

FRAGILE SIGNS OF CRITICALITY IN THE NUCLEAR MULTIFRAGMENTATION

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(Received March 22, 2002)

Deviations from an idealized equilibrium phase transition picture in nuclear multifragmentation is studied in terms of the entropic index. We investigate different heat-capacity features in the canonical quantum statistical fragmentation model generalized in the framework of non-extensive thermostatics, and show that in this model the negative branch of heat capacity in quasi-peripheral Au+Au reactions is consistent with the dominance of non-extensive effects in these reactions.

PACS numbers: 25.70.Pq, 05.20.-y, 05.70.Jk, 24.60.Ky

1. Introduction

The nuclear multifragmentation process is studied in the energetic collisions of Heavy Ions (HI). In these collisions, strongly off-equilibrium transient system is formed which equilibrates at the later stage of the reaction due to dissipative processes. Perfectly equilibrated system, as assumed in most theoretical descriptions of the multifragmentation decay of the hot residue, is most probably never attained. Moreover, standard equilibrium thermodynamics is valid only for sufficiently short-ranged interactions, what is not the case for Coulomb field. This would not be a serious problem if the nuclear fragmentation process does not show any sign of the criticality [1, 2]. Indeed, the nonextensivity of weakly off-equilibrium finite systems may qualitatively modify both the picture of the two phase coexistence and signatures of the critical behavior in small systems [3]. On the other hand, these nonextensivity corrections to Boltzmann–Gibbs Statistical Mechanics (BGSM) have no measurable effects on standard signatures

of the equilibrium such as the particle/fragment kinetic energy spectra or angular distributions [3]. Neither the caloric curve nor the negative heat-capacity branch measurements [1, 2], both put forward as an evidence for the nuclear liquid-gas phase transition, can be interpreted unambiguously. The non-extensive effects due to the Coulomb interaction/non-Markovian memory effects or the multi-fractal phase boundary conditions [4, 5], which are crucial *only* in the critical region [3], not only constitute an integral part of the physics of HI collisions but also provide an essential limitation to our understanding of the multifragmentation process in the BGSM framework.

Non-extensive features can be studied in the molecular dynamics framework. The effect of equilibration in a confining wall has been investigated recently in excited Lennard–Jones systems [6]. In the system of classical spins with infinite range interaction [7], it has been shown that before going to the Boltzmann–Gibbs equilibrium state, the system reaches the metastable state with non-Maxwellian velocity distribution whose lifetime increases indefinitely with increasing number of spins. Hence, in the thermodynamical limit the system may be indefinitely trapped in a non-Boltzmann–Gibbs state.

Analytical studies of the non-extensive spin model with infinite-range ferromagnetic interaction and repulsive correlations [8] showed clearly the development of a new weak-ferromagnetic phase and an unusual first order phase transition from paramagnetic to weak-ferromagnetic phase in which a discontinuity of the averaged order parameter appears even for finite number of spins.

All these intriguing results obtained in schematic models, confirm that the applicability of standard equilibrium ensembles for the description of the dynamically induced fragmentation process in the presence of long range interactions/correlations requires further studies.

In the case of nuclear multifragmentation, a practical and sufficiently realistic approach to this problem would be to use those (extensive) models which describe physics of limiting two phases outside of the critical region, and then to develop a flexible phenomenological parametrization of non-generic non-extensive effects in the critical region. It is the aim of this work to illustrate this problematic in the thermodynamic canonical model which is extended in the framework of the generalized thermostatistics [3].

