Phase Coexistence in Multifragmentation?

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The charge (Z) distributions from intermediate energy heavy-ion reactions depend upon the multiplicity n of intermediate mass fragments through a factor of the form e^{-cnZ}. Experimentally c starts from zero at low values of the transverse energy E_t and reaches a saturation value at high E_t. In a liquid-gas phase diagram $c = 0$ for the saturated vapor, while $c > 0$ for the unsaturated vapor. It is suggested that in the $c \approx 0$ regime the source evaporates down to a sizable remnant, while for $c > 0$ the source vaporizes completely. Percolation of finite systems and nuclear evaporation portray a behavior similar to that observed experimentally.

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Multifragmentation has been frequently associated with the liquid-vapor phase transition. A large amount of theoretical work [1–7] describes multifragmentation as due either to droplet formation near criticality (critical opalescence) or to the appearance of both liquid and vapor phases after crossing the spinodal line. Percolation theories have been developed to mimic critical behavior in finite systems [8,9]. Behavior near criticality has been inferred from the experimentally observed power-law dependence of the intermediate mass fragment (IMF, $3 \leq Z \leq 20$) charge distribution [10–13]. More recently, critical exponents have been extracted from the dependence of the moments of the charge distribution upon quantities, like the charge multiplicity, that are typically associated with temperature and/or excitation energy [14].

One of the goals of multifragmentation studies is to map the nuclear phase diagram, in particular, the boundaries of the phase coexistence region. In this Letter we report evidence for a possible transition from a two-phase to a one-phase regime obtained from the analysis of charge distributions and their dependence upon IMF multiplicity.

It has been observed [15,16] that, for a large number of reactions, the IMF multiplicities are binomially distributed, and that the probability P_n to emit n IMFs can be reduced to a one-fragment emission probability p. This is true at all transverse energies ($E_t = \sum_i E_i \sin^2 \theta_i$, where the sum is over all particles in an event and E_i and θ_i are the kinetic energy and polar angle of particle i, respectively) [15,16]. The resulting one-fragment emission probability appears to have a Boltzmann form $p = e^{-B/T}$, assuming that the temperature $T \propto \sqrt{E_t}$ and B represents an average barrier for fragment emission. The thermal nature of p was inferred from the observation of linear Arrhenius plots ($\ln p \propto 1/\sqrt{E_t}$), where E_t is assumed to be proportional to excitation energy.

The reducibility of P_n to p was verified empirically for fragments with $Z \geq 3$. The implications of such reducibility for the charge distributions as a function of the fragment multiplicity n were discussed in Ref. [17]. In this work it was found that the experimental charge distributions for any value of n and E_t could be expressed by the equation

$$P_n(Z) \propto \exp -\left[\frac{-F(Z)}{\sqrt{E_t}} + ncZ\right], \qquad (1)$$

where $F(Z)$ represents a universal function of Z.

The dependence of $P_n(Z)$ on n is illustrated in Fig. 1 for the reaction ^{36}Ar + ^{197}Au at $E/A = 110$ MeV. In the top panel, the different symbols show the charge distributions for different fragment multiplicities n at a fixed $E_t = 650$ MeV. The slight differences in the n gated charge distributions nearly disappear when plotted in the “reduced” form $F(Z)$ shown in the middle panel.

Again, assuming E_t proportional to the excitation energy of the source, Eq. (1) can be written in terms of the temperature T:

$$\begin{aligned} P_n(Z) &\propto \exp -\left(\frac{B(Z)}{T} + ncZ\right) \\ &= \exp -\left(\frac{\Delta E(Z)}{T} + \Delta S(n,Z)\right). \end{aligned} \qquad (2)$$

The first term in the exponent was interpreted [17] as an energy or enthalpy term, associated with the energy (enthalpy) needed to form a fragment. The second term was claimed to point to an asymptotic entropy associated with the combinatorial structure of multifragmentation. It was observed that a term of this form arises naturally in the charge distribution obtained by the least biased breaking of

