## Multifragmentation and the symmetry term of the nuclear equation of state

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We investigate the possibility of extracting the symmetry energy from multifragmentation data. The applicability of the grandcanonical formula earlier proposed by Ono *et al.* [Phys. Rev. C **68**, 051601(R) (2003)] in the case of finite excited nuclei is tested within a microcanonical framework. Relatively good results are obtained except for large residual nuclei, especially when large sources are highly excited. Effects of secondary particle emission and the extent in which relevant information may be inferred from experimental observables are finally discussed.

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## I. INTRODUCTION

Isospin dependent phenomena are attracting increasing interest as they hold the promise of revealing the asymmetry term of the nuclear equation of state. At normal nuclear density this quantity dictates the structure of the neutron-rich and neutron deficient isotopes, while in other domains of density and temperature its behavior is reflected in a variety of astrophysical phenomena, such as the structure and evolution of neutron stars and the dynamics of supernovae explosions. In nuclear multifragmentation reactions, the asymmetry term influences the neutron-proton composition of the break-up fragments.

Interpreting multifragmentation in the light of first-order phase transitions in multicomponent systems, the neutron enrichment of the gas phase with respect to the liquid phase comes out as a natural consequence of Gibbs equilibrium criteria and a connection between phases chemical composition and the symmetry term can be established [1,2]. Interestingly enough, the phenomenon of isospin fractionation which is systematically observed in analyses of multifragmentation data [3–7], seems to be a generic feature of phase separation independent of the equilibrium Gibbs construction [8]. Indeed, dynamical models of heavy ion collisions [9–13] where fragment formation is essentially ruled by the out of equilibrium process of spinodal decomposition also exhibit fractionation. Adopting an equilibrium scenario for the break-up stage of a multifragmenting system, Ono et al. [12] derive an approximate grandcanonical expression which connects the symmetry term with the isotopic composition of fragments obtained in the break-up stage of two sources with similar sizes in identical thermodynamical states and differing in their isospin content,

$$C_{\rm sym} = -\frac{\alpha_{12}T}{4\left[\left(\frac{Z_1}{A_1}\right)^2 - \left(\frac{Z_2}{A_2}\right)^2\right]},$$
 (1)

under the hypothesis that the isotopic distributions are essentially Gaussian and that the free energies contain only bulk terms. Here,  $\alpha_{12}$  is the isoscaling slope parameter,  $Z_i/A_i$ stands for the isospin asymmetry of a fragment produced by the source i(=1, 2) and T is the temperature of the decaying systems.

In the limit of vanishing temperature, fractionation can be neglected and  $Z_i/A_i$  can be replaced by the corresponding quantity of the sources [7] giving

$$C_{\rm sym} = -\frac{\alpha_{12}T}{4\left[\left(\frac{Z}{A}\right)_{s1}^2 - \left(\frac{Z}{A}\right)_{s2}^2\right]}.$$
 (2)

This expression, which was first proposed in the framework of the EES model [14], as well as Eq. (1), has been used extensively on experimental data and results ranging from values compatible with the ground state bulk symmetry term  $(\approx 25 \text{ MeV})$  to about half this value have been interpreted as sign of dilute matter at freeze-out. Thus,  $C_{\text{sym}} = 14 \text{ MeV}$ has been obtained in FRS experiments using midperipheral collisions of 1 GeV/nucleon <sup>124,136</sup>Xe beams on Pb targets [15]; values decreasing from 25 to 15 MeV have resulted from a variety of heavy ion collisions induced by a few tens MeV/nucleon projectiles studied by the Texas A&M group [16–19], while from the fragmentation of excited target residues produced in the peripheral collisions of <sup>12</sup>C on <sup>112,124</sup>Sn at 300 and 600 MeV/nucleon incident energies the INDRA-ALADIN collaboration extracts a symmetry coefficient which decreases from 25 MeV for peripheral collisions to 15 MeV for central collisions [20].

