Diffusion-limited aggregation as a growth process statistically invariant under an infinitesimal affine transformation of mass and multiple spatial coordinates

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This work has two major results. First, it proposes a model of diffusion-limited aggregation (DLA) that accounts for the diversity of mass dependence exponents of four characteristic lengths: radius of gyration, major and minor asymmetry axes, and width of active growth zone. Second, it uses the agreement of model predictions with simulation data to show that the invariance of DLA growth probability under an infinitesimal affine transformation of mass and several growth zone spatial coordinates is consistent with the results of simulation data, to a high degree of approximation, in the range of aggregate mass from 200 to 50 000 for the first three of the above characteristic lengths, and in the range 1100–440 000 for the active zone width. [S1063-651X(97)09909-1]

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Diffusion-limited aggregation (DLA), a process by which an aggregate of particles grows by the addition of a succession of randomly walking particles [1], generates growth patterns that are similar to patterns observed as arising from a range of widely different physical processes, such as electrodeposition [2], dielectric breakdown [3], and viscous fingering [4].

Investigators have used a variety of approaches in attempting to understand this process. One category of approach has been to model the process in terms of the mechanism of a field of diffusing particles [5–7]. A second has been to model the process by imposing the simplifying property of scale invariance on a growth process. These include methods of real-space renormalization group and branch competition [8–10]. While these approaches have been successful in explaining a number of features of DLA, important properties of this process are not yet understood.

Of these latter properties, the present paper will be concerned with those related to the fact that different characteristic lengths of the process, such as radius of gyration, major and minor asymmetry axes, and width of the active growth region, appear to depend on the aggregate mass (number of particles) through different exponents [11–14]. A number of these lengths have recently been involved in a discussion of whether DLA can be described by a single scaling exponent or whether it has a more complex scaling [15]. Specifically, I shall propose a model of DLA which attempts to account for the characteristic length mass dependence described by the following.

(1) The aggregates are asymmetric. Quantifying the asymmetry by a moment of inertia analysis leads to a description of the aggregates in terms of a major axis $r_>$ and a minor axis $r_<$ [11].

(2) If the aggregates are described using the characteristic lengths of radius of gyration r_g [12–13], major and minor asymmetry axes $r_>$ and $r_<$ [11], and the width of the active zone of growth w [12–14], then the dependence of each of these quantities on aggregate mass can be described by a power law $l \propto m^{\beta}$, where l represents one of the lengths, and the exponent β represents its mass dependence.

(3) The value of β is different for each of the characteristic lengths. It also appears to vary with mass for the major and minor axes and for the active zone width, but appears to be nearly constant for the radius of gyration. Table I summarizes the values of these exponents, given by numerical simulations.

My approach to writing a model of DLA is suggested by the asymmetry of the aggregates. One can use as a starting point the usual relationship between the radius of gyration r_g and mass m, which is given in terms of fractal dimension D by

$$r_g \propto m^{1/D}. \tag{1}$$

Though DLA is an asymmetric process, it is usually treated as involving only the one spatial (radial) dimension. Investigators have found [16] that the probability $p(m,r)dr$ that a particle will be added at mass m in a interval between radius r and $r+dr$ of the active growth region is, to an order of approximation, invariant under the affine [17] transformation F_0 given by

$$F_0: \ m \rightarrow \lambda m$$
$$r \rightarrow \lambda^{1/D} r, \tag{2}$$

where λ is a real positive number.

The asymmetry of aggregates, however, suggests that they might be more accurately described in terms of several spatial dimensions, which would allow taking this asymmetry into account. Each of the coordinates corresponding to a spatial dimension would then represent a different direction of growth away from the center of the aggregate. In analogy to the invariance relationship given above, one might then expect that the probability that a particle will be added at mass m to a small region of the aggregate active zone, at a position written in terms of these new coordinates, would be invariant under a scaling transformation of mass and several spatial dimensions.

In order to illustrate this point let us perform this generalization in following intuitive way. In Eq. (2) the affine

TABLE I. Numerical simulation values for the mass dependence exponent β are listed for four aggregate characteristic lengths, together with the mass range over which they were measured. Quantities in parentheses after exponent values are estimated uncertainties.

Characteristic length	Scaling exponent	Range of mass	Reference
Radius of gyration	0.584	100–50 000	[13]
	0.582 (0.002)	100–50 000	[11]
	0.588 (0.003)	100–2000	[11]
	0.5830 (0.0014)	10,000–1 000 000	[12]
	0.5832 (0.0014)	100,000–1 000 000	[12]
Major axis	0.567 (0.004)	100–50 000	[11]
	0.570 (0.014)	10,000–50 000	[11]
	0.555 (0.018)	100–2000	[11]
Minor axis	0.606 (0.004)	100–50 000	[11]
	0.593 (0.011)	10 000–50 000	[11]
	0.621 (0.019)	100–2000	[11]
Width of active zone	0.48 (0.01)	100–3000	[14]
	$0.48-0.54$	100–50 000	[13]
	$0.48-0.56$	1100–600 000	[12]

transformation F_0 performing the scaling can be represented by a matrix multiplying a two-dimensional column vector with components m and r. In general, an affine transformation performing scaling in a multidimensional coordinate system can be represented by a nonsingular matrix whose elements are real numbers [18]. When we change the description of the aggregate by replacing the radial coordinate r by a set of N spatial coordinates $x_1,x_2,\ldots,x_N$, we can generalize the affine transformation F_0 by replacing it with a higher dimensional affine transformation, call it F, which, since it performs scaling, can again be represented by a nonsingular matrix multiplying an $(N+1)$-dimensional vector $\xi=(m,x_1,x_2,\ldots,x_N)$. We then have the linear generalized transformation F given by

$$\xi' = F\xi, \tag{3}$$

where ξ' is another vector representing position in the active zone.

