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Convex to concave transition and invariant distribution of segment lengths in many-walker anisotropic diffusion-limited aggregation

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We present numerical studies of an on-lattice many-walker diffusion-limited aggregation model. For asymptotic late stage growth, the ensemble averaged envelope exhibits a convex to concave transition. This transition resembles morphology transitions in other diffusion-limited systems but we do not detect a change in the functional form of the growth velocity. We also find that the distribution of the segment lengths is invariant under changes of the supersaturation.

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In the past few years, studies of diffusive patterning [1–3] have focused on morphology selection and morphological properties [3–10]. It has been proposed that all observed patterns can be grouped into a small set of "essential shapes" or *morphologies* [4], each having its own characteristic geometric features. Motivated by experimental observations in the Hele-Shaw cell, Ben-Jacob *et al.* [4] proposed the existence of a morphology determination principle: In the presence of anisotropy, both tip-splitting and dendritic solutions exist, but the fastest growing solution is the dynamically selected one. In general, if more than one morphology is a possible solution, only the fastest growing one is globally stable and hence will be observed. The existence of a morphology selection principle would imply the existence of a morphology diagram (analogous to phase diagrams in equilibrium).

In processes like solidification [11], electrochemical deposition [12,13], growth of bacterial colonies [14,15], and others, different shapes are observed for different values of the control parameter (e.g., undercooling, supersaturation, etc.). The observation of various shapes is a necessary, but not a sufficient condition, for the existence of morphology selection. The change in shape could be a crossover rather than a transition. That is, for each value of the driving force a unique solution (shape) exists. The space of control parameters could still be divided according to some classification of the observed patterns. However, in this case the boundaries between different regimes would be fuzzy, and with no sharp transitions as the boundaries are crossed (by varying the growth parameters). The second possibility is that more than one morphology is a possible solution, but only one solution is dynamically selected [3]. In that case, sharp transitions (morphology transitions) are predicted upon crossing the boundaries. Note that by "sharp transitions" we consider simultaneous change of many of the pattern's properties such as growth velocity, branch width, correlation functions, etc.

The noise, inherent in experimental systems far from equilibrium, makes the distinction between a sharp crossover and a smeared transition unpractical. At present, morphology diagrams have been constructed for a variety of experimental systems [12,13,16–19]. These findings, while supportive, are yet not a clear answer regarding the questions of transition versus crossover. Additional support for a morphology transition is the demonstration of coexistence of morphologies [6]. As a step to distinguish between a transition and a crossover, it was proposed [4] to correlate the change in shape with changes in dynamical response functions, for example the (average) growth velocity. Indeed, both discontinuities, and changes in the slope of the velocity (as a function of the driving force) were observed in experimental systems upon crossing the boundaries between morphologies [3,4,12,13,20]. The above principle was also used as a base for a theoretical approach of morphology transitions [8].

Recently, a correlation between the patterns and a geometrical characterization was found using the diffusion-transition model [5] inspired by solidification from supersaturated solution. An envelope of the pattern was defined and found to be shape preserving and to propagate at constant velocity. A change in the shape of the growing pattern which was first detected by "artistic" view was characterized as the *simultaneous occurrence* of (1) a change in the scaling of the growth velocity, and (2) a change of the envelope shape from convex to concave. This convex to concave transition was found also in experiments of liquid crystals [19], for a mean-field model of many-walker diffusion-limited aggregation (DLA) [7], for the phase-field model [10], and for experiments and simulations of growth of bacterial colonies [21].

Since its construction in 1981, the Witten and Sander DLA model [22] started a new active field of research [23]. Its simplicity in formulation and nontrivial asymptotic behavior made it one of the most studied and canonical models of nonequilibrium processes. The high level of noise and in addition a clear asymptotic behavior made it an example of the "victory" of the dynamics over the noise. Can we find morphology transitions in a simple derivative of the DLA model? For such a study, we need the possibility to vary a thermodynamic force and to observe the response of the system to such changes. As the thermodynamic force in DLA (the difference between the chemical potential of the cluster and of the walkers) is infinity (since there is no equilibrium concentration of walkers [24]), we choose to work on many-walker DLA [25–27]. Here we vary the supersaturation, and study the response

