Nuclear Multifragmentation Time Scale and Fluctuations of the Largest Fragment Size

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(Received 3 October 2012; revised manuscript received 26 February 2013; published 25 April 2013)

Distributions of the largest fragment charge, Z_{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_{max} distributions are characteristic of the multifragmentation time scale, which is largely determined by the onset of radial expansion in this energy range.

DOI: [10.1103/PhysRevLett.110.172701](http://dx.doi.org/10.1103/PhysRevLett.110.172701) PACS numbers: 25.70.Pq, 64.60.an

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 quasisimultaneous break-up of finite pieces of excited nuclear matter [[1](#page-4-1)[–7\]](#page-4-2). This so-called "nuclear multifragmentation'' is a fascinating process [\[8](#page-4-3)] which has long been associated with a predicted liquid-gas coexistence region in the nuclear matter phase diagram at subcritical temperatures and subsaturation densities [\[9–](#page-4-4)[11](#page-4-5)]. Statistical $[12–19]$ $[12–19]$ and dynamical $[20–24]$ $[20–24]$ $[20–24]$ aspects have been widely studied, and evidence supports equally well either a continuous phase transition of the liquid-gas universality class [\[13](#page-4-10)[,14,](#page-4-11)[25](#page-4-12)[–30\]](#page-4-13), a discontinuous (''first-order'') transition occurring within the coexistence region [[29](#page-4-14)–[35](#page-4-15)], or the survival of initial-state correlations in a purely dynamical picture [[36,](#page-4-16)[37](#page-4-17)]. This state of affairs demonstrates well 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](#page-4-18)[–40\]](#page-5-0) whose order may indeed change with the size of the system [\[29](#page-4-14)[,30\]](#page-4-13), along with the importance of long-range Coulomb forces $[4,19,41,42]$ $[4,19,41,42]$ $[4,19,41,42]$ $[4,19,41,42]$, and presence of dynamical effects such as radial flow [[43](#page-5-3)–[49](#page-5-4)].

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](#page-5-5)], we used the model-independent universal fluctuations theory [\[51\]](#page-5-6) 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 Ref. [[52](#page-5-7)]: it is an aggregation phenomenon (''condensation of vapor''), not a frag-mentation process ("shattering of glass"). Next [[53](#page-5-8)], we studied the order parameter distributions for a wide range of data and showed that, to a first approximation, they tend toward one of two laws: the Gaussian distribution of the central limit theorem at the lowest energies, or the Gumbel distribution [\[54,](#page-5-9)[55\]](#page-5-10) of extreme value statistics [[56](#page-5-11)] 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](#page-5-12)].

In this letter we will study in more detail the transition from one regime to the other, using new data on the largest fragment distributions for multifragmentation 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 systems around a critical point.

Experimental analysis.—Collisions of $^{129}Xe + ^{nat}Sn$ were measured using the INDRA 4π charged product array [\[58\]](#page-5-13) 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 Refs. [\[53,](#page-5-8)[59\]](#page-5-14). Data used here were taken during two separate campaigns: beam energies of 25, 32, 39, 45, and 50 MeV/A were previously analyzed in Refs. [[50,](#page-5-5)[53](#page-5-8)]; 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 Ref. [\[50\]](#page-5-5), 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_{max} . We therefore look for events which maximize the quantity ($Et_{12} \times Z_{tot}$): $Et₁₂$ is the total transverse energy of light charged particles $(Z = 1, 2)$, which increases with collision centrality [\[60\]](#page-5-15), while Z_{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 $(Et_{12} \times Z_{tot})$ distribution measured with the on-line trigger condition (charged product multiplicity $M \ge 4$).

We begin by examining the scaling properties of Z_{max} fluctuations including the three new data points at 27, 29, and 35 MeV/A. Figure [1](#page-1-0) shows the correlations between the first two cumulant moments of the Z_{max} distribution $(\langle Z_{\text{max}}\rangle^2$ and $\sigma^2)$, for each beam energy. As in Fig. 1(c) of Ref. [\[50\]](#page-5-5), the data fall on two branches, $\sigma^2 \sim \langle Z_{\text{max}} \rangle^{2\Delta}$, with different values for the scaling parameter, Δ [\[51,](#page-5-6)[61\]](#page-5-16): $\Delta \sim 1$ above 32 MeV/A, and $\Delta \sim 1/2$ below 32 MeV/A.

FIG. 1 (color online). (Symbols) Log-log correlation between the first two cumulant moments ($\langle Z_{\text{max}}\rangle^2$ and σ^2) of the order parameter distribution; (Lines) linear fits performed in the range 50–32 MeV/A (resp. 32–25 MeV/A) which correspond to a slope $\Delta \sim 1$ (resp. $\Delta \sim 1/2$) (see text).

The new data points present a consistent behavior which follows the systematic scaling trend.

The order parameter distributions (largest fragment charge, Z_{max} , distributions) for four bombarding energies are presented in Figs. $2(a)-2(d)$. 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_{max} distribution at any beam energy is an admixture of a Gaussian and a Gumbel distribution,

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

with $0 < \eta < 1$ and $x = Z_{\text{max}}$. As a first test of its validity, fits to the Z_{max} distribution using Eq. [\(1](#page-1-2)) are shown in Fig. [2.](#page-1-3) 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 χ^2 values for all energies lie between approximately 4 and 10, and can further be reduced if Z_{max} distributions are smoothed to remove odd-even staggering of the yields [[62](#page-5-17)]: in that case all χ^2 values are close to 2, except for data at 32 MeV/A for which a significantly larger value (χ^2 ~ 4) is obtained. These values are significantly better than those obtained for single-component fits (as in Ref. [\[53\]](#page-5-8)).

