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Additivity property and emergence of power laws in nonequilibrium steady states

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We show that an equilibriumlike additivity property can remarkably lead to power-law distributions observed frequently in a wide class of out-of-equilibrium systems. The additivity property can determine the full scaling form of the distribution functions and the associated exponents. The asymptotic behavior of these distributions is solely governed by branch-cut singularity in the variance of subsystem mass. To substantiate these claims, we explicitly calculate, using the additivity property, subsystem mass distributions in a wide class of previously studied mass aggregation models as well as in their variants. These results could help in the thermodynamic characterization of nonequilibrium critical phenomena.

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I. INTRODUCTION

Simple power-law scaling is ubiquitous in nature [1]. It appears in the distribution of drainage areas of rivers [2], droplet size [3,4], the size of clusters formed in polymerization processes [5], rain size [6], the size of fragments in fractured solids [7], population and wealth [8,9], stock market fluctuations [10], etc. Evidently, power laws, which are usually associated with criticality through the emergence of a diverging length scale, are observed in widely unrelated systems, suggesting the existence of some broad underlying principle. Recent evidence that living systems might be operating, independent of most of the microscopic details, in the vicinity of a critical regime [11] indeed invokes further questions, e.g., how and why systems adapt to near criticality.

There have been several attempts to reveal the origin of power laws in nature through studies of paradigmatic nonequilibrium models, the most appealing being sandpile [12–14] and mass aggregation models [15–20]. Many of these models, where there is a conservation law or, in the case of violation, the law is weakly violated in the sense that the systems are slowly driven, are intimately connected to each other. For example, the mass aggregation models [17–19,21] are connected to the directed Abelian sandpile model [22] or to the models of river networks [2].

In this paper we argue that power-law distributions in out-of-equilibrium systems can arise simply from an additivity property, the tenet of equilibrium thermodynamics. We find that the divergence in the response function is the key: Diverging fluctuations can in principle arise from distributions other than simple power laws, which are however prohibited if one imposes additivity and the consequent fluctuation-response (FR) relation. The response function determines the full scaling form of the distribution at as well as away from criticality and critical exponents originate from the singularity in the response function. To demonstrate this, we consider mass aggregation models that are known to have a nonequilibrium steady state with scale-invariant structures. At all mass densities, the distribution function $P_v(m)$ of mass m in a subsystem of volume v, which is obtained solely from the FR relation, is shown to have a scaling form $P_v(m) \sim m^{-\tau} \exp(\tilde{\mu} m)$. The quantity $\tilde{\mu}(\rho) = \mu(\rho) - \mu(\rho_c)$, the inverse of a cutoff mass $m^*(\rho) = -1/\tilde{\mu}(\rho)$, is an analog of the equilibriumlike chemical potential and provides a useful thermodynamic interpretation of the emergence of power laws in nonequilibrium steady states. The exponent τ and the critical properties of chemical potential $\mu(\rho)$ arise from a multiple-pole or branch-cut singularity in the variance at a critical mass density ρ_c. As the critical density is approached $\rho \to \rho_c$, the nonequilibrium chemical potential vanishes $\mu(\rho) \to 0$, leading to pure power laws. Beyond the critical density $\rho > \rho_c$, there is a gas-liquid-like phase coexistence.

The above result immediately provides answer to why the $m^{-5/2}$ power law, at or away from criticality, appears so often in mass aggregation models, especially in higher dimensions, at all densities and irrespective of whether or not the motion of the diffusing masses is biased [19,21,23,24]. Interestingly, the same power law appears in the k-mer distribution in the classic Flory-Stockmayer [25] theory of polymerization and also in the particle-number distribution in a three-dimensional ideal Bose gas near the critical point, irrespective of whether the systems are in or out of equilibrium, thus indicating a universality. We demonstrate that the $m^{-5/2}$ law is a consequence of a simple-pole singularity in the variance. The whole analysis is extended also to nonconserved-mass aggregation models. We validate our theory by explicitly calculating mass distributions in previously studied mass aggregation models and their variants and by comparing them with simulations.

The organization of the paper is as follows. In Sec. II A we discuss the additivity property. In Sec. II B we discuss the connection between singularity in the variance and the asymptotic behavior of the mass distribution function. In Sec. III we illustrate our analytic methods in a broad class of model systems, in both conserved-mass aggregation models and the nonconserved versions. In Sec. IV we summarize our results with a concluding perspective.

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II. THEORY

A. Additivity property

We start by invoking an additivity property that a wide class of systems, irrespective of whether they are in or out of equilibrium, could possess. Consider a continuous-mass (generalization to discrete masses is straightforward) transport process on a lattice of size V and then divide the system into $\nu = V/v$ identical subsystems or cells, the kth subsystem with mass m_k, where the total mass $M = \sum_k m_k$ remains conserved. Provided the subsystems are large compared to the spatial correlation length, they could be considered statistically almost independent [26–29]. In that case, the joint subsystem mass distribution in the steady state can be written in a product form

$$\mathcal{P}[\{m_k\}] \simeq \frac{\prod_{k=1}^{\nu} w_v(m_k)}{Z(M,V)} \delta\left(\sum_k m_k - M\right), \tag{1}$$

where $w_v(m_k)$ is an unknown weight factor [to be determined later; see Eq. (13)] depending only on the subsystem mass m_k, $Z = \prod_k \int dm_k w_v(m_k) \delta(\sum_k m_k - M) \equiv \exp[-V f(\rho)]$ is the partition sum, $f(\rho)$ is a nonequilibrium free energy density, and $\rho = M/V$ is the mass density. The product form in Eq. (1) amounts to an equilibriumlike additivity property, in the sense that a free energy function $F = -\sum_k \ln w_v(m_k)$ is minimized in the macrostate.

Using standard statistical mechanics [30], Eq. (1) leads to the probability distribution $\text{Prob}[m_k \in (m, m+dm)] = P_v(m)dm$ for the subsystem mass where

$$P_v(m) = \frac{w_v(m) e^{\mu m}}{\mathcal{Z}}, \tag{2}$$

with $\mu(\rho)$ a nonequilibrium chemical potential and $\mathcal{Z}$ the normalization constant. The weight factor $w_v(m)$ and chemical potential $\mu(\rho) = df/d\rho$ can be obtained using a fluctuation-response relation [26–29,31]

$$\frac{d\rho}{d\mu} = \sigma^2(\rho), \tag{3}$$

where the scaled variance $\sigma^2(\rho) = (1/v)(\langle m_k^2 \rangle - \langle m_k \rangle^2)$ in the limit of $v \gg 1$. The free energy density function $f(\rho)$ can be obtained through the relation $\mu(\rho) = df/d\rho$, i.e., $f(\rho) = \int \mu(\rho) d\rho + \beta$, with the chemical potential $\mu(\rho) = \int 1/\sigma^2(\rho) d\rho + \alpha$ [obtained from Eq. (3)] and α and β arbitrary constants of integration. Then the Laplace transform of $w_v(m)$ is written as $\tilde{w}_v(s) = \int w_v(m) \exp(-sm) dm \equiv e^{-\lambda_v(s)}$, i.e.,

$$e^{-\lambda_v(s)} = \int w_v(m) e^{-sm} dm. \tag{4}$$

The function $\lambda_v(s)$ can be obtained from a Legendre transform of the free energy density function $f(\rho)$ [32],

$$\lambda_v(s) = v[\inf_\rho \{f(\rho) + s\rho\}] = v[f(\rho^*) + s\rho^*], \tag{5}$$

where $\rho^*(s)$ is the solution of

$$s = -\mu(\rho^*). \tag{6}$$

As discussed later, Eq. (5) requires concavity and differentiability of $f(\rho)$. In the discrete case, the weight factor can be calculated as $w_v(m) = (1/2\pi i) \int_C \tilde{w}_v(z)/z^{m+1} dz$, where $\tilde{w}_v(z) = \sum_{m=0}^{\infty} z^m w_v(m)$ is obtained from $\tilde{w}(s)$ by substituting $s = -\ln z$ and C is a suitably chosen contour in the complex z plane.