In terms of vector ξ, the assumption that the growth probability of the active zone is invariant under the scaling transformation F can be written

$$p(F\xi)d\Omega(F\xi)=p(\xi)d\Omega, \tag{4}$$

where $p(\xi)d\Omega$ is the probability of a particle being added to an aggregate, at mass m in the active growth zone in an infinitesimal region $d\Omega$, and $d\Omega(F\xi)$ is the infinitesimal region transformed by operator F. This invariance assumption acts as a requirement on change of the growth probability as mass increases. It can be used to calculate equations of motion for ensemble averages of active zone coordinates directly. Intuitively, one would expect that the application of this scaling transformation to the set of active zone positions would then describe the statistical spatial growth of the aggregates as mass increases. That is, the invariance assumption determines how positions in the active zone ξ will move, on the average, as mass increases.

Let us illustrate this point in an intuitive way using the transformation of equation (2). First, writing the infinitesimal form of transformation F_0, for λ close to unity, and then, because the scaling invariance is statistical, applying this transformation to a vector with components m and $\langle r\rangle$, where $\langle r\rangle$ is ensemble average of the radius r, we have the equation of motion

$$m\frac{\partial}{\partial m}\langle r\rangle=(1/D)\langle r\rangle \tag{5}$$

for $\langle r\rangle$, which can be identified with the average radius of deposition and is proportional to the radius of gyration. This result is just another form of the radius of gyration-mass relationship of Eq. (1). If the aggregates were symmetrical, it would be a statistical model of their growth. It is a linear equation, inheriting this property from the linearity of the affine transformation of Eq. (2). It has a constant coefficient on the right-hand side which, through its dependence on the fractal dimension D, contains the growth dynamics, including complex interactions between branches whose representation in a more mechanical model, one would expect, would be nonlinear. It describes the active zone in terms of a single characteristic length, radius of gyration, by averaging over its complex branching and multifractal structure.

The main results of this paper are a consequence of performing a not dissimilar derivation of equations of motion, this time using, in place of Eq. (2), the analogous multidimensional transformation of Eq. (3). The resulting equations will be seen to share many of the properties of Eq. (5), and in fact will be just an extension of Eq. (5) to several spatial dimensions. Specifically, these equations are linear in the spatial coordinates. In place of a constant multiplying a single coordinate on the right-hand side, one obtains a constant matrix multiplying a vector of spatial coordinate elements. Like the multiplicative constant in Eq. (5), this matrix can be interpreted as containing the growth dynamics. Now, however, this information is distributed among its elements, the diagonal elements interpreted as representing growth along a single coordinate, and the nondiagonal elements as representing interactions between growth measured along different coordinates.

The linearity of these equations is inherited from the linearity of the affine transformation of Eq. (3). It results from the severe restriction of statistical scaling invariance placed on the structure of the equations of motion. It allows them to represent complex dynamics by constants, and gives rise to algebraic solutions. However, this should not seem surprising in light of the fact that Eq. (5), the radius of gyration-mass relationship, also represents this dynamics by a constant.

These equations of motion will be seen to statistically describe the active zone, of a two-dimensional aggregate, as a vector of four averaged spatial coordinates, a large enough number to represent the asymmetry, in terms of a major and a minor axis, and the characteristic lengths radius of gyration and active zone width. Again, this should not be surprising in light of the fact that Eq. (5) also statistically describes the active zone in terms of the radius of gyration.

These equations do not predict the growth of an individual aggregate. They are consequences of the invariance of the growth probability under a symmetry transformation. They are therefore statistical equations, as is Eq. (5).

Since the multidimensional affine transformation leads to equations of motion for aggregate growth, one would expect that in choosing the detailed form and parameters of the multidimensional transformation, one could look for clues among the physical properties of the growth process. This will be discussed below.

Note that the model makes no assumptions about physical structures of aggregate growth, such as cells or branches, or about a growth mechanism, such as an interaction of branches through a Laplacian field. It instead is written in terms of distances to the active zone, measured along various coordinates, and the scaling symmetry of the active zone as mass increases. If interactions between growth processes are discussed, they are mentioned only as interpretations of terms occurring in the symmetry transformation. As Eq. (5) indicates, symmetry terms containing interaction information may bear little resemblance to mechanical interaction terms.

The following discussion gives a more detailed account of the construction of the proposed DLA model and simulation evidence to support it. Section I below defines a scaling transformation of mass and spatial coordinates under which one might expect DLA to be invariant, and writes an infinitesimal form of the transformation in terms of differential equations relating changes in active zone position and aggregate mass. Section II derives equations of motion for characteristic lengths and discusses physical considerations involved in determining model parameters. Section III compares the results of the model with the data of DLA numerical simulations. Section IV is a summary and discussion of the results.

I. DEFINITION OF A SCALING TRANSFORMATION OF MASS AND SPATIAL COORDINATES

I have assumed that position in the active zone of the aggregate growth process can be written in terms of a set of N spatial coordinates x_i, $i=1,2,\ldots,N$, and have defined a vector $\xi=(m,x_1,x_2,\ldots,x_N)$ as a compact way of writing active zone position at a given aggregate mass m.

