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Stochastic aggregation: scaling properties

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Abstract. We study the scaling properties of stochastic aggregation processes in one dimension. Numerical simulations for both diffusive and ballistic transport show that the mass distribution is characterized by two independent non-trivial exponents corresponding to the survival probability of particles and monomers. The overall behaviour agrees qualitatively with the mean-field theory. This theory also provides a useful approximation for the decay exponents, as well as the limiting mass distribution.

1. Introduction

In the preceding study, we introduced a stochastic aggregation process involving both active and passive clusters [1]. We generalized the Smoluchowski rate equations and obtained exact results for several kernels. In this paper, we apply stochastic aggregation to reaction–diffusion, coarsening and ballistic agglomeration problems. Our goal is to examine the range of validity of the mean-field results, and to determine whether the overall scaling behaviour extends to low-dimensional systems.

The rate equations approach is mean field in nature, i.e. it is valid only when spatial correlations are absent. Formally, it is applicable only in an infinite spatial dimension, or in the presence of an effective mixing mechanism. This mean-field theory should also be *asymptotically* exact when the spatial dimension is sufficiently high. In low spatial dimensions, however, significant spatial correlations eventually develop, and the rate-equation approach does not apply in the long-time limit. We therefore focus on one-dimensional systems where the spatial correlations are most pronounced.

We have performed numerical simulations of stochastic aggregation processes with both diffusive and ballistic particle transport. The simulations show that the scaling behaviour suggested by the mean-field theory is indeed generic, as it extends to one-dimensional systems. We find that two non-trivial model-dependent exponents characterize the survival probabilities of the particles and monomers, respectively. The Smoluchowski theory provides reasonable estimates for these exponents.

Additionally, we have studied the limiting mass distribution of passive clusters. Surprisingly, over a substantial mass range, this distribution depends only weakly on the underlying transport mechanism. Furthermore, mean-field theory provides an excellent approximation for the limiting mass distribution.

The rest of this paper is organized as follows. The general scaling behaviour is outlined in section 2. Predictions of the mean-field theory are summarized in section 3.

Numerical simulations of stochastic aggregation processes with diffusive and ballistic transport mechanisms are described in sections 4 and 5, respectively. A discussion of the results is presented in section 6.

2. Scaling properties

Stochastic aggregation involves two types of clusters: active and passive [1]. Initially, the system consists of active monomers only. When two active clusters merge, the newly born aggregate remains active with probability p , or becomes passive (i.e. it never aggregates again) with probability $q = 1 - p$. Eventually, all active clusters are depleted and the system consists only of passive clusters. This process can be viewed as an aggregation–annihilation process since it interpolates between aggregation ($p = 1$) and annihilation ($p = 0$)[†].

Quantities of interest include $A_k(t)$ and $P_k(t)$, the distributions of active and passive clusters at time t , as well as the final distribution of passive clusters, $P_k(\infty)$. As shown in the preceding paper, two conservation laws underlie this system. The first is mass conservation: $\sum k[A_k(t) + P_k(t)] = \text{constant}$. The second conservation law reflects the fact that changes in the overall densities are coupled: $qA(t) + (1+q)P(t) = \text{constant}$, where $A(t) = \sum A_k(t)$ and $P(t) = \sum P_k(t)$ are the number densities of active and passive clusters, respectively.

Therefore, it is sufficient to study the time evolution of the number density and the mass density of the active clusters, $A(t)$ and $M(t) = \sum kA_k(t)$, respectively. The latter quantity is the survival probability of an active particle, i.e. the probability that it still belongs to an active cluster at time t . The Smoluchowski theory suggests that both quantities decay algebraically in the long-time limit

$$A(t) \sim t^{-\nu} \quad M(t) \sim t^{-\nu\psi}. \quad (1)$$

As will be shown, this, as well as other scaling properties suggested by this theory, holds qualitatively even for low-dimensional stochastic aggregation processes. While the decay exponent ν is typically robust in that it depends only on the major characteristics of the process such as the spatial dimension or the transport mechanism, the exponent $\psi \equiv \psi(p)$ is non-universal as it depends on the details of the model, i.e. on the probability p . In turn, this implies a non-universal growth law for the average mass of an active cluster $\langle k \rangle = M/A \sim t^{\nu(1-\psi)}$.

