Off-lattice noise reduced diffusion-limited aggregation in three dimensions

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Using off-lattice noise reduction, it is possible to estimate the asymptotic properties of diffusion-limited aggregation clusters grown in three dimensions with greater accuracy than would otherwise be possible. The fractal dimension of these aggregates is found to be 2.50 ± 0.01, in agreement with earlier studies, and the asymptotic value of the relative penetration depth is $\xi/R_{dep}=0.122\pm0.002$. The multipole powers of the growth measure also exhibit universal asymptotes. The fixed point noise reduction is estimated to be $\epsilon^f\sim0.0035$, meaning that large clusters can be identified with a low noise regime. The slowest correction to scaling exponents are measured for a number of properties of the clusters, and the exponent for the relative penetration depth and quadrupole moment are found to be significantly different from each other. The relative penetration depth exhibits the slowest correction to scaling of all quantities, which is consistent with a theoretical result derived in two dimensions. We also note fast corrections to scaling, whose limited relevance is consistent with the requirement that clusters grow far enough in radius to support sufficient scales of ramification.

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Diffusion-limited aggregation (DLA) [1] is an extensively studied model of diffusion limited growth which appears to capture the essential features of many different physical growth phenomena [2–5]. However, the fractals generated have evaded a complete understanding for many years and there has recently been controversy over whether diffusion-limited aggregates are truly fractal [6].

DLA is modeled [1] by allowing particles to randomly walk from a sphere surrounding the cluster, one at a time, until they contact the cluster, at which point they are irreversibly stuck. Detailed study [7] has shown that DLA growth in two dimensions does approach true fractal scaling, but with slowly decaying corrections to scaling of the form

$$Q_N = Q_\infty + C_Q N^{-\nu}, \tag{1}$$

where Q_N is some property of the cluster, tending towards the value Q_∞ as the number of particles in the cluster, N, tends to infinity. Here the correction to scaling exponent ν is expected to exhibit some universality while the constant C_Q will not. For aggregates grown in two dimensions, it has been argued [7] that there should be no quantity whose correction to scaling is slower than that of the relative penetration depth, ξ/R_{dep}, where R_{dep} is the average radius at which new particles are deposited and ξ is the standard deviation of the same. Here and below we take as origin the centroid of the depositing particles.

When studying DLA grown on a lattice, reducing the shot noise associated with the growth being by discrete particles [8] has proved valuable in understanding the asymptotic properties of clusters, complicated by their sensitivity to lattice anisotropy [9,10]. Using a conformal map from the unit circle to the boundary of a growing cluster, Hastings and Levitov [11] introduced a technique for implementing a noise reduction scheme for DLA clusters grown off-lattice. Rather than adding particles to the cluster, bumps were added to the conformal map. Ball *et al.* [12] built on this work, allowing crescent-shaped bumps to be added to DLA clusters without the need for a conformal map. In this approach, the particles are allowed to diffuse normally until they contact the cluster. At this point the new particle is touching the cluster, and the distance between the center of the new particle and the center of the particle it contacted in the cluster is equal to the diameter of one particle. To implement noise reduction, this distance is reduced by a factor $A<1$, so that the new particle is deposited partially inside the cluster: the effect is to protrude a shallow bump of height A on the cluster perimeter. Since this method does not rely on a conformal map it allows the growth of noise reduced DLA clusters off-lattice in any dimension.

Most of the work on DLA has been restricted to two dimensions. Meakin pioneered work on DLA in higher dimensions, growing clusters in dimensions up to $d=8$ [13]. Much of that work has focused on estimating the fractal dimension of DLA clusters and the scaling of the relative penetration depth [14–16], yet firm conclusions have proved difficult. There has also been some progress measuring the multifractal spectrum of DLA in three dimensions [17–19]. However, Davidovitch *et al.* [20] recently claimed that all previous attempts to measure $f(\alpha)$ in two dimensions are poorly converged, so the early three dimensional measurements should be taken with caution. Other work has also examined DLA on a cubic lattice in the limit of zero noise [21], and the extension of the fixed scale transformation to 3 dimensions [22].

