Aggregation with Multiple Conservation Laws

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Aggregation processes with an arbitrary number of conserved quantities are investigated. On the mean-field level, an exact solution for the size distribution is obtained. The asymptotic form of this solution exhibits nontrivial "double" scaling. While processes with one conserved quantity are governed by a single scale, processes with multiple conservation laws exhibit an additional diffusion-like scale. The theory is applied to ballistic aggregation with mass and momentum conserving collisions and to diffusive aggregation with multiple species.

I. INTRODUCTION

Irreversible aggregation processes underly many natural phenomena including, *e. g.*, polymerization [1], gelation [2], island growth [3-4] and aerosols [5]. The classical rate theory of Smoluchowski describes the kinetics of such processes [6-9]. Recently, scaling [10-16] and exact [17-23] theoretical studies showed that spatial correlations play a crucial role in low dimensions. While the above examples are diffusive driven, there are physical situations such as formation of large-scale structure of the universe [24] and clustering in traffic flows [25], where the aggregates move ballistically. So far, theories of ballistic aggregation [26] have been restricted to scaling arguments [26-30].

In the ballistic aggregation process both the mass and the momentum are conserved. In polymerization processes involving copolymers, each monomer species mass is a conserved quantity. Hence, we study aggregation processes with multiple conservation laws. The simplest example for such a system is aggregation with k distinct species. Both the multivariate distribution and the single variable distributions are of interest. We present exact solutions to the time dependent and the steady state mean-field rate equations. Although they are straightforward generalizations to the well known solutions they exhibit interesting behaviors. An asymptotic analysis shows that fluctuations associated with a single conserved quantity are Gaussian in nature. As a result, an additional "diffusive" size scale emerges.

We apply the above theory to ballistic aggregation as well as diffusive aggregation. In the case of ballistic aggregation, we use an approximate collision rate to obtain a solution to the Boltzmann equation in arbitrary dimension. While this approach agrees with the scaling arguments, it suggests that for a given mass, the momentum distribution is Gaussian. We compare these predictions with one and two dimensional simulations. Furthermore, we consider steady state properties of the aggregation process by introducing input of particles. For homogeneous input, a novel time scale describing the density relaxation is found. In the case of a localized input, clustering occurs only for $d \leq 2$. Additionally, we apply the theory to two-species aggregation with diffusing particles. Using the density dependent reaction rate, we obtain the leading scaling behavior of the two relevant mass scales.

This paper is organized as follows. In section II, we present exact solutions of the rate equation theory. We investigate time dependent as well as steady state properties of the process. We then apply the theory to ballistic aggregation with momentum conserving collisions (section III) and to diffusive driven aggregation (section IV). We conclude with a discussion and suggestions for further research in section V.

II. THEORY

Following the above discussion, there are aggregation processes where several physical quantities are conserved. In such processes, it is natural to label the aggregates by a "mass" vector $\mathbf{m} \equiv (m_1, \cdots, m_k)$, where every component represents a conservation law. Let us denote the probability distribution function for particles of mass $\mathbf{m}$ at time t by $P(\mathbf{m}, t)$. Mean field theory of the binary reaction process assumes that reaction proceeds with a rate proportional to the product of the reactants densities. Thus the mean field approximation neglects spatial correlations and therefore typically holds in dimensions larger than some critical dimension d_c [10, 14-16]. The rate equations [6] are written as

$$\frac{dP(\mathbf{m},t)}{dt} = \sum_{\{m_i'\}} P(\mathbf{m}',t)P(\mathbf{m}-\mathbf{m}',t) - 2P(\mathbf{m},t)\sum_{\{m_i'\}} P(\mathbf{m}',t), \qquad (1)$$

where the sum is carried over all k variables m_i', $1 \leq i \leq k$. The loss term prefactor reflects the fact that two particles are lost in each collision. One can verify that the rate equations conserve each mass separately, *i. e.*, $\sum m_i P(\mathbf{m},t) \equiv \sum m_i P_0(\mathbf{m})$, where $P_0(\mathbf{m})$ is the initial distribution. In writing Eq. (1) we have implicitly assumed that the rate $K(\mathbf{m},\mathbf{n})$ at which the reaction $(\mathbf{m}) + (\mathbf{n}) \to (\mathbf{m}+\mathbf{n})$ proceeds does not depend on masses of the reactants, $K(\mathbf{m},\mathbf{n}) = \text{const}$. This constant can be set to unity without loss of generality. For an arbitrary reaction rate kernel $K(\mathbf{m},\mathbf{n})$, Eq. (1) is easily generalized so that, *e. g.*, the gain term is replaced by $\sum K(\mathbf{m}',\mathbf{m}-\mathbf{m}')P(\mathbf{m}',t)P(\mathbf{m}-\mathbf{m}',t)$.

