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Nuclear multifragmentation time-scale and fluctuations of largest fragment size

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Distributions of the largest fragment charge, $Z_{\text{max}}$, in multifragmentation reactions around the Fermi energy can be decomposed into a sum of a Gaussian and a Gumbel distribution, whereas at much higher or lower energies one or the other distribution is asymptotically dominant. We demonstrate the same generic behavior for the largest cluster size in critical aggregation models for small systems, in or out of equilibrium, around the critical point. By analogy with the time-dependent irreversible aggregation model, we infer that $Z_{\text{max}}$ distributions are characteristic of the multifragmentation time-scale, which is largely determined by the onset of radial expansion in this energy range.

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Introduction In central heavy-ion collisions at beam energies of $\sim$20-150 MeV/A multiple production of nuclear fragments can be observed, compatible with the quasi-simultaneous break-up of finite pieces of excited nuclear matter \[1-7\]. This so-called “nuclear multifragmentation” is a fascinating process \[8\] which has long been associated with a predicted liquid-gas coexistence region in the nuclear matter phase diagram at sub-critical temperatures and sub-saturation densities \[9-11\]. Statistical \[12-19\] and dynamical \[20-24\] aspects have been widely studied, and evidences supporting equally well either a continuous phase transition of the liquid-gas universality class \[13,14,25,30\], a discontinuous (“first-order”) transition occurring within the coexistence region \[24,35\], or indeed the survival of initial-state correlations in a purely dynamical picture \[36,37\] have been presented. This state of affairs well demonstrates the difficulty of quantitatively identifying a phase transition in small systems such as atomic nuclei, where finite-size effects blur the nature of the transition \[38-40\] whose order may indeed change with the size of the system \[23,30\], along with the importance of long-range Coulomb forces \[4,19,41,42\], and presence of dynamical effects such as radial flow \[43-49\].

In this context we have tried to establish generic features of multifragmentation in order to deduce its nature in a less model-dependent way. In our previous works \[50\], we used the model-independent universal fluctuations theory \[51\] to determine that the order parameter of nuclear multifragmentation is the size of the largest fragment of each partition. This means that we know to which class of generic cluster models nuclear multifragmentation belongs, answering the question raised in \[52\]: it is an aggregation phenomenon (“condensation of vapor”), not a fragmentation process (“shattering of glass”). Next \[53\], we studied the order parameter distributions for a wide range of data and showed that, to a first approximation, they tend towards one of two laws: the Gaussian distribution of the central limit theorem at the lowest energies, or the Gumbel distribution \[54,55\] of extreme value statistics \[56\] at the highest. The system-size dependence of the energy of transition from one regime to the other was mapped out and tentatively associated with the observed behavior of limiting temperatures for finite nuclei \[57\].

In this letter we will study in more detail the transition from one regime to the other, using new data on largest fragment distributions in central collisions at bombarding energies intermediate between the two asymptotic regimes. We will show that, in the transition region, these distributions are better approximated by an admixture of the two asymptotic distributions with proportions which evolve with the bombarding energy. We will then compare this behavior with that seen in two generic models of aggregation, for finite sys-
Experimental analysis

Collisions of $^{129}$Xe+$^{nat}$Sn were measured using the INDRA 4π charged product array [58] at the GANIL accelerator facility. This detector, composed of 336 detection cells arranged according to 17 rings centered on the beam axis, covers 90% of the solid angle and can identify fragments from hydrogen to uranium with low thresholds. More details can be found in [58]. Data used here were taken during two separate campaigns: beam energies of 25, 29, 35, 45 and 50 MeV/A were previously analyzed in [50, 53]; measurements at beam energies of 27, 29, and 35 MeV/A were subsequently performed specifically in order to probe the energy range around the change of scaling regime observed in [50], and are presented here for the first time.

