Universal features of the order-parameter fluctuations: Reversible and irreversible aggregation

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We discuss the universal scaling laws of order-parameter fluctuations in any system in which a second-order critical behavior can be identified. These scaling laws can be derived rigorously for equilibrium systems when combined with a finite-size scaling analysis. The relation between the order parameter, the criticality, and the scaling law of fluctuations has been established, and the connection between the scaling function and the critical exponents has been found. We give examples in out-of-equilibrium aggregation models such as the Smoluchowski kinetic equations, or at-equilibrium Ising and percolation models.

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I. INTRODUCTION

Fluctuations in many physical processes are difficult to analyze because they develop dynamically and often retain the memory of initial conditions. On the other hand, strong fluctuations are ubiquitous, as shown by examples like hadronization in a strong interaction physics, polymerization, colloid aggregation, aerosol coalescence, or the formation of large scale structures in the Universe. With the advent of recently developed advanced detection systems, the study of large fluctuations in physical observables became accessible in ''small systems'' such as formed in ultrarelativistic collisions of hadrons, leptons, and nuclei, in heavy-ion collisions at intermediate energies, or in collisions of atomic aggregates [1].

In theoretical studies, it is often assumed that fluctuations are irrelevant. In this spirit, many aggregation processes have been studied in the mean-field approximation [2]. This problem was recently revisited [3]. It was shown that, contrary to the usual belief, fluctuations in the size distribution of the largest cluster are generally large in the aggregation processes. Large fluctuations in the cluster multiplicity distribution were also reported in the binary fragmentation process with the inactivation mechanism [4].

This paper deals with features of fluctuations of physical quantities in an N-body, d-dimensional system, with N essentially *finite*. Moreover, the system is not necessarily at thermodynamic equilibrium. Both these aspects of our approach are important in many areas of physics where, for example, small and strongly fluctuating systems are produced in violent collision processes. Consequently, these systems are short lived, and the typical time scales are such that standard methods of equilibrium statistical physics may not be applicable.

We are particularly interested in self-similar systems such as fractal objects or thermodynamic systems at a second-order phase transition. ''Self-similarity'' means, in particular, that one is unable to define the characteristic length $\sim(N^*)^{1/d}$, where N^* is the characteristic size, which could be associated with the disappearance of fluctuations. Our aim in this work is to discuss universal scaling laws of fluctuations of different observables in self-similar systems. In particular, we shall consider the order-parameter fluctuations in any system, both equilibrium and nonequilibrium ones, in which the second-order critical behavior can be identified. These considerations will provide an understanding of the relation between the order parameter, the criticality, and the scaling law of fluctuations. The notion of *the relevant variable* (*the relevant observable*) for a discussion of critical behavior in finite systems will appear from this discussion.

The paper is organized as follows. In Sec. II, the order-parameter fluctuations in statistical systems are analyzed, and the relation with the finite-size scaling analysis and the Widom's hypothesis is developed in details [5] (Sec. II B). The generic features of the tail of the scaling function is addressed in Sec. II C. In Secs. II E – II H, the generalized scaling of observable quantities , the Δ scaling, is discussed, and the reasons for the deviations from the limiting cases $\Delta=1$ and 1/2 are presented.

Sections III–VI are devoted to the detailed discussion of several well known generic models, using the results and methods of analysis proposed in Sec. II. In Sec. III, the noncritical fragmentation model is discussed which exhibits the power-law cluster size distribution. Results obtained in the Potts model are discussed in Sec. IV. Section V is devoted to a discussion of the reversible aggregation, as modeled by the percolation model. Both realistic three-dimensional bond percolation and mean-field percolation on the Bethe lattice are considered. Irreversible aggregation and the fluctuation properties in the Smoluchowski kinetic model are discussed in Sec. VI. Finally, the main conclusions are given in Sec. VII [6,7].

II. ORDER-PARAMETER FLUCTUATIONS

A. Correlation function argument

Let us call m the observable under investigation. For the reason of a presentation, we shall restrict ourselves to the case where m is a scalar quantity and takes real positive values. (In fact, this restriction is not a true limitation and, generally, one can consider $|m|^2$ as well.) Fluctuations of the order parameter in thermodynamic systems are expected to have different properties at the critical point and outside of it. Far from the critical behavior, the correlations are short ranged. Fluctuations of the extensive order parameter m in this case resemble the ergodic Brownian motion of this vari-

able in its proper configuration space. Consequently, $\langle(m-\langle m\rangle)^2\rangle/\langle m\rangle$ is roughly a constant, meaning it is independent of the number of constituents in the sample. Conversely, close to the second-order transition point, the fluctuations are correlated throughout the whole system and the correlation length ξ for the infinite system becomes infinite as well. In such a system let us now define the deviation ϵ of the driven parameter from its critical value [e.g., in thermodynamical systems $\epsilon=(T-T_c)/T_c$, where T_c is the critical temperature], such that $\epsilon<0$ and $\epsilon>0$ in the ordered and disordered phases, respectively. Moreover, let us define the local value of the order parameter m and the field h conjugated to m. The following isotropic correlation function is introduced:

$$\sigma_n(\epsilon,h,\vec{r}_1,\ldots\vec{r}_{n-1})=\langle m(\vec{r}_o)m(\vec{r}_o-\vec{r}_1)\cdots m(\vec{r}_o-\vec{r}_{n-1})\rangle. \tag{1}$$

Notation $\langle\cdots\rangle$ in the above expression denotes the thermodynamic average at a given ϵ and over the position $\vec{r}_o$. For vanishing ϵ, all the length scales disappear and the correlation length ξ diverges algebraically as

$$\xi\sim\epsilon^{-\nu},$$

with a universal exponent ν, which depends only on the universality class of the transition. Moreover, the scaling description of second-order critical phenomena leads to the fundamental postulate that the thermodynamic potential G verifies

$$G(\lambda\epsilon,\lambda^{2-\alpha-\beta}h)\sim\lambda^{2-\alpha}G(\epsilon,h), \tag{2}$$

and this defines the two universal exponents α and β. Let us now go back to the correlation function σ_n defined in Eq. (1). The integral of σ_n over the $n-1$ space variables: $\vec{r}_1,\ldots,\vec{r}_{n-1}$, is equal to the nth derivative of G with respect to the field h. Hence, to be consistent with Eq. (2), we must obtain the scaling relation

$$\sigma_n(\lambda\epsilon,\lambda^{2-\alpha-\beta}h,\lambda^{-\nu}\vec{r}_1,\ldots,\lambda^{-\nu}\vec{r}_{n-1}) \sim\lambda^{n\beta}\sigma_n(\epsilon,h,\vec{r}_1,\ldots,\vec{r}_{n-1}). \tag{3}$$

For $n=1$, we recover the well-known scaling behavior of the averaged order parameter with the critical exponent β. For any integer n, setting all the space variables equal to $\vec{0}$ in the formula (3) leads to the scaling of powers of the local order parameter :

$$\langle m^n\rangle(\lambda\epsilon,\lambda^{2-\alpha-\beta}h)\sim\lambda^{n\beta}\langle m^n\rangle(\epsilon,h). \tag{4}$$

As a consequence, if we let $h=0$ and $\lambda=1/\epsilon$, one finds that the quantities $\langle m^n\rangle/\langle m\rangle^n$ are independent of ϵ, when close to the transition. This means, by the finite-size analysis, that this ratio is independent of the size N of the system at the transition point.

Let us now introduce the cumulants κ_q from the general formula for the moment expansion of the order-parameter probability distribution $P[m]$:

$$\ln\left(\sum_{m=0}^{\infty}P[m]\exp(mu)\right)=\sum_{q=0}^{\infty}\frac{u^q}{q!}\kappa_q. \tag{5}$$

Expanding the left-hand side of the above expression in the power series in u, and comparing corresponding powers on the left- and right hand-sides, one derives the relations between ordinary moments of $P[m]$ and the cumulant moments :

$$\begin{aligned}\kappa_1&=\langle m\rangle,\\ \kappa_2&=\langle m^2\rangle-\langle m\rangle^2,\\ \kappa_3&=\langle m^3\rangle-3\langle m^2\rangle\langle m\rangle+2\langle m\rangle^3,\\ \kappa_4&=\langle m^4\rangle-4\langle m^3\rangle\langle m\rangle-3\langle m^2\rangle^2\\ &\quad+12\langle m^2\rangle\langle m\rangle^2-6\langle m\rangle^4,\ldots\end{aligned} \tag{6}$$

In the case of a second-order phase transition, as a result of scaling relations (4), all cumulant moments scale like

$$\kappa_q\sim\langle m\rangle^q.$$

Consequently, the generating function of the m-probability distribution [Eq. (5)] is only a function of the reduced variable $\langle m\rangle u$. This can be written as

$$P[m]=\frac{1}{2\pi}\int_0^{2\pi}\hat{G}(iu)\exp(-imu)du, \tag{7}$$

where $\hat{G}$ is the generating function :

$$\hat{G}(u)=\sum_{m=0}^{\infty}P[m]\exp(mu).$$

In the case when $\langle m\rangle$ tends to ∞ but $m/\langle m\rangle$ remains finite, we can rewrite formula (7) in a more compact form,

$$\langle m\rangle P[m]=\Phi\left(\frac{m}{\langle m\rangle}\right), \tag{8}$$

which is valid at the critical point. Φ is the scaling function of the single reduced variable $m/\langle m\rangle$. As stated above, we can express this scaling as

$$\sum_{m=0}^{\infty}P_N[m]\exp(mu)=\Psi(\langle m\rangle u), \tag{9}$$

which is the necessary and sufficient condition for the applicability of scaling law (8) [8]. This result also implies that if this scaling occurs for fluctuations of the parameter m, then it holds also for fluctuations of any power

$$X=N^a m^b$$

of this parameter as well. This is a consequence of Eq. (4) and

$$P_N[X]dX=P_N[m]dm.$$

Up to now, we did not specify reasons for changing $\langle m\rangle$. Indeed, under the condition that the scaling framework of the second-order phase transition holds, the scaling relation (8) is valid independently of the explicit reasons for changing $\langle m\rangle$, and independently of any phenomenological details. In

other words, an explicit relation between the size N of the system and $\langle m \rangle$ need not be known at this stage. In Sec. II B we shall show how to derive supplementary information about Φ when the system is at a pseudocritical point.

