Multiple-contact diffusion-limited-aggregation model

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New diffusion-limited growth models are described in which growth occurs at the site which is first contacted by k (k is a small integer) random walkers launched from outside the growing cluster and adsorbed on contact with the cluster. This model is closely related to the dielectric-breakdown model in which the growth probability is proportional to the kth power of the field gradient at the surface of the growing cluster. For the case $k=2$ our results for the radius-of-gyration exponent β are consistent with the theoretical predictions of Turkevich and Sher. However, our results do indicate that the main assumption of the theory of Turkevich and Sher (the formation of a diamond shape on the square lattice) is not correct in this case. The clusters grow into a crosslike shape in which the increase in length and width of the arms of the cross with increasing cluster size are described by different exponents. A similar anisotropy is not observed from the largest clusters which can be grown on hexagonal lattices for the cases $k=2$, 3, and 4. For $k=2$ the radius-of-gyration exponent has effective values of about 0.73 and 0.70 for the largest clusters which can be grown on square and hexagonal lattices, respectively.

INTRODUCTION

Considerable interest was generated in nonequilibrium random growth models following the demonstration by Witten and Sander[1] that a simple diffusion-limited aggregation (DLA) model leads to the formation of structures which seem to possess interesting, nontrivial, scaling and universality properties.[1-3] In this model particles (often represented by occupied lattice sites) are added to a growing cluster or aggregate of particles using random walk trajectories. In recent years a variety of models more or less closely related to the Witten-Sander model have been developed and applied to a broad range of physical phenomena.[4,5] In one of these models the growth probability at each unoccupied surface site adjacent to an occupied site on the growing cluster (i.e., at each growth site) is obtained by solving Laplace's equation ($\nabla^2\phi=0$) subject to absorbing boundary conditions ($\phi=0$) on the growing cluster and a fixed value for the potential ($\phi=1$) at some distant boundary.[6,7] Laplace's equation is solved numerically in a discretized form using standard relaxation methods.[6,7,8] The growth probability at each of the surface sites is then assumed to be proportional to some power (ϵ) of potential (ϕ) or proportional to the same power of the potential gradient ($\nabla\phi$) (since $\phi=0$ on the cluster, these two procedures are essentially equivalent):

$$P\sim\phi^{\epsilon}\,. \tag{1}$$

In other words, the growth probability is proportional to some power of the harmonic measure. This model was first introduced by Niemeyer *et al.*[6] to describe dielectric breakdown phenomena (here ϕ is the electric potential) and was developed independently by Meakin[7] to describe growth of biological systems (here ϕ is the concentration of some growth controlling chemical).

In the case $\epsilon=1$ this dielectric breakdown[6] or diffusion-limited growth[7] model becomes equivalent to the Witten-Sander model for diffusion-limited aggregation (apart from some details which depend on how the models are implemented and are not believed to change the scaling properties of the aggregates). In general, these models lead to fractal-like structures in which the fractal dimensionality (D) (Ref. 9) varies continuously with the exponent ϵ [Eq. (1)]. Because of this characteristic, these models have the potential of being able to describe a broader range of phenomena than the original Witten-Sander model.

Based on some early, rather small scale, simulations,[1-3] it has generally been assumed that these models are universal in the sense that they lead to structures which can be described in terms of fractal dimensionalities or scaling exponents which do not depend on model details. This idea has recently been challenged by Turkevich and Sher[10] who have developed a new theory for DLA based on the idea that the fractal dimensionality is determined by power-law singularities in the distribution of growth probabilities and that the strength of these singularities is determined by the structure of the lattice. Based on these ideas, they predict that $D=\frac{5}{3}$ for a two-dimensional (2D) square lattice and $\frac{7}{4}$ for 2D triangular and hexagonal lattices. Recent large scale computer simulations[11] are not inconsistent with $D=\frac{5}{3}$ for the square lattice but do not support a value of $\frac{7}{4}$ for the triangular and hexagonal lattices. However, in this case the discrepancy between theory and simulation can be rationalized on the basis of the concepts embodied in the theory of Turkevich and Sher.[11]

