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STATISTICAL MULTIFRAGMENTATION MODEL WITHIN THE EXTENDED MORPHOLOGICAL THERMODYNAMICS APPROACH

On the basis of the morphological thermodynamics, we develop an exactly solvable version of the statistical multifragmentation model for the nuclear liquid-gas phase transition. It is shown that the hard-core repulsion between spherical nuclei generates only the bulk (volume), surface, and curvature parts of the free energy of the nucleus, while the Gaussian curvature one does not appear in the derivation. The phase diagram of the nuclear liquid-gas phase transition is studied for a truncated version of the developed model.

K e y w o r d s: morphological thermodynamics, induced surface and curvature tensions, equation of state, nuclear liquid-gas phase transition, statistical multifragmentation.

1. Introduction

Over the past 20 years, the morphological thermodynamics approach has been extensively developed in condensed matter physics to describe the behavior of dense 3- and 2-dimensional fluids [1]. It is based on the Hadwiger theorem [2, 3] and can be formulated as: the total free energy decrease $-\Delta\Omega$ of a convex rigid body $\boldsymbol{B}$ inserted into a fluid can be completely described by four thermodynamic characteristics such as the pressure p, the mean surface tension coefficient Σ, the mean curvature tension coefficient K, and the bending rigidity coefficient Ψ, i.e.,

$$-\Delta\Omega = pv_B + \Sigma s_B + Kc_B + \Psi x_B, \tag{1}$$

where the quantities v_B, s_B, c_B, x_B are, respectively, the volume of $\boldsymbol{B}$, its surface, the mean curvature integrated over the surface of the rigid body, and the mean Gaussian curvature also integrated over the surface of $\boldsymbol{B}$. The last two quantities are defined via the two local principal curvature radii R_{c1} and R_{c2} as $c_B = \int_{\partial\mathcal{B}} d^2r \frac{1}{2}\left[\frac{1}{R_{c1}} + \frac{1}{R_{c2}}\right]$ and $x_B = \int_{\partial\mathcal{B}} d^2r \frac{1}{R_{c1}R_{c2}}$ (the Euler characteristic). Such a treatment is usually justified, if the system exists relatively away from the critical point and, simultaneously, from wetting and drying transitions [1, 3].

Independently of the morphological thermodynamics, its analog in the grand canonical ensemble widely known as the induced surface and curvature tensions (ISCT) and the equation of state (EoS) was developed recently in [4–8]. This approach was successfully applied in Ref. [4] to model the properties of the one- and two-component mixtures of classical hard spheres and hard discs. The ISCT EoS was developed not only for classical particles, but also for quantum ones [5, 6] and for relativistic particles that experience the Lorentz contraction [8]. Very recently, the grand canonical ensemble formulation of morphological thermodynamics was also worked out for very small system volumes of about 100 fm³, which are typical of the nuclear reactions [7].

Here, we reformulate the exactly solvable version of the statistical multifragmentation model (SMM) [9, 10] using the postulates of morphological thermodynamics. This is a highly nontrivial extension of the morphological thermodynamics to the infinite number of degrees of freedom which characterize the nuclear clusters of k nucleons with $k = 1, 2, 3, \ldots$. It corresponds to an important extension of the usual morphological thermodynamics to a new field, namely, to a vicinity of the (tri)critical endpoint of nuclear matter. In addition, we will discuss a possible extension of the suggested model which will improve the description of the liquid phase of nuclear matter at high packing fractions.

The work is organized as follows. In Sect. 1, we heuristically derive the ISCT EoS for the SMM using the requirements of morphological thermodynamics. In Sect. 2, a truncated version of the ISCT EoS for the SMM is discussed, and our results are summarized in Conclusions.

2. Derivation of ISCT EoS for SMM

We start our discussion from the one-component gas of Boltzmann particles with hard-core repulsion. The potential energy $U(r)$ of the interaction of such particles depends on the distance r between their centers as

$$U(r) = \begin{cases} \infty & \text{for } r \le 2R, \\ 0 & \text{for } r > 2R. \end{cases} \tag{2}$$

Hence, the quantity R is the hard-core radius. Since the typical temperatures of the nuclear liquid-gas phase transition (PT) are below 20 MeV, i.e. much smaller than the nucleon mass, one can safely use the non-relativistic treatment [9, 10].

