Critical Behavior in the Coexistence Region of Finite Systems

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The liquid-gas phase transition in finite systems is studied within a lattice gas model in the canonical ensemble. In the coexistence region, the existence of conservation laws is shown to result in anomalies in the associated equation of state leading, for example, to negative compressibility due to surface effects. The associated partitions exhibit scaling behavior inside the coexistence zone. When the thermodynamical limit is taken this scaling disappears while the anomaly of the equation of state becomes the usual nonanalytical behavior. Therefore, in the fragmentation of small systems such as nuclei the experimentally observed critical behavior is demonstrated to be compatible with a first order phase transition because of finite size effects. [S0031-9007(99)08488-4]

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Two of the most fascinating phenomena in the thermodynamics of complex systems are phase transitions and critical behavior [1,2]. Over the years, it has been recognized that phase transitions exhibit generic properties. In particular, second order phase transitions have been classified in various universality classes which depend upon few characteristics of the considered system such as the dimensionality of the space and of the order parameter. In the same class, various systems present a similar phenomenology and, in particular, they have identical critical exponents. This remark has opened a wide range of applications for exact computer Monte Carlo simulations of simplified models as the Ising model [3] which may be used to study generic properties of the universality class to which they belong.

On the other hand, the experimental study of phase transitions in finite systems has recently attracted a strong interest from various communities. Bose condensates with a small number of particles and vaporization of small hot systems such as atomic clusters or atomic nuclei are examples of recent attempts to study phase transitions in finite systems.

In the nuclear case, considering the analogy between the nucleon-nucleon force and a van der Waals interaction, it is expected that the equation of state (EOS) presents the characteristics of a liquid-gas phase transition [4]. In recent years, different indications of such a transition have been accumulated [5–8]. Statistical equilibrium models, extremely successful in reproducing multifragmentation patterns, systematically suggest low freeze out densities [7–9] which would indicate that the transition occurs in the middle of the coexistence region.

On the other hand, the observation of critical behaviors like power laws in the charge distribution of the multifragmenting system [6] has been interpreted as an evidence of a second order phase transition. Indeed, in the Fisher droplet model a power law behavior is expected at the critical point when the liquid and vapor chemical potentials are equal and the surface tension is zero [10,11]. Recently, power laws have been also reported along the Kertész line [12] at higher temperature and densities [13] inside the single fluid part of the phase diagram. The presence at the same time of first and second order transition signals is still a puzzle and the density at which the partitioning of the system occurs is still an open question.

In this paper we shall concentrate on the liquid-gas phase transition in a canonical framework. The use of the canonical constraint allows an unambiguous definition of the phase diagram even for a finite system and a direct exploration of the coexistence region. The main result is that a critical behavior in fragment observables can be consistent with phase coexistence and the occurrence of a low freeze-out density due to finite size effects. We shall show that the partitions of small systems in the coexistence region may mimic a critical behavior. This conclusion will be reached through the use of the finite size scaling properties of fragment size distributions. The critical signals inside the coexistence region appear to be suppressed in the thermodynamical limit so that the reported effect can be considered as a specific signature of phase transitions in finite systems.

In order to reach this conclusion we shall study an exactly solvable model for second and first order phase transitions, the lattice gas model of Yang and Lee [14]. This model in the grand-canonical formalism is equivalent to the canonical ensemble of an Ising model in the presence of an external magnetic field. It has already been applied to nuclear physics in Refs. [13,15–17]. In the canonical ensemble only an approximate [18] sampling has been employed by the authors of Ref. [16]. In this work we use the standard Metropolis sampling [19] where the configurations are chosen according to the canonical probability $\exp(-\beta E)$ and are weighted evenly.

In our implementation the n sites of a lattice are characterized by an occupation number τ which is defined as $\tau = 0(1)$ for a vacancy (nucleon). Particles occupying nearest neighboring sites interact with a constant energy ϵ. The Hamiltonian is given by

$$H = \sum_{i=1}^{n} \frac{p_i^2}{2m} \tau_i^2 + \sum_{i \neq j} \epsilon \tau_i \tau_j . \tag{1}$$

Events are generated by placing a number A_0 of particles (corresponding to a density $\rho = A_0/n$) on a three-dimensional cubic lattice according to the statistical canonical probability. In order to avoid surface effects periodic boundary conditions are imposed.

