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Thermodynamic analysis of multifragmentation phenomena

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Based on a similarity of the Van der Waals and nucleon-nucleon interaction, the known thermodynamic relations for ordinary liquids are used to analyze the possible nuclear-system phase trajectories in the proton-induced nuclear multifragmentation phenomena. A number of decay channels suggesting the most appropriate qualitative picture are proposed.

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I. INTRODUCTION

The experimental observation of multiple intermediate mass fragments (IMF) is often linked to the nuclear liquid-gas transition [1–3]. The present study, to a large extent, was inspired by the growing understanding of the reactions in which excited nuclei break up into intermediate size fragments. The interest in this subject was triggered by the importance of the multifragmentation phenomena for understanding nuclear compressibility and the equation of state of finite nuclear matter [4]. Most remarkably, these studies have much in common [5–7] with different neighboring fields of physics, such as condensed matter and statistical mechanics.

Starting from the pioneering works of Finn *et al.* [8] and up to present days, much effort has been put forward to investigate multifragmentation in nuclear collisions [9–11]. Many models have been devised to study the behavior of nuclear systems in the hadron-nuclear collisions. Some existing models are related to statistical description based on multibody phase-space calculations [12], whereas others describe the dynamic evolution of the systems via molecular dynamics [13,14]. The main concepts of the statistical approach were formulated in the 1980s [15] and in recent years the new approach is of high interest. An important theoretical effort has been made to investigate signatures of phase transitions in small systems [16,17]. Consequently, there are a number of studies that link the IMF emission process with the spinodal decomposition characteristic of the infinite ordinary systems [6,13,18,19]. At the same time attempts were made to describe multifragmentation as a sequential evaporation process [20,21]. Unfortunately, the concept of the compound nuclei cannot be applied at high excitation energies, $E^* \geqslant 3$ MeV/nucleon [11]. The existing analysis of the multifragmentation data proves that the decay mechanism through sequential evaporation from a compound nucleus is impossible [22,23]. It should be mentioned that the extended models of the compound nucleus (e.g., the harmonic-interaction Fermi gas model and the expanding-emitting source model) do not explain the results either [11,24,25]. Therefore, at high excitation energies (higher than the multifragmentation threshold $E_th \sim 2-4$ MeV/nucleon [11]) a simultaneous emission is the preferable assumption.

Though the debate related to the multifragmentation phenomenon mechanism is ongoing, the important point is that the current state of the data suggests the liquid-gas phase transition to occur in nuclear systems [18]. From this point of view it is important that the thermodynamic behavior of the nuclear matter has much in common with that of the fluids. The physical reason is the qualitative similarity of the Van der Waals and nucleon-nucleon interaction [26–28]. Despite the tremendous difference in the energy and space scales, their equations of state are very similar. This fact is illustrated in Fig. 1, where isotherms for an equation of state corresponding to nuclear forces (Skyrme effective interaction and finite temperature Hartree-Fock theory [29]) are shown, exhibiting the maximum-minimum structure typical of the Van der Waals equation of state. The apparent similarity of the properties of different media suggests the use of the classical thermodynamic approach to the nuclear systems [30].

However, at this point much uncertainty regarding the nature of the intermediate mass fragments emission remains. In particular, the underlying mechanism of the nuclear multifragmentation is not clear.

II. PHASE DIAGRAM ANALYSIS

In this paper we focus our analysis on the proton-induced nuclear multifragmentation. In subsequent parts of the paper we study different possible phase trajectories of the excited nuclear system at the Pressure-Volume (P-V) plane. We could have conducted precise calculations for the fragment spectra, time scales, etc., but our objective here is different. Our aim is to get a qualitative picture of the phenomenon considering the boundedness of the system and the existing laws which describe the behavior of the ordinary liquids in the metastable and spinodal states [31].

Let us assume our system before the collision to be in point L of the phase diagram (Fig. 2).

Its further evolution crucially depends on the excitation energy, mass number, and energy release conditions. This position suggests two different groups of phase trajectories, namely, the single-phase transition (marked with the dashed line) and two-phase transitions (marked with the solid line). It is obvious that there could be a mixture of two decay channels when different parts of the system are found in different areas of the phase diagram. Thus one may say that nuclear multifragmentation is a nonlinear phenomenon which means that the qualitative picture is different depending on the system parameters.