B. Singularity in variance and mass distribution

Importantly, the distribution function $P_v(m)$ is determined solely by the functional form of the scaled variance $\sigma^2(\rho)$. We argue below that singular response functions generate only power-law distributions. Other functional form of the mass distribution $P_v(m)$ with diverging variance is also possible [33], which we show, however, is not allowed if the FR relation holds. In this paper we mainly focus on multipole singularity at a finite density ρ_c,

$$\sigma^2(\rho) = \begin{cases} \frac{g(\rho)}{(\rho_c - \rho)^n} & \text{for } \rho < \rho_c \\ \infty & \text{otherwise.} \end{cases} \tag{7}$$

This form, with $0 < n < \infty$, is relevant in the context of a wide class of mass aggregation models as discussed in Sec. III. The analytic part $g(\rho)$ is not particularly relevant in determining the asymptotic form of the distribution $P_v(m)$, however it contributes to the exact form of $P_v(m)$ (discussed in Sec. II B 4). In fact, other kinds of singularities, such as logarithmic singularity $\sigma^2(\rho) \sim [\ln(\rho_c - \rho)]^p$ or exponential singularity $\exp[(\rho_c - \rho)^{-p}]$ where $p > 0$, $1/|\rho - \rho_c|^n$, and the case with $n < 0$ can also arise. One can show that they all lead to power laws, possibly with logarithmic corrections to the power-law scaling (discussed in the following sections).

The divergence in the variance, as in Eq. (7) (or in the cases of logarithmic and exponential divergence), indeed has broad implications, not only in conserved-mass aggregation models but also in nonconserved versions. Note that the FR relation in Eq. (3) implies that free energy density $f(\rho)$ is not a strictly concave function of ρ and has a linear branch of slope $\mu(\rho_c)$ for $\rho \geqslant \rho_c$. Moreover, $f''(\rho = \rho_c) = \mu'(\rho = \rho_c) = 0$ (a prime denotes the derivative with respect to ρ) implies a point of inflection in the f-ρ curve at $\rho = \rho_c$. That is, the free energy density function can be written as

$$f(x) = \begin{cases} \int \mu(x) dx & \text{for } x < \rho_c \\ \mu(\rho_c)(x - \rho_c) + f(\rho_c) & \text{otherwise.} \end{cases} \tag{8}$$

Consequently, the Legendre transform of $f(\rho)$ develops a branch-cut singularity [see Eq. (11)]; for a schematic representation of the above analysis, see Fig. 1. This construction of a nonequilibrium free energy function $f(\rho)$ from a general thermodynamic consideration readily explains the phase coexistence between a fluid and a condensate, as observed in the past in many out-of-equilibrium systems (discussed in Sec. III).

1. Multipole singularity

To analyze the behavior of $\lambda_v(s)$ in the case of Eq. (7), we integrate Eq. (3) near $\rho = \rho_c$ and obtain

$$\mu(\rho) \simeq -\frac{(\rho_c - \rho)^{n+1}}{(n+1) g(\rho_c)} [1 + \mathcal{O}(\rho_c - \rho)] + \alpha, \tag{9}$$

which gives $(\rho_c - \rho^*) \simeq [(n+1) g(\rho_c)(s + \alpha)]^{1/(1+n)}$ by using Eqs. (6) and (9). Integrating the chemical potential $\mu(\rho)$,

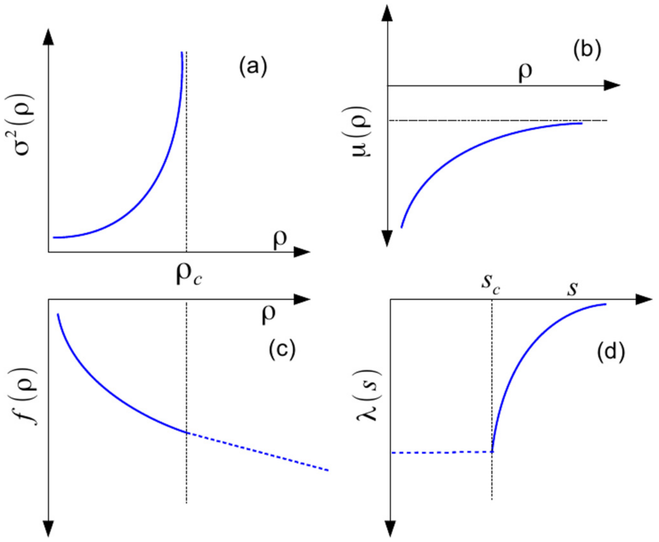

FIG. 1. (Color online) Schematic representation of condensation transition: (a) variance $\sigma^2(\rho)$ as a function of density ρ, (b) chemical potential $\mu(\rho)$ as a function of ρ, (c) free energy density $f(\rho)$ as a function of ρ, and (d) Legendre transform $\lambda(s)$ of the free energy density as a function of s.

we get the free energy function

$$f(\rho) \simeq \frac{(\rho_c - \rho)^{n+2}}{(n+1)(n+2)g(\rho_c)} + \alpha\rho + \beta \tag{10}$$

and write $\lambda_v(s) = v[f(\rho^*) + s\rho^*]$, to leading order, as

$$\lambda_v(s) \simeq v[a_0 + a_1(s+\alpha) + a_2(s+\alpha)^{(n+2)/(n+1)}], \tag{11}$$

where a_0, a_1, and a_2 are constants. Thus, we obtain $\tilde{w}(s) = \exp[-\lambda_v(s)] \simeq \text{const} \times [1 - va_1(s+\alpha) - va_2(s+\alpha)^{1+1/(1+n)}]$ to leading orders of $s+\alpha$, implying

$$w_v(m) \sim \frac{e^{-\alpha m}}{m^\tau},$$

where, for large subsystem masses $m \gg v$, the power-law exponent τ in the denominator is given by

$$\tau = \left[2 + \frac{1}{(1+n)}\right], \tag{12}$$

with the inequality $2 < \tau < 3$ [since $0 < n < \infty$ in Eq. (7)]. This translates into the mass distribution having a scaling form

$$P_v(m) \propto \frac{1}{m^\tau} e^{\tilde{\mu}(\rho)m} \equiv \frac{1}{(m^*)^\tau} \Phi\left(\frac{m}{m^*}\right), \tag{13}$$

where $\tilde{\mu}(\rho) = \int_{\rho_c}^{\rho} 1/\sigma^2(\rho)d\rho = \mu(\rho) - \mu(\rho_c)$ is an effective chemical potential, the inverse of which gives a cutoff $m^* = -1/\tilde{\mu}$ in the distribution, and the scaling function $\Phi(x) = x^{-\tau}\exp(-x)$. Later we explicitly calculate $\tilde{\mu}(\rho)$ in specific model systems. Note that $\tilde{\mu}(\rho_c) = 0$ at the critical point $\rho = \rho_c$ and consequently $P_v(m)$ becomes a pure power law. Moreover, by defining a critical exponent $\delta = 1 + n$ as $\tilde{\mu}(\rho) \sim (\rho_c - \rho)^\delta$, we get a scaling relation $\delta(\tau - 2) = 1$.

