Caloric Curves and Energy Fluctuations in the Microcanonical Liquid-Gas Phase Transition

Ph. Chomaz,[1] V. Duflot,[1,2] and F. Gulminelli[2]
[1]*GANIL (DSM-CEA/IN2P3-CNRS), B.P. 5027, F-14021 Caen cedex, France*
[2]*LPC Caen, (IN2P3-CNRS/ISMRA et Universite), F-14050 Caen cedex, France*
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In this paper we study a microcanonical lattice gas model with a constrained average volume. We show that the caloric curve explicitly depends on the considered transformation of the volume with the excitation energy and so does not bear direct information on the characteristics of the phase transition. Conversely, partial energy fluctuations are demonstrated to be a direct measure of the equation of state. Since the heat capacity has a negative branch in the phase transition region, the presence of abnormally large kinetic energy fluctuations is a signal of the liquid-gas phase transition.

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One of the most important challenges of heavy ion physics is the identification and characterization of the nuclear liquid-gas phase transition. Since nuclei contain only a few hundreds of particles at most we are forced to address the general problem of the definition and identification of phase transitions in finite systems. This is the case of many other microscopic or mesoscopic systems: well-known examples are melting and vaporization of metallic clusters, Bose condensation of atoms in traps, and deconfinement of dense hadronic matter towards quark and gluon plasma. It has been proposed that the phase transition in finite systems within the microcanonical ensemble [1,2] may be signed through an anomalous backbending behavior in the functional relationship between the temperature and the excitation energy, the so-called caloric curve. Therefore a first order phase transition should correspond to a negative branch for the heat capacity.

Many different measurements of the nuclear caloric curve have been performed [3], showing however quite different behaviors. Simultaneously it has been shown that negative heat capacities can be signed through the occurrence of abnormally large kinetic energy fluctuations [4,5]. This new signal of a first order phase transition has been applied to multifragmentation data and a liquid-gas phase transition has been tentatively identified [6]. Statistical models [1,2,7] which are very successful in reproducing experimental data also show important fluctuations which support this idea [6]. The link between the observation of monotonic caloric curves and the measure of negative heat capacities is the puzzle we want to address in this Letter.

In the liquid-gas phase transition the volume is directly related to the order parameter, which means that a second state variable has to be introduced in order to specify the volume. In this paper we illustrate within the lattice gas model numerically solvable without any approximation that if the system passes through coexistence it can be traced back from the study of kinetic energy fluctuations since this signal is directly related to the equation of state. On the other hand, we show that the caloric curve is a more indirect way to look for the phase transition because it depends not only on the equation of state but also on the considered path in the state variables plane.

In our implementation of the lattice gas model of Lee and Yang [8] the L^3 sites of a cubic lattice are characterized by an occupation number τ which is defined as $\tau = 0$ for a vacancy, and $\tau = 1$ for a nucleon. Particles occupying nearest neighboring sites interact with an energy ϵ. The Hamiltonian is given by

$$H = \sum_i \frac{p_i^2}{2m} \tau_i + \frac{1}{2} \sum_i \sum_{j \in \mathcal{N}_i} \epsilon \tau_i \tau_j , \qquad (1)$$

where the second sum runs only over the closest neighbors $\mathcal{N}_i$ of i. The coupling constant $\epsilon = -5.5$ MeV is fixed so as to reproduce the saturation energy.

