Variability of the form and of the harmonic measure for small off-off-lattice diffusion-limited aggregates

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The harmonic measure on off-lattice diffusion-limited (DL) aggregates is defined in detail. The formulation requires the continuum Laplacian and the regularization of divergences in the density of the measure due to folds between atoms. An ultraviolet cutoff is needed for numerical simulations, but in this *off-off-lattice* formulation, the cutoff can be varied at will. The cutoff's effects on the distribution of the Hölders α is studied. We find that the right-hand tail of the distribution of α is very dependent on the lower lattice cutoff. However, the cutoff *does not* effect the transformations that collapse the distributions corresponding to clusters of different sizes. The shapes of off-lattice DL aggregates giving rise to extremely small minimal harmonic measures are illustrated. We discuss the notion of the smallest harmonic measure in an ensemble of clusters, and also the scaling properties of the distribution of α on small clusters.

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I. INTRODUCTION

On diffusion-limited (DL) aggregates [1], and for that matter on the boundary of any two-dimensional domain, the harmonic measure is affected by local geometry. In particular, the smallest harmonic measures are related to the shapes of the "fjords" by the Beurling equality [2–4]. From this equality, but also more directly from the scale invariance of the Laplace equation, it follows that a fjord's Euclidean depth affects but does not determine how small the harmonic measure is at its bottom. It is also known that the number of fjords of given Euclidean depth increases with decreasing depth [5]. However, the measure in a small fjord depends strongly on details of the construction of DL aggregates. In the case of lattice diffusion-limited aggregation (DLA), the constraints due to the lattice drastically limit the possible morphologies of the clusters, especially on smaller scales. For example, the lattice introduces a small-wavelength (ultraviolet) cutoff in the potential field, whose effects have never been studied properly. This paper shows that this cutoff has a significant influence on the distribution of the harmonic measure, and therefore, on the right-hand side of the $f(\alpha)$ curve that corresponds to the negative moments [6–11]. When the boundary of a domain is linearly self-similar [12,13], the $f(\alpha)$ curve is known to provide a quantitative description of the rules of self-similarity of the harmonic measure. In DLA, the right-hand side of the $f(\alpha)$ does not exist. But the failure of the $f(\alpha)$ to exist does not necessarily imply that the measure is not geometrically self-similar [10]. Instead of $f(\alpha)$, new limit distributions and their corresponding collapse rules may be needed to describe the rules of self-similarity [11].

We have argued elsewhere that the observed sample variability of the small probabilities and also the large variability within individual clusters are due to fluctuations in cluster geometry [2,4]. These fluctuations are severely affected when the DL aggregate is grown on a lattice. The cutoff most severely restricts the smaller scale fluctuations, and thus most significantly affects the smaller clusters. However, our findings suggest that the effects of the cutoff do not decrease with cluster size.

Therefore a much more careful model of DLA is needed if one wishes to study the effects of the lattice cutoff on geometric fluctuations, and in particular, on the right-hand tail of the distribution $f(\alpha)$. The model we use is a combination of two well-known concepts: off-lattice DLA [14] and the continuum Laplacian potential. We will refer to this model as *off-off-lattice DLA*, to indicate that the Brownian motion of the atoms proceeds off lattice, and that the harmonic measure is derived from the (off-lattice) continuum Laplacian.

II. OFF-OFF-LATTICE DLA

Consider off-lattice DLA in two Euclidean dimensions. The center of a disk of diameter Δ, called an *atom* or particle, performs a Brownian motion until its perimeter intersects the perimeter of one of the disks forming a cluster. At that instant, the moving atom becomes part of the cluster, and a new Brownian atom is released at a randomly chosen point away from the cluster. An example of an eight-particle cluster is shown in Fig. 1.