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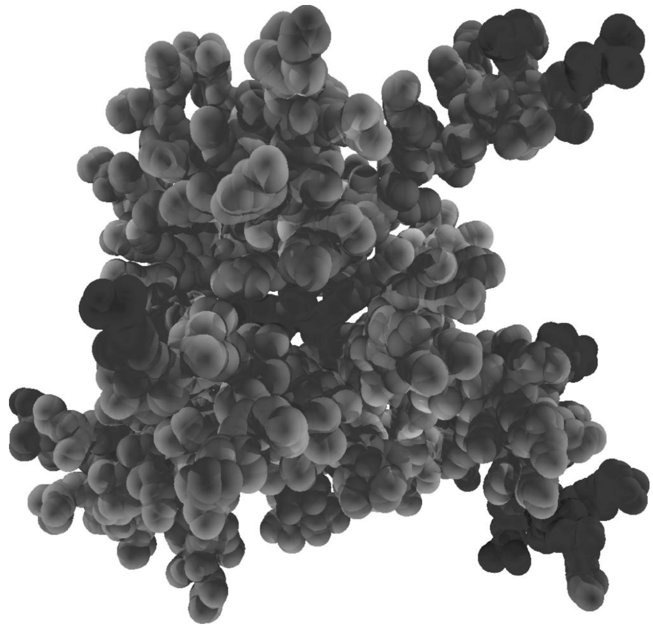

FIG. 1. A DLA cluster grown in three dimensions with $N=10^4$ particles and noise reduction factor $A=0.1$, where the different shading indicates a different time of deposition on the cluster.

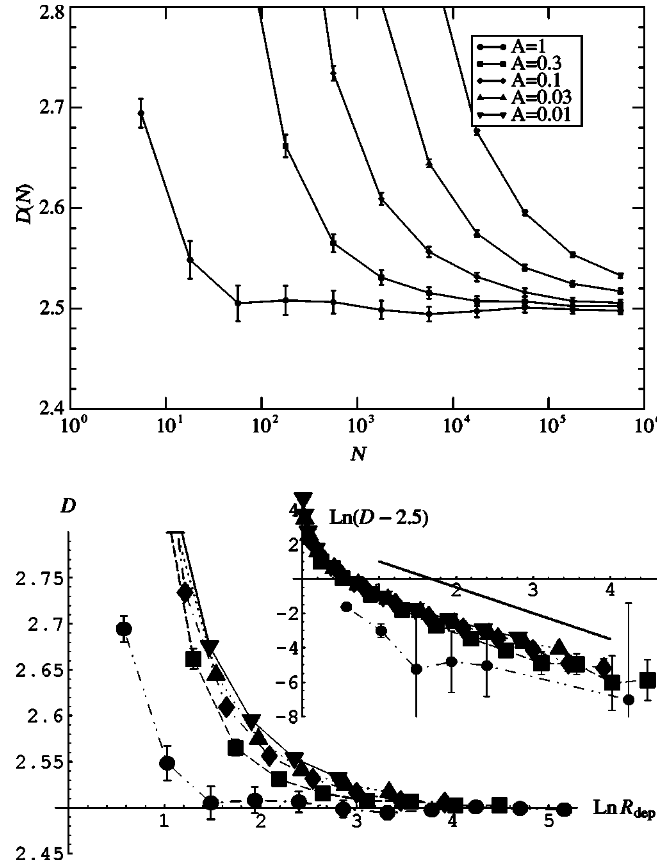

FIG. 2. The measured fractal dimension D of DLA clusters, estimated by taking the local slope of $\ln N$ vs $\ln R_{dep}$. Plotted vs N in the upper panel, the results for different levels of noise reduction A are initially well separated but converging to a universal dimension of $D=2.50\pm0.01$. The noise reduced results come much more together when plotted vs $\ln R_{dep}$ as in the lower panel, suggesting that it is fundamentally radius which governs the departure of D from its asymptote. The inset shows $\ln(D-2.5)$ vs $\ln R_{dep}$ and the guideline drawn corresponds to $D-2.5\propto R_{dep}^{-1.5}$.

In this paper we exploit the new noise reduction techniques to explore the convergence to scaling of DLA in three dimensions. We grew 1000 DLA clusters off-lattice in three dimensions spanning five different values of the noise reduction $A=1$, 0.3, 0.1, 0.03, 0.01, and an example cluster with $N=10^4$ particles and $A=0.1$ is shown in Fig. 1. At integer values of $\log_{\sqrt{10}}N$, the growth of the clusters was suspended and 10^5 probe particles were "fired at" the cluster: these probe particles were allowed to diffuse freely, one at a time, until they contacted the cluster, at which point their location was recorded and the particle was deleted. In this way the growing properties of the clusters (such as the penetration depth and multipole moments) were estimated. The code used is a direct descendant of that of Meakin [14], which in turn builds on the computational tricks of Ball and Brady [23] to speed up computation. While the code is truly lattice free, the smallest step size that a particle was allowed to take was set equal to one particle radius. Comparisons in 2 dimensions between this and an algorithm which uses a much smaller minimum step size have shown that any effect that this has on the results is the same order as the noise in the measurements attributable to intercluster variability [24].