In the grand canonical ensemble with the pressure p, the van der Waals (VdW) EoS of particles with hard-core repulsion can be written as

$$p = T\phi(T) \exp\left(\frac{\mu - a\,p}{T}\right), \tag{3}$$

$$\phi(T) = g \int \frac{d^3k}{(2\pi\hbar)^3} \exp\left(-\frac{\mathbf{k}^2}{2mT}\right), \tag{4}$$

where T is the system temperature, and μ is the chemical potential. In Eq. (3), the parameter

$$a = \int d^3r \left[1 - \exp\left(-U(r)/T\right)\right] \equiv 4v \equiv$$
$$\equiv v + sR \equiv v + \frac{1}{2}sR + \frac{1}{2}cR^2 \tag{5}$$

denotes the second virial coefficient, where $v = \frac{4}{3}\pi R^3$ denotes the eigenvolume of particles, while $s = 4\pi R^2$ and $c = 4\pi R$ denote, respectively, their eigensurface and double eigenperimeter. In Eq. (4), $\phi(T)$ is the thermal density of particles with mass m and the degeneracy factor g.

According to morphological thermodynamics [1–3], the free energy of a rigid particle should be written as $f = vp + s\Sigma + cK$, and, hence, the grand canonical pressure (3) should be generalized as

$$p = T\phi(T) \exp\left(\frac{\mu - (vp + s\Sigma + cK)}{T}\right). \tag{6}$$

But this equation should be supplemented by the equations for the surface tension coefficient Σ and the curvature tension one K induced by hard-core repulsion. In Ref. [4], one can find how the equations for Σ and K can be derived rigorously. Here, we generalize the heuristic derivation of Ref. [10] by including, into our treatment, the curvature tension coefficient K.

Since, in the SMM, the nuclear clusters can consist of any positive number of nucleons, we consider the system of particles of N sorts with the hard-core radii R_k, $k = 1, 2, ..., N \to \infty$. The virial expansion of the gas pressure up to the second order can be cast as [10]

$$p = T\sum_{k=1}^{N} \phi_k e^{\frac{\mu_k}{T}} \left[1 - \sum_{n=1}^{N} a_{kn}\phi_n e^{\frac{\mu_n}{T}}\right], \tag{7}$$

$$\phi_n(T) = g_n \int \frac{d^3k}{(2\pi\hbar)^3} \exp\left[-\frac{\mathbf{k}^2}{2m_nT}\right] =$$
$$= g_n \left[\frac{m_nT}{2\pi\hbar^2}\right]^{\frac{3}{2}}, \tag{8}$$

where $\phi_n(T)$ is the thermal density of particles of the degeneracy g_n, mass $m_n = nm_1$ with $m_1 = 940$ MeV. The second virial coefficient a_{kn} which has the meaning of excluded volume per particle is given by

$$a_{kn} = \frac{2}{3}\pi(R_k + R_n)^3 =$$
$$= \frac{2}{3}\pi(R_k^3 + 3R_k^2 R_n + 3R_k R_n^2 + R_n^3). \tag{9}$$

First, consider the low densities at which expansion (7) is valid. Substituting the second virial coefficients (9) into Eq. (7) and regrouping the terms with the

same powers of R_k of a k-nucleon fragment, we can write

$$p = T \sum_{k=1}^{N} \phi_k e^{\frac{\mu_k}{T}} \left[1 - \frac{4}{3} \pi R_k^3 \sum_{n=1}^{N} \phi_n e^{\frac{\mu_n}{T}} - \right.$$
$$\left. - \frac{4\pi R_k^2}{2} \sum_{n=1}^{N} R_n \phi_n e^{\frac{\mu_n}{T}} - \frac{4\pi R_k}{2} \sum_{n=1}^{N} R_n^2 \phi_n e^{\frac{\mu_n}{T}} \right]. \quad (10)$$

Apparently, for low densities, each sum in the square brackets of Eq. (10) can be identified as

$$\sum_{n=1}^{N} \phi_n e^{\frac{\mu_n}{T}} = \frac{p}{T}, \quad (11)$$

$$\frac{1}{2} \sum_{n=1}^{N} R_n \phi_n e^{\frac{\mu_n}{T}} = \frac{\Sigma}{T}, \quad (12)$$

$$\frac{1}{2} \sum_{n=1}^{N} R_n^2 \phi_n e^{\frac{\mu_n}{T}} = \frac{K}{T}. \quad (13)$$