For the system to follow a scaling behaviour, the average mass must be the only relevant scale in the long-time limit, and, conversely, any scale characterizing the initial mass distribution must eventually be ‘erased’. In other words, the mass distribution is characterized by a single rescaled variable

$$A_k(t) \sim t^{\nu(\psi-2)} F(k t^{\nu(\psi-1)}) \quad (2)$$

with the time-dependent prefactor fixed by the decay laws (1).

This scaling behaviour is similar to that found for deterministic aggregation–annihilation processes [2–4] and for aggregation–annihilation of domains in coarsening processes [5–7]. These studies suggest that another independent exponent describes the decay of small clusters. Specifically, the monomer density decays according to

$$A_1(t) \sim t^{-\nu\delta} \quad (3)$$

with a model-dependent exponent $\delta \equiv \delta(p)$. The monomer density decay reflects the small-argument behaviour of the scaling function $F(\xi) \sim \xi^\sigma$ with $\delta - 1 = (1 - \psi)(1 + \sigma)$. One of our main results is that the mass distribution of active clusters is described by a set of

[†] Somewhat similar deterministic aggregation–annihilation processes have been investigated in [2–4]

non-trivial exponents (ψ, δ) . These exponents can be viewed as persistence exponents [8, 9] as they characterize the survival probability of an active particle, and an active monomer [10].

Several properties of the scaling exponents are general. For instance, the inequalities $\psi \leq 1 \leq \delta$ hold since $A_1 \leq \sum A_k \leq \sum kA_k$. The two exponents are equal, $\psi = \delta = 1$, in the annihilation case ($p = 0$), since $A_k(t) = A(t)\delta_{k,1}$. In the aggregation limit ($p = 1$) the mass density of active clusters is conserved and therefore $\psi = 0$.

We now turn to the mass distribution of passive clusters. The Smoluchowski theory suggests that the same scaling form underlies both mass distributions

$$P_k(t) \sim t^{\nu(\psi-2)} G(k t^{\nu(\psi-1)}). \tag{4}$$

In contrast to the active-cluster distribution, the passive-cluster distribution approaches a non-trivial final distribution $P_k(\infty)$. Such a *time-independent* final distribution is consistent with the above scaling form only when the scaling function diverges, $F(\xi) \sim \xi^{-\gamma}$ in the limit $\xi \rightarrow 0$, with $\gamma = (2 - \psi)/(1 - \psi)$. As a result, the final mass distribution of passive clusters decays algebraically in the large-mass limit

$$P_k(\infty) \sim k^{-\gamma} \quad \text{with} \quad \gamma = \frac{2 - \psi}{1 - \psi}. \tag{5}$$

At a given time t , this decay is realized for clusters whose mass k does not exceed the characteristic mass $k^* \sim t^{\nu(1-\psi)}$. Note also that $0 < \psi < 1$ implies $2 < \gamma < \infty$. Generally, the mass conservation restricts the large-mass decay exponent to $\gamma > 2$. Since the ψ exponent varies between 0 and 1, we see that the entire range of acceptable exponents is realized by tuning the probability p .

3. Mean-field theory

It is well established that spatial correlations can be safely neglected only in spatial dimensions larger than an upper critical dimension, $d > d_c$ [11]. For example, for irreversible aggregation with mass-independent diffusion and reaction rates, one has $d_c = 2$; for a general aggregation process, however, the upper critical dimension may be arbitrarily large [12]. Below the upper critical dimension, substantial spatial correlations develop, and the most important features, including the scaling exponents and the scaling functions, are changed. Generally, the lower the spatial dimension, the larger the difference from the mean-field predictions.

Although the Smoluchowski-rate-equations approach does not apply in low spatial dimensions, it can still serve as a useful approximation after an appropriate modification. This can be accomplished by replacing the overall reaction rate with an effective density-dependent reaction rate $r \equiv r(A)$

$$\begin{aligned} \frac{dA_k}{dt} &= r \left(\frac{1}{2} p \sum_{i+j=k} A_i A_j - A_k A \right) \\ \frac{dP_k}{dt} &= r \left(\frac{1}{2} q \sum_{i+j=k} A_i A_j \right). \end{aligned} \tag{6}$$