Turkevich and Sher have also used their theory to make predictions concerning the dielectric breakdown model. For the case $\epsilon=2$ they find $D=\frac{4}{3}$ for a square lattice, and for $\epsilon=0.5$ they find $D=\frac{11}{12}$ (1.833). In their simulations carried out using the dielectric breakdown model,

Niemeyer *et al.*[6] found $D \cong 1.6$ for $\epsilon = 2$ and $D = 1.89 \pm 0.01$ for $\epsilon = \frac{1}{2}$, while Meakin[7] found $D \cong 1.40$ for $\epsilon = 2.0$ and $D \cong 1.90$ for $\epsilon = 0.5$ using an essentially identical model. Unfortunately, large amounts of computer time are required for complete relaxation of the field (ϕ) in these simulations (in particular, a large number of relaxation cycles are needed for large ϵ in order to obtain accurate values for the growth probabilities). In addition, the fixed boundary conditions ($\phi = 1$) are imposed at distances of only 2–3 R_{max} from the growth site or seed (where R_{max} is the maximum radius of the cluster) in current algorithms. Although it might be possible to make substantial improvements to the algorithms used for the dielectric breakdown[6] and diffusion-limited growth[7] models, it does not seem probable that results from large numbers of large clusters will be available in the near future. In this paper some results are presented which have been obtained from a closely related model.

In this new, multiple-contact model, particles are launched one at a time from outside of the region containing the cluster and follow random walk trajectories. These trajectories are terminated if they reach a distance of 100 R_{max} from the cluster or enter an unoccupied site adjacent to an occupied site (a growth site). A record is kept of how may times each of the growth sites is occupied, and the first growth site to be contacted k times is occupied (k is a small integer). After each growth event the whole procedure is repeated with each growth site having an initially zero contact score. This model is not equivalent to the dielectric breakdown model, but in the limit of large cluster sizes (where all of the growth probabilities for the growth sites are small) the growth probabilities for a site with probability P will be proportional to P^k, and we expect the asymptotic scaling behavior for the clusters obtained from this model to be the same as that for the dielectric breakdown model with $\epsilon = k$.

COMPUTER MODELS

All of the simulations used in connection with the work described in this paper were carried out using two-dimensional square or hexagonal lattices. At the beginning of a simulation a site in the center of the lattice is filled to represent the "seed" or growth site. Particles are released, one at a time, in the vicinity of the cluster and allowed to undergo random walks. If the mobile particle undergoing a random walk moves into an unoccupied lattice site which is a nearest neighbor of an occupied site (i.e., a "growth" site), the random walk trajectory is stopped and a new trajectory is started. A record is kept of how many times each of the growth sites has been contacted. The first growth site to be contacted k times is occupied and the procedure described above is repeated. After each growth event the number of contacts for each of the growth sites is set to zero.

At later stages in the simulation, when a cluster of occupied sites has been found, the particle trajectories are started from a random position outside of the region occupied by the cluster[2,3,12] and are terminated if a growth site is reached or if the particle reaches a distance greater than 100 R_{max} from the seed or growth site where R_{max} is the maximum radius of the cluster. In order to reduce computer time requirements, the particles are allowed to take large off-lattice jumps if they are situated a large distance from any occupied site on the cluster.[12] However, if the particles reach a position within 6–7 lattice units from any occupied site on the cluster, they are transferred to the nearest lattice site and the random walk continues on the lattice until the particle either enters a growth site or moves a distance greater than 6–7 lattice units away from the cluster. Thus, in the vicinity of the cluster, these models work like ordinary lattice models for diffusion-limited aggregation.[2,3]

At larger distances from the cluster the length of an off-lattice step is restricted so that the particle cannot penetrate more than one lattice unit into the region in which the on-lattice walk occurs. An illustration of this model, for the case of a square lattice, is given in Fig. 1. The motivation for this somewhat elaborate procedure is to obtain a model which will give results which are indistinguishable from those obtained using completely on-lattice random walks while retaining the efficiency inherent in long off-lattice steps.[12] The procedures used in these models are very similar to those which have been used recently to grow large numbers of quite large ($\sim 10^5$ particles or sites) DLA aggregates.[12] In both the models used in connection with the work described in this paper and the improved models for diffusion-limited aggregation, an underlying lattice is used to inform the random walker how long its next step may be. Since this procedure does not make efficient use of computer storage capabilities, the size of clusters which can be generated is limited. However, that is not an important practical consideration since clusters of the size used in this work required 1–2 hours of CPU time on an IBM 3081 computer to generate.