The basic assumption of the proposed model is then that the probability $p(\xi)d\Omega$ of a particle being added to an aggregate, at mass m in the active growth zone in an infinitesimal region $d\Omega$, and at a position given by the coordinates $x_1,x_2,\ldots,x_N$, is invariant under a scaling of mass and spatial coordinates. In analogy to the affine transformation in mass and one spatial dimension of Eq. (2), I perform this scaling by an affine transformation in mass and N spatial dimensions. Affine transformations are linear transformations that represent operations including magnifications and translations of coordinates [18]. As mentioned above, an affine transformation performing scaling in a multidimensional coordinate system can be represented by a nonsingular matrix whose elements are real numbers. This is indicated above as transformation F of Eq. (3). One can write the elements of this matrix, in analogy to Eq. (2), as

$$F_{ij}=\lambda^{A_{ij}}, \tag{6}$$

where λ is a real number greater than unity, and the A_{ij} determine matrix element values, with $i=0,1,2,\ldots,N$, $j=0,1,2,\ldots,N$.

Let us now write the application of the transformation F to vector ξ as a differential equation relating infinitesimal changes in active zone coordinates $x_1,x_2,\ldots,x_N$ as mass increases incrementally. This result will be used in Sec. II to write an expression for the invariance of the growth probability under the coordinate transformation F. The latter relation will in turn be used to derive equations of motion for statistical moments of active zone position. Note that this section contains no DLA equations of motion, but only expressions for a mathematical transformation of coordinates.

Let us first restrict transformation F so that it induces only an infinitesimal change in mass, that is, λ will be infinitesimally close to unity; these results will then not apply to aggregates with a small number of particles. We obtain, in this case, the following result for the rate of change, with respect to λ, in active zone position (given by coordinate vector ξ) that is produced by application of the operator F:

$$\frac{\partial \xi_i}{\partial \lambda}=\sum_j A_{ij}\xi_j, \tag{7}$$

where $i=0,1,2,\ldots,N$, $j=0,1,2,\ldots,N$. If we restrict the transformation to eliminate what would seem to be the unphysical situation where mass and spatial coordinates interact directly through the elements A_{0j} and A_{j0} with $j\neq 0$, we can set

$$A_{0j}=A_{j0}=0, \quad j\neq 0. \tag{8a}$$

In addition, we can choose

$$A_{00}=1 \tag{8b}$$

without losing generality, because this choice just gives the relation between mass and arbitrary scaling parameter λ. We then have the following result for the variation with respect to mass of the spatial coordinates under application of the operator F:

$$\frac{\partial}{\partial \mu}x_i=\sum_j A_{ij}x_j, \tag{9}$$

where $\mu = \ln(m)$. Now, however, $i = 1, \ldots, N$ and $j = 1, \ldots, N$. Here, the transformation has been written in the form of differential equations relating change in each of the active zone spatial coordinates x_i to change in log of mass μ.

II. A MODEL OF AGGREGATE GROWTH

Using the coordinate transformation F, in the incremental form of Eq. (9), will allow us to derive statistical equations of motion for the active zone coordinates and characteristic aggregate lengths. First, let us write the invariance of the growth probability $p(x,\mu)d\Omega$ under the coordinate transformation F, where x represents the coordinate set $x_1, x_2, \ldots x_N$, and a prime on a quantity indicates it has been incrementally changed according transformation of Eq. (9). The invariance of the probability is represented by the equation

$$\sum_{i,j} \frac{\partial p(x,\mu)}{\partial x_i} A_{ij} x_j + \frac{\partial p(x,\mu)}{\partial \mu} + p(x,\mu) \frac{\partial}{\partial \mu'} [J(x,\mu')]_{\mu'=\mu} = 0, \tag{10a}$$

where J, given by

$$d\Omega(x',\mu') = J(x',\mu') d\Omega(x,\mu), \tag{10b}$$

is the Jacobian determinant of the transformation. Use of Eq. (10), in a direct calculation of the change in coordinate moments induced by the transformation, then yields one set of equations of motion for the ensemble averages $\langle x_i \rangle$ of the aggregate coordinates and another for their two-point cumulants $\langle x_i x_j \rangle_c$. The coordinate ensemble averages lead to equations of motion for radius of gyration and major and minor asymmetry axes, and the two-point cumulants lead to equations for active zone width. Appendix A contains a discussion of how active zone is defined and measured, and how it is related to the two-point cumulants.

Performing the direct calculations mentioned above, one arrives at equations for the ensemble averages,

$$\frac{\partial}{\partial \mu} \langle x_i \rangle = \sum_j A_{ij} \langle x_j \rangle, \tag{11}$$

with $i = 1, \ldots, 4$. The equations for the cumulants can, by making use of their symmetry under interchange of the coordinate indices, be written as the following set of ten equations:

$$\frac{\partial}{\partial \mu} \langle x_i x_j \rangle_c = \sum_k A_{ik} \langle x_k x_j \rangle_c + \sum_k A_{jk} \langle x_k x_i \rangle_c, \tag{12}$$

where $j \geqslant i$, and $i,j = 1, \ldots, 4$. If, further, the ten index pairs $\{i,j\}$ are treated as a single index, then Eqs. (12) can be written in the form

$$\frac{\partial}{\partial \mu} \langle x_i x_j \rangle_c = \sum_{(l,n)} B_{(i,j)(l,n)} \langle x_l x_n \rangle_c, \tag{13}$$

where the elements of matrix B are given in terms of the matrix elements of A, and the sum is over all ten index pairs $\{l,n\}$.