We are primarily interested in situations where aggregation is independent of mass, and therefore we use a mass-independent rate kernel. The reaction rate $r(A)$ is model dependent. In reaction-diffusion processes, the reaction rate decays algebraically with the density (see, for example, [13, 14]). Assuming $r(A) \sim A^\alpha$ yields $\frac{dA}{dt} \sim -A^{\alpha+2}$, then the density decay exponent is found to be

$$\nu = \frac{1}{1 + \alpha}. \tag{7}$$

In general, a reduction in the reaction rate, i.e. $\alpha > 0$, leads to a slowing down in the density decay rate, $\nu < 1$. Apart from the change in ν , all other aspects of this approximation are identical to the Smoluchowski theory with a constant-rate kernel. Indeed, the above rate equations reduce to the Smoluchowski rate equations with a redefined time variable, $t \rightarrow \tau = \int_0^t dt' r(t')$. In particular, the scaling exponents ψ and δ are independent of α :

$$\psi = 2 \frac{1-p}{2-p} \quad \delta = \frac{2}{2-p}. \quad (8)$$

One can verify the expected limiting behaviours $\psi(1) = 0$ and $\psi(0) = \delta(0) = 1$. Furthermore, the scaling functions are as in the constant-kernel solution [1], and for example, $F(\xi)$ is purely exponential. The corresponding small-argument exponents $\gamma = 2/p$ and $\sigma = 0$ follow from ψ and δ using the aforementioned scaling relations. The final mass distribution of passive clusters is independent of the reaction rate r [1]:

$$P_k(\infty) = \frac{q}{p} \frac{\Gamma(1+2/p) \Gamma(k)}{\Gamma(k+2/p)}. \quad (9)$$

We now compare these mean-field predictions with simulation results for one-dimensional stochastic aggregation where the spatial correlations are most pronounced. We also examine the role of the aggregates' transport mechanism by considering both diffusive and ballistic transport.

4. Diffusive transport

In diffusive stochastic aggregation, identical particles are placed onto a d -dimensional lattice. All particles perform independent random walks, i.e. they hop to a randomly chosen nearest-neighbour site with a constant rate. If this site is occupied, the two particles coalesce irreversibly, and with probability p the resulting aggregate remains active, while with probability $q = 1 - p$ it becomes passive. Effectively, passive particles are removed from the system.

In the case of single-species reaction–diffusion processes, the effective reaction rate can be obtained from dimensional analysis. Equation (6) implies $[r] = [L]^d [T]^{-1}$, and since the reaction rate can only be a function of the diffusion coefficient $[D] = [L]^2 [T]^{-1}$ and the density $[A] = [L]^{-d}$, one finds $r \propto DA^{(2-d)/d}$. Hence, $\alpha = (2-d)/d$ and equation (7) yield the correct decay exponents $\nu = d/2$ [11] below the upper critical dimension $d_c = 2$.

To examine the above scaling picture we performed numerical simulations of diffusive stochastic aggregation processes in one dimension. Unless noted otherwise, the data were obtained from an average over 10 independent realizations in a system of size $L = 10^7$ with periodic boundary conditions. Initially, all sites were occupied. First, we verified that the number density, the mass density and the monomer density indeed decay algebraically in the long-time limit, in accordance with equations (1) and (3). The case $p = \frac{1}{2}$ is shown in figure 1, and the corresponding decay exponents were found: $\nu = 0.500(1)$, $\psi = 0.6193(3)$ and $\delta = 1.460(2)$. Mean-field theory correctly predicts $\nu = \frac{1}{2}$. Furthermore, the predictions $\psi = \frac{2}{3}$ and $\delta = \frac{4}{3}$ provide a reasonable approximation. One can compare with the case of disordered (Sinai) diffusion where a real-space decimation procedure [15] was used to determine the *exact* values of these exponents [7]. Remarkably, the disorder case exponent $\psi = 0.61937$ is in excellent agreement with the simulation value. There is a small discrepancy with the second exponent $\delta = 1.47041$. In addition, we verified that the densities of active and