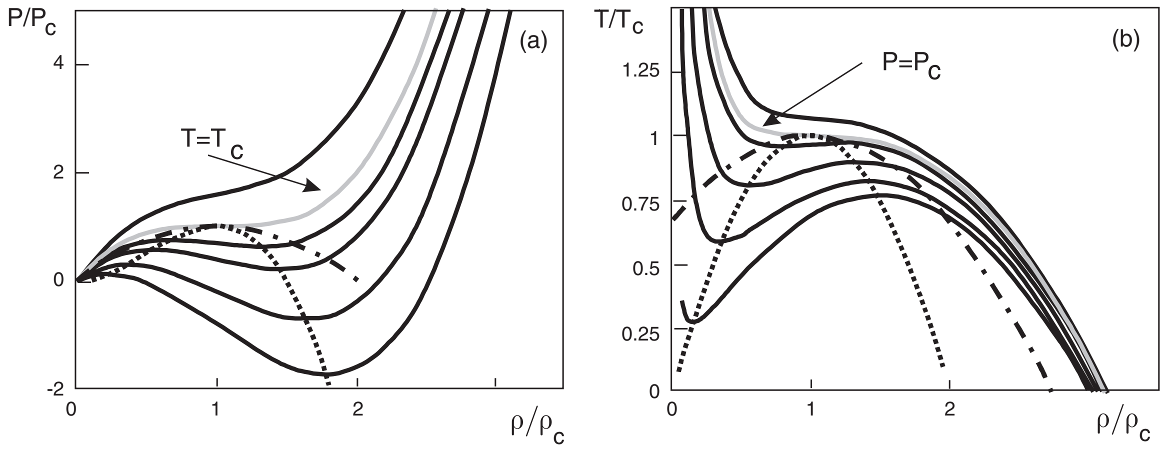

FIG. 1. Eqnarray of state relating the pressure (a) or the temperature (b) and the density (normalized to critical values) in nuclear matter. The curves correspond to isotherms (a) and isobars (b). The dash-dotted lines are the coexistence ones and the dotted lines are the spinodal ones (from Ref. [30]).

Let us take a closer look at the different multifragmentation mechanisms.

A. Single-phase transition

A particulary interesting set of phase trajectories that stands up in the study of multifragmentation phenomenon is that where the trajectories do not cross the binodal (LAC- and LGE-type curves in Fig. 2). The necessary condition for realizing the above trajectories is heating the system to supercritical temperature in order to get over the critical point and binodal line. The necessary condition for such a process in terms of entropy is the following [32]:

$$S_h = S_0 + \int_{T_0}^{T_h} C_v(V_0, T) d \ln T > S_c. \tag{1}$$

In the case of ordinary liquids quantitative estimates show that the energy introduced into the system Q should be at least twice as large as the evaporation energy $Q \geqslant 2Q_{\text{evaporation}}$. [33] The second condition to be met is the relation of the thermalization time τ_{term} with the characteristic parameters of the system

$$\tau_{\text{term}} < \tau_{\text{gd}} = \frac{r_{\text{characteristic}}}{a_0}. \tag{2}$$

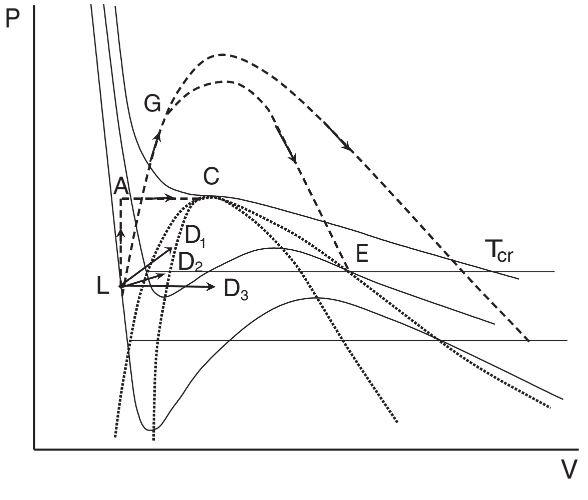

FIG. 2. Possible phase trajectories of the nuclear system in the proton-induced multifragmentation phenomenon.

Let us conduct a step-by-step analysis of the process. There could be a number of different phase trajectories responsible for the single-phase process. The first group includes isochoric heating at the start with the adiabatic expansion to follow. In order to meet such processes the fragmentation time τ_f should be much longer than the collision time:

$$\tau_{\text{col}} \ll \tau_f. \tag{3}$$

At the same time a different mechanism that does not involve either isochoric heating or adiabatic expansion may be realized. In this case the single-phase process is possible when the increase in the pressure ΔP in a period of time $\tau \leqslant \frac{r_0}{a_0}$ exceeds the equilibrium pressure in the outer medium. Therefore we have the explosion with the energy source inside the system. The condition for the characteristic times in that case is

$$\tau_{\text{col}} \sim \tau_f. \tag{4}$$

Use of the criteria (1)–(4) with regard to the experimental data leads to a preliminary decision concerning the applicability of the model.