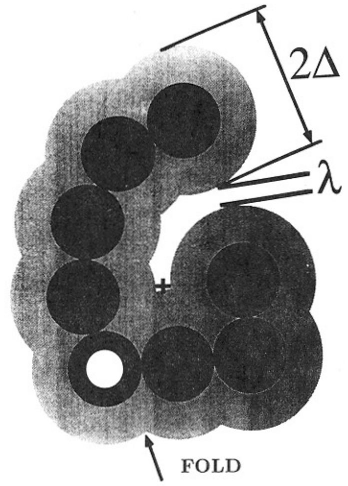

FIG. 1. The black disks of diameter Δ are the atoms forming the cluster. The seed atom is marked in white. A new Brownian atom will stick to this cluster when its center touches the boundary of the gray Δ neighborhood. This set is the union of disks of diameter 2Δ at the centers of the atoms in the cluster. The harmonic measure is supported by the Δ perimeter, and in this particular case, it attains its smallest density near the cross. Since the opening λ of the fjord connecting the cross to the outside can be arbitrarily small, the Laplacian potential should be known with infinite spatial precision. See also Fig. 4.

Kakutani [15] established a relation between Brownian motion and Laplacian potentials. He studied the probability of a Brownian *point* hitting any point in a subset of the perimeter of an off-lattice DL aggregate in the Euclidean plane, and showed that it equalled the harmonic measure of that subset. The density of this measure is the normalized Laplacian charge density on the perimeter of the cluster; it is proportional to the gradient of the Laplacian potential on the boundary, i.e., to the electric field.

Let us recall that, when the cluster is viewed as a collection of atom centers, its Δ *neighborhood* (Fig. 1) is the set formed by replacing these centers by disks of diameter 2Δ. The Δ *perimeter* is the external boundary of the Δ neighborhood; that is, the boundary accessible from infinity. Thus the probability of the perimeter of a Brownian *atom* of diameter Δ intersecting the perimeter of the cluster is given by the normalized harmonic measure on the Δ perimeter of the cluster.

In the square lattice Laplacian formulation of DLA [16,17], the Laplace equation is solved on a lattice with a spacing equal to the side of the square atoms (or bonds) forming the cluster. Solving the Laplace on a much finer lattice than the particle "diameter" would at best yield more attractive graphics. The above definition of a continuum DL aggregate shows, however, that the atom size does not (by any means) set an upper limit to the accuracy to which the harmonic measure needs to be known. Figure 1 shows that, ideally, the position of atom 8 should be known with infinite precision, because λ can take any value ≥ 0. The Laplacian potential should also be known with infinite spatial accuracy; that is, the continuum Laplacian potential is needed. In practice (e.g., in the numerical simulations to be discussed below), a finite precision in both these quantities results from, and roughly equals, the lower cutoff σ that must be imposed on the Brownian step lengths. We shall refer to σ as the *sticking distance* or sticking precision, and measure it, and all other distances, in units of atomic diameter $\Delta=1$.

In contrast with on-lattice DLA, off-off-lattice DLA has the following features: (1) the number of distinct N-atom ($N>2$) configurations is infinite, (2) there is no lattice anisotropy, (3) (as exemplified by the configuration in Fig. 1) the smallest possible harmonic density in clusters of approximately five or more atoms is 0^+, and the maximum density Hölder is ∞ (see below), and (4) in on-lattice DLA, *only* the larger scale features of the larger fjords are more or less independent on the lattice. So, in small on-lattice DLA, both the constraints on the possible cluster configurations which are due to the lattice, and those more genuine ones, which are due to the small number of atoms, are mixed. On off-off-lattice DLA, the only finite size effects are due to the small number of atoms: therefore we expect the asymptotic behavior to transpire more rapidly.

A. Numerical simulation and regularization

Our numerical simulation of off-off-lattice DLA grows off-lattice clusters using Brownian atoms that take off-lattice steps of size σ in units of atomic diameter Δ. An atom sticks to the cluster when its center steps within a distance $\leq\sigma$ of the Δ perimeter.

The cluster is then embedded in a square lattice with lattice constant δ; that is, the diameter of an atom is $1/\delta$ lattice units. On this lattice, the Laplacian potential is estimated using an iterative numerical procedure. At the point marked by a cross in Fig. 1 the harmonic measure will be reliable, if and only if the sticking precision σ is smaller than the smallest neck width in the fjord. If the lattice constant is larger than the sticking precision, that is, $\delta>\sigma$, the harmonic measure at the bottom of a fjord with neck widths smaller than σ is automatically excluded from the geometric support of the measure. Thus, when $\delta>\sigma$, the accuracy of even the smallest positive values of the measure is thus guaranteed. The majority of our simulations use $\sigma=0.01$ and $\delta=0.1$.