I. FRACTAL DIMENSION AND APPROACH TO SCALING

We calculate the effective value of fractal dimension from the local slope of the average radius of deposition vs number of particles N according to

$$D=\frac{\ln(N_2)-\ln(N_1)}{\ln(R_{dep}(N_2))-\ln(R_{dep}(N_1))}, \tag{2}$$

where the properties are measured at two different cluster sizes N_1 and N_2. For the clusters grown, the value of the fractal dimension is shown in Fig. 2. The dimensions estimated for each value of A appear to be converging to a common value of $D=2.50\pm0.01$ which is consistent with previous computational estimates [13]. The data for $A=0.03$ and $A=0.01$ are less well converged, and the results could be made more accurate with data for larger N.

Plotting the fractal dimension data against radius R_{dep} gives a much more coherent convergence of the noise reduced growth data, as shown in the lower panel of Fig. 2. Thus it appears that the convergence of the fractal dimension in noise reduced growth is primarily governed by the radius attained: we suggest this is simply reflects the need for noise reduced clusters to develop sufficient branching structure. The value of radius required (of order 10 particle diameters for accurate D) is qualitatively reasonable and we discuss a refinement of this below in relation to angular fine structure. The approach of D to scaling also exhibits some dependence of amplitude on the level of noise reduction, but this is comparatively weak. For all noise reductions (including none) the variation of $D-2.5$ with radius is roughly consistent with a power law, $D-2.5\propto R_{dep}^{-1.5}\propto N^{-0.6}$ for $\ln(R_{dep})>1$. It is important that this is a relatively rapid convergence to scaling, much more rapid than the slow corrections to scaling discussed below.

II. CRITICAL AMPLITUDES AND THEIR CORRECTIONS TO SCALING

We now consider the approach to fixed point behaviour for DLA in three dimensions, in terms of dimensionless quantities which can be directly measured (as opposed to being inferred by differentiating measured data). In two dimensions it has been shown that no property of the cluster has a slower correction to scaling than the relative penetration depth and suggested that all properties should show influence of this slowest correction [7].

To measure the slowest correction to scaling exponent of a quantity, we used differential plots which proved effective for DLA in two dimensions [12]. For some converging quantity Q_N, which displays a single correction to scaling [Eq. (1)], then

$$\frac{dQ_N}{d\ln(N)} = -\nu(Q_N - Q_\infty), \tag{3}$$

so a plot of $dQ/d\ln(N)$ against Q should exhibit a straight line with slope $-\nu$, intercepting the x axis at the asymptotic value Q_∞. We approximated the differential by

$$\frac{dQ}{d\ln(N)} \simeq \frac{Q_{N_2} - Q_{N_1}}{\ln(N_2/N_1)} \tag{4}$$

and its statistical error by

$$\sigma\left(\frac{dQ}{d\ln(N)}\right) \simeq \frac{\sqrt{\sigma^2(Q_{N_2}) + \sigma^2(Q_{N_1})}}{\ln(N_2/N_1)}, \tag{5}$$

where $\sigma(Q_N)$ is the standard error in Q_N, and the differentials are plotted against the interpolated value $Q=(Q_{N_2}+Q_{N_1})/2$.

The relative penetration depth is simply the relative spread of radius over which walkers are deposited, at fixed cluster size, computed as standard deviation of radius of deposition divided by its mean. The application of the above differential analysis to this ratio is shown in Fig. 3. The key feature is the universal linear asymptote, corresponding to limiting values of the relative penetration depth and its leading correction to scaling index, independent of the level of noise reduction. As in two dimensions the limiting value is very modest, $\xi/R_{dep}|_\infty = 0.122 \pm 0.002$, with the result that different moments of the deposition distribution give very similar mean radii.

The slope of the common asymptote in Fig. 3 gives a correction to scaling exponent $\nu = 0.22 \pm 0.03$. This corresponds to a much slower correction to scaling than the effects in the fractal dimension plots discussed in the preceding section, and as a result it is asymptotically dominant over those. The asymptotic dominance of the slow correction to scaling is also directly confirmed by noting that the corresponding points in Fig. 3 come from radius $R_{dep} > 10$: this is clearly inside the regime where the fast corrections cease to be important in D.