B. Widom's hypothesis and the finite-size scaling argument

The hypothesis of Widom states that, in the thermodynamic limit of a system at thermal equilibrium, the free energy density close to the critical point scales as [5]

$$f(\lambda^{\beta}\eta, \lambda\epsilon) \sim \lambda^{2-\alpha} f(\eta, \epsilon), \tag{10}$$

where α, and β are the usual critical exponents, η is the intensive order parameter, and λ is the scale parameter. Even though finite systems do not exhibit critical behavior, their properties may nevertheless resemble those of infinite systems if the correlation length ξ is larger than or comparable to the typical length L of the system. In this case, one speaks about the pseudocritical point in a finite system at a distance

$$\epsilon \sim c N^{-1/\nu d} \tag{11}$$

from the true critical point [9]. The quantity N in Eq. (11) is the size of the d-dimensional system, and c is some dimensionless constant which can be either positive or negative. This constant c is negative if a maximum of the finite-size susceptibility or of any other divergent macroscopic quantity in the thermodynamic limit lies in an ordered phase, while c is positive if this maximum is in a disordered phase. One can then derive the finite-size scaling of the total free energy

$$F(\eta, \epsilon, N) = N f(\eta, \epsilon)$$

at the pseudocritical point

$$F_c(\eta, N) \sim f(\eta N^{\beta/(2-\alpha)}). \tag{12}$$

In deriving Eq. (12), we used the hyperscaling relation

$$2 - \alpha = \nu d.$$

The canonical probability density of the order parameter $P_N[\eta]$ is given by [10]

$$P_N[\eta] = \frac{1}{Z_N} \exp[-\beta_T F(\eta, \epsilon, N)], \tag{13}$$

where the coefficient $\beta_T (\equiv 1/T)$ is independent of η (T is the temperature of the system). Using Eq. (13), one may calculate the most probable value of the order parameter, which is the solution to the equation

$$\frac{\partial P_N[\eta]}{\partial \eta} = 0,$$

as well as the average value of the order parameter and the partition function

$$Z_N \sim N^{-\beta/(2-\alpha)} \sim \langle \eta \rangle \sim \eta^*. \tag{14}$$

η^* in Eq. (14) denotes the most probable value of the order parameter. The average value of the order parameter vanishes for large values of N, since both β and $2 - \alpha = 2\beta + \gamma$ are positive. The probability density $P_N[\eta]$ then obeys the scaling law, which is formally identical to Eq. (8),

$$\langle \eta \rangle P_N[\eta] = \Phi\left(\frac{\eta}{\langle \eta \rangle} \right) \equiv \Phi(z), \tag{15}$$

where, in addition,

$$\Phi(z) \sim \exp[-\beta_T f(az, c)]. \tag{16}$$

In the above formula, we have omitted the temperature-dependent multiplicative factor which can be determined by the normalization of $P_N[\eta]$. Coefficients a and c may both depend on β_T. We can then rewrite scaling (15) in a standard form for the extensive order parameter $m = N\eta$,

$$\langle m \rangle P_N[m] = \Phi(z_{(1)}), \tag{17}$$

with the scaling variable $z_{(1)}$ defined by

$$z_{(1)} = \frac{m - m^*}{\langle m \rangle}. \tag{18}$$

m^* denotes the most probable value of the extensive order parameter. We call Eqs. (17) and (18) *the first-scaling law*. The scaling domain is defined by this asymptotic behavior of $P_N[m]$ when $m \to \infty$ and $\langle m \rangle \to \infty$, but $z_{(1)}$ has a finite value. The normalization of the probability distribution $P_N[\eta]$ and the definition of the average value of m provide the two constraints

$$\lim \int_{-m^*/\langle m \rangle}^{\infty} \Phi(z_{(1)}) dz_{(1)} = 1,$$

$$\lim \int_{-m^*/\langle m \rangle}^{\infty} z_{(1)} \Phi(z_{(1)}) dz_{(1)} = 0.$$

The first-scaling law [Eq. (15)] is a consequence of the self-similarity of the statistical system. Self-similarity here means that the fluctuations of the *reduced* order parameter $\eta/\langle \eta \rangle$ at different scales characterized by different values of the intensive order parameter $\langle \eta \rangle$, have *identical properties*. This is a qualitative explanation for this scaling.

The logarithm of scaling function (16) corresponds to the noncritical free energy density at the renormalized distance $\epsilon = c$ from the critical point. If it happens that the order parameter is related to the number of fragments, as in the fragmentation-inactivation-binary (FIB) process [11,12], then Eq. (15) can be written in an equivalent form to Koba-Nielsen-Olesen (KNO) scaling [13], proposed some time ago as an ultimate symmetry of the S matrix in relativistic field theory [14] . The multiplicity distribution of produced particles has been intensely studied in strong interaction physics, where the simple behavior of much of the data on the hadron-multiplicity distribution seems to point to some universality which is independent of the particular dynamical process.

If, instead of a real positive scalar, the parameter under investigation is a vector of dimensionality n, $\vec{m} = [m_1, \ldots, m_n]$, then the first-scaling law [Eqs. (17) and (18)] takes a more general form :

$$\langle|\vec{m}|\rangle^n P_N[\vec{m}]=\Phi(\vec{z}_{(1)}), \tag{19}$$

with

$$\vec{z}_{(1)}=\frac{\vec{m}-\vec{m}^*}{\langle|\vec{m}|\rangle}. \tag{20}$$

The scaling limit in Eq. (19) is defined by the asymptotic behavior of $P_N[\vec{m}]$ when $m_i\to\infty$ $(i=1,\ldots,N)$ and $\langle|\vec{m}|\rangle\to\infty$, but $z_{i(1)}$ $(i=1,\ldots,N)$ have finite values.

C. Tail of the scaling function

The scaling function Φ introduced in the first-scaling law (17) has some typical features which are reminiscent of a non-Gaussian critical distribution of the order parameter. In this section, we are interested in the behavior of the scaling function for large values of the reduced parameter $m/\langle m\rangle$, so we have to study the system subject to a small field h conjugated to the order parameter. This breaks the symmetry of the distribution by shifting m and $\langle m\rangle$ toward larger values. More precisely, let us write the probability of obtaining the value η of an intensive order parameter at a distance ϵ from the critical point as

$$P_N[\eta,\epsilon,h]=P_N[\eta,\epsilon,0]\exp(\beta_T\eta Nh). \tag{21}$$

up to now, we have studied the behavior of $P_N[\eta,\epsilon,0]$ for which the first-scaling law holds when $\epsilon=0$ (the critical point) or $\epsilon=cN^{-1/\nu d}$ (the pseudocritical point). Substituting Eq. (15) and using Eq. (14), we obtain

$$P_N[\eta,0,h]=N^{\beta/(2-\alpha)}\exp[\ln\Phi(\eta N^{\beta/(2-\alpha)})+\beta_T\eta Nh].$$

The most probable value

$$\eta^*\sim h^{1/\delta}$$

of the order parameter, in the limit of a small external field h, is given by the maximum of the term in the exponential. Since

$$\delta=\frac{2-\alpha-\beta}{\beta},$$

we obtain

$$\ln\Phi(h^{1/\delta}N^{1/(\delta+1)})\sim-\beta_T h^{1/\delta}Nh. \tag{22}$$

Relation (22) is valid for any value of N if and only if

$$\Phi(z)\sim\exp(-az^{\delta+1})\equiv\exp(-az^{\hat{\nu}}), \tag{23}$$

with the coefficient a which depends on the temperature regularly.

One can express this relation in a different way. The anomalous dimension for an extensive quantity $m=N\eta$ can be defined as

$$g=\lim_{N\to\infty}g_N=\lim_{N\to\infty}\frac{d}{d\ln N}(\ln\langle m\rangle). \tag{24}$$

One can see that due to both Eq. (14) and the Rushbrooke relation between critical exponents,

$$\alpha+2\beta+\gamma=2,$$

the anomalous dimension is

$$g=1-\frac{\beta}{\gamma+2\beta}. \tag{25}$$

Since both α and β are positive, g is contained between 1/2 and 1 for equilibrium systems at the critical point of the second-order phase transition . Because of these additional relations between critical exponents, one may note that the behavior of a tail of scaling function (23) is governed by the exponent

$$\hat{\nu}=\frac{1}{1-g}=\delta+1=\frac{2-\alpha}{\beta}, \tag{26}$$

which is always larger than 2. The limiting case $\hat{\nu}=2$, i.e., the Gaussian tail [see Eq. (23)], corresponds in this framework to the noncritical system.

Finally, let us mention in passing that whenever cluster size can be defined in a system exhibiting the second-order phase transition, as e.g., in the cases of percolation, the Ising model, or the Fisher droplet model, the exponent τ of the power-law cluster-size distribution

$$n_s\sim s^{-\tau}$$

satisfies additional relations [15]:

$$\gamma+\beta=\frac{1}{\sigma},$$

$$\gamma+2\beta=\frac{\tau-1}{\sigma},$$

which yields

$$g=\frac{1}{\tau-1} \tag{27}$$

and

$$\hat{\nu}=\frac{\tau-1}{\tau-2} \tag{28}$$

Since g at the second-order equilibrium phase transition is contained between 1/2 and 1, the allowed values of exponent τ at the critical point are $2<\tau<3$, and the normalized cluster-size distribution

$$\sum_{s=1}^{N}sn_s=N$$

is

$$n_s=Ns^{-\tau}.$$

Consequently, whenever τ is defined, we obtain whether the studied equilibrium system is at a second-order phase transi-

tion and whether the considered extensive quantity can be identified with the order parameter of this transition.