2. *Logarithmic singularity*

Now we consider the case of logarithmic singularity where the variance $\sigma^2(\rho)$ diverges logarithmically as given by

$$\sigma^2(\rho) = \begin{cases} g(\rho)[\ln(\rho_c - \rho)]^p & \text{for } \rho < \rho_c \\ \infty & \text{otherwise.} \end{cases} \tag{14}$$

Integrating Eq. (3) near $\rho = \rho_c$, we obtain the chemical potential, to leading order of $\rho_c - \rho$,

$$\mu(\rho) \simeq -\frac{(\rho_c - \rho)}{g(\rho_c)[\ln(\rho_c - \rho)]^p} + \alpha, \tag{15}$$

which gives $(\rho_c - \rho^*) \simeq g(\rho_c)[\ln(\rho_c - \rho^*)]^p(s+\alpha)$ from Eq. (6). The free energy density is obtained by integrating the above chemical potential $\mu(\rho)$,

$$f(\rho) \simeq -\frac{(\rho_c - \rho)^2}{2g(\rho_c)[\ln(\rho_c - \rho)]^p} + \alpha\rho + \beta, \tag{16}$$

and, accordingly, its Legendre transform, to leading orders,

$$\lambda_v(s) \simeq v[a_0 + a_1(s+\alpha) + a_2(s+\alpha)^2\{\ln(s+\alpha)\}^p]. \tag{17}$$

For large mass $m \gg v$, this implies that the weight factor has a functional form of a power law with logarithmic correction

$$w_v(m) \sim \frac{(\ln m)^{p-1}}{m^3} e^{-\alpha m}$$

and the corresponding mass distribution function

$$P_v(m) \propto \frac{(\ln m)^{p-1}}{m^3} e^{\tilde{\mu}(\rho)m}, \tag{18}$$

where the effective chemical potential $\tilde{\mu}(\rho) = \mu(\rho) - \mu(\rho_c)$ and the power-law exponent in the denominator is $\tau = 3$, the borderline case of Eqs. (7) and (12) with $n = 0$.

3. *Exponential singularity*

We also consider the case where the variance diverges exponentially, $\sigma^2(\rho) \sim \exp(\rho_c - \rho)^{-p}$ for $\rho < \rho_c$ and $\sigma^2(\rho) = \infty$ otherwise. The analysis is similar to the ones given above. Substituting the chemical potential $\mu(\rho) \simeq \text{const} \times (\rho_c - \rho)^{p+1}\exp[-(\rho_c - \rho)^{-p}] + \alpha$ in Eq. (6) and solving to leading order of $s+\alpha$, we get $(\rho_c - \rho^*) \sim \{\ln(s+\alpha)\}^{-1/p}$ and consequently

$$\lambda_v(s) \simeq v[a_0 + a_1(s+\alpha) + a_2(s+\alpha)\{\ln(s+\alpha)\}^{-1/p}]. \tag{19}$$

For large mass $m \gg v$, this leads to the mass distribution function

$$P_v(m) \propto \frac{(\ln m)^{-1-1/p}}{m^2} e^{\tilde{\mu}(\rho)m}, \tag{20}$$

where $\tilde{\mu}(\rho) = \mu(\rho) - \mu(\rho_c)$ and the power-law exponent in the denominator is $\tau = 2$, the borderline case of Eqs. (7) and (12) with $n = \infty$.

4. *Subsystem mass distribution*

For any finite v, it is not easy to find the distribution function $P_v(m)$ at small or intermediate values of masses

$m \sim v$ because, in that case, one must invert Eq. (4) using inverse Laplace transform, i.e., by evaluating the integral

$$w_v(m) = \frac{1}{2\pi i}\int_C e^{-\lambda_v(s)+ms}ds, \tag{21}$$

on the complex s plane; the contour C on the complex plane should be chosen such that the integral converges. However, in the models we consider here, it is not possible to get an exact functional form of $\lambda_v(s)$ for all s, which actually involves solving the transcendental equation (6). However, for large subsystem size $v \gg 1$, the analysis simplifies as the function $-(1/v)\ln w_v(m)$ is simply related to $\lambda_v(s)/v$ by the Legendre transformation [32] and therefore, to leading order of m, is the free energy density function $f(m/v)$ itself, which has already been constructed in Eq. (8) (see Fig. 1 and the related discussion). The subsystem mass distribution function now can be written as

$$P_v(m) \propto \frac{e^{-vf(m/v)+\mu(\rho)m}}{(a+m)^{2.5}}, \tag{22}$$

where the denominator is essentially a subleading correction to the free energy function, with $a \sim \mathcal{O}(v)$ being a model-dependent cutoff mass. The correction term is obtained from the fact that, for large masses $m \gg v$, the free energy function $f(x)$ has a linear branch [see Eq. (8)] and the mass distribution function must have the asymptotic form $P_v(m) \sim m^{-\tau}\exp(\tilde{\mu}m)$ as in Eq. (13).

III. MODELS AND ILLUSTRATION

A. Conserved-mass aggregation models

We now substantiate the above claims in a broad class of nonequilibrium models that have been studied intensively in the past couple of decades. In this paper we mainly focus on the models having multipole singularity in the variance. Other singularities, e.g., exponential or logarithmic, are possible, however they are not as common as the multipole one. For the purpose of illustration, we specifically consider the case with $n = 1$ and mass distribution at a single site, i.e., $v = 1$. We define a generalized version of the conserved-mass aggregation models (CMAMs) studied in [19–21,34], for simplicity on a one-dimensional lattice of L sites. We mainly focus on symmetric mass transfer, i.e., masses are transferred symmetrically to either of the neighbors. Here masses (or particles) diffuse, fragment, and coalesce stochastically with either of the nearest-neighbor masses according to the following dynamical rules: (i) diffusion of mass m_i at site i to $i \pm 1$ with mass-dependent rate $D(m_i)$, where $m_i \to 0$ and $m_{i\pm1} \to m_{i\pm1} + m_i$, and (ii) fragmentation of a discrete mass Δ at site i, provided $\Delta \leqslant m_i$, and coalescence of the mass to either of the sites $i \pm 1$ with mass-dependent rate $w(\Delta)$, where $m_i \to m_i - \Delta$ and $m_{i\pm1} \to m_{i\pm1} + \Delta$ with $\Delta = 1,2,\ldots$ (continuous Δ considered later). The total mass $M = \sum_{i=1}^{V} m_i$ is conserved in this process. Even for these simple dynamical rules, the steady-state weight in general is not exactly known. However, as spatial correlations are small, the additivity property as in Eq. (1), to a good approximation, is expected to hold.

We calculate the variance $\sigma^2(\rho)$ of mass at a single site in various special cases, using the additivity property (1) with $v = 1$. We take the diffusion rate $D(m_i) = 1$, independent of mass m_i, $w(\Delta = 1) = w_1$ (rate of single-particle chipping), $w(\Delta = m_i - 1) = w_2$ (rate of all-but-one-particle chipping), and $w(\Delta) = 0$ otherwise.

