directions in which the convexity of the entropy function sets in and which are identical with the directions of the eigenvectors of the Hessian of the entropy corresponding to zero eigenvalue. These bars indicate also that the instability will develop along a direction of joint changes in the iso-asymmetry I and energy and that upon “over-stepping” the boundary line 2, more iso-rich portions of the system would acquire more excitation energy at the expense of the more iso-poor portions and would expand thermally. There is no mechanism which would prevent then a portion of the iso-rich matter from reaching the line 4 of “no return” and leaving behind a more iso-symmetric metastable residue somewhere on the “attractor” line 2. This “attractor” line is a potential experimental observable, were experiments possible on bulk matter. However, a similar plot for a finite nucleus obtained with a realistic EOS could provide true guidance as to the experimental efforts to pin down the subtleties of iso-asymmetry energy in sub-normal matter densities. Note that, significantly, there is no line shown in Fig. 4 where the second derivative of entropy with respect to I would turn positive and, thus indicate the hypothetical onset of a purely chemical spinodal instability. This is so because the second derivative in question stays here negative over the whole domain where the system is stable with respect to uniform expansion and, thus, the system here never becomes unstable purely chemically.

The behavior inferred from the appearance of the lines in Fig. 4 is clearly that of classical distillation and it differs from what is commonly termed as iso-spin fractionation in that here the escaping vapors are never in equilibrium with the residue

6. Discussion

Boiling is a very common phenomenon observed in everyday life with respect to water. Quite obviously, water in an open container at room temperature is unstable such that it evaporates with time. Yet, its metastability allows one to treat it as a microcanonical object on the time scales needed to perform measurements of its temperature, specific density, specific heat, rate of evaporation, and other characteristics of interest. With increasing temperature the rate of evaporation increases and water expands in volume, but still is sufficiently metastable to allow quantitative measurements of its properties. The situation changes drastically at the boiling-point temperature, where the system begins behave chaotically, with bubbles forming throughout the volume, rapidly growing in size, making it to the surface, and then vaporizing into the open space. Importantly, the temperature here stays constant regardless of the heat supplied, until all of the water has vaporized. Also, it appears absolutely impossible to reach the critical point at 374° of temperature, 218 atm of pressure and a volume of 13.5 L/kg when heating water in an open kettle.

Similarly to the example of water, in nuclear thermodynamics, the fundamental concept of compound nucleus fully relies on the metastability and on practical microcanonical equilibrium expressed in the Boltzmann entropy. Yet, all practical implementations of the concept of a metastable compound system have failed to recognize the fact that such system must come to boiling if nothing else prevents it from being excited to the boiling-point excitation energy. This failure has its roots in the overlooking of the importance of thermal expansion in the behavior of excited nuclei. One may posit that the true thermal expansion and the resulting boiling are the most overlooked phenomena in nuclear thermodynamics. This true thermal expansion is based fully on the sound and time-proven notion of maximum Boltzmann entropy and is to be contrasted with non-thermal forms of expansion such as, e.g., the purely hypothetical isentropic expansion and purely hypothetical forms of expansion considered in some multifragmentation models under the “umbrella” of what is called the freezeout volume.

As discussed in the present study, boiling phenomenon arises directly from thermal expansion that tends to reduce the growth of temperature with increasing excitation energy per nucleon. Due to the peculiarity of van der Waals type matter, the trend of the growth of temperature with energy is not only slowed down but reversed at certain excitation energy per nucleon, corresponding to boiling. The essence of boiling is then in thermal expansion of matter to the boiling-point volume and then, when extra energy is available, expansion of portions of matter first via infinitesimally small fluctuations in excitation energy distribution (and the matching fluctuations in matter density) and then via an “entropy-driven” growth of the resulting micro-bubbles into macroscopic bubbles at the expense of the energy derived from the surrounding liquid. This driven growth lasts until the point of “no return” is reached where having accumulated enough energy these bubbles are able to expand into the open space on their own, without need for a further supply of energy. The hallmark signature of such a phenomenon is a residue at constant temperature independent of the initial energy content. This hallmark signature has been observed systematically in various experiments.[9, 10] Further, it is clear from the modeling that the escaping vapors will be at a temperature lower than that of the residue and corresponding to the point where the instability against uniform expansion sets in, another signature worth an experimental effort to confirm. Because of the latter, boiling shows a relatively low latent heat as compared with classical liquid-gas transition ending in an equilibrated two-phase system.

For finite nuclei, the presence of a diffuse surface domain has profound effects on the boiling temperature, significantly lowering that temperature with respect to that for the bulk matter. This appears quite natural as the surface domain is relatively weakly bound and is thus expected to lose stability at lower temperatures. Furthermore, the surface boiling has a phenomenology much different from that of bulk boiling and in the model calculations does not occur via formation of expanding bubbles. Rather, portions of the surface domain diffuse smoothly away as they derive the energy from neighboring sections of the surface domain. While it is not possible to tell from the thermodynamic modeling how exactly boiling happens, it appears certain that parts of the system are forced to leave the system, leaving behind a metastable residues which then undergoes statistical decay either via particle emission or Coulomb fragmentation into two or more fragments, all showing in experiments statistical source temperatures of the boiling residue, i.e., the boiling-point temperatures.

In iso-asymmetric systems, the boiling takes first a form of distillation when with increasing initial excitation energy, the system rids itself progressively of excess neutrons. The residues that are left behind have higher temperatures for lower iso-asymmetries, reflecting the dependence of the temperature of the thermo-chemical spinodal instability on the system isospin. Eventually, when the system has shed all of its excess neutrons, the iso-symmetric azeotrope boils uniformly at constant temperature. Here, it is worth extending model calculations on finite systems such that quantitative predictions can be compared to experimental observations.

In summary, boiling is a very unique and interesting phenomenon, whether in bulk or finite and whether in iso-neutral or iso-asymmetric systems. It is rich in detectable consequences, rich in theoretical implications, and constitutes potentially a fertile arena for mind-challenging speculations. It is also a phenomenon whose very presence in highly excited nuclear systems, while obvious and inevitable and with a sound confirmation in series of unique experimental observations, still awaits recognition by the nuclear science community.

Acknowledgments

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