In finite systems, the various statistical ensembles are not equivalent. The elementary ensemble is the microcanonical ensemble because its entropy is directly related to the density of states. Moreover in practical applications the microcanonical ensemble is most adapted since the total energy can almost always be defined and/or measured on an event per event basis. For systems undergoing a liquid-gas phase transition the volume is also an essential degree of freedom. Various ensembles [5,9–14] with a volume fixed through boundary conditions have already been considered in the lattice gas model context. In actual experiments however the volume is not defined through boundary conditions. At best, an average size of the fragmenting system can be inferred from experimental observables. From the theoretical point of view one is therefore forced to consider a statistical ensemble for which the volume can fluctuate from event to event around an average value [7]. Different choices can be made to measure the average size of the system. We present here results with the one body volume observable proportional to the cubic radius [15] $\hat{\mathbf{V}} = \frac{4\pi}{3A} \sum_{i=1}^{A} r_i^3 \tau_i$ where r_i is the distance to the center of the lattice, but the main results of the paper do not depend on this choice. Introducing the associated Lagrange multiplier λ in order to constrain a specific value for the average volume when maximizing the entropy (to get the least biased statistical ensemble), we come to a microcanonical ensemble defined through the partition function

$$Z_\lambda(E) = \sum_V W_V(E)\exp(-\lambda A V) \qquad (2)$$

with the density of states $W_V(E)$ having a energy E and a size V.

In the statistical ensemble (2) the energy E and the Lagrange conjugate of the volume observable λ represent the two state variables of the system. They are associated with two equations of state, giving the microcanonical temperature $T_\lambda^{-1} \equiv \partial_E \log Z_\lambda$ and the average volume $\langle V\rangle_\lambda$ as a function of E and λ. λ can be formally related to a pressure via $P_\lambda = T_\lambda \lambda$. The ensemble (2) is not the standard microcanonical ensemble defined by the entropy $S = \log W_V(E)$. To avoid misunderstandings, we shall in the following note the temperature, pressure, and average volume by T_λ, P_λ, and $\langle V\rangle_\lambda$.

In the calculations shown below a number $A = 216$ of particles is fixed while L^3 is large enough (typically greater than 20^3 lattice sites) so that the boundary conditions do not affect the calculations with a constraining λ. The microcanonical results are obtained through a sorting of canonical events generated with a standard Metropolis sampling (for more details see [12,13]). If N states are sampled with a given constraining β and λ, the canonical energy distribution of events reads

$$N_{\beta,\lambda}(E) = \frac{N}{z_{\beta,\lambda}} Z_\lambda(E)\exp(-\beta E), \qquad (3)$$

where $z_{\beta,\lambda}$ is the canonical partition sum. $\log N_{\beta,\lambda}(E)$ directly leads to the microcanonical equation of state at constant λ so that

$$T_\lambda^{-1}(E) \equiv \frac{\partial S_\lambda(E)}{\partial E} = \beta + \frac{\partial \log N_{\beta,\lambda}(E)}{\partial E}. \qquad (4)$$

Since this equation is valid for every β, we can use many canonical samplings at different β to derive the same microcanonical caloric curve. The agreement between the different curves provides a strong test of the numerical sampling [5]. Other tests of the Metropolis procedure can be found in Ref. [16].

The various isotherms in the $(P_\lambda, \langle V\rangle_\lambda)$ plane are displayed in Fig. 1. Far from coexistence the curves $P_\lambda(\langle V\rangle_\lambda)$ at constant temperature are monotonous. However, when we get close to the coexistence region we observe an anomalous backbending. The physical origin of this negative compressibility is completely different from the usual mean field result which reflects the instability of the homogenous system with respect to phase separation. In our exact calculation which naturally includes all inhomogeneous partitions conversely this feature corresponds to equilibrium under specific conservation laws (here mass number and energy) [9,16]. A backbending is also apparent in the two-dimensional caloric curves, $T_\lambda(E)$ (Fig. 2) leading to a negative branch in the microcanonical heat capacity at constant λ: $C_\lambda^{-1} = \partial T_\lambda(E)/\partial E$.

