Finite size and Coulomb corrections: from nuclei to nuclear liquid vapor phase diagram

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The difficulties encountered with Coulomb and finite size effects in generalizing to the infinite system phase diagram are considered. Methods to correct for the Coulomb effect exactly and to eliminate finite size effects are described.

I. INTRODUCTION

Since the birth of the liquid drop model, which occurred more than 60 years ago, nuclei have been recognized as charged drops of a van der Waals like fluid. Soon after, the concept of cold uncharged, symmetric nuclear matter was introduced.

The experimental characterization of cold nuclear matter began by setting the surface, symmetry, and Coulomb terms of the liquid drop expression to zero and retaining just the volume term. This, together with the independent measurement of nuclear radii (already inferable from the surface and Coulomb coefficients), defined the fundamental properties of cold symmetric nuclear matter, namely its binding energy and density at saturation.

Thus, the Coulomb interaction was reasonably eliminated from the picture in order to dispose of a troublesome inessential divergence which, however remains all pervasive in the experimental realm.

The experimental extension to higher temperatures was hampered both by the lack of a suitable container which became necessary to accommodate the vapor phase (shown to exist by the soon discovered neutron and proton evaporation) and by the not obviously generalizable finite size effects.

In this paper we consider the problem of obtaining the infinite symmetric uncharged nuclear matter phase diagram from a thermal nuclear reaction. In the first part we shall consider the Coulomb interaction which, because of its long range makes the definition of phases problematic. We shall show how to obviate this problem. In the second part we shall deal with the finite size problem by means of a new method, the complement method, which shall permit a straightforward extrapolation to the infinite system.

II. THE COULOMB INTERACTION

In this part we consider the problem of the Coulomb interaction and the problem of the container, and we present a simple and natural solution to both.

Some attempts have been made to characterize the phase transition both theoretically and experimentally. Many of the theoretical approaches are based upon numerical simulations of finite lattice systems. From these studies negative heat capacities have been claimed to be the signal of the phase transitions in nuclei and other finite systems [1]. We found it enlightening to use a simple extension of thermodynamics which incorporates the finiteness of the nuclear system through the surface and other liquid drop terms [2].

We showed that a negative heat capacity can be trivially expected in terms of the change of the droplet (nuclear) binding energy with the decreasing size of the evaporating droplet (nucleus). This can be seen most clearly in the case of an isobaric transition:

$$dp = \left.\frac{\partial p}{\partial A}\right|_T dA + \left.\frac{\partial p}{\partial T}\right|_A dT = 0. \qquad (1)$$

Since from the Clapeyron equation

$$\left.\frac{\partial p}{\partial A}\right|_T \approx -\frac{p}{T}\left.\frac{\partial \Delta H_m}{\partial A}\right|_T \qquad (2)$$

and

$$\left.\frac{\partial p}{\partial T}\right|_A \approx p\frac{\Delta H_m}{T^2} \qquad (3)$$

we can obtain the desired result for the isobaric dependence of the transition temperature on the size of the system

$$\left.\frac{\partial T}{\partial A}\right|_p = T\left.\frac{\partial \log \Delta H_m}{\partial A}\right|_T. \qquad (4)$$

In all the above, A is the size of the system, ΔH_m is the molar enthalpy of vaporization ($\Delta H_m \approx \partial B(A)/\partial A + T$), and $B(A)$ is the binding energy of a cluster of size A.

In a droplet of a van der Waals liquid, ΔH_m increases with increasing size A and saturates for infinite systems. Thus Eq. (4) says that, at fixed pressure p the coexistence temperature decreases as the droplet (nucleus) evaporates, leading to a downward slope of the caloric curve and to a negative heat capacity.

However, in nuclei surface effects are not the only ones to be considered [2]. Coulomb effects become progressively more important with increasing A, Z and eventually, at about $A=30$ they reverse the surface trend prevailing at small A values. The inescapable conclusion is that, within the scope of the Clapeyron equation, no negative heat capacities should be observed for $A > 30$, in contradiction with claims to the contrary [1].