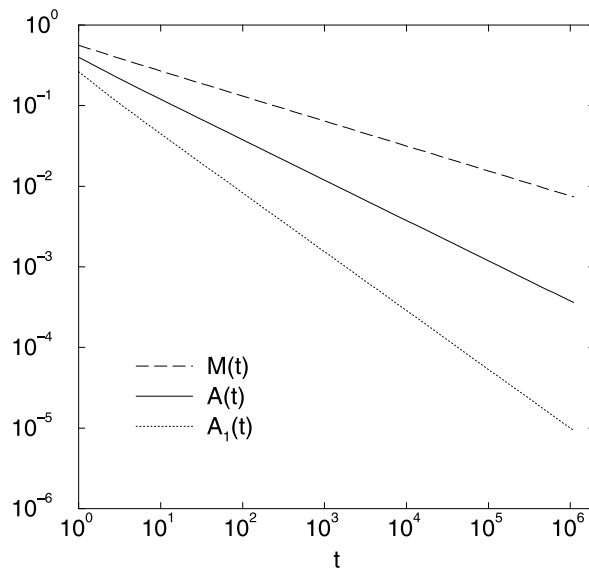


Figure 1. The number density, mass density and monomer density versus time for $p = \frac{1}{2}$.

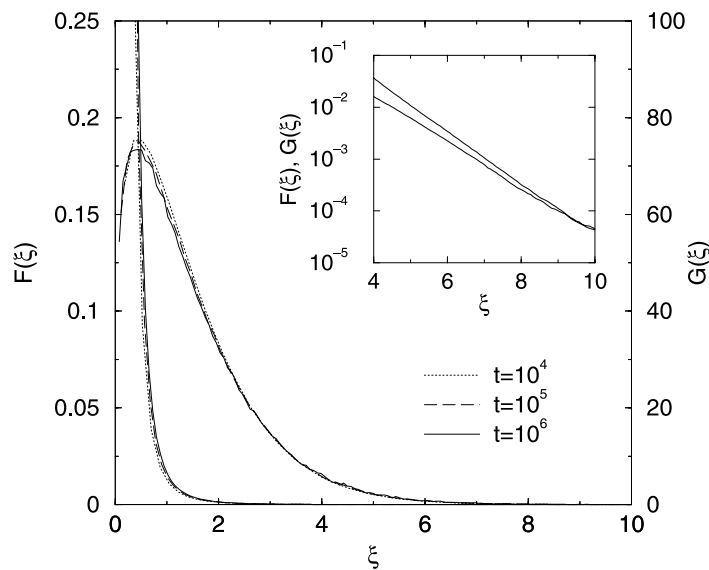


Figure 2. Scaling of the active and passive mass distributions. Shown are the scaling functions $F(\xi) \equiv t^{\nu(2-\psi)} A_k(t)$ and $G(\xi) \equiv t^{\nu(2-\psi)} P_k(t)$, versus the scaling variable $\xi = kt^{\nu(1-\psi)}$ at three different times $t = 10^4, 10^5$ and 10^6 . Different scales correspond to $F(\xi)$ and $G(\xi)$ in the main figure since the latter diverges at the origin. The data represent an average over 10^3 independent realizations in a system of size $L = 10^6$ for the case $p = \frac{1}{2}$. The exponent value $\psi = 0.619$ was used. The tail of the distribution is shown in the inset.

passive clusters follow the scaling forms of equations (2) and (4), respectively (see figure 2). In agreement with the mean-field theory, the scaling functions decay exponentially for large masses.

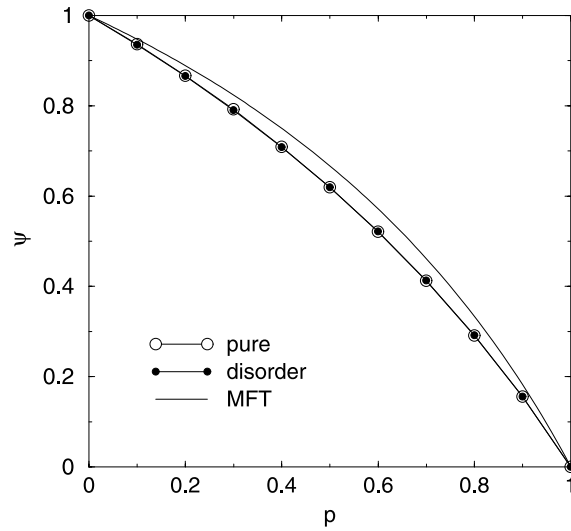


Figure 3. The exponent ψ versus p . Monte Carlo simulation results for the pure diffusion case are compared with the mean-field theory (8) and the exact value for the disordered case. The latter is obtained from $U(-2/(2-p), 2\psi, 2) = 0$ [7], where $U(a, b, z)$ is the confluent hypergeometric function.