Let us define, for example, the multiplicity as the total number of clusters. (The definition of clusters also includes the monomers.) The cluster multiplicity cannot be an order parameter of these kind of equilibrium phase transitions because with $2<\tau<3$,

$$\sum_{s=1}^{N} n_s \sim N,$$

which means that the average multiplicity scales as the total mass of the system at the transition point, i.e., $g=1$. On the other hand, the size of the largest cluster is a natural order parameter for these kinds of phase transitions. In this case we have

$$\langle s_{max}\rangle \sim N^{1/(\tau-1)}, \tag{29}$$

which is a direct consequence of

$$\sum_{s=\langle s_{max}\rangle}^{N} n_s \sim 1,$$

i.e., that there is in the average only one largest cluster. Moreover, relation (27) derived for the second-order critical phenomenon is correctly recovered. One should emphasize, that relation (29) is very general, and its derivation does not depend on the assumption of thermodynamic equilibrium. In other words, relation (29) between the anomalous dimension and the exponent τ, is also valid for the off-equilibrium second-order phase transitions. We shall return to this point in Sec. VI.

The cluster multiplicity could be the order parameter whenever $\tau<2$, though this cannot occur in equilibrium phase transitions. Note that this argument, to sort among different candidates for the order parameter, requires only a knowledge of τ, i.e., the complete information about the critical process is superfluous. We shall use this argument later in the case of percolation model and Smoluchowski model of gelation. Finally, we shall see below in the Mekjian model that we may have a power-law size distribution with $\tau<2$, in the absence of a phase transition governed by the multiplicity, as the order parameter.

D. Landau-Ginzburg theory of phase transitions

Let us consider the Landau-Ginzburg (LG) theory as an exactly solvable example of a second-order phase transition. The homogeneous LG free energy density is

$$f(\eta)=\epsilon\eta^2+b\eta^4+\cdots,$$

where b is a positive constant. The most probable value of the order parameter η in the disordered phase ($\epsilon>0$) is implicitly set to 0. It is more convenient to work with the extensive order parameter $m=N\eta$ when dealing with finite systems. The probability of a state m for a given ϵ is [10]

$$P_N[m]=\frac{1}{Z_N}\exp\left[-\beta_T\left(\epsilon\frac{m^2}{N}+b\frac{m^4}{N^3}-\cdots\right)\right]. \tag{30}$$

Z_N is defined by the normalization of $P_N[m]$. To remain consistent with other sections of this paper and without loss of generality, we now consider the case where m is positive. We will admit that N is so large that the first two terms in the free energy expansion are sufficient to study the phase transition. At the critical point $\epsilon=0$, the leading term of the free energy density is proportional to m^4. Standard integrations yield the values for the partition function Z_N and the average value of the order parameter $\langle m\rangle$, both proportional to $N^{3/4}$. Introducing there into Eq. (30), one finds

$$\langle m\rangle P_N[m]=\frac{4\sqrt{\pi}}{\Gamma^2[1/4]}\exp\left[-\frac{\pi^2}{\Gamma^4[1/4]}\left(\frac{m}{\langle m\rangle}\right)^4\right], \tag{31}$$

which has the form of Eq. (8). Note that the scaling function $\Phi(z)\sim\exp(-z^4)$ decreases very quickly as one moves away from the most probable value. This result is consistent with the analysis done in Sec. II C.

The pseudocritical point is the value of ϵ for which the finite-size thermal susceptibility reaches its maximum. Writing that the inverse of this susceptibility is the second derivative of the free energy with respect to the order parameter, one finds

$$\epsilon=-6\frac{\Gamma[3/4]}{\Gamma[1/4]}\left(\frac{b}{\beta_T N}\right)^{1/2}. \tag{32}$$

This result is correct at first order in $N^{-1/2}$. Replacing ϵ in Eq. (30) by Eq. (32) leads to the scaling form of $P_N[m]$:

$$\langle m\rangle P_N[m]=A\exp\left[-\frac{\Gamma[3/4]^2}{\Gamma[1/4]^2}((m/\langle m\rangle)^4-6(m/\langle m\rangle)^2)\right], \tag{33}$$

where A denotes a normalization constant. We recover indeed the first-scaling law, with the exponential tail $\exp(-az^4)$, for the large arguments.

Outside of the critical point in the disordered phase ($\epsilon>0$), the leading term of the free energy is proportional to m^2, and the probability distribution $P_N[m]$ is essentially Gaussian. Deriving, as previously, the values of Z_N and $\langle m\rangle$ (both behave like $N^{1/2}$ in this case), we obtain the scaling form

$$\langle m\rangle P_N[m]=\frac{4}{\pi}\exp\left[-\frac{4}{\pi}\left(\frac{m}{\langle m\rangle}\right)^2\right], \tag{34}$$

which is still under the form of Eq. (15) but with a Gaussian scaling function which is reminiscent of the Gaussian fluctuations.

Finally, in the low temperature regime ($\epsilon<0$), the most probable value of the order parameter is positive:

$$m^*=\sqrt{-\frac{\epsilon}{2b}}N.$$

Developing $P_N[m]$ in Eq. (30) around this point leads to the expression

$$m^{*1/2}P_N[m] \simeq \left(-2\frac{\epsilon^3}{b\pi^2}\right)^{1/4} \exp\left(\epsilon\sqrt{-2\frac{\epsilon}{b}}\frac{(m-m^*)^2}{m^*}\right), \tag{35}$$

which is no longer in the standard form [Eq. (15)]. In this case, the average value of the order parameter $\langle m\rangle$ is of the same order of magnitude as its most probable value m^*, and one can rewrite Eq. (35) in the scaling form

$$\langle m\rangle^{1/2}P_N[m] \sim \exp\left(-a\frac{(m-m^*)^2}{\langle m\rangle}\right), \tag{36}$$

where a is a positive constant. This particular scaling form will be discussed later in detail.

E. Δ-scaling law

One may ask what happens if the observable quantity is not the order parameter but an N-dependent function of the order parameter like

$$m = N^{a_1} - N^{a_2}\eta, \tag{37}$$

where

$$a_1 > g + a_2 - 1. \tag{38}$$

The latter condition ensures that the order parameter does not determine the leading behavior of m. For large N,

$$\langle m\rangle \sim N^{a_1}.$$

Writing Eq. (17) with m instead of η, and taking into account that

$$P_N[\eta]d\eta = P_N[m]dm,$$

one finds the generalized law:

$$\langle m\rangle^{\Delta}P_N[m] = \Phi(z_{(\Delta)}) \equiv \Phi\left(\frac{m-m^*}{\langle m\rangle^{\Delta}}\right), \tag{39}$$

where

$$\Delta = \frac{g + a_2 - 1}{a_1} < 1.$$

This generalized law will be called in *the* Δ*-scaling law* what follows. The scaling function $\Phi(z_{(\Delta)})$ depends only on one scaled variable:

$$z_{(\Delta)} = \frac{m-m^*}{\langle m\rangle^{\Delta}}. \tag{40}$$

The normalization of the probability distribution $P_N[m]$ and the definition of the average value of m provide two constraints

$$\lim \int_{-\langle m\rangle^{1-\Delta}}^{\infty} \Phi(z_\Delta)dz_\Delta = 1,$$

$$\lim \int_{-\langle m\rangle^{1-\Delta}}^{\infty} z_\Delta\Phi(z_\Delta)dz_\Delta = 0,$$

which are consistent with $\Delta \leqslant 1$, because the scaling function Φ is positive. The scaling function $\Phi(z_{(\Delta)})$ in Eq. (39) has a form identical to $\Phi(z_{(1)})$, except for the inversion of the abscissa axis. In particular, its tail for large $z_{(\Delta)}$ has the same form

$$\Phi(z_{(\Delta)}) \sim \exp(-z_{(\Delta)}^{\hat{\nu}}) = \exp(-z_{(\Delta)}^{1/(1-g)}) \tag{41}$$

as given in Eq. (23). One should mention in passing that if

$$a_1 < g + a_2 - 1$$

in Eq. (37), then

$$\langle m\rangle \sim N^{a_2}\langle\eta\rangle$$

and $\Delta = 1$, following the remark of Sec. II A.

According to Eq. (16), the logarithm of scaling function $\Phi(z_{(\Delta)})$,

$$\ln\Phi(z_{(\Delta)}) = -\beta_T f(az_{(\Delta)}, c),$$

is related to the *noncritical* free energy f, in either ordered ($c>0$) or disordered ($c<0$) phases.

As an important example, we see from Eqs. (37) and (39) that the Δ scaling of the extensive variable

$$\hat{m} = N(1-\eta) \equiv N\hat{\eta}$$

can be used to determine the anomalous dimension, since in this case $\Delta = g$. For this reason, $\hat{m}$ is a very useful variable in all phenomenological studies. The importance of $\hat{m}$ in the percolation studies is well established [20]. At the phase transition,

$$\langle N\hat{\eta}\rangle \sim N,$$

but the finite-size corrections are *algebraic*.

F. Off-critical scaling

$\Delta = 1/2$, with a nearly Gaussian function $\Phi(z_{(\Delta)})$, is a particular case of a Δ scaling associated with noncritical systems [16]. This limit,

$$\langle m\rangle^{1/2}P_N[m] = \Phi\left(\frac{m-m^*}{\langle m\rangle^{1/2}}\right) \equiv \Phi(z_{(1/2)}), \tag{42}$$

which is called *the second-scaling law*, has been found in the shattering phase of the non-equilibrium FIB process [8] and in the ''gaseous'' phase of the equilibrium percolation process [16]. We should also recall that this form of scaling function has been seen for a LG model in the low-temperature regime [see Eq. (36)].

More generally, let us now suppose that the extensive parameter m is not critical, i.e., either the system is in a critical state but the parameter m is not critical, or the system is outside the critical region. The value of m at the equilibrium is obtained by minimizing the free energy. The free energy F is analytical in the variable m close to its most probable value m^*:

$$F \sim N^{-1}(m-m^*)^2. \tag{43}$$

Using Eq. (43), one obtains

$$\langle m\rangle \sim \mu^* N,$$

where μ^* is a positive (finite) number which is independent of N, and

$$Z_N \sim N^{1/2} \sim \langle m\rangle^{1/2}. \tag{44}$$

The probability density $P_N[m]$ verifies the second-scaling law [Eq. (39)]:

$$\langle m\rangle^{1/2} P_N[m] = \exp\left[-\beta_T \mu^* \left(\frac{m-\langle m\rangle}{\langle m\rangle^{1/2}} \right)^2 \right] \equiv \Phi(z_{(1/2)}). \tag{45}$$

This is a particular case of the Δ-scaling law ($\Delta = 1/2$) and the scaling function is now Gaussian [17]. This scaling [Eq. (42)] holds for $\langle m\rangle \sim N$, but now with *exponential* finite-size corrections. This is a principal difference from the finite-size corrections and/or the Δ scaling. The above arguments apply to any second-order phase transition. In particular, they are not limited to the LG theory of phase transitions [see Eq. (36)].