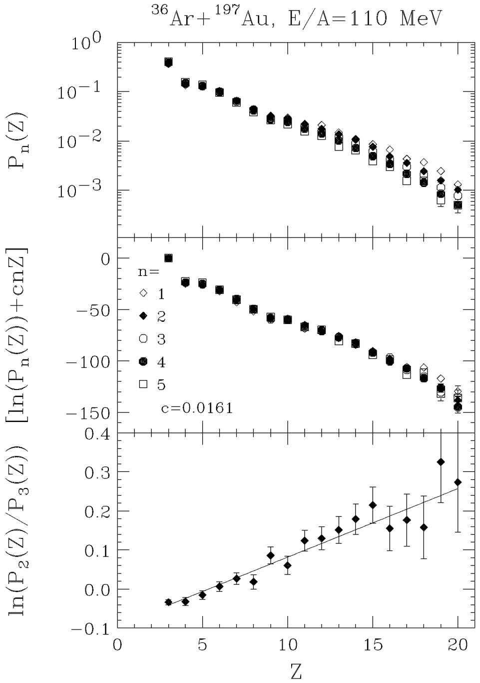

FIG. 1. Top panel: the n gated charge distributions $P_n(Z)$ for the reaction ^{36}Ar + ^{197}Au at $E/A = 110$ MeV. The charge distributions were constructed from events with $E_t = 650 \pm 20$ MeV and $n = 1-5$. Middle panel: the "reduced" charge distribution [17] for the same data using the indicated value of c. (The data here are normalized at $Z = 3$.) Bottom panel: the log of the ratio of $P_2(Z)/P_3(Z)$. The slope corresponds to c for $n = 2$ [see Eq. (4)]. The statistical error bars are shown for errors larger than the symbol size.

an integer Z_0 into n fragments. Such a Z distribution is given approximately by

$$P(Z) = \frac{n^2}{Z_0} e^{-nZ/Z_0} = cn^2 e^{-cnZ}. \tag{3}$$

While this form obviously implies charge conservation, it is not necessary that charge conservation be implemented as suggested by Eq. (3). In fact, it is easy to envisage a regime where the quantity c should be zero. Sequential thermal emission is a case in point. Since any fragment does not know how many other fragments will follow its emission, its charge distribution cannot reflect the requirement of an unbiased partition of the total charge among n fragments. We have in mind a liquid drop evaporating fragments of different size and binding energy. "Charge conservation" will affect the distribution minimally, unless evaporation consumes the entire system, and even then, not in the sense of an unbiased partition. A simulation in which fragments with different barriers are allowed to be emitted sequentially according to the binomial scheme of Ref. [15] yields indeed $c = 0$ if a residue survives.

On the other hand, in a simultaneous emission controlled by an n-fragment transition state [18], fragments would be strongly aware of each other and would reflect such an awareness through the charge distribution.

The question then arises whether $c = 0$ or $c > 0$, or even better, whether one can identify a transition from a regime for which $c = 0$ to a new regime for which $c > 0$. To answer this question, we have studied the charge distributions as a function of fragment multiplicity n and transverse energy E_t for a number of systems and excitation energies. Specifically, we will present data for the reaction ^{36}Ar + ^{197}Au at $E/A = 80$ and 110 MeV and the reaction ^{129}Xe + ^{197}Au at $E/A = 50$ and 60 MeV.

Several approaches were used to extract c from the charge distributions. If the charge distributions are exponential [as is sometimes the case, $P_n(Z) \propto e^{-\alpha_n Z}$], it is sufficient to extract from them the exponential coefficient α_n. From the n dependence of α_n, the quantity c is readily extracted [17]. A more general approach which does not depend on any specific form for the charge distribution is to construct at each E_t the ratio

$$\frac{P_n(Z)}{P_{n+1}(Z)} \propto e^{cZ}. \tag{4}$$

A value of c can be extracted for each n by taking the log of this ratio and finding the slope of the resulting graph (see bottom panel of Fig. 1). A weighted average (over all IMF multiplicities n) for c can then be constructed at all E_t. Alternatively, a χ^2 can be constructed in terms of the differences in $F(Z)$ [see Eq. (1)] between any pairs of n values and minimized as a function of c. These procedures yield essentially the same results which are reported in Fig. 2 for the ^{129}Xe + ^{197}Au and ^{36}Ar + ^{197}Au reactions.

It is interesting to notice that for all reactions and bombarding energies the quantity c starts at or near zero, it increases with increasing E_t for small E_t values, and seems to saturate to a constant value at large E_t.