The thermodynamics of the model is calculated from a direct evaluation of the partition sum

$$Z(A_0, \beta) = \sum_E W(E) e^{-\beta E}, \tag{2}$$

where $W(E)$ is the degeneracy of the state with energy E. This evaluation is performed via an iterative procedure [20]. At a temperature $T = 1/\beta$, the number of sampled realizations of the system with an energy E is

$$N(E, \beta) = N \frac{1}{Z(A_0, \beta)} W(E) e^{-\beta E}, \tag{3}$$

where N is the total number of sampled states. From the comparison of two different temperatures $T_1 = 1/\beta_1$ and $T_2 = 1/\beta_2$ one then has

$$Z(A_0, \beta_1) = Z(A_0, \beta_2) \frac{N(E, \beta_1)}{N(E, \beta_2)} e^{-E(\beta_2 - \beta_1)}, \tag{4}$$

which is valid for all the different energy bins. In order to profit from all the available data we can compute the partition sum as an average of the above relation over the various energy bins weighted by $\sqrt{N(E, \beta_1) N(E, \beta_2)}$.

Z is obtained iteratively via Eq. (4) with an initial normalization to the infinite temperature limit where the partition sum is $Z(A_0, 0) = n!/A_0!(n - A_0)!$. The equation of state can then be obtained from a numerical derivative of the free energy, $F(A_0, T) = Af(A_0, T) = T \log Z(A_0, \beta)$. In Fig. 1 thermodynamical variables are shown for a lattice of size $n = 216$ at a subcritical temperature. In the infinite system the thermodynamical potential must be convex. However, for the free energy of the finite system (Fig. 1c) we observe a concave pattern due to the surface energy between the liquid and the vapor in the mixed phase. This behavior induces a backbending of the chemical potential μ (Fig. 1a). In infinite systems one would expect the chemical potential to stay constant inside the coexistence region. This corresponds to a nonanalytical behavior of F which can be obtained only when the volume diverges. In a finite system the partition sum is always analytical and the chemical potential presents a smooth backbending. This "anomaly" of an intensive variable (e.g., μ) as a function of its extensive conjugated variable (e.g., A_0) or equivalently the nonconvex behavior of the thermodynamical potential can be considered as the signature of the phase transition in a finite system. This behavior can be observed only if in the considered sampling a conservation law applies to the extensive variable. An analogous backbending was found by Gross in the temperature versus energy plot when computed in the microcanonical ensemble [21].

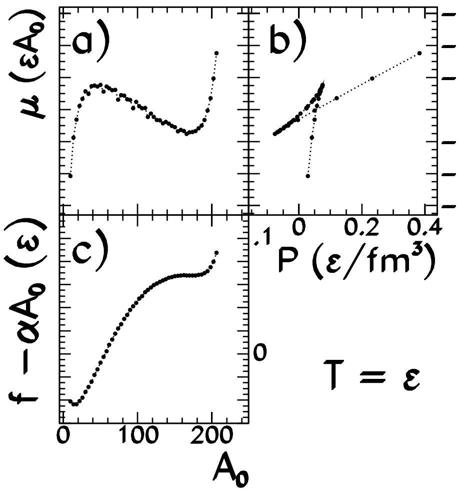

FIG. 1. Chemical potential, pressure, and free energy as a function of the particle number at temperature $T = \epsilon$ for a cubic lattice of size $n = 216$.

In the presented results the limit of the coexistence zone can be accurately spotted on the chemical potential versus pressure plane (Fig. 1b). At low temperature, one can clearly observe two branches characteristic of a gas phase at low pressure and of a liquid phase at high density. The coexistence of two phases disappears above a critical temperature leaving a single smooth fluid isotherm. By making a systematic analysis of the crossing points of the two branches, the coexistence zone can be accurately evaluated leading to a critical temperature $T_c = 1.22\epsilon$ and a critical exponent $\beta = 0.31$ for the temperature dependence of the order parameter $\rho - \rho_c = (T - T_c)^\beta$. The corresponding values for $n = 512$ are $T_c = 1.20\epsilon$ and $\beta = 0.31$. These values have to be compared with the expected value in the thermodynamical limit $T_c^\infty = 1.12\epsilon$, $\beta^\infty = 0.33$.