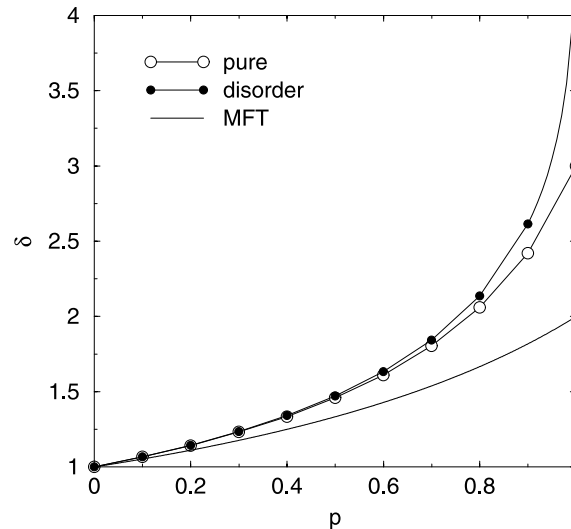


Figure 4. The exponent δ versus p . Monte Carlo simulation results for the pure diffusion case are compared with the mean-field theory (8) and the exact value for the disordered case obtained from $U(-2(1-p)/(2-p), 2\delta, 2) = 0$ [7].

We also analysed how the exponents vary with the probability p , as shown in figures 3 and 4. The exact exponents for the disordered case found by Le Doussal and Monthus [7] provide an excellent approximation (within 0.1%) for ψ . In the case of δ , a different behaviour emerges in the aggregation limit, $p \rightarrow 1$, where the exact value is $\delta = 3$ [16], and the disagreement with both mean-field theory and the disordered case are most pronounced.

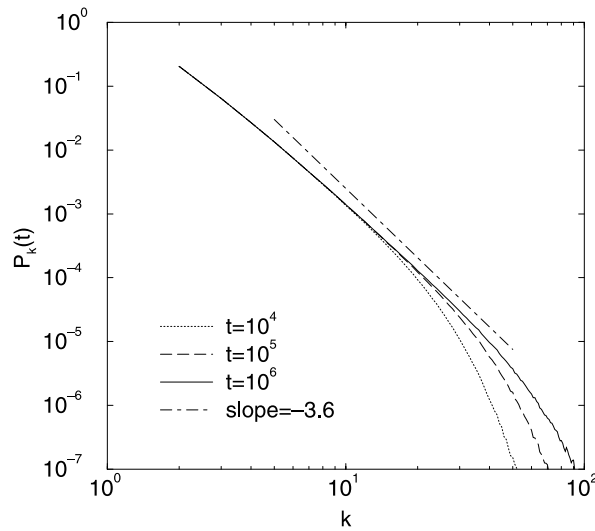


Figure 5. The distribution $P_k(t)$ versus k for three different times $t = 10^4$, 10^5 and 10^6 . The typical mass at these three times is proportional to $k^* \equiv t^{\nu/(1-\psi)} \propto 6$, 10 and 16 , respectively. Hence, the distribution is fully developed only over a short range of masses. The data represent an average over 10^3 realizations in a system of size $L = 10^6$ with $p = \frac{1}{2}$.

The above scaling arguments suggest that the limiting mass distribution of passive clusters decays algebraically with the exponent $\gamma = (2 - \psi)/(1 - \psi)$. For $p = \frac{1}{2}$, one therefore expects $\gamma \cong 3.627$ (compare with $\gamma = 3.62722$ and $\gamma = 4$, predicted by the disordered case and the mean-field theory). This corresponds to a very strong suppression of large masses, and it is therefore much more difficult to confirm this behaviour numerically. Nevertheless, our simulations (figure 5) are consistent with a power-law decay with an exponent $\gamma \cong 3.6$.

In one dimension, the diffusion-controlled stochastic aggregation is equivalent to the Potts model with zero-temperature Glauber dynamics [17]. For the Q -state Potts model with spatially uncorrelated initial conditions, aggregation of domain walls occurs with probability $p = (Q - 2)/(Q - 1)$, and annihilation occurs with probability $q = 1/(Q - 1)$. Therefore, the above can be reformulated in terms of domain walls rather than aggregates. In the coarsening context, the domain wall mass (or number) distribution is dual to domain number distribution [5–7].