This behavior can be compared to that of a fluid crossing from the region of liquid-vapor coexistence (univariant system) to the region of overheated and unsaturated vapor (bivariant system, see Fig. 3). In the coexistence region, the properties of the saturated vapor cannot depend on the total mass of fluid. The presence of the liquid phase guarantees mass conservation at all average densities for any given temperature. A change in mean density (volume) merely changes the relative amount of the liquid and vapor, without altering the properties of the saturated vapor. Hence the vapor properties, and, in particular, the cluster size distributions, cannot reflect the total mass or even the mean density of the system. In our notation, $c = 0$.

On the other hand, in the region of unsaturated vapor, there is no liquid to ensure mass conservation. Thus the vapor itself must take care of this conservation, at least grand canonically. In our notation, $c > 0$. In other words we can associate $c = 0$ with thermodynamic univariance, and $c > 0$ with bivariance.

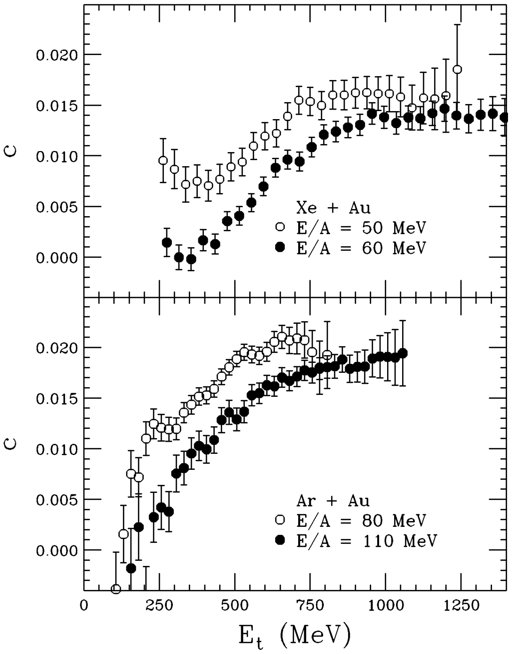

FIG. 2. Plots of the coefficient c vs E_t for the reactions ^{129}Xe + ^{197}Au at $E/A = 50$ and 60 MeV (top panel) and ^{36}Ar + ^{197}Au at $E/A = 80$ and 110 MeV (bottom panel). The error bars are statistical.

These considerations lead to the immediate identification: $c \propto \mu_L - \mu_V$ in the Fischer model [1], where μ_L and μ_V are the chemical potentials of the liquid and the vapor, respectively. Thus, in the coexistence region $(\mu_L = \mu_V)$ $c = 0$, while for the overheated vapor $(\mu_V < \mu_L)$ $c > 0$.

This description should not be taken too literally, for a variety of reasons, one of which is the finiteness of the system. The $c = 0$ regime may just signify an evaporativelike emission from a source which survives as a charge conserving residue (liquid), while the $c > 0$ regime may signify the complete vaporization of the source.

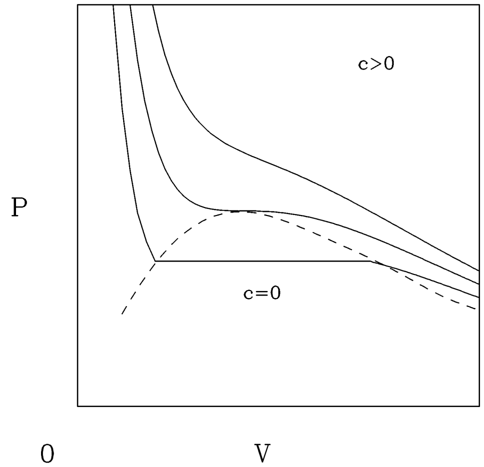

FIG. 3. Liquid-gas phase diagram. The top of the dashed line is the critical point. The area below the dashed line ($c = 0$) is the region where mixtures of the liquid and gas phases coexist.

To test these ideas in finite systems, we have considered a finite percolating system and a system evaporating according to a thermal binomial scheme [15,19]. Percolation calculations [8] were performed for systems of $Z_0 = 97$, 160, and 400 as a function of the percentage of bonds broken (p_b). Values of c were extracted [using Eq. (4)] as a function of p_b.