Approximation (11) is valid for the low densities, since defining the second virial coefficient with the help of Eqs. (11) and (13), one modifies the third and higher virial coefficients which, at these densities, can be neglected. Therefore, substituting Eqs. (11) and (13) into the right-hand side of Eq. (10), we obtain

$$p = T \sum_{k=1}^{N} \phi_k e^{\frac{\mu_k}{T}} \times$$
$$\times \left[1 - \frac{4}{3} \pi R_k^3 \frac{p}{T} - 4\pi R_k^2 \frac{\Sigma}{T} - 4\pi R_k \frac{K}{T} \right] \simeq$$
$$\simeq T \sum_{k=1}^{N} \phi_k \exp\left[\frac{\mu_k - v_k p - s_k \Sigma - c_k K}{T} \right], \quad (14)$$

where, in the last step of derivation, we used the approximation $1 - x \simeq \exp(-x)$ which is valid for low densities. In Eq. (14), we introduced the eigenvolume $v_k = \frac{4}{3}\pi R_k^3$, the eigensurface $s_k = 4\pi R_k^2$, and double eigenperimeter $c_k = 4\pi R_k$ of the k-nucleon cluster with the hard-core radius $R_k = R_1 k^{\frac{1}{3}}$ and $R_1 \simeq 0.72$ fm.

Comparing Eqs. (6) and (14), we conclude that the latter is a multicomponent version of the grand canonical pressure obtained in accord with the morphological thermodynamics. Using the same logic, we can generalize expressions (13) for the coefficients of

induced surface tension Σ and induced curvature tension K in accordance with the morphological thermodynamics and write

$$\Sigma = TA \sum_{k=1}^{N} R_k \phi_k \exp\left[\frac{\mu_k - v_k p - s_k \Sigma - c_k K}{T} \right], \quad (15)$$

$$K = TB \sum_{k=1}^{N} R_k^2 \phi_k \exp\left[\frac{\mu_k - v_k p - s_k \Sigma - c_k K}{T} \right], \quad (16)$$

where the coefficients $A = 1/2$ and $B = 1/2$ will be considered as the adjustable parameters. In Refs. [4–6] it is shown that system (14)–(16) is the VdW EoS which can be improved at high densities by inserting the set of parameters $\{\alpha_k > 1\}$ and $\{\beta_k > 1\}$ for each sort of particles into the equations for Σ and K as

$$\frac{\Sigma}{T} = A \sum_{k=1}^{N} R_k \phi_k \times$$
$$\times \exp\left[\frac{\mu_k - v_k p - \alpha_k s_k \Sigma - c_k K}{T} \right], \quad (17)$$

$$\frac{K}{T} = B \sum_{k=1}^{N} R_k^2 \phi_k \times$$
$$\times \exp\left[\frac{\mu_k - v_k p - \alpha_k s_k \Sigma - \beta_k c_k K}{T} \right], \quad (18)$$

which can be now extrapolated to any particle number densities. The principal difference of the obtained system (14)-(16) from the ones analyzed previously in Refs. [4–6] is that the degeneracy factor g_k of the k-nucleon cluster is a statistical partition of the ensemble of clusters with the same mean volume v_k, but different shapes [10–12]. For $k \gg 1$, this internal partition of large clusters of k-nucleons can be expressed in terms of the mean surface s_k [10–12]. Here, we generalize the internal partition g_k to high pressures by including the mean double perimeter c_k in accord with the morphological thermodynamics and write it as

$$g_{k \gg 1} = \frac{1}{k^{\tau + \frac{3}{2}}} \exp\left[\frac{v_k p_L - s_k \sigma_0(T) - c_k K_0(T)}{T} \right], \quad (19)$$

where p_L is the pressure of nuclear liquid (internal pressure of large custers), $\sigma_0(T)$ is the T dependent proper surface tension coefficient, and $K_0(T)$ is proper curvature tension coefficient, while τ is the Fisher exponent [11]. Eq. (19) is a generalization of the expression found in Ref. [10] which allows us to

account for the effects of proper curvature tension. It is necessary to stress that, without inclusion of the proper curvature tension coefficient in Eq. (19), it is impossible to compensate the induced surface tension one and, hence, in such a model, the (tri)critical point does not exist.