1. Case I: The CMAM with $\mathbf{w_1 = 1, w_2 = 0}$

For $w_1 = 1$ and $w_2 = 0$ and symmetric mass transfer, the model becomes the symmetric one studied in [19,21] (our model is a variant of those studied in [15,16]). For $\rho \leqslant \rho_c$, using the additivity property, we exactly calculated the variance and consequently chemical potential with the critical density $\rho_c = \sqrt{2} - 1$. The variance is given by (for details, see the Appendix, Sec. 1)

$$\begin{aligned}\sigma^2(\rho) &= \frac{\rho(1+\rho)(1+\rho^2)}{1-2\rho-\rho^2} \\ &= \frac{\rho(1+\rho)(1+\rho^2)}{(\rho_c-\rho)(\sqrt{2}+1+\rho)},\end{aligned} \tag{23}$$

with $\rho_c = \sqrt{2} - 1$, for which one can obtain a chemical potential $\mu(\rho)$ and free energy function $f(\rho)$, by integrating the fluctuation-response relation as in Eq. (2)

$$\mu(\rho) = \int \frac{1}{\sigma^2(\rho)}d\rho = -2\tan^{-1}\rho + \ln\left(\frac{\rho}{1+\rho}\right) + \alpha \tag{24}$$

and, upon one more integration,

$$\begin{aligned}f(\rho) = \int \mu(\rho)d\rho &= -2\rho\tan^{-1}\rho + \rho\ln\left(\frac{\rho}{1+\rho}\right) \\ &\quad - \ln\left(\frac{1+\rho}{1+\rho^2}\right) + \alpha\rho + \beta,\end{aligned} \tag{25}$$

where α and β are two arbitrary constants of integration. For large mass $m \gg 1$, the mass distribution function is calculated to be $P_1(m) \propto m^{-5/2}\exp\{[\mu(\rho) - \mu(\rho_c)]m\}$ (for details, see the Appendix, Sec. 2).

In Fig. 2(a) we have plotted the single-site ($v = 1$) mass distribution function $P_1(m)$, obtained from simulations, as a function of mass m for various values of densities $\rho = 0.1, 0.2, 0.3, 0.414$, and 1.0 and compared them with the theoretical expression in Eq. (13). The theoretical results are in quite good agreement with the simulation results, especially at large mass $m \gg 1$. In Fig. 3(a) we have plotted the subsystem mass distribution function $P_v(m)$, with $v = 100$, for densities $\rho = 0.1$ and 0.2 and compared them with the theoretical expression in Eq. (22) with the cutoff mass $a \approx 20$; the agreement between simulations and theory is reasonably good.

2. Case II: The CMAM with $\mathbf{w_1 = 0, w_2 = 1}$

The CMAM with $w_1 = 0, w_2 = 1$ is a variant of the models studied in [20]. In this case, for $\rho \leqslant \rho_c$, the variance and chemical potential can be exactly obtained using the additivity property (for details see the Appendix, Sec. 1). The variance is given by

$$\begin{aligned}\sigma^2(\rho) &= \frac{\rho(1-\rho)(2\rho^2-2\rho+1)}{2\rho^2-4\rho+1} \\ &= \frac{\rho(1-\rho)(2\rho^2-2\rho+1)}{(\rho_c-\rho)(2+\sqrt{2}-2\rho)}.\end{aligned} \tag{26}$$

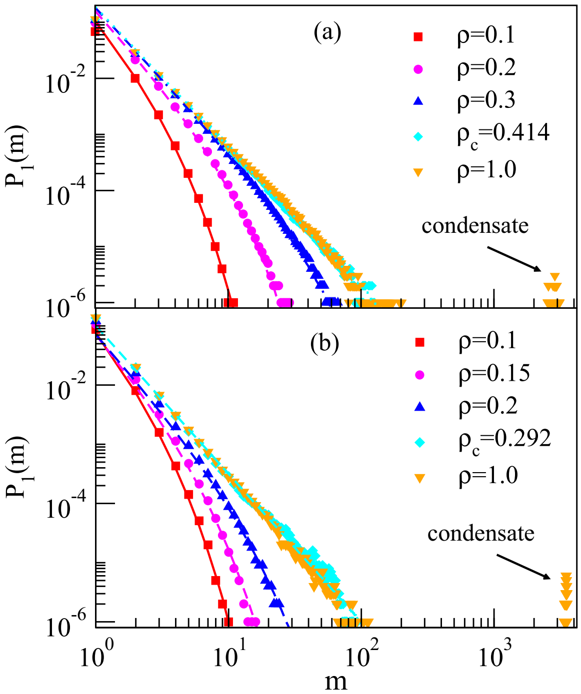

FIG. 2. (Color online) Single-site ($v = 1$) mass distribution functions $P_1(m)$ (points represent simulations) in CMAMs compared with the analytic expression in Eq. (13) (lines represent theory) for various densities and (a) mass chipping rates $w_1 = 1$ and $w_2 = 0$ and (b) mass chipping rates $w_1 = 0$ and $w_2 = 1$. In all cases, the mass diffusion rate $D = 1$ and the system size $L = 5000$.

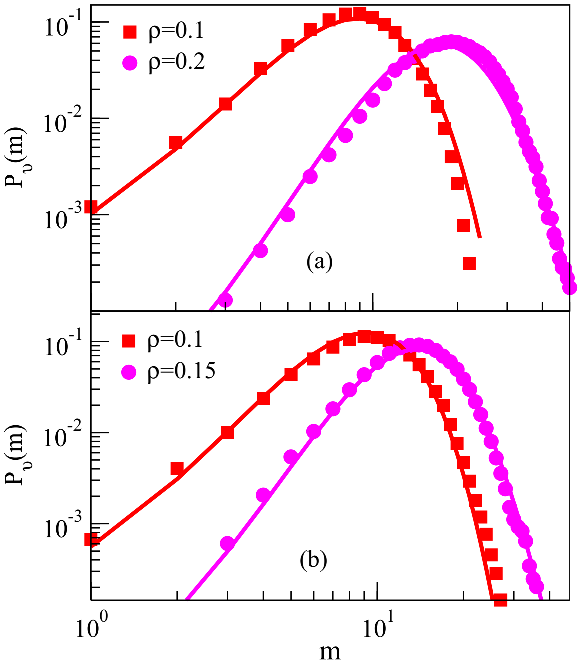

FIG. 3. (Color online) Subsystem mass distribution functions $P_v(m)$ (points represent simulations) in CMAMs compared with the analytic expression in Eq. (22) (lines represent theory) for various densities and cutoff mass (a) mass chipping rates $w_1 = 1$ and $w_2 = 0$ and $a \approx 20$ and (b) mass chipping rates $w_1 = 0$ and $w_2 = 1$ and cutoff mass $a \approx 25$. In all cases, the mass diffusion rate $D = 1$, the system size $L = 10^5$, and the subsystem size $v = 100$.