This result is predicated upon the dominant emission of monomers (neutrons, protons), *i.e.* very small clusters. Should the nucleus statistically break up into large fragments, the upper limit for the observation of negative heat capacities would be correspondingly displaced upwards. However, not only are large fragments rare at low temperature, but they never dominate at any temperature below the critical temperature. In Ising models, finite or not, the average cluster size in the "gas phase" hardly rises above unity. The same holds true for experiments [3, 4]. It seems impossible to escape these thermodynamic conclusions.

Alternatively, the position of the maximum can be displaced towards larger masses by inhibiting the effect of Coulomb by means of a box. If we take a spherical box of size nR where R is the radius of a nucleus, we can immediately write down the liquid drop formula for a nucleus confined in it

$$B = a_v A + a_s A^{2/3} + a_c Z^2 \left(\frac{1}{A^{1/3}} - \frac{1}{nA^{1/3}} \right) \quad (5)$$

where a_v, a_s and a_c are the volume, surface and Coulomb coefficients respectively. Letting $\mu = \partial B/\partial A \approx \partial \Delta H_m/\partial A$, $Z = KA$ and solving for $\partial \mu/\partial A = \partial^2 B/\partial A^2 = 0$, we find that the largest mass number for which negative heat capacities can occur is

$$\hat{A} = \frac{1}{5} \frac{a_s}{a_c} \frac{1}{K^2} \frac{n}{n-1}. \quad (6)$$

Without a box, $n \to \infty$ and $\hat{A} \approx 29$. If the maximum is desired at $\hat{A} = 208$ then $n \approx 1.16$. This means that the system must be confined in a box barely larger than itself. This extreme requirement shows that in order to obtain the desired result one must literally kill the Coulomb effect. A justification is needed for such an extreme assumption.

However, one may question whether the role of the Coulomb interaction is merely that of decreasing the binding energy. The long range nature of this force may compel us to analyze its role in more detail in first order phase transitions. As will be shown below, the problems are serious and threaten our ability to define a true first order phase transition with any generality in the presence of such a force.

Let us introduce the Coulomb interaction in the problem of a drop and its vapor.

The Coulomb interaction can be split into three parts: 1) the drop self energy; 2) the drop-vapor interaction energy; and 3) the vapor self energy.

The drop self energy, for a finite bound or metastable drop, is easily calculated and does not constitute a problem.

For the drop-vapor interaction, we consider a probe cluster which we can carry from the interior of the drop to infinity. The potential energy experienced in the process depends upon the particle's charge/mass and is shown schematically for charged particles in Fig. 1.

If the particle has zero charge, a step is observed at the droplet radius equal to the particle binding energy. For charges greater than zero, a maximum B_c is observed at the approximate distance of the two droplets in contact. From there the potential decreases according to the Coulomb law, settling at infinity to a value equal to the binding energy of the particle, Q.

In this case, where we assume that any particle of any size is bound ($Q < 0$) and we forget about problem 3), there is no difficulty in defining a gas phase in equilibrium with the droplet at infinity constituted by particles of all sizes whose abundance is controlled by the respective binding energies. The intervening Coulomb barrier B_c does not alter the equilibrium, although it may slow its achievement. In this case the vapor is mostly monomers. The coexistence pressure described by the Clapeyron equation ($dp/dT = \Delta H_m/T\Delta V_m$). The molar enthalpy ΔH_m accounts for both surface and Coulomb terms thus providing an adequate description of the liquid vapor coexistence. (ΔV_m is the molar volume).

Let us now consider the case in which the probe particle becomes unbound to the droplet above some Z value, due to the Coulomb interaction. The situation becomes as depicted in the right panel of Fig. 1. In this case the droplet is metastable and the ground state of the system consists of two or more pieces of the original drop at infinity. This is true already at $T = 0$. Thus it is not possible to speak properly of this drop in statistical equilibrium with its vapor, since the drop itself is metastable. For a gold nucleus, the ground state is at least as complicated as three fragments of approximately size 60 nucleons at infinity. This "true" ground state is hundreds of MeV below the mass of the gold nucleus. In any statistical calculation, at any reasonable temperature, one can expect a liquid-like phase consisting of a configuration similar to the true ground state in equilibrium with some vapor. A metastable gold-like drop is an immensely improbable configuration because of the great energy chasm mentioned above. The probability of such a configuration is $P = \exp(-\Delta E/T)$ where ΔE is the energy difference between the metastable state and the ground state. Estimating $\Delta E \approx 135$ MeV and a temperature of 4 MeV we obtain $P \approx e^{-34}$ or approximately 2×10^{-15}.