Existence of the curvature and even of the Gaussian curvature terms in the Bethe–Weizsäcker formula for the binding energy of large nuclei at zero temperature has been discussed for about fifty years [13–15], but, no definite conclusion about its existence was reached so far. In this work, the induced surface tension coefficient is derived similarly to the surface one, but our analysis shows no room to introduce the Gaussian curvature terms. Therefore, we do not consider it for the SMM.

The T-dependence of both the proper and the induced surface tension coefficients of nuclear clusters have been discussed for a few decades [9, 12, 14–16]. Although there exist several parametrizations for $\sigma_0(T)$ and for $\sigma_{\rm tot} = \sigma_0(T) + \Sigma$, we use the linear T-dependence of $\sigma_0(T)$, since it is obtained within an exactly solvable model for the surface partition [12]. In addition, a thorough analysis of experimental data performed in Ref. [16] shows that there is a wide range of temperatures at which the total surface tension coefficient $\sigma_{\rm tot}$ is a linear function of T. Below, it is shown that even a truncated version of the ISCT EoS provides the existence of two regions of such T-dependence of $\sigma_{\rm tot}$.

3. Truncated ISCT EoS for Low Densities

System (14)–(16) can be used to describe the properties of nuclear matter at high particle number densities and to clarify a principal question what is the value of total surface tension coefficient $\sigma_{\rm tot}$ at supercritical temperatures. In the famous Fisher droplet model [11] and in the solvable version of SMM [9], it is assumed that $\sigma_{\rm tot} = 0$ for $T \geq T_{\rm cep}$, while, in the SMM with compressible nuclear liquid, it is argued that $\sigma_{\rm tot} < 0$ for $T > T_{\rm cep}$, while $\sigma_{\rm tot} = 0$ at $T = T_{\rm cep}$. Apparently, this question can be answered only experimentally. On the other hand, it is expected that the critical point of nuclear matter is located at particle number densities about $\rho_c \simeq \frac{\rho_0}{3}$ [10, 14], where $\rho_0 \simeq 0.16$ fm^{-3} is the normal nuclear density. Therefore, to study the properties of nuclear matter in the vicinity of the critical endpoint, it is sufficient to work out a simpler EoS.

Using the fact that, at low particle number densities, the contributions of surface and curvature tensions to the second virial coefficient are the same, i.e., $\sum_{k=1}^{N} \phi_k e^{\frac{\mu_k}{T}} R_k^2 \Sigma = \sum_{k=1}^{N} \phi_k e^{\frac{\mu_k}{T}} R_k K$, one can account for the curvature tension effects by doubling down the contribution of Σ on the right-hand side of Eqs. (14) and (17) [10]. Hence, instead of the system (14)–(16), one can write

$$p = T \sum_{k=1}^{N} \phi_k \exp\left[\frac{\mu_k - v_k p - s_k \Sigma}{T}\right], \tag{20}$$

$$\Sigma = T \sum_{k=1}^{N} R_k \phi_k \exp\left[\frac{\mu_k - v_k p - \alpha s_k \Sigma}{T}\right], \tag{21}$$

where the parameter $\alpha = 1.5$ is chosen according to Ref. [10]. To parametrize the degeneracy of large clusters according to Eq. (19) and to account for the fact that nucleons do not have binding energy, we assume the thermal density of k-nucleon fragments to be

$$\phi_1 = z_1 \left[\frac{mT}{2\pi\hbar^2}\right]^{\frac{3}{2}} \exp\left[-\frac{\sigma_0(T)}{T}\right], \tag{22}$$

$$\phi_{k\geq 2} = \frac{1}{k^\tau} \left[\frac{mT}{2\pi\hbar^2}\right]^{\frac{3}{2}} \exp\left[\frac{(k p_L V_1 - \mu_k) - \sigma_0(T) k^{\frac{2}{3}}}{T}\right], \tag{23}$$