As stated in the Introduction, we dwelt on the qualitative picture which should be the first step in choosing the most appropriate phase trajectory. What are the main features the decay channel of this kind? First of al one should bear in mind that it is the most effective one regarding the time necessary for the liquid-vapor transition [33]. The next thing of importance is unlike the other mechanisms the medium flow is realized with the distinct frontier and the intense shockwave formed in the process. In supercritical explosions the most significant part of the energy is expended on shockwave formation, while in the two-phase processes the energy is spent for the work in the sphere-expanding process against the interparticle forces. The main feature of the overcritical explosion is the very high evaporation rate $X_{\mathrm{exp}} \to 1$. In such a process the whole system is evaporated not the way it is observed in the proton induced multifragmentation experiments. There are two possibilities to obtain condensed fragments. The first one is the recondensation in the metastable region (E-type points in Fig. 2). For the ordinary liquids the maximum predicted recondensation rate is 0.4 [31,32]. As for the nuclear matter the recondensation process is rather uncertain, as we have very few particles moving far away after the expansion. The second one is the overcritical explosion of the inner part of the nuclei but not the entire system. In this case the shockwave that is formed in the system may cause the mechanical breakdown of the shell around the supercritical region. This process, which may be accompanied by the metastable boiling of the shell and recondensation of the supercritical vapor, would result in system fragmentation with a polydisperse spectrum of fragments.

We investigated several qualitative features of single-phase transitions and compared them with the data from the multifragmentation experiments. Great differences between these two pictures are apparent in the evaporation rate in the case of the overcritical explosion in the entire volume. At the same time they demonstrate similarity in the time scales. Summing up all the main qualitative features of the phenomenon, one comes to the conclusion that possible scenarios involving the single-phase transitions are as follows:

(1) The overcritical explosion of the inner part of the system followed by the mechanical breakdown of the shell.

(2) The overcritical explosion of the inner part of the system with the shell being heated by the shockwaves, causing metastable states.

We should also mention another mechanism, the one where the liquid breakdown is caused by the rarefication of the matter due to cavitation. Unfortunately, the mechanism is not suitable in our case regarding the fragments spectra.

B. Metastable boiling

The decay mechanism we discuss here could be represented by a number of different trajectories leading to the metastable region (e.g., LD_1, LD_2-type transitions at Fig. 1). It may be either overheating under the equilibrium pressure or the subcooled liquid stretching due to the gas-dynamic expansion of the high-pressure regions. When the system enters the metastable region of the phase diagram it is possible to write down the relation linking the parameters of the process [31–33]:

$$\int_0^{\tau_f} dt \int_{V_l(t)} J[t, T(\vec{r}, t)] m_v(\tau_f - t) dV = X_{\mathrm{exp}} M, \quad (5)$$

where V_l is the metastable liquid volume, $J = NB_k \exp\{-\Delta\Phi - \frac{t}{\tau_r}\}$ is the nucleation rate, and $m_v = \frac{4}{3}\pi r_v^3(t)\rho_v(t)$ is the vapor mass in the gas bubble, where the radius is defined by the problem of vapor-sphere expansion in the incompressible fluid within the droplet.

The condition for the explosive boiling of the overheated liquid is as follows:

$$\int_0^{\tau_f} dt \int_{V_l(t)} J[t, T(\vec{r}, t), P(\vec{r}, t)] dV \gg 1. \quad (6)$$

In case of the metastable boiling, the important characteristic parameter is the number of bubbles at the explosion time [31]:

$$N_{\mathrm{be}} = \int_0^{\tau_f} J(t) dt. \quad (7)$$

Relations (5)–(7) could be treated as criteria for the decision if the metastable boiling is possible in the system. They also make it possible to analyze the details of the process. Thus three different scenarios are underway.

The first one corresponds to $N_{\mathrm{be}} \sim 1$. In this case there is one or several big vapor bubbles and we obtain $X_{\mathrm{exp}} \sim 1$, which means almost full evaporation and very small fragments. The resulting fragments spectrum is monodisperse. At the same time the system is transformed into a sphere with the shell of finite size containing a vapor inside when only the inner parts of the system are in the metastable state. In certain cases the instability of the shell and its breakdown could be observed. In this case the fragments size will be proportional to the breaking perturbation amplitude and the resulting fragments spectrum will be monodisperse.

The next case is $N_{\mathrm{be}} \gg 1$. This leads to a large quantity of vapor bubbles of different size. The system fragmentation is the result of forefront instability between the growing bubbles. The number and size distribution of the fragments should be correlated with the number and size distribution of the vapor bubbles. The number of fragments and the number of bubbles are related as

$$N_f = \frac{N_{\mathrm{be}} \rho_v (1 - X_{\mathrm{exp}})}{\rho_l X_{\mathrm{exp}}}. \quad (8)$$

The resulting fragment spectrum will be polydisperse. In the case of the small inner part of the system involved in the process, we should observe the similar picture of the mechanical breakdown and monodisperse spectrum as for $N_{\mathrm{be}} \sim 1$ but with different fragment size.