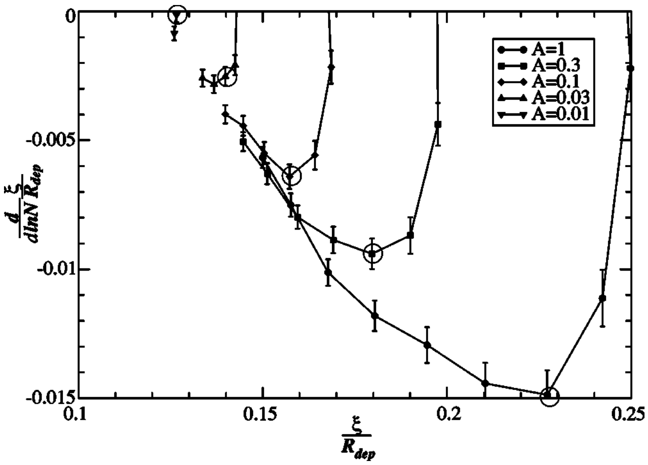

FIG. 3. Differential plot of relative penetration depth ξ/R_{dep} (horizontal scale) against its own derivative with respect to ln N (vertical scale). The intercept with zero derivative indicates the asymptotic value of ξ/R_{dep} for infinite N is given by $\xi/R_{dep}|_\infty = 0.122 \pm 0.002$ and the common limiting slope of the plots indicates a correction to scaling exponent of $\nu = 0.22 \pm 0.03$. Growth at different levels of noise reduction $A \geqslant 0.03$ is consistent with universal values of asymptote and exponent, while $A = 0.01$ appears to start and remain close to the "fixed point" value of ξ/R_{dep}. The circled points correspond to the first value of radius above 10 for each curve, being $R_{dep} = 11.0$, 14.1, 12.7, 11.3, and 10.5 (in order of increasing noise reduction from $A = 1$), showing how the onset of universal correction to scaling is primarily set by radius. The corresponding values of N are given by $N = 562$, 5620, 17800, 56200, and 178000 and along each curve points are spaced by factors of $\sqrt{10}$.

In the regime dominated by it, the slow correction to scaling in the relative penetration depth is reduced in amplitude by increasing noise reduction, even comparing at fixed N. This can be seen in Fig. 3 by comparing the leftmost point of each curve, corresponding to $N = 10^6$ walkers per cluster in each case. This is in contrast to the fast corrections to scaling exhibited in the fractal dimension D, which largely track radius and hence require more walkers at a given level of noise reduction to fall below a given threshold. The combination of these two observations leads to the least noise reduction ($A = 1$) giving us the clearest plot and slope for the slowest correction to scaling and its exponent: this is the case where the slowest correction has largest amplitude and where the fast correction to scaling is least obtrusive.

To more fully characterise the DLA clusters we measured the multipole powers, since the corresponding multipole moments provide an orthogonal set which may be used to totally describe the growing properties of the clusters. In three dimensions the multipole moments are estimated by (see [25], Chap. 4)

$$q_{l,m} = \frac{1}{n}\sum_{i=1,n} r_i^l Y_{l,m}(\theta_i, \phi_i) \tag{6}$$

using n probe particles which contact the cluster at (r_i, θ_i, ϕ_i) for $i = 1$ to n. We normalized the multipole power as