where the chemical potential of a k-nucleon fragment is $\mu_k = k\mu$ [9, 10], and value of $\tau = 1.9$ provides the existence of the 1-st order PT for $T < T_{\rm cep}$ [10]. In Eq. (22), $z_1 = 4$ is the degeneracy factor of nucleons. For $k \geq 1$, one finds $v_k = V_1 k = \frac{4}{3}\pi R_k^3$ with $V_1 = \frac{1}{\rho_0}$. For $\phi_{k\geq 2}$ the binding energy is included into the pressure of the liquid phase:

$$p_L = \frac{\mu + W_{\rm F}(T) + W_0 + a_\nu \left[\mu + W_0\right]^\nu}{V_1}, \tag{24}$$

with $\nu = 2, 3, 4$. The binding energy per nucleon is given by $W(T) = W_0 + W_{\rm F}(T)$, where $W_0 = 16$ MeV is the bulk binding energy per nucleon at $T = 0$, and $W_{\rm F}(T) = \frac{T^2}{\varepsilon_0}$ (with $\varepsilon_0 = 16$ MeV) accounts for the Fermi motion of nucleons inside a nucleus at $T > 0$ [9]. The constant a_ν in Eq. (24) should be found by requiring that, at $T = 0$ and normal nuclear density, $\rho_L = \frac{\partial p_L}{\partial \mu} = \rho_0$, the liquid pressure is zero [10]. For definiteness, in this work, we assume that
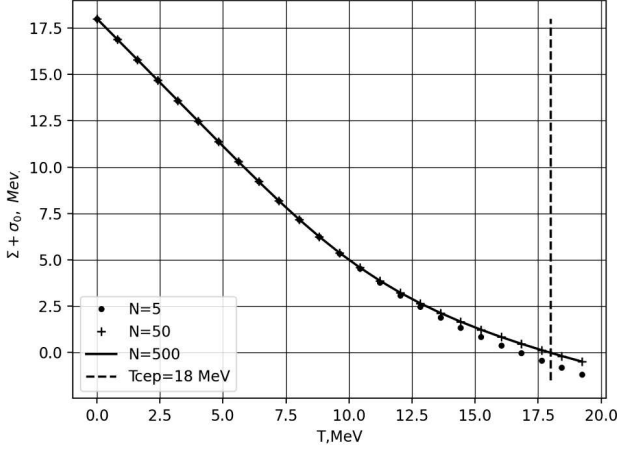
Fig. 1. Total surface tension coefficient $\sigma_{\text{tot}}(\mu_c(T), T)$ at the PT curve $\mu_c(T)$ as a function of T is shown for several sizes of the largest nucleus N. A vertical line defines the critical temperature $T_{\text{cep}} = 18$ MeV

$a_2 = 1.261 \times 10^{-2}$ MeV^{-1} and $\nu = 2$. Hence, the system (20)–(21) becomes

$$\frac{p}{T} = \left[\frac{mT}{2\pi\hbar^2}\right]^{\frac{3}{2}} \sum_{k=1}^{N} \frac{b_k}{k^\tau} \exp\left[\frac{(p_L - p)V_1 k - (\Sigma + \sigma_0)k^{\frac{2}{3}}}{T}\right],$$
(25)

$$\frac{\Sigma}{3V_1 T} = \left[\frac{mT}{2\pi\hbar^2}\right]^{\frac{3}{2}} \sum_{k=1}^{N} \frac{b_k}{k^{\tau - \frac{1}{3}}} \times$$

$$\times \exp\left[\frac{(p_L - p)V_1 k - (\alpha\Sigma + \sigma_0)k^{\frac{2}{3}}}{T}\right],$$
(26)

where the degeneracies b_k are defined as $b_1 = 4\exp\left[\frac{-W(T)}{T}\right]$ and $b_{k>1} = 1$. The proper surface tension coefficient $\sigma_0 = \sigma_{01} - \sigma_{02}\frac{T}{T_{\text{cep}}}$ is chosen according to the exact result found for the surface partition [12], where $\sigma_{01} = 18$ MeV, $\sigma_{02} = 24.76$ MeV, $T_{\text{cep}} = 18$ MeV.