2. The non-extensive canonical quantum statistical model of nuclear multifragmentation

A starting point could be any reasonable thermodynamic fragmentation model (for the list of examples see *e.g.* [9, 10]). This choice offers several advantages, such as a correct quantum statistics and a correct definition of

both nuclear fragments and their binding energies. The Coulomb and surface effects can be tuned by analyzing the observable quantities far outside of the critical region and fragment excitations can be included, if necessary. Several models of this kind had an unquestionable success in describing reaction products and their properties from the regime of particle evaporation at low excitation energies to the explosion at about 5–10 MeV/nucleon [9, 10]. The new class of non-extensive thermodynamical models can be formulated in the framework of the Tsallis Generalized Statistical Mechanics (TGSM) [4]. TGSM is based on an alternative definition for the entropy of a system whose i -th microscopic state has probability \hat{p}_i

$$S_q = k \frac{1 - \sum_i \hat{p}_i^q}{q - 1} = k \frac{\sum_i \hat{p}_i - \sum_i \hat{p}_i^q}{q - 1}, \quad k > 0, \quad (1)$$

where k is the conventional positive constant. The entropic index q defines statistics and the normalization condition

$$\sum_i \hat{p}_i = 1 \quad (2)$$

is used to get the second equality in (1). The limit $q = 1$ corresponds to the BGSM. The main difference between BGSM and TGSM is the non-additivity of entropy in the TGSM. For two independent subsystems A , B , *i.e.* such that the joint probability of $A + B$ is factorized into $\hat{p}_{A+B} = \hat{p}_A \hat{p}_B$, the global entropy

$$S_q(A + B) = S_q(A) + S_q(B) + \frac{(1 - q)S_q(A)S_q(B)}{k} \quad (3)$$

is not equal to the sum of the subsystem entropies. In particular, the entropy is always concave for $q > 0$, and this is the case discussed in the present paper. Different works have shown that the above described statistical mechanics retains much of the formal structure of the standard theory [11]. Many important properties have been shown to be q -invariant. Among them, we have the Legendre transformation structure of thermodynamics and the H-theorem (macroscopic time irreversibility). Considering that the essence of the second law of thermodynamics is the concavity (see *e.g.* Ref. [12] and references quoted therein), the mentioned properties of S_q indicate that there are no problems with this law in TGSM.

The entropy (1) was discussed recently in terms of the incomplete information theory [13]. The condition (2) means that all possible physical states are both well-known and counted. However, in complex systems we often do not know all interactions and/or cannot find the exact Hamiltonian and,

therefore, exact values of physical quantities are not accessible. In other words, part of the information is lost and the normalization (2) is violated because the set of the countable states becomes incomplete. By changing probabilities in Eq. (2) into effective ones $\hat{p}_i \rightarrow \hat{p}_i^q$, one arrives again at Tsallis definition of the entropy. The parameter $q_1 \equiv q - 1$ is related in this formulation to an extra entropy due to the neglected interactions. This is the essential reason for introducing TGSM with a phenomenologically adjusted parameter q . The situation encountered experimentally in the nuclear multifragmentation process, where effects of thermal and chemical non-equilibrium, expansion of decaying system, long-ranged Coulomb interaction, various memory effects of system dynamical history, or effects of complicated interphase boundary are present, can be better addressed in the framework of generalized statistical mechanics where the entropic index parameter allows to correct the BGSM framework for missing physically important effects.

The TGSM is relevant if the microscopic interactions in the system are long-ranged and/or the effective microscopic memory is long-ranged and/or the geometry of the system is fractal [4,5,14]. In the *super-additive regime* ($1 - q > 0$), independent subsystems A and B will tend to join together increasing in this way the entropy of the whole system. On the contrary, in the *sub-additive regime* ($1 - q < 0$), the system increases its entropy by fragmenting into the separate subsystems [8]. This is a natural regime for non-extensive systems with long-ranged repulsive interactions, such as formed in the collisions of atomic nuclei or atomic clusters. These ideas agree with the results of Landsberg *et al.* [15] who studied fragmentation process in connection with the thermodynamics of black holes.