There is a simple pole at the critical density $\rho_c = 1 - 1/\sqrt{2}$. By integrating the fluctuation-response relation (3), we get the chemical potential

$$\mu(\rho) = 2\tan^{-1}(1-2\rho) - \ln\left[\frac{1}{2\rho(1-\rho)} - 1\right] + \alpha \qquad (27)$$

and then the free energy density

$$f(\rho) = 2\rho\tan^{-1}(1-2\rho) - \ln(1-\rho) + \ln(1-2\rho+2\rho^2) - \rho\ln\left[\frac{1}{2\rho(1-\rho)} - 1\right] + \alpha\rho + \beta. \qquad (28)$$

In Fig. 2(b) we have plotted the single-site ($v = 1$) mass distribution function $P_1(m)$, obtained from simulations, for various values of densities $\rho = 0.1$, 0.15, 0.2, 0.292, and 1.0. We find that the simulation results agree remarkably well with the analytical scaling form $P_1(m) \propto m^{-5/2}\exp\{[\mu(\rho) - \mu(\rho_c)]m\}$ as in Eq. (13) with $\tau = 5/2$ (for details of the derivation, see the Appendix, Sec. 2). In Fig. 3(b) we have plotted the subsystem mass distribution function $P_v(m)$ for $v = 100$ for densities $\rho = 0.1$ and 0.15 and compared them with theory, Eq. (22) with $a \approx 25$; the agreement between simulations and theory is reasonably good.

3. Other cases

We have also studied, through simulations, various other cases (with $D = 1$): case III, $w(\Delta = 1) = w_1$, $w(\Delta = 2) = w_2$, and $w(\Delta) = 0$ otherwise; case IV, a discrete-mass model with $w(\Delta) = \exp(-\Delta)$; and case V, a continuous-mass model with $w(\Delta) = \exp(-\Delta)$. In these cases, in the absence of an analytical expression of $\sigma^2(\rho)$, we checked in simulations (see Fig. 4) that the variance near the critical point indeed has the behavior $\sigma^2 \sim (\rho_c - \rho)^{-n}$, with $n = 1$, which therefore leads to the same power-law exponent $\tau = 5/2$.

One can also define an asymmetric version of the CMAMs discussed above. In one dimension, there are some nontrivial spatial correlations and the above mean-field analysis fails to capture the mass fluctuations in the system. However, in higher dimensions, the above results qualitatively remain the same also for the asymmetric mass transfer and is consistent with [24].

Interestingly, the exponent $\tau = 5/2$ appears also in the distribution of particle numbers in an ideal Bose gas in three dimensions near the critical point where Bose-Einstein condensation (BEC) occurs. This could be easily understood from the fact that the particle-number fluctuation in the case of a three-dimensional Bose gas has the same critical behavior $\sigma^2(\rho) \sim (\rho_c - \rho)^{-n}$, with $n = 1$, as in these mean-field nonequilibrium systems having negligible spatial correlations. That, on a mean-field level, the nonequilibrium aggregation models belong to the universality class of equilibrium Bose gas in three dimensions, so far has not been realized.

It is quite instructive to consider a limiting case of Eq. (7) with $n = 0$, $\rho_c = \infty$, and $g(\rho) \sim \rho^{1-\delta}$ at large density, i.e., the variance $\sigma^2(\rho) \sim \rho^{1-\delta}$, with $\delta < -1$, diverges algebraically at infinite density. As there is no singularity in the variance at any finite density, our analysis quite straightforwardly shows that condensation transition cannot occur, consistent with the past observations in the CMAM with mass-dependent diffusion [35]. Asymptotic scaling of the mass distribution can be

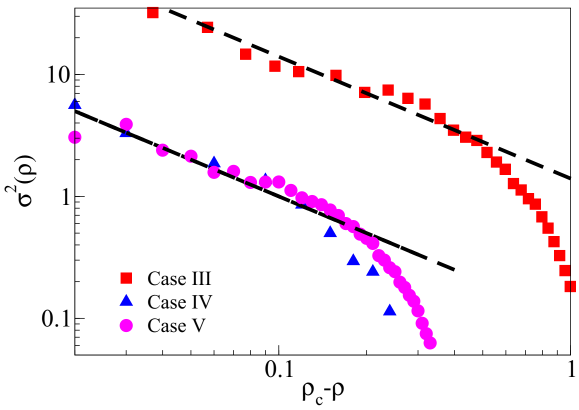

FIG. 4. (Color online) Variance $\sigma^2(\rho)$ vs $(\rho_c - \rho)$. The black line is const $\times$ $(\rho_c - \rho)^{-n}$ with $n = 1$. Red rectangles are for case III (one- and two-particle fragmentation), blue triangles are for case IV [$\Delta = 1,2,\ldots$ is discrete with fragmentation rate $w(\Delta) = \exp(-\Delta)$], and red circles are for case V [$\Delta > 0$ is continuous with fragmentation rate $w(\Delta) = \exp(-\Delta)$]. The diffusion rate $D(m) = 1$ throughout.

obtained as follows. Using Eq. (3), we get $\mu(\rho) \sim \rho^{\delta}$ (setting $\alpha = 0$ without loss of any generality) and, consequently, a Laplace transform of the weight factor $w_v(m)$,

$$\tilde{w}_v(s) \simeq a_0 + a_1 s^{1+1/\delta}, \tag{29}$$

immediately leading to mass distributions having a scaling form $P_v(m) \propto m^{-\tau}\exp(\tilde{\mu}m) \equiv (m^*)^{-\tau}\Phi(m/m^*)$. Here the scaling function $\Phi(x) = x^{-\tau}\exp(-x)$ with $m^* \sim \rho^{-\delta}$ and power law exponent $\tau = 2 + 1/\delta$ with $1 < \tau < 2$ (as $\delta < -1$), leading to a relation $\delta(\tau - 2) = 1$. The scaling form was numerically observed in [35]. Interestingly, the borderline case with $\delta = -1$ generates gamma distributions, which are found in a broad class of mass transport processes [31] and have been also observed in a limiting case of the conserved-mass aggregation models studied in Ref. [35].

B. Nonconserved-mass aggregation models

In this section we discuss a nonconserved version of the mass aggregation models where systems can exchange mass, though weakly, with environment. In this case, in addition to the earlier defined processes of (i) diffusion and (ii) fragmentation of masses, a particle now can be adsorbed with rate q and desorbed at a site with rate p, provided the site is occupied, where $p,q \to 0$ (i.e., weak exchange) with the ratio $r = q/p$ finite. Due to adsorption and desorption processes, total mass in the system is not conserved. This model is related to several models studied in the past for finite p and q [17,19,34,36]. Interestingly, in the limit of $p,q \to 0$, mass fluctuation in a nonconserved model can be obtained from the occupation probability of a site in its conserved version (i.e., $p = q = 0$) [37,38]. Let us first define, in the space of total mass M, a transition rate $T_{M+1,M}$ from mass M to $M+1$. In the steady state, the probability $P(M)$ that the system has mass M satisfies a balance condition

$$P(M)T_{M+1,M} = P(M+1)T_{M,M+1},$$

where the mass distribution $P(M)$ can be obtained as

$$P(M+1) = \left[\prod_{M=0}^{M} \frac{T(M \to M+1)}{T(M+1 \to M)}\right] P(0). \tag{30}$$

As the ratio of transition rates can be written as

$$\frac{T_{M+1,M}}{T_{M,M+1}} = \frac{q}{p\mathcal{S}(\rho)},$$

where $\mathcal{S}(\rho)$ is the occupation probability and $\rho = M/V$, the distribution function can be written, up to a normalization factor, as

$$P(M) \propto \exp\left(\sum_M [\ln(q/p\mathcal{S})]\right)$$
$$\simeq \exp\left(-V\int_0^{\rho} d\rho[\mu(\rho) - \mu_0]\right), \tag{31}$$

where $\mu_0 = \ln(q/p)$ is an effective chemical potential and $f(\rho) = \int d\rho\, \mu(\rho) = \int d\rho \ln \mathcal{S}(\rho)$ is an effective free energy (canonical) density function. The steady-state mass density as a function of adsorption to desorption ratio $r = q/p$ can be obtained by minimizing the grand potential or the large deviation function for the density fluctuation $h(\rho) = f(\rho) - \mu_0\rho$, leading to the relation $\mathcal{S}(\rho) = r$ (for details, see the Appendix, Sec. 3).