One might argue that our point is made from energetic rather than free energy considerations. Let us consider, then, the transition from a condensed phase (liquid-like) to a dilute phase (vapor-like). For an infinitesimal isothermal transfer, the variation of the free energy must be zero

$$\Delta F = \Delta E - T\Delta S = 0. \quad (7)$$

As we go from liquid to vapor, $\Delta E > 0$ for a typical fluid, but this energy increase is compensated by an equivalent increase in entropy, due to the increase in molar volume.

However, if ΔE is negative, due to the Coulomb

effect, we need a <u>decrease</u> in entropy which is hardly compatible with expansion.

The conclusion is that a statistical equilibrium first order phase coexistence and phase transition is not definable for any droplet that has unbound channels.

This Coulomb effect seems truly devastating since it does not allow one to define nuclear phase transitions much above $A \approx 30$.

However there may be a solution to this difficulty. If we consider the emission of particles with a sizable charge, we notice that a large Coulomb barrier B_c is present. For $T << B_c$ these channels may be considered effectively closed. Consequently the unbound channels may not play a role on a suitably short time scale. Then a phase transition may still be definable in an approximate way. But, of course, we reach again the previous conclusion that for $A > 30$ heat capacities must be <u>positive</u> and therefore claims of <u>negative</u> heat capacities for large nuclei [1, 5] find here a most serious objection.

Let us consider now part 3) of the Coulomb energy, namely the vapor self energy. As we said above, it diverges for an infinite amount of vapor. For a dilute vapor, we could consider a small portion such that the intrinsic self energy/nucleon is much less than the temperature T. Alternatively, we could consider a finite box containing a finite system. Unfortunately, at any other distance smaller than infinity the result depends annoyingly on the size (and shape!) of the container and on whether the drop is confined or not in a specified location of the container – a rather inelegant and non-general situation leading to confusing questions about true equilibrium. In any case, it is clear that the Coulomb term makes the definition of phase coexistence and phase transition intractable and ill-posed.

A solution to these difficulties can be arrived at by asking a slightly different question: is there a way to obtain experimentally the signal and characterization of the phase diagram (transition) of a nucleus as if the Coulomb interaction were not there?

Any attempt to define and characterize both phases in the presence of the Coulomb interaction depends on the shape and size of a confining volume applied from without. But nature actually provides this "confining volume" for us. Any particle trying to leave the nucleus is "boxed in" by a barrier (B_s) which depends on the particle under consideration and on the residual nucleus (or the "complement"). The top of this potential barrier is close in shape to the potential of two objects, particle and complement, in near contact. The tops of these barriers are actually conditional saddle points [6], conditional in the sense that the mass asymmetry is considered frozen.

According to standard transition state theory in any thermal decay all these saddles are in statistical equilibrium with the droplet. The decay rates give direct information on their population which is controlled by a Boltzmann factor $\exp(-B_s/T)$. In particular
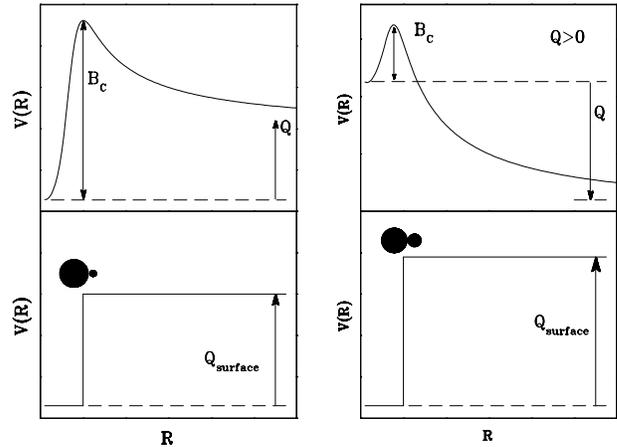

FIG. 1: A schematic representation of the Coulomb correction when the emitted fragment is bound (left panels) and unbound (right panels). In both cases one can remove the Coulomb energy of the saddle configuration and calculate the Q value using surface energies only (bottom panels). The resulting hypothetical gas will be composed of fragments that are bound to the droplet ($Q_{\text{surface}} < 0$) for all fragment partitions.

for large enough B_s the observed experimental abundances are directly related to first chance emission and thus to the transition state rate.