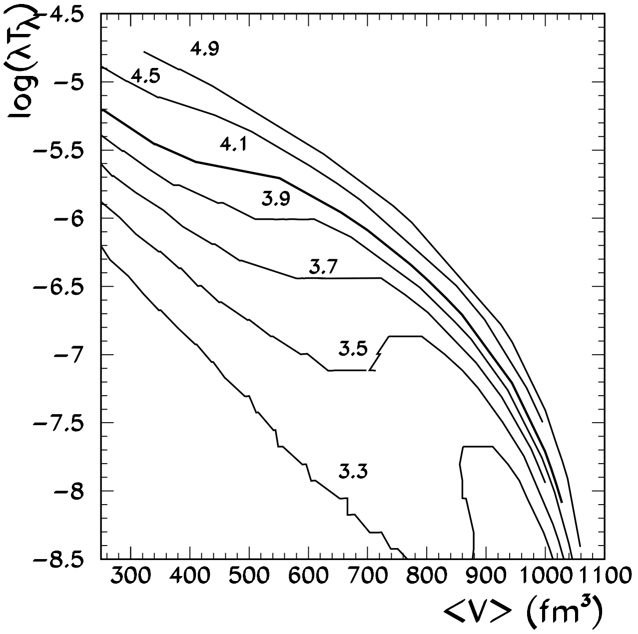

FIG. 1. Correlation between pressure $P_\lambda = \lambda T$ and volume for a system of 216 particles in the ensemble (2). The microcanonical temperature is indicated on each isotherm. The thick line gives the critical isotherm.

The microcanonical heat capacity can be measured using partial energy fluctuations [4]. The total energy E can be decomposed into two independent components, its kinetic (E_K) and interaction energy (E_I): $E = E_K + E_I$.

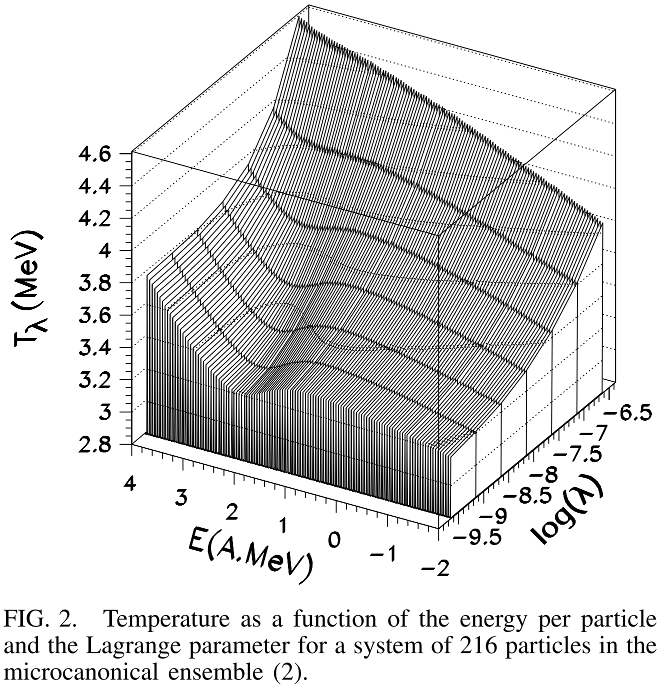

FIG. 2. Temperature as a function of the energy per particle and the Lagrange parameter for a system of 216 particles in the microcanonical ensemble (2).

Since the energy partition directly depends on the partial entropies S_K and S_I the kinetic energy variance can be related in the Gaussian approximation to the heat capacities [4,17]

$$\sigma_K^2 \simeq T^2 \frac{C_K C_I}{C_K + C_I}, \tag{5}$$

where C_K and C_I are the kinetic and interaction microcanonical heat capacities calculated for the most probable energy partition characterized by a microcanonical temperature T. Equation (5) can be inverted to extract from the observed fluctuations an approximation for the heat capacity [4]

