Fragile Signs of Criticality in the Nuclear Multifragmentation

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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 model of nuclear multifragmentation generalized in the framework of Tsallis nonextensive thermostatistics. We find that the negative branch of heat capacity observed in quasi-peripheral Au+Au collisions is caused primarily by the non-generic nonextensivity effects.

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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. This would not be a serious problem if the nuclear fragmentation process does not show any sign of the ‘criticality’ [1]. 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 [4]. Neither the caloric curve nor the negative heat-capacity branch measurements [5], both put forward as an evidence for the nuclear liquid-gas phase transition, can be interpreted unambiguously. The nonextensive effects due to the long-range interaction/non-Markovian memory effects or the multifractal phase boundary conditions [6], which are crucial only in the ‘critical region’ and cannot be reliably estimated, not only constitute an integral part of the physics of HI collisions but also provide an essential limitation to the understanding of the multifragmentation based on the BGSM. The practical solution to this problem would be to use those models of excited nuclear matter which describe physics of limiting two phases outside of the ‘critical region’ and to parameterize the critical region in simple terms. It is the aim of this work to illustrate this problematic in the thermodynamic (canonical) model of the fragmentation which is extended to include the nonextensive effects in the framework of the generalized thermostatistics [3].

A reasonable starting point could be any ‘realistic’ thermodynamic fragmentation model (for the list of examples see [7]). This choice offers several advantages, such as correct quantum statistics, correct definition of fragment sizes and the fragment 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 [3]. The new class of nonextensive thermodynamical models, can be formulated in the framework of the Tsallis generalized statistical mechanics (TGS) [8]. TGS is based on an alternative definition for the equilibrium entropy of a system whose ith microscopic state has probability $\hat{p}_i$:

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

where $q$ is the entropic index and the normalization condition

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

is used to get the second equality in (1). The limit $q = 1$ corresponds to the BGSM. It is easy to verified that such general properties as non-negativity, concavity and so on are preserved by this new entropy definition. The main difference between BGSM and TGS is in the non-additivity of entropy in the TGS. Indeed, 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 in TGS:

$$S_q(A + B) = S_q(A) + S_q(B) + (1 - q)S_q(A)S_q(B)/k,$$

is not equal to the sum of the subsystem entropies.

The entropic index $q$ sometimes can be related in explicit way to other basic quantities such as internal energy or free energy of the system. Recently the Tsallis definition of entropy [9] was reinterpreted in terms of incomplete information theory [10]. The condition $\{\}$ means that all the possible physical states are well-known and...
counted. But for complicated systems we often in prac-
tice do not know all interactions, cannot find the exact
Hamiltonian, the exact solution of equation of motion
and the exact values of physical quantities. Therefore, a
part of information is lost and the normalization [1] is
violated because the set of the countable states becom-
ies incomplete. By redefining the real probability in Eq.(3)
to effective one as \( \hat{p} \rightarrow \hat{p}^q \), one can keep the Tsallis defini-
tion of the nonextensive canonical quantum statistical
BGSM.

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limit

Here the difference \( q_1 \equiv q - 1 \) is related to the extra
entropy of the system due to neglected interactions. In
this case the essential question is not the preferential use
of microcanonical ensemble (for which the entropy is ex-
tremized with keeping only the normalization restriction)
or canonical one (when an additional restriction for the
energy conservation in average is applied) but rather the
point which interactions are included effectively by in-
introducing a free parameter \( q \). The situation discussed
we meet in the multifragmentation where effects of ther-
mal and chemical non-equilibrium, slow expansion of de-
caying system as well as possible multifractal behavior
of interphase boundary are out of consideration in the
BGSM.

Our further consideration is based on the canonical
multifragmentation model [9]. The canonical ensemble
method in TGSM was introduced in [9]. The main ingre-
dient of the nonextensive quantum canonical statistical
model of nuclear multifragmentation [9] is the expression
for the fragment partition function:

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

where \( a \) and \( z \) are the fragment mass number and
the fragment charge number, respectively. The fragment pa-
tition probability equals:

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

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.(4) recovers the familiar expres-
sion: \( \hat{p}_{\vec{p}}(a, z) = \exp(-\beta \varepsilon_{\vec{p}}(a, z))/\omega_1(a, z) \). The internal
energy \( U \), which includes the fragment binding energy
and the fragment excitation energy, the tempera-
dependent surface energy, and the Coulomb interaction
between fragments in the Wigner-Seitz approximation,
is parameterized as in [9]. In the dilute gas approxima-
tion [9], the partition function of a whole system can be
written as:

\[
Q_q(A, Z) = \sum_{\{\hat{n}\}} \prod_{a,z} \frac{\omega_q(a, z)^{N(a, z)}}{N_{\hat{n}}(a, z)!}
\]  

where the sum runs over the ensemble \( \Pi_{A, Z} \) of different
partitions of \( A \) and \( Z \) of the decaying system : \( \{\hat{n}\} = \{N_{\hat{n}}(1, 0), N_{\hat{n}}(1, 1), \ldots, N_{\hat{n}}(A, Z)\} \) 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 [11] can be applied providing exact expression for \( Q_q(A, Z) \).

Given the partition function, the mean value of any
quantity is [10]:

\[
\langle \mathcal{O} \rangle_q = \sum_{\vec{p}} \mathcal{O}_{\vec{p}} \hat{p}^q \cdot (6)
\]

In order to ensure the proper normalization of
q–averages [10], it is better to work with the generalized
averages [10]:

\[
\langle \mathcal{O} \rangle_q = \langle \mathcal{O} \rangle_q / q > (7)
\]

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 in-
dex: \( q^* = 1 + (q - 1)/q \). In particular, the total average
energy and pressure of the system become:

\[
\mathcal{E}_q = \sum_{a,z} \langle N(a, z) \rangle_q > q^{*\times} \varepsilon(a, z) > q^* (8)
\]

\[
P_q = \sum_{a,z} \langle N(a, z) \rangle_q > q^{*\times} P(a, z) > q^* (9)
\]

where \( \varepsilon(a, z) > q \) and \( P(a, z) > q \) are given by:

\[
\langle \varepsilon(a, z) \rangle_q = \frac{\partial}{\partial \beta} \left( \frac{1 - [\omega_q(a, z)]^{q_1}}{q_1} \right) (10)
\]

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

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

\[
\langle N(a, z) \rangle_q AZ = \omega_q(a, z) Q_q(A - a, Z - z) \frac{Q_q(A, Z)}{Q_q(A, Z)} (12)
\]

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

\[
C_V = \beta^2 \left\{ \sum_{a,z} \langle \Delta(a; a', z') \rangle_q > q^* \varepsilon(a, z) > q^* \times
\right.
\]

\[
\left. \times \langle \varepsilon(a', z') \rangle_q > q^* + \sum_{a,z} \langle N(a, z) \rangle_q > q^{*\times} \times
\right.
\]

\[
\left. \times \left[ \langle \varepsilon^2(a, z) \rangle_q > q^* - \varepsilon(a, z) > q^* \right] \right\} (13)
\]

where:

\[
\langle \Delta(a; a', z') \rangle_q = \langle N(a, z) N(a', z') \rangle_q AZ - \langle N(a, z) \rangle_q AZ < N(a', z') \rangle_q AZ (14)
\]

and:

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above the BGSM limit leads to an upwards shift of the zeros appearing 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.

\[
< N(a,z)N(a',z') >_{q\text{AZ}} = \omega_q(a,z)\omega_q(a',z') \frac{Q_q(A - a - a', Z - z - z')}{Q_q(A, Z)} + \delta_{aa'}\delta_{zz'}\omega_q(a,z) \frac{Q_q(A - a, Z - z)}{Q_q(A, Z)}.
\]

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

\[
\frac{1}{\kappa_T} = -\beta V_f \left[ \sum_{a,z} \sum_{a',z'} < \Delta(a; a', z') >_q \times \right.
\times < p(a, z) >_q < p(a', z') >_q + \sum_{a,z} < N(a, z) >_q A_Z \frac{1}{\beta^2} \left( \frac{\partial < p(a, z) >_q}{\partial V_f} \mid_T \right)
\]

and:

\[
\frac{\partial P_q}{\partial T} \mid_{V_f} = \beta^2 \left[ \sum_{a,z} \sum_{a',z'} < \Delta(a; a', z') >_q \times \right.
\times < p(a, z) >_q < \varepsilon(a', z') >_q + \sum_{a,z} < N(a, z) >_q A_Z \frac{1}{\beta^2} \left( \frac{\partial < p(a, z) >_q}{\partial T} \mid_V \right).
\]

