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# Theory of diffusion-limited growth

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We review and extend the field-theoretic approach to diffusion-limited growth at a finite background walker density. This approach leads to an Ito-type stochastic evolution equation with a multiplicative noise term. We show that a consistent mean-field (i.e., deterministic) reduction of this problem contains an unexpected low-density cutoff induced by the net probability drift due to the aforementioned noise. At the conclusion, implications of these findings for a first-principles theory of diffusion-limited-aggregation fractals are discussed.

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The formation of spatial patterns via diffusion-limited growth continues to be a topic of great interest [1]. These nonequilibrium systems form structures via the evolution of a phase boundary under the influence of transport effects and microscopic attachment kinetics. The types of patterns which form seem to be independent of many of the details of any specific experimental realization of this type of process. Because of this, similar structures have been observed in crystal growth [2], viscous fingering [3], electrochemical deposition [4], and most recently bacterial colony growth [5].

One of the most popular models for studying these growth processes has been the diffusion-limited-aggregation (DLA) simulation of Witten and Sander [6]. Their original algorithm involving one walker which always sticks to a growing cluster has been generalized to include finite walker density, nontrivial sticking probabilities, etc. [7]. The most important aspect of this work has long been the ease with which this approach can produce disordered, fractal structures which are remarkably realistic. Despite many attempts, however, there is still lacking a first-principles theory which can explain DLA results regarding fractal scaling [8].

The purpose of this paper is to take a step in this direction. Specifically, we will reformulate (finite walker density) DLA as a stochastic evolution equation, extending the long-ignored work of Peliti [9], Shapir [10], and others [11] on field-theoretic approaches to DLA. We will then show that a proper continuum limit of this equation leads to a new mean-field theory. Crucial to this result is the realization that multiplicative noise can give rise to unexpected stabilization effects, when the underlying discrete time process leads to the Ito interpretation of a stochastic equation. This concept will emerge clearly in what follows.

Before proceeding, we would like to discuss the connection of this work to recent ideas regarding modified mean-field theory for DLA. As shown in several recent papers, experiments which measure the DLA probability distribution as a function of imposed geometry, walker density, anistropy, etc., cannot be explained by using the familiar Witten-Sander mean-field equation [6]

$$\begin{aligned} \dot{\rho} &= u(\rho + a^2\nabla^2\rho), \\ \dot{u} &= \nabla^2 u - \dot{\rho}. \end{aligned} \tag{1}$$

Instead, a phenomenological modification was proposed [12] which artificially introduced a reduction in the cluster growth rate (by replacing the cluster density $\rho$ by $\rho^\gamma$, $\gamma > 1$ in the $\rho$ evolution equation). This model then agreed rather well with the aforementioned findings [12–14]. As we shall see, one can now *derive* a mean-field theory with a true reduction in the small $\rho$ growth rate; it takes a slightly different form than was postulated in those papers, but will have the same desired effect and hence will agree with all experimental findings to date. We will return to a discussion of this at the conclusion of this paper.

First, we review the field-theoretic approach to DLA. The basic idea of Peliti [9] and others [11] is to associate an occupation number Fock space with the state of the aggregation process at some (discrete) time $t$. If the probability of the system having a given configuration of walkers $\{n_\mathbf{x}\}$ and cluster particles $\{m_\mathbf{x}\}$ ($\mathbf{x}$ is a lattice site) is given as $p(\{n\},\ \{m\})$, the associated state is simply

$$|\Psi_t\rangle = \sum_{\{n\}\{m\}} p(\{n\},\ \{m\}) \prod_\mathbf{x} |n_\mathbf{x}; m_\mathbf{x}\rangle. \tag{2}$$

Then, the evolution of the system via a given Markov process can be used to define a Liouvillian operator.

$$e^{\mathcal{L}\ \Delta t}|\Psi_t\rangle = |\Psi_{t+\Delta t}\rangle.$$

The only difference between this and a more familiar quantum evolution problem is that one does not take squares of amplitudes to get probabilities; these are given merely by overlaps of the system state with the "bare" state $\prod_\mathbf{x}\langle n_\mathbf{x}; m_\mathbf{x}|$.