Writing the solutions to the equation sets (11) and (13) is simplified by the fact that both have the same form, given by the vector equation

$$\frac{\partial}{\partial \mu} u = Lu, \tag{14}$$

where L represents either matrix A or B, and where u indicates the vector whose elements are either the averaged coordinates $\langle x_i \rangle$, for matrix A, or the cumulants $\langle x_i x_j \rangle_c$, for matrix B. The well-known solution of this equation is then given by

$$u(\mu) = \sum_l e^*(l) \cdot u(\mu_0) \exp[l(\mu - \mu_0)], \tag{15}$$

where $u(\mu)$ is the solution at mass parameter μ, $u(\mu_0)$ is the initial value of the solution at μ_0, $e(l)$ is an eigenvector of the matrix L corresponding to the eigenvalue l. For the case of a non-self-adjoint matrix B, $e^*(l)$ is an eigenvector of the adjoint of matrix L corresponding to the eigenvalue l; for the case of a self-adjoint matrix A, $e^*(l) = e(l)$. The quantity $e^*(l) \cdot u(\mu_0)$ is the inner product the vectors $e^*(l)$ and $u(\mu_0)$, and the summation is carried out over all eigenvalues of the matrix.

As argued in the introduction, one would expect that in choosing the parameter values of the multidimensional transformation, and therefore of the equations of motion, one could look for clues among the physical properties of the growth process. Specifically, the fact that the mass dependence exponents of the major and minor axes differ by only a fraction of their average value suggests that their difference is caused by an interaction of growth along different spatial dimensions. Let us identify this interaction with the off-diagonal elements in the equations of motion, that is, off-diagonal elements of matrix A. Similarly, the fact that these two mass dependence exponents differ from the space-filling value of one-half by only a fraction of this value suggests that, if these growth processes were mathematically isolated, they would have identical mass-dependent exponents, and that each would be space filling. Let us then identify the motion of the isolated processes with the diagonal elements of A. A comparison of the analogous expressions in Eqs. (2) and (6) would then lead us to set each of the diagonal elements of A to the space-filling value of one-half.

The spatial coordinates x_i, as described above, represent directions of growth away from the aggregate seed particle. Because the asymmetry of the aggregates is described adequately, in rectangular coordinates, by the magnitudes and mass-dependent exponents of their major and minor axes [11], it would seem that one can define the spatial coordinates x_i as four positive distances from the seed particle to positions in the active growth region, each in a direction at a 90° angle from its two neighbors. That is, if a two-dimensional rectangular coordinate system were centered at the seed particle, the coordinates x_i would lie along all four positive and negative coordinate axes.

TABLE II. Initial conditions, used with either Eqs. (11) or (13), in calculating the growth of four aggregate characteristic lengths are listed along with the mass for which they are given.

Characteristic length	mass	$\langle x_{\mathrm{odd}}\rangle$	$\langle x_{\mathrm{even}}\rangle$	$\langle x^2_{\mathrm{even\ or\ odd}}\rangle/\langle x_{\mathrm{even}}x_{\mathrm{odd}}\rangle$
Radius of gyration [11]	200	10.47	10.47	
Major, minor axes [11]	200	8.54	5.93	
Width of active zone [21]	1096			−8.7

Finally, if one of these growth processes interact with others, the most reasonable expectation is that it would interact with its neighbors to its right and left.

With these assumptions, the elements of the matrix A, for $1 \leqslant i,\ j \leqslant 4$, take the following values

$$A_{ij}=\begin{cases} v_0, & i=j \\ v_1, & |i-j|=1 \quad \text{or} \quad i=1, j=4 \quad \text{or} \quad j=1, i=4 \\ 0 & \text{otherwise,} \end{cases} \tag{16}$$

where v_0 represents the space-filling value of one-half, and v_1 is an undetermined constant representing the interaction between a growth process and its neighbors. Since the elements of matrix A are known, matrix B can be found. An expression for matrix B is given in Appendix B.

III. COMPARISON WITH NUMERICAL SIMULATIONS

In summary, the above argument, by starting with the assumption that the DLA growth zone is invariant under the infinitesimal affine transformation defined by Eqs. (3), (6), (8), and (16), describes a model of aggregate growth which implies the equations of motion, given by Eqs. (11) and (13), for the growth of the characteristic aggregate lengths. The equations of motion have the form of an initial value problem with one undetermined parameter v_1. The following discussion (1) determines these initial conditions from the numerical simulation data, (2) finds a value for parameter v_1, and (3) displays the evidence that the predictions for growth of the characteristic lengths from their initial values are consistent, to a high degree, within the mass range considered, with the assumption of affine symmetry.

For the calculation of radius of gyration and major and minor axes, the mass range considered here is 200–50 000, which includes all but the lowest part of the mass range of the data in Ref. [11]. Initial conditions for these quantities are set at mass equal to 200. For the active zone width, the mass range is 1100–440 000, corresponding to values of μ in the range 7–13, which is approximately the mass range of the data in Ref. [12]. The initial condition is set at μ equal to 7. In both cases, including lower mass values would not further test the proposed model, because it does not apply to aggregates of small mass.

Initial conditions for calculating a characteristic length are chosen to reflect the experimental situation, including the relevant coordinate average or cumulant, used to measure that quantity. Table II is a summary of the initial conditions for Eqs. (11) and (13) together with the references from which the data was taken. When choosing initial conditions for calculating the angular independent radius of gyration, I take the coordinates $x_1,\ldots x_4$ to be fixed in space, the values of their ensemble averages to be independent of individual aggregate orientation, and each average to be equal to the radius of gyration. The initial conditions for these coordinates are then given directly by the simulation results [11].