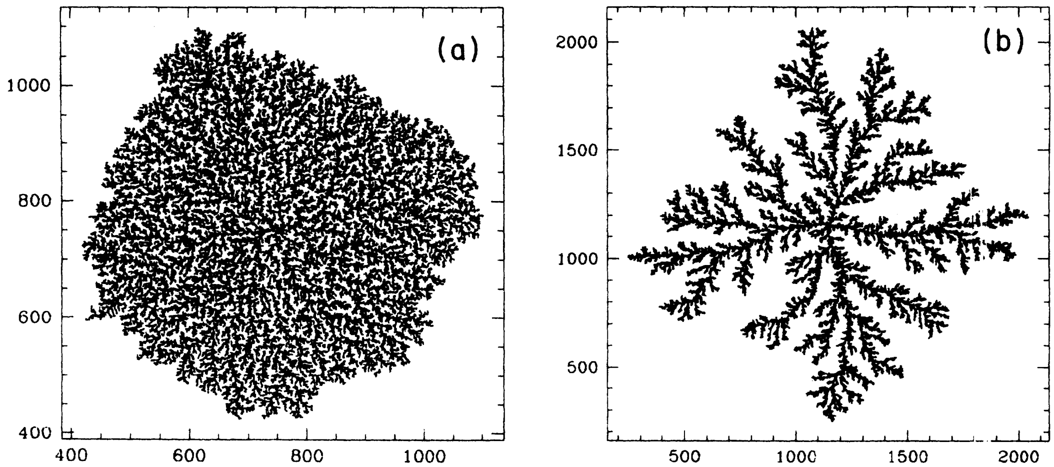

FIG. 1. Two realizations of the many-walker DLA cluster. (a) $\Delta = 0.35$ and the number of particles is 108 544. (b) $\Delta = 0.05$ and the number of particles is 174 331.

of the growth pattern and specifically the envelope and the growth velocity.

Several models for many-walker DLA were studied [25–27], mainly for channel geometry. We use here an extension of the original Witten and Sander [22] DLA model, for many walkers which move simultaneously. The "atoms" are inside a squared box, each performing a random walk on a square lattice until it lands on a site next to the aggregate and is added to it. In order to set the diffusion constant to be unity, the lattice constant is set to unity and, during each cycle, each atom moves four times. Initially the aggregate contains one atom in the middle of the box, and random walkers are spread randomly inside the box with uniform density Δ. The box size is chosen to be larger than the decay length of the diffusion field. [As explained by Uwaha and Saito [25], the asymptotic growth velocity is related to a characteristic correlation length (the diffusion length) $\xi \equiv 1/v$.] Note that two or more atoms may have the same coordinates, however, the results do not change qualitatively if the atoms are treated as avoiding walkers.

In Fig. 1 we show two clusters which were grown for two different levels of supersaturation. For high supersaturation [Fig. 1(a)], the macroscopic pattern is similar to a compact Eden-like [28] structure with additional details on the mesoscale (branches level) organization. For lower supersaturation [Fig. 1(b)], the structure is less dense and the fourfold symmetry is more pronounced. Note that a concave shape of anisotropic DLA (i.e., in the limit $\Delta \to 0$) is familiar from the work of Meakin *et al.* [29].

Next we look for a geometrical characterization of the growing patterns using their envelopes. To do so we construct the ensemble average field [5,30] by projecting many clusters (which were grown using the same parameters but different seeds for the random number generator). The contour lines of the ensemble average field define the envelope. The dependence of the envelope shape on supersaturation is presented in Fig. 2. For high supersaturation the envelope is almost circular. For lower supersaturation the envelope tends to a square shape, and as it decreases further there is a transition to a concave pattern. There is no general explanation for the concave to convex transition. There is an explanation for the occurrence of convex patterns at high supersaturation and concave patterns at low supersaturation for infinitely noise-reduced DLA [31,32]. In this case, at high supersaturation ($\Delta = 1$) the model is equivalent to the Eden model where the growth patterns are all convex [33]. For a low level of supersaturation there exists an analytical solution which is concave [34]. For a quantitative geometric measure (whether the pattern is convex or concave) one can use the dynamical density $\rho(t) \equiv \frac{M(r)}{2R^2_{max}(t)\Delta}$ (where R_{max} is the distance from the center of the cluster to the farthest point which belongs to the cluster). Since the growing cluster advances at constant velocity (as shown below), the cluster density behind the growing front should be exactly equal

