Negative heat capacity in the critical region of nuclear fragmentation: an experimental evidence of the liquid-gas phase transition

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An experimental indication of negative heat capacity in excited nuclear systems is inferred from the event by event study of energy fluctuations in Au quasi-projectile sources formed in Au + Au collisions at 35 A.MeV. The excited source configuration is reconstructed through a calorimetric analysis of its de-excitation products. Fragment partitions show signs of a critical behavior at about 5 A.MeV excitation energy. In the same energy range the heat capacity shows a negative branch providing a direct evidence of a first order liquid gas phase transition.

Phase transitions are the prototype of a complex system behavior which goes beyond the simple sum of individual properties. In macroscopic systems the thermostatical potential presents non analytical behaviors which unambiguously marks a phase transition. Non analytical behaviors of infinite systems originate from anomalies of the thermostatical potentials in finite systems. Specifically in microcanonical finite systems, the entropy is known to present a convex intruder in 1-st order phase transitions associated to a negative heat capacity. Non statistical potential presents non analytical behaviors which unambiguously marks a phase transition. Non statistical potential presents non analytical behaviors which unambiguously marks a phase transition.

A 2-nd order phase transition is characterized by a symmetry breaking. In this case, since the volume is directly related to the liquid-gas case, since the volume is directly related to the temperature it should be considered as another order parameter. The fluctuations around a maximum $E_t$ of the total level density $W_t$ is the folding product of the two partial level densities $W_{t,i}$.

$$C_t^{-1} = -T^2 \partial^2 S_t / \partial E_t^2$$

where $C_t^{-1} = C_1^{-1} + C_2^{-1}$ are the heat capacities calculated for the most probable energy partition $(E_1, E_t - E_1)$.

The fluctuations around a maximum $E_t$ of the total level density $W_t$ is the folding product of the two partial level densities $W_{t,i}$.

$$\sigma_t^{-2} = (C_1^{-1} + C_2^{-1}) T^{-2}$$

In the absence of a phase transition because of the microcanonical constraint one expects the fluctuations $\sigma_t^2$ to be smaller than the canonical expectation $C_t T^2$ and the total heat capacity to be positive. Phase transitions are signed by poles and negative heat capacities corresponding to anomalously large fluctuations $C_t^{-1} \geq C_t T^2$.

A word of caution is here necessary. Convex intruders are expected for phase transitions of systems characterized by a unique extensive observable e.g. the total energy, as for example in the case of the melting transition which is characterized by a symmetry breaking. In the liquid-gas case, since the volume is directly related to the order parameter it should be considered as another...
extensive variable. In the experimental situation, since the break up volume is at best known only in average, the pressure, interpreted as the Lagrange multiplier associated with the volume observable, appears to be the relevant state variable together with the total deposited energy. In such a case the energy fluctuations are related to $C_p$ and a convex intruder is expected. In principle one could argue that the same information on the heat capacity can be obtained by taking the derivative of the correlation between the temperature and the excitation energy (the so called caloric curve). We want to stress that fluctuations are a characteristic of the state and so depend on the pertinent state variable while the caloric curve $T(E)$ depends upon the specific thermodynamical transformation from one state to another. Therefore, the information obtained by taking the derivative of the measured caloric curve may differ from the information coming from the fluctuations.

Here we report on a study on the de-excitation properties of $Au$ quasi-projectiles formed within an excitation energy range from 1 to 8 A.MeV. The experiment was performed at the K1200-NSCL Cyclotron of the Michigan State University. Beams of $Au$ ions at 35 A.MeV incident energy were used to bombard $Au$ foils. The MUL-TICS and MINIBALL arrays were coupled to measure light charged particles and fragments with a geometric acceptance greater than 87% of $4\pi$. For experimental details see ref. [9]. Peripheral collisions of a predominantly binary character have been selected by requiring the velocity of the largest fragment in each event to be at least 75% of the beam velocity. For each event, the fragments were considered as originating from the quasi-projectile if forward emitted in the centre of mass reference frame. The contribution of light particles is then added. To avoid pollution from other sources than the quasi-projectile, the backward emitted particles are substituted by the symmetric of the forward emission in the quasi-projectile reference frame. The total excitation energy of the source is measured from calorimetry on an event by event basis. This allows a sorting of the events as a function of energy as well as excitation energy per nucleon of the source $E^*/A_0$ (events falling into the selected area). One can recognize the critical distribution in the form of a power law $\tau - 2 = \beta/\gamma(1 + \beta/\gamma)$. Similar results were already found by the EOS collaboration [13].

Experimentally, information on critical exponents can be inferred from the analysis of the Campi scatter plot which gives the correlation between the heaviest fragment produced in each event and the corresponding second moment of the charge distribution $[11]$. This plot, presented in Fig. 2a) for our data, shows two branches characteristic of a subcritical regime and a supercritical one. The zone where the two branches join corresponds to the critical region. The critical exponents can be extracted from the mean values of the moments of the Campi plot, giving $\beta/\gamma = 0.29 \pm 0.01$. Fig. 2b) [2] shows the charge distribution in the critical region of the Campi plot (events falling into the selected area). One can recognize the critical distribution in the form of a power law of exponent $\tau = 2.13 \pm 0.08$ in agreement within the errors with the value of $\beta/\gamma$, according to the scaling relation $\tau - 2 = \beta/\gamma(1 + \beta/\gamma)$. Similar results were already found by the EOS collaboration [13].