For choosing major and minor axis initial conditions, I take the coordinates $x_1,\ldots x_4$ to be fixed in the body of the aggregate, and take the ensemble averages of the aggregate to have inversion symmetry about the seed particle. I also take each of the ensemble averages of coordinates x_1 and x_3 to be equal to that of the major axis and, similarly, take those of x_2 and x_4 to be equal to that of the minor axis. The initial conditions for these coordinates are also given directly by the simulation results [11].

For active zone initial conditions, I again use coordinates fixed in space because active zone width is also independent of individual aggregate orientation. For Eq. (13), in which active zone initial conditions are to be used, there are two independent quantities that enter into these initial conditions: the same-coordinate cumulant $\langle x_i^2\rangle_c$ and the cross-coordinate cumulant $\langle x_i x_j\rangle_c$, $i\neq j$. Actually, in order to calculate β for the active zone width, as is done here, all that is needed is the ratio of these the quantities.

Because I am not aware of any previous measurements of this ratio, I found a value using a DLA computer simulation. In order to make clear how these cumulants were measured, note that in a single aggregate each coordinate $x_1,\ldots x_4$ represents the active zone position, at a given mass, measured along one the four orthogonal half-axes. Therefore, for example, the same-coordinate cumulant $\langle x_1^2\rangle_c$ involves averages of the distance x_1 from the seed particle to the active zone on half-axis number 1. The cross-coordinate cumulant $\langle x_1 x_2\rangle_c$ involves averages of the distances x_1 and x_2 to the active zone, where both distances are from the same aggregate at the same mass.

Using the simulation, I first found values for the two cumulants at mass intervals of 200 in the mass range 600 to 1600 using 200 aggregates for each point [19]. On a log-log plot of each cumulant against mass, a linear regression gave a best fit to the data and a value for each cumulant at μ equal to 7. The resulting ratio of same-coordinate to cross-coordinate cumulant at that value of μ is approximately -8.7. Data for the cross-coordinate cumulant contain a good deal of randomness. Taking this into account gives approximate uncertainty limits of $-8.7+3.8$ for the upper limit and $-8.7-7.0$ for the lower limit.

Using the initial conditions above, one can find a value for the undetermined parameter v_1. The most straightforward and accurate way seems to be to fit the calculated value of the exponent β for radius of gyration r_g to its high mass simulated value. The reason is that the model-predicted value for β is a constant, and its simulation value appears to be nearly constant over the range for which it has been measured. The simulation value is constant to within $\frac{1}{2}$% for masses above approximately 1800, and appears to oscillate about its high mass value for masses down to about 1000 [12]. The result of this fitting procedure is that $v_1=0.0415$,

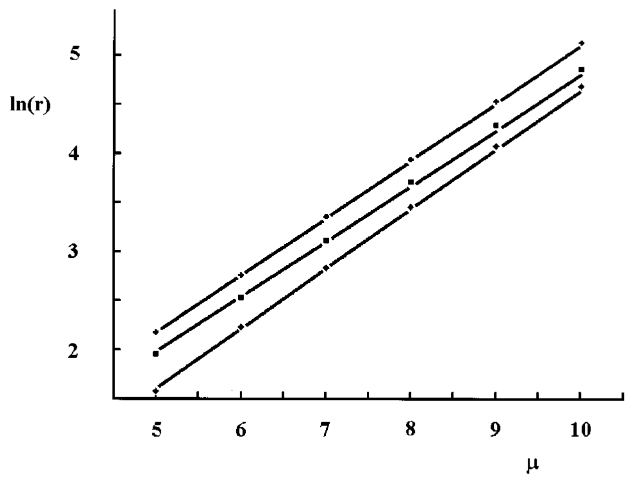

FIG. 1. The log of each of three characteristic lengths is plotted against log of mass μ. The lines represent calculated results of the proposed model, and the points represent results of numerical simulations from Ref. [11]. The major axis $r_>$ is represented by the top line and the points, $+$; the radius of gyration r_g is represented by the middle line and the points, ■; the minor axis $r_<$ is represented by the bottom line and the points, ♦.

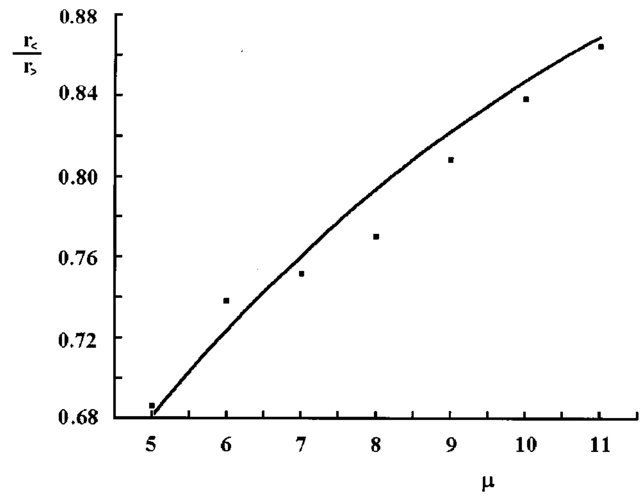

FIG. 2. The ratio of the minor to the major symmetry axis $r_< / r_>$ is plotted against the log of mass μ. The line represents the calculated results of the proposed model, and the points represent the results of numerical simulations from Ref. [11].

corresponding to an exponent $\beta = 0.5830$ for the radius of gyration.