Until now, the analysis has been exact. However, it may not always be possible to exactly calculate the occupation probability $\mathcal{S}(\rho)$. For the purpose of demonstration, let us proceed by considering a model with diffusion and fragmentation rate as in case I. We obtain an approximate expression, obtained within mean-field theory, of the occupation probability (see the Appendix, Sec. 3)

$$\mathcal{S}(\rho) = \frac{\rho(1-\rho)}{(1+\rho)}.$$

Equation (31) implies the subsystem mass distribution having a form $P_v(m) \propto w_v(m)\exp(\mu m)$ and consequently a FR relation as in Eq. (3) follows. Then, for $\rho < \rho_c$ or equivalently for $r < r_c$, one can immediately calculate the scaled variance as

$$\sigma^2(\rho) = \left(\frac{d\mu_0}{d\rho}\right)^{-1} = \frac{\rho(1-\rho)(1+\rho)}{1 - 2\rho - \rho^2}, \tag{32}$$

where critical density $\rho_c = \sqrt{2} - 1$. The variance in the nonconserved case is different from that in the conserved-mass case, implying that the canonical and grand canonical ensembles are not equivalent [37,38]. However, the nature of singularity in the variance remains the same near criticality where $\sigma^2(\rho) \sim (\rho_c - \rho)^{-n}$ with $n = 1$. Therefore, the additivity property leads to the same power-law scaling in the single-site mass distribution $P_1(m) \sim m^{-\tau}\exp(\tilde{\mu}m)$ for large m, where $\tau = 5/2$ and $\tilde{\mu} = \mu_0 - \ln \mathcal{S}(\rho_c) = \ln(r/r_c)$ with $r_c = \mathcal{S}(\rho_c)$.

The above results are consistent with what was found, on the mean-field level, for general p and q in the in-out model [34], a special case of the above nonconserved model with $w = 0$. One can interpret the results in the light of equilibrium BEC: The critical density signifies that, for $r > r_c = \mathcal{S}(\rho_c) = 3 - 2\sqrt{2}$, there is a condensate as in the BEC. In the grand-canonical setting (i.e., with no mass conservation), that would imply a phase with a diverging mass density, similar to the Takayasu phase where mass density actually diverges. For p and q finite, the form of the subsystem mass distribution as written in Eq. (31) remains the same, but only the expression of $\mathcal{S}(\rho)$, due to the presence of spatial correlations, is different. However, the similarity to the BEC still persists.

IV. SUMMARY AND CONCLUDING PERSPECTIVE

In this paper we argued that an additivity property can possibly explain why simple power-law scaling appears generically in nonequilibrium steady states with short-range correlations. We demonstrate that the existence of a fluctuation-response relation, a direct consequence of additivity, with a singular response function leads to power-law distributions with nontrivial exponents. The simplest form of the singularity, a simple pole, gives rise to the exponent 5/2, which was often observed in the past in apparently unrelated systems. We substantiate the claims by analytically calculating the response function, which diverges as critical point is approached, in paradigmatic nonequilibrium mass aggregation models and the corresponding single-site and subsystem mass distributions. Most remarkably, the analysis, being independent of dynamical rules in a particular system, equally extends to critical properties in equilibrium and nonequilibrium.

Thermodynamic characterization of phase coexistence in driven systems is a fundamental problem in statistical physics and has been addressed in the past [27,28,39–43], either numerically or analytically for exactly known steady states mostly having a product measure. From that perspective, it is quite encouraging that, even when steady-state weights are *a priori* not known, our analytical method not only gives insights into the steady-state structure but can also be applied to identify a chemical potential, which equalizes in the coexisting phases and whose vanishing at criticality gives rise to pure power laws.

Note that, in our formulation, the mass distribution functions, though approximate, have been calculated solely from the knowledge of the variance. This formulation is perhaps not surprising in equilibrium where the free energy function (or entropy, for an isolated system) essentially determines the fluctuation properties of a system. However, in the nonequilibrium scenario, it is *a priori* not clear that such an equilibrium thermodynamic approach can indeed be applied in systems having a steady state with nontrivial spatial structure. Here it is worth mentioning that one requires, in principle, all the moments to specify a probability distribution function. However, the additivity property, provided it holds, puts a strong constraint on the mass distribution function $P_v(m)$ through a fluctuation-response relation and thus helps to uniquely determine $P_v(m)$, only from the knowledge of the variance as a function of density.

We believe that our analysis, being based on a general thermodynamic principle, would be applicable in many other driven systems where phase coexistence is known to occur (e.g., in active matters [44,45]). As a concluding remark, we mention that the additivity property is expected to be quite generic for systems having short-range correlations and therefore it would be indeed interesting to actually verify additivity, through the predictions concerning fluctuations, on a case-by-case basis. Also, it remains to be seen whether the principle of additivity can be extended to systems having long-range spatial correlations, at least in the cases where the strength of these correlations is weak.

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APPENDIX

In this appendix we provide the details of the calculations to obtain the mass distributions, using the additivity property, in mass aggregation models (both conserved and nonconserved versions), which were studied over the past couple of decades. The generalized models introduced here cover some of those studied in the past and their variants [15,16,19–21,24,34].

1. Calculation of variance in conserved-mass aggregation models

We define here a class of CMAMs on a one-dimensional lattice with a periodic boundary and calculate the variance of mass at a single site in the steady state, assuming that the additivity property (1) holds. For, simplicity, we consider only the discrete-mass cases.

The mass at each site undergoes either diffusion (where whole of the mass is transferred to either of neighboring sites) or chipping, with certain transition rates; in the models considered below, there are two types of chipping process. The diffusing mass or the chipped-off mass coalesce with the mass at either of the neighboring sites with a preassigned rate. In this process, the total mass of the system is conserved.

Provided a site i is occupied, particles hop to either of the nearest-neighbor sites according to the transition rates specified below.

(a) Diffusion with rate 1. All particles at a site i hop with rate 1 to the left or right, i.e., $m_i \to 0$ and $m_{i\pm1} \to m_{i\pm1} + m_i$.

(b) Chipping with rate w_1. This chipping process involves a particle at site i being chipped off and thrown to the left or right neighbor, i.e., $m_i \to (m_i - 1)$ and $m_{i\pm1} \to m_{i\pm1} + 1$.

(c) Chipping with rate w_2. This chipping process involves $m_i - 1$ particles going to either the left or right neighbor and the rest of the particles remaining at site i, i.e., $m_i \to 1$ and $m_{i\pm1} \to m_{i\pm1} + m_i - 1$.