For the specific case of "transparent" DLA, the probability changes by two dynamical processes. First, walkers can move to a neighboring site. This changes the state by removing a walker from site $\mathbf{x}$ and adding one to site $\mathbf{x}+\boldsymbol{\alpha}$ when $\boldsymbol{\alpha}$ is one of $(\pm a\hat{\mathbf{x}},\ \pm a\hat{\mathbf{y}})$. This process causes an increase in probability at time $t+\Delta t$ at some site if there is a walker at a neighboring site at time $t$ and a decrease in probability at time $t+\Delta t$ of any time $t$ walker-occupied site, due to diffusion away from the site. It is clear that this can be implemented by

$$\mathcal{L}_{\rm walk} = D \sum_{\mathbf{x},\boldsymbol{\alpha}} \left(\pi_{\mathbf{x}+\boldsymbol{\alpha},n} - \pi_{\mathbf{x},n}\right) a_{\mathbf{x},n} \tag{3}$$

where $[\pi_{\mathbf{x},n},\ a_{\mathbf{x}',n}] = \delta_{\mathbf{x}\,\mathbf{x}'}$ are the usual commutation relations for creation and destruction operators $\pi, a$.

The other process in transparent DLA is the conversion of a walker to a cluster particle if it has any nearest neighbors which already belong to the cluster (i.e., have $m \neq 0$). Again, there are both positive and negative contributions to the probability evolution. This process can be described by

$$\mathcal{L}_{\rm stick} = \mu \sum_{\mathbf{x},\boldsymbol{\alpha}} \left(\pi_{\mathbf{x},m} - \pi_{\mathbf{x},n}\right) a_{x,n}\, \pi_{\mathbf{x}+\boldsymbol{\alpha},m}\, a_{\mathbf{x}+\boldsymbol{\alpha},m}, \tag{4}$$

where we have also introduced operators which create and destroy cluster particles. The sum of these two pieces constitutes the entire Liouvillian. In more recent work [15], Sandow and Trimper have added additional factors to enforce the nontransparent one particle per site growth of standard DLA; we will operate under the assumption that the university class is insensitive to this change in the microscopic dynamics.

Given this operator formulation, it is straightforward to derive a path integral for the time evolution of the system. The final result derived by Peliti [9] is that the evolution of the system from time 0 to time $t$ is given by

$$\int D\rho D\hat{\rho} D\phi D\hat{\phi}\ \exp - \sum_t \left[ \sum_{\mathbf{x}} \left( i\hat{\phi}_{\mathbf{x}}\dot{\phi}_{\mathbf{x}} + i\hat{\rho}_{\mathbf{x}}\dot{\rho}_{\mathbf{x}} - iD\,\hat{\phi}_{\mathbf{x}}\,\boldsymbol{\nabla}^2\,\phi_{\mathbf{x}}\right) - i\mu \sum_{\mathbf{x},\boldsymbol{\alpha}} \left(\hat{\rho}_{\mathbf{x}} - \hat{\phi}_{\mathbf{x}}\right)\phi_{\mathbf{x}}\left(1 + i\hat{\rho}_{\mathbf{x}+\boldsymbol{\alpha}}\right)\rho_{\mathbf{x}+\boldsymbol{\alpha}} \right]\Delta t. \tag{5}$$

Here $\boldsymbol{\nabla}^2\phi_x$ is the lattice Laplacian $\sum_{\alpha}(\phi_{\mathbf{x}+\boldsymbol{\alpha}} - \phi_{\mathbf{x}})$ and $\dot{\phi} = (\phi_{t+\Delta t} - \phi_t)/\Delta t$. $\phi$ and $\rho$ have the interpretation of walker and cluster densities, respectively.