G. Finite-size crossover effects

The discussion of Sec. II F is valid for systems at a critical (and pseudocritical) point, or far from a critical point in an ordered phase. Let us suppose now that the system is prepared such that

$$\langle m\rangle \sim N^{g'}, \quad g' < 1$$

and g' is not an anomalous exponent. Here we would like to study how the finite system evolves when the control parameter ϵ tends slowly to 0, namely,

$$\epsilon \sim N^{2g'-2}.$$

We shall address this question in the mean-field approximation using the LG theory. Let us first write down the average value

$$\langle m\rangle = \frac{\int_0^\infty m \exp(-\epsilon m^2/N - b m^4/N^3)\, dm}{\int_0^\infty \exp(-\epsilon m^2/N - b m^4/N^3)\, dm}. \tag{46}$$

Hence, writing this definition with the new driving parameter $\epsilon' = \epsilon N^{1/2}$ and using the rescaled variable $m' = m/N^{3/4}$, the average value of m can be put into the form

$$\langle m\rangle = N^{3/4} \psi(\epsilon N^{1/2}),$$

while its most probable value is

$$m^* = \sqrt{-\frac{\epsilon}{2b}}\, N.$$

If the exponent g' is not too small, i.e., if ϵ does not vanish too quickly, the two quantities: $\langle m\rangle$ and m^* have to coincide. This is because the exponential weight term in Eq. (46) diverges as $\sim \exp(\epsilon^2 N/4b)$ when

$$\epsilon N^{1/2} \sim N^{(4g'-3)/2} \tag{47}$$

becomes large with increasing N. As a consequence, the common behavior of $\langle m\rangle$ and m^* is $\sim N^{g'}$. The scaling form [Eq. (35)] in this case is

$$\langle m\rangle^{\Delta} P_N[m] \sim \exp\left(-c \frac{(m-m^*)^2}{\langle m\rangle^{2\Delta}} \right), \tag{48}$$

with c a positive constant, and

$$\Delta = \frac{3}{2} g' - 1.$$

Here we recover the two cases previously discussed in Sec. II B. When $g' = 1$, i.e., when $\epsilon = \text{const}$, then this is the second-scaling law. When $g' = 3/4$, then this is the first-scaling law, since the finite system is still in the critical region ($g' = g$). In between these two limiting cases, Δ scaling holds, with $1/2 < \Delta < 1$. Note also that the scaling function in Eq. (48) has a Gaussian form, even for $\Delta > 1/2$, which is quite different from the case [Eq. (23)] of Sec. II C.

H. Summary: panorama of the Δ-scaling for thermodynamic systems

Several features of finite systems are important if one wants to study either the criticality of the corresponding infinite system or the distance from the critical point. Here one should point out the Δ scaling (this includes the first-scaling law $\Delta = 1$ as well), the form of the tail of the scaling function Φ, and the anomalous exponent. All these features are closely related to the properties of the scaling function which characterizes the finite system at the equilibrium. If the infinite system experiences a second-order phase transition, and if m is the scalar order parameter or the shifted scalar order parameter [Eq. (37)], then:

At the critical point, the corresponding finite system exhibits the first-scaling law if m is an order parameter, or the Δ-scaling law if m is a shifted order parameter. In both cases, the tail of the scaling function $\sim \exp(-z^{\hat{\nu}})$ is characterized by a large value of the exponent $\hat{\nu} = 1/(1-g) > 2$, with g being the anomalous exponent, i.e., the exponent characterizing the decrease of the extensive order parameter with the size N of the finite system. The values of Δ are restricted to $0 < \Delta \leqslant 1$, and the anomalous exponent g takes values in between 1/2 and 1 for a second-order at-equilibrium phase transition.

Far from the critical point, finite system exhibits the second-scaling law with the Gaussian tail of the scaling function.

Close to the critical point, when $\epsilon \to 0$ if $N \to \infty$, the finite system exhibits a crossover phenomenon from the first-scaling law to the second-scaling law by the continuous Δ-scaling law with a Gaussian shape of the scaling function. One should remember here that the precise dependence of $\epsilon(N)$ is irrelevant provided that $g < g' < 1$, i.e., that the conditional point does not approach 0 move quickly than the pseudocritical point. This last remark is important in phe-

nomenological applications of scaling theory to situations where the N dependence of the conditional point is governed by the external control parameter, with an unknown relation to the system size. Last but not least, if the parameter m is not singular at the transition, then all properties of its probability distribution are the same as in the case of noncritical systems.

In phenomenological applications, it is often difficult to obtain the probability distribution with sufficient accuracy for values of scaling variable which are far from the most probable value, since this corresponds to very small probabilities. It is then more judicious to work with moments of the distribution instead of with the distribution itself. For example, when the system undergoes Δ scaling, the properly normalized cumulant moments [Eq. (6)]

$$\frac{\kappa_q}{(\kappa_1)^{q\Delta}} \sim \text{const} \tag{49}$$

are independent of the size of the system. An important consequence is that the generating function of the m distribution, $\hat{G}(u)=\Sigma P[m]\exp(mu)$, is a function of the reduced variable $\langle m\rangle^{\Delta}u$ only, generalizing a remark of Sec. II A for the generating function in the first-scaling case.

III. A NONCRITICAL MODEL: MEKJIAN MODEL

The Mekjian fragmentation model is an equilibrium model which describes the decomposition of system into an ensemble of fragments. The statistical weights for every configuration of fragments are given explicitly in this model. If n_s denotes the number of fragments of size s with the size conservation, $N=\Sigma_s s n_s$, the weight function for the configuration $\{n_s\}$ is given by [18]:

$$W_N(\{n_s\},x)=\prod_{s=1}^{N}\frac{s x^{n_s}}{n_s! s^{n_s}(x+s-1)},$$

with x a real control parameter. Many exact results can be obtained in this simple model. Here we are interested in the multiplicity distribution $P_N[m]$, where the fragment multiplicity is $m=\Sigma_s n_s$. We can show that [19]:

$$P_N[m]=x^m|S_N^{(m)}|\frac{\Gamma(x)}{\Gamma(N+x)},$$

where $|S_N^{(m)}|$ are signless Stirling numbers of the first kind. Then knowing the generating function for these Stirling numbers,

$$\sum_{m=0}^{\infty}P_N[m]e^{mu}=\frac{\Gamma(x)\Gamma(xe^u+N)}{\Gamma(xe^u)\Gamma(x+N)}, \tag{50}$$

one obtains the average value of m:

$$\langle m\rangle = x\sum_{s=1}^{N}\frac{1}{x+s}=x\ln N+(x-1)\gamma-\psi(x)+O(1/N).$$

Moreover, making an asymptotic development of Eq. (50) for large N and small s, one obtains

$$\sum_{m=0}^{\infty}P_N[m]e^{mu}\simeq N^{x(e^u-1)}. \tag{51}$$

The latter approximation is known to be correct for finite values of u [20]. This means that $P_N[m]$ is approximately a Poisson m distribution with parameter $x\ln N$. In the leading order we then have $\langle m\rangle\simeq m^*$. Inverting Eq. (50) to obtain $P_N[m]$ as a Fourier transform, and making N large, yields the scaling formula

$$\langle m\rangle^{1/2}P_N[m]=\frac{1}{\sqrt{\pi}}\exp\left[-\frac{(m-\langle m\rangle)^2}{2\langle m\rangle}-(x-1)[\gamma-\psi(x)]\frac{m-\langle m\rangle}{\langle m\rangle}+O\left(\frac{1}{\langle m\rangle}\right)\right]. \tag{52}$$

This is nothing else but the second-scaling law (42) for the multiplicity distribution when N becomes large enough, because $\langle m\rangle\simeq m^*$. When $\langle m\rangle$ is large enough, the second term in Eq. (52) is always very small compared to the first one for a finite x [21].

Different fixed values of the control parameter x mimic different situations of the fragmentation. For $x\ll 1$, one has the situation of a fused system. For $x\sim 0.5$, the fragmentation resembles the evaporation of light fragments. The limit $x\gg 1$ corresponds to a complete dissociation of the mass into light fragments (monomers). Each of these situations is characterized by a different fragment-size distribution. The case $x=1$ is particular in this model, since it leads to a power-law size distribution [Eq. (27)] with an exponent $\tau=1$. Following discussion in Sec. II C, the cluster multiplicity could be the order parameter. On the other hand, the second-order equilibrium phase transition is associated with $2<\tau<3$, which implies that the equilibrium model of Mekjian is a noncritical model. Indeed, that is what can also be seen in the cluster-multiplicity scaling law [Eq.(52)]. Hence the power-law cluster-size distribution alone does not guarantee that the system exhibits a critical behavior of any kind [12].