Then, applying the solution given in Eq. (15) to Eq. (11), together with the appropriate initial conditions, gives the mass dependence for the growth of the radius of gyration and of the major and minor axes. These results, shown in Figs. 1 and 2, agree well with simulation results. Figure 1 shows the variation of the three characteristic lengths with mass, and Fig. 2 shows the variation of the ratio of minor to major axis with mass. Also, calculated values for the mass-dependence exponent β for the major and minor axes, shown in Table III, agree well (to within 1.3% or better) with simulation results in the three mass ranges 100–2000, 10 000–50 000, and 100–50 000. Both the calculated and simulated values of β for major and minor axes vary monotonically over the three mass ranges. For the major axis, the calculated value increases by 1.6%, while the simulated value increases by 0.9%; and for the minor axis, both values decrease, the calculated value, by 2.5% and the simulated, by 4.6% [11]. In contrast, as mentioned above, the calculated value of β for the radius of gyration is constant in the three mass ranges, while the simulation value is nearly constant [12].

As can be seen in Fig. 1, calculated values for all three characteristic lengths appear to increase at a slightly slower rate than simulated values. A possible explanation is that initial conditions are set here at the small mass value of 200. If they are set at a higher value, such as 1100, the discrepancy decreases.

In an analogous way to Eq. (11), the solution of Eq. (13) yields the mass dependence of the cumulants $\langle x_i^2 \rangle_c$ for $i = 1 \ldots 4$. These quantities, all equal because of the symmetry of the initial conditions, are each equal to the square of the width of the active zone. Therefore, the derivative with respect to μ of the natural logarithm of the square root of any $\langle x_i^2 \rangle_c$ is equal to the mass dependence exponent β of the active zone width. Shown in Fig. 3, for the mass range of about 1100–440 000 [20], the calculated values of β have the correct qualitative increase with mass, and also agree well with simulation values.

Although the simulated value for the initial condition used to solve Eq. (13) has a great deal of uncertainty, the solution to the equation (in Fig. 3) is only weakly dependent on its value. For example, at μ equal to 7, this uncertainty could

TABLE III. This list compares results for the mass dependence exponents β of three characteristic aggregate lengths given by the proposed model and by numerical simulations. The results are listed with the mass range for which they were measured for calculated. References are to numerical simulations. Quantities in parentheses after simulation exponent values are estimated uncertainties.

Characteristic length	Range of mass	Model value	Simulation value	Reference
Radius of gyration	200–50 000	0.5830	0.5830 (0.0014)	[12]
Major axis	200–2000	0.561	0.555 (0.018)	[11]
	10 000–50,000	0.570	0.570 (0.014)	[11]
	200–50 000	0.566	0.567 (0.004)	[11]
Minor axis	200–2000	0.613	0.621 (0.019)	[11]
	10 000–50 000	0.598	0.593 (0.011)	[11]
	200–50 000	0.606	0.606 (0.004)	[11]

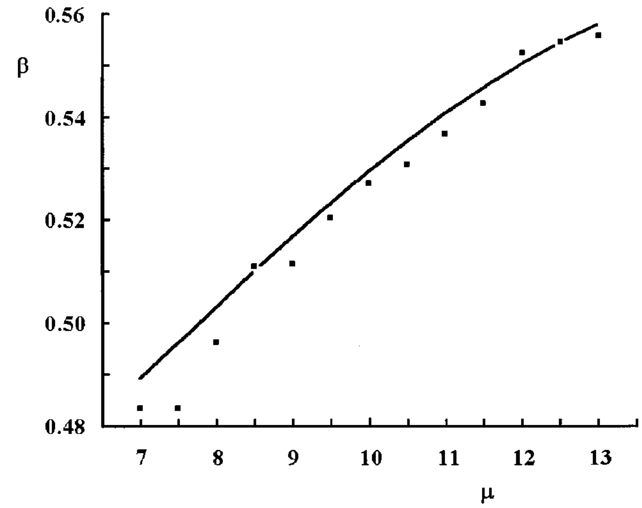

FIG. 3. The mass dependence exponent β for the aggregate active zone width is plotted against log of mass μ. The line represents the calculated results of the proposed model, and the points represent the results of numerical simulation from Ref. [12].

change active zone width by as much as 1.6% and at μ equal to 13, it could change width by as much as 0.8%.

In addition to the agreement of calculated characteristic lengths with numerical simulation results, the calculated values have clear geometric interpretations within the eigenvector space of the matrices A and B. Table IV shows the eigenvectors, and corresponding eigenvalues, that contribute to the growth of the aggregate characteristic lengths. For the radius of gyration, only the angular independent eigenvector contributes to its growth, because of the angular independence of the initial conditions. The corresponding eigenvalue 0.5830 is therefore equal to the mass dependence exponent β of this characteristic length. For the major and minor axes, both the angular independent eigenvector and the inversion symmetric vector contribute to growth, because of the inversion symmetry of the initial conditions. The eigenvalues of these two vectors, 0.5830 and 0.4170, respectively, therefore, both contribute to the exponent β for the major and minor axes. This has the consequences that (1) neither β, for the two axes, is constant, and that (2) the β of the minor axis has a larger value than either of the two contributing eigenvalues, because of the partial cancellation of the two contributions. For the active zone width, only the two angular independent eigenvectors, with eigenvalues 1.166 and 0.834, contribute to growth, again because of the angular independence of the initial conditions. The combination of these two values of mass dependence accounts for the variation in the active zone width exponent β, within a mass range of 1100–440 000, from a low value of about 0.48 to a high value of about 0.56. These limits are comparable to half the values of the two contributing eigenvalues, that is, 0.417 and 0.583.