Assuming, in Eqs. (25, 26), that $p = p_L$, one obtains the equations for the PT curve:

$$\frac{p_L}{T} = \left[\frac{mT}{2\pi\hbar^2}\right]^{\frac{3}{2}} \sum_{k=1}^{N} \frac{b_k}{k^\tau} \exp\left[-\frac{(\Sigma + \sigma_0)}{T}k^{\frac{2}{3}}\right],$$
(27)

$$\frac{\Sigma}{3V_1 T} = \left[\frac{mT}{2\pi\hbar^2}\right]^{\frac{3}{2}} \sum_{k=1}^{N} \frac{b_k}{k^{\tau - \frac{1}{3}}} \exp\left[-\frac{(\alpha\Sigma + \sigma_0)}{T}k^{\frac{2}{3}}\right].$$
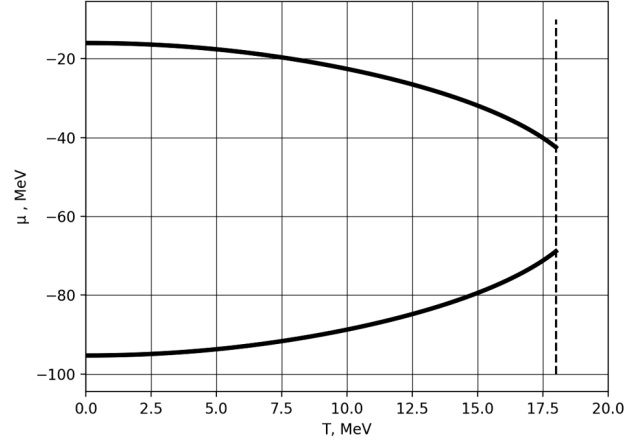(28)

Fig. 2. The PT curves $\mu_c(T)$ in the plane of temperature T and nuclear chemical potential μ

Solving Eq. (28) for Σ and substituting it into Eq. (27), one gets the pressure $p_c = p_L$ at the PT curve. Then Eq. (24) for μ with $\nu = 2$ can be written as

$$\mu^2 + \mu(1 + 2a_2 W_0) + W(T) + a_2 W_0^2 - p_c V_1 = 0. \quad (29)$$

Solving Eq. (29) for μ, one obtains $\mu_c(T)$ at the PT curve. The T-dependence of the total surface tension coefficient $\sigma_{\text{tot}}(\mu_c(T), T) = \Sigma + \sigma_0$ is shown in Fig. 1, while the function $\mu_c(T)$ is presented in Fig. 2. From Fig. 1, one can see that $\sigma_{\text{tot}}(\mu, T)$ vanishes at $T = T_{\text{cep}} = 18$ MeV even for a small size of the largest nucleus $N = 50$, although the true PT exists only for $N \to \infty$.

In Fig. 2, the solutions $\mu_c(T)$ of Eq. (29) are shown. It is evident that the upper curve describes the gas-liquid PT curve for nuclear matter, while the lower one corresponds to the unphysical solution.

4. Conclusions

In this work, we develop an exactly solvable version of the statistical mutifragmentation model for nuclear liquid-gas PT using the requirements of morphological thermodynamics. By evaluating the second virial coefficients, we explicitly demonstrate that the hard-core repulsion between spherical nuclei generates only the bulk (volume), surface, and curvature parts of the free energy of nuclei and does not produce the Gaussian curvature one. For a truncated version of the developed ISCT EoS, we studied the $T - \mu$ phase diagram of nuclear liquid-gas PT for several sizes of the largest nucleus.

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МОДЕЛЬ СТАТИСТИЧНОЇ МУЛЬТИФРАГМЕНТАЦІЇ В РАМКАХ РОЗШИРЕНОГО ПІДХОДУ МОРФОЛОГІЧНОЇ ТЕРМОДИНАМІКИ

На основі морфологічної термодинаміки розвинуто точно розв'язувану модель статистичної мультифрагментації для ядерного фазового переходу типу рідина–пара. Продемонстровано, що відштовхування типу твердого кора між сферичними ядрами генерує лише об'ємну, поверхневу та пов'язану із кривиною частини вільної енергії ядра, тоді як частина гауссової кривини у виведенні не фігурує. Фазову діаграму ядерного фазового переходу типу рідина–пара досліджено в рамках спрощеної версії розробленої моделі.

К л ю ч о в і с л о в а: морфологічна термодинаміка, індукований поверхневий натяг та натяг кривини, рівняння стану, ядерний фазовий перехід типу рідина–пара, статистична мультифрагментація.