To solve Eq. (1) we introduce the generating function, $F(\mathbf{z},t)$, defined by

$$F(\mathbf{z},t) = \sum_{\{m_i\}} \mathbf{z}^{\mathbf{m}} P(\mathbf{m},t). \tag{2}$$

In the above equation we have used the shorthand notations $\mathbf{z} \equiv (z_1,\cdots,z_k)$ and $\mathbf{z}^{\mathbf{m}} \equiv z_1^{m_1}\cdots z_k^{m_k}$. Moments of the distribution function are readily obtained by evaluating the generating function and its derivatives in the vicinity of the point $\mathbf{z} = \mathbf{1} \equiv (1,\ldots,1)$. For example, the total cluster density is given by $N(t) = F(\mathbf{1},t)$. The equation describing the temporal evolution of the generating function $F(\mathbf{z},t)$ can be evaluated by a proper summation of the rate equation. This yields

$$\frac{dF}{dt} = F^2 - 2FN. \tag{3}$$

As a preliminary step, we evaluate the time dependence of the density. The corresponding rate equation is obtained by evaluating Eq. (3) at $\mathbf{z} = \mathbf{1}$,

$$\frac{dN}{dt} = -N^2. \tag{4}$$

Without loss of generality we set the initial density to unity and therefore we have $N(t) = 1/(1+t)$. We also note that by subtracting Eq. (4) from Eq. (3), a simple differential equation for the quantity $F-N$, emerges, $d(F-N)/dt = (F-N)^2$. This equation is readily solved to find

$$F(\mathbf{z},t) = \frac{F_0(\mathbf{z})}{(1+t)\,(1+t-tF_0(\mathbf{z}))}, \tag{5}$$

where the notation $F_0(\mathbf{z}) \equiv F(\mathbf{z},t=0)$ has been used. Eq. (5) represents the general solution to Eq. (1). Indeed, it is a simple generalization of the well known solution [6] for the single mass aggregation process.

We consider the simplest multivariate case,

$$P_0(\mathbf{m}) = k^{-1}\sum_{i=1}^{k}\delta(m_i-1)\prod_{j\neq i}\delta(m_j). \tag{6}$$

These initial conditions imply $F_0 \equiv \bar{z} = (z_1+\cdots+z_k)/k$ and from Eq. (5) we have

$$F(\mathbf{z},t) = \frac{\bar{z}}{(1+t)\,(1+t-t\bar{z})}. \tag{7}$$

A possible application of these initial conditions is to aggregation processes involving k distinct species with equal initial densities. Although this symmetric situation appears to be too simple at first, it contains the necessary ingredients for exploring the long time kinetics.

Before we investigate the multivariate distribution, let us first study properties of the following distribution function

$$P(m_+,t) = \sum_{\{m_i\}} P(\mathbf{m},t)\delta\left(m_+ - (m_1+\cdots+m_k)\right). \tag{8}$$

This distribution corresponds to the sum variable $m_+ = m_1+\cdots+m_k$. It is useful to introduce the generating function $F(z,t) = \sum z^{m_+}P(m_+,t)$. This generating function can be obtained from $F(\mathbf{z},t)$ by replacing $\bar{z}$ with z. Expansion of $F(z,t) = z/(1+t)(1+t-tz)$ in powers of z yields the sum distribution

$$P(m_+,t) = \frac{t^{m_+-1}}{(1+t)^{m_++1}}. \tag{9}$$

The variable m_+ ignores the "identity" of the different conserved variables and thus, the problem reduces to ordinary aggregation. In the long time limit, $m_+ \sim t$, and the corresponding distribution is given by

$$P(m_+,t) \simeq t^{-2}\exp(-m_+/t). \tag{10}$$

Using the scaling variable, $M_+ = m_+/t$, this distribution can be also written in terms of a scaling function $P(m_+,t) \simeq t^{-2}\Psi(M_+)$, with $\Psi(x) = \exp(-x)$.

The multivariate probability distribution function can be now found by expanding $F(\mathbf{z},t)$ and comparing with Eq. (2),

$$P(\mathbf{m},t) = P(m_+,t)g(\mathbf{m}) \quad \text{with} \quad g(\mathbf{m}) = \frac{k^{-m_+}(m_+)!}{m_1!\cdots m_k!}. \tag{11}$$

Note that $\sum_{\{m_i\}}g(\mathbf{m}) = 1$. The above expression is the explicit solution to the mean field equations. However, its long time nature is of particular interest and we proceed with an asymptotic analysis.

To study the asymptotic properties of $P(\mathbf{m},t)$, we concentrate on the case $k = 2$ and then generalize the results to arbitrary k. The time independent geometric factor reads, $g(m_1,m_2) = 2^{-m_+}(m_1+m_2)!/m_1!m_2! \sim m_+^{-1/2}\exp\big(-(m_1-m_2)^2$ The limit of large masses, $m_1, m_2 \gg 1$, is the relevant one since the masses grow indefinitely. Using the limiting form of $P(m_+,t)$, we arrive at the asymptotic form of the mass density,

$$P(m_1,m_2,t) \sim t^{-2}m_+^{-1/2}\exp(-m_+/t)\exp(-m_-^2/2m_+), \tag{12}$$

with $m_\pm = m_1 \pm m_2$. Two mass scales govern the mass distribution, $m_+ \sim t$ and $|m_-| \sim \sqrt{m_+} \sim \sqrt{t}$. Furthermore, introduction of the scaling variables, $M_+ = m_+/t$ and $M_- = m_-/\sqrt{t}$, enables us to write the solution in a convenient scaling form