Our further considerations are based on the canonical multifragmentation model [16] which is a generalization of phenomenologically successful statistical multifragmentation model of Bondorf *et al.* [10]. The canonical ensemble method in TGSM was introduced in [17]. In this case, the entropy S_q is extremized with the conditions (2) and

$$\sum_{i=1} p_i^q \varepsilon_i = U_q,$$

where ε_i is the energy of the i -th microscopic state and U_q is the generalized average energy. The main ingredient of the non-extensive canonical quantum statistical model of nuclear multifragmentation [3] is the expression for the fragment partition function

$$\omega_q(a, z) = \sum_{\vec{p}} [1 + q_1 \beta \varepsilon_{\vec{p}}(a, z)]^{-1/q_1}, \quad (4)$$

where a and z are the fragment mass number and the fragment charge number, respectively. The fragment partition probability equals

$$\hat{p}_{\vec{p}}(a, z) = [\omega_q(a, z)]^{-1} [1 + q_1 \beta \varepsilon_{\vec{p}}(a, z)]^{-1/q_1}, \quad (5)$$

where $\varepsilon_{\vec{p}}(a, z) = p^2/2M + U(a, z)$ and $\beta \equiv 1/T$. In the limit $q_1 \rightarrow 0$, Eq. (5) recovers the familiar expression $\hat{p}_{\vec{p}}(a, z) = \exp(-\beta \varepsilon_{\vec{p}}(a, z))/\omega_1(a, z)$. The internal energy U , which includes the fragment binding energy and the fragment excitation energy, the temperature-dependent surface energy, and the Coulomb interaction between fragments in the Wigner–Seitz approximation, is parameterized as in [10]. In the dilute gas approximation [18], the partition function of a whole system can be written as

$$\mathcal{Q}_q(A, Z) = \sum_{\hat{n} \in \Pi_{A,Z}} \prod_{a,z} \frac{[\omega_q(a, z)]^{N_{\hat{n}}(a,z)}}{N_{\hat{n}}(a, z)!}, \quad (6)$$

where the sum runs over the ensemble $\Pi_{A,Z}$ of different partitions of A and Z of the decaying system $\{\hat{n}\} = \{N_{\hat{n}}(1, 0), N_{\hat{n}}(1, 1), \dots, N_{\hat{n}}(A, Z)\}$ and $N_{\hat{n}}(a, z)$ is the number of fragments (a, z) in the partition $\{\hat{n}\}$. In this approximation, the recurrence relation technique [16,19] can be applied providing exact expression for $\mathcal{Q}_q(A, Z)$ [3].

Given the partition function, the mean value of any quantity is [4]

$$\langle \mathcal{O} \rangle_q = \sum_{\vec{p}} \mathcal{O}_{\vec{p}} \hat{p}_{\vec{p}}^q. \quad (7)$$

In order to ensure the proper normalization of q -averages (7), it is better to work with the generalized averages [17]

$$\langle\langle \mathcal{O} \rangle\rangle_q = \frac{\langle \mathcal{O} \rangle_q}{\langle 1 \rangle_q}. \quad (8)$$

These normalized mean values exhibit all convenient properties of the original mean values. Moreover, the TGSM can be reformulated in terms of ordinary linear mean values calculated for the renormalized entropic index $q^* = 1 + (q - 1)/q$. In particular, the total average energy and pressure of the system become

$$\mathcal{E}_q = \sum_{a,z} \langle N(a, z) \rangle_{q^*AZ} \langle \varepsilon(a, z) \rangle_{q^*}, \quad (9)$$

$$P_q = \sum_{a,z} \langle N(a, z) \rangle_{q^*AZ} \langle p(a, z) \rangle_{q^*}, \quad (10)$$

where $\langle \varepsilon(a, z) \rangle_q$ and $\langle p(a, z) \rangle_q$ are given by

$$\langle \varepsilon(a, z) \rangle_q = -\frac{\partial}{\partial \beta} \left(\frac{1 - [\omega_q(a, z)]^{-q_1}}{q_1} \right), \tag{11}$$

$$\langle p(a, z) \rangle_q = \frac{1}{\beta} \frac{\partial}{\partial V_f} \left(\frac{1 - [\omega_q(a, z)]^{-q_1}}{q_1} \right), \tag{12}$$

and the average multiplicity of (a, z) -fragments in the fragmentation of system (A, Z) is

$$\langle N(a, z) \rangle_{qAZ} = \omega_q(a, z) \frac{\mathcal{Q}_q(A - a, Z - z)}{\mathcal{Q}_q(A, Z)}. \tag{13}$$