5. Ballistic transport

The situation in which particles move ballistically involves a number of complications. Firstly, while the annihilation limit is uniquely defined [18–23], the aggregation limit has various realizations. In traffic flows the velocity of a newly born cluster is the smaller of the two velocities [24], whereas in application to astrophysics and granular gases the velocity follows from momentum conservation [25, 26]. Secondly, the numerical results for the annihilation case [19] and analytical results for the traffic case [24] show that the initial conditions are ‘remembered’ forever, in contrast to the diffusive case. Specifically, the small-velocity characteristics of the initial velocity distribution influence the long-time asymptotic behaviour, including the scaling exponents.

We consider the momentum-conserving case, also known as ‘ballistic aggregation’ or ‘sticky gas’ [27–33]. The initial velocities are assigned according to the distribution $P_0(v)$. The mass (momentum) of a newly born cluster is equal to the sum of masses (momenta) of the two colliding clusters. After an agglomeration event, the newborn particle remains active with probability p , or becomes passive with probability $q = 1 - p$.

To apply the Smoluchowski-rate-equations approach, we again use dimensional analysis to calculate the decay exponent ν . The collision rate is $r \sim va^{d-1}$, where v is the typical velocity and a is the typical radius of an aggregate. A particle of radius a contains of the order a^d monomers whose initial momenta are uncorrelated. Momentum conservation therefore implies $v \sim a^{-d/2}$. Using $a^d \sim M/A \sim A^{\psi-1}$ gives the collision rate $r \sim a^{(d-2)/2} \sim A^{(d-2)(\psi-1)/2d}$. From equation (7) one finds

$$\nu = \frac{2d}{d+2+\psi(d-2)} \quad (10)$$

with ψ given by equation (8). Apart from the exponent ν , features such as the exponential mass distribution and the exponents ψ and δ are given by the mean-field theory outlined above. In two dimensions, the collision rate does not depend on ψ and hence the asymptotic behaviour $A \sim t^{-1}$ agrees with that found for deterministic ballistic agglomeration [27]. For $d \neq 2$, stochastic and deterministic asymptotics differ: stochasticity enhances the decay of the number density A for $d < 2$ and weakens it for $d > 2$. A more detailed mean-field theory can be carried. It yields a factorizing joint mass-velocity distribution, with an exponential mass distribution, and a Gaussian velocity distribution [28, 31].

In the aggregation case, $\psi = 0$ and therefore the correct scaling exponent $\nu = 2d/(d+2)$ [27] is recovered from equation (10). For the annihilation case, however, initial conditions are ‘remembered’ forever and therefore the above dimensional arguments no longer hold. The predicted exponent in the annihilation case is always mean-field $\nu = 1$, while one-dimensional numerical simulations yield an exponent continuously varying from 0 to 1 depending on the initial velocity distribution $P_0(v)$, e.g., $\nu \approx 0.8$ for uniform initial distributions [19, 22].

We have simulated the stochastic aggregation process on a one-dimensional ring with 10^6 particles. The initial velocity distribution was uniform in $[-1, 1]$. We measured the scaling exponent ψ via the scaling relation $M \sim A^\psi$ rather than directly versus time, since the exponent $\nu(p)$ is not known analytically. We have found that the mean-field prediction, $\psi = (2-2p)/(2-p)$, provides a reasonable approximation for the exponent ψ , as shown in figure 6. Furthermore, this approximation should improve in higher dimensions.

We compared the mean-field prediction for the mass distribution of passive clusters, equation (9), with the numerically obtained distributions in both ballistic and diffusive cases. Interestingly, the rate equations provide an excellent approximation for small and moderate masses (see figure 7). Given that the discrepancy in ψ is maximal for the case $p = \frac{1}{2}$, one may expect an even better approximation for other values of p . Noting the strong decay of this distribution, the contribution of very large masses is extremely small; for example, $P_{100}(\infty) \approx 2.4 \times 10^{-7}$ for $p = \frac{1}{2}$. Hence, the most pronounced part of the distribution is well approximated by the rate-equations theory. Surprisingly, the transport mechanism does not play an important role as far as the final mass distribution of passive clusters is concerned.

