Aggregation in the presence of sources and sinks: A scaling theory

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A scaling generalization of the Smoluchowski equation is used to treat fluctuation effects in aggregation problems. In particular, we investigate the diffusion-limited cluster-cluster aggregation subject to the condition that single particles are fed into the system at a constant rate, h, while clusters larger than a fixed size are removed. Considering the zero-feed-rate limit as a critical point, we find that h plays the role of an external field conjugate to the order parameter which turns out to be the cluster density. The cluster density obeys dynamic scaling and, because of the finiteness of a kinetic coefficient, the dynamic critical exponents are expressible in terms of a static exponent. The exponents are determined by arguing that the zero-feed-rate process is in one universality class with the $A+A\rightarrow 0$ diffusive annihilation problem. Our scaling theory is in agreement with available Monte Carlo simulation data.

I. INTRODUCTION

Irreversible aggregation appears to be the principal process underlying a variety of phenomena such as colloid growth, polymerization, aerosol formation, red-blood-cell aggregation, nucleation at phase transitions, etc.[1,2] From a general point of view, aggregation is interesting because it produces scale invariant clusters even in its simplest variant when particles and clusters execute a random walk and stick irreversibly on contact (diffusion-limited cluster-cluster aggregation[3,4]). This scale invariance can be described in terms of anomalous dimensions such as, e.g., the fractal dimension[5] and the task of an analytical theory would be the calculation of the exponents determining those dimensions. This is not an easy task, however. As evidenced by Monte Carlo simulations,[6,7] the cluster-cluster aggregation is qualitatively similar to the relaxation towards equilibrium at a critical point but an additional complication is also present. The dynamic scaling observed for relatively short times might break down as a result of the appearance of an infinite spanning cluster (gelation).[7,8]

In order to bring the analogy with equilibrium critical phenomena closer and to make the methods developed for that case applicable, one would like to modify the cluster-cluster aggregation so that the system would relax to a well-defined steady state. Furthermore, it would be desirable to eliminate the possibility of gelation since it has no relevance to the problem of aggregation in the regime when the average distance between the clusters is much larger than the radius of the clusters. A possible modification consists of removing the larger clusters and creating smaller ones according to some rules. Removal processes such as sedimentation and outflow from a chemical reactor are present in real systems and, as indicated by the rate-equation studies of aggregation,[9,10] gelation is prevented if the removal rate is sufficiently weighted toward larger clusters. Particle sources may also play an important role in aggregation processes, e.g., there is inflow into a chemical reactor or seeds for secondary aerosol formation may be produced by photooxidation. In the presence of sources and sinks, the system usually evolves to a steady state which has been investigated in detail using the Smoluchowski equation approach. E.g., theorems giving the conditions for the existence of the steady state are provided in Ref. 10 and 11 while numerical studies and comparisons with experiments[12] can be found in Refs. 13 and 14.

In this paper we consider cluster-cluster aggregation with the simplest choice of sources and sinks. Single particles are fed into the system homogeneously and at a fixed rate h while clusters containing more than a prescribed number (k_0) of particles are eliminated instantaneously. Our aim with the study of this simple model is to suggest that the zero-feed-rate limit ($h=0$) may be considered as a critical point and consequently, around this point, the Smoluchowski equation approach may be generalized into a scaling description of aggregation. This is the topic of Sec. II where we shall see that the feed-rate plays the role of an external field conjugate to the order parameter which might be taken to be the cluster density n.

The results of the scaling theory can be summarized in two scaling laws. The first one

$$\Delta'+\frac{1}{\delta}=1 \tag{1}$$

relates the exponent δ of the steady-state cluster density

$$\bar{n}_h \sim h^{1/\delta} \tag{2}$$

to that of the relaxation time at fixed h:

$$\tau_h \sim h^{-\Delta'} . \tag{3}$$

This scaling law relating a static and a dynamic exponent is similar in spirit to the $\Delta=\gamma$ relationship in equilibrium systems where Δ is the critical exponent of the relaxation time of the order parameter while γ is the susceptibility exponent. Apart from the scaling assumption, the derivation of both of these relationships involve the assumption

that the appropriate kinetic coefficient is not singular at the critical point. We shall argue that this is a valid assumption for cluster-cluster aggregation. Supporting evidence comes from Monte Carlo simulations[15] of diffusion-limited cluster-cluster aggregation where the scaling law (1) is found to hold in $d=1$, 2, and 3 dimensions.

The second scaling law

$$\zeta\Delta'=\frac{1}{\delta} \tag{4}$$

is based solely on the scaling assumption. It involves a third exponent ζ which characterizes the power-law decay of the cluster density at $h=0$

$$n(t)\sim t^{-\zeta} . \tag{5}$$

This scaling law is useful because, as is discussed in Sec. III, the $h=0$ dynamics can be thought of as a diffusive annihilation problem[16] and the exponent ζ is well known for that case. There are exact results[17] for $d=1$ and various analytical approaches[18] and Monte Carlo simulations[19–21] indicate that the critical dimension d_c is two. For $d>d_c$ one has $\zeta=1$ while

$$\zeta=\frac{d}{2} \tag{6}$$

for $d<2$. Then all the exponents are determined since δ and Δ' follows from Eqs. (1) and (4)

$$\delta=\frac{d+2}{d},\quad \Delta'=\frac{2}{d+2},\quad d<2 \tag{7}$$

while mean-field results which can be derived from the Smoluchowski equation apply for $d>2$

$$\delta=2,\quad \Delta'=\tfrac{1}{2} . \tag{8}$$

Comparing these predictions [Eqs. (7) and (8)] with the Monte Carlo results,[15] one can see good agreement for $d=1$ and 3 but considerable deviation is observed for $d=2$. The disagreement might be due to the fact that $d_c=2$ and consequently the logarithmic corrections to scaling make the finite-size analysis difficult in $d=2$. Of course, it is not excluded that, as argued in Ref. 15, the fractal nature of the clusters and the presence of clusters of different size may make the process distinct from diffusion-limited annihilation.

II. SCALING GENERALIZATION OF THE SMOLUCHOWSKI EQUATION

The first mean-field-like treatment of irreversible coagulation is due to Smoluchowski[22,23] who assumed that clusters coalesce through binary collisions and proposed the following rate equation to describe the time evolution of the concentration $n_k(t)$ of k-particle clusters:

$$\frac{dn_k(t)}{dt}=\tfrac{1}{2}\sum_{\substack{i,j\\ i+j=k}} K_{ij}n_i(t)n_j(t)-n_k(t)\sum_j K_{jk}n_j(t) . \tag{9}$$

Here the first sum gives the increase of $n_k(t)$ as a result of the coalescence of clusters satisfying the condition $i+j=k$ while the second sum accounts for the decrease of $n_k(t)$ due to the collision of the k-particle clusters with clusters of any size. An appropriate choice of the collision kernel K_{ij} takes partial care of such details of the process as how the diffusivity of the clusters affects the probability of their meeting and how the collision cross section depends on the size of the clusters. The spatial fluctuations in the cluster distribution are completely neglected and this is why Eq. (9) is considered to be the mean-field theory of aggregation.[24]

The generalization of Eq. (9) to include sources and sinks is straightforward.[9–14,25] In our case, the production of single particles at a fixed rate h (particles per unit volume per unit time) is taken into account by adding an $h\delta_{k1}$ term to the right-hand side of Eq. (9) while the instantaneous elimination of clusters containing more than k_0 particles means that we must set $n_k(t)\equiv 0$ for $k>k_0$. Thus the equation to study is as follows:

$$\frac{d}{dt}n_k(t)=h\delta_{k1}+\tfrac{1}{2}\sum_{\substack{i,j\\ i+j=k\le k_0}} K_{ij}n_i(t)n_j(t) -n_k(t)\sum_{j=1}^{k_0} K_{jk}n_j(t),\quad k=1,2,\ldots,k_0 . \tag{10}$$

This equation has simple scaling properties with respect to the feed rate h. Introducing scaled time and scaled cluster-size distribution

$$\tilde{t}=h^{1/2}t,\quad \tilde{n}_k(\tilde{t})=\frac{n_k(t)}{h^{1/2}} \tag{11}$$

one can see that h is eliminated from Eq. (10), and consequently, the solution of that equation can be written in the form

$$n_k(t,h)=h^{1/2}\phi_k(h^{1/2}t) . \tag{12}$$

This form is exact only if $n_j(0)\sim h^{1/2}$. Otherwise ϕ_k also depends on h through the initial conditions $\tilde{n}_j(0)=n_j(0)/h^{1/2}$. We believe, however, that the long-time behavior and the steady-state properties are insensitive to the initial conditions and thus, for our purposes, Eq. (12) gives an adequate description of the system. Since the cluster density is given by

$$n(t,h)=\sum_{k=1}^{k_0} n_k(t,h) , \tag{13}$$

substitution of (12) into (13) gives a scaling form for the time-dependent cluster density

$$n(t,h)=h^{1/2}\phi(h^{1/2}t) . \tag{14}$$

It follows from Eq. (14) that if the system reaches a steady state in the $t\to\infty$ limit then the steady-state cluster density $\bar{n}$ scales as

$$\bar{n}\sim h^{1/\delta}\sim h^{1/2} , \tag{15}$$

and we obtain $\delta=2$, a result which has been known from previous studies.[13,14] It also follows from Eq. (14) that, in

the $h \to 0$ limit, $n(t,h)$ approaches its steady-state value $\bar{n}=0$ by power-law decay

$$n(t,0) \sim t^{-\zeta} \sim t^{-1} , \tag{16}$$

i.e., mean-field predicts $\zeta=1$.

Finally, the relaxation time to a steady state at a fixed h can be calculated as the area under the relaxation function

$$\psi(t,h)=[n(t,h)-n(\infty,h)]/[n(0,h)-n(\infty,h)]$$

which gives

$$\tau_h=\int_0^\infty \psi(t)dt \approx \int_0^\infty \frac{\phi(h^{1/2}t)-\phi(\infty)}{\phi(0)-\phi(\infty)}dt \sim h^{-\Delta'} \sim h^{-1/2} , \tag{17}$$

hence the mean-field value of Δ' is $\frac{1}{2}$. It should be noted here that in calculating τ_h it is assumed that $n(0,h)-n(\infty,h) \sim h^{1/2}$. This assumption ensures that the initial state is close to the final steady state and consequently τ_h measures the linear ("equilibrium") relaxation time. Otherwise one would deal with the so-called nonlinear relaxation time[26] which diverges with an exponent distinct from Δ'.[27,28] We also note that the possible integrability problems in (17) may be avoided by defining τ_h as the time for ψ to relax to $\frac{1}{2}$. The result is again $\tau_h \sim h^{-1/2}$.

Comparing the mean-field results ($1/\delta=0.5$, $\Delta'=0.5$) with the available Monte Carlo data[15] ($1/\delta=0.33\pm0.02$; 0.40 ± 0.04; 0.48 ± 0.05 and $\Delta'=0.67\pm0.03$; 0.62 ± 0.05; 0.57 ± 0.05 for $d=1,2,3$, respectively) one can find satisfactory agreement only in $d=3$. Otherwise a pattern familiar from the theory of critical phenomena emerges, the discrepancy increases as the dimensionality is lowered, thus indicating the enhanced importance of the neglected spatial fluctuations.

Since the singularity of the relaxation time [Eq. (17) and the power-law decay of $n(t)$ (Eq. (16)] suggests that the $h=0$ point should be regarded as a critical point, one can follow the theory of critical phenomena and treat the spatial fluctuations on a phenomenological level by a scaling generalization of the Smoluchowski equation. To do this, note that Eq. (10) can be written in the form

$$\frac{d}{dt}n_k(t)=h\delta_{k1}-G_k(n_1,n_2,\ldots,n_{k_0}) \tag{18}$$

where G_k is a homogeneous function of second degree

$$G_k(\lambda n_1,\lambda n_2,\ldots,\lambda n_{k_0})=\lambda^2 G_k(n_1,n_2,\ldots,n_{k_0}) . \tag{19}$$

The simplest generalization of Eq. (18) is to keep the degree of homogeneity as a free parameter

$$G_k(\lambda n_1,\lambda n_2,\ldots,\lambda n_{k_0})=\lambda^\delta G_k(n_1,n_2,\ldots,n_{k_0}) . \tag{20}$$

The scaling analysis of the resulting equation is similar to that of the original Smoluchowski equation. Changing to scaled time and scaled cluster-size distribution

$$\tilde{t}=h^{1-1/\delta}t, \quad \tilde{n}_k(\tilde{t})=\frac{n_k(t)}{h^{1/\delta}} , \tag{21}$$

one eliminates h from Eq. (18) and thus the solution for large t can be written as

$$n_k(t,h)=h^{1/\delta}\phi_k(h^{1-1/\delta}t) . \tag{22}$$

As a consequence we obtain the cluster density in a scaled form

$$n(t,h)=h^{1/\delta}\phi(h^{1-1/\delta}t) \tag{23}$$

which is in agreement with the Monte Carlo simulations.[15] As in the mean-field case, Eq. (23) implies $\bar{n} \sim h^{1/\delta}$, $n(t,0) \sim t^{-\zeta}$, and $\tau_h \sim h^{-\Delta'}$ but now the critical indexes are not determined explicitly. Instead, they are expressed through the homogeneity index δ, i.e., we obtain two scaling laws

$$\Delta'+\frac{1}{\delta}=1 \tag{24}$$

and

$$\zeta\Delta'=\frac{1}{\delta} \tag{25}$$

connecting the three exponents. Monte Carlo simulation[15] are in agreement with the scaling law (24) and since Eq. (25) is based only on the scaling form (23) there is implicit confirmation of this scaling law as well.

At first sight it might seem strange that one parameter (δ) determines both the static and dynamic exponents of the system. To understand this, let us briefly recall how the scaling theory is extended to dynamics near an equilibrium critical point. For a purely relaxational system with a one-component order parameter m (model A in Ref. 29; for example, let us think of a uniaxial ferromagnet in a magnetic field H), one assumes that the time evolution towards the minimum free energy configuration is described by a van Hove–type equation[28,30,31]

$$\frac{dm}{dt}=\Gamma\left[H-\frac{\partial F}{\partial m}\right] . \tag{26}$$

Here F is the appropriate free energy which is assumed to obey scaling near the critical point. In particular, restricting ourselves to the critical isotherm we have[32]

$$\frac{\partial F}{\partial m} \approx H_0 m^\delta . \tag{27}$$

In general, the kinetic coefficient Γ is also singular at the critical point and also obeys scaling. On the critical isotherm it is assumed to have the form

$$\Gamma(m) \approx \Gamma_0 m^\sigma . \tag{28}$$

Equation (26) together with (27) and (28) determines the scaling properties of the system on the critical isotherm. Quantities such as the equilibrium value of magnetization ($\bar{m} \sim H^{1/\delta}$), the decay at the critical point [$m(t) \sim t^{-\zeta}, \zeta=(\delta+\sigma-1)^{-1}$], and the relaxation time [$\tau_H \sim H^{-\Delta'}, \Delta'=(\delta+\sigma-1)/\delta$] can be calculated and one can see that δ determines the static properties while both σ and δ enter into the expression for the dynamic exponents. If the kinetic coefficient is not singular ($\sigma=0$) then the static exponent (δ) determines the dynamic behavior as well and we obtain scaling laws identical to

those found for aggregation.

To understand why the kinetic coefficient is constant in the case of aggregation, let us derive an equation for the average density of clusters $n(t)$ which seems to play a role similar to that of the order parameter $m(t)$. Returning to Eq. (18) and summing it over $k=1,2,\ldots,k_0$, one finds

$$\frac{dn}{dt}=h-\sum_{k=1}^{k_0} G_k(n_1,n_2,\ldots,n_{k_0}) . \tag{29}$$

Now we make an approximation used in deriving equations like Eq. (26); namely, we assume that the sum on the right-hand side depends only on n:

$$\frac{dn}{dt}=h-G(n) . \tag{30}$$

This equation ought to reproduce the scaling behavior as obtained from the cluster-size distribution treatment [Eq. (23)] so the function $G(n)$ should behave as $G(n)\sim G_0 n^{\delta}$ for small n. Thus we have arrived at a special form of Eq. (26), namely for aggregation we have $\Gamma=1$. It should be emphasized that this feature is not a consequence of the approximations. There is a basic difference between H and h in Eqs. (26) and (30). The magnetic field tends to align the magnetic moments along the field but its effect appears through a complicated interplay with the thermal fluctuations in the system. Thus the timescale on which the effect of H is felt depends on the magnitude of fluctuations and this is why Γ in Eq. (26) might become singular at the critical point. The effect of h, on the other hand, appears on a timescale which is independent of the aggregation process. The feed rate h just describes how often the single-particle clusters are added to the system. So, if an equation of the form (26) can be used for calculating the scaling properties of the cluster density in the aggregation processes, then $\Gamma=1$ necessarily in that equation.

One might question, of course, the existence of Eq. (26) since the memory effects which are known to be amplified near a critical point are neglected. In some circumstances (e.g., near four dimension in case of a ferromagnet), the memory effects can be taken care of by renormalizing the kinetic coefficient.[29,33] The main point of the above argument, however, is that the kinetic coefficient is not renormalized in the case of aggregation and thus one might expect that the dynamic critical exponents can be expressed through the static ones. Monte Carlo simulations[15] seem to indicate that at least in case of diffusion-limited cluster-cluster aggregation, this expectation is justified.

III. ANALOGIES WITH ANNIHILATION PROBLEMS

As discussed in Sec. II, the absence of a singularity in the kinetic coefficient Γ means that only one of the exponents δ, Δ', and ζ has to be determined, the other two follow from the scaling laws (24) and (25). The most promising way to proceed seems to be the calculation of ζ. This exponent describes the decay of cluster density when no particles are fed into the system, a case which is reminiscent of the annihilation problems[16] much studied in chemistry. Indeed, when $k_0=1$ and the two particle clusters disappear from the system, the aggregation process corresponds to the

$$A+A\rightarrow 0$$

chemical reaction where A and 0 denotes the active and inert molecules, respectively. When the A molecules move around by diffusion, this reaction is called diffusive annihilation[16] and has been studied extensively.[17–20] The one-dimensional model can be solved exactly[17] with the result

$$\zeta=\tfrac{1}{2} \quad (d=1) . \tag{31}$$

It is remarkable that, as rather extensive Monte Carlo simulations[15] indicate, the same result holds for a one-dimensional diffusive cluster-cluster aggregation model[21] where the colliding clusters coalesce into a cluster occupying only one lattice site.

No exact result is available for $d>1$ but the common view, based on dimensional analysis,[18] analytical and scaling approaches,[18,34] and Monte Carlo data,[19,20] is that $d_c=2$ for diffusive annihilation. Mean-field theory [$\zeta=1$, Eq. (16)] applies above $d_c=2$ and a simple functional form $\zeta=d/2$ seems to hold for $d<2$.

If we assume now that the universality class is not changed when going from $k_0=1$ to any other finite value of k_0 then we have ζ and consequently Δ' and δ as discussed in the Introduction [Eqs. (7) and (8)]. We cannot provide a rigorous justification for the above assumption but the following points will clarify the ideas which form the basis for it.

(i) For $k_0>1$, we have clusters of different size in the system so the process superficially resembles the

$$A+B\rightarrow 0$$

annihilation problem which is not in the same universality class with the $A+A\rightarrow 0$ reaction.[18,20] The distinction is not surprising because it is known from the theory of critical dynamics[29] that the conservation laws play an important role in determining the universality classes and the two processes are distinct because there are no conservation laws in the $A+A\rightarrow 0$ case while the difference between the number of A and B atoms is conserved in the $A+B\rightarrow 0$ reaction. Looking now at diffusion-limited cluster-cluster aggregation from the point of view of conservation laws, one can see that no conservation laws appear when going from $k_0=1$ to $1<k_0<\infty$ so the process should be in the same universality class as the $A+A\rightarrow 0$ diffusive annihilation.

(ii) It has been observed[4,35] that if the diffusivity of clusters decreases with increasing cluster size then the process of diffusion-limited cluster-cluster aggregation is dominated by the sticking of clusters of almost the same size. This means that after a while only clusters containing more than $k_0/2$ particles are present in the system and consequently all collisions lead to annihilation. Thus, in the long-time limit, the process is equivalent to the $A+A\rightarrow 0$ type diffusive annihilation.

It should be noted, however, that the existence of a time-dependent characteristic cluster size is not a necessary condition for the equivalence with the $A+A\rightarrow 0$ reaction. If the diffusivity of the clusters is independent of their size, then the cluster-size distribution is not bell-shaped but monotonically decreasing with increasing clus-

ter size.[6] Nevertheless, the same exponents Δ' and δ are obtained[15] as in the case of size-dependent diffusivity.

(iii) The Monte Carlo simulations[15] lend numerical support to the universality assumption. The agreement with the predictions following from that assumption is good in $d=1$ and 3 and the discrepancy in $d=2$ probably can be explained by the presence of logarithmic corrections to scaling at the critical dimension.

In closing, we note that although the above arguments are quite convincing and thus the exponents δ, Δ', and ζ might be considered to be determined, one thing remained unaccomplished. For large k_0, one expects that the clusters grown in the system have the same fractal structure as the clusters of similar size grown without the presence of the sources and sinks. The intriguing question of what is the relationship (if any) between the fractal dimension of the clusters and the exponents δ, Δ', and ζ remained unanswered.

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