$$P(m_1,m_2,t) \sim t^{-5/2}\Phi(M_+,M_-). \tag{13}$$

The scaling function Φ depends on the scaling variables only,

$$\Phi(x,y) = x^{-1/2}\exp(-x)\exp(-y^2/2x). \tag{14}$$

Interestingly, each of the variables, m_1 and m_2, exhibit simple scaling: $m_1, m_2 \sim t$. Additionally, the moments $M_{\mathbf{n}}(t)$ of the distribution function, $M_{\mathbf{n}}(t) = \sum_{\{m_i\}}\mathbf{m}^{\mathbf{n}}P(\mathbf{m},t)$, are described asymptotically by linear exponents. By taking derivatives of the generating function, one can show that $M_{\mathbf{n}}(t) \sim t^{\alpha(\mathbf{n})}$ with $\alpha(\mathbf{n}) = n_+ - 1$. This behavior agrees with ordinary scaling and therefore one could naively expect single-size scaling to hold. However, from the complete form of the mass distribution we learn that it is *impossible* to write the scaling solution in terms of a simple scaling function, $P(m_1,m_2) \sim t^{-2}\Phi(m_1/t, m_2/t)$. Such a scaling solution would imply that the process has only one intrinsic size scale. Instead, this problem exhibits "double-scaling", as the process is governed by two distinct size scales. The second scale $|m_-| \sim \sqrt{t}$ is hidden, *e. g.*, it does not clearly appear in the moments of the distribution function. The mass difference is reminiscent of a diffusive scale for the following reason. The mass difference, $m_- = m_1 - m_2$, is also a conserved variable, since each of its components is conserved. Hence, for an aggregate of total mass m_+, the mass difference is a sum of m_+ random variables. According to the central limit theorem this variable is Gaussian and thus, $m_- \sim \sqrt{m_+}$.

The above results can be generalized to arbitrary k. The generalized mass difference variable is $m_-^2 = k^{-1}\sum_{i,j}(m_i - m_j)^2$ and the mass distribution follows the scaling form

$$P(\mathbf{m},t) \sim t^{-(k+3)/2}\Phi(M_+,M_-), \tag{15}$$

with the scaling function

$$\Phi(x,y) = x^{-(k-1)/2}\exp(-x)\exp(-y^2/2x). \tag{16}$$

The above results can be also generalized to situations with asymmetric initial conditions, *i. e.*, the initial conditions are not invariant with respect to a permutation of the mass variables, $\{m_i\}$. Denoting by $M_i = \sum_{\{m_i\}} m_i P_0(\mathbf{m})$ the $i^{\rm th}$ conserved mass, one can easily show that with the transformation $m_i \to m_i/M_i$ and $P(\mathbf{m}) \to M_1 \cdots M_k P(\mathbf{m})$, the system reduces to the symmetric case. For this transformation to be valid, the process must be truly multivariate, $M_i \neq 0$. In this case, the prefactor $g(\mathbf{m})$ equals probabilities associated with a biased random walk in a k-dimensional "mass" space.

Previous results have been established for a constant rate kernel, $K(\mathbf{m},\mathbf{n}) = \text{const}$. Although the most general situation is hardly tractable, in the case of sum-variable dependent reaction rate, $K(\mathbf{m},\mathbf{n}) \equiv K(m_+,n_+)$, the sum-variable distribution function $P(m_+,t)$ satisfies a simpler single-variable Smoluchowski equation. In this case, the complete multivariate distribution function $P(\mathbf{m},t)$ is related to $P(m_+,t)$ by Eq. (11). Therefore solvable variants of the single-variable Smoluchowski equation provide exact solutions for multivariate Smoluchowski equation with the corresponding reaction rates. For the usual Smoluchowski equation, exact solutions have been found for three types of reaction rates: $K(m,n) = \text{const}$, $K(m,n) = m+n$, $K(m,n) = mn$, and for their linear combinations [6,5,31]. In particular, for the sum-kernel $K(\mathbf{m},\mathbf{n}) = \frac{1}{2}(m_+ + n_+)$, subject to the monodisperse initial conditions of Eq. (6), we again arrive at the exact solution of Eq. (11) with

$$P(m_+,t) = \exp\left[-t - m_+(1-e^{-t})\right]\frac{\left[m_+(1-e^{-t})\right]^{m_+-1}}{(m_+)!}. \tag{17}$$

Similarly, one can find an exact solution for the product kernel.