Assuming transition rates are Poissonian, we have the following stochastic update rules where mass $m_i(t + dt)$ at time $t + dt$ takes a particular value, depending on mass $m_i(t)$ at time t, with certain probabilities as shown below: loss terms at site i,

$$m_i(t+dt) = \begin{cases} 0 & \text{with probability } dt \\ m_i(t) - 1 + \delta_{m_i(t),0} & \text{with probability } w_1 dt \\ 1 - \delta_{m_i(t),0} & \text{with probability } w_2 dt; \end{cases}$$

gain terms from the $(i-1)$th site,

$$m_i(t+dt)=\begin{cases} m_i(t)+m_{i-1}(t) & \text{with probability } \frac{dt}{2}\\ m_i(t)+1-\delta_{m_{i-1}(t),0} & \text{with probability } w_1\frac{dt}{2}\\ m_i(t)+m_{i-1}(t)-1+\delta_{m_{i-1}(t),0} & \text{with probability } w_2\frac{dt}{2}; \end{cases}$$

gain terms from the $(i+1)$th site,

$$m_i(t+dt)=\begin{cases} m_i(t)+m_{i+1}(t) & \text{with probability } \frac{dt}{2}\\ m_i(t)+1-\delta_{m_{i+1}(t),0} & \text{with probability } w_1\frac{dt}{2}\\ m_i(t)+m_{i+1}(t)-1+\delta_{m_{i+1}(t),0} & \text{with probability } w_2\frac{dt}{2}; \end{cases}$$

and mass remains unchanged at site i,

$$m_i(t+dt)=m_i(t) \quad \text{with probability } (1-2dt-2w_1dt-2w_2dt).$$

Now we define the occupation probability $\langle(1-\delta_{m_j,0})\rangle=\mathcal{S}(\rho)$, i.e., the probability that a site is occupied. We deal with steady-state averages throughout. We assume that the additivity property [as in Eq. (1)] is valid at the single-site level and therefore the n-point ($n\geqslant 2$) correlation factorizes.

(i) The nth-moment equation. The time evolution of the nth moment $\langle m_i^n\rangle$ can be written as

$$\begin{aligned}\langle m_i^n(t+dt)\rangle=\langle m_i^n(t)\rangle=&\ \langle[m_i(t)-1+\delta_{m_i(t),0}]^n\rangle w_1dt+\langle[m_i(t)+m_{i-1}(t)]^n\rangle\frac{dt}{2}+\langle[1-\delta_{m_i(t),0}]^n\rangle w_2dt\\ &+\langle[m_i(t)+1-\delta_{m_{i-1}(t),0}]^n\rangle w_1\frac{dt}{2}+\langle[m_i(t)+m_{i-1}(t)-1+\delta_{m_{i-1}(t),0}]^n\rangle w_2\frac{dt}{2}\\ &+\langle[m_i(t)+m_{i+1}(t)]^n\rangle\frac{dt}{2}+\langle[m_i(t)+1-\delta_{m_{i+1}(t),0}]^n\rangle w_1\frac{dt}{2}\\ &+\langle[m_i(t)+m_{i+1}(t)-1+\delta_{m_{i+1}(t),0}]^n\rangle w_2\frac{dt}{2}\\ &+\langle m_i^n(t)\rangle(1-2dt-2w_1dt-2w_2dt),\end{aligned}\tag{A1}$$

which, in the steady state where $\langle m_i^n(t+dt)\rangle=\langle m_i^n(t)\rangle$, gives a Bogoliubov-Born-Green-Kirkwood-Yvon hierarchy where n-point correlations are coupled to $(n+1)$-point correlations. To get a closed set of equations for the moments, we use the factorization property of n-point correlations. As mentioned in this paper, the mass distributions are solely obtained from the response function (or the variance of the mass distribution) and therefore we are interested in only calculating the variance, or equivalently the second moment, which can be done as follows.

(ii) Second-moment equation. If we set $n=2$ in the above equation, the second moment $\langle m_i^2\rangle$ however cancels out from the above equation. Using factorization of the two-point correlation, i.e., $\langle m_im_j\rangle\approx\rho^2$ for $i\neq j$, we get an expression for the occupation probability $\mathcal{S}(\rho)$ as a function of mass density ρ,

$$\rho^2(1+w_2)=w_+(\rho-\mathcal{S})-w_-\rho\mathcal{S},\tag{A2}$$

where $w_\pm=w_1\pm w_2$. This gives

$$\mathcal{S}(\rho)=\frac{w_+\rho-(1+w_2)\rho^2}{w_++w_-\rho}.\tag{A3}$$

(iii) Third-moment equation. Similarly, for $n=3$, we obtain an equation where the third moment $\langle m_i^3\rangle$ cancels out and we actually get, using factorization of both two-point and three-point correlations, a relation for the second moment

$$\langle m^2\rangle=\rho\frac{w_+(1+\mathcal{S})-2w_2\rho}{w_+-2(1+w_2)\rho-w_-\mathcal{S}}.\tag{A4}$$

Using the expression of the occupation probability in Eq. (A3), we obtain

$$\langle m^2\rangle=\rho\frac{w_+^2+2w_+w_-\rho-\left(w_++3w_1w_2-w_2^2\right)\rho^2}{w_+^2-2w_+(1+w_2)\rho-w_-(1+w_2)\rho^2},\tag{A5}$$

which leads to the desired expression of the variance as a function of density

$$\sigma^2(\rho)=\frac{w_+^2\rho+w_+(w_1-3w_2)\rho^2}{w_+^2-2w_+(1+w_2)\rho-w_-(1+w_2)\rho^2}+\frac{\left(w_+-w_1w_2+3w_2^2\right)\rho^3+w_-(1+w_2)\rho^4}{w_+^2-2w_+(1+w_2)\rho-w_-(1+w_2)\rho^2}.\tag{A6}$$

The variance $\sigma^2(\rho)$ has a singularity at $\rho=\rho_c$, i.e., it diverges at a critical density $\rho=\rho_c$, which can be obtained by setting the denominator of Eq. (A6) equal to zero and solving

$$w_+^2-2w_+(1+w_2)\rho_c-w_-(1+w_2)\rho_c^2=0.\tag{A7}$$

This gives a simple pole at the critical density

$$\rho_c=\frac{w_+}{w_-}\left(\sqrt{1+\frac{w_-}{1+w_2}}-1\right).\tag{A8}$$

The nonequilibrium free energy function can be calculated by integrating the nonequilibrium chemical potential with respect to density ρ,

$$\mu(\rho) = \frac{df}{d\rho} \Rightarrow f(\rho) = \int \mu(\rho) d\rho. \tag{A9}$$