IV. SUMMARY AND DISCUSSION

This work has two major results. It proposes a model of DLA that accounts for the diversity of mass dependence exponents of four characteristic lengths: radius of gyration, major and minor asymmetry axes, and width of active growth zone. It uses the agreement of model predictions with simulation data to show that the invariance of DLA growth probability under an infinitesimal affine transformation of mass and several growth zone spatial coordinates is consistent with the results of simulation data.

The limitations of the model include that it provides only a statistical picture of DLA. It makes testable statements about only the characteristic lengths of the active growth zone, all of which are of order of magnitude of the aggregate itself. It does not apply to small aggregates or the early growth period of large aggregates; initial conditions for the equations of motion, then, must be supplied by simulation. It has been tested for only the ranges of mass where characteristic length data are available.

However, the model does predict the change, as mass increases, of the four characteristic lengths with a high degree of accuracy. It predicts quantitative relationships among fractal dimension, aggregate asymmetry, and the multiple mass-dependent exponents.

The affine symmetry portrays aggregate growth, not as a detailed dynamics of aggregate structures, but as the result of scaling invariance of the growth process in multiple spatial dimensions, that is, as the result of an overall restriction of a lack of preferred length parameter. Multiple spatial dimensions, however, allow the symmetry transformation to have terms that lead to interactions between growth processes along different coordinates. These interactions provide an explanation of the complex behavior of aggregate growth. They arise within the restriction of scaling invariance and take on a different mathematical form from the interactions of the detailed dynamics.

The model contributes to the ongoing discussion of whether DLA is characterized by a single scaling exponent, with complications arising from finite-size effects such as random fluctuations, or whether DLA has a more complex scaling property. By its success in accounting for character-

TABLE IV. Two eigenvectors of matrix A and two of matrix B contribute to the growth of the four characteristic lengths. The components e_i ($i=1-4$ for A and $i=11,12,\ldots,44$ for B) of each of these is listed here along with its corresponding eigenvalue.

Matrix	eigenvalue	Eigenvector of A: e_{odd}	Eigenvector of A: e_{even}	Eigenvector of B: $e_{11},e_{13},e_{22},e_{24},e_{33},e_{44}$	Eigenvector of B: $e_{12},e_{14},e_{23},e_{34}$
A	0.5830	0.5	0.5		
	0.4170	0.5	-0.5		
B	1.166			0.323	0.323
	0.834			0.323	-0.323

istic length growth, it suggests that complex scaling, that is, invariance under the multidimensional affine transformation, is a reasonable explanation for the diversity of mass dependence exponents among the four characteristic lengths. It gives a physical picture of how this scaling arises in terms of growth processes which, if isolated would have simple scaling properties, but instead interact and form a more complex aggregate scaling relationship.

The model therefore describes DLA as a process in which the different scaling exponents of the characteristic lengths arise from deterministic dynamics. But it predicts that this complex scaling will only occur for finite mass. In the limit of very large mass, it predicts that all characteristic lengths will scale with the same exponent, that is, all growth terms in the solution of the equations of motion will become negligible except for those corresponding to the largest eigenvalue.

In spite of the differences between the picture of DLA given by the proposed model and that of the early intuitive view of DLA as simple scale invariant growth, both have a form of scaling symmetry under which the growth is statistically invariant and which simplifies the view of the process.

APPENDIX A: WIDTH OF ACTIVE GROWTH ZONE

The active growth zone in DLA can be understood intuitively as the fragmented outer region of a single aggregate where particles are added [11], or statistically as a distribution of locations, over an ensemble of aggregates of given mass, at which the last particle is added [14]. The purpose of this discussion is an attempt to relate these two views of active zone, and to determine their relationship to the proposed model.

Let us first show that the proposed model uses the statistical view of active zone. Let us examine Eq. (2) of a paper by Plischke and Racz [14] that contains the original definition of width of the active zone. Written below (numbered as PR 2) in its original notation, this equation consists of definitions of mean deposition radius $\overline{r_N}$ and the square of active zone width ξ_N^2 (please note that their quantity ξ_N^2 is not the vector ξ used above):

$$\overline{r_N}=\frac{1}{M}\sum_{i=1}^{M} r_N(i), \quad \xi_N^2=\overline{(r_N-\overline{r_N})^2}. \tag{PR2}$$

The authors define $r_N(i)$ as ''the deposition radius of the Nth particle in the ith cluster.'' In other words, the mean deposition radius is defined as an average value of the radius taken over an ensemble of aggregates. More importantly for our purposes, the notation indicates that the square of active zone width is also an aggregate ensemble average, of squares of deviations of the deposition radius from its mean. That is, it is the variance of the deposition radius over the ensemble. In terms of the coordinates x_i of the proposed model, which represent position in the aggregate active zone, it would then seem that active zone width can be represented by the cumulants $\langle x_i^2\rangle_c$, and its equation of motion is given by the equations of motion for the two-point cumulants $\langle x_i x_j\rangle_c$. The present paper uses active zone width simulation results of Tolman and Meakin [12], Meakin and Sander [13], and Plischke and Racz [14], all of which use the statistical view of active zone and the original statistical definition of its width.

What then is the relationship between the intuitive and statistical views? Since the statistical view of the active zone tells us only about ensemble averages and ensemble fluctuations, one might ask if it is capable of describing the width of the active zone considered as the fragmented outer region of a single aggregate where particles are added? The following consideration indicates that there is not such a great difference, as the above reasoning might suggest, between active zone in an individual aggregate and active zone as an ensemble average.