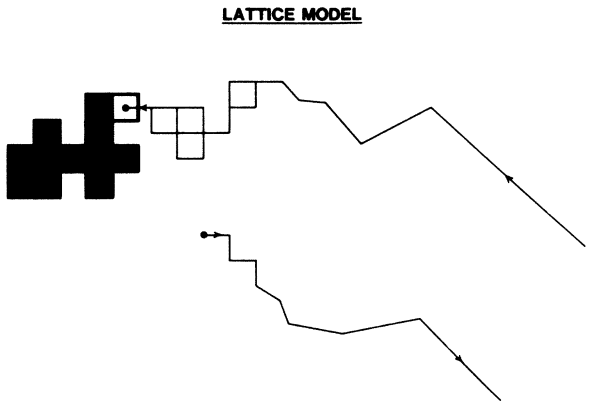

FIG. 1. Early stage in a simulation carried out using a square lattice. The occupied sites comprising the cluster are shaded and a typical particle trajectory is shown. The growth site which is contacted by the particle and at which the particle trajectory terminates is outlined. The on-lattice steps in the vicinity of the cluster and the larger off-lattice steps further from the cluster are shown.

RESULTS

Square lattice models

14 000-site clusters were grown on a square lattice using the two-contact model ($k=2$). Figure 2 shows a superposition of 30 of these clusters. The most remarkable characteristic of this figure is the way in which the density is concentrated along the axes of the lattice to form a "cross" shape. A similar, but much smaller, concentration of density along the axes of the lattice has been observed for DLA clusters ($k=1$) generated on a square lattice.[13,14] Similar results have been obtained using the three- and four-contact models. Figure 3 shows a superposition of forty 5,000-site clusters grown using the four-contact model ($k=4$). In this case the concentration of density along the lattice axes is even more pronounced.

A total of seventy-eight 14 000-site clusters were generated using the two-contact ($k=2$) model. Seventy-eight 7500 site clusters were generated using the $k=3$ model, and forty-five 5,000 site clusters were generated using the $k=4$ model. For all three models the dependence of the effective exponent, β, which describes how the radius of gyration (R_g) increases with cluster mass (M) has been investigated:

$$R_g \sim M^{\beta} . \tag{2}$$

The effective values of β were obtained by least-squares fitting straight lines to the coordinates $(\ln(R_g), \ln(M))$ over various ranges of cluster mass (bins). For "bin" 8, clusters in the size range $M_{\max}/2 \le M \le M_{\max}$ were used, for bin 7, clusters in the size range $M_{\max}/4 \le M \le M_{\max}/2$ were used, and for each successively smaller bin number the cluster masses were smaller by a factor of 2. Figure 4 shows some of the results obtained from all three square lattice models.

For the two-contact model the effective value of β is weakly dependent on the cluster mass (by an amount barely larger than the statistical uncertainties) and the dependence of β on M is not inconsistent with a limiting ($M \to \infty$) value of 0.75 predicted by the theory of Turkevich and Sher. However, it is clear from Fig. 2 that these clusters are not homogeneous fractals and that they do not have a square or diamond shape characteristic of the lattice. For $k>2$ the dependence of β on cluster mass is more pronounced. It is difficult from the results shown in Fig. 4 to even guess what the limiting value for the exponent β might be. However, our results do suggest limiting ($M \to \infty$) values greater than 0.8 for $k=2$ and greater than 0.9 for $k=3$.

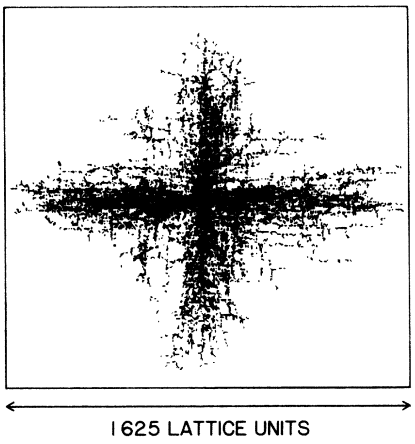

FIG. 3. Superposition of 40 5000-site clusters grown on a square lattice using the four-contact ($k=4$) model. For each cluster every tenth lattice site to be occupied during the growth process is shown.