We now wish to rewrite this as a set of coupled stochastic partial differential equations. This was first suggested by Shapir [10], but our calculation differs in some important technical details. To do this, we express

$$\exp\left[-\mu(\hat{\rho}_{\mathbf{x}} - \hat{\phi}_{\mathbf{x}})\phi_{\mathbf{x}}\,\rho_{\mathbf{x}+\boldsymbol{\alpha}}\,\hat{\rho}_{\mathbf{x}+\boldsymbol{\alpha}}\,\Delta t\right] = \int D\eta\, D\eta^+ \exp\left[-\eta\eta^+ + i(\hat{\rho}_{\mathbf{x}} - \hat{\phi}_{\mathbf{x}})\eta\,\sqrt{\mu\phi_{\mathbf{x}}\rho_{\mathbf{x}+\boldsymbol{\alpha}}\Delta t}\right.$$
$$\left. + i\eta^+\hat{\rho}_{\mathbf{x}+\boldsymbol{\alpha}}\,\sqrt{\mu\phi_{\mathbf{x}}\rho_{\mathbf{x}+\boldsymbol{\alpha}}\,\Delta t}\right]. \tag{6}$$

To prove this, we notice that the integral over $\eta$ sets

$$\eta^+ = i(\hat{\rho}_{\mathbf{x}} - \hat{\phi}_{\mathbf{x}})\,\sqrt{\mu\phi_{\mathbf{x}}\,\rho_{\mathbf{x}+\boldsymbol{\alpha}}\Delta t},$$

which converts the last term in the exponential to that of the left-hand side.

We now imagine introducing an independent pair of noises $\eta, \eta^+$ for each directed link labeled by $\mathbf{x}, \boldsymbol{\alpha}$. Doing this allows us to obtain a path integral which is linear in both $\hat{\rho}$, $\hat{\phi}$; hence the functional integration over these objects reduces to a $\delta$ function on the $\phi$ and $\rho$ variables. The evolution is thus equivalent to the classical equations

$$\begin{aligned}\dot{\phi}_{\mathbf{x}} &= D\boldsymbol{\nabla}^2\phi_{\mathbf{x}} - \mu\left(\sum_{\boldsymbol{\alpha}}\rho_{\mathbf{x}+\boldsymbol{\alpha}}\right)\phi_{\mathbf{x}} - \sum_{\boldsymbol{\alpha}}\tilde{\eta}_{\mathbf{x},\boldsymbol{\alpha}}\,\sqrt{\mu\phi_{\mathbf{x}}\,\rho_{\mathbf{x}+\boldsymbol{\alpha}}},\\ \dot{\rho}_{\mathbf{x}} &= \mu\sum_{\boldsymbol{\alpha}}\rho_{\mathbf{x}+\boldsymbol{\alpha}}\,\phi_{\mathbf{x}} + \sum_{\boldsymbol{\alpha}}\tilde{\eta}_{\mathbf{x},\boldsymbol{\alpha}}\,\sqrt{\mu\phi_{\mathbf{x}}\,\rho_{\mathbf{x}+\boldsymbol{\alpha}}}\\ &\quad + \sum_{\boldsymbol{\alpha}}\tilde{\eta}^+_{\mathbf{x}+\boldsymbol{\alpha},-\boldsymbol{\alpha}}\,\sqrt{\mu\phi_{\mathbf{x}+\boldsymbol{\alpha}}\,\rho_{\mathbf{x}}},\end{aligned} \tag{7}$$

where we have now defined a noise field $\tilde{\eta} \equiv \eta/\sqrt{\Delta t}$, which will become white in the continuous time limit. As $\mathbf{x} \to \infty$, we must impose $\rho = 0$ and $\phi \to \Delta$, the background walker density.