$$C \simeq C_K + C_I \simeq \frac{C_K^2}{C_K - \sigma_K^2/T^2}. \tag{6}$$

Even if in the following Eq. (6) appears to be very accurate, it should be noticed that it is only the leading order of a Gaussian approximation and that correction terms can be evaluated as discussed in [4]. From Eq. (6) we can see that when the heat capacity becomes negative σ_K overcomes the canonical expectation $\sigma_K^2/T^2 = C_K$. In the classical model $C_K \approx (3/2)A$. It is amazing to observe that the constraint of energy conservation leads in the phase transition region to larger fluctuations than in the canonical case where the total energy is free to fluctuate. This is because the kinetic energy part is forced to share the total available energy with the interaction part. When the interaction part presents a negative heat capacity the jump from liquid to gas induces strong fluctuations in the energy partitioning. The normalized fluctuations σ_K^2/T_λ^2 obtained in the microcanonical ensemble with a constrained average volume, $\langle V \rangle_\lambda$, are shown in the energy-λ plane in Fig. 3 together with the isotherms. One can clearly see that up to the critical temperature the fluctuations are abnormally large in the coexistence region. From Figs. 2 and 3 it is apparent that the phase transition signal is visible in the temperature as well as in the fluctuation observable.

However the experimentally measured caloric curves are not bidimensional. Indeed, even if different sources with different excitation energies can be prepared, the other thermodynamical parameters are not controlled. In particular, an average value for the freeze-out volume of a selected ensemble of events can be deduced from interferometry and correlation measurements or through comparisons with statistical models but it cannot be varied independently of the deposited energy. This means that experiments are sampling a monodimensional curve on the equation of state surface characterized by a relation between E and λ: $\lambda(E)$. The resulting caloric curve therefore depends on the actual path in the thermodynamical parameters plane. As an example the behavior of the temperature as a function of energy at a constant pressure or a constant average volume in the subcritical region is displayed in the upper part of Fig. 4. In the first case the path $\lambda(E)$ is found solving the implicit equation $P_\lambda(E) = \text{const}$ while in the second case the relation $\langle V \rangle_\lambda(E) = \text{const}$ is used. In the coexistence region the isobars are almost identical to the iso-λ's since P_λ and λ differ only by the temperature which is almost constant in the phase transition region, and a backbending is clearly seen. On the other hand, at constant average volume a smooth behavior is observed with a slope change entering the gas phase, as expected from general thermodynamics (see also [10,14]). This is due to the fact that the λ parameter varies rapidly in the coexistence region if we ask to have $\langle V \rangle = \text{const}$

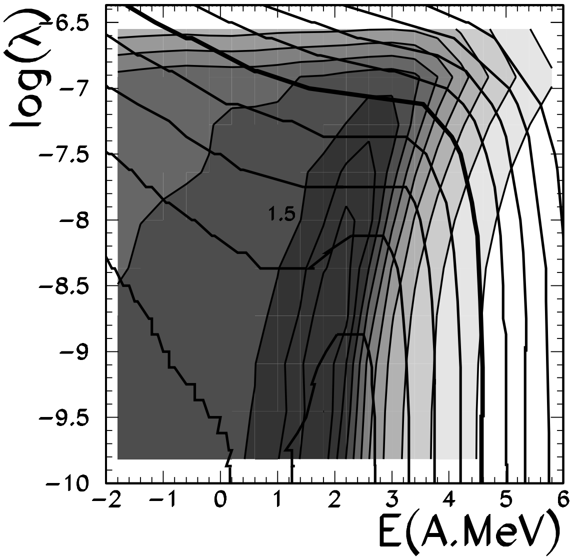

FIG. 3. Isotherms and grey contour plot of the normalized kinetic energy fluctuations in the Lagrange parameter versus energy plane. The level corresponding to the canonical expectation $\sigma_1^2/T^2 = 1.5$ is shown. Thick line: critical isotherm.