We now investigate the steady state properties of the aggregation process by introducing input of particles into the system. For simplicity, we consider homogeneous input with rate h and restrict ourselves to the constant rate kernel. The governing equations are modified by adding an input term,

$$\begin{aligned}\frac{dP(\mathbf{m},t)}{dt} &= \sum_{\{m_i'\}} P(\mathbf{m}',t)P(\mathbf{m}-\mathbf{m}',t) \\ &\quad - 2P(\mathbf{m},t)\sum_{\{m_i'\}} P(\mathbf{m}',t) + hR(\mathbf{m}).\end{aligned} \tag{18}$$

The input function, $R(\mathbf{m})$, satisfies the normalization condition $\sum_{\{m_i\}} R(\mathbf{m}) = 1$ since the total input $h\sum_{\{m_i\}} R(\mathbf{m})$ is equal to h.

The solution to the above equation parallels the solution to the time dependent problem. We denote the steady state distribution by $P(\mathbf{m})$ and the steady state generating function by $F(\mathbf{z})$. This generating function is obtained by eliminating the time derivative of Eq. (18), $F^2 - 2FN + hR = 0$, where $R \equiv R(\mathbf{z}) = \sum_{\{m_i\}} \mathbf{z}^{\mathbf{m}} R(\mathbf{m})$. The normalization condition $R(\mathbf{1}) = 1$ is satisfied by the input function, and consequently the solution to the generating function reads

$$F(\mathbf{z}) = \sqrt{h}\left(1 - \sqrt{1 - R(\mathbf{z})}\right). \tag{19}$$

The total monomer density is given by $N = F(\mathbf{1})$ and thus,

$$N(h) = \sqrt{h}. \tag{20}$$

In analogy with the initial conditions of Eq. (6), we consider the input $R(\mathbf{m}) = P_0(\mathbf{m})$, which in turn implies $R(\mathbf{z}) = \bar{z}$. In this case, the steady state generating function is simply $F(\mathbf{z}) = \sqrt{h}\big(1 - \sqrt{1-\bar{z}}\big)$. Expanding in powers of the variables z_i, and comparing with the definition of the generating function, one finds the steady state distribution

$$P(\mathbf{m}) = P(m_+)g(\mathbf{m}), \quad P(m_+) = \frac{(2m_+)!}{(2m_+ - 1)(2^{m_+}m_+!)^2}. \tag{21}$$

There is a strong similarity with the time dependent counterpart, $P(\mathbf{m},t)$. The distribution among aggregates with the same total mass, m_+, is given by the same combinatorial factor $g(\mathbf{m})$ which has already appeared in solution to the time dependent problem. For large masses, the distribution describing the sum variable, m_+, is an algebraic one, $N^{-1}P(m_+) \sim m_+^{-3/2}$. We can write now the asymptotic form of the complete steady state distribution,

$$P(\mathbf{m}) \equiv P(m_+, m_-) \sim \sqrt{h}\; m_+^{-(k+2)/2} \exp(-m_-^2/2m_+). \tag{22}$$

For the time dependent problem, an additional size scale emerges for the mass difference variable, $m_- \sim \sqrt{m_+}$. In the steady state, on the other hand, the total mass diverges and thus, the central limit theorem does not apply. However, the similarity between the two cases is still strong as for a fixed m_+, the mass difference distribution function is Gaussian.

For completeness, we briefly discuss the relaxation properties of the density towards the steady state. It is possible to obtain these properties by incorporating the time dependent density, $N \sim t^{-\alpha}$, and the steady state solution, $N \sim h^{\gamma}$, into a single expression. Let us assume the scaling form [17]

$$N(h,t) \sim t^{-\alpha}\psi(t/\tau), \tag{23}$$

with the relaxation time $\tau \sim h^{-\beta}$. In the limit of a vanishing input rate the time dependent solution must be recovered and thus, $\psi(x) \to 1$ as $x \to 0$. In the long time limit steady state is approached and thus, $\psi(x) \sim x^{\alpha}$ for $x \gg 1$. Comparing with the definition of the relaxation time the exponent relation

$$\gamma = \alpha\beta \tag{24}$$

is found. The exponent $\beta = 1/2$, is obtained from the decay exponent, $\alpha = 1$, and the steady state exponent, $\gamma = 1/2$. Hence, the relaxation time diverges in the limit of a vanishing input rate, $\tau \sim h^{-1/2}$. This result can be obtained directly by solving the equation $dN/dt = -N^2 + h$ which is readily performed to find $N(h,t) = \sqrt{h}\tanh\left((t+t_0)\sqrt{h}\right)$. The time shift t_0 is determined by the initial density, $N(t=0) = \sqrt{h}\tanh(\sqrt{h}t_0)$. We prefer the above scaling argument since it is applicable to a wide class of problems.

III. BALLISTIC AGGREGATION

Ballistic aggregation is a natural and important example of a process with multiple conservation laws. Both the mass and the momentum are conserved quantities and thus, there are $d+1$ conservation laws in d dimension [26]. A collision between two particles results in an aggregate whose mass as well as momentum are given by a sum over its components. One can view this system as a gas of sticky particles, *i. e.*, an inelastic gas with a vanishing restitution coefficient. Heuristic arguments predict certain scaling properties of the system. However, other aspects of this problem, such as the mass-momentum distribution function remain unsolved. Our theory is well suited for this problem and yields an approximate form for the particle distribution function.