Finally, there may be the case when $N_{\mathrm{be}} > 1$, the intermediate and most intriguing case. It includes the features of both asymptotical cases, and the fragment spectrum could be polydisperse. In the case of the small inner part of the system involved, such a process suggests mechanical breakdown with polydisperse spectra of fragments close to the experimental data and quite low evaporation rates X_{exp}.

As a result of the explosive boiling, the vapor-liquid mixture is produced with the pressure and temperature exceeding their equilibrium values, which leads to the gas-dynamic expansion of the mixture [34]. In this case the initial parameters (P, V, r_c) can be taken from the vapor thermodynamic analysis at the explosion moment.

We want to point out that when the system is in the metastable region there can be a different fragmentation mechanism, namely:

(1) Fragmentation caused by metastable boiling itself.

(2) Mechanical breakdown of the shell due to the metastable boiling in the inner parts of the system.

(3) Forefront instability between the growing bubbles.

Let us consider the disadvantages of such a mechanism. First of all one should keep in mind the boundedness of the system. Metastable boiling is a well-known mechanism for the infinite system, but there are still some questions in the case of small systems. The second thing to bear in mind is the time scale of the process. We are not providing precise calculations in this work, but metastable boiling is a much less effective mechanism regarding the time needed for the process compared to the single-phase transition.

C. Spinodal decomposition

The main characteristic of that decay channel is that the system enters into the absolute instability area (e.g., LD_3-type transitions at Fig. 2). This means there is mechanical instability of the system to the small fluctuations [18,31,35]. The comparison of the experimental data (time scales, qualitative picture) of the nuclear multifragmentation [7,36] with those for the spinodal decomposition [31] suggests that the mechanism is valid for the description of the phenomenon. Unfortunately there are some peculiarities that restrict the validity of the spinodal decomposition model. Let us have a look at the underlying mechanism.

The main idea of the spinodal decomposition is the system instability to the small fluctuations. It is a well-known mechanism for the binary solutions at the thermodynamic limit. In contrast to the metastable state, even very small nucleation centers should lead to the phase transition. In this case it is important to provide the analysis of the existing energy changes in the system. Cahn's theory of separation by spinodal decomposition [35] for the changes in free energy presents the following:

$$\Delta F = \int \left[\frac{1}{2} \left(\frac{\partial^2 f}{\partial c^2} (c - c_0)^2 \right) + K (\nabla c)^2 \right] dV, \tag{9}$$

where f is the free-energy density of homogeneous material of composition c, and $K(\nabla c)^2$ is the additional free-energy density if the material is in a gradient in composition. Therefore the system is unstable to the Fourier components with $\beta < \beta_c = (-\frac{\partial^2 f/\partial c^2}{2K})^{1/2}$ or sufficiently large wavelength, as it decreases the system free energy when it is in the unstable region. This result shows that for the smaller wavelengths there is no decrease in the system free energy and there is no sign change in the diffusion coefficients. In this case there is no reason for the mechanical instability and the system behaves very much like that in the metastable state. Some experiments with capillary show (from private communications with V. P. Skripov) the metastablelike behavior of the ordinary liquids in the spinodal region for the bounded systems.

As the nucleus is quite a bounded system, the question arises of whether it is big enough to have wavelengths needed for the spinodal decomposition (especially if the process takes place not in the entire system but in a small part of it). We should also notice that for the instability to develop in the system it should stay long enough in the spinodal region for the initial fluctuations to be amplified. Therefore it is not yet clear if the spinodal separation has all the necessary features to explain the experimental data. All this indicates that spinodal decomposition is not the best agent to explain nuclear multifragmentation.

III. CONCLUSIONS

We have analyzed the possible phase trajectories of the nuclear system that could have corresponded to the multifragmentation phenomenon. We figured out the most appropriate mechanisms of the multifragmentation in nuclear systems from a thermodynamic point of view. It appeared that not all of the mechanisms could be realized in nuclear systems because of their size. Some mechanisms do not meet the requirements for the time needed for the process. Summing up all pros and cons of different decay channel qualitative characteristics we suggest the following:

(1) Spinodal decomposition could not be responsible for the multifragmentation phenomena because of the system size.

(2) The most appropriate decay channel is the mechanical breakdown of the shell in the single-phase process that may be followed by metastable boiling.

(3) There also could be a mechanism based on the metastable boiling of the inner part of the system.

In order to choose the correct one from the last two, quantitative analysis is needed.

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