



Caloric curves and energy fluctuations in the microcanonical liquid-gas phase transition

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In this paper we study a microcanonical lattice gas model with a constrained volume. We show that the caloric curve explicitly depends on the considered transformation and does not bear direct informations 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 robust signature of the liquid gas phase transition.

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 or superfluid phase transition of quantum fluids, 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] may be signed through an anomalous back-bending behavior in the functional relationship between the temperature and the excitation energy deposited in the system, the so called caloric curve. The derivative of the caloric curve being by definition the heat capacity of the system, a first order phase transition should correspond to a negative branch for the heat capacity.

Many different measurements of the caloric curve have been performed [2], showing however quite different and generally smooth behaviors. Recently it has been shown that negative heat capacities can be signed also through the occurrence of abnormally large kinetic energy fluctuations [3,4]. 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 [5]. The link between the observation of monotonic and smooth caloric curves and the measure of negative heat capacities is therefore still a puzzle.

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. The phase transition will then be univocally signed by the abnormal convexity of the thermodynamical potential [1,6] in the state variables plane which reflects on the different equations of state. In this paper we show within an exactly solvable model that if the system passes through coexistence the anomaly can be traced back from the study of kinetic energy fluctu-

ations 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 transformation in the state variables plane.

In our implementation of the Lattice Gas Model of Lee and Yang [7] the N 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

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

where the second sum runs only over the closest neighbors \mathcal{N}_i of i . The coupling constant $\epsilon = -5.5 \text{ MeV}$ is fixed so 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. For systems undergoing a liquid-gas phase transition the volume is also an essential degree of freedom. Grandcanonical [8], canonical [9,6,10] and microcanonical [11,4] ensembles with a volume fixed through sharp or periodic boundary conditions have already been considered in the lattice gas model context. In actual experiments however the volume cannot be defined through boundary conditions but is an experimental observable known at best in average. 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. The one-body volume observable can be defined as

$$\hat{V} = \frac{4\pi}{3} \sum_{i=1}^N r_i^3 \tau_i \quad (2)$$

where r_i is the distance to the center of the lattice. Introducing the associated Lagrange multiplier λ in order to constrain a specific value for the average volume [12] when maximizing the entropy, we come to the microcanonical ensemble defined through the partition function

$$Z_\lambda(E) = \sum_V W_V(E) \exp(-\lambda V) \quad (3)$$

with the density of states $W_V(E)$.

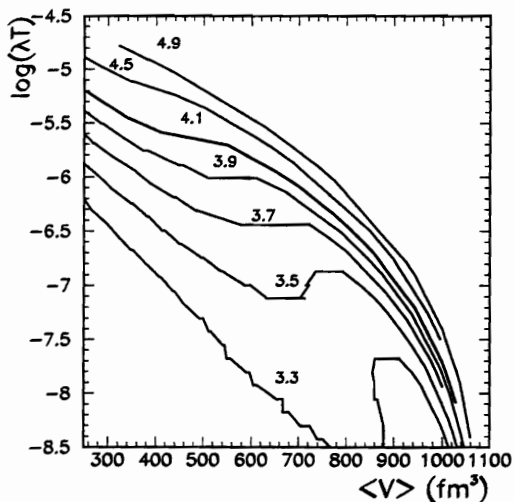


FIG. 1. Correlation between pressure and volume for a system of 216 particles in the microcanonical ensemble with fluctuating volume. The microcanonical temperature is indicated on each isotherm. The thick line gives the critical isotherm.

In the statistical ensemble (3) the energy E and the Lagrange conjugate of the volume λ represent the two state variables of the system. They are associated to two equations of states, giving the microcanonical temperature $T_\lambda^{-1} \equiv \partial_E \log Z_\lambda$ and the average volume $\langle V \rangle$ as a function of E and λ . It should be noticed that λ can be formally related to a pressure via $P = T_\lambda \lambda$.

In the calculations shown below a number $A = 216$ of particles is fixed. The numerical realization of the model is a three dimensional cubic lattice characterized by a size large enough so that the boundary conditions do not affect the calculations with a constraining λ . The microcanonical results are obtained through a simple sorting of canonical events. Canonical events are generated with a standard Metropolis sampling. If N states are sampled with a given constraining β and λ , the canonical energy distribution of events reads

$$N_{\beta,\lambda}(E) = \frac{N}{z_{\beta,\lambda}} Z_\lambda(E) \exp(-\beta E) \quad (4)$$

where $z_{\beta,\lambda}$ is the canonical partition sum. The logarithm

of the energy distribution $N_{\beta,\lambda}(E)$ directly leads to the microcanonical equation of state at constant λ

$$T_\lambda^{-1}(E) \equiv \frac{\partial S_\lambda(E)}{\partial E} = \beta + \frac{\partial \log N_{\beta,\lambda}(E)}{\partial E} \quad (5)$$

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 [4].

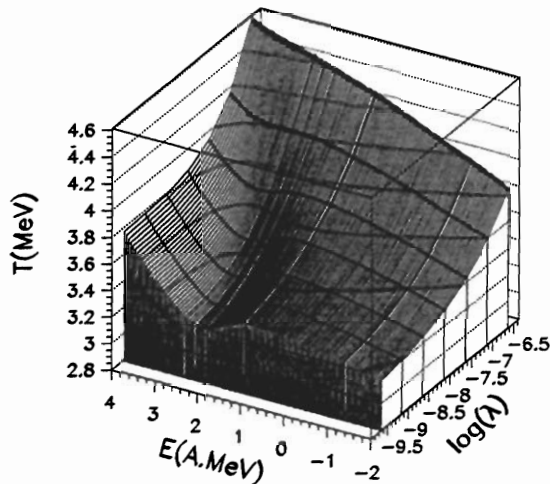


FIG. 2. Temperature as a function of the energy per particle and the Lagrange parameter for a system of 216 particles in the microcanonical ensemble with fluctuating volume.

The various isotherms in the $(P, \langle V \rangle)$ plane are displayed in Figure 1. Far from coexistence the curves $P(\langle V \rangle)$ at constant temperature are monotonous. However, when we get close to the coexistence region we observe an anomalous backbending. This induces a negative branch in the compressibility which has already been observed in canonical lattice gas calculations at constant volume [6,13]. The physical origin of this anomaly is completely different from the backbendings systematically found in the mean field approximation which simply reflect the instability of the homogenous system respect to phase separation. In our exact calculation which naturally includes inhomogeneous partitions conversely this feature corresponds to equilibrium under specific conservation laws (here mass number and energy) [6,13]. In the microcanonical case the same anomaly is apparent in the equation of state providing the temperature T as a function of the state variables (λ, E) as shown in the two-dimensional caloric curves displayed in figure 2. The backbending of the temperature surface induces a neg-

active branch in the microcanonical heat capacity at constant λ : $C_\lambda^{-1} = \partial T_\lambda(E) / \partial E$.

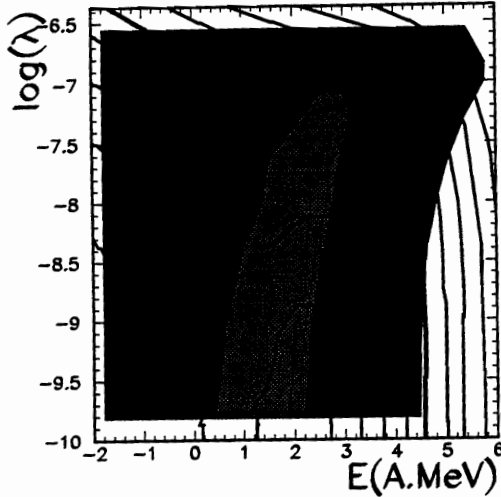


FIG. 3. Isotherms and contour plot of the normalized kinetic energy fluctuations in the Lagrange parameter versus energy plane. The level corresponding to the canonical expectation $\sigma_K^2/T^2 = 1.5$ is shown. Thick line: critical isotherm.

It has been recently proposed that the microcanonical heat capacity can be measured using partial energy fluctuations [3]. The total energy E of the considered system can be decomposed into two independent components, its kinetic (E_K) and interaction energy (E_I): $E = E_K + E_I$. Since the energy partition directly depends on the partial entropies S_K and S_I the kinetic energy variance can be calculated from the partial heat capacities

$$\sigma_K^2 = T^2 \frac{C_K C_I}{C_K + C_I} \quad (6)$$

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 (6) can be inverted to extract from the observed fluctuations the heat capacity

$$C \simeq C_K + C_I = \frac{C_K^2}{C_K - \sigma_K^2/T^2} \quad (7)$$

From eq.(7) we can see that when the heat capacity becomes negative σ_K overcomes the canonical expectation $\sigma_K^2/T^2 = C_K$. In our classical model Maxwell statistics implies a constant value for $C_K = 3/2 \cdot A$. 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 jumps from liquid to gas induce strong fluctuations in the energy partitioning. These anomalously large kinetic energy fluctuations are a signature of the convex intruder in the entropy surface and therefore of the first order phase transition. The normalized fluctuations σ_K^2/T^2 obtained in the microcanonical ensemble with a constrained volume are shown in the energy- λ plane in figure 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 figures 2 and 3 it is apparent that the phase transition signal is visible in the temperature as well as in the fluctuation observable.

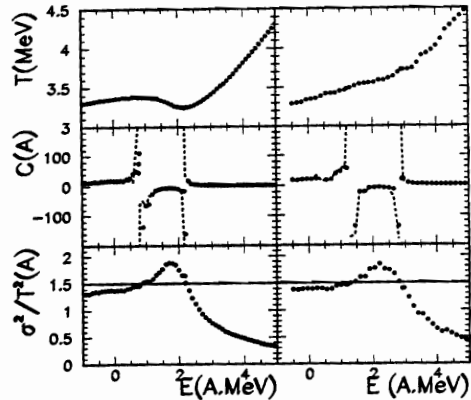


FIG. 4. Thermodynamical quantities in the microcanonical ensemble for a transformation at constant pressure (left part) and at constant volume (right part). Upper panels: caloric curve. Lower panels: normalized kinetic energy fluctuations compared to the canonical expectation (lines). Medium panels: heat capacity (symbols) compared to the estimation through eq.(7) (lines).

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 even if they can be measured. 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. The resulting caloric curve therefore depends on the actual transformation in the thermodynam-

ical 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 are displayed in the upper part of figure 4. At constant pressure the caloric curves are steeper than the ones at constant λ presented in figure 2 when the system is in the liquid or in the vapor phase; in the coexistence region the isobars are almost identical to the iso- λ 's since P 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 volume a smooth behavior is observed with a slope change entering the gas phase, as expected from general thermodynamics (see also [8,11]). This is due to the fact that the λ parameter varies rapidly in the coexistence region (see fig.1). From these examples one clearly sees that the various transformations leads to very different caloric curves. More generally, it is clear that the backbending of the temperature surface can 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.

On the other side partial energy fluctuations are a state variable which does not depend on the transformation from one state to another and can directly give access to the equation of state. From figure 3 we can see that in the whole phase transition region the microcanonical fluctuations present a strong maximum which exceeds the canonical value: an anomalously large fluctuation signal will be always seen if the system undergoes a first order phase transition, independent of the path. As an example the lower part of figure 4 shows a constant P or $\langle V \rangle$ cut of the bidimensional fluctuation surface shown in figure 3. The quantitative behavior of the heat capacity as a function of energy depends on the specific transformation, but at each point the heat capacity extracted from fluctuations is a direct measure of the underlying equation of state. This is clearly demonstrated in the medium part of figure 4 where the symbols, which represent the heat capacity C_λ extracted from the equation of state eq.(5), are in very good agreement with the lines which correspond to the fluctuation estimation eq.(7).

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 only if 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 state variable plane. In an experimental situation this transformation cannot be controlled and is even hardly defined. Conversely, we have shown that partial energy

fluctuations are a direct measure of the heat capacities. This observable depends on the considered state and is a direct measure of the EOS. Considering a statistical ensemble of states characterized by total energy and an average volume we have shown that abnormal fluctuations are a robust 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 [14].

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 excitation energy can be measured on an event-by-event basis; 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 constraining box implies that this variable can be known at best in average. 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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- [1] D. H. E. Gross and P.A.Hervieux, *Z.Phys.* D35 (1995) 27; D.H.E. Gross, *Phys. Rep.* 279 (1997) 119; D.H.E. Gross and E.Votyakov, *cond-mat* 9911257.
 - [2] J. Pochodzalla et al., *Phys.Rev.Lett.* 75 (1995) 1040; Y. G. Ma et al, *Phys.Lett. B* 390 (1997) 41; M.D'Agostino et al., *Nucl. Phys. A*650 (1999) 329.
 - [3] P.Chomaz and F.Gulminelli, *Nucl. Phys. A*647, 153 (1999).
 - [4] F.Gulminelli, P.Chomaz and V.Duflot, preprint LPCC 99-17 and submitted to *Europhys.Lett.*
 - [5] M. D'Agostino et al., *nucl-ex/9906004* and *Phys.Lett.B*, in press.
 - [6] F.Gulminelli and P.Chomaz, *Phys. Rev. Lett.* 82 (1999) 1402.
 - [7] C.N. Yang and T.D. Lee, *Phys.Rev.* 87 (1952) 410.
 - [8] X. Campi and H. Krivine, *Nucl. Phys. A* 620 (1997) 46 and *Physica A*, in press.
 - [9] J. Pan and S. Das Gupta, *Phys.Rev. C* 51 (1995) 1384; *Phys.Rev.Lett.* 80 (1998)1182.
 - [10] J.M.Carmona, J.Richert, A.Tarancon, *Nucl. Phys. A* 643 (1998) 115.
 - [11] J.M.Carmona, N.Michel, J.Richert, P.Wagner, *nucl-th/9910061*.
 - [12] V.Duflot, Ph.Chomaz and F.Gulminelli, preprint GANIL P 99-29 and *Phys.Lett.B*, in press.
 - [13] F.Gulminelli and Ph.Chomaz, *Int. Journ. Mod. Phys. E* vol.8, n.6 (1999) 1.
 - [14] D.Lynden-Bell, *Proc. of the XXth IUPAP Int.Conf on Stat. Phys., Paris, July 20-24 (1998)* and *cond-mat/9812172*.