IV. EXAMPLE: POTTS MODEL

A generalization of the magnetic Ising spin model was proposed by Domb [22], and studied in details by Potts [23]. In this model, one considers a system of N sites in d-dimensional space. The magnetic state of each site i is characterized by a quantity called a spin (say, s_i). Each spin is of the same constant modulus, and points to one of the q equally spaced directions, labeled from 0 to $q-1$. The ferromagnetic short-ranged Potts Hamiltonian is then

$$H_q=-J\sum_{i,j}\delta(s_i,s_j), \tag{53}$$

where δ is the Kronecker symbol, and J is the positive coupling constant. The sum is restricted to the nearest-neighbor pairs. The site percolation corresponds to the $q=1$ Potts model, and the ferromagnetic Ising model to the $q=2$ case. This model is one of the simplest nontrivial critical thermodynamic N-body systems, and many exact or accurate results

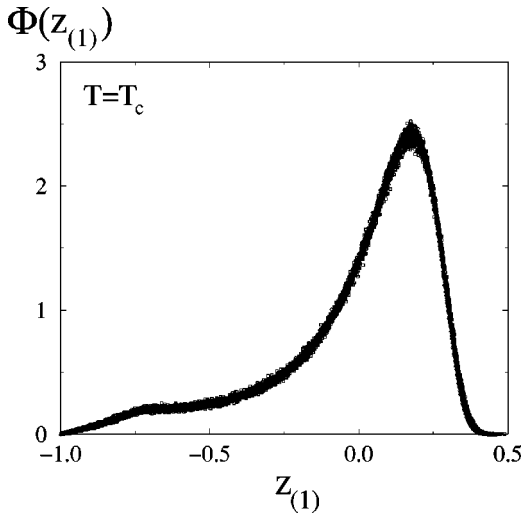

FIG. 1. Scaled m distribution for the three-state Potts model ($q=3$) on a two-dimensional $L\times L$ square lattice, at a critical temperature. Three sizes are shown: $L=32$ (stars), $L=48$ (circles), and $L=64$ (squares). The thermalization is achieved after 4×10^6 Monte Carlo steps, starting from the initial disordered system.

are known for standard values of (d,q). In particular, there exists a value $q_c(d)$ [for example, $q_c(2)=4$], for which, when $q\leqslant q_c(d)$, such an interacting system experiences a second-order phase transition at a finite critical temperature [for example, $\beta_c J=\ln(1+\sqrt{q})$ at $d=2$], while for $q>q_c(d)$ the transition is a first-order one.

Here let us consider the case of a second order phase transition. All the scalings described above should hold. We first have to define the order parameter for the system. If, for a given configuration of the system, we call N_k the number of sites in the state k, where k varies from 0 to $q-1$, then the order parameter m is given by

$$m=\frac{q(N_{max}/N)-1}{q-1}. \tag{54}$$

N_{max} in Eq. (54) is defined as the maximum of all N_k's. Figure 1 shows the m distribution at the critical temperature in the $(d,q)=(2,3)$ case, in the first-scaling form. The scaling is recovered very precisely, even for such small system sizes such as 64×64. Note also the complicated shape of the scaling curve [24,25].

We can discuss this scaling here in the slightly different context of correlated variables [26]. Let us consider, for simplicity, the Ising model [i.e., $(d,q)=(d,2)$ case]. The extensive order parameter is just the sum of N *correlated* variables: $M=\Sigma s_i$. When the system is disordered, the spins are correlated at a short distance ξ ($\xi/N\to0$ at the thermodynamic limit), and their mean value is zero. The central limit theorem tells then that the distribution of the random variable $M/\sqrt{N}$ is Gaussian when N becomes large, with zero mean and finite variance. This can also be expressed by the asymptotic law

$$\langle M^2\rangle P[M^2]=\sqrt{\frac{\langle M^2\rangle}{2\pi M^2}}\exp\left(-\frac{M^2}{2\langle M^2\rangle}\right), \tag{55}$$

which is correct in the first-scaling form, with a Gaussian shape and the trivial anomalous exponent $g=1/2$ [see Eq. (34)].

On the other hand, if the system is in the ordered phase, the average value of the individual spins is finite, say $\langle s_i\rangle=m$, and the same reasoning can be used for the variable $(M-Nm)/\sqrt{N}$. This variable is of zero mean and finite variance, and is short range correlated. Thus, its fluctuations are Gaussian, and can be put in a second-scaling form:

$$\langle M\rangle^{1/2}P[M]=\frac{1}{\sqrt{2\pi}}\exp\left(-\frac{(M-\langle M\rangle)^2}{2\langle M\rangle}\right). \tag{56}$$

Of course, the most interesting case corresponds to the critical temperature. At this point, the spins are correlated throughout the system, and the magnetization cannot be evaluated by the central limit theorem. Instead, we can remark that the spin-correlations are a power law,

$$\langle s_{\vec{r}_o}s_{\vec{r}+\vec{r}_o}\rangle\sim\frac{1}{r^{d-2+\eta}},$$

with η a critical exponent whose value should be between $2-d$ and 2. Looking at the total magnetization as the sum of N correlated variables, one obtains

$$\langle M^2\rangle=\sum_{i,j}\langle s_is_j\rangle=\sum_i\langle s_i^2\rangle+N\int_1^L\langle s_{\vec{0}}s_{\vec{r}}\rangle r^{d-1}dr, \tag{57}$$

with $L\sim N^{1/d}$ the typical macroscopic length of the system. This means that

$$\langle M^2\rangle\sim N^{1+[(2-\eta)/d]}$$

as the leading behavior. This nontrivial anomalous exponent

$$g=\frac{1}{2}+\frac{2-\eta}{2d},$$

between 1/2 and 1, is here the sign of the criticality. The first-scaling law should hold in this case, as for the $(d,q)=(2,3)$ Potts model discussed above, but the scaling function should be different since it depends on the precise form of the interactions. Only the tail can be linked to another critical exponent, as it has been written in Sec. II C.

V. REVERSIBLE AGGREGATION PROCESS-PERCOLATION MODEL

The percolation model can be defined as follows. In a box (a part of the regular lattice), each site corresponds to a monomer, and a proportion p of active bonds is set randomly between sites (the bond percolation model). Such a network results in a distribution of clusters defined as an ensemble of occupied sites connected by active bonds. For a definite value of p, say p_{cr}, a giant cluster almost surely spans the whole box. The sol-gel transition corresponds to the appearance of ''infinite'' cluster (gel) at a finite time. Infinite in this context means that the gel contains a finite fraction of the total mass of the system. The sol-gel transition in finite systems can be suitably studied using moments of the number-

size distribution n_s, i.e., the number of finite clusters of a size s,

$$M_k' = \sum_{s<s_{max}} s^k n_s, \tag{58}$$

where the summation is performed over all clusters with the exception of the largest cluster $s \equiv s_{max}$. The superscript $'$ recalls this constraint on summation in Eq. (58). The mass of the largest cluster is then $N - M_1'$, with

$$N = \sum_{all\ s} s n_s.$$

In infinite systems, one works with the normalized moments of the concentration-size distribution c_s, i.e., the concentration of clusters of size s,

$$m_k' = \sum_{s} s^k c_s, \tag{59}$$

where the summation in Eq. (59) runs over all finite clusters. Generally, concentrations are normalized such that

$$c_s = \lim_{N\to\infty} \frac{n_s}{N}.$$

The probability that a monomer belongs to the infinite cluster (the gel) is equal to $1 - m_1'$, with

$$m_k' = \lim_{N\to\infty} \frac{M_k'}{N}.$$

For example, in the thermodynamic limit when the size of the box becomes infinite, a finite fraction of the total number of vertices belongs to this cluster. Therefore, we obtain the results $m_1' = 1$ for $p < p_{cr}$ and $m_1' < 1$ for $p > p_{cr}$. Moreover, m_1' is a decreasing function of the occupation probability. This typical behavior is commonly (and incorrectly) called ''the failure of mass conservation,'' but, as stated before, m_1' is more simply the probability for a vertex to belong to some finite cluster.

A. Percolation on the Bethe lattice

The bond percolation on the Bethe lattice with a coordination number $\hat{z}$, has been solved by Fisher and Essam [27]. Here the main result we are interested in, is the concentration-size distribution [27]

$$c_s = \hat{z} \frac{[(\hat{z}-1)s]!}{[(\hat{z}-2)s+2]!\,s!} p^{s-1} (1-p)^{(\hat{z}-2)s+\hat{z}},$$

and the first normalized moment

$$m_1' = \left(\frac{1-p}{1-p^*} \right)^{2\hat{z}-2},$$

with p^* being the smallest solution of the equation:

$$p^*(1-p^*)^{\hat{z}-2} = p(1-p)^{\hat{z}-2}.$$

Let us define

$$p_{cr} \equiv \frac{1}{\hat{z}-1}.$$

For $p < p_{cr}$, the only solution of the above equation is $p^* = p$, but when p is larger than p_{cr}, then there is a smaller nontrivial solution which behaves as $p_{cr} - |p - p_{cr}|$ near p_{cr}. Above this threshold, the moment m_1' is smaller than 1, and behaves approximately as

$$m_1' \simeq 1 - \frac{2(p-p_{cr})}{1-p_{cr}}.$$

The marginal case $\hat{z} = 2$ corresponds to the linear-chain case.

Coming back to the concentrations, we can see that for large values of the size s, the following Stirling approximation holds:

$$c_s \sim s^{-5/2} \exp(-\alpha s),$$

with α given by

$$\alpha = \ln\left[\frac{p}{p_{cr}} \left(\frac{1-p}{1-p_{cr}} \right)^{\hat{z}-2} \right].$$

For this model, a power-law behavior of the concentrations is seen at the threshold p_{cr}, namely, $c_s \sim s^{-\tau}$, with $\tau = 5/2$. Outside this threshold, an exponential cutoff is always present [28]. This sort of critical behavior at an equilibrium is analogous to the thermal critical phenomena, and in particular, there exist two independent critical exponents, for example, τ and σ. The latter one is the exponent of the mean cluster-size divergence. Together, the two critical exponents $\tau = 5/2$ and $\sigma = 1$ completely describe the critical features.

This singular behavior is due to the appearance of a giant cluster, the so-called percolation cluster, at the transition point. More precisely, in the infinite system the probability for a given site to belong to this infinite cluster is zero below the critical threshold p_{cr}, and positive above it. This probability is nonanalytical at the critical point. Because of this behavior, the extensive order parameter defined for finite systems is just the size of the largest cluster s_{max}.