The function $\lambda_v(s) = -\ln \tilde{w}(s)$, which is the Legendre transform of the free energy density $f(\rho)$, can be obtained as

$$\lambda_v(s) = v[f(\rho^*) + s\rho^*], \tag{A10}$$

where ρ^* is the solution of

$$s = -\mu(\rho^*). \tag{A11}$$

2. Calculation of mass distribution in the conserved-mass aggregation model

Here we provide the essential steps of the calculations to obtain the single-site (i.e., $v = 1$) mass distribution function $P_1(m) \propto w_1(m)\exp[\mu(\rho)m]$, where $w_1(m)$ is the single-site weight factor and $\mu(\rho)$ is a chemical potential. We first analyze the behavior of $\lambda_1(s)$ near the singularity at $s = s_c$ by expanding $\mu(\rho)$ and $f(\rho)$ near critical density in the power series of $\rho - \rho_c$, where $\rho - \rho_c < 0$ is small,

$$\begin{aligned} \mu(\rho) &= \mu(\rho_c) + \frac{\mu''(\rho_c)}{2}(\rho - \rho_c)^2 + \cdots, \\ f(\rho) &= f(\rho_c) + \mu(\rho_c)(\rho - \rho_c) + \frac{f'''(\rho_c)}{3!}(\rho - \rho_c)^3 + \cdots, \end{aligned} \tag{A12}$$

where we have used Eq. (A9) and $\mu'(\rho_c) = f''(\rho_c) = 0$ (see Fig. 1). Using Eq. (A11) in Eq. (A12) and substituting $s + \mu(\rho_c) \simeq -\mu''(\rho_c)(\rho^* - \rho_c)^2/2$, we get

$$\rho^* - \rho_c = -\sqrt{\frac{2}{|\mu''(\rho_c)|}}(s - s_c)^{1/2}, \tag{A13}$$

where $s_c = -\mu(\rho_c)$ and $\mu''(\rho_c) < 0$. Therefore, $\lambda_1(s) = f(\rho^*) + s\rho^*$ near $s = s_c$, in the leading order of $s - s_c$, can be approximated as

$$\begin{aligned} \lambda_1(s) &\simeq \left[f(\rho_c) - s_c(\rho^* - \rho_c) + \frac{f'''(\rho_c)}{3!}(\rho^* - \rho_c)^3 \right] + s\rho^* \\ &= \lambda_1(s_c) + \rho^*(s - s_c) + \frac{f'''(\rho_c)}{3!}(\rho^* - \rho_c)^3 \\ &= [a_0 + a_1(s - s_c) + a_2(s - s_c)^{3/2}], \end{aligned} \tag{A14}$$

where $a_0 = \lambda_1(s_c) = f(\rho_c) + s_c\rho_c$, $a_1 = \rho_c$, and $a_2 = -(2/3)\sqrt{2/|\mu''(\rho_c)|}$. The inverse Laplace transform of the weight factor $w_1(m)$ can be written as

$$\tilde{w}_1(s) = e^{-\lambda_1(s)} \simeq e^{-a_0}[1 - a_1(s - s_c) - a_2(s - s_c)^{3/2}], \tag{A15}$$

which, for $m \gg 1$, translates into

$$w_1(m) \sim \frac{e^{s_c m}}{m^{5/2}}. \tag{A16}$$

Consequently, the mass distribution can be written as

$$P_1(m) \sim \frac{e^{s_c m}}{m^{5/2}} e^{\mu(\rho)m} = \frac{e^{-(\alpha+\mu_0(\rho_c))m}}{m^{5/2}} e^{[\mu_0(\rho)+\alpha]m}, \tag{A17}$$

$$P_1(m) \sim \frac{1}{m^{5/2}} e^{[\mu_0(\rho)-\mu_0(\rho_c)]m}. \tag{A18}$$

Note that effective chemical potential $\tilde{\mu}(\rho) = \mu_0(\rho) - \mu_0(\rho_c)$ is zero at the critical density $\rho_c = \sqrt{2} - 1$. The mass distribution in Eq. (A18) is precisely what was found in [19] at $\rho = \rho_c$ and describes the simulation data remarkably well (see Fig. 1).

3. Calculation of mass distribution in the absence of mass conservation

As shown in the paper, the probability distribution function $P(M)$ of total mass M can be written, up to a normalization factor, as

$$P(M) = \text{const} \times \exp\left(-V \int_0^\rho d\rho[\mu(\rho) - \mu_0]\right). \tag{A19}$$

If we assume that the joint mass distribution $\mathcal{P}[\{m_i\}]$ has a product form on the single-site level ($v = 1$), i.e., the product of the single-site mass distribution function $p(m_i)$,

$$\mathcal{P}[\{m_i\}] = \prod_{i=1}^{V} p(m_i), \tag{A20}$$

the probability distribution function $P(M)$ of mass M in the system can be written as

$$P(M) = \prod_{i=1}^{V}\left[\int dm_i\, p(m_i)\right]\delta\left(M - \sum_i m_i\right). \tag{A21}$$

From the Laplace transform $\tilde{P}(s) = \int dM P(M) \exp(-sM) = [\tilde{p}(s)]^V$ of the mass distribution $P(M)$, the Laplace transform $\tilde{p}(s) = \int dm_i\, p(m_i)\exp(-sm_i)$ of the single-site mass distribution $p(m)$ can be written as

$$\tilde{p}(s) = \text{const} \times e^{-\lambda_1(s)}, \tag{A22}$$

where

$$\lambda_1(s) = \inf_\rho[h(\rho) + s\rho]. \tag{A23}$$

Here we have used inverse transform

$$\tilde{P}(s) = \text{const} \times \int d\rho\, e^{-V[h(\rho)+s\rho]}, \tag{A24}$$

which has been obtained from Eq. (A19) and where the grand potential or the large deviation function for the density fluctuation $h(\rho) = f(\rho) - \mu_0\rho = \int_0^\rho [\mu(\rho) - \mu_0]d\rho$ and chemical potential $\mu(\rho) = \ln \mathcal{S}(\rho) = \ln[\rho(1 - \rho)/(1 + \rho)]$, as given in the present paper. Note that the function $\mathcal{S}(\rho)$ is the occupation probability in the conserved-mass aggregation model and has been obtained by setting $w_1 = 1$ and $w_2 = 0$ in Eq. (A3).

Now the function $\lambda_1(s)$, the Legendre transform of the grand potential $h(\rho)$, can be written as

$$\lambda_1(s) = h(\rho^*) + s\rho^*, \tag{A25}$$

where ρ^* is the root of the equation $d[h(\rho) + s\rho]/d\rho = 0$ or $\mu(\rho^*) - \mu_0 + s = 0$, i.e., ρ^* is the root of

$$\ln\left[\frac{\rho^*(1 - \rho^*)}{1 + \rho^*}\right] = \mu_0 - s. \tag{A26}$$

The critical density is obtained by setting the scaled variance as $\sigma^2(\rho) = (d\mu_0/d\rho)^{-1} = \infty$ or $1/\sigma^2(\rho) = 0$,

$$\frac{(1 - 2\rho_c - \rho_c^2)}{\rho_c(1 - \rho_c)(1 + \rho_c)} = 0, \tag{A27}$$

and thus $\rho_c = \sqrt{2} - 1$. In the macrostate (most probable state) we have $\mathcal{S}(\rho) = r$, implying that the critical density is related to the ratio $r = q/p$ through $\mathcal{S}(\rho_c) = r_c$. To obtain the large-mass behavior, we expand $\mu(\rho)$ around $\rho = \rho_c$,

$$\mu(\rho) = \mu(\rho_c) + \frac{\mu''(\rho_c)}{2}(\rho - \rho_c)^2, \tag{A28}$$

to obtain

$$s - s_c \simeq \frac{|\mu''(\rho_c)|}{2}(\rho^* - \rho_c)^2, \tag{A29}$$

$$\lambda_1(s) \simeq a_0 + a_1(s - s_c) + a_2(s - s_c)^{3/2}, \tag{A30}$$

to leading order in $\rho^* - \rho_c$, where $s_c = \mu_0 - \mu(\rho_c)$, leading to the desired result in the paper,

$$p(m) \sim \frac{1}{m^{5/2}} e^{s_c m} = \frac{1}{m^{5/2}} e^{[\mu_0 - \mu(\rho_c)]m}. \tag{A31}$$

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