Hexagonal lattice models

A series of simulations similar to those described above were carried out using hexagonal rather than square lattices. Sixty-seven 15 000-site clusters were generated us-

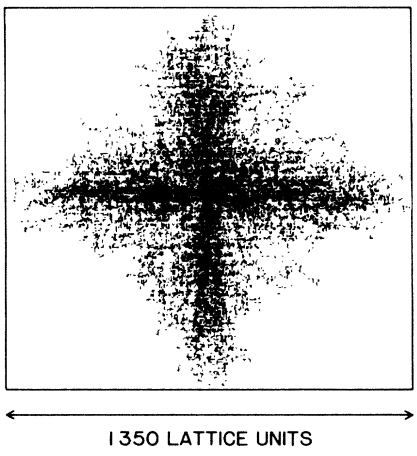

FIG. 2. Superposition of thirty 14 000-site clusters grown on a square lattice using the two-contact ($k=2$) model. For each cluster every tenth particle (occupied lattice site) is shown.

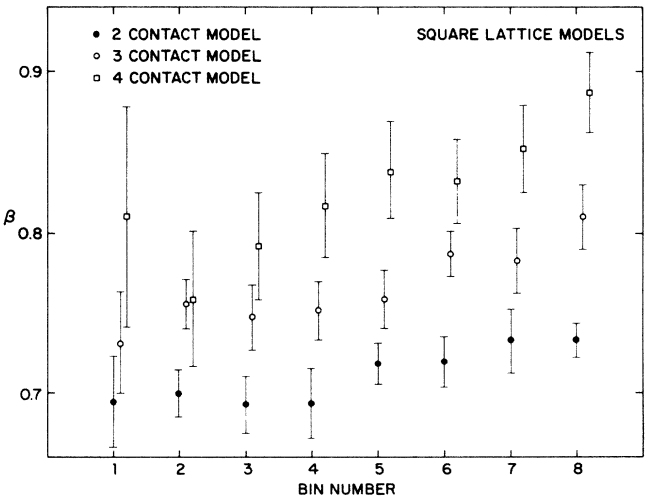

FIG. 4. Dependence of the effective radius-of-gyration exponent (β) on cluster mass (number of occupied lattice sites) for the square lattice model.

ing the two-contact model, seventy-one 7500-site clusters were generated using the three-contact model, and forty-seven 5000-site clusters were generated using the four-contact model. Figure 5 shows a superposition of 10% of the occupied lattice sites from forty simulations carried out using the four-contact hexagonal lattice model. This figure should be compared with Fig. 3 obtained using the corresponding square lattice model. There is no indication in Fig. 5 that the density is concentrated along the axes of the hexagonal lattice or that growth occurs preferentially along these axes. However, it is possible that anisotropies of this type might develop in much larger clusters grown on a hexagonal lattice. Figure 6 shows the dependence of the effective radius of gyration exponent (β) on the cluster size for all three hexagonal lattice models. In the case of the two-contact model β seems to be almost independent of cluster size and has a value of about 0.7 for all cluster sizes. For clusters in the size range 75–15 000 lattice sites a value of 0.700 ± 0.005 is obtained for β assuming that Eq. (2) describes the dependence of R_g on M and that β is mass independent. For the three-contact hexagonal lattice model, β is dependent on cluster mass for small clusters but seems to reach a limiting value of about 0.80 for clusters containing more than about 500 occupied lattice sites. For the four-contact model, β is increasing with increasing cluster mass for the largest clusters we are able to generate. In this case it is difficult to estimate a limiting ($M\rightarrow\infty$) value for β, but our results do suggest a value greater than 0.85.