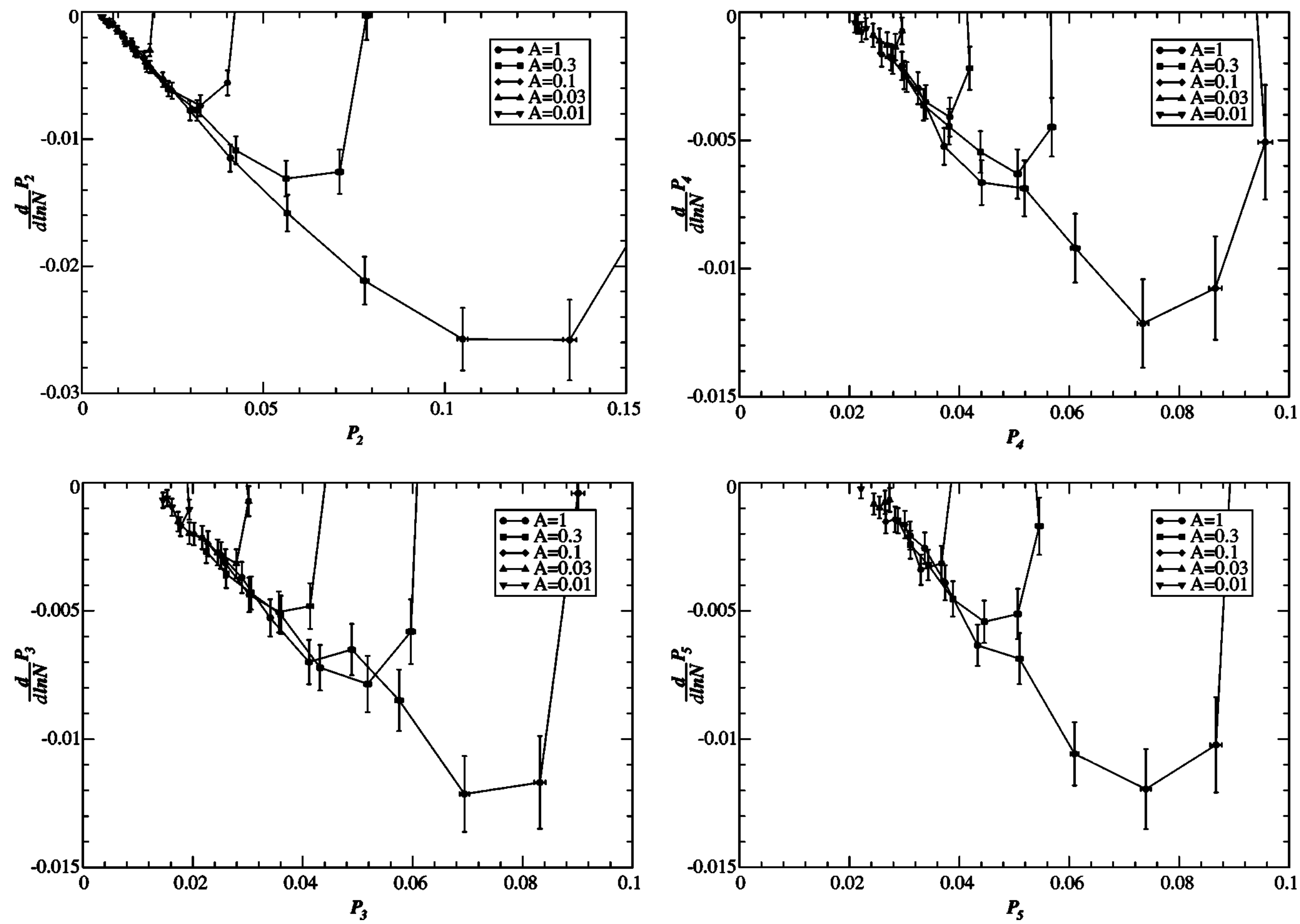

FIG. 4. Differential plots for the first four (nontrivial) multipole moments $P_2 - P_5$. All of the plots exhibit universal asymptotic values, corresponding to the extrapolation to zero derivative, and universal limiting slope corresponding to their correction to scaling exponent. The correction to scaling exponents were estimated by eye as $\nu(P_2)=0.32\pm0.02$, $\nu(P_3)=0.24\pm0.03$, $\nu(P_4)=0.26\pm0.06$, and $\nu(P_5)=0.29\pm0.05$, where the errors represent the maximum believable error.

$$P_l = \frac{\sum_{m=-l}^{l} |q_{l,m}|^2}{(2l+1)R_{\mathrm{eff}}^{2l}}, \tag{7}$$

where the effective radius is in turn given by

$$\frac{1}{R_{\mathrm{eff}}} = \frac{1}{n}\sum_i \frac{1}{r_i}. \tag{8}$$

The definition of R_{eff} is such that it gives the radius of spherical target of equivalent absorption strength to the cluster, constituting a natural choice of radius relating to the zeroth multipole sector.

Note that for each measurement we used the center of charge as origin, meaning zero dipole moments and hence $P_1 = 0$; otherwise there is confusion between cluster shape and drift of the cluster center (albeit the latter is rather negligible).

The first and important feature of our differential plots is again the indication of limiting asymptotic values Q_∞ [corresponding to $dQ/d\ln(N)=0$] and approaching slopes which are universal, independent of the level of noise reduction. This is shown for the multipole powers $P_2 – P_5$ in Fig. 4, and also for the relative variability (between clusters at fixed N) of extremal cluster radius in Fig. 5. The universality of the asymptotic values of all these plots is strong indication that the limiting distribution of cluster shape is universal.