If these values really correspond to the break-up stage of the decay, the implications are dramatic. First, if the primary fragments at the equilibrated freeze-out are diluted, statistical models which described successfully a wealth of experimental data over more than two decades should be completely reformulated. Starting from the geometrical extension of fragments which dictates the free volume, to the binding and internal excitation energies which enter energy balance and affect fragments partitions, all quantities need serious reconsideration. If, on the other hand, this diluteness would reflect inter-fragment interactions at breakup, as advanced by Botvina *et al.* [7], the situation would be even more severe as it would refute the fundamental hypothesis of statistical models, namely the lack of any fragment interaction except the Coulomb one. Before leaving the discussion on the values of the symmetry energy other than the one corresponding to the ground state and their implications and approach the possibility to access this quantity in multifragmentation data, we recall that values of the symmetry energy decreasing with temperature have been obtained in Ref. [21] for asymmetric nuclear matter within a self-consistent model with an effective interaction while, more recently, shell model Monte Carlo calculations for nuclei with mass around A = 60 indicate the opposite trend [22].

Given the implications of the above-discussed measurements, the present work aims to investigate the possibility to extract the symmetry term from multifragmentation data using Eq. (1). Sticking to the equilibrium hypothesis, Ono's formula has three drawbacks: (a) it is grandcanonical while the grandcanonical approximation is known to be acceptable only for relatively light fragments emitted by large systems with high excitation energies, (b) it does not account for full mass dependence of the binding energy, and (c) it holds for the break-up stage of the reaction, impossible to access experimentally. Equation (2), which was employed to obtain  $C_{\text{sym}}$  values as low as  $\approx 10 \text{ MeV}$  [20], contains the additional approximation of neglecting isospin fractionation.

A quantitative estimation of the possible deformations induced by these effects can be done in the framework of a microcanonical equilibrium model [23]. The less serious objection against Eq. (1), possible deviations produced by omitting the contributions of surface [24], Coulomb and asymmetry terms, may be overcome taking into account the full dependence of the binding energy on the system size and will be addressed first.

Thus, adopting the grandcanonical expression of the isotopic yield of an emitted cluster with N neutrons and Z protons,

$$Y(N, Z) \propto \exp\left[\frac{1}{T}(B(N, Z) + \mu_n N + \mu_p Z)\right], \quad (3)$$

where  $\mu_n$  and  $\mu_p$  stand for neutron and proton chemical potentials and B(N, Z) represents the binding energy, we use as in Ref. [12] the Gaussian shape of Y(N, Z) distributions as a function of N(Z) when Z(N) is kept fixed to approximate the value of N(Z) corresponding to the maximum of the distribution with its average value. Under these assumptions, the equation

$$\frac{\partial(\ln Y(N,Z))}{\partial N} = 0 = \frac{1}{T} \left[ \frac{\partial B(N,Z)}{\partial N} |_{Z} + \mu_{n} \right], \quad (4)$$

applied for two isospin different similar nuclei in identical equilibrium states, relates the isoscaling slope parameter  $\alpha$  with the difference of partial derivatives of B(A, Z) as a function of N.

If we consider for the binding energy a liquid drop parametrization including surface and Coulomb terms,

$$B(A, Z) = (a_v A - a_s A^{2/3}) - (a_i a_v A - a_i a_s A^{2/3}) I^2 + a_c Z^2 / A^{1/3} + a_a Z^2 / A,$$
(5)

and account for full mass dependence of the bulk+surface, isospin-dependent, and Coulomb contributions, the resulting equation will acquire additional terms corresponding to surface, Coulomb, and asymmetry energies,

$$C_{\rm sym}$$

$$= \frac{-\alpha_{12}T + \frac{2}{3}a_{s}(\langle A_{1}\rangle^{-1/3} - \langle A_{2}\rangle^{-1/3})}{4\left[\left(\frac{Z}{\langle A_{1}\rangle}\right)^{2} - \left(\frac{Z}{\langle A_{2}\rangle}\right)^{2}\right]} + \frac{\frac{1}{3}Z^{2}a_{c}(\langle A_{1}\rangle^{-4/3} - \langle A_{2}\rangle^{-4/3}) + a_{a}Z^{2}(\langle A_{1}\rangle^{-2} - \langle A_{2}\rangle^{-2})}{4\left[\left(\frac{Z}{\langle A_{1}\rangle}\right)^{2} - \left(\frac{Z}{\langle A_{2}\rangle}\right)^{2}\right]} + \frac{1}{3}\frac{a_{i}a_{s}(\langle A_{1}^{-1/3}\rangle\langle I_{1}\rangle^{2} - \langle A_{2}^{-1/3}\rangle\langle I_{2}\rangle^{2})}{4\left[\left(\frac{Z}{\langle A_{1}\rangle}\right)^{2} - \left(\frac{Z}{\langle A_{2}\rangle}\right)^{2}\right]}.$$
(6)

The  $\langle A \rangle^{-n}$  (n > 0) dependence of these quantities rend the corrections negligible for heavy fragmentation products [12] and quantitative results will be presented for a case relevant to most multifragmentation reactions.