Anisotropy of clusters generated by the square lattice models

Figures 2 and 3 show a strong anisotropy in the density distribution associated with the square lattice models. This anisotropy is reminiscent of that found by Ball *et al.*[15] for DLA clusters grown on a square lattice with a higher sticking probability for addition of particles in two of the four possible directions on the lattice. Similar results have also been found by Meakin[13] for DLA on a hexagonal lattice with sticking in three of the six possible directions. In the case of the square lattice simulations Ball *et al.* found that compact needle-like objects were formed (in the limit of large cluster mass) for any amount of anisotropy in the sticking probabilities. They showed that the strength in the "easy" direction could be described by $X\sim M^{2/3}$ and growth in the "hard" direction by $X\sim M^{1/3}$. In our case the growth in the easy direction, along the axes of the square lattice, will be described (in the limit $M\rightarrow\infty$) by the relationship

$$X\sim M^{\beta}, \tag{3}$$

where β is the same exponent which describes the growth of the radius of gyration [Eq. (2)]. To characterize the growth in the "hard" direction the distance Y from the closest of the lattice axes has been measured for particles which have been added to the clusters during small intervals in the growth process ($M\pm\delta M/2$). The quantity Y can be regarded as the mean distance from the active zone[16] to the nearest axis of the lattice (with the origin at the growth site). The results shown for the two-contact model are shown in Fig. 7. Our data indicate that in this case

$$Y\sim M^{\gamma}, \tag{4}$$

where the exponent γ has a value of about 0.6. The fact that γ is significantly smaller than β ($\beta>0.7$) indicates that the shape of the clusters will become more and more anisotropic as they get larger and larger (i.e., we have two divergent length scales). In the anisotropic sticking probability model of Ball *et al.* the quantity $\beta+\gamma$ has an asymptotic limit of 1.0 and the clusters are asymptotically compact. In our case $\beta+\gamma$ is considerably larger than 1 ($\beta+\gamma>1.3$). From least-squares-fitting straight lines to

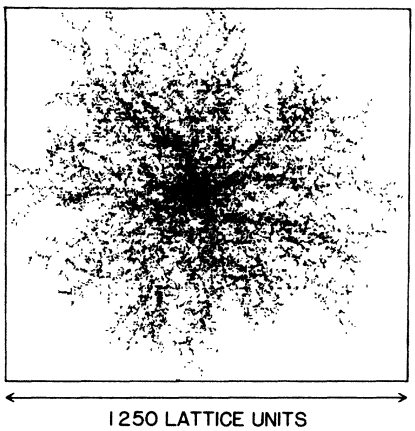

FIG. 5. Superposition of 10% of the sites from 40 clusters generated on hexagonal lattices using the four-contact model. Each cluster contains 7500 sites.

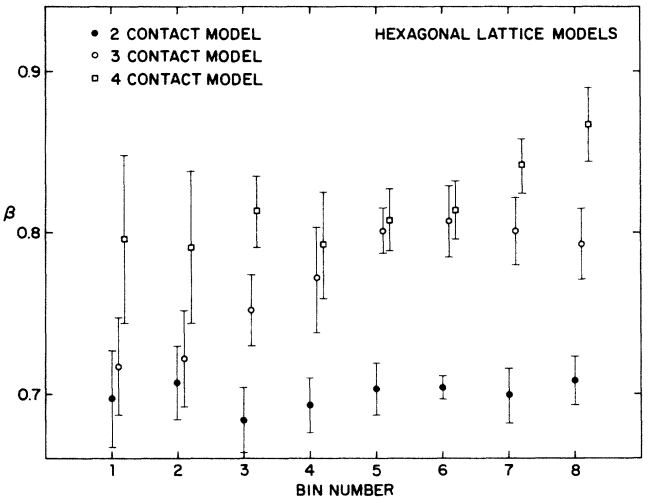

FIG. 6. Effective radius-of-gyration exponents (β) for various ranges of cluster mass obtained assuming a linear dependence of $\ln(R_g)$ on $\ln(M)$. This figure shows the results from all three hexagonal lattice models. Bin 8 corresponds to clusters in the size range $M_{\max}/2\leq M\leq M_{\max}$, bin 7 corresponds to clusters in the size range $M_{\max}/4\leq M\leq M_{\max}/2$ occupied lattice sites, and for each successively smaller bin number the clusters are smaller by a factor of 2.