Now B_s is composed by $B_s = E^s_{\text{surface}} - E^{gs}_{\text{surface}} + E^s_{\text{Coul}} - E^{gs}_{\text{Coul}}$, where E^s_{surface} and E^{gs}_{surface} are the surface energies of the saddle and ground state respectively, and E^s_{Coul} and E^{gs}_{Coul} are the Coulomb energies for the same two configurations. Since the Coulomb energies can be easily estimated assuming a two touching spheres configuration for the saddle and one sphere configuration for the droplet (see Fig. 1), we can correct the rates by dividing away the Boltzmann factor containing the Coulomb terms and be left with only the rates/abundances pertaining to the decay of an uncharged drop, for which all channels are bound by the extra surface energy $Q_{\text{surface}} = E^s_{\text{surface}} - E^{gs}_{\text{surface}}$. These rates are now independent of distance and are proportional to the effective partial concentration of the hypothetical gas in equilibrium.

We speak of a virtual gas phase because it is not and it need not be present. This picture of a free evaporation of a droplet in vacuum neatly bypasses also the need for a <u>physical</u> presence of the vapor. The resulting situation is that described by the Fisher droplet model [7] for the composition of a saturated vapor in equilibrium with a liquid droplet. The Fisher droplet model can be directly co-opted to describe the (first chance) fragment abundances of a nuclear physics experiment after correction for Coulomb effects. From it, it is easy to obtain the coexistence diagram for any nuclear system deprived of the Coulomb interaction [3, 4]. This is in the same spirit as in nuclear matter calculations in which neutrons and protons are consid-

ered as distinct particles, but without any Coulomb interaction.

III. THE COMPLEMENT

We present here a general method to deal with finite size effects in phase transitions. In the case of liquid-vapor phase coexistence, a dilute nearly ideal vapor phase is in equilibrium with a dense liquid-like phase. The vapor pressure of a drop can be calculated by correcting the molar vaporization enthalpy to include the surface energy of the drop. We introduce here the concept of the complement (the residual drop which remains after a cluster has been emitted) in order to further quantify finite size effects and to generalize the theory to better describe the cluster yields from extremely small systems. The complement approach consists of evaluating the change in free energy occuring when a particle or cluster is moved from one (finite) phase to another. In the case of a liquid drop in equilibrium with its vapor, this is done by extracting a vapor particle of any given size from the drop and evaluating the energy and entropy changes associated with <u>both</u> the vapor particle <u>and</u> the residual liquid drop (complement).

This detailed accounting can be easily generalized to incorporate other energy terms common in the nuclear case, such as symmetry energy, Coulomb energy and even angular momentum [8, 9]. In order to demonstrate the power of this method, we apply it to the Ising model, where a great deal of work exists on the subject of finiteness, in particular on the dependence of critical quantities on the lattice size [10, 11].

We concentrate first on the canonical lattice gas representation of the Ising model: a fixed number of up spins represents the occupied lattice sites (i.e., matter in the form of monomers, dimers, large drops etc.), while down spins are empty space. In a finite lattice, we can fix the mean density in such a way that, below the coexistence temperature T_0, there is a large cluster or drop of a certain size in equilibrium with its vapor, constituted mostly by monomers. At the lowest temperatures the up spins congregate into one liquid drop in a vacuum. At higher temperatures the vacuum is filled with a vapor made from clusters of up spins. Clusters were identified via the Coniglio-Klein algorithm [12] to insure that they were physical (i.e., cluster distributions return Ising critical exponents and not percolation exponents). The largest cluster was identified as the liquid drop. Care was taken to eschew from the vapor the many monomers forming the surface thickness of the macroscopic drop.