The most important fact about this set of equations is that the multiplicative noise is defined via the Ito prescription [16] of evaluating the multiplicative factor $\sqrt{\mu\phi_{\mathbf{x}}\rho_{\mathbf{x}+\boldsymbol{\alpha}}}$ at time $t$ where $\dot{\rho}_x \equiv (\rho_{x,t+\Delta t} - \rho_{x,t})/\Delta t$. We now want to take the continuum limit (in time). It is well known in the literature on stochastic equations [17] that the Ito prescription differs from the more symmetric Stratonovich approach by an additional contribution to the deterministic piece of the above equation. The easiest way to derive this extra piece is to return to the path integral and recognize that it is equivalent to the Ito form Fokker-Planck equation for the probability functional $P[\phi,\rho]$

$$\begin{aligned}\frac{\partial P[\phi,\rho]}{\partial t} &= -\sum_{\mathbf{x}}\left(\frac{\partial}{\partial\rho_{\mathbf{x}}} - \frac{\partial}{\partial\phi_{\mathbf{x}}}\right)\left[\mu\sum_{\boldsymbol{\alpha}}\rho_{\mathbf{x}+\boldsymbol{\alpha}}\,\phi_{\mathbf{x}}\,P[\phi,\rho]\right] - \sum_{\mathbf{x}}\frac{\partial}{\partial\phi_{\mathbf{x}}}(D\boldsymbol{\nabla}^2\phi P[\phi,\rho])\\ &\quad + \sum_{\mathbf{x},\boldsymbol{\alpha}}\left(\frac{\partial}{\partial\rho_{\mathbf{x}}} - \frac{\partial}{\partial\phi_{\mathbf{x}}}\right)\frac{\partial}{\partial\rho_{\mathbf{x}+\boldsymbol{\alpha}}}(\mu\,\rho_{\mathbf{x}+\boldsymbol{\alpha}}\,\phi_{\mathbf{x}}\,P[\phi,\rho]).\end{aligned} \tag{8}$$

Going exactly to the Stratonovich version is difficult because one must find the square root of the (symmetrized form of the) second derivative operator; if, however, we approximate $\rho_{\mathbf{x}+\boldsymbol{\alpha}} \sim \rho_{\mathbf{x}}$ in this diffusive term, we obtain the Stratonovich form equation [18]

$$\frac{\partial P}{\partial t} = -\sum_{\mathbf{x}} \left(\frac{\partial}{\partial \rho_{\mathbf{x}}} - \frac{\partial}{\partial \phi_{\mathbf{x}}}\right) [U_{\text{eff},\mathbf{x}} P] - \sum_{\mathbf{x}} \frac{\partial}{\partial \phi_{\mathbf{x}}} [D\nabla^2 \phi P] + 2d \sum_{\mathbf{x}} \left(\frac{\partial}{\partial \rho_{\mathbf{x}}} - \frac{\partial}{\partial \phi_{\mathbf{x}}}\right) \sqrt{\mu \rho_{\mathbf{x}} \phi_{\mathbf{x}}}\ \frac{\partial}{\partial \rho_{\mathbf{x}}} \sqrt{\mu \rho_{\mathbf{x}}\, \phi_{\mathbf{x}}}\, P, \quad (9)$$

where the drift term now takes the form

$$U_{\text{eff},\mathbf{x}} = \mu\, \phi_{\mathbf{x}} \sum_{\boldsymbol{\alpha}} [\rho_{\mathbf{x}+\boldsymbol{\alpha}} - 1/2\theta(\rho)]. \quad (10)$$

Note that the $\theta$ function arose because whenever $\rho_{\mathbf{x}+\boldsymbol{\alpha}} \simeq \rho_{\mathbf{x}} = 0$ there is no quadratic (in $\hat{\rho}$) term in the path integral and hence no fluctuation induced contribution to the deterministic part of the equation of motion.