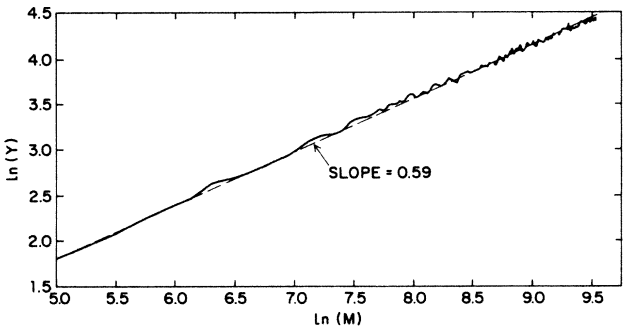

FIG. 7. Dependence of the mean deposition distance (Y) measured from the nearest lattice axis for the two-contact model clusters grown on a square lattice.

the coordinates ($\ln(M)$,$\ln(Y)$) a value of 0.58 ± 0.04 was obtained for the two-contact model. Similar measurements have been made for the three- and four-contact models. In these cases we find $\gamma=0.60\pm0.05$ for $k=3$ and $\gamma=0.63\pm0.09$ for $k=4$ for the square lattice model. Similar measurements have not been made for the one-contact (ordinary DLA) model. However, in this case a value close to 0.6 would almost certainly be obtained for clusters in the size range of a few thousand up to 100 000 occupied lattice sites.[11] Consequently, it seems that $\gamma=0.6$ for the $k=1$, 2, 3, and 4 square lattice models.

Correlation functions

For all six models the two-point density-density correlation functions[1] $C(r)$ have been measured as a function of distance (r). Figure 8 shows results obtained from the square lattice models. Despite the complexity of the structure of these clusters, the dependence of $C(r)$ on r seems to follow a simple power law

$$C(r)\sim r^{-\alpha} \tag{5}$$

over a substantial range of length scales from a few lattice units to more than 50 lattice units for the $k=2$ model and more than 100 lattice units for the $k=3$ and $k=4$ models. By least-squares fitting straight lines to the coordinates ($\ln(r)$, $\ln[C(r)]$) over the range $5\leq r\leq 50$ lattice units the exponent α was estimated to have a value of 0.587 ± 0.005 for the $k=2$ model, 0.724 ± 0.004 for $k=3$, and 0.792 ± 0.003 for $k=4$.

Similar results have been obtained for the hexagonal lattice models (Fig. 9). In this case the dependence of $\ln[C(r)]$ on $\ln(r)$ is almost linear for lengths ranging from a few lattice units to more than 100 lattice units. For distances r in the range $5\leq r\leq 100$ lattice units, the results $\alpha=0.578\pm0.010$ for $k=2$, $\alpha=0.728\pm0.011$ for $k=3$, and $\alpha=0.800\pm0.016$ for $k=4$ were obtained. Within the statistical uncertainties (95% confidence limit ranges) both the square lattice and hexagonal lattice models give the same values for the exponent α for any particular k in the range $k=2$–4.

DISCUSSION

The structure of the clusters grown on a hexagonal lattice using these multiple contact DLA models seems to approximate much more closely that of a self-similar fractal than does the structure of clusters grown on a square lattice. For a self-similar fractal the radius of gyration exponent β and the density-density correlation function exponent α are related by

$$D=1/\beta=d-\alpha \tag{6}$$

or

$$\alpha=d-1/\beta\,. \tag{7}$$

In the case of the hexagonal lattice models these relationships are satisfied quite well at least for $k=2$ and 3. For $k=2$ we found $\alpha=0.578\pm0.010$ and $d-1/\beta=0.571\pm0.01$. For $k=3$,$\alpha=0.728\pm0.011$ and $d-1/\beta\simeq0.75$. For the $k=4$ model it is difficult to estimate a limiting ($M\rightarrow\infty$) value for β. However, our results for α (0.792 ± 0.003) would imply a value of about 0.83 for the radius of gyration exponent. For the largest clusters we can generate, β is larger than 0.83 and seems to be increasing with increasing cluster size.