The equilibrium of a small droplet with its coexisting vapor is relevant to the nuclear case. If we keep the drop size fixed, at any given temperature T sufficiently below the critical temperature T_c, the equilibrium of the system is not changed by extending the lattice to infinity, or by reducing it down to a size sufficient to
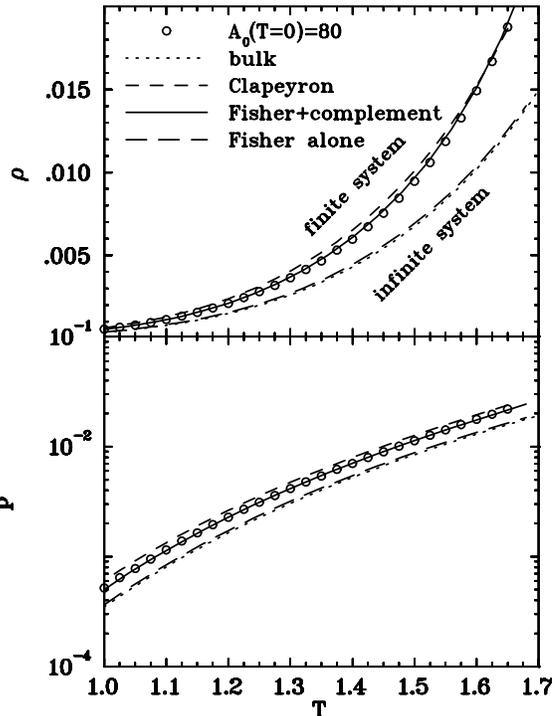

FIG. 2: The density ρ and pressure p of a vapor in coexistence with a finite drop A_0 whose size at $T = 0$ is 80 (symbols) from 2d Ising calculations with a lattice length $L = 40$ at fixed magnetization. The dotted lines are the analytic bulk solutions known for the Ising model in two dimensions [13]. The solid line comes from fits to the cluster yields using the modified Fisher formula to include the complement (see Eq. (13)). The long dashed curve comes from the normal Fisher description (see Eq. (10)) of the cluster yields. The medium dashed curve represents the integral of the Clapeyron equation with a simple accounting of the molar surface enthalpy (see Eq. (8)).

hold the drop comfortably away from the walls. The size of the drop A_0, and not of the lattice L, is here of paramount importance, and the finite size effects we worry about are those of the drop.

Lord Rayleigh used the Clapeyron equation $dp/dT = \Delta H_m / T \Delta V_m$ with the molar enthalpy of vaporization $\Delta H_m = \Delta H_m^0 + \frac{2}{3} c A_0^{-1/3}$ modified from its bulk form (ΔH_m^0) to account for the molar surface enthalpy of the drop. For a dilute vapor and constant ΔH_m^0, this integrates to

$$p(T, A_0) = p_\infty(T) \exp\left(\frac{\sigma c}{A_0^{1-\sigma} T}\right). \qquad (8)$$

where $p_\infty(T)$ is the vapor pressure in coexistence with an infinite liquid and σ is the surface to volume ratio ($\sigma \approx 2/3$ for 3D and $1/2$ for 2D). The bulk solution $p_\infty(T)$ is semi-analytic for 2D lattice gas calculations [14]. Thus we expect that the pressure of the lattice gas vapor in equilibrium with a finite drop (symbols in bottom panel of Fig. 2) should be greater than that of the vapor in equilibrium with an infinite

liquid (dotted line). This is the case, and the vapor pressures for equilibrium with a finite drop (symbols) are qualitatively described by Eq. (8) (medium dashed curve). The droplet pressures were calculated assuming an ideal gas $p(T, A_0) = T \sum_A n_A(T, A_0)$, where n_A is the concentration of clusters of size A. The expected enhancement above p_∞ is given approximately by $\exp(8c/15A_0^{7/15}T)$ for two dimensions (with $c = 8$ and $\sigma = 8/15$).