Let the Δ perimeter $\gamma(t)$ of an off-lattice cluster be parametrized by arclength, so that $\gamma(0)=\gamma(l(N))$, where $l(N)$ is the Euclidean length of this perimeter. In [13], we have argued in detail that the study of the scaling properties of the harmonic measure on growing fractal boundaries should be based on the density Hölder defined by $\alpha(t)=-\ln d\mu(t)$, where $d\mu(t)$ is the harmonic density at perimeter site $\gamma(t)$. However, at the folds in $\gamma(t)$, see Fig. 1, the harmonic density is 0, so that $\alpha=\infty$. In order to avoid the resulting anomalous contributions, we consider the *ε-regularized density*

$$d\mu_i(\epsilon)=\frac{1}{\epsilon}\mu\left[i-\frac{\epsilon}{2\delta},i+\frac{\epsilon}{2\delta}\right],$$

with $i=1,\ldots,l(N)$. Large density Hölders due to the presence of folds disappear for ϵ equal to, e.g., 0.1 times the circumference 2π of the Δ perimeter of a single particle. On the other hand, the genuine large α's due to screening by fjords of Euclidean depth larger than ϵ will remain.

Numerically, it does not make sense to take $\epsilon<2\delta$, since the Laplacian potential is only known for spatial resolutions larger than δ. In these cases, the regularization is provided by the finite cutoff δ of the underlying lattice. The "site probabilities" usually used in the study of the growth probability distribution on DLA are a special case, which may be referred to as lattice regularization; in this case, the lattice constant, regularization constant, and sticking distance are all equal to the particle diameter: $\delta=\epsilon=\sigma=\Delta$.

III. GEOMETRIC FORM OF SAMPLES WITH EXTREME HARMONIC DENSITIES

For a fixed lattice constant, the smallest harmonic density is found when a slit of width 2δ is encountered in a cluster whose length is approximately half the number of its atoms. These configurations occur with very small probability, and the ensemble sizes needed to sample them increase rapidly with increasing number of atoms. Figure 2 illustrates the three- to eight-atom configurations which give rise to the smallest harmonic density out of ensembles of 800 clusters. The Laplacian potential was estimated with lattice constant $\delta=0.2$. Even though the ensemble size is not large enough, it is apparent that the smallest density is carried by a growing slit. (The eight-atom cluster in Fig. 1 occurred in a larger ensemble.)