The slopes of these same plots indicate the correction to scaling exponents, which also appear to be universal with respect to the noise reduction. Figure 6 shows the measured values of the correction to scaling exponents for each of the quantities plotted (and all multipole moments measured). The exponent for the quadrupole power P_2 is significantly different from the exponents for ξ/R_{dep} and P_3. There is no quantity which shows a slower correction to scaling than ξ/R_{dep}, which suggests that the result found by Somfai *et al.* [7] in some sense also applies to clusters grown in three dimensions.

The values of the correction to scaling exponents are least precise for the highest multipole moments, as these are most sensitive to the fine structure of the cluster. Intriguingly, the asymptotic value of the relative penetration depth for DLA clusters in three dimensions is equal within measurement error to that of the two dimensional case: see Fig. 3 here and Fig. 3 in [12]. The correction to scaling exponent for ξ/R_{dep} is around $\frac{2}{3}$ the value of the exponent for clusters grown in

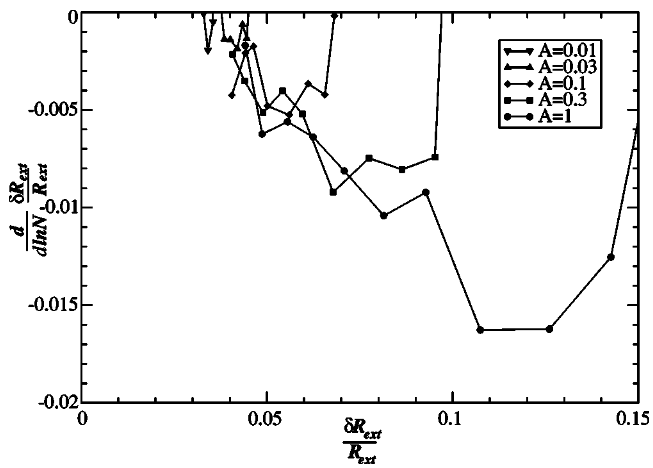

FIG. 5. Differential plot of the relative fluctuation in extremal radius, $\delta R_{ext}/R_{ext}$. The asymptotic value is $\delta R_{ext}/R_{ext}|_{\infty} = 0.032 \pm 0.004$ which leads to an estimate of the fixed point noise reduction of $\epsilon^* = 0.0064 \pm 0.0016$.

two dimensions, indicating that DLA clusters in three dimensions are considerably slower to mature.

III. RESOLUTION OF ANGULAR STRUCTURE

We have also made a limited analysis of how the multipole powers depart from universal correction to scaling behaviour at smaller N. For the differential plot of each multipole power, at each value of noise reduction $A=0.03$ to $A=1$, we identified the first point lying on the universal curve. Quantified in terms of the corresponding cluster radius R_l these vary systematically with the multipole index l, as shown in Fig. 7. Their dependence on A is weak and below the (substantial) scatter, so values from different A have been combined statistically at each l. (For the highest level of noise reduction $A=0.01$ and $l \geqslant 6$, the differential plots generally approach the fixed point from the opposite direction, making it hard to decide R_l consistently; we therefore excluded $A=0.01$ from this analysis.)

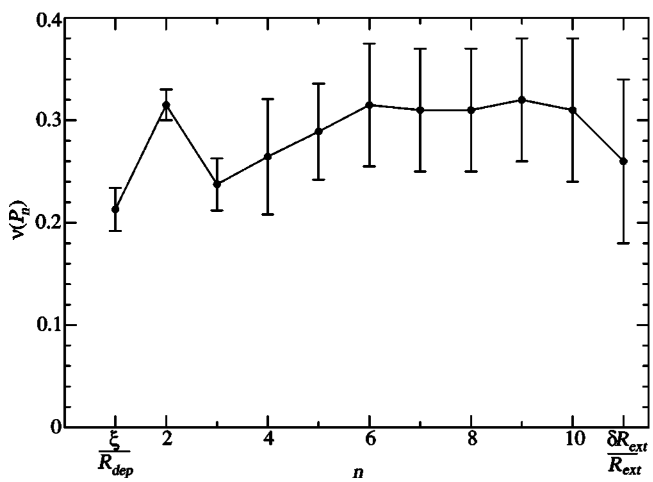

FIG. 6. The correction to scaling exponents obtained from differential plots of different multipole powers, and the two other quantities marked. The exponent for the quadrupole power P_2 is significantly different from the exponents measured for ξ/R_{dep} and P_3.