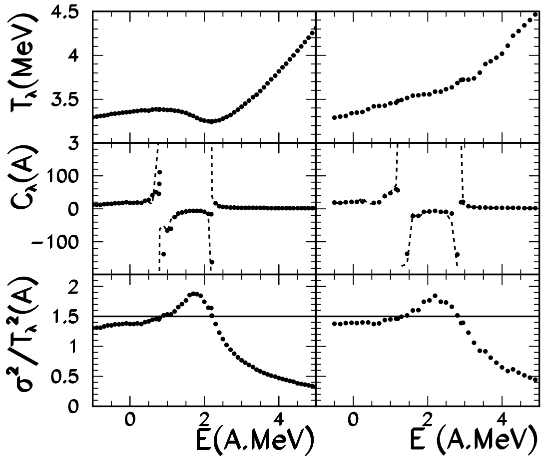

FIG. 4. Thermodynamical quantities in the ensemble (2) for a transformation at constant pressure (left part) and at constant average volume (right part). Upper panels: caloric curve. Lower panels: normalized kinetic energy fluctuations compared to the canonical expectation (lines). Middle panels: heat capacity (symbols) compared to the estimation through Eq. (6) (lines).

(see Fig. 1). The backbending of the temperature surface can thus be avoided depending on the path of the considered transformation and the phase transition signal can be hidden in the observation of the caloric curve. From these examples one clearly sees that the derivative of the caloric curve does not give directly the heat capacity of the considered ensemble ($C_\lambda^{-1} = \partial T/\partial E|_\lambda$) but contains an extra term which explicitly depends on the transformation $\lambda(E)$

$$\frac{d\mathbf{T}}{dE} = \left.\frac{\partial T}{\partial E}\right|_\lambda + \left.\frac{\partial T}{\partial \lambda}\right|_E \frac{\partial \lambda}{\partial E}. \qquad (7)$$

If $\lambda(E)$ follows a constant pressure path $\frac{d\mathbf{T}}{dE} = \frac{1}{C_P}$. If a constant volume transformation $\langle V \rangle_{\lambda(E)}(E) = \text{const}$ is followed, $\frac{d\mathbf{T}}{dE} = \frac{1}{C_V}$. On the other side partial energy fluctuations depend only upon the state and not on the path from one state to another and can directly give access to the equation of state $C_\lambda^{-1} = \partial T/\partial E|_\lambda$ using Eq. (6). From Fig. 3 we can see that in the whole phase transition region the microcanonical fluctuations become anomalously large. This signals that the system undergoes a first order phase transition, independently of the path. As an example the lower part of Fig. 4 shows a constant P_λ or $\langle V \rangle_\lambda$ cut of the bidimensional fluctuation surface shown in Fig. 3. The comparison of the exact heat capacity C_λ with the fluctuation approximation (middle part of Fig. 4) illustrates the accuracy of the estimation (6).

To summarize, in finite systems the equation of state depends explicitly on the considered statistical ensemble of events. In particular, a negative heat capacity is a well-defined signal of a first order phase transition when events are sorted in constant excitation energy bins. In the case of the liquid-gas phase transition one is forced to introduce a second thermodynamical variable in order to specify the volume of the system. Then a monodimensional curve such as the measured caloric curves can be misleading. Indeed, many different caloric curves can be generated depending on the path followed in the thermodynamical variable plane [7]. In a theoretical model this is not a problem since all paths can be studied and so the bidimensional equation of state can be inferred. One may even study the direction of the strongest curvature anomaly of the thermodynamical potential which could be a way to define the order parameter [4,18]. However, in an experimental situation this transformation cannot be controlled and is even hardly defined. Conversely, we have shown that partial energy fluctuations can provide a direct measure of the heat capacities. Considering a statistical ensemble of states characterized by total energy and an average volume we have shown that abnormal fluctuations are a signal of a first order phase transition. In this case kinetic energy fluctuations in the phase transition region are indeed related to the isobar heat capacity C_P which is known to diverge.

The connection to experimental fragmentation data is straightforward. The microcanonical ensemble is relevant for the analysis of experimental data because of the absence of a heat bath and since using calorimetry techniques the events can thus be sorted in constant energy bins, i.e., in microcanonical ensembles. As far as the freeze-out volume is concerned, the absence of a confining box implies that this variable can be known at best in average, leading to the statistical ensemble (2). Therefore, we expect that the partial energy fluctuations will present a strong anomaly if the multifragmenting nuclear system is undergoing a liquid-gas phase transition.

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