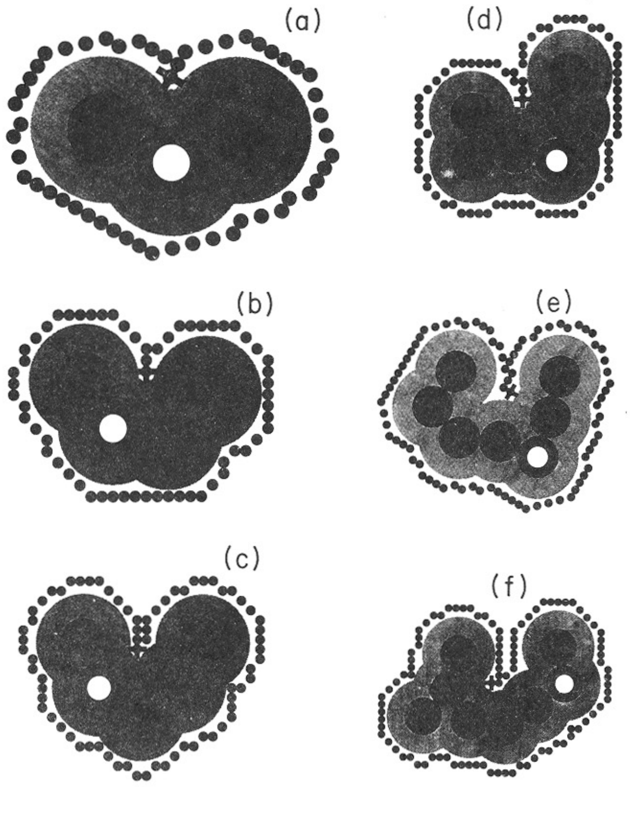

FIG. 2. The configurations having the smallest harmonic densities out of ensembles of three- through eight-atom clusters. Each ensemble contains 800 clusters for which the Laplacian was estimated using a square lattice with spacing $\delta=0.2$. In the presence of such a lower cutoff on the spatial precision of the Laplacian, the smallest density occurs in slit-like morphologies.

However, it should be kept in mind that the notion of a specific morphology, giving rise to the smallest harmonic density, loses its meaning in the continuum limit of the Laplacian for $\delta\to 0$. This is because small densities can be due to either elongated fjords, very narrow necks, or a combination of the two.

Figure 3 juxtaposes the configurations of smallest harmonic density corresponding to large ensembles of clusters of different sizes N. The neighborhood with the smallest density is marked with a cross. The smallest neck in the fjords bearing these small densities is the same in all the clusters, because the size of the grid used to estimate the potential was $\delta=0.1$ in all cases. Such a grid does not allow the estimation of the potential for necks smaller than 1% of the atomic diameter. This is illustrated in Fig. 4 for a 32-atom configuration; all the sites of the lattice used in the estimation of the harmonic density around the Δ perimeter of the cluster have been drawn. Clearly, this estimate of the density is completely inadequate in the upper-right fjord. The contribution of this cluster to the right-hand side of the distribution of α would have been very different had δ been somewhat smaller.

Figures 3 and 4 also illustrate the enormous sample variability of the smallest harmonic measures. In a run of 26 000 clusters of 32 atoms, we found the values of the smallest density to be scattered between 10^{-4} and 10^{-28}. The larger clusters in Fig. 3 may look like "typical" DL-aggregate clusters, but the distribution of their harmonic measure is not "typical."

IV. THE DISTRIBUTION OF THE HÖLDERS α

Denote by $P(\alpha)d\alpha$ the probability that the density Hölder of a randomly picked point on the Δ perimeter of one of the clusters in the ensemble lies between α and $\alpha+d\alpha$. In Fig. 5, $\ln P(\alpha)$ is plotted against $\ln(\alpha)$ for ensembles of eight-atom clusters with different values of the ultraviolet lattice cutoff δ. The values of the lattice constants are $\delta=0.2$, 0.1, 0.05, and 0.025, and the ensemble sizes are 32 600, 150 300, 10 350, and 240, respectively. The dependence of the right-hand tail on δ is perspicuous. The straightness of these tails seems to suggest that $P(\alpha)\sim\alpha^{-x}$. The estimated values of the slopes x are 8.0, 6.7, 6.2, and 5.5 for the respective values of δ. The fact that $\epsilon=2\pi/10$ excludes the notion that the increase in x is due to a better articulation of the folds. Neither do the reported slopes and the behavior of the tails depend on the ensemble sizes, except (of course) for the far-right ends. This dependence on δ seems to be due to the above-illustrated fact that fjords with increasingly narrow subatomic necks are sampled for decreasing δ.

To find a theoretical upper bound of this behavior of x, we replace the ensembles of size n of N-particle clusters by slits of width θ and length $L=N/2$. Since the DLA growth rules do not favor closed loops, the most extreme