The heat capacity at a constant volume ($= \partial \mathcal{E}_q / \partial T |_{V_f}$) is:

$$\begin{aligned} C_V = & \beta^2 \left\{ \sum_{a,z} \sum_{a',z'} \langle \Delta(az; a'z') \rangle_{q^*} \langle \varepsilon(a, z) \rangle_{q^*} \langle \varepsilon(a', z') \rangle_{q^*} \right. \\ & \left. + \sum_{a,z} \langle N(a, z) \rangle_{q^*AZ} \left[\langle \varepsilon^2(a, z) \rangle_{q^*} - \langle \varepsilon(a, z) \rangle_{q^*}^2 \right] \right\}, \tag{14} \end{aligned}$$

where

$$\langle \Delta(az; a'z') \rangle_q \equiv \langle N(a, z)N(a', z') \rangle_{qAZ} - \langle N(a, z) \rangle_{qAZ} \langle N(a', z') \rangle_{qAZ}, \tag{15}$$

and

$$\begin{aligned} & \langle N(a, z)N(a', z') \rangle_{qAZ} \\ & = \omega_q(a, z)\omega_q(a', z') \frac{\mathcal{Q}_q(A - a - a', Z - z - z')}{\mathcal{Q}_q(A, Z)} \\ & \quad + \delta_{aa'}\delta_{zz'}\omega_q(a, z) \frac{\mathcal{Q}_q(A - a, Z - z)}{\mathcal{Q}_q(A, Z)}. \tag{16} \end{aligned}$$

The heat capacity at a constant pressure C_P ($= \partial(\mathcal{E}_q + \mathcal{P}_q V_f) / \partial T |_{\mathcal{P}_q}$) can be calculated using the relation $C_P - C_V = TV_f \kappa_T (\partial \mathcal{P}_q / \partial T |_{V_f})^2$, where κ_T stands for the isothermal compressibility ($= -(1/V_f) \partial V_f / \partial \mathcal{P}_q |_T$)

$$\begin{aligned} \frac{1}{\kappa_T} = & -\beta V_f \left[\sum_{a,z} \sum_{a',z'} \langle \Delta(az; a'z') \rangle_{q^*} \langle p(a, z) \rangle_{q^*} \langle p(a', z') \rangle_{q^*} \right. \\ & \left. + \sum_{a,z} \langle N(a, z) \rangle_{q^*AZ} \frac{1}{\beta} \left(\frac{\partial \langle p(a, z) \rangle_{q^*}}{\partial V_f} \Big|_T \right) \right] \tag{17} \end{aligned}$$

and

$$\begin{aligned} \frac{\partial \mathcal{P}_q}{\partial T} \Big|_{V_f} = & \beta^2 \left[\sum_{a,z} \sum_{a',z'} \langle \Delta(a,z; a',z') \rangle_{q^*} \langle p(a,z) \rangle_{q^*} \langle \varepsilon(a',z') \rangle_{q^*} \right. \\ & \left. + \sum_{a,z} \langle N(a,z) \rangle_{q^*} AZ \frac{1}{\beta^2} \left(\frac{\partial \langle p(a,z) \rangle_{q^*}}{\partial T} \Big|_{V_f} \right) \right]. \end{aligned} \quad (18)$$

One should stress that all these thermodynamical quantities are calculated *exactly*, without using the Monte Carlo technique.