As discussed in Sec. II C, the corresponding finite-size order parameter scales as

$$s_{max} \sim N^{2/3}.$$

Even though the system experiences a second-order critical phenomenon, fluctuations of the multiplicity distribution remain small and the KNO scaling does not hold. Of course, m_0' is not in this case an order parameter since $\tau > 2$ even though there is some irregularity in its behavior passing the threshold. This nonanalyticity can be illustrated by an exact result for the bond percolation on the Bethe lattice. In this mean-field case, the normalized zeroth-moment is

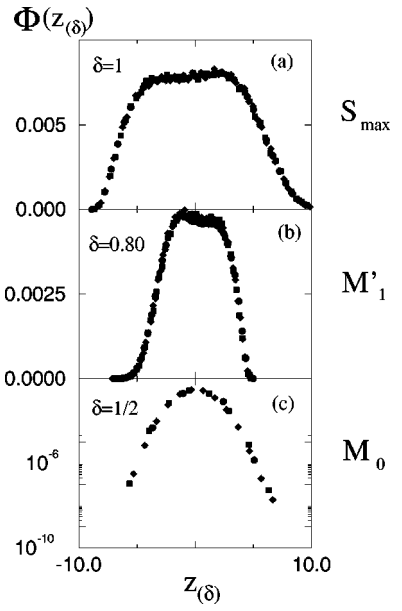

FIG. 2. (a) The first scaling of s_{max} distributions at the percolation threshold ($p=p_{cr}=0.2488$) of the three-dimensional bond percolation for lattices of different sizes: $N=14^3$ (diamonds), $N=20^3$ (squares), and $N=32^3$ (circles). The data correspond to 10^5 events. (b) The Δ scaling of the distributions of $M_1'=N-s_{max}$ for the same conditions as in (a). (c) The second scaling of the multiplicity distributions plotted on a log-linear scale (i.e., $\log(\langle M_0\rangle^{1/2}P[M_0])$ vs $z_{(1/2)}$) for the same conditions as in (a).

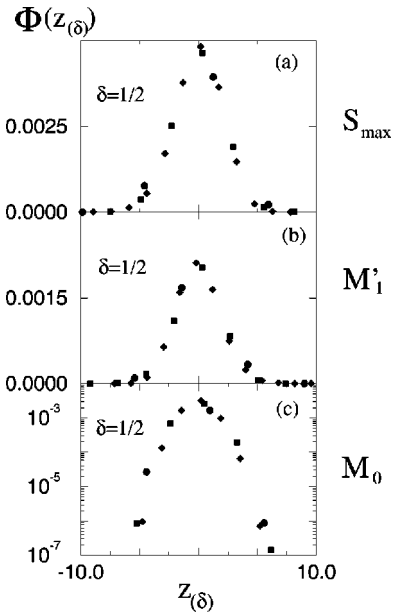

FIG. 3. (a) The second scaling of s_{max} distributions above the percolation threshold ($p=0.35$) of the three-dimensional bond percolation for lattices of different sizes: $N=14^3$ (diamonds), $N=20^3$ (squares), and $N=32^3$ (circles). The calculated data correspond to 10^5 events. (b) The second scaling of the distributions of $M_1'=N-s_{max}$ for the same conditions as in (a). (c) The second scaling of the multiplicity distributions plotted on a log-linear scale (i.e., $\log(\langle M_0\rangle^{1/2}P[M_0])$ vs $z_{(1/2)}$) for the same conditions as in (a).

$$m_0'=\left(1-\frac{\hat{z}}{2}p^*\right)\left(\frac{1-p}{1-p^*}\right)^{2\hat{z}-2}$$

$$\simeq\frac{\hat{z}-2}{2(\hat{z}-1)}-(\hat{z}-1)\epsilon+\left(1-\frac{\hat{z}}{2}\right)|\epsilon|,$$

with $\epsilon=p-p_{cr}$ and $\epsilon\ll 1$. There is a jump of the first derivative of m_0' with respect to p: $-\hat{z}/2$ for $p\to p_{cr}^-$ and $(4-3\hat{z})/2$ for $p\to p_{cr}^+$.

B. Three-dimensional percolation

As shown by Botet *et al.* [16], the multiplicity distribution for the three-dimensional bond percolation model on the cubic lattice at the infinite-network percolation threshold exhibits a Δ scaling with $\Delta=1/2$, and hence the fragment multiplicity is not related to the order parameter in this process. This is shown in Fig. 2(c) as a typical example of noncritical parameter scaling. Note that the multiplicity distributions in Fig. 2(c) are plotted in a semi-logarithmic form to show clearly the Gaussian behavior (a parabolic shape on the figure). The proper order parameter for this model is the normalized mass of the gel phase, i.e., the mass of the largest cluster divided by the total mass of the system s_{max}/N. Different probability distributions $P_N[s_{max}/N]$ for different system sizes N can be all compressed into a unique characteristic function [see Fig. 2(a)]

$$\left\langle\frac{s_{max}}{N}\right\rangle P_N\left[\frac{s_{max}}{N}\right]=\Phi\left(\frac{s_{max}-\langle s_{max}\rangle}{\langle s_{max}\rangle}\right),$$

which is analogous to the KNO scaling function. As an application of the results developed in Sec. II E, Fig. 2(b) shows the Δ scaling for the shifted order parameter $M_1'=N-s_{max}$. The value of Δ ($=0.8$) is consistent with the value of the anomalous dimension [Eq. (25)], $g=0.8435$, for the accepted values of the critical exponents β,γ in the three-dimensional percolation [15]. One should also remember that Δ has been extracted from a small size ($N=14^3$, 20^3, and 32^3) percolation network calculations at the *infinite-network* percolation threshold. This explains a small difference between the value for Δ from the scaling analysis, and the expected value $\Delta=g$ in the infinite network.

According to the results derived above for the second-order phase transition, the second scaling should hold outside of the critical point. This is correctly realized with the three variables s_{max}, M_1', and M_0 for large or small values of the probability p. Figure 3 shows such results for the value $p=0.35$.

Finally, it is instructive to see how the first scaling is disappearing when the value of p is slightly shifted away from its critical value. Fig. 4 illustrates the deviations from the first scaling for the values of a parameter p close to p_{cr}, on both sides of p_{cr}. Even very close to the critical point, these deviations are quite significant, and can easily be seen in this representation.

VI. IRREVERSIBLE AGGREGATION PROCESS — AN EXAMPLE OF THE SMOLUCHOWSKI KINETIC MODEL

The irreversible sol-gel transition can be modelled using the coupled nonlinear differential equations in distributions

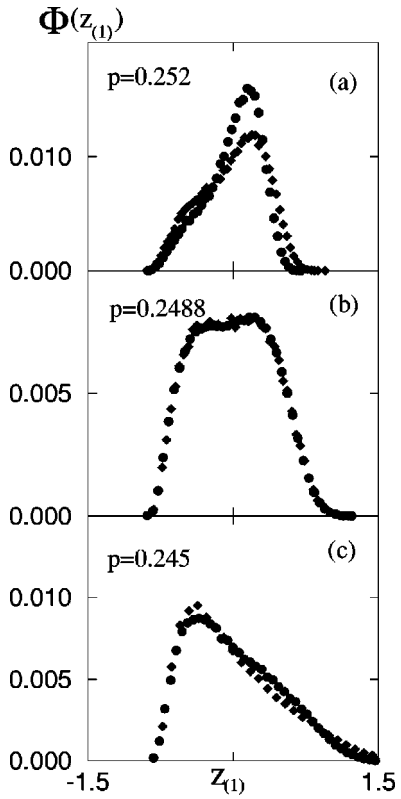

FIG. 4. (a) The s_{max} distributions are plotted in the first-scaling form for parameters p close to the percolation threshold of the three-dimensional bond percolation: (a) $p=0.252$, (b) $p=p_{cr}=0.2488$, and (c) $p=0.245$. The calculations are done for lattices of different sizes: $N=14^3$ (diamonds), and $N=32^3$ (circles). The calculated data correspond to 10^5 events.

c_s of clusters of mass s per unit volume (the Smoluchowski equations [29]):

$$\frac{dc_s}{dt}=\frac{1}{2}\sum_{i+j=s}K_{i,j}c_ic_j-\sum_j K_{s,j}c_sc_j. \tag{60}$$

Coefficients $K_{i,j}$ represent the probability of aggregation per unit of time between two clusters of mass i and j . The Smoluchowski equations are derived from the master equation in the mean-field approximation [30]:

$$\langle c_kc_l\rangle=\langle c_k\rangle\langle c_l\rangle.$$

The time t includes both diffusion and reaction times [31]. Equations (60) suppose the irreversibility of the aggregation, i.e., the cluster fragmentation is excluded. One should note, however, that the sum over j in Eq. (60) does not include the infinite cluster (gel), because

$$c_{j=\infty}=1/\infty=0.$$

Experimentally known aggregation kernels K_{ij} are homogeneous functions [32],

$$K_{ai,aj}=a^{\lambda}K_{i,j},$$

with λ being the homogeneity index. Perhaps the simplest physically relevant example of the homogeneous kernel is $K_{i,j}=(ij)^{\mu}$. It has been shown in this case that if μ is larger than 1/2, then there exists a time t_{cr} ($t_{cr}<\infty$) such that $m_1'=1$ for $t\leqslant t_{cr}$ but $m_1'<1$ for $t>t_{cr}$ [33,34].

Let us now consider the case $K_{i,j}=(ij)^{\mu}$ with $\mu=1$ in more details. It was shown in this case [33] that the critical gelation time is : $t_{cr}=1$, and the solutions for the size distribution of Smoluchowski equations with the monodisperse initial condition are [35].

$$c_s=\frac{s^{s-2}}{s!}e^{-st}t^{s-1} \quad \text{for} \quad t\leqslant 1,$$

$$c_s=\frac{s^{s-2}}{s!}\frac{\exp(-s)}{t} \quad \text{for} \quad t>1.$$

The asymptotic solutions for large s are

$$c_s\sim\frac{1}{t\sqrt{2\pi}}s^{-5/2}\exp[-s(t-1+\ln t)] \quad \text{for} \quad t\leqslant 1,$$

$$c_s\sim\frac{1}{t\sqrt{2\pi}}s^{-5/2} \quad \text{for} \quad t>1. \tag{61}$$

Note that the power-law behavior ($\tau=5/2$) is present for $t>1$, and not only at the threshold. The whole distribution of finite-size clusters evolves self-similarly, and the appearance of a power-law behavior is not a sign of a critical behavior but a specific characteristics of the gelation phase.

The solutions for the first normalized moment are

$$m_1'=1 \quad \text{for} \quad t\leqslant 1,$$

$$m_1'=\frac{1}{t} \quad \text{for} \quad t>1.$$

With those asymptotic forms of c_s, one can calculate the gel fractions in the infinite system before and after the critical point:

$$m_G=0 \quad \text{for} \quad t\leqslant 1,$$

$$m_G=1-\frac{1}{t} \quad \text{for} \quad t>1.$$

It has been shown that the gelation is analogous to the dynamical critical phenomenon with [36]:

$$m_G=\lim_{N\to\infty}\frac{1}{N}\langle s_{max}\rangle$$

as the order parameter. For one realization, s_{max} corresponds to the mass of the gel above $t_c=1$.