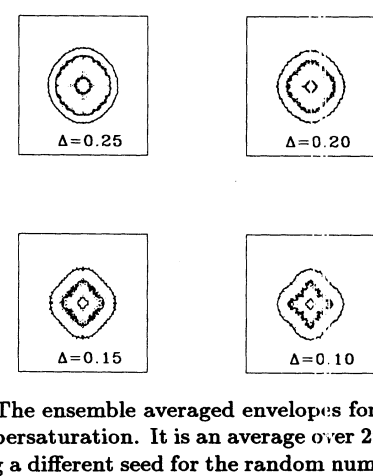

FIG. 2. The ensemble averaged envelopes for four different levels of supersaturation. It is an average over 200 realizations each having a different seed for the random number generator. We use the fact that the growing patterns symmetry reflects the lattice anisotropy. Therefore we can average over the fourfold symmetry and reveal a smooth envelope [6]. The envelope shape changes from convex for $\Delta = 0.25$ to concave at $\Delta = 0.1$. Three contour lines (0.1,0.2,0.3) are plotted for each Δ.

to Δ [25]. Therefore, for late stage asymptotic growth, $\rho(t) \geq 1$ for convex patterns and $\rho(t) < 1$ for concave patterns [10]. This quantity, while an important quantitative measure, converges very slowly and requires large scale simulations. It was checked in our system and found to approach the right values.

As stated above, a correlation between the morphology and the rate of propagation of the envelope was observed in many systems. As the envelope shape is maintained (and the growth velocity is not the same in the different directions) we use the maximal growth velocity $v(t) \equiv \frac{dR_{max}(t)}{dt}$ as the envelope velocity. The asymptotic growth velocity, $\bar{v}$, is presented in Fig. 3 on a log-log scale. The velocity scales with the supersaturation as $\bar{v} \propto \Delta^{2.88}$. There is no detectable change of this scaling law as the envelope changes from convex to concave (around $\Delta = 0.15$) in agreement with the results of Ref. [7].

An additional characteristic of morphology transitions is the change of the mesoscale ordering. A simple measure for such ordering is the distribution of the segment lengths in the growing cluster. Here, we consider a segment to be a sequence of consecutive particles along a straight line. Note that each particle which belongs to the cluster is measured twice as being part of a segment in the x direction and in the y direction. In Fig. 4 we plot a histogram of the number of segments of length L (which we denote as n_L). That is, we define the frequency of segments length as

$$g(L) \equiv \frac{n_L}{\sum_i n_i}. \tag{1}$$

Each histogram is normalized by the total number of segments in the cluster. Surprisingly, the histograms coincide for all levels of supersaturation Δ (note that the data become noisy for large L as the frequency of such an event is very small and numerical fluctuations become

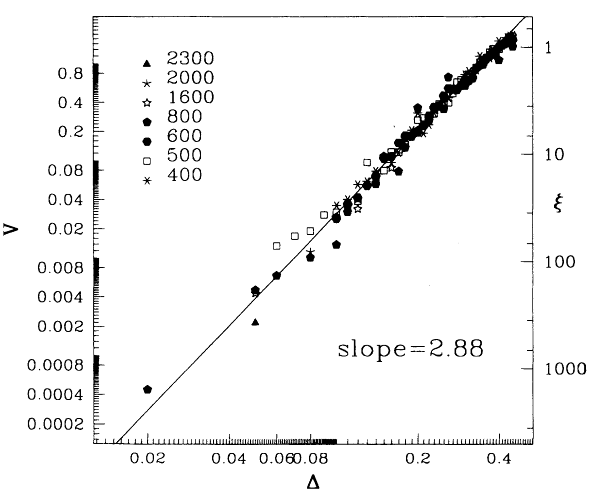

FIG. 3. The asymptotic growth velocity $\bar{v}$ and diffusion length $\xi \equiv 1/v$ (shown on the right axis), versus supersaturation on a log-log scale. No traceable change in the scaling of the velocity is observed as the pattern is changed from convex (above $\Delta \sim 0.15$) to concave (below $\Delta \sim 0.15$). The point style marks the system size.

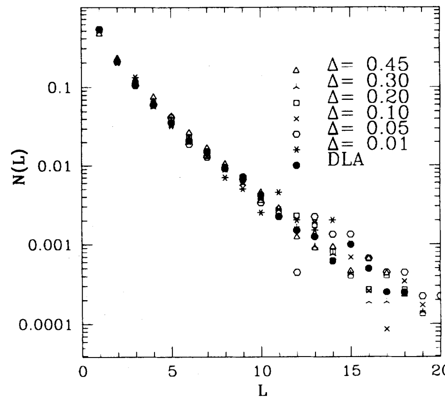

FIG. 4. Distribution of segments length rescaled by the number of particles in the cluster on a semilogarithmic plot. The distribution function is independent of the supersaturation. The DLA distribution function is also the same as for finite supersaturation.

important). To show the validity of this behavior for infinitesimally small Δ, we performed a simulation of on-lattice DLA (where each particle is released sequentially at infinity). The histogram of this case also coincides with all the above histograms of finite Δ. This aspect of the mesoscale organization is invariant under changes of Δ, and is a basic feature of the growth process. Note that for $0.2 \leq \Delta \leq 0.45$ the noiseless part of the distribution ($L \leq 10$) includes segments of length larger than the diffusion length (shown on the right hand side of Fig. 3). Moreover, preliminary observation shows that the segment lengths distribution for a noise-reduced many-walker DLA process [35] and for many-walker DLA on a triangular lattice is also independent of the supersaturation. However, the functional form of the distribution of segment lengths in those cases differs from our $g(L)$.

A simple and important consequence of the above property is that the mass of the cluster can be expressed as

$$M = \frac{1}{2}\sum_i n_i i = \frac{1}{2}\sum_i \frac{g(i)}{g(k)} n_k i \tag{2}$$

for any value of k. We define $\mathcal{G}_k \equiv \frac{\sum_i g(i) i}{2g(k)}$ and find that, for each value of k,

$$n_k = \frac{M}{\mathcal{G}_k}, \tag{3}$$

where $\mathcal{G}_k$ is a known quantity and is independent of Δ. That is, one can relate the total mass to the number of segments of each length and vice versa. This property is also correct for parts of the cluster. Thus it seems that this invariant property can be used as a hint for an alternative mean-field description of the many-walker DLA [36].

How can we understand the above observations and the difference between them and the diffusion-transition model? One possibility is that the change from convex to concave envelope is indeed a morphology transition similar to the one in the diffusion-transition model. We do not observe a simultaneous discontinuity of the velocity and mesoscale organization. However, as the whole process depends strongly on stochastic noise, the above quantities may be smeared and the transition cannot be traced. Alternatively, the change in the envelope shape may not be a morphology transition. It may be that a change in the velocity scaling can be observed only when one varies the thermodynamic driving force (the chemical potential difference) between the two phases. As the chemical potential difference in this system is infinity (irrespective of Δ), we cannot find in this system a morphology transition as a function of Δ. Moreover, the fact that we observe a convex to concave change of the envelope suggests that this property is not a sign for a morphology transition. It may be a property of diffusive patterning in two dimensions when anisotropy is present which is manifested in the diffusion-transition model as a morphology transition via the selection of tip-splitting or dendritic growth out of the possible solutions. Here there is no morphology transition as there is no competition between two different solutions (similar to tip-splitting growth and dendritic growth in the diffusion-transition model).

To conclude, this work calls for more studies of envelope vs velocity vs mesoscale organization, especially when surface tension and noise reduction are included. Also the segment lengths distribution is an intriguing tool for experimental observation.

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[1] D. A. Kessler, J. Koplik, and H. Levine, Adv. Phys. **37**, 255 (1988).
[2] J. S. Langer, Science **243**, 1150 (1989).
[3] E. Ben-Jacob and P. Garik, Nature (London) **343**, 523 (1990).
[4] E. Ben-Jacob, P. Garik, T. Muller, and D. Grier, Phys. Rev. A **38**, 1370 (1988).
[5] O. Shochet, K. Kassner, E. Ben-Jacob, S. G. Lipson, and H. M. Müller-Krumbhaar, Physica A **181**, 136 (1992); **197**, 87 (1992).
[6] O. Shochet and E. Ben–Jacob, Phys. Rev. E **48**, R4168 (1993).
[7] Y. Tu, H. Levine, and D. Ridgway, Phys. Rev. Lett. **71**, 3838 (1994).
[8] E. Brener, H. M. Müller-Krumbhaar, and D. E. Temkin, Europhys. Lett. **17**, 535 (1992).
[9] T. Ihle and H. Müller-Krumbhaar, Phys. Rev. Lett. **70**, 3083 (1993).
[10] R. Kupferman, O. Shochet, and E. Ben-Jacob, Phys. Rev. E (to be published).
[11] W. Kurz and D. J. Fisher, *Fundamentals of Solidification*, 3rd ed. (Trans Tech, Switzerland, 1989).
[12] Y. Sawada, A. Dougherty, and J. P. Gollub, Phys. Rev. Lett. **56**, 1260 (1986).
[13] D. G. Grier, E. Ben-Jacob, R. Clarke, and L. M. Sander, Phys. Rev. Lett. **56**, 1264 (1986).
[14] E. Ben-Jacob, H. Shmueli, O. Shochet, and A. Tenenbaum, Physica A **187**, 378 (1992).
[15] E. Ben-Jacob, O. Avidan, A. Tenenbaum, and O. Shochet, Physica A **202**, 1 (1994).
[16] E. Ben-Jacob, R. Godbey, N. D. Goldenfeld, J. Koplik, H. Levine, T. Mueller, and L. M. Sander, Phys. Rev. Lett. **55**, 1315 (1985).
[17] V. Horvath, T. Vicsek, and J. Kertesz, Phys. Rev. A **35**, 2353 (1987).
[18] A. Buka and P. Palffy-Muhoray, Phys. Rev. A **36**, 1527 (1987).
[19] P. Oswald, J. Bechhoefer, and F. Melo, Mater. Res. Bull. **16**, 38 (1991).
[20] S. K. Chan, H. H. Reimer, and M. Kahlweit, J. Cryst. Growth **32**, 303 (1976).
[21] E. Ben-Jacob, O. Shochet, A. Tenenbaum, I. Cohen, A. Czirók, and T. Vicsek (unpublished).
[22] T. A. Witten and L. M. Sander, Phys. Rev. Lett. **47**, 1400 (1981).
[23] T. Vicsek, *Fractal Growth Phenomena* (World Scientific, New York, 1989).
[24] Y. Saito and T. Ueta, Phys. Rev. A **40**, 3408 (1989).
[25] M. Uwaha and Y. Saito, Phys. Rev. A **40**, 4716 (1989).
[26] R.F. Voss, J. Stat. Phys. **36**, 861 (1984).
[27] P. Meakin, Physica A **153**, 1 (1988).
[28] M. Eden, in *Proceedings of the 4th Berkeley Symposium on Mathematical Statistics and Probability*, edited by F. Neyman (University of California Press, Berkeley, 1961), Vol. 4, p. 223.
[29] P. Meakin, R. C. Ball, P. Ramanlal, and L. M. Sander, Phys. Rev. A **35**, 5233 (1987).
[30] A. Arneodo, Y. Couder, G. Grasswau, V. Hakim, and M. Rabaud, Phys. Rev. Lett. **63**, 984 (1989).
[31] C. Tang, Phys. Rev. A **31**, 1977 (1985).
[32] J. Kertesz and T. Vicsek, J. Phys. A: Math. Gen. **19**, L257 (1986).
[33] D. E. Wolf, J. Phys. A: Math. Gen. **20**, 1251 (1987).
[34] J.-P. Eckmann, P. Meakin, I. Proccaccia, and R. Zeitak, Phys. Rev. A **39**, 3185 (1989).
[35] D. Kessler (private communication).
[36] O. Shochet, E. Ben-Jacob, and Z. Schuss (unpublished).