3. Discussion of the results

The upper part in Fig. 1 shows the temperature dependence of the pressure for various entropic indices q in systems with $A_0 = 100, 200$ and 300 nucleons and $Z_0 = 0.4A_0$ protons. All numerical results shown in this work

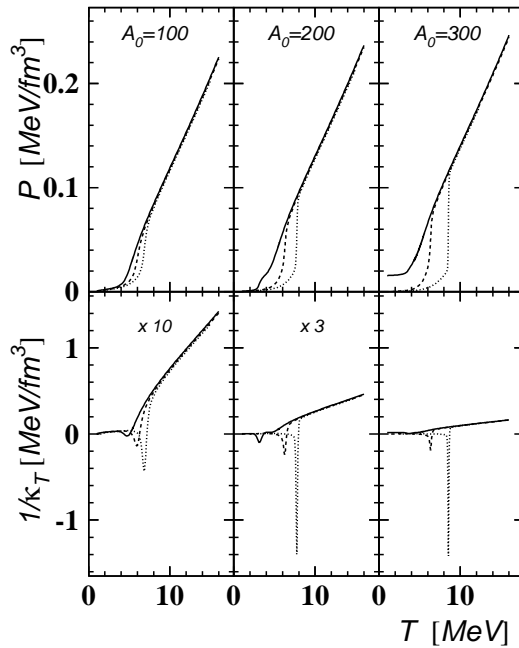


Fig. 1. The dependence of the pressure (upper part) and the inverse isothermal compressibility (lower part) on the temperature T plotted for system of different sizes and different entropic indices q : 1.0 (solid line), 1.0005 (dashed line), 1.001 (dotted line). The freeze-out volume V_f corresponds to $\rho_f \equiv A_0/V_f = \rho_0/4$. The calculated values of $1/k_T$ are multiplied by factors 10 and 3 for $A_0=100$ and 200 , respectively.

have been obtained for the density $\rho_f \equiv A_0/V_f = \rho_0/4$. In the bottom part, the temperature dependence of the inverse thermal compressibility $1/\kappa_T$ is shown. Zero of $1/\kappa_T$ ($\partial\mathcal{P}_q/\partial V_f|_{T=0}$) corresponds to the pole of C_P and defines the boundary of the two phase coexistence region. Negative heat capacity C_P within the canonical ensemble provide a signal of the first-order phase transition. This is a counterpart of negative micro-canonical heat capacity in a certain energy range [20]. For $q > 1$, there exists a region of temperatures where $1/\kappa_T$ is negative and, hence, C_P becomes negative between the poles. In the BGSM limit, $1/\kappa_T$ has zeros for $A_0 = 100, 200$, whereas in heavier systems these zeros appear only in the subadditive regime $q > 1$, *i.e.* in the typical situation of fragmentating residue with long-ranged interactions [8,15]

An essential part of the pressure and, hence, of $1/\kappa_T$ is the Coulomb term. The inverse compressibility $1/\kappa_T$ never vanishes when the Coulomb term is neglected [16]. Since the Coulomb contribution to the pressure and the inverse thermal compressibility decreases in the Wigner–Seitz approximation roughly as $A_0^{-1/3}$, this particular signature may not be seen in heavy systems in the BGSM limit. Existing data do not allow yet to pin down the A_0 -dependence of the criticality signatures. Nevertheless, Fig. 1 demonstrates how fragile is the Boltzmann–Gibbs equilibrium critical behavior in finite systems. Small increase of q above the BGSM limit leads to an upwards shift of the critical temperature T_c which, for the same value of q , is higher in heavier systems. All these important changes take place in a narrow range of temperatures around T_c , beyond which the fragmenting system closely follows the BGSM limit.

Fig. 2 present the heat capacities C_V and C_P as a function of the excitation energy $E^* = \mathcal{E}_q(T, V_f) - \mathcal{E}_q(T = 0, V_0)$, where V_f is the freeze-out volume, $V_0 = A_0/\rho_0$ and ρ_0 is the equilibrium density at $T = 0$. C_V is a smooth positive function of the excitation energy for all values of q . The peak of $C_V(E^*)$, whose position is associated with the critical temperature T_c , becomes more pronounced for higher q . Fig. 3 compares the heat capacity C_P versus E^*/A_0 for systems of different sizes $A_0 = 200$ and $A_0 = 300$. These results can be compared with those for system $A_0 = 100$ shown in the bottom of Fig. 2. In the BGSM limit, the negative branch of C_P is seen only for $A_0 \lesssim 200$. With increasing A_0 , its position moves towards lower excitation energies.