The approximations of Eqs. (1) and (2) may be judged within a microcanonical multifragmentation model [23]. We use the MMM version [25] where the equilibrated multifragmenting source is characterized by its mass  $A_s$ , charge  $Z_s$ , excitation energy E, total momentum P, total angular momentum L, and freeze-out volume V. All configurations  $C: \{\{A_i, Z_i, \epsilon_i, p_i, r_i\}, \{i = 1, \dots, NF\}\}$  permitted by the specific microcanonical conservation laws are spanned by a Metropolis trajectory and average values of physical quantities are calculated numerically on the basis of the statistical weight of each configuration C. Break-up fragments are treated as normal nuclear density malleable objects described by a zero-temperature binding energy as in Eq. (5) where  $a_v =$ 15.4941 MeV,  $a_s = 17.9439$  MeV,  $a_i = 1.7826, a_c =$ -0.7053 MeV, and  $a_a = 1.1530$  MeV [26]. This liquid drop description is consistent with a semiclassical Thomas-Fermi approximation [27] or hot Hartree-Fock [28], where the effect of temperature is a modified occupation of the single particle eigenstates of the mean field Hamiltonian. The finite temperature fragment energy functional in this approach is thus modified respect to the ground state only for the internal excitation energy  $(\epsilon)$  coming from the occupation of continuum states. To avoid double counting of the free particles states [29], the internal energy is cut at the binding level. In our microcanonical formalism, the temperature is univocally defined through the thermodynamic equality  $T^{-1} =$  $\partial S/\partial E = \langle (3N/2 - 4)/K \rangle$ , where K represents the thermal kinetic energy, N is the product multiplicity and the last equality stems from the equipartition theorem applied to a non interacting system of clusters [30]. It is worthwhile to mention that, due to the high energy cutoff in the cluster level densities, the internal fragment excitation energy cannot be used to estimate the temperature, and a thermometer based upon internal fragment properties would severely underestimate the thermodynamic temperature. In the experimental evaluation of  $C_{\rm sym}$  conversely, the temperature is also estimated from fragment properties or model calculations, which may lead to an extra source of uncertainty.

The break-up stage of the decay which contains all information relevant for the equation of state is completed with

a particle evaporation stage which simulates the subsequent disintegration of the excited fragments [25]. This step is important as well as it shows the capacity of experimentally measurable quantities to access the physics at breakup.

The predictive power of Eqs. (1) and (6) has been systematically checked by confronting their results with the input symmetry energy which enters fragment definition via the binding energy. We have considered a variety of situations in which the size of the emitting sources was varied between A = 230 and A = 100, the freeze-out volume covered the usually accepted interval ( $V = 4V_0 - 8V_0$ ) and the excitation energy ranged between 2 and 10 MeV/nucleon. The lower limit of the source size for which Eqs. (1), (6), and (2) hold was conditioned by the possibility to calculate  $\alpha$  out of the isotopic composition of light emitted fragments. Indeed, for small sources  $\alpha$  manifests a relatively strong dependence on the emitted cluster size, its calculation as an average value getting disputable [31]. For completeness, the stability of the above mentioned formulas was checked against sources isospin variation as well. The conclusions are the same and only few illustrative cases will be considered in the following.

Figure 1 illustrates the magnitude of the additional terms of Eq. (6) corresponding to corrections due to surface, Coulomb, and isospin energies as a function of the charge of the considered fragment in a case typical for nuclear multifragmentation reactions, (210, 82) and (190,82) with  $V = 4V_0$  ( $V_0$  is the volume at normal nuclear density) and E = 6 MeV/nucleon. The values of the order of unity rend these corrections negligible even for light fragments, in agreement with expectations of Ref. [12]. For this reason, the discussion on the possibility to extract the symmetry energy from multifragmentation data will address only the original Eq. (1).