For finite sizes, one makes the usual assumption that there exists a characteristic size which diverges at the transition, say

$$N_c\sim|t-1|^{-1/\sigma_N}, \tag{62}$$

such that for the mass gel in a finite system one has

$$\frac{1}{N}\langle s_{max}\rangle\sim(t-1)f\left(\frac{N}{N_c}\right) \quad \text{for} \quad t\geqslant 1.$$

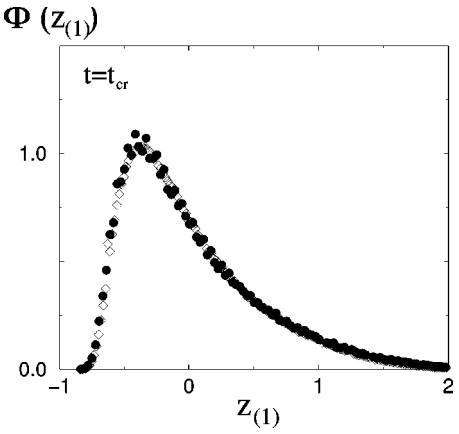

FIG. 5. The first scaling of the s_{max} variable in the Smoluchowski kinetic model with the kernel $K_{ij}=ij$ at the critical time $t=t_{cr}=1$. The calculations are performed for two system sizes: $N=2^{10}$ (diamonds) and $N=2^{14}$ (circles). Each data set corresponds to 10^5 independent events.

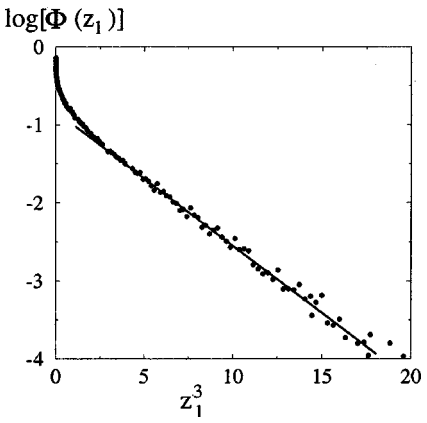

FIG. 7. The plot of the large $z_{(1)}$ tail of a decimal logarithm of the scaling function, $\log \Phi(z_{(1)})$, against $z_{(1)}^3$ for the system size $N=2^{12}$. The calculated data correspond to 10^5 independent events. The solid line shows the dependence $\Phi(z_{(1)}\sim\exp(-z_{(1)}^3)$, which is expected from the value of the anomalous dimension $g=2/3$.

In particular, at the gelation time one has

$$\langle s_{max}\rangle \sim N^{1-\sigma_N}\sim N^g. \tag{63}$$

Using formula (29), which is valid both for equilibrium and nonequilibrium systems, one can calculate the anomalous dimension. Given the value of τ [see Eq. (61)], one finds $g=2/3$. Hence $\sigma_N=1/3$ can be deduced from Eq. (63). The average value of the order parameter $\langle s_{max}\rangle$ increases logarithmically for $t<1$, and becomes a finite portion of the system size when $t>1$.

The illustration of the above discussion is shown in Figs. 5 and 6. Figure 5 shows the distribution of s_{max} in the first-scaling variables for systems of different sizes. The results have been obtained in the Smoluchowski model with the kernel $K_{ij}=ij$, at the critical time $t=t_{cr}=1$. Fluctuation properties of s_{max}, outside of the critical time $t=2t_{cr}$, are shown in Fig. 6. The remaining parameters of the Smoluchowski calculations are the same as used in the calculations shown in Fig. 5. In this case, the data for different system sizes collapse into the universal curve in the scaling variables

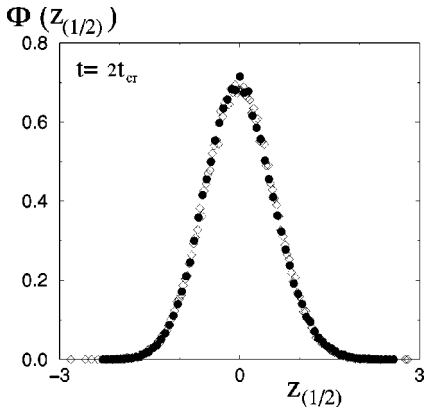

FIG. 6. The second scaling of the s_{max} variable in the Smoluchowski kinetic model with the kernel $K_{ij}=ij$ above the critical time $t=2t_{cr}=2$. The calculations are performed for two system sizes: $N=2^{10}$ (diamonds) and $N=2^{14}$ (circles). Each data set corresponds to 10^5 independent events.

with $\Delta=1/2$. One should keep in mind that in both cases, the fragment-size distribution is a power law with $\tau=5/2$ [see Eq. (61)].

The relation between the form of tail of the scaling function and the anomalous dimension [Eq. (26)] was derived analytically in Sec. II C for equilibrium systems at the second-order phase transition. For nonequilibrium systems, we do not have an equally rigorous derivation (also see Sec. VI A). On the other hand, one may expect that the relation between the N dependence of the average value of the order parameter and the asymptotic form of the scaling function in the limit $N\to\infty$, i.e., between $\hat{\nu}$ and g, is connected to the asymptotic stability of the limit distributions. Actually, there is a very close connection between the renormalization group ideas and the limit theorems in the probability theory [37]. If true, then relation (26) could be valid in a more general framework than the one provided by the equilibrium statistical mechanics. To check this assertion, in Fig. 7 we show the plot of a logarithm of the scaling function $\Phi(z_{(1)})$ (see Fig. 5) versus $z_{(1)}^3$ for large values of $z_{(1)}$. If relation (26) is also valid for the nonequilibrium sol-gel second-order phase transition, then $\Phi(z_{(1)})\sim\exp(-z^3)$, and the tail of the scaling function should be a straight line in Fig. 7. That is indeed the case.

Figures 8 and 9 show the Δ scaling for the shifted order-parameter variable $M_1'=N-s_{max}$. Results of the Smoluchowski calculations, with the kernel $K_{ij}=ij$, are shown at $t=t_{cr}$ (see Fig. 8), and at $t=2t_{cr}$ (see Fig. 9). One sees that the M_1' distribution exhibits a qualitative change while going from the critical time $t=t_{cr}$, where $\Delta=0.67$, to $t=2t_{cr}$ for which $\Delta=1/2$. At $t=t_{cr}$, the value of Δ obtained by superimposing different M_1' distributions in the scaling plot [Eq. (39)], agrees perfectly with the value of the anomalous dimension g $(=2/3)$.

By comparing Figs. 5 and 6 and 8 and 9, one may also see that the effect of changing the variable $s_{max}\to M_1'$ is seen only at the critical time (compare Figs. 5 and 8) where $(\Delta=1)\to(\Delta=0.67)$, and is absent above the critical time (compare Figs. 6 and 9) where $\Delta(=1/2)$ remains unchanged.

Finally, in Fig. 10 we show the size-dependence of the

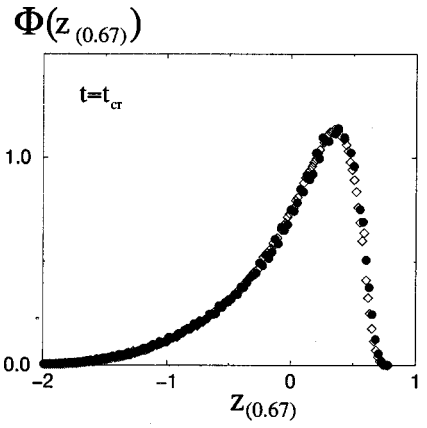

FIG. 8. The Δ scaling of the distributions of the M_1' shifted order parameter in the Smoluchowski kinetic model with a kernel $K_{ij}=ij$ at the critical time $t=t_{cr}=1$. Two system sizes are considered: $N=2^{10}$ (diamonds) and $N=2^{14}$ (circles). The calculated data correspond to 10^5 independent events.

M_1' distributions at $t=t_{cr}$, when the distributions are plotted in the ''wrong'' variables of the second-scaling $\Delta=1/2$. The distributions for two system sizes are clearly displaced , showing the sensitivity of the scaling analysis and the failure of the second scaling.

A. Origin of fluctuations and the argument of Van Kampen

Ω expansion is a systematic expansion of the master equations in powers of $1/N$ [2]. Lushnikov was the first to express the generating functions as the contour integrals for quantities like the moments M_k' [38]. Then Van Dongen and Ernst [39] used an Ω expansion to calculate these integrals explicitly for the moments M_k' in some simple cases like $K_{ij}=ij$. For example, the result for M_1' can be expressed in terms of the generating function for the s_{max}-distribution $P_N[s_{max}]$, as

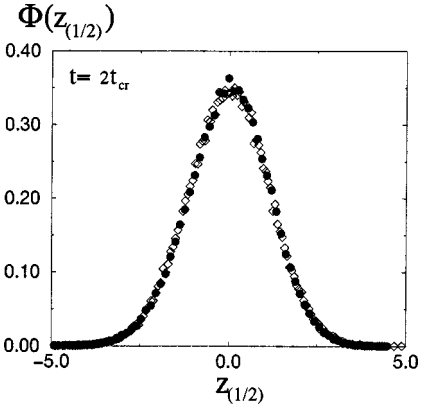

FIG. 9. The second scaling of the distributions of M_1' shifted order parameter in the Smoluchowski kinetic model with the kernel $K_{ij}=ij$ above the critical time, at $t=2t_{cr}$. Two system sizes are considered: $N=2^{10}$ (diamonds) and $N=2^{14}$ (circles). The calculated data correspond to 10^5 independent events.