We start by discussing the one dimensional case and then generalize to higher dimensions. We denote the probability distribution function for particles of mass m and momentum p at time t by $P(m,p)$ (we suppress the time variable). The *stosszahlansatz* Boltzmann equation, which describes the temporal evolution of this density, must conserve mass and momentum. Additionally, the collision rate between two particles is given by their velocity difference and hence, the Boltzmann equation reads

$$\begin{aligned}\frac{dP(m,p)}{dt} &= \sum_{m',p'} |v'-v''|\, P(m',p')P(m-m',p-p') \\ &\quad -2P(m,p)\sum_{m',p'} |v-v'|\, P(m',p'),\end{aligned} \tag{25}$$

where $v'-v'' = p'/m' - (p-p')/(m-m')$ and $v - v' = p/m - p'/m'$. This approximation ignores possible spatial correlations and is generally uncontrolled. By replacing the kernel terms $|v-v'|$ and $|v'-v''|$ with their average value $\langle v\rangle$, an approximate Boltzmann equation is written,

$$\frac{dP(m,p)}{dt} = \langle v\rangle \left(\sum P(m',p')P(m-m',p-p') - 2P(m,p)\sum P(m',p')\right). \tag{26}$$

Although this approximation is generally unjustified, it enables a solution for the mass-momentum distribution. The time-dependent factor $\langle v\rangle$ can be absorbed into a novel time variable, T, defined by $dT/dt = \langle v\rangle$. The resulting Boltzmann equation is equivalent to Eq. (1).

For simplicity we consider an initial system of identical particles with zero average momentum. Without loss of generality we assume that typical initial quantities such as the mass and the momentum equal unity. Hence, the initial distribution function, $P_0(m,p)$, is given by

$$P_0(m,p) = \delta(m-1)\big(\delta(p-1) + \delta(p+1)\big)/2. \tag{27}$$

In principle, the general solution of Eq. (26) can be obtained using the generating function method. However, with these specific initial conditions, the process reduces to the two-mass process discussed in the preceding section. Indeed, by identifying m with the total mass $m_+ = m_1 + m_2$ and p with the mass difference $m_- = m_1 - m_2$, the initial conditions Eq. (6) and Eq. (27) are the same. Moreover, identical rate equations describe the time evolution of both $P(m_1, m_2)$ and $P(m, p)$. From the solution of Eq. (11), the mass-momentum distribution function is found in terms of the time variable T,

$$P(p,m) = \frac{T^{m-1}}{(1+T)^{m+1}} \; \frac{2^{-m} m!}{((m+p)/2)!\,((m-p)/2)!}. \quad (28)$$

Using the asymptotic scaling properties of the solution $m \sim T$ and $p \sim \sqrt{T}$, we can rewrite the solution in terms of the observable time t. Since $dT/dt \sim v \sim p/m \sim T^{-1/2}$, one has $T \sim t^{2/3}$. Hence, the well known the scaling laws

$$m \sim t^{2/3} \qquad \text{and} \qquad p \sim t^{1/3}, \quad (29)$$

are recovered. Asymptotically, the mass-momentum distribution is given by the following form

$$P(p,m) \sim \langle m \rangle^{-2} m^{-1/2} \exp\Big(-\frac{m}{\langle m \rangle} - \frac{p^2}{2m}\Big), \quad (30)$$

with the average mass $\langle m \rangle \sim t^{2/3}$. As discussed in the previous section, the momentum p is a sum of m independent variables. As a result, for a fixed mass m the momentum distribution is Gaussian and $p \sim \sqrt{m}$. From Eq. (10), the mass distribution is [27] $P(m) = \langle m \rangle^{-2} \exp\big(-m/\langle m \rangle\big)$. Direct integration of Eq. (30) shows that the momentum distribution is also purely exponential, $P(p) = \big(\langle m \rangle \langle |p| \rangle\big)^{-1} \exp(-|p|/\langle |p| \rangle)$, with the typical momentum, $\langle |p| \rangle = \sqrt{\langle m \rangle / 2}$. On the other hand, the velocity distribution is algebraic for large velocities $p(v) \sim |v|^{-3}$.

It is interesting to compare these predictions with numerical simulations. Carnevale *et al.* [26] established a scaling behavior of Eq. (29) heuristically and confirmed it numerically. They also reported a distribution that is reminiscent of Eq. (30). Their numeric form resembles Eq. (30) in that the mass distribution is exponential and in that for a fixed mass the velocity distribution is Gaussian. However, there is a significant difference between the two forms as the simulation data suggests that the velocity distribution is independent of mass. This observation is an intriguing one, since for a fixed mass m the typical velocity is mass dependent $v(m) \sim m^{-1/2}$. Jiang and Leyvraz [28] also studied the mass distribution and found that it is singular near the origin, $P(m) \sim m^{-1/2}$ for $m \ll \langle m \rangle$, in contradiction with our approximate theory. Curiously, the mass-momentum distribution contains an identical singularity. However, this singularity disappears when the momentum is integrated over. We conclude that further numeric investigation of quantities such as velocity correlations between neighboring aggregates and moments of the mass-velocity distribution are needed to better the understanding of one-dimensional ballistic aggregation.