A qualitative indication that the observed phase transition has a thermodynamical origin comes from the fact that, in the subcritical regime, an exponent $\beta = 0.33 \pm 0.04$ can be obtained by fitting with a power law the charge of the heaviest fragment as a function of the excitation energy per nucleon $E^*/A_0$ of the quasi-projectile (Fig.2c)), which means that $E^*/A_0$ can be considered as a critical parameter.
FIG. 2. (color) a) Campi scatter plot: Logarithm of the charge of the largest fragment in each event ($Z_{\text{big}}$) as a function of the normalized second moment of the charge distribution. c) Logarithm of $Z_{\text{big}}$ as a function of the excitation energy of the source. b) Charge distribution for the critical region of the Campi scatter plot (dashed contours of panels a) and c)). The solid symbols represent the mean correlation. The full lines are fits resulting from power-law behaviors.

The extracted values of the critical exponents are perfectly compatible with a liquid-gas transition but also with a geometrical percolation type of transition. Moreover, as found in the Lattice Gas Model [14] for finite systems, a critical behavior in fragment observables can also be consistent with a phase coexistence of a 1-st order phase transition. Indeed 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 with approximately the same critical exponents as at the critical point. In other words the analysis of the charge distribution in terms of critical behavior and critical exponents is a sign of a phase transition but does not allow to determine the corresponding universality class nor the associated order. Therefore, a more direct information about the order of the transition is mandatory.

To further progress on this point we shall now examine the partial energy fluctuations which are a direct observable to explicitly test thermodynamical equilibrium and extract thermodynamical state variables from the experimental data. In order to exploit eq. (3), one has to find a suitable decomposition of the total energy $E_i$ into $E_1$ and $E_2$. In the case of nuclear fragmentation data, the partition of the total energy is complicated by the fact that thermal equilibrium fragments are produced hot, while the detected fragments are collected at infinity after secondary de-excitation, i.e. with lower mass. Moreover, because of the presence of the long range Coulomb interaction, asymptotic kinetic energies have to be corrected from the Coulomb boost. To take into account these distortions, primary partitions have been reconstructed by applying the energy balance at the production time event by event:

$$m_0 + E^* = E_1 + E_2$$

where $m_0$ is the mass of the source, $E^*$ the excitation energy calculated via calorimetry, and the energy $E_2$ can be chosen as:

$$E_2 = \sum_{i=1}^{M} m_i + E_{\text{coul}}$$

Here $m_i$ is the mass excess of primary fragment $i$, $M$ is the event multiplicity at the production time, and $E_{\text{coul}}$ is the Coulomb energy of the partition.

FIG. 3. (color) a),c) Partial energy per nucleon $E_1/A_0$ as a function of the excitation energy $E^*/A_0$. The full line gives the average values $\langle E_1/A_0 \rangle$. b), d) Normalized variance of $E_1/A_0$ as a function of $E^*/A_0$. Left (right) panels refer to the freeze-out hypotheses I (II) (see text).

The primary masses have been obtained by sharing the charge detected in the form of light particles and neutrons among the detected fragments, following two extreme freeze-out hypotheses [2,7]. In the first assumption (I) primary fragments ($Z \geq 3$) have a charge-to-mass ratio as in the entrance channel, so that they de-excite only through neutron evaporation and all the light charged particles are considered as primary. In the second hypothesis (II) the totality of both neutrons and light charged particles is shared among final fragments. The experimental correlation between the partial energy $E_1/A_0$ and the excitation energy per nucleon of the source $E^*/A_0$ is shown in Fig.3a), c) for the two freeze-out hypotheses. The normalized variance of $E_1/A_0$, in bins of $E^*/A_0$, is represented in Fig.3b), d). The two
freeze-out hypotheses are in qualitative agreement, and the presence of peaks of Fig.3 b, d) indicate a phase transition governed by the equilibrium between the kinetic and potential degrees of freedom, i.e., a thermodynamical phase transition, in both cases. Finally, the actual microcanonical temperature $T$ can be obtained from $\langle E_1 \rangle$ by inverting the kinetic equation of state

$$\langle E_1 \rangle = \left( \sum_{i=1}^{M} a_i \right) T^2 + \left( \frac{3}{2} (M - 1) \right) T \tag{5}$$

where $a_i$ is the fragment dependent level density parameter. The brackets $\langle \cdot \rangle$ indicate the average on the events with the same $E^*$. This also allows the extraction of the kinetic heat capacity $C_1$ obtained by taking the numerical derivative of $\langle E_1 \rangle$ with respect to $T$.

It is important to remark that the thermodynamical transition energy interval corresponds to the critical region of the Campi scatter plot: the observation of power laws and critical behaviors in the fragment charge distribution is directly correlated to the occurrence of anomalous fluctuations indicating a negative heat capacity. These two pieces of informations which correspond to the theoretical expectations indicate that a thermodynamical first order phase transition has taken place in finite nuclear systems formed in nuclear reactions.

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