To complete our derivation, we rescale $\rho$ and $\phi$ by $D/\mu$ and rescale time by $1/D$. This leads to the final (Stratonovich form) stochastic differential equation

$$\dot{\rho}_{\mathbf{x}} = \phi_{\mathbf{x}} \sum_{\boldsymbol{\alpha}} \left(\rho_{\mathbf{x}+\boldsymbol{\alpha}} - \frac{\mu}{2D}\,\theta(\rho_{\mathbf{x}})\right) + \mathcal{N}\sqrt{\frac{\mu}{D}},$$
$$\dot{\phi}_{\mathbf{x}} = \nabla^2 \phi_{\mathbf{x}} - \dot{\rho}_{\mathbf{x}} + \mathcal{N}\sqrt{\frac{\mu}{D}}, \quad (11)$$

where $\mathcal{N}$ represents noise terms. Our mean-field theory then amounts to simply dropping the noise terms. It only makes sense to do this in the Stratonovich form because here the noise represents *symmetric* diffusion around the deterministic path. This has been extensively discussed [19] in the context of the stabilization effect of multiplicative noise in cases where it multiplies the field; this occurs, for example, in the Landau equation with a fluctuating control parameter [20]. In these systems, the Ito prescription gives rise to a shifted threshhold which is due precisely to the extra term which comes about from going to the Stratonovich form. What is different here is that the noise is proportional to the square root of the field and hence gives rise to a constant (field independent) shift, as long as $\rho \neq 0$.

We thus see explicitly that the microscopic theory gives rise to a minimum growth density proportional to $\mu/D$. Physically, this ratio represents how fast the cluster changes as compared to how fast the diffusion field can react. Because of this low-density cutoff, we expect that this model will exhibit the same qualitative behavior as the $\gamma > 1$ phenemenological model alluded to earlier. Specifically, three connected facts should emerge.

(a) If we take the limit $\Delta \to 0$ and keep the flux fixed (formally this is just arrived at by eliminating the $\dot{\phi}_x$ term and replacing $\phi \to \Delta$ at $\infty$ by $\phi \to Jx$), this model exhibits a stable steady-state solution with some value $\rho(x \to -\infty) \equiv \rho_0$ which depends on $\mu/D$ and vanishes as $\mu/D \to 0$.

(b) At finite $\Delta$, the steady-state solution vanishes at some value $\Delta^*$ which is of order of $\rho_0$. This is actually a direct consequence of the last point [21].

(c) There is a Mullins-Sekerka instability [22] at some $\Delta^{**} > \Delta^*$ which is also at order $\rho_0$.

We have checked items (a) and (b) by regularizing the $\theta(\rho)$ function (as an arctangent) and numerically integrating the equations of motion. A discussion of why these facts are necessary in order to explain results to date on walker probability distributions and on morphology transitions has been presented elsewhere [12–14].

The fact that $\mu/D$ gives rise to a lower limit on where the mean-field theory can maintain stability is not surprising, in light of the fact that lowering $\Delta$ means that correlating the motion of the interface on longer and longer length scales requires that the cluster field change more and more slowly as compared to the walker probability distribution — in fact, in the original DLA model, each infinitesimal change in the cluster gives rise to a completely instantaneously updated walker field, formally corresponding to $D \to \infty$.

It is interesting to speculate on the prospects for going beyond mean-field theory so as to include fluctuation effects. The simplest extension would involve some type of random-phase approximation, which would lead to a coupled set of equations for the mean fields and two-point correlations for the field fluctuations. This idea will be pursued elsewhere.

So, where does this leave us in our quest for a theory of DLA? Our perspective is as follows. We can now justify all the qualitative results (which agreed with direct experimental findings) which emerged from the mean-field theory approach in which a density cutoff was introduced in a phenemenological manner. Next, true DLA simulations clearly produce a stable interface on a large enough length scale for any value of $\Delta > 0$; this is just a restatement of the fact that there is always an upper length to fractal scaling at finite walker density. Our results indicate that a field-theoretic approach starting from a mean-field description and adding fluctuations in afterward will only be valid if we require $\Delta \gg \mu/D$; recovering fractal scaling, say in the Uwaha-Saito form [23]

$$v \sim \Delta^{1/d-d_f} \text{ as } \Delta \to 0$$

for the interface velocity $v$, will clearly require taking the limit $\mu/D \to 0$. Exactly how to take into account fluctuations so as to alter the mean-field prediction $v \sim \Delta$ (for $1 \gg \Delta \gg \mu/D$) or equivalently $d_f = d - 1$ is still a significant challenge.

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