When measuring the width of active zone according to the definition of Plischke and Racz, one would generate a collection of aggregates of a mass m, and record the radial distance at which particle m is added in each of them. And, when measuring it in a more intuitive way, one might take an aggregate of mass $m-1$ and list the radial distances of all possible positions at which particle m might attach itself. Both methods would produce a list of the set of possible locations at which particle m might be attached, that is, they would both chart the (intuitive) active zone of a mass m aggregate. They would both seem to give comparable results.

The statistical and intuitive views of width of active zone, seen in terms of how they are applied in practice, then appear to be different ways of characterizing the same aggregate physical region. They are not incompatible views.

Still, any differences between the two views should not be crucial for the purposes of the present paper. This paper has the limited purpose of proposing a statistical model of DLA, and comparing its predictions with published data in order to test the model. Considering the logic only, the choice of view of active zone width will be appropriate for this purpose as long as the view it uses is the same as that of the references with which its predictions are compared. The paper compares its data with that of Tolman and Meakin [12], who refer one to the statistical definition of width of active zone as given by Plischke and Racz [14]. For the limited purposes of the present paper, therefore, use of the statistical view is appropriate and accomplishes the intended result of testing the proposed model.

APPENDIX B: MATRIX B

The ten-dimensional matrix B is given by the following. The definitions of both v_0 and v_1 are given in Sec. II, and the rows and columns are numbered by the index pairs $\{i,j\}=11,12,\ldots,44$, for which $j\geqslant i$,

$$B=\begin{pmatrix}
2v_0 & 2v_1 & 0 & 2v_1 & 0 & 0 & 0 & 0 & 0 & 0\\
v_1 & 2v_0 & v_1 & 0 & v_1 & 0 & v_1 & 0 & 0 & 0\\
0 & v_1 & 2v_0 & v_1 & 0 & v_1 & 0 & 0 & v_1 & 0\\
v_1 & 0 & v_1 & 2v_0 & 0 & 0 & v_1 & 0 & 0 & v_1\\
0 & 2v_1 & 0 & 0 & 2v_0 & 2v_1 & 0 & 0 & 0 & 0\\
0 & 0 & v_1 & 0 & v_1 & 2v_0 & v_1 & v_1 & 0 & 0\\
0 & v_1 & 0 & v_1 & 0 & v_1 & 2v_0 & 0 & v_1 & 0\\
0 & 0 & 0 & 0 & 0 & 2v_1 & 0 & 2v_0 & 2v_1 & 0\\
0 & 0 & v_1 & 0 & 0 & 0 & v_1 & v_1 & 2v_0 & v_1\\
0 & 0 & 0 & 2v_1 & 0 & 0 & 0 & 0 & 2v_1 & 2v_0
\end{pmatrix}. \tag{B1}$$

[1] T. A. Witten, Jr. and L. M. Sander, Phys. Rev. Lett. **47**, 1400 (1981).

[2] D. Grier, E. Ben-Jacob, Roy Clarke, and L. M. Sander, Phys. Rev. Lett. **56**, 1264 (1986).

[3] L. Niemeyer, L. Pietronero, and H. J. Wiesmann, Phys. Rev. Lett. **52**, 1033 (1984).

[4] D. Bensimon, L. P. Kadanoff, S. Liang, B. I. Shrainman, and C. Tang, Rev. Mod. Phys. **58**, 977 (1986).

[5] M. Muthukumar, Phys. Rev. Lett. **50**, 839 (1983).

[6] Herbert Levine and Yuhai Tu, Phys. Rev. E **48**, R4207 (1993).

[7] Raphael Blumenfeld, Phys. Rev. E **50**, 2952 (1994); Raphael Blumenfeld and Robin C. Ball, Phys. Rev. A **51**, 3434 (1995).

[8] X. R. Wang, Y. Shapir, and M. Rubinstein, Phys. Rev. A **39**, 5974 (1989).

[9] L. Pietronero, A. Erzan, and C. Evertsz, Phys. Rev. Lett. **61**, 861 (1988).

[10] Thomas C. Halsey and Michael Leibig, Phys. Rev. A **46**, 7793 (1992); Thomas C. Halsey, Phys. Rev. Lett. **72**, 1228 (1994).

[11] Peter Garik, Phys. Rev. A **32**, 1275 (1985).

[12] Susan Tolman and Paul Meakin, Phys. Rev. A **40**, 428 (1989).

[13] P. Meakin and L. M. Sander, Phys. Rev. Lett. **54**, 2053 (1985).

[14] M. Plischke and Z. Racz, Phys. Rev. Lett. **54**, 2054 (1985).

[15] Chi-Hang Lam, Phys. Rev. E **52**, 2841 (1995).

[16] Bayard K. Johnson, Robert F. Sekerka, and Michael P. Foley, Phys. Rev. E **52**, 796 (1995).

[17] Benoit B. Mandelbrot, *Fractals: Form, Chance, and Dimension* (Freeman, San Francisco, 1977), p. 276.

[18] Ernst Snapper and Robert J. Troyer, *Metric Affine Geometry* (Academic, New York, 1971), pp. 73–96.

[19] I used an on-lattice simulation. For aggregates of these masses, it is thought that there is no difference between on-lattice and off-lattice simulations: F. Argoul, A. Arneodo, J. Elezgaray, G. Grasseau and R. Murenzi, Phys. Rev. A **41**, 5537 (1990).

[20] The mass range 1100–440 000 differs from the Ref. [12] mass range of approximately 1100–600 000 only in that the independent variable in Fig. 3 is the logarithm of mass, and that, because the simulated values are represented by points at intervals of unity, no point corresponds to a mass of 600 000.

[21] See Sec. III above.