A similar line of reasoning applies in d dimensions. We assume that the density of an aggregate is constant, and thus, as the aggregation process evolves, the size of an aggregate grows indefinitely. Initially, only monomers with unit momentum occupy the system. Since the collision rate is also proportional to the surface area of an aggregate, the typical collision rate is va^{d-1}, with the radius $a \sim m^{1/d}$. The density satisfies the approximate Boltzmann equation

$$\frac{dP(m,\mathbf{p})}{dt} = \langle v a^{d-1} \rangle \quad (31)$$
$$\Big(\sum P(m',\mathbf{p}')P(m-m',\mathbf{p}-\mathbf{p}') - 2P(m,\mathbf{p})\sum P(m',\mathbf{p}')\Big),$$

where $\mathbf{p}$ is the d-dimensional momentum. Repeating the above analysis yield the leading scaling behavior for the mass and the momentum,

$$m \sim t^{2d/(d+2)} \qquad \text{and} \qquad |\mathbf{p}| \sim t^{d/(d+2)}. \quad (32)$$

The distribution function is a simple generalization of Eq. (31),

$$P(m,\mathbf{p}) \sim \langle m \rangle^{-2} m^{-d/2} \exp\Big(-\frac{m}{\langle m \rangle} - \frac{d|\mathbf{p}|^2}{2m}\Big), \quad (33)$$

with the average mass $\langle m \rangle \sim t^{2d/(d+2)}$. Eq. (32) suggests that ballistic aggregation has no upper critical dimension. Hence, it is not clear whether our approximation holds in sufficiently large dimension, as is the case for diffusive aggregation. In a recent study of the two-dimensional gas of sticky particles [30], some deviations from the mean-field predictions have been observed. The simulations [30] revealed that the growth exponent varies up to 10% as the initial density was varried. The growth exponent of Eq. (32) was recovered for sufficiently high initial densities. In this limit, multiple coalescence events dominate and mean-field thoery is appropriate. In general, an exponential mass distribution and a Boltzmann energy distribution were found, in agreement with mean-field theory. We conclude that despite the crude nature of the approximation, it provides good estimates for the leading asymptotic behavior as well as the various distribution functions.

Steady state can be achieved by adding particles to the system with rate h. We consider homogeneous and isotropic input of particles with unit mass and unit momentum. From Eq. (21), the distribution function reads,

$$P(m,p) = \sqrt{h}\; m^{-(d+3)/2} \exp\Big(-\frac{d|\mathbf{p}|^2}{2m}\Big). \quad (34)$$

Furthermore, the mass distribution is given by $N^{-1}P(m) \sim m^{-3/2}$, with the density $N \sim \sqrt{h}$. Note that the velocity kernel $\langle v \rangle$ is not important in the steady state since $\langle v \rangle \propto \sum P(m)v(m) \sim \int m^{-3/2} m^{-1/2}$ is finite.

One can also study the relaxation properties of the system. The relaxation towards the steady state becomes slower and slower as the input rate vanishes. Following the analysis of Eq. (23), the corresponding relaxation exponent is obtained,

$$\tau(h) \sim h^{-\beta}, \qquad \text{with} \qquad \beta = \frac{d+2}{4d}. \tag{35}$$

However, only in the low input rate limit, $h \ll 1$, steady state is achieved. Indeed, the previous description is valid in the low coverage limit, *i. e.*, when $t \ll t_c$ with $t_c \sim h^{-1}$, while for $t > t_c$ the space is covered by a single "superparticle". Since the relaxation exponent β is less than one, the time scales τ and t_c are well separated in the low input rate limit. Thus, the steady state distribution of Eq. (34) provides an intermediate asymptotics valid for $\tau \ll t \ll t_c$.

In contrast, the nature of steady state caused by a spatially *localized* particle input is a truly asymptotic one. The spatial dependence of the density distribution is of particular interest in this problem. We consider a spherically symmetric point-like source of ballistic particles with unit mass and unit momentum. Let $P(m, \mathbf{p}, r) = P(m, \mathbf{p}, r, t = \infty)$ be the steady-state radial mass-momentum density. For $d > 2$ the reaction is in fact irrelevant away from the source [20]. The density satisfies the convection equation, $d\big(r^{d-1}P(m, \mathbf{p}, r)\big)/dr = 0$, and thus, the concentration decays as $r^{-(d-1)}$. So in the *inhomogeneous* ballistic problem, $d = 2$ is a critical dimension above which the reaction merely leads to the renormalization of the strength of the source.