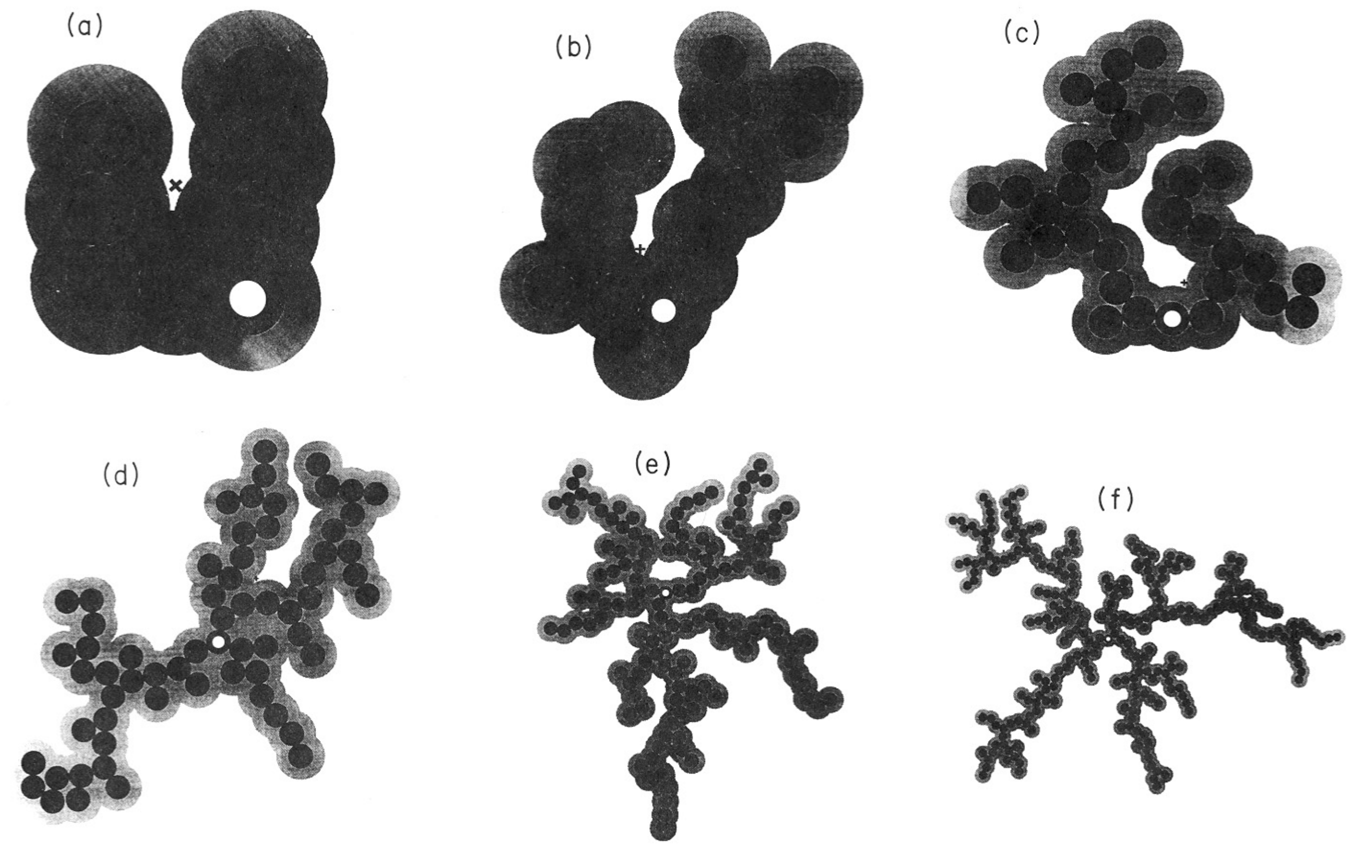

FIG. 3. Configurations bearing the smallest harmonic densities out of large ensembles of clusters with 8, 16, 32, 64, 128, and 256 atoms. The harmonic measure has been estimated with lattice spacing $\delta=0.1$. The ensemble sizes are 150 000, 42 200, 26 000, 10 000, 5000, and 1000, respectively. The orders of magnitude of the densities are 10^{-13}, 10^{-21}, 10^{-28}, 10^{-26}, 10^{-28}, and 10^{-30}.

distribution of θ that one could imagine would behave for small θ as $\mathrm{Prob}(\Theta\leq\theta)\sim\theta$ (Θ is a random variable). The harmonic measure at the bottom of such a slit is known to be proportional to $e^{-cL/\theta}$. It follows that $\alpha\sim L/\theta$ which shows that the right-hand tail of the distribution of α is bounded from above by $\mathrm{Prob}(A\geq\alpha)\sim\alpha^{-1}$; that is, $-x\leq-2$.