In order to get a deeper insight into the partitions inside the coexistence region we have studied the fragment size distribution. The definition of clusters has been extensively discussed in the literature [13,16]. The first idea is to group all the connected sites (Ising clusters). However, it has been known for a long time that this is not the proper way to define clusters in the lattice gas model since it does not fulfill the requirement that the correlation length should diverge at the critical point (see [13]). Using renormalization group arguments, Coniglio and Klein proposed to combine the above site percolation with an additional bond percolation algorithm using a temperature dependent bond breaking probability [22] $p(T) = e^{-\epsilon/2T}$. We have employed an almost equivalent way to define clusters [13,16] which consists of introducing a kinetic term in the Boltzmann probability and then breaking the bond between two nucleons as soon as the kinetic energy of their relative motion $\vec{p}_r^{\,2}/2\mu$ exceeds the binding energy ϵ.

From finite size scaling one expects that at temperatures close to the critical point the size distribution scales as [2,23]

$$\frac{dN}{dA}(A,T) = A^{-\tau} f\big(A^{\sigma}(T - T_c)\big), \tag{5}$$

where f is a scaling function and τ, σ are critical exponents. Two values of the universal function are of particular interest:

(i) Its value at the origin $f(0) = f_0$ since it is associated with the critical point

$$\frac{dN}{dA}(A,T_c) = A^{-\tau} f_0 . \tag{6}$$

(ii) Its maximum value $f(x_{\max}) = f_{\max}$. For each size A there exists a temperature $T_{\max}(A) = T_c - x_{\max}A^{-\sigma}$ for which the production is maximum

$$\left.\frac{dN}{dA}\right|_{\max}(A) = \frac{dN}{dA}\big(A, T_{\max}(A)\big) = A^{-\tau} f_{\max} . \tag{7}$$

To test the validity of Eq. (5) for the partitions computed at constant density we have used the following procedure. The maximum production yield $dN/dA|_{\max}$ of a species of size A as a function of A is fitted to a power law of exponent $\tau_{\max}$. Then T_c can be obtained as the temperature at which the power law fit to the size distribution $dN/dA(A)$ gives an exponent $\tau = \tau_{\max}$. In finite systems the minimum value of τ is systematically lower than $\tau_{\max}$; therefore, two temperatures fulfill this condition. However, only the lowest one leads to a scaling behavior and can be identified with T_c. The temperature $T_{\max}(A)$ at which the production yield of size A is maximum is fitted to a power law of exponent σ. At the thermodynamical critical density $\rho_c = 1/2$ we observe that the scaling behavior of intermediate mass fragments is remarkably fulfilled over a wide range of temperatures even far away from the critical temperature (Fig. 2, central panel). The extracted values of the parameters at the density $\rho_c = 1/2$ for $n = 512$ are very close to the exponents of the 3D-Ising universality class (see Table I). These results are fully compatible with the values $\beta = 0.31$, $T_c = 1.2\epsilon$ extracted from the coexistence curve using the relation $\beta = (\tau - 2)/\sigma$.

We have performed the same analysis at different densities. As illustrated in Fig. 2, a scaling behavior is clearly seen at supercritical as well as subcritical densities. For various densities, typical mass distributions at temperatures close to the critical temperature are displayed in Fig. 3 (upper part). The critical exponents exhibit a small density dependence (shown in Table I for a few selected density points) while the critical temperature increases with density and coincides with the critical temperature of the coexistence curve only at the thermodynamical critical density ρ_c (Fig. 3, lower part). For all data points presented in Fig. 3 the quality of finite size scaling is comparable to the one shown in Fig. 2. The values of the critical exponents are also compatible with the experimental values recently reported in Ref. [6] in the case of nuclear fragmentation.

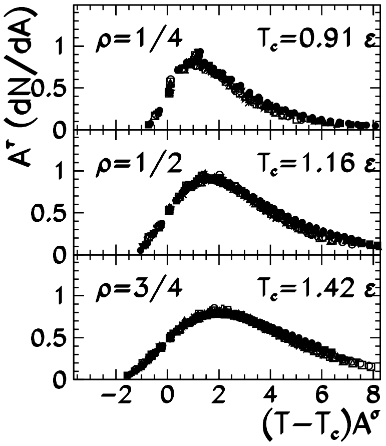

FIG. 2. Scaling function for clusters of size $6 \le A \le 30$ and temperature $0.36 \le T/\epsilon \le 3.6$ obtained from Eq. (5) at three different densities for a cubic lattice of size $n = 512$.