We want to study central collisions, requiring the geometrical overlap between projectile and target to be as close as possible to total, while detecting all fragments event by event in order to correctly measure the distribution of the largest fragment charge, $Z_{\text{max}}$. We therefore look for events which maximize the quantity $(E_{t_2} \times Z_{\text{tot}})$: $E_{t_2}$ is the total transverse energy of light charged particles ($Z = 1, 2$), which increases with collision centrality [60], while $Z_{\text{tot}}$ is the sum of the atomic numbers of all detected charged products in each event. For each beam energy we define a centrality cut corresponding to the last percentile of the $(E_{t_2})$ energy we define a centrality cut corresponding to the detected charged products in each event. For each beam multiplicity $M \geq 4$.

We begin by examining the scaling properties of $Z_{\text{max}}$ fluctuations including the three new data points at 27, 29 and 35 MeV/A. Figure 1 shows the correlations between the first two cumulant moments of the $Z_{\text{max}}$ distribution ($(Z_{\text{max}})^2$ and $\sigma^2$), for each beam energy. As in Fig.1(c) of [50], the data fall on two branches, $\sigma^2 \sim (Z_{\text{max}})^{2A}$, with different values for the scaling parameter, $\Delta$ [50]: $\Delta \sim 1$ above 32 MeV/A, and $\Delta \sim 1/2$ below 32 MeV/A. The new data points present a consistent behavior which follows the systematic scaling trend.

The order parameter distributions (largest fragment charge, $Z_{\text{max}}$, distributions) for four bombarding energies are presented in Fig.2(a-d). They become more asymmetric with increasing bombarding energy, tending towards the asymptotic Gumbel distribution at 50 MeV/A. At intermediate energies (see e.g. new data at 29 MeV/A, Fig.2(b)) the distribution is clearly of neither one or the other asymptotic forms, leading us to propose the following ansatz: the effective $Z_{\text{max}}$ distribution at any beam energy is an admixture of a Gaussian and a Gumbel distribution,

$$f(x) = \eta f_{\text{Gauss}}(x) + (1-\eta)f_{\text{Gumbel}}(x)$$

(1)

with $0 < \eta < 1$ and $x = Z_{\text{max}}$. As a first test of its validity, fits to the $Z_{\text{max}}$ distribution using Eq.(1) are shown in Fig.2. It should be noted that in these fits the positions and widths of the two components, as well as their relative weights, were left as free parameters. Reduced $\chi^2$ values for all energies lie between approximately 4 and 10, and can further be reduced if $Z_{\text{max}}$ distributions are smoothed to remove odd-even staggering of the yields [62]: in that case all $\chi^2$ values are close to 2, except for data at 32 MeV/A for which a significantly larger value ($\chi^2 \sim 4$) is obtained. These values are significantly better than those obtained for single-component fits (as in
The quantity $R = 2\eta - 1$ has been defined in such a way that $R = \pm 1$ for pure Gauss/Gumbel distributions, and $R = 0$ when both are equally important. Its evolution over the bombarding energy range 25-50 MeV/A is presented in Fig. 3(e). The vertical bars show an estimated uncertainty coming from the fitting procedure. The monotonically decreasing value of $R$ reflects the continuous evolution of the form of the order parameter distribution. It is interesting to note that the value $R = 0$ is reached between 29 and 32 MeV/A, in the same bombarding energy range as the change of $\Delta$-scaling (Fig. 3).

**Theoretical models** We will now consider some results of generic aggregation models in order to see if our ansatz, Eq. (1), is just a convenient fitting function or if it can have some physical interpretation.