6. Discussion

We have investigated diffusion- and ballistic-controlled stochastic aggregation in one dimension. We have seen that the rate-equations approach captures the overall scaling

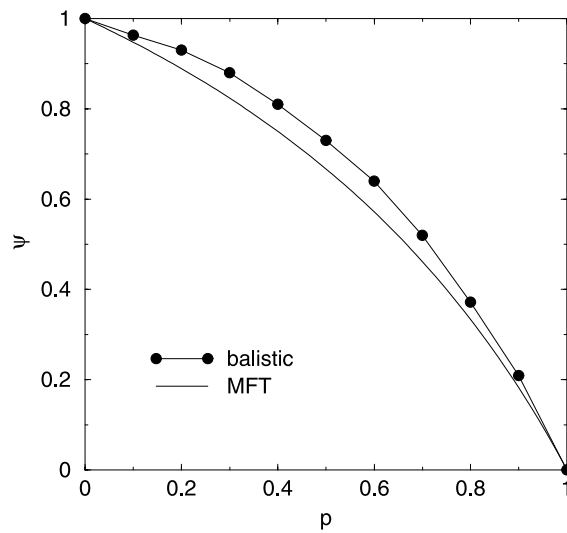


Figure 6. The scaling exponent $\psi(p)$ versus p for ballistic aggregation compared with the mean-field value of equation (8).

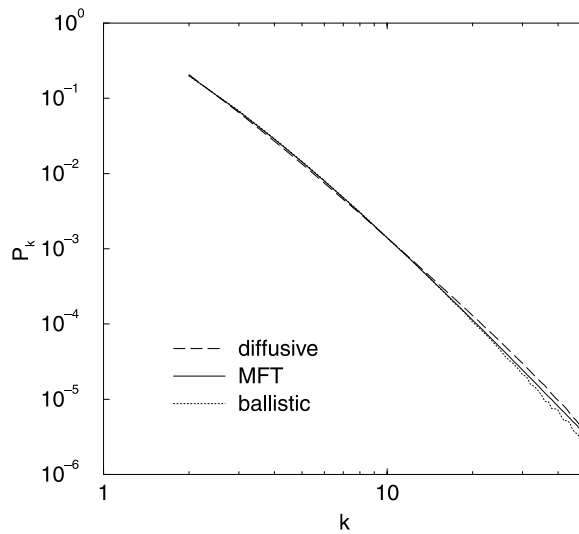


Figure 7. The final distribution of passive clusters for the $p = \frac{1}{2}$ stochastic aggregation with diffusive and ballistic transport. Also shown is the mean-field distribution $P_k(\infty) = 24/[k(k+1)(k+2)(k+3)]$.

behaviour and additionally it provides reasonable estimates for the decay exponents. In general, the mass distribution is characterized by two non-trivial model-dependent decay exponents.

In the diffusion-controlled case, the exponent ψ underlying the survival probability of a particle is in excellent agreement with the exact results from the disordered case. In fact, one cannot dismiss the possibility that the disordered and the pure values are identical, based on numerics alone. However, there is an evident discrepancy in the exponent δ as the disordered case exponent diverges logarithmically in the aggregation limit. Stochastic aggregation is

equivalent to domain coarsening in the zero-temperature Potts–Glauber model. The above exponents (ψ , δ) characterize the domain-wall number distribution in analogy with (ψ_D , δ_D) for the domain number distribution [5]. In the latter case as well, the exact values calculated for the disordered case provide an excellent approximation for the domain exponents. In general, the particle survival probability exponent ψ is robust, while the monomer survival probability exponent δ is very sensitive to the details of the process.

In the ballistic-controlled case, we have shown that even in the absence of a consistent mean-field theory, some characteristics such as the exponent ψ are well approximated by the rate equations. Understanding of reaction processes with an underlying ballistic transport remains largely incomplete. The asymptotic behaviour is highly sensitive to the initial conditions, and the critical dimension is apparently infinite. In fact, exact analytical results are available mainly in the aggregation limit [24, 32, 33].

The most intriguing property of the stochastic aggregation is the profound lack of universality. Indeed, the weak dependence on the transport mechanism is in contrast to the strong dependence on the parameter p . For example, our numerical results show that the final distribution of passive clusters is very close in diffusion- and ballistic-controlled situations. Another very impressive manifestation of this is the excellent agreement between the values of the exponent $\psi(p)$ in the disordered and pure cases.

Acknowledgments

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