Figure 2 presents break-up stage predictions of MMM for the asymmetry term as a function of the emitted fragment charge. Results of Eqs. (1) and (2) are shown for the multifragmenting nuclei [(130, 50) and (110, 50)] (upper panel) and [(210, 82) and (190, 82)] (lower panel) with  $V = 4V_0$ and excitation energy ranging from 2 to 10 MeV/nucleon. The



FIG. 1. (Color online) MMM predictions for the additional surface (dashed), Coulomb (dotted), and isospin (dot and dash) contributions to the symmetry energy as a function of Z at breakup. The solid curve depicts the sum of the above-mentioned terms. The equilibrated systems are (210, 82) and (190, 82) with  $V = 4V_0$  and excitation energy 6 MeV/nucleon.



FIG. 2. (Color online) MMM predictions for the symmetry energy as a function of Z at breakup. The equilibrated systems are [(130, 50) and (110, 50)] (upper panel) and [(210, 82) and (190, 82)] (lower panel) with  $V = 4V_0$  and excitation energies ranging from 2 to 10 MeV/nucleon. Solid curve: input symmetry energy of the model.  $C_{\text{sym}}$  calculated according to Eq. (1) is represented with symbols while results of Eq. (2) are illustrated with dotted and dashed lines.

MMM results (symbols) are compared to the input symmetry energy of the model,  $a_i a_v - a_i a_s A^{-1/3}$  (solid curve). As one may notice, Eq. (1) shows a remarkable overall stability against excitation energy variations but its behavior while modifying the source size or excitation energy deserves a more attentive investigation. Firstly, Eq. (1) shows a systematic overestimation of the real value by up to 4 MeV for the largest fragments. With the increase of the source size and excitation energy, the overestimation slightly diminishes so that, for the Z = 82 sources and the highest considered excitation energy, 10 MeV/nucleon, the calculated  $C_{\text{sym}}$  exceeds by less than 1 MeV the real value. This is an interesting manifestation of the applicability conditions of grandcanonical approaches in the case of small systems. Thus, for relatively light fragments emitted by highly excited large systems, grandcanonical formulas give reasonable values while more modest results are obtained at low excitation energies and for fragments commensurable with the source size. This result should not be surprising given that even the values of fragment multiplicities are known to depend significantly on the employed statistical treatment as illustrated, for instance, by the grandcanonical and canonical versions of the analytically solvable thermodynamic model of multifragmentation of Ref. [32] where the values converge exclusively for relatively light fragments emitted by highly excited sources.

The bending of the curves calculated with Eq. (1) for the Z = 50 sources at 2 and 4 MeV/nucleon is due to the mass and charge conservation specific of a microcanonical ensemble. Similar effect manifests also for the Z = 80 case, but the limited charge domain in the right panel of Fig. 2 hides it. The residual slight overestimation of the input value of  $C_{\text{sym}}$  in a domain where the grandcanonical approximation should in principle be acceptable remains an open question and will be addressed in the future. However, we have checked that it does not depend on the value of the high energy cutoff  $\exp(-\epsilon/\tau)$  of the fragment internal density, as similar results are obtained when  $\tau \to \infty$ .

As expected, Eq. (2) which approximates the isospin content of fragments to the one of the source gives good results only for very heavy fragments and low excitation energy. However in the fragmentation regime it leads to a systematic underestimation of the symmetry energy. In particular, Fig. 2 suggests that the low value  $C_{\text{sym}} \approx 15$  MeV extracted in some experimental analyses [15,20] may be compatible with standard ground state values for the symmetry energy. In this respect it is interesting to remark that the use of Eq. (2) produces an unphysical apparent reduction of  $C_{\text{sym}}$  with increasing excitation, similar to the findings of Ref. [20]. The explanation of this evolution in the model lays in the steep diminish of  $\alpha$  not compensated by the temperature increase [31].

Figure 2 was obtained for a specific choice of the fragmenting sources. However it was proved in Ref. [31] that the isoscaling parameter  $\alpha$  manifests a complex dependence on the considered pair of sources and their equilibrium state under the microcanonical constraint. This fact requires a detailed investigation of the persistence of Eq. (1) under various freeze-out volumes, source isospin combinations, and source sizes. Therefore, Fig. 3 presents the results of Eq. (1) for the same sources [(210, 82) and (190, 82)] with 6 MeV/nucleon excitation energy when the freeze-out volume has different values ( $V = 4V_0$  and  $V = 8V_0$ ) (upper panels), while the behavior with respect to sources isospin modification is represented considering three pairs of Z = 82nuclei [(190, 82), (200, 82) and (210, 82)] at  $V = 4V_0$  and 6 MeV/nucleon excitation energy (lower panels). As in the previous case, the solid curve illustrates the  $(a_i a_v - a_i a_s A^{-1/3})$ term. The conclusions confirm the stability of Eq. (1) while modifying source isospin and freeze-out volume. A more increased predictive power is expected for larger sources with a more advanced fragmentation, where the grandcanonical approximation is more reasonable.