First of all, let us consider the most well-known and widely-used model of this kind, percolation [63]. Indeed, percolation models have long been used in the analysis and interpretation of multifragmentation data [28, 64–69]. In bond percolation, each lattice site corresponds to a monomer, and a proportion $p$ of active bonds is set randomly between sites. Then clusters of size $s$ are defined as an ensemble of $s$ occupied sites connected by active bonds. For a definite value of $p = p_c$, a macroscopic cluster appears, corresponding to the sol-gel transition. The order parameter of the transition is the size of the largest cluster, $s_{\text{max}}$, and it is known that for sub-critical finite lattices, $s_{\text{max}}$ has a Gumbel distribution [70], while at criticality in the mean-field limit $s_{\text{max}}$ follows the Kolmogorov-Smirnov (K-S) distribution [71].

Fig. 3(a-d) show $s_{\text{max}}$ distributions calculated for cubic $6 \times 6 \times 6$ lattices with bond probabilities $p$ above and below the critical value of $p_c = 0.312$. For $p \ll p_c$, the distribution is well described by the Gumbel distribution alone, as expected [70], while for $p \gg p_c$, the distribution approaches a Gaussian form. In the critical region the distribution of $s_{\text{max}}$ takes on a non-trivial form which is rather well fitted by the ansatz of Eq. (1). It should be noted that deviations close to $p = p_c$ are to be expected, as in this case correlations cannot be neglected as required for either Gumbellian or Gaussian statistics to be strictly valid. Nevertheless the ansatz of Eq. (1) is a good approximation to the order parameter distribution for all values of $p$.

Fig. 3(e) presents the evolution of the relative strengths of the two components, $R$, in the critical domain. It varies smoothly from $-1$ to $+1$ with increasing $p$. The value $R = 0$ corresponds to very large fluctuations of $s_{\text{max}}$ (see Fig. 3(c)), but is reached for $p > p_c$, not at the infinite-lattice critical point, due to the finite size of the lattice. It is interesting to note that the value $R \approx -0.45$ at $p = p_c$ is very close to the value obtained from a fit to the K-S distribution with Eq. (1).

Percolation is an equilibrium model of reversible aggregation. The irreversible sol-gel transition can be modeled using the coupled nonlinear differential equations in the concentrations of clusters of mass $s$ per unit volume, known as the Smoluchowski equations [72]. The probability of aggregation per unit time between clusters is a homogeneous function of cluster masses, and after a time $t_c$, called the critical gelation time, an “infinite” cluster appears. It was shown in [61] that for this model $s_{\text{max}}$ exhibits the $\Delta = 1$ scaling at $t = t_c$, and $\Delta = 1/2$ scaling with a symmetric distribution for $t \gg t_c$. Fig. 3(a–d) present the $s_{\text{max}}$ distributions obtained at different times for a system of total mass $N = 216$. These distributions and their evolution over time are very similar to those obtained from percolation as a function of the bond probability, Fig. 3. Fits to these distributions using Eq. (1) are shown, and once again they are a good approximation for $s_{\text{max}}$ distributions both far from and around the critical region. Fig. 3(e) shows the relative strengths of the two components, $R$, obtained from the fits as a function of time: as in the case of percolation calculations, $R = 0$.
does not coincide with the infinite-system critical gelation time (in fact it occurs between $t = t_c$ and the time of maximum fluctuations). Also, it may be remarked that at the critical time, the value of $R$ is very close to the K-S distribution one, and indeed independent of the size, $N$, of the system. Moreover, these results are unchanged if a fragmentation kernel is included in the equations, i.e. if clusters can also split into smaller pieces \[ 73 \].

**Discussion** We have shown that there is a strong similarity between $Z_{\text{max}}$ distributions for central collisions of $^{129}$Xe+$^{\text{nat}}$Sn and the order parameter distributions obtained for two very different generic aggregation models in their critical domain. We would now like to understand this similarity. The percolation model offers little scope for interpretation, as all physics is “hidden” in the bond probability, $p$. On the other hand, the physical picture of clusters being built up over time by agglomeration described by the Smoluchowski equations recalls microscopic approaches in which fragments result from the spinodal decomposition of the hot, expanding nuclear matter formed by the head-on collision of two nuclei \[ 74–77 \].