It should be noted that the critical density ρ_c in the non-extensive fragmentation model [3] is relatively high. The global critical point (ρ_c, T_c, P_c) for $A_0 = 100$ corresponds to $\rho_c/\rho_0 = 0.547, 0.783$ and 0.925 for $q = 1, 1.0005$ and $q = 1.001$, respectively. This tendency is a direct consequence of subadditivity of the entropy which increases the instability of the system and extends the domain of multifragmentation instability towards higher den-

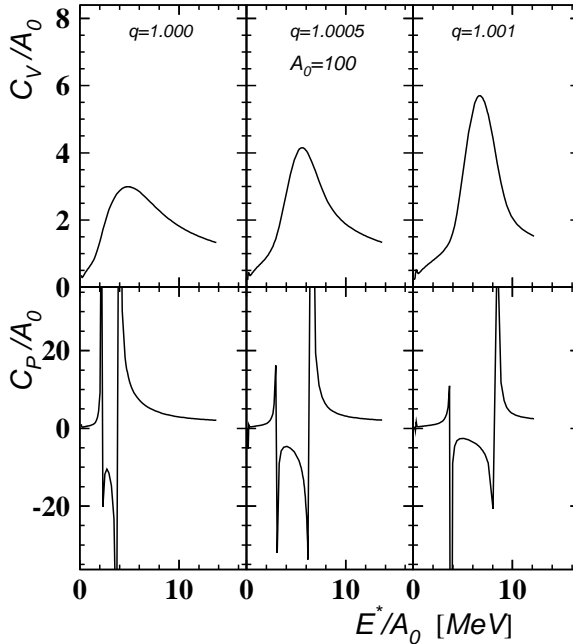


Fig. 2. The specific heat at a constant volume C_V (upper part) and at a constant pressure C_P (lower part) are plotted *versus* the excitation energy per nucleon for various entropic indices q in the system with $A_0 = 100$ and $Z_0 = 40$. The freeze-out volume V_f corresponds to $\rho_f \equiv A_0/V_f = \rho_0/4$.

sities. One should be aware that for densities higher than $\sim \rho_0/2$, the Wigner–Seitz approximation becomes less accurate and there is a need for devising a better approximation [10, 21]. In any case, all results shown in Figs. 1–3 are obtained in the safe region of low densities. For $A_0 = 200$, the global critical point exists only for $q = 1$ whose value of $\rho_c/\rho_0 = 0.904$ is close to that obtained in statistical multifragmentation model using the recurrence relation technique [22].

The description of nuclear matter in terms of the Van der Waals fluid [22] (see also [23]) yields much lower critical densities ($\rho_c \approx 0.3\rho_0$). In this model, the boundary of the coexistence region on the diagram $P(\rho, T)$ has a bell-like shape and the line $\rho = V\rho_f$ crosses it in a single point. Consequently, the negative branch of heat capacity is not seen. In the non-extensive fragmentation model [3], the boundary of the coexistence region is skew with the top tilted towards higher ρ what allows for two crossings with the line $\rho = \rho_f$ and leads to the negative branch of C_P .