For $d \le 2$, away from the source, particles have typically collided many times and they move with a velocity close to unity in the radial direction. Thus, collisions occur with rate proportional to the rms velocity v. The steady-state radial distribution $P(m, \mathbf{p}, r)$ satisfies

$$\begin{aligned} &r^{1-d}\frac{d}{dr}\big(r^{d-1}P(m,\mathbf{p})\big) = \langle v a^{d-1}\rangle \\ &\Big(\sum P(m',\mathbf{p}')P(m-m',\mathbf{p}-\mathbf{p}') - 2P(m,\mathbf{p})\sum P(m',\mathbf{p}')\Big). \end{aligned} \tag{36}$$

It is not difficult to verify that with the transformation $r^{d-1}P(m, \mathbf{p}) \to P(m, \mathbf{p})$ and $R = \int^r r'^{(1-d)} dr' \to t$, the steady-state equation reduces to the time-dependent equation (31). Therefore when $d < 2$, the variable $R \sim r^{2-d}$ plays the role of time and from Eq. (32), $m(r) \sim R^{2d/(2+d)} \sim r^{2d(2-d)/(2+d)}$. As $d \to 2$, the exponent describing the mass growth vanishes, indicating that $d = 2$ is indeed the critical dimension, above which the typical mass far from the source is constant. At the critical dimension $d = 2$, logarithmic behavior occurs, $R \sim \log(r)$, and consequently, $m(r) \sim R \sim \log(r)$. Hence, for $d \le 2$ clustering is significant and the typical mass is a growing function of the distance from the source.

To determine the mass-momentum distribution, we tacitly impose boundary conditions similar to the initial conditions of the time dependent problem, $r_0^{d-1}P(m, \mathbf{p}, r_0) = \delta(m-1)\delta(|\mathbf{p}| - 1)$. The steady-state density far from the source reads

$$\begin{aligned} P(m,\mathbf{p},r) &\sim r^{-(d-1)}\langle m(r)\rangle^{-2} m^{-d/2} \\ &\exp\left(-\frac{m}{\langle m(r)\rangle} - \frac{d(|\mathbf{p}|-m)^2}{2m}\right) \quad d \le 2, \end{aligned} \tag{37}$$

with $\langle m(r)\rangle \sim r^{2d(2-d)/(2+d)}$ for $d < 2$ and $\langle m(r)\rangle \sim \log(r)$ for $d = 2$. For a fixed mass, the momentum distribution is Gaussian and as a result, the rms momentum is characterized by $\sqrt{m(r)}$. By integration of the mass-momentum density, one finds the concentration, $N(r) \sim 1/(r^{(d-1)}\langle m(r)\rangle)$. As a result, $N(r) \sim r^{(2-5d+d^2)/(2+d)}$ for $d < 2$, and $N(r) \sim 1/\big(r\log(r)\big)$ for $d = 2$.

Returning to the time dependent problem, the steady state solution holds for $r < t$ only, while for a larger the space is essentially empty. It is useful to estimate the total number of clusters $\mathcal{N}(t) \sim \int_0^t r^{d-1}N(r)$. For $d < 2$ one finds $\mathcal{N}(t) \sim t^{(2d^2-3d+2)/(d+2)}$, and for $d = 2$ one has $\mathcal{N}(t) \sim t/\log(t)$. In the ballistic regime, $d > 2$, collisions do not cause a significant reduction in the number of clusters, and the total number of clusters grows linearly in time.

To summarize, we write the leading asymptotic behaviors of the mass

$$m(r) \sim \begin{cases} r^{2d(2-d)/(2+d)} & d < 2; \\ \log(r) & d = 2; \\ 1 & d > 2, \end{cases} \tag{38}$$

the density

$$N(r) \sim \begin{cases} r^{-(5d-d^2-2)/(2+d)} & d < 2; \\ r^{-1}[\log(r)]^{-1} & d = 2; \\ r^{-(d-1)} & d > 2, \end{cases} \tag{39}$$

and the total number of clusters

$$\mathcal{N}(t) \sim \begin{cases} t^{(2d^2-3d+2)/(2+d)} & d < 2; \\ t/\log(t) & d = 2; \\ t & d > 2. \end{cases} \tag{40}$$

The typical rms momentum behaves as $\sqrt{m(r)}$. Numerical simulations agree with the above in $d = 1$ [26]. It will be interesting to test these predictions in higher dimensions.

IV. DIFFUSIVE AGGREGATION

In this section, we consider the diffusive driven aggregation processes involving k distinct species. Each of the species masses is conserved and thus there are k conservation laws. We apply the rate theory described in Sec. II and also investigate low dimensional systems where the rate equation description is expected to fail [15]. In all dimensions we find that in addition to the typical mass scale, there exists an additional mass scale.

In spatial dimensions larger than the critical dimension, $d_c = 2$, Smoluchowski rate theory is exact. In dimensions lower than the critical dimension, spatial correlations are significant asymptotically. Particles are repelling each other, and the effective reaction rate, κ, depends on the density [20]

$$\kappa \sim \begin{cases} N^{2/d-1} & d < 2; \\ 1/|\log(N)| & d = 2; \\ 1 & d > 2. \end{cases} \tag{41}$$

This reaction rate can be absorbed into a suitably defined time variable T,

$$\frac{dT}{dt} = \kappa. \tag{42}$$

With this novel time, Smoluchowski rate equation reduces to Eq. (1).

For $d \leq d_c$, this approximation yields erroneous results for the mass distribution. Nevertheless, it produces correct asymptotic scaling behavior for quantities such as the typical mass. Thus for the general case $d \leq d_c$ we just quote the leading asymptotic behaviors while in one dimension we provide exact results. Let us consider the case where there are two species, say A and B. A cluster is characterized by the respective masses of its components, m_A and m_B. The typical total mass is given by $m_A + m_B \sim T$, or equivalently,

$$m_A + m_B \sim \begin{cases} t^{d/2} & d < 2; \\ t/\log(t) & d = 2; \\ t & d > 2. \end{cases} \tag{43}$$

On the other hand, since the mass difference is a Gaussian variable, for a fixed total mass, one has $|m_A - m_B| \sim \sqrt{m_A + m_B}$. This additional mass scale can be also expressed in terms of time,

$$|m_A - m_B| \sim \begin{cases} t^{d/4} & d < 2; \\ t^{1/2}[\log(t)]^{-1/2} & d = 2; \\ t^{1/2} & d > 2. \end{cases} \tag{44}$$

Note also that the present two-mass aggregation process can be mapped onto a two-species aggregation-annihilation process with two conservation laws [23]. Indeed, the above results agree with analytical and numerical findings of Refs. [23,32].

In one dimension, it is possible to derive a complete analytical solution. We consider a linear lattice on which point clusters hop randomly from site to nearest neighbor sites. We assume that the diffusion coefficient D does not depend on cluster's mass. We also assume that initially each site is occupied by some monomer, A or B with equal probability. The multivariate distribution function can be expressed in the form of Eq. (11), with the sum variable given by the Spouge's solution [19] of the ordinary one dimensional diffusion-controlled aggregation problem,

$$P(m_+, t) = e^{-4Dt}\big[I_{m_+-1}(4Dt) - I_{m_++1}(4Dt)\big], \tag{45}$$

where I_n denotes a modified Bessel function. Introducing now the scaling variables, $M_+ = m_+/\sqrt{8Dt}$ and $M_- = m_-/(8Dt)^{1/4}$, one can rewrite the solution in a convenient scaling form

$$P(m_1, m_2, t) \sim t^{-5/4}\Phi(M_+, M_-), \tag{46}$$

with the scaling function Φ

$$\Phi(x, y) = \sqrt{x}\exp\Big(-x^2 - \frac{y^2}{2x}\Big). \tag{47}$$

Finally, we consider diffusive aggregation in a system with a steady spatially localized monomer input. We assume that monomers of all types are added at random with an equal rate. Making use of exact and scaling results for the corresponding ordinary aggregation [20], we solve for our case. This inhomogeneous system is characterized by two critical dimensions, the usual "homogeneous" critical dimension $d_c = 2$ and an additional critical dimension $d^c = 4$ which demarcates the pure diffusion regime $d > 4$ (particles do not affect each other far away from the source) and the diffusion-reaction regime $d \leq 4$. The system approaches steady state as $t \to \infty$. In the diffusion-reaction regime, the density distribution function approaches a power-law in r, where r is the distance from the source. The borderline cases $d = d_c = 2$ and $d = d^c = 4$ should be treated more carefully since logarithmic factors appear. We write the final results for the typical total mass

$$m_A + m_B \sim \begin{cases} r^2 & d < 2; \\ r^2/\log(r) & d = 2; \\ r^{4-d} & 2 < d < 4; \\ \log(r) & d = 4; \\ 1 & d > 4. \end{cases} \tag{48}$$

For a fixed total mass $m_A + m_B$, the mass difference is again Gaussian and consequently, $|m_A - m_B| \sim \sqrt{m_A + m_B}$.

V. CONCLUSIONS

In summary, we have investigated irreversible aggregation with many conservation laws. The solution to this process is characterized by the Gaussian statistics of the fluctuations in a given conserved quantity. The process is governed asymptotically by two size scales. A typical aggregation-induced scale characterizes the total mass, while a novel diffusive scale characterizes individual masses. The latter scale is hidden, *i. e.*, it is not present in the moments of the multivariate distribution. Application to a "sticky gas" suggests a Boltzmann velocity distribution for a fixed mass. In addition, the mass distribution is exponential. By comparing our predictions with available numerical results we have found that the present approximate theory gives a good description of the sticky gas. We have also investigated steady-state properties by introducing a localized source. We have observed two different behaviors, the ballistic-reaction regime for $d \leq 2$, and the pure ballistic regime for $d > 2$. Application to diffusive aggregation with more than one species also exhibit a novel "diffusive" size scale.

This study suggests different avenues for further investigation. The theory might be applicable to problems such as catalysis and chemical reactions with many species. It will be also interesting to analyze the rate equations with more realistic reaction rates. An important question to be addressed is how robust is the Gaussian nature of the fluctuations statistics. It is plausible that spatial correlations introduce nontrivial internal arrangement of the clusters, leading to more complicated statistics. It is also plausible that even on the rate equation level but with the reaction rate not expressible as a function of the sum-variables only, different asymptotic behavior emerges. This could explain why for a dual fragmentation process the multivariate generalization produces an infinite set of scales [33-35] compared to the two scales in the models of multivariate aggregation we have examined in this study.

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