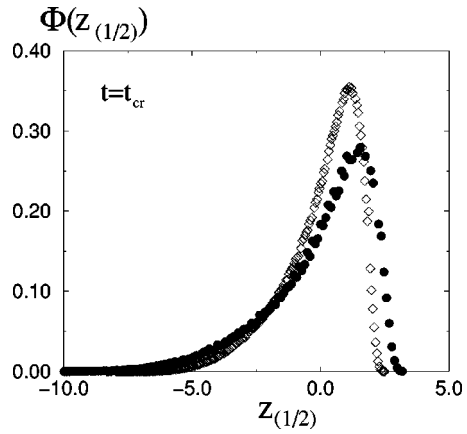

FIG. 10. The distributions of the M_1' shifted order parameter in the Smoluchowski kinetic model with kernel $K_{ij}=ij$, at the critical time $t=t_{cr}=1$, are plotted in the scaling variables of second scaling. Two system sizes are considered: $N=2^{10}$ (diamonds) and $N=2^{14}$ (circles). The calculated data correspond to 10^5 independent events. The failure of the second scaling is clearly visible.

$$\sum_{s_{max}} P_N[s_{max}]e^{s_{max}u}=\frac{N!}{2i\pi}e^{-Nu}\oint\frac{dz}{z^{N+1}}\times\exp\left[\sum_{s=1}^{N}\frac{c_s}{N^{s-1}}e^{s(N-s)/(2N)}(ze^{-u})^s\right]$$

Then using then the identity,

$$\frac{\partial^n}{\partial u^n}\left[\exp\left(\sum_s \alpha_s e^{su}\right)\right]=\sum\frac{n!}{1!^{a_1}a_1!\cdots n!^{a_n}a_n!}\times\left[\sum_s \alpha_s s^1 e^{su}\right]^{a_1}\cdots\left[\sum_s \alpha_s s^n e^{su}\right]^{a_n}\times\exp\left(\sum_s \alpha_s e^{su}\right),$$

where the sum runs over different sets $\{a_1,\ldots,a_n\}$ with the constraint : $a_1+\cdots+a_n=n$, and the particular result written down by Van Dongen and Ernst for the $K_{ij}=ij$-case [39],

$$\exp\left[\sum_{s=1}^{N}\frac{c_s}{N^{s-1}}\exp\left[\frac{s(N-s)}{2N}\right]z^s\right]=\sum_{s=1}^{N}\frac{z^s}{s!}\exp\left[-\frac{1}{2}st\frac{1-s}{N}\right]+O(1/N),$$

we can find

$$\sum_{s_{max}} P_N[s_{max}]\exp(s_{max}u)=\exp\left[N\left(\sum_{k=1}^{\infty}\frac{m_k'(-u)^k}{k!}+u\right)\right]. \tag{64}$$

Having the Ω expansion of the generating function of the s_{max} distribution, we can make a conclusion about scaling at the gelation point. The moments m'_k of the size distribution for infinite systems are known to diverge near the gelation time [40] as

$$m'_k \sim |t-1|^{3-2k}.$$

For finite systems, using $\sigma_N = 1/3$, one obtains

$$m'_k \sim |t-1|^{-2k+3} f_k(N|t-1|^3) \sim N^{(2k-3)/3} \tag{65}$$

at the gelation time. We have then found the asymptotic result

$$N m'_k \simeq a_k \langle s_{max} \rangle^k, \tag{66}$$

where a_k's are some positive constants. Inserting Eq. (66) into Eq. (64), one can show that the generating function for s_{max} is the function of a single variable $\langle s_{max} \rangle u$, which is a sufficient condition for the validity of the first-scaling law [Eq. (9)].

We can have informations on similar scalings for various moment distributions. Ω expansion leads to the results

$$\langle M'^2_k \rangle - \langle M'_k \rangle^2 = N m'_{2k} - (1-t) m'^2_{k+1},$$

$$\langle M'_k \rangle = N m'_k \tag{67}$$

for the values of k when all the quantities are defined. At the transition ($t=1$), relation (65) allows one to calculate the reduced moments m'_k. The results can be written in the compact form

$$\frac{\langle M'^2_k \rangle - \langle M'_k \rangle^2}{\langle M'_k \rangle^{2\Delta}} \sim \text{const},$$

with the following values of exponent Δ :

$$\Delta = 1/2 \quad \text{for} \quad k \leqslant 3/4,$$

$$\Delta = 2k/3 \quad \text{for} \quad 3/4 \leqslant k \leqslant 3/2,$$

$$\Delta = 1 \quad \text{for} \quad 3/2 \leqslant k.$$

These are indications of Δ scaling according to Eq. (49) in Sec. II H. More precisely, the moments of order $k < 3/4$ are not critical (the second-scaling law), the moments of order k between 3/4 and 3/2 exhibit Δ scaling with $\Delta = 2k/3$. In particular, for $k=1$, one recovers the correct value $\Delta = g = 2/3$ corresponding to the general argument of a shifted order parameter [Eq. (37)] with $a_1 = a_2 = 1$. Finally, when the value of k is larger than 3/2, we obtain the first-scaling law for the distribution of moments M'_k. This is also a consequence of the shifted order-parameter argument, since, in these cases,

$$\langle M'_k \rangle \sim \langle s^k_{max} \rangle.$$

Far from the critical point, all the reduced moments m'_k are independent of N, since the correlation size.

$$\frac{1}{t-1+\ln t}$$

[see Eq. (61)] is finite. Then, for any value of k, the second-scaling law holds, as expected from the general theory.

The above results about Δ scaling for various moments of the size distribution in the Smoluchowski model, with kernel $K_{ij} = ij$, are not complete, since the arguments involve only the second cumulant moment κ_2. In principle, as shown in Sec. II H, all cumulants should be investigated. So, even though many exact results are known in this model, the complete analytical solution is not yet available.

The same study as presented above for gelling systems, can be performed also for nongelling systems. An example of this kind is obtained for $K_{ij} = i + j$. In this case, the size distribution is a power law with the exponent $\tau = 3/2$ [41] and, following the discussion in Sec. II C, the cluster multiplicity can be the order parameter. One can analytically derive, that the multiplicity distribution is binomial,

$$P_N[M_0,t] = \binom{N-1}{M_0-1} [1-\exp(-Nt)]^{N-M_0} \times \exp[-(M_0-1)Nt],$$

and can be approximated for $N \to \infty$, and for a finite value of $\langle M_0 \rangle / N$, by

$$\langle M_0 \rangle^{1/2} P_N[M_0,t] \sim \frac{1}{\sqrt{2\pi}(1-e^{-Nt})} \times \exp\left(-\frac{1}{2(1-e^{-Nt})} \frac{(M-\langle M_0 \rangle)^2}{\langle M_0 \rangle} \right), \tag{68}$$

which corresponds to the second scaling. One may note, that this binomial distribution is exactly equivalent to the bond percolation on a Bethe lattice with the occupation probability:

$$p = 1 - \exp(-Nt).$$

In spite of self-similar features in the fragment-size distribution at an infinite time, one does not see any critical behavior in the cluster multiplicity distribution at any time in the nongelling aggregation systems. This confirms the observation, made in Sec. III A for the Mekjian equilibrium model, that the power-law size distribution alone does not guarantee that the system exhibits a critical behavior.

The insight gained from the numerical simulations of Smoluchowski equations, and the evidence from exact results for both gelling and nongelling aggregation systems, provide strong hints that the discussion of Sec. II H is valid not only for equilibrium systems but also for nonequilibrium ones. We see the same significance of the Δ scaling in nonequilibrium systems as found in thermodynamic systems, not only at the critical point but also close to the critical point or even far from it. We believe that this universality, which is common to equilibrium and nonequilibrium systems, has a deeper foundation in the relation between renormalization group ideas for self-similar systems and the limit theorems of

probability theory for the asymptotic scaling laws of order-parameter distributions. The concept of statistical equilibrium does not intervene at this level. One should also remember that the universality discussed in this work is associated with only one critical exponent, and certainly does not exhaust all the singularity properties of the thermodynamical potential in the second-order thermal phase transition.

VII. CONCLUSIONS

In this paper we have presented a theory of universal scaling laws of order-parameter fluctuations in any system in which the second-order critical behavior exist. These scaling laws, called Δ-scaling laws, are rigorously derived for the equilibrium systems. Moreover, both analytical and numerical evidence is also presented in favor of a general validity of the Δ-scaling laws for off-equilibrium processes which exhibit a critical phenomenon of second order. The method works very efficiently if the ''observables'' are known numerically, either by means of numerical simulations or experimental data.

In this work, we have discussed different aggregation models, both reversible and irreversible ones, finding the same connection between the scaling function properties and the anomalous dimension (the critical exponents). These results can be important in a phenomenological analysis of ''the critical behavior'' in finite systems, where the critical exponent analysis is dubious and, moreover, the precise mechanism of the process may be unknown. In these cases, the Δ-scaling analysis allows one to select both the relevant observable and the interesting initial conditions, which lead to a ''pseudocritical'' behavior in the studied process. Another interesting aspect of the Δ-scaling analysis is the possibility of compressing data and, hence, the elimination of redundant dependences in the data for parameters like the system size, the total energy, the total momentum, etc., This provides an obligatory intermediate step in any phenomenological analysis before the laws governing complicated dynamics can be found. Finally, one should stress that the scaling laws discussed in this paper are independent of whether one deals with an equilibrium process or an off-equilibrium process. This is a crucial advantage in studies of short-lived systems. Examples of multifragmentation processes in collisions of atomic nuclei or atomic clusters illustrate this problem well [1]. In the absence of thermal equilibrium, which is a theoretical hypothesis difficult to verify in dynamically formed short-lived systems, we simply do not have any other tool at our disposal to address reliably the question of possible ''criticality'' of the studied process.

As stated above before, the Δ scaling analysis developed in this work provides an alternative to critical exponent analysis in equilibrium systems, and is the only tool for the analysis of the nonequilibrium systems. All essential information can be deduced from the scaling function, the value of the Δ parameter, the form of the tail of the scaling function and the value of the anomalous exponent. With this information it is possible to find out whether the studied process is at the critical point, in its neighborhood, or far away from it. The reference point in this analysis is the self-similarity of the system. A generalization of the above scaling theory to discontinuous phase-order transitions, for which the characteristic length can be defined, is in progress. Our preliminary experience with systems which exhibit a first-order phase transition show, however, that the Δ scaling cannot be defined in these systems [42].

ACKNOWLEDGMENT

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