It has already been observed that power laws and minima of the function $\tau(T)$ from power law fits of the size distribution are not characteristic of the critical point solely but occur also at supercritical densities along the Kertész line [13] and at some subcritical densities at lower temperatures [16,24]. However, the possibility to observe finite size scaling in small systems inside the coexistence region is demonstrated here for the first time. The presented results show, in agreement with the work of Pan *et al.* [24], that there is no contradiction between the scenario of nuclear fragmentation inside the coexistence or the spinodal region associated with a first order phase transition and the observed critical signals [6] in fragment observables. However, contrary to Ref. [24], we have shown that the critical temperature extracted from fragment observables has no thermodynamical meaning at low density. As a matter of fact, the physical origin of the scaling behavior at subcritical densities lies on the finite size of the system and not on the existence of critical fluctuations of all sizes induced by a divergence of the

TABLE I. Critical exponents for a lattice size $n = 512$.

Density	τ	σ
1/6	2.16 ± 0.11	0.58 ± 0.07
1/3	2.19 ± 0.07	0.70 ± 0.10
1/2	2.20 ± 0.05	0.61 ± 0.16
2/3	2.21 ± 0.05	0.56 ± 0.07
5/6	2.25 ± 0.05	0.63 ± 0.07

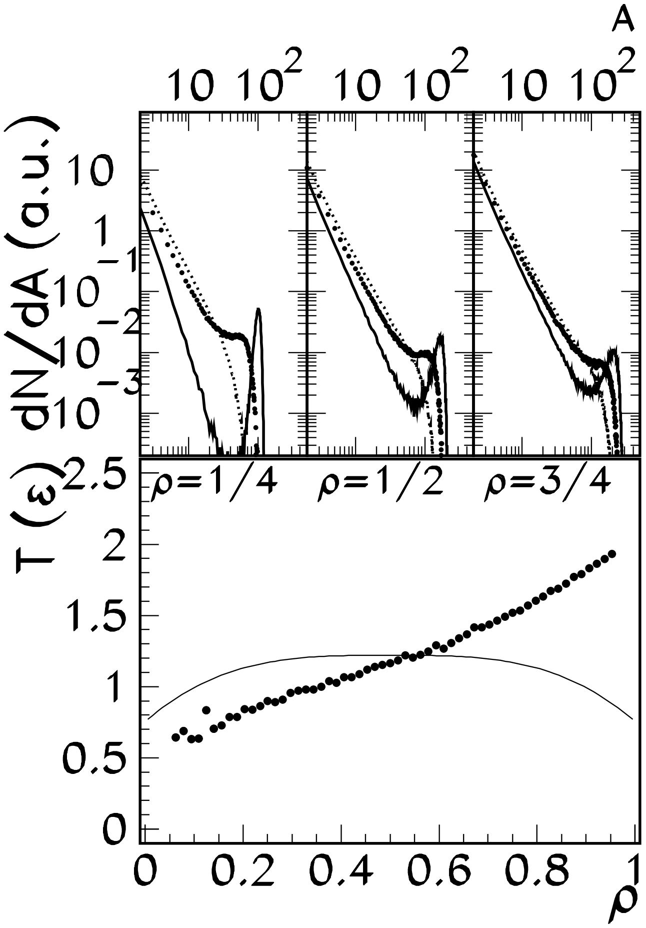

FIG. 3. Lower part: (full line) coexistence line calculated using the partition sum; (symbols) critical curve from fragment size distributions. Upper part: cluster size distributions at three different densities and temperatures $T = T_c(\rho)$ (symbols), $T = T_c(\rho) \pm 0.2\epsilon$ (lines). The lattice size is $n = 512$.

correlation length. In fact, in the coexistence region of small systems the liquid cluster is not much larger than the vapor fragments and may mimic critical fluctuations of the mass distribution. Going to the thermodynamical limit, inside the coexistence zone the liquid piece becomes infinite so that it clearly separates from the intermediate mass fragments. The observed power law in the coexistence region is then suppressed when going to the infinite size limit and the critical line abruptly stops on the coexistence boundary at the critical point. In practice this starts to be observed in systems with approximately $n = 125\,000$: Inside the coexistence region a large fragment well separated from vapor clusters is clearly observed in the mass distribution. Therefore, below the critical density the power law and the related scaling is never achieved.

In this paper we have shown that the liquid-gas phase transition in finite systems is associated with nontrivial partitions. They induce modifications of the partition sum resulting in anomalies in the equation of state such as negative compressibility, which can be used to unambiguously define phase transition even in small systems. We have stressed the fact that these anomalies are due to finite size effects and will result in nonanalytical behaviors only when the thermodynamical limit is reached.

These partitions in small systems present scaling behavior with critical parameters close to the ones expected for the liquid-gas universality class. The scaling inside the coexistence region disappears as soon as large systems are considered.

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