The results are shown in Fig. 4. Guided by the insight gleaned from the approximate solution to Euler's problem [see Eq. (3)], we have scaled the extracted values of c by the source size Z_0 in order to remove this leading dependence and to evidentiate the true finite size effects. For values of p_b smaller than the critical (percolating) value ($p_b^{\text{crit}} \approx 0.75$ for an infinite system), we find $c = 0$. This is the region in which a large (percolating) cluster is present. As p_b goes above its critical value, the value of c increases, and eventually saturates in a way very similar to that observed experimentally.

Notice that although the phase transition in the infinite system is second order at $p = p_c$, here the region for which $c = 0$ mimics a first order phase transition.

Before proceeding, let us remind ourselves that charge conservation is *not* a finite-size effect. For instance, the chemical potential, introduced in statistical mechanics to conserve mass, survives the thermodynamical limit and retains its meaning for an infinite system, despite the fact that the extensive thermodynamic quantities go to infinity. In our case, while it is true that c goes to zero or that $1/c$ goes to infinity, it is also true that the product cZ_0 tends to a finite limit nearly independent of Z_0.

Our analysis is not directly comparable to the Euler solution [Eq. (3)], since we have restricted ourselves to a

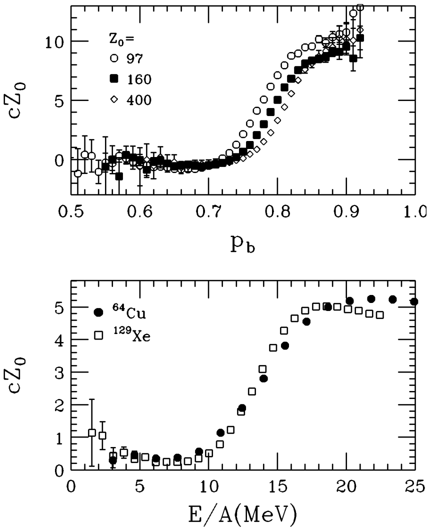

FIG. 4. Top: A plot of cZ_0 versus the percentage of broken bonds p_b from a percolation calculation [8] for three systems $Z_0 = 97$ (circles), 160 (squares), and 400 (diamonds). Bottom: A plot of cZ_0 versus excitation energy per nucleon from a binomial evaporation calculation [19] for ^{64}Cu and ^{129}Xe. The statistical error bars are shown for errors larger than the symbol size.

limited region ($3 \leq Z \leq 20$) of the total charge distribution for our study of how the source is partitioned into different IMF multiplicities. Furthermore, Eq. (3) and the associated dependence of c upon Z_0 are characteristic of a one-dimensional percolation model. Therefore, it is not unexpected that c appears to be proportional, but not equal, to $1/Z_0$ in the three-dimensional percolation calculation reported in Fig. 4.

An evaporation calculation was also carried out for the nuclei ^{64}Cu and ^{129}Xe according to the thermal binomial scheme [15,19]. The only constraint introduced was to prevent at every step the emission of fragments larger than the available source. The resulting charge distributions are very well reproduced by Eq. (1). The extracted quantity cZ_0 is plotted in the bottom panel of Fig. 4 as a function of excitation energy per nucleon. In both cases cZ_0 goes from 0 to a positive finite value (equal for both nuclei) as the energy increases. The region where $c = 0$ is readily identified with the region where a large residue survives. On the other hand, when $c > 0$ there is no surviving residue.

These results are in striking agreement with those obtained for percolation. For both kinds of finite systems, the univariant regime ($c = 0$) is associated with the presence of a residue, while the bivariate regime ($c > 0$) with the absence of a residue.

In conclusion, we have presented extensive evidence for an n dependence of charge distributions of the form given by Eq. (1). We have shown that the parameter c increases from near zero at low E_t (excitation energy) to a saturation value at high E_t. Using the analogy of liquid-vapor equilibrium we have argued that $c = 0$ may indicate the presence of two phases (liquid-vapor), while $c > 0$ may indicate the presence of one phase (unsaturated vapor). We have shown that percolation calculations for finite systems can be analyzed in the same way as the data and portray the same dependence for c as one moves from the region where a percolating cluster is present to one where such a cluster is absent. Similar results are obtained from an evaporation calculation.

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