In this framework, we can infer that the $Z_{\text{max}}$ distribution and its form, quantified by the ratio $R$, reflect the time-scale of fragment formation, whose determining factor is the amount of collective radial expansion which increases with bombarding energy \[ 8 \]. It has been shown that for central $^{129}$Xe+$^{\text{nat}}$Sn reactions the onset of significant radial expansion occurs for beam energies above 25 MeV/A \[ 53 \]. The similarity between Figs. 2 and 3 can therefore be understood in terms of fragment size distributions being determined on shorter and shorter time-scales due to increasingly rapid expansion.

We can use this interpretation to understand the system mass-dependence of the energy of transition from $\Delta = 1/2$ to $\Delta = 1$ scaling presented in \[ 53 \]: for $^{58}$Ni+$^{58}$Ni collisions, measured from 32 MeV/A to 90 MeV/A, a change of $\Delta$-scaling and of the form of the $Z_{\text{max}}$ distribution were observed, as for $^{129}$Xe+$^{\text{nat}}$Sn but at a higher bombarding energy of 52 MeV/A; for the lighter system $^{36}$Ar+KCl, studied from 32 MeV/A to 74 MeV/A, only the $\Delta = 1/2$ regime was observed, with quasi-Gaussian $Z_{\text{max}}$ distributions; on the other hand, for the much heavier $^{197}$Au+$^{197}$Au system, at bombarding energies between 40 MeV/A and 80 MeV/A, only the $\Delta = 1$ regime occurs, with Gumbel $Z_{\text{max}}$ distributions.

Radial expansion in central heavy-ion collisions occurs after significant compression of the incoming nuclear fluid, and as such depends not only on static nuclear matter properties such as incompressibility, but also on transport properties such as the degree of stopping achieved in the collision \[ 78 \]. The latter increases with the mass of the colliding nuclei, as shown in \[ 73 \]. Thus for light systems, such as $^{36}$Ar+KCl or $^{58}$Ni+$^{58}$Ni, the bombarding energy required to achieve sufficient initial compression for there to be significant radial expansion is higher than for the heavier systems like $^{129}$Xe+$^{\text{nat}}$Sn and $^{197}$Au+$^{197}$Au. This explains why the $\Delta$-scaling transition occurs at higher energy for $^{58}$Ni+$^{58}$Ni than for $^{129}$Xe+$^{\text{nat}}$Sn. For the very light $^{36}$Ar+KCl system the threshold is higher still than for $^{58}$Ni+$^{58}$Ni, outside the range of measured bombarding energies. On the other hand, for $^{197}$Au+$^{197}$Au both the greater initial compression and the far larger Coulomb contribution may come into play in order to reduce the fragment formation time-scale even at the lowest bombarding energy.

**Conclusions** We have shown that, for finite systems, the largest cluster size distribution in critical aggregation models is an admixture of the two asymptotic distributions observed far below and above the critical region. This result holds true for both equilibrium (percolation) and out-of-equilibrium (irreversible aggregation) models. A similar decomposition has been shown for the experimentally-observed charge distribution of the largest fragment per event produced in nuclear multifragmentation, indicating that the critical domain lies around...
$E_{\text{beam}} \approx 30\text{MeV/A for the }^{129}\text{Xe}^{+}\text{natSn} \text{ system. By analogy with the irreversible aggregation model, where the form of the order parameter distribution depends on the time-scale of the process, we interpret such criticality along with the corresponding change of $\Delta$-scaling as the onset of an “explosive” multifragmentation regime in which initially compressed heated nuclear liquid clusters in the presence of significant radial expansion}$. The mass-dependence of the energy at which the onset occurs for different (symmetric) systems is related to nuclear stopping and hence to transport properties of hot nuclear matter. Such an overall picture is both consistent with, and provides a link between, recent results on nuclear matter. Corresponding author; Electronic address: john.frankland@ganil.fr

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