The tail behavior of this bound is the same as that of the Cauchy distribution. However, it should be noted that there is no obvious relation between the upper bound to the tail behavior of α for small clusters discussed here and the results in [11]. This will be discussed further in the next section.

One might have thought that the contribution of fjords with narrow subatomic necks becomes negligible for larger aggregates. But, in comparing the plots of the probability density $P(\alpha)$ for $\delta=0.1$ and 0.2 in Fig. 6, we

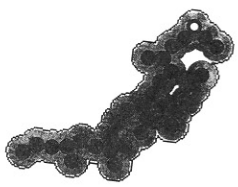

FIG. 4. A 32-atom cluster out of the same ensemble as in Fig. 3. But now we have those with the largest smallest densities, namely, a density of the order 10^{-4}. This illustrates the large sample variability of the small densities. The figure also illustrates the drastic nature of a lower cutoff on the spatial precision of the Laplacian potential. The spatial precision of 1% of the atomic diameter is clearly insufficient to handle the upper-right fjord with a narrow neck. See also Fig. 1.

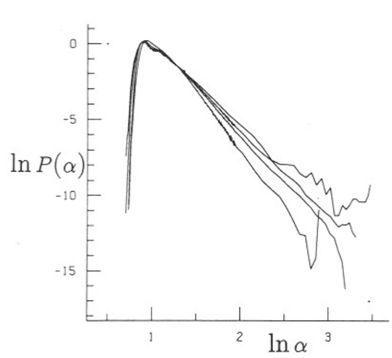

FIG. 5. Plots of the logarithm of the probability density $P(\alpha)$ vs the logarithm of the density Hölder $\alpha=-\ln d\mu$ for eight-atom clusters with different values of the lattice constant δ. The density of the harmonic measure was regularized with $\epsilon=2\pi/10$. Starting from the bottom, the right-hand tails correspond to $\delta=0.2$, 0.1, 0.05, and 0.025. The straightness of these tails seems to indicate that $P(\alpha)\sim\alpha^{-x}$ for eight-atom clusters with $x\approx8.0$, 6.7, 6.2, and 5.5.

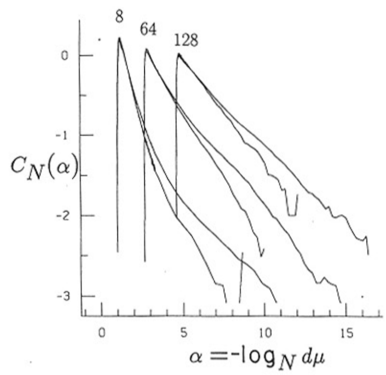

FIG. 6. Study of the effect of an increasing spatial resolution of the Laplacian on the right-hand tail of the distribution of α, for the harmonic measure on clusters of different sizes. From left to right, the pairs of curves are for $N=8$, 64, and 128. For clarity, we shifted the 64 and 128 pairs by 2 and 4, respectively, to the right. The lower-right tail in each of the pairs is for lattice constant $\delta=0.2$ and the upper for $\delta=0.1$. We plotted $C_N(\alpha)=\log_N P(\alpha)$ vs the normalized Hölder $\alpha=-\log_N d\mu$ in order to compare the results for different cluster sizes.

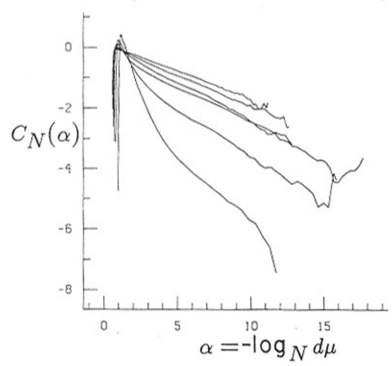

FIG. 7. A Cramèr plot $\log_N P(\alpha)$ vs $\alpha=-\log_N d\mu$, where $N=8,16,32,64,128,256$ are the numbers of particles in the clusters. For a restricted multifractal, the different curves are known to collapse to a single curve which is a linearly translated $f(\alpha)$ curve. The left-hand sides of the curves show a tendency to collapse for increasing cluster sizes. This is in complete agreement with previous results [11]. The right-hand side is not supposed to collapse, because $f(\alpha)$ is left sided.