If the preceding figures suggest that for large systems and high excitation energies, Eqs. (1) and (6) may allow one to extract the symmetry energy coefficient at the fragmentation stage from isoscaling observables, it is important to stress that not only the average fragment isotopic composition, but also the isoscaling parameter  $\alpha$  should be known at the time of fragment formation, while the measured  $\alpha$  value may have been distorted by sequential decay. The effect of secondary decay on the isoscaling parameter is explored in Fig. 4. Since in the MMM model the value of  $\alpha$  is larger in the asymptotic stage of the reaction than at breakup [31], Eqs. (1) and (6) lead to an overestimation of the symmetry energy coefficient.



FIG. 3. (Color online) MMM predictions for the symmetry energy as a function of Z at breakup as obtained using Eq. (1) (open symbols). The stability against freeze-out volume variation ( $V = 4V_0$ and  $V = 8V_0$ ) is illustrated for the equilibrated systems (190, 82) and (210, 82) with 6 MeV/nucleon excitation energy (upper panel). The stability against sources isospin modification is checked for three pairs of Z = 82 nuclei with  $V = 4V_0$  and 6 MeV/nucleon excitation energy (lower panel). Results of Eq. (2) are indicated using dotted and dashed lines.

If fractionation is neglected using Eq. (2) these two errors tend to compensate and the extracted values of  $C_{\text{sym}}$  range from 20 to 25 MeV, in better agreement with the real value (if sufficiently heavy isotopes are analyzed for the surface influence to be negligible, which is not the case in present fragmentation data). It is important to stress that this compensation entirely relies on the evolution of  $\alpha$  from hot to cold fragments. Unfortunately, in this respect, information in the



FIG. 4. (Color online) MMM predictions for the symmetry energy as a function of Z in the asymptotic stage of the decay as obtained using Eq. (1) (open symbols). The equilibrated systems are (190, 82) and (210, 82) with  $V = 4V_0$  and excitation energies ranging from 2 to 10 MeV/nucleon. Results of Eq. (2) are indicated using dotted and dashed lines. In all cases, asymptotic values of  $\alpha$ and break-up values of  $Z/\langle A \rangle$  have been used.

literature is controversial. Thus, MSU-SMM [33] and Isospin Quantum Molecular Dynamics (IQMD) [34] plead in favor of negligible contribution of sequential evaporation on  $\alpha$  as stated in Refs. [14] and [35], respectively. Stochastic mean field [36], Antisymmetrized Molecular Dynamics (AMD) [12], and the Markov-chain SMM [37] with  $C_{\text{sym}} > 15$  MeV indicate  $\alpha$ values decreasing from the breakup to the asymptotic stage [20,38,39]. Finally, MMM, EES [40] and the Markov-chain SMM with  $C_{\text{sym}} < 15$  MeV predict the opposite effect:  $\alpha$ values increasing from the breakup to the asymptotic stage [14,20,31]. The origin of these discrepancies could by due to isospin population of break-up fragments, their excitation energy and secondary decay procedure as well, but a pertinent discussion of this issue goes much beyond the limits of present paper.

To summarize, we investigated the possibility of inferring the symmetry energy coefficient from isoscaling observables in multifragmentation reactions. Our results indicate that, in the limit of large systems and high excitation energies where the use of a grandcanonical approach is reasonable, Eq. (1) is a reliable tool to determine  $C_{\text{sym}}$  out of the isotopic

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composition of the break-up fragments but overestimations of few MeV are expected for the systems usually produced in multifragmentation reactions. In particular low values of  $C_{\text{sym}}$  respect to the standard saturation density bulk value may be interpreted as an effect of surface contributions to the symmetry energy.

Conversely, if we assume that the density of break-up fragments is close to their ground state density [23], the symmetry energy coefficient can be considered as known and isoscaling measurements can be used to probe fractionation. In particular spinodal decomposition is predicted to lead to higher fractionation than phase separation at equilibrium [41], and this effect could be accessed through isoscaling observables. To realize this ambitious program it is essential to experimentally control the effect of side feeding on the  $\alpha$  isoscaling parameter.

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