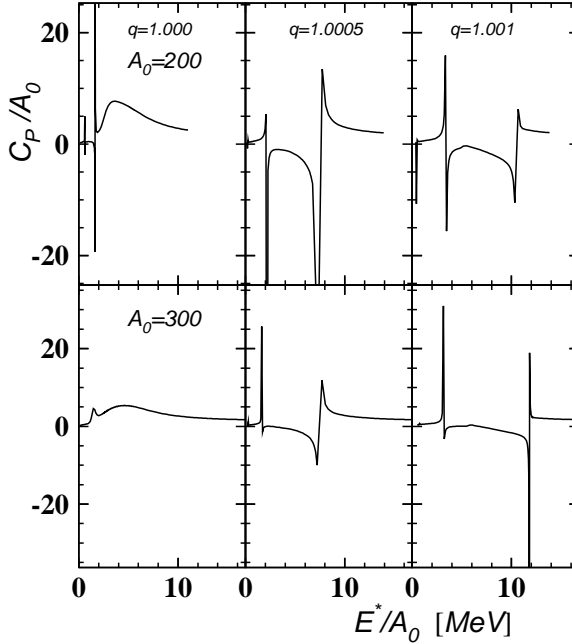


Fig. 3. The specific heat at a constant pressure C_P is plotted *versus* the excitation energy per nucleon for various entropic indices q in the systems with $A_0 = 200$, $Z_0 = 80$ (upper part) and $A_0 = 300$, $Z_0 = 120$ (lower part). The freeze-out volume V_f corresponds to $\rho_f \equiv A_0/V_f = \rho_0/4$.

4. Conclusions

The phase transition in the statistical nuclear multifragmentation models tends to disappear in heavy systems due to the weakening of the Coulomb contribution. This effect can be compensated by the non-extensive features of entropy due to either long-range correlations/memory effects or the fractality of the liquid-gas interphase, which both tend to strengthen signatures of the first order phase transition. The application of non-extensive canonical statistical fragmentation model [3] for the understanding of experimental caloric curve [1] and negative heat capacity [24] in the critical region, consistently indicates deviation from the BGSM picture of the phase transition and $q \gtrsim 1.0005$. This tiny variation of q , which cannot be detected either in the particle/fragment kinetic energy distributions or in the angular distributions, have strong measurable effects on the event-by-event energy fluctuations of particles/fragments in the region of phase coexistence. Hence, the mass-dependence of the criticality signatures is determined by a subtle conspiracy between the Coulomb contribution to the pressure and the non-extensive features of the entropy.

For $q > 1$, the negative branch of C_P is seen both in light and heavy systems. The range of excitation energies corresponding to $C_P < 0$ increases with increasing A_0 . However, in heavy systems the negative branch of C_P appears *uniquely* for $q > 1$. Both extension and localization of the negative branch of C_P in quasi-peripheral Au+Au collisions at 35A.MeV [24], closely resemble results of non-extensive fragmentation model for $q \simeq 1.0005$ and $A_0 = 200$ (see Fig. 3). The position of singularity of C_P at higher excitation energies increases sensitively both with the entropic index q and with the source size. Hence, a successful description of this data for a maximal size of quasi-projectile source ($A_0 \lesssim 200$) [24], allows to find a lowest limit on the value of the entropic index and, hence, on the deviation from the BGSM limit, within the framework of the statistical multifragmentation model.

There are many sources of non-extensivity in mesoscopic systems. Some of them, *e.g.* the formation of liquid-gas (fractal) interphase [25], have been pointed out in the microcanonical studies [25,26]. Most of these effects are non-generic what provides a principal obstacle in the meaningful characterization of nuclear multifragmentation data in the critical region using an ideal picture of BGSM. In the framework of the non-extensive multifragmentation model [3], the entropic index $q \sim 1.0005$ seems to be consistent with both the caloric curve [1] and the negative heat capacity [24] data, in spite of completely different kinematical conditions in these measurements. Surprisingly, the excitation energy of higher singularity of C_P seems to be the same, both in quasi-peripheral Au+Au collisions at 35A MeV [24] and in central Xe+Sn collisions at 32A MeV [27] and agrees with $q \simeq 1.0005$. These results show a large utility of such simple and flexible non-extensive statistical multifragmentation models to correlate different experimental data not only far outside of the critical zone, but also in the critical region itself.

The work was supported by the IN2P3-JINR agreement no. 00-49.

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