find no numerical evidence for a decreasing contribution by such fjords for increasing cluster sizes. The pairs of curves in Fig. 6, considered from left to right, correspond to $N=8$, 64, and 128. For the sake of clarity, the 64 and 128 pairs have been shifted to the right by 2 and 4, respectively. From the left to the right tail, the numbers of clusters in the ensembles are $\approx 156\,000$, 33 000, 10 000, 1600, 2000, and 400. In each pair, the lower curve refers to $\delta=0.2$, and the top curve to $\delta=0.1$. A plot of $\log_N P(\alpha)$ versus the normalized Hölder $\alpha=-\log_N d\mu$, which one of us has shown to be a Cramèr plot [11,13,18], is known to give a collapse for the very simplest restricted multifractals, and therefore allows a more realistic comparison between results for different cluster sizes. The dependence of the slopes of the right tails on δ does not seem to diminish with increasing cluster size. This, combined with the fact that the smallest necks (most sensitive to δ) occur most frequently in the shorter fjords and that—independently of the cluster size—these fjords outnumber the larger ones, suggests that the part of the distribution of α on the right-hand side of its maximum is very dependent on the ultraviolet cutoff δ (here we take σ to be 0). The implications for the $f(\alpha)$ distribution are discussed in the next section.

V. MULTIFRACTALITY OF THE HARMONIC MEASURE

Our study of off-off-lattice DLA is primarily motivated by our previous studies of the self-similarity properties of the harmonic measure on DL-aggregate boundaries. However, the clusters studied here are too small to draw conclusions as to the asymptotic properties of the harmonic measure's self-similarity. In our previous study [11] of the multifractality of the harmonic measure on DL aggregates, we gave numerical evidence for the left-sidedness of $f(\alpha)$. We showed that for 3125, 6250, 12 500, and 50 000 particle square-lattice clusters, the left-hand side of the distribution of α collapses under the Cramèr rescaling rule, which is known to collapse the whole distribution for restricted multifractal measures such as the binomial. For DLA, however, the right-hand side is not collapsed by this rule but by a different rule we referred to as "Cauchy rescaling."

For the small clusters discussed here, neither the Cramèr nor the Cauchy rescaling collapse the distributions. This is shown in Fig. 7 for the Cramèr rescaling. There is nevertheless a clear tendency for the left-hand sides of the distributions to collapse. On the other hand, a Cauchy plot of the same data (not shown) tends to collapse the right-hand tails for the larger clusters ($N<256$). Finite size effects could have been expected, and it comes as no surprise that these collapses do not work for small clusters.

The dependence of the right-hand tail of the distribution of α on the ultraviolet cutoff δ, does not affect the applicability of the collapse rules. Plotting the distributions in Fig. 7 for a different value of δ results in a similar behavior, even though the shapes of the right-hand tails differ (see Fig. 6). In fact, we have simulations for many different cluster sizes ($N=3-150\,000$), numbers of clusters ($n=1-100\,000$), values of δ ($\delta=1-0.05$), mostly off off lattice, but also square lattice. When combined, they very clearly show Cramèr collapse for the left-hand sides, and Cauchy collapse for the right-hand tails, independently of δ [19].

Clearly, the ultraviolet cutoff δ is a necessity for numerical simulations. It is, however, not part of the formal definition of off-off-lattice DLA. The strong dependence of the right-hand tail of the distribution of α on δ is artificial, and although we have a qualitative understanding of its origin, a more quantitative understanding is presently lacking. What is encouraging though, is that in establishing the self-similarity of the harmonic measure, one is not primarily interested in the exact shape of the distribution, but in its scaling properties; that is, in the collapse rule.

VI. DECOMPOSITION OF LARGE CLUSTERS INTO UNCORRELATED EIGHT-ATOM CLUSTERS AND THE BEHAVIOR OF THE SMALLEST HARMONIC DENSITIES

We now address the behavior of the smallