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Mean-Field Theory for Diffusion-Limited Cluster Formation

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A mean-field theory for the diffusion-controlled cluster formation is presented by considering the competition among the different portions of a growing cluster for the incoming diffusive particles. This competition is shown to introduce a screening length which depends inversely on the density of the cluster. The Hausdorff dimensionality D of these clusters is shown to be $(d^2+1)/(d+1)$ where d is the Euclidean dimensionality. This result is in excellent agreement with that of the computer simulations of Witten and Sander and of Meakin.

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In an attempt to describe the growth of clusters of small particles, Witten and Sander[1] recently simulated a diffusion-limited aggregation in $d=2$ on a computer and compared with other models such as Eden growth,[2] dendritic growth,[3] random animals,[4] and percolating clusters.[5] Meakin[6] also has independently performed simulations of diffusion-controlled cluster growth similar to those of Witten and Sander for $d=2$, 3, and 4. These simulations start with a single seed particle at the origin of a lattice. A second particle is added at some random site at a large distance from the origin. This particle undergoes a random walk on the lattice until it reaches a site adjacent to the seed and becomes part of the growing cluster. A third particle is then introduced at a random distant point and it walks randomly until it joins the cluster, and so forth. The simulations show that the clusters are critical objects having scale-independent correlations over an arbitrarily large range of distances and with the Hausdorff dimension[7] D of 1.68 ± 0.07, 2.51 ± 0.26, and 3.32 ± 0.10 for $d=2$, 3, and 4, respectively. D is defined for an N-particle aggregate by

$$N \propto R^D, \tag{1}$$

where R is the radius of gyration of the cluster.

I present a mean-field theory in an attempt to calculate D of these objects. The most important aspect of the growth of these clusters is that the various portions of the growing clusters compete for the incoming diffusive particle. The propagator for the diffusion of the individual particles has long-range inverse distance dependence. The competition between the various centers of the growing cluster for the incoming diffusing particles screens this long-range distance dependence. I calculate the screening length ξ by considering a large collection of small spheres within the coherent-potential approximation (CPA). I then deduce D by calculating the effective rate constant for the cluster to absorb the screened incident diffusive field.

First we consider the effect of the competition among N randomly distributed absorbers in a volume V for the diffusing density field from the macroscopic boundary. For simplicity, the absorbers are taken to be spheres of radius a. Let $d=3$; the generalization to other dimensions will follow. At steady state, the rate of production of density field due to external sources is just compensated by its removal by the absorbers. In the presence of the absorbers, the diffusive field satisfies

$$D_0 \nabla^2 u(\vec{r}) = \varphi(\vec{r}) + \sum_{i=1}^{N} \int d\Omega_i\, \delta(\vec{r} - \vec{R}_i)\sigma_i(\Omega_i), \tag{2}$$

where D_0 is the diffusion constant of the density field and $\varphi(\vec{r})$ is an auxiliary field to account for external boundary effects. $\varphi(\vec{r})$ defines the density field in the absence of any absorber. The second term on the right-hand side of Eq. (2) accounts for the presence of the absorbers and gives the net depletion of the density field due to all the absorbers. The ith absorber absorbs the density field with strength $\sigma_i(\Omega_i)$ at any space point on the surface of i, i.e., $\vec{R}_i = \vec{R}_i^{\,0} + \vec{r}_i(\Omega_i)$, where $\vec{R}_i^{\,0}$ is the position vector of the center of mass of i and $\vec{r}_i(\Omega_i)$ is the position vector of this surface point from $\vec{R}_i^{\,0}$; Ω_i denotes the orientation of $\vec{r}_i$.

Equation (2) expresses the microscopic density field at any space point $\vec{r}$ in terms of the various $\{\sigma_i\}$ and the problem reduces to determining these $\{\sigma_i\}$ in terms of φ. This is accomplished by employing the boundary condition of complete absorption of the density field at the surface of the absorbers,

$$u(\vec{R}_i) = 0. \tag{3}$$

More general boundary conditions can be used without any conceptual difficulties, but we restrict ourselves to this simple boundary condition. Although CPA, which is used below, is meaningful for the case of weak absorbers, the qualitative results are not affected within CPA whether Eq. (3) or a more general boundary condition is used in the actual calculations. The above boundary condition is directly related to the sticking probability in Meakin's simulations.

As we are interested in the macroscopic average density field, we average Eq. (2) over a distribution of $\{\vec{R}_i^{\,0}\}$ to obtain

$$D_0 \nabla^2 \langle u(\vec{r})\rangle = \varphi(\vec{r}) + \left\langle \sum_{i=1}^{N} \int d\Omega_i\, \delta(\vec{r} - \vec{R}_i)\sigma_i(\Omega_i)\right\rangle \equiv \varphi(\vec{r}) - \int d^3r'\, \Sigma(\vec{r} - \vec{r}\,')\langle u(\vec{r}\,')\rangle, \tag{4}$$

where the angular brackets denote the average over the appropriate distribution function for the absorbers. In the second equality of Eq. (4) a macroscopic kernel Σ has been defined so that Eq. (4) has the form of a linear law. With the introduction of the Fourier space transform of a field $f(\vec{r})$ as $f(\vec{k}) = \int d^3r f(\vec{r}) \exp(-i\vec{k}\cdot\vec{r})$, Eq. (4) becomes

$$[D_0 k^2 + \Sigma(\vec{k})]\langle u(\vec{k})\rangle = f(\vec{k}). \tag{5}$$

The mathematical structure of Eq. (5) implies that the k^2 part of $\Sigma(\vec{k})$ gives the change in the diffusion constant $D(\rho) - D_0$, where $D(\rho)$ is the density-dependent diffusion coefficient with $\rho = N/V$. Also, the constant term in Σ, i.e., $\Sigma(k=0)$, defines the screening length $\xi(\rho)$ for the diffusion:

$$\frac{1}{2}\frac{d^2\Sigma(k)}{dk^2}\bigg|_{k=0} = D(\rho) - D_0, \tag{6a}$$

$$\Sigma(k=0) \equiv D_0 \xi^{-2}(\rho). \tag{6b}$$

$\Sigma(k=0)$ is often called the rate constant,[8] K, for the diffusion-controlled process. Thus, a knowledge of the kernel Σ leads to the values of the diffusion and rate constants for a given ρ. Here, we confine ourselves to the calculation of ξ although caution must be exercised regarding the self-consistent dependence[9] on $D(\rho)$.

Although $\Sigma(\vec{k})$ can be calculated as a diagrammatic series expansion in ρ and then summing the diagrams, I present a simple effective-medium argument to obtain $\Sigma(\vec{k})$. By considering a single absorber which absorbs an effective field $u(\Sigma(\vec{k}))$, where $\Sigma(\vec{k})$ is yet unknown, we calculate Σ_1, the contribution of one such absorber to $\Sigma(\vec{k})$. Since $\Sigma(\vec{k})$ is N times Σ_1 which in turn depends on $\Sigma(\vec{k})$,

$$\Sigma = N\Sigma_1(\Sigma), \tag{7}$$

the requirement of self-consistency leads to Σ.

Σ_1 is calculated as follows. The equation for the density field, when a single absorber is present in an effective medium, is

$$D_0\nabla^2 u(\vec{r}) + \int d^3r'\,\Sigma(\vec{r}-\vec{r}\,')u(\vec{r}\,') = f(\vec{r}) + \int d\Omega_1\,\delta(\vec{r}-\vec{R})\sigma_1(\Omega_1) \tag{8}$$

with the boundary condition

$$u(\vec{R}_1) = 0. \tag{9}$$

Combining Eqs. (8) and (9), we get

$$0 = \int d^3r'\,G(\vec{R}_1-\vec{r}\,') + \int d\Omega_1'\,G(\vec{R}_1-\vec{R}_1')\sigma_1(\Omega_1'), \tag{10}$$

where

$$G(\vec{r}) = \int d^3k\,(2\pi)^{-3}\,G(k)\exp(+i\vec{k}\cdot\vec{r}),$$

$$G(\vec{k}) = [D_0k^2 + \Sigma(\vec{k})]^{-1}. \tag{11}$$

By defining the inverse operator G^{-1} on the surface of the sphere 1 according to

$$\int d\Omega_1'\,G^{-1}(\vec{R}_1,\vec{R}_1')G(\vec{R}_1',\vec{R}_1'') = \delta(\vec{R}_1-\vec{R}_1'') \tag{12}$$

we get from Eq. (10)

$$\sigma_1(\Omega_1) = -\int d^3r'\,d\Omega_1'\,G^{-1}(\Omega_1,\Omega_1')\,\delta(\vec{R}_1'-\vec{r}\,')\tilde{u}(\vec{r}\,'), \tag{13}$$

where

$$\tilde{u}(\vec{r}) = \int d^3r'\,G(\vec{r}-\vec{r}\,')f(\vec{r}\,').$$

Substitution of Eq. (13) into Eq. (8) yields

$$u(\vec{r}) = \tilde{u}(\vec{r}) - \int d^3r'\,d\Omega_1 d\Omega_1'\,\delta(\vec{r}-\vec{R}_1)G^{-1}(\Omega_1,\Omega_1')\delta(\vec{R}_1'-\vec{r}\,')\tilde{u}(\vec{r}\,'). \tag{14}$$

Therefore $\Sigma_1(\vec{k})$ follows as

$$\begin{aligned}\Sigma_1(\vec{k}) &= V^{-1}\int d(\vec{r}-\vec{r}\,')\,d\Omega_1 d\Omega_1'\exp[-i\vec{k}\cdot(\vec{r}-\vec{r}\,')]\,\delta(\vec{r}-\Omega_1)G^{-1}(\Omega_1,\Omega_1')\,\delta(\Omega_1'-\vec{r}\,')\\ &= V^{-1}\int d\Omega_1 d\Omega_1'\,G^{-1}(\Omega_1,\Omega_1')\exp[-i\vec{k}\cdot(\Omega_1-\Omega_1')],\end{aligned} \tag{15}$$

where G^{-1} depends on Σ through Eqs. (11) and (12) and V appears as a result of the averaging over the position of the center of mass of the single sphere over the whole volume. Expanding G^{-1} and G in spherical harmonics and combining Eqs. (7), (11), (12), and (15) we get

$$\Sigma(k=0) = 4\pi\rho G_0^{-1}, \tag{16}$$

with

$$G_0 = \frac{1}{4\pi}\int d\Omega_1 d\Omega_1'\int d^3k\,(2\pi)^{-3}[D_0k^2+\Sigma(\vec{k})]^{-1}\exp[i\vec{k}\cdot(\Omega_1-\Omega_1')] = (D_0a)^{-1}I_{1/2}(a/\xi)K_{1/2}(a/\xi), \tag{17}$$

where I_p and K_p are the modified Bessel functions of order p. In getting the second equality of Eq. (17), Eq. (6b) is utilized and the local structures of the problem reflected by the large-k part of Σ have been ignored. The k^2 term of Σ only alters D_0 as pointed out above and does not change[9] the power-law dependence between ξ and ρ.

Substitution of Eqs. (17) into Eqs. (16) and (6b) gives

$$\xi^{-2} = 8\pi a^2\rho\xi^{-1}[1-\exp(-2a/\xi)], \quad d=3. \tag{18}$$

For very large N, ξ becomes small so that the dependence of ξ on ρ and a is

$$\xi^{-1} \sim \rho a^2, \quad d=3. \tag{19}$$

Thus the competition effect between the various absorbers in a certain volume for the incoming diffusive field leads to an inverse density dependence for the screening length within the CPA description. It is to be noted that if the competition were completely absent then there is no screening ($\xi\to\infty$).

The results of Eqs. (18) and (19) can be generalized for other $d \geq 2$. The analysis is similar to the above and the result is

$$\Sigma(k=0) \sim \rho\left(\int_0^\infty dk\,\frac{k^{d-1}}{k^2+\xi^{-2}}\int_0^\pi d\theta \int_0^\pi d\theta' \sin^{d-2}\theta \sin^{d-2}\theta' \exp[-ika(\cos\theta - \cos\theta')]\right)^{-1} \quad (20)$$

$$\sim \rho a^{d-2}/I_{d/2-1}(a/\xi)K_{d/2-1}(a/\xi) \quad (21)$$

$$\sim \rho a^{d-1}\xi^{-1}[1+O(\xi/a)], \quad (22)$$

where Eq. (22) is obtained by considering the large-N limit. The use of Eq. (6b) in Eq. (22) gives the inverse dependence of ξ,

$$\xi^{-1} \sim \rho a^{d-1} \quad (23)$$

for all $d \geqslant 2$.

I now present a mean-field argument to calculate D. Consider an aggregate of N constituent spheres each of radius a. Let R be the unknown radius of gyration of the aggregate. Since ξ is the screening length, we perform a coarse graining over a distance ξ so that the renormalized radius of the aggregate is Ra/ξ. Taking the cluster as a single sphere of radius Ra/ξ absorbing the screened diffusive field, the rate constant is given by the d-dimensional version of Eq. (15) with the radius a replaced by Ra/ξ,

$$\hat{K} \sim \left(V\int_0^\infty dk\,\frac{k^{d-1}}{k^2+\xi^{-2}}\int_0^\pi d\theta \int_0^\pi d\theta' \sin^{d-2}\theta \sin^{d-2}\theta' \exp[-ikRa\xi^{-1}(\cos\theta - \cos\theta')]\right)^{-1} \sim \xi^{-d}R^{-1}, \quad (24)$$

where the large-N limit is taken as in Eq. (22) and $V = R^d$ is utilized. Since D_0 and a are microscopic quantities independent of N, they are left out in Eq. (24). $\hat{K}$ is defined also as a phenomenological rate constant for the cluster growth,

$$\partial R/\partial t \equiv \hat{K}R. \quad (25)$$

Since every particle in the computer simulations of Witten and Sander and of Meakin is made sure to be absorbed by the growing cluster before the next particle is introduced into the system, the role of the time variable is played by N in this problem. In view of Eq. (1), $\hat{K}$ follows from Eqs. (24) and (25) as

$$\hat{K} \sim 1/N \sim \xi^{-d}R^{-1}. \quad (26)$$

Because $\xi \sim \rho^{-1} \sim R^d/N$ [see Eq. (23)], it follows from Eq. (26) that

$$D = (d^2+1)/(d+1). \quad (27)$$

This result is in excellent agreement with that of the simulations quoted earlier. However, for higher dimensions, our value of d/D does not support the universal value of $\frac{6}{5}$ suggested by Meakin.[6]

In summary, I present a simple mean-field theory to describe the fractal nature of the diffusion-limited cluster growth. The main assumptions are as follows: (i) All particles of the growing cluster compete for the incident particle at every stage of growth. (ii) The collective behavior due to this competition is obtained by treating the various particles as small spheres and using CPA. (iii) The overall growth of the cluster does not depend on the structural details within distances of the order of the screening length. (iv) The time variable is proportional to the number of particles in the cluster (since the details of the actual dynamics of any particle during the period between its introduction into the system and its absorption are irrelevant in the simulations). The main reason for the agreement between our CPA (where density fluctuations are ignored) and the simulations is perhaps due to the very low densities in these ramified clusters.

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