FIG. 2 (color online). (a)–(d) (dots) Z_{max} distributions for 129 Xe + nat Sn central collisions for different beam energies; (full line) fit using Eq. [\(1\)](#page-1-2); (red dashed line) Gumbel component; (blue dotted line) Gaussian component. (e) Relative strengths of the two components, R (see text), as a function of the beam energy.

The quantity $R = 2\eta - 1$ has been defined in such a way that $R = \pm 1$ for pure Gauss or 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. $2(e)$. 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 Δ -scaling (Fig. [1](#page-1-0)).

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

First, let us consider the most well-known and widelyused model of this kind, percolation [\[63\]](#page-5-18). Indeed, percolation models have long been used in the analysis and interpretation of multifragmentation data [\[28](#page-4-20)[,64–](#page-5-19)[69](#page-5-20)]. 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_{max} , and it is known that for subcritical finite lattices, s_{max} has a Gumbel distribution [\[70\]](#page-5-21), while at criticality in the mean-field limit s_{max} follows the Kolmogorov-Smirnov (K-S) distribution [\[71\]](#page-5-22).

Figures $3(a)$ –[3\(d\)](#page-2-0) show s_{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 [\[70\]](#page-5-21), while for $p \gg p_c$ the distribution approaches a Gaussian form. In the critical region the distribution of s_{max} takes on a nontrivial 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.

Figure $3(e)$ 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_{max} [see Fig. [3\(c\)\]](#page-2-0), but is reached for $p > p_c$, not at the infinitelattice 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\)](#page-1-2).

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 [\[72\]](#page-5-23). 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 Ref. $[61]$ $[61]$ $[61]$ that for this model s_{max} exhibits the $\Delta = 1$ scaling at $t = t_c$, and $\Delta = 1/2$ scaling with a symmetric distribution for $t \gg t_c$. Figures [4\(a\)](#page-3-0)–[4\(d\)](#page-3-0) present the s_{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.](#page-2-1) Fits to these distributions using Eq. ([1](#page-1-2)) are shown, and once again they are a good approximation for s_{max} distributions both far from and around the critical region. Figure $4(e)$ 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](#page-5-24)].

FIG. 3 (color online). (a)–(d) (dots) s_{max} distributions for bond percolation on a $6 \times 6 \times 6$ lattice for different values of the bond probability, p; (full line) fit using Eq. [\(1\)](#page-1-2); (red dashed line) Gumbel component; (blue dotted line) Gaussian component. (e) Relative strengths of the two components, R (see text), as a function of p. Vertical line indicates the critical bond probability $p_c = 0.312$.

FIG. 4 (color online). (a)–(d) (dots) s_{max} distributions for irreversible aggregation in a system of size $N = 216$ for different times; (full line) fit using Eq. ([1](#page-1-2)); (red dashed line) Gumbel component; (blue dotted line) Gaussian component. (e) Relative strengths of the two components, R (see text), as a function of time with respect to t_c . Vertical line indicates critical gelation time.

Discussion.—We have shown that there is a strong similarity between Z_{max} distributions for central collisions of $129Xe + nat$ 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](#page-5-25)–[77\]](#page-5-26).

In this framework, we can infer that the Z_{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\]](#page-4-3). It has been shown that for central $^{129}Xe + \frac{nat}{5n}$ reactions the onset of significant radial expansion occurs for beam energies above [2](#page-1-3)5 MeV/A $[59]$ $[59]$ $[59]$. The similarity between Fig. 2 and [4](#page-3-1) 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 Ref. [\[53\]](#page-5-8): for ⁵⁸Ni + ⁵⁸Ni collisions, measured from 32 MeV/A to 90 MeV/A, a change of Δ -scaling and of the form of the Z_{max} distribution were observed, as for $^{129}Xe + \frac{nat}{Sn}$ but at a higher bombarding energy of 52 MeV/A; for the lighter system $36Ar + KCl$, studied from 32 MeV/A to 74 MeV/A, only the $\Delta = 1/2$ regime was observed, with quasi-Gaussian Z_{max} distributions; on the other hand, for the much heavier $197Au + 197Au$ system, at bombarding energies between 40 MeV/A and 80 MeV/A, only the $\Delta = 1$ regime occurs, with Gumbel Z_{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](#page-5-27)]. The latter increases with the mass of the colliding nuclei, as shown in Ref. [\[79](#page-5-28)]. 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 $12\overline{9}Xe + n a t$ and ¹⁹⁷Au + ¹⁹⁷Au. This explains why the Δ -scaling transition occurs at higher energy for ⁵⁸Ni + ⁵⁸Ni than for ¹²⁹Xe + nat Sn. For the very light $36Ar + KCl$ system the threshold is higher still than for $58Ni + 58Ni$, outside the range of measured bombarding energies. On the other hand, for $197Au + 197Au$ 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 MeV/A for the $^{129}Xe + nat$ Sn 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 Δ -scaling as the onset of an ''explosive'' multifragmentation regime in which initially compressed heated nuclear liquid clusterizes in the presence of significant radial expansion [\[46,](#page-5-29)[80\]](#page-5-30). The massdependence 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 the role of radial expansion in nuclear multifragmentation [\[59](#page-5-14)[,81\]](#page-5-31) and the systematic study of nuclear stopping around the Fermi energy [[79](#page-5-28)].

The authors would like to thank the staff of the GANIL Accelerator facility for their continued support during the experiments. D. G. gratefully acknowledges the financial support of the Commissariat à l'Énergie Atomique and the Conseil Régional de Basse-Normandie. This work was also supported by the Centre National de la Recherche Scientifique and by the Ministère de l'Education Nationale.

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