For the square lattice models our results for the radius of gyration exponent β and the exponent γ, which describes how the mean deposition distance from the closest lattice axis increases with increasing cluster size, seem to have distinctly different values. This result indicates that these structures are not self-similar fractals but may be described as self-affine fractals. This result may be related to the observation that the density-density correlations for ordinary DLA clusters have different dependencies on

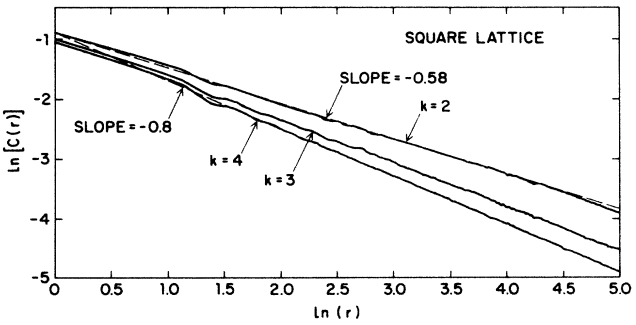

FIG. 8. Two-point density-density correlation functions $[C(r)]$ obtained from the $k=2$, 3, and 4 square lattice models.

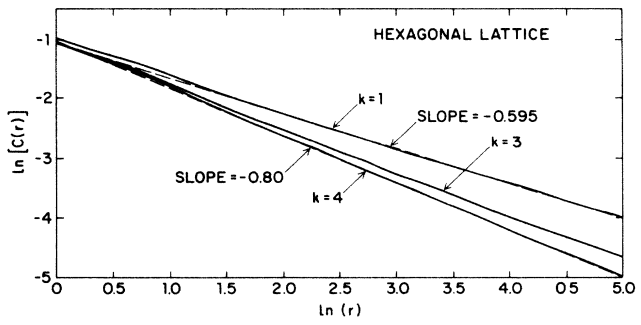

FIG. 9. Two-point density-density correlation functions $[C(r)]$ obtained from the hexagonal lattice models.

distance measured in the radial and tangential directions which can be described by two different effective exponents.[17,18]

The apparent qualitative differences between clusters grown on hexagonal and square lattices may, of course, be more apparent than real. It may be that clusters must be grown to a very much larger size on hexagonal lattices before the much weaker anisotropy associated with these lattices is able to control the form of the clusters. For very small clusters the values of the radius-of-gyration exponent, β, are quite similar for the hexagonal and square lattice models for all three cases ($k=2$, 3, and 4). In fact, this behavior also extends to the $k=1$ (DLA) case, where β is essentially cluster-size independent for the hexagonal lattice models but increases slightly with increasing cluster mass for the square lattice models.[13] For relatively small clusters, β has a value close to 0.585 for both models.

One of the main objectives of this work was to test the theoretical predictions of Turkevich and Sher.[10] While the results obtained for the radius-of-gyration exponents for the $k=2$ case are not inconsistent with this theory, it is clear that one of the main assumptions of the theory (the diamondlike shape of the clusters) is not correct. We also seem to be seeing evidence for the nonuniversal behavior predicted by Turkevich and Sher. However, since there is clear evidence that we have not reached the asymptotic ($M\rightarrow\infty$) limit in our simulations, it is not clear how our results should be interpreted. Clearly, much larger scale simulations would be desirable. Our present algorithms are limited by both computer time and storage requirements. Algorithms of the type described by Ball and Brady[14,19] make much more efficient use of computer storage capabilities, but it is doubtful if very much larger clusters could be grown using these algorithms because of the computer time requirements.

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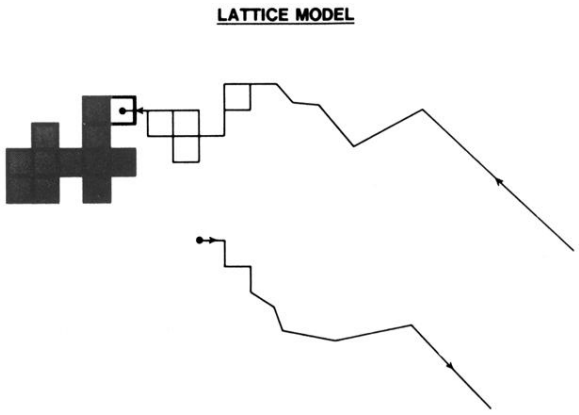

FIG. 1. Early stage in a simulation carried out using a square lattice. The occupied sites comprising the cluster are shaded and a typical particle trajectory is shown. The growth site which is contacted by the particle and at which the particle trajectory terminates is outlined. The on-lattice steps in the vicinity of the cluster and the larger off-lattice steps further from the cluster are shown.