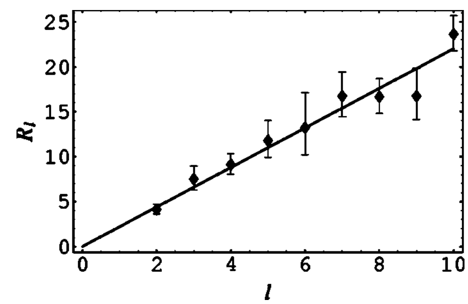

FIG. 7. For each multipole power we identified radius R_l where its differential plot became dominated by the universal slow correction to scaling. This judgement was made by eye separately for each value of noise reduction $A=1$ to $A=0.03$, and the error bars shown correspond to error in the mean of $\ln R_l$ when separate values from different noise reduction values were combined statistically. The dependence on noise reduction level was weak. The guideline shows the consistency of the data with a simple linear dependence of R_l on the multipole index l, explained in the text by a simple spatial resolution threshold.

The plot of R_l in Fig. 7 is clearly consistent with a simple linear dependence on l, and this in turn has a simple interpretation in terms of the smallest cluster radius at which the features probed by the l'th angular harmonics can be supported. The cluster cannot support features finer than the particle size, which at radius R corresponds to angular scale $1/R$, while angular harmonics of order l are sensitive to features of angular scale $1/l$. Matching these leads to $R_l \propto l$ in agreement with our data.

Thus we are again led to the conclusion that the regime of universal correction to scaling sets in once the cluster is large enough to support sufficient ramification. The thresholds of R seen for the convergence of the fractal dimension and the relative penetration depth correspond to the threshold on R seen for P_3. While this is at first sight rather low, it is reasonable when one notes from Fig. 1 that these clusters have relatively few major arms, equivalent to their major angular features being at large angles.

IV. FIXED POINT NOISE REDUCTION

It is clear from differential plots such as Fig. 3 that the noise reduction "controls" the slowest correction to scaling. For low values of A this correction to scaling is strongly reduced, and we may write the behavior of this correction as follows:

$$Q_N = Q_\infty + C_Q(A) N^{-\nu}. \tag{9}$$

Other corrections to scaling need not depend on A, but it is quite evident that the amplitude of the slowest correction to scaling is strongly affected by it. If there is some value of A for which all $C_Q(A)$ are zero, then this value of the noise reduction would correspond to the fixed point of a renormalization scheme (see [10]). The plots for P_2–P_4 suggest that the fixed point noise reduction is $A^f < 0.01$ and plots of

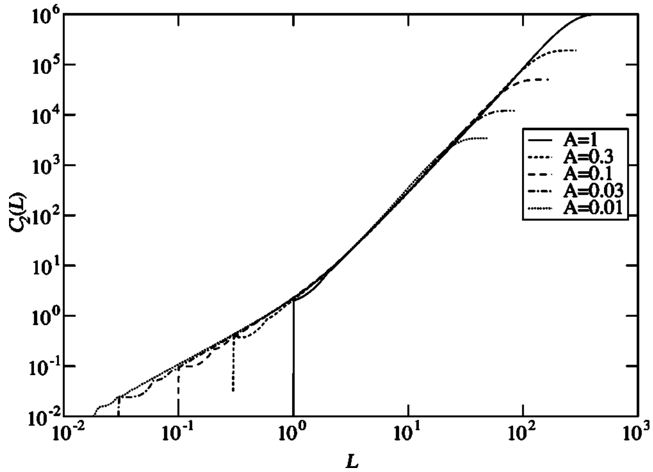

FIG. 8. The two point correlation function for clusters grown in three dimensions, scaled by effective noise reduction factors, ϵ^{eff}. This noise reduction is chosen so that a data collapse is seen for small L.

$\xi/R_{dep}, P_5$ are unclear as to the value of the fixed point noise reduction. Hence, one estimates that

$$A^f \leqslant 0.01. \tag{10}$$

The noise reduction of the fixed point can also be estimated from the asymptotic properties of DLA clusters using the Barker and Ball formula [10]

$$\epsilon^* = D^2 \left(\frac{\delta R_{ext}}{R_{ext}} \right)^2, \tag{11}$$

where R_{ext} is the extremal cluster radius (the radius of the furthest cluster particle from the seed particle) and δR_{ext} is the cluster to cluster variability of R_{ext}. This was originally cast in terms of the variability of N at fixed radius R_{ext} and in two dimensions it was shown to be important to tune out lattice anisotropy [10], yet subsequent work restricted to just 2×2 renormalization cells (and with uncontrolled lattice effects) claimed fixed point noise reduction values of order unity [26]. More recently direct off-lattice measurements using Eq. (11) were shown to give $\epsilon^* = 0.0036 \pm 0.0006$ in two dimensions [12], in agreement with the trend of values in [10]. For off-lattice DLA in three dimensions, we now have from Fig. 5 that the asymptotic value of the relative variability of extremal cluster radius is $\delta R_{ext}/R_{ext}|_\infty = 0.032 \pm 0.004$. By the methodology of reference [12] this leads to

$$\epsilon^* = 0.0064 \pm 0.0016, \tag{12}$$

in three dimensions, which is consistent with the estimated value of A^f.