A similar enhancement is expected for the density

$$\rho(T, A_0) = \rho_\infty(T) \exp\left(\frac{\sigma c}{A_0^{1-\sigma}T}\right) \qquad (9)$$

where ρ is calculated as $\rho(T, A_0) = \sum_A A n_A(T, A_0)$ and $\rho_\infty(T)$ is given by the Onsager solution [13] for the magnetization. A comparison is given in the upper panel of Fig. 2. The densities of Eq. (9) qualitatively describe the calculated densities.

Now we examine the vapor in detail rather than globally. Time-honored theories for non-ideal gases (physical cluster theories) assume that the monomer-monomer interaction exhausts itself in the formation of physical clusters in equilibrium with the liquid and themselves. The epigon and most famous of these theories is Fisher's theory of clusterization, or Fisher's drop model [7]. The cluster concentration is described by

$$\begin{aligned} n_A(T) &= g(A) \exp\left(-\frac{c_0 A^\sigma}{T}\right) \\ &= q_0 A^{-\tau} \exp\left(c_0 A^\sigma \left[\frac{1}{T_c} - \frac{1}{T}\right]\right) \quad (10) \end{aligned}$$

where q_0 is the normalization, $c_0 A^\sigma$ is the surface energy of the cluster and $g(A)$ is the degeneracy of the clusters of size A (the number of ways the cluster A can be realized through different surface configurations) and is approximately $A^{-\tau} \exp(c_0 A^\sigma / T_c)$. The resulting entropy $S(A)$ is given by

$$S(A) \approx \ln g(A) \approx -\tau \ln A + \frac{c_0 A^\sigma}{T_c}. \qquad (11)$$

Eq. (11) is a remarkably felicitous asymptotic expansion. The presence of a leading term in S proportional to A^σ permits the vanishing of the cluster free energy at a $T = T_c$ independent of cluster size. This expression, valid for a vapor in equilibrium with the infinite liquid, needs to be generalized for a vapor in equilibrium with a finite drop. This can be done by making a preliminary observation: in Fisher's expression the abundance of each cluster in the vapor is Poissonianly distributed by construction [15]. This is because the resulting partial vapors are considered as non-interacting. In the Ising model this same situation prevails to a surprising degree as can be seen in refs. [15, 16]. The Poisson nature of the multiplicity distributions allows us to introduce the concept of the complement.

Consider a vapor in equilibrium with a drop of its liquid. The system may be a physical system or the Ising realization of it. For each cluster of the vapor we can make the mental exercise of extracting it from the liquid, determining the change in entropy and energy of the drop and cluster system, and then putting it back in the liquid (the equilibrium condition), as if all other clusters of the vapor did not exist. Fisher's expression can be generalized for a drop of size A_0 in equilibrium with its vapor as follows

$$\begin{aligned} n_A(T) &= \frac{g(A)g(A_0 - A)}{g(A_0)} \\ &\quad \exp\left\{-\frac{c_0}{T}[A^\sigma + (A_0 - A)^\sigma - A_0^\sigma]\right\} (12) \end{aligned}$$

or

$$n_A(T) = q_0 \left[\frac{A(A_0 - A)}{A_0}\right]^{-\tau} e^{[A^\sigma + (A_0 - A)^\sigma - A_0^\sigma]\left(\frac{c_0}{T_c} - \frac{c_0}{T}\right)}. \qquad (13)$$

In other words, we treat the "complement" $(A_0 - A)$ in the same fashion as a cluster. The resulting expression reduces to Eq. (10) when A_0 tends to infinity. While different than the standard Fisher expression, Eq. (13) still admits the <u>same</u> T_c as that of the infinite system. This is because the A_0, A dependence of the surface energy finds its <u>exact</u> counterpart in that of the entropy.

Our generalized form of the Fisher expression (Eq. (13)) can be used to fit vapor concentrations in equilibrium with a finite droplet. We can then look directly at a "Fisher" like scaling plot of all the $n_A(T)$ in detail. Given Eq. (13) we can use any drop size and automatically correct for it by plotting

$$\frac{n_A(T)}{\left(\frac{A[A_0 - A]}{A_0}\right)^{-\tau}} \qquad (14)$$

versus

$$c_0(A^\sigma + [A_0 - A]^\sigma - A_0^\sigma)\left(\frac{1}{T_c} - \frac{1}{T}\right). \qquad (15)$$

This is shown extensively in Fig. 3 for a variety of vapors in equilibria with drops of different sizes in two dimensions. The accurate scaling that is observed vouches for the complete generality of Eq. (13) and for a satisfactory result in our initial quest for a finite size treatment. The value of the critical temperature adjusted in the fit is $T_c = 2.28 \pm 0.02$. This is to be compared with the theoretical value of 2.269. This good agreement is remarkable. Using the fits to the vapor concentrations, we have have summed the Fisher concentrations of Eq. (13) (modified for the complement) to calculate the pressure and density of the vapor in coexistence with a droplet of size 80 (the solid curves in Fig. 2). The agreement is excellent. Furthermore, we can then take the calculated Fisher concentrations in the limit that A_0 goes to infinity
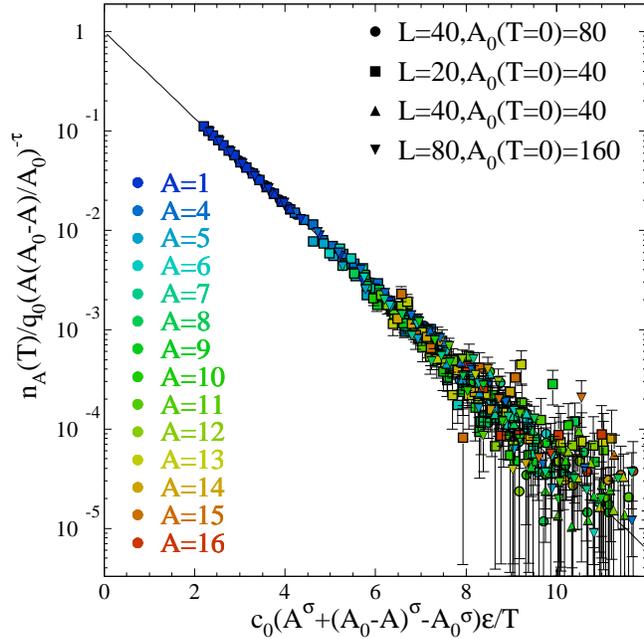
FIG. 3: The scaled cluster yields as a function of the surface energy with the complement for four different combinations of lattice size L and mean density ρ. The critical temperature dependence on lattice size is taken into account as $T_c(L) = T_c(\infty)(1 - .13/L)$. The T_c dependence on size of the drop is taken into account by the complement. Fitting parameters and results were $T_c(\infty) = 2.28 \pm 0.02$ (compare with the Onsager result of $2/\ln(1 + \sqrt{2}) = 2.26919\ldots$), the surface energy coefficient $c_0 = 8.23 \pm 0.03$ (expected value of 8).

(Eq. (10)) and predict the pressure and density in coexistence with a bulk liquid (the long-dashed curves in Fig. 2). This extrapolation agrees well with the known analytic results. Employing the complement along with the Fisher drop model allows one to describe quantitatively vapors in equilibrium with finite drops as well as bulk matter.

The generality of the modified Fisher description of Eq. (13) is of course true only in the coexistence region. Should one extend the scaling using distributions in the bivariant (single phase) region outside the coexistence (two phase) region, one would do that at his/her peril, as the Fisher description is not defined for that. The drastic change in cluster distribution that occurs as one samples calculations from inside and outside the coexistence region, together with the lack of accounting for the complement, explains the recent results of references [17] and [18] which report at different average densities good Fisher plots pointing to very different T_c values.

The critical temperature determined by this procedure does not change with drop size. Yet a study of the unconstrained Ising model at different sizes without periodic boundary conditions shows the well known scaling $\frac{T_c(L)}{T_c^\infty} \propto 1 - L^{-1/\nu}$. We have incorporated this correlation-length scaling into our fits.

The complement approach presented here should account not only for finite size effects in the surface energy but also the Coulomb and symmetry terms contained in the liquid drop model as well. Thus we can obtain a generalized Fisher formula applicable to any system we choose. Fitting such an expression to experimental data and using standard liquid drop parameters we can extract, in a straightforward manner, the critical temperature of the symmetric uncharged infinite nuclear matter in a consistent way [9].

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