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

The upper part of 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. In the bottom part, the temperature dependence of the inverse thermal compressibility $1/\kappa_T$ is shown. Zero of $1/\kappa_T (\partial P_q/\partial V_f \mid_T = 0)$ corresponds to the pole of $C_P$ and defines the boundary of the two phase coexistence region. 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 for $q > 1$. 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. 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. 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. 1](http://example.com/fig1.png)

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

Fig. 2 present the heat capacities $C_V$ and $C_P$ as a function of the average excitation energy: $E^* = E_q(T, V_f) - E_q(T = 0, V_0)$, where $V_f$ is the freeze-out volume, $V_0 = A_0/\rho_0$ and $\rho_0$ 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 $C_P$ vs $E^*/A_0$ for systems of different sizes: $A_0 = 200$ and $A_0 = 300$. In the BGSM limit, the negative branch of $C_P$ is seen only for $A_0 \leq 200$. With increasing $A_0$, its position moves towards lower excitation energies. It should be noted that the critical density $\rho_c$ in the nonextensive fragmentation model is relatively high. The ‘global’ critical point $(V_c, T_c, \rho_c)$ 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. 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 \cite{13}. For \( q > 1 \), the system is always found inside of the two phase coexistence region for all \( \rho < \rho_0 \). This is consistent with the picture of nonextensivity driven first order phase transition.

![Graph showing specific heat at constant volume and pressure](image)

**FIG. 2.** The specific heat at a constant volume \( C_V \) (the upper part) and at a constant pressure \( C_P \) (the lower part) are plotted vs the excitation energy per nucleon for various entropic indices \( q \) in the system with \( A_0 = 100 \) and \( Z_0 = 40 \).

The description of nuclear matter in terms of the Van der Waals fluid \cite{12} (see also \cite{13}) yields much lower critical densities \( (\rho_c \approx 0.3\rho_0) \). In this model, the boundary of the coexistence region has a bell-like shape and the line \( V = V_f \) crosses it in a single point. Consequently, the nonextensive branch of heat capacity is not seen. In the nonextensive fragmentation model \cite{3}, the boundary of the coexistence region is skew with the top tilted towards smaller \( V \) what allows for two crossings with the line \( V = V_f \) and leads to the negative branch of \( C_P \).

In conclusion, 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 nonextensive 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 nonextensive canonical statistical fragmentation model \cite{3} for the understanding of experimental ‘caloric’ curve data \cite{11} and the negative heat capacity data \cite{14} 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 competition between Coulomb repulsive interactions and nonextensive features of the entropy.

![Graph showing specific heat at constant pressure](image)

**FIG. 3.** The specific heat at a constant pressure \( C_P \) is plotted vs the excitation energy per nucleon for various entropic indices \( q \) in the systems with \( A_0 = 200, Z_0 = 80 \) (the upper part) and \( A_0 = 300, Z_0 = 120 \) (the lower part).

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 \cite{14}, closely resemble results of nonextensive fragmentation model for \( q \approx 1.0005 \) and \( A_0 = 200 \) (see Fig. 3). This suggests that the observed effect is caused primarily by the nonextensive features of the entropy. The position of singularity of \( C_P \) at higher excitation energies increases sensitively both with the entropic index \( q \) and with the source size. Correct description of its experimental value \cite{14} by the nonextensive fragmentation model assuming maximal possible size of quasi-projectile source in this experiment \((A_0 \lesssim 200)\) \cite{14}, means that we have determined lower limit for \( q \).

There are many sources of nonextensivity in mesoscopic systems. Some of them, e.g. the formation of liquid-gas (fractal) interphase \cite{5}, have been pointed out in the microcanonical studies \cite{13,16}. Most of these effects are non-generic and, moreover, they are hard to
quantify. This is a principal obstacle in the meaningful characterization of nuclear multifragmentation data in the 'critical region' in the idealized picture of BGSM. On the other hand, the same value of entropic index \( q \sim 1.0005 \) seems to be consistent with both the caloric curve \([1]\) and the negative heat capacity \([14]\) data, in spite of completely different kinematical conditions in these measurements. Moreover, the excitation energy of higher singularity of \( C_p \) seems to be the same both in quasi-peripheral Au+Au collisions at 35A.MeV \([14]\) and in central Xe+Sn collisions at 32A.MeV \([17]\) and agrees with \( q \approx 1.0005 \). This surprising universality remains a puzzle at present.

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