For a noise reduction of A, one would naively assume that it would require of order N/A particles to grow a cluster with the same radius as a non-noise reduced cluster of N particles. Our data below show that this is a systematic underestimate, so that each value of A corresponds to a more severe noise reduction than expected. Figure 8 shows the two point correlation function for DLA clusters grown at different noise reductions. The graphs have been shifted vertically so that all the curves collapse to a single line. From the shift factors used, we estimate the effective noise reductions to be

$$\begin{aligned} A = 1, &\quad \epsilon^{\mathrm{eff}} = 1, \\ A = 0.3, &\quad \epsilon^{\mathrm{eff}} = 0.19, \\ A = 0.1, &\quad \epsilon^{\mathrm{eff}} = 0.05, \\ A = 0.03, &\quad \epsilon^{\mathrm{eff}} = 0.012, \\ A = 0.01, &\quad \epsilon^{\mathrm{eff}} = 0.0034. \end{aligned} \tag{13}$$

Hence one concludes that the fixed point noise reduction in Eq. (10) should be adjusted to

$$\epsilon^f \leqslant 0.0035. \tag{14}$$

The values for ϵ^f and ϵ^* differ by a factor of 2, demonstrating that the identification process is open to some errors. If, as indicated by the results in Fig. 6, a single slowest correction to scaling exponent does not control the scaling of all parameters, then a theoretical renormalization scheme would need more than one parameter to match this behavior. If more than one parameter is indeed relevant, then our arguments to match ϵ^f and ϵ^* based on noise reduction alone is certainly not expected to be perfect.

V. CONCLUSION

The growth off-lattice of noise-reduced diffusion-limited aggregates in three dimensions has been considered and shown to exhibit universality with respect to noise reduction. The fractal dimension is found to be $D = 2.50 \pm 0.01$ which agrees with previous computational [13] and mean field [27] estimates. The penetration depth scales with the radius, and the asymptotic value of the relative penetration depth is $\xi/R_{dep} = 0.122 \pm 0.002$ which overlaps the value found for clusters grown in two dimensions [12]. The convergence of the multipole powers provides a very strong indication that DLA cluster growth, in three dimensions and off-lattice, converges to a universal distribution of cluster shapes.

The relative penetration depth exhibited the slowest correction to scaling, $N^{0.22\pm0.03}$. Some multipole powers and also the relative fluctuations in extremal radius exhibited correction to scaling exponents which could be consistent with the same. However not all quantities exhibit the influence of the slowest correction to scaling and in particular the convergence to scaling of the dipole power, P_2, appears significantly faster than that of either ξ/R_{dep} or P_3.

Reducing the input noise by factors up to 100, by growing clusters in shallow bumps, clearly reduces the amplitude of the leading correction to scaling. This supports in three dimensions the idea of Barker and Ball [10] that the intrinsic fluctuation level is the physical origin of that slowest correction to scaling. We estimated the fixed point noise reduction to be $\epsilon^f \sim 0.0035$ and this is close to the value estimated using the Barker and Ball [10] formula in terms of relative fluctuation in extremal radius.

All of the above discussion relies on growing clusters beyond the influence of much faster corrections to scaling with $\nu \simeq 0.6$, as most dominantly observed in the fractal dimension. These have little sensitivity to noise reduction when plotted in terms of the radius of growth. This and the variation of their relevance on the order of multipole power studied is consistent with the trivial requirement that the cluster must grow large enough to support the required scales of ramification.

Taken together our results support the hypothesis that isotropic DLA in three dimensions approaches a simple ensemble of statistically self-similar clusters, with a rather slow approach to scaling which is associated with the level of local geometric fluctuation. From this point of view, a quantitative model of the convergence of that fluctuation level appears to be the outstanding challenge in understanding isotropic DLA (in any dimension). Another important challenge for three dimensions, for which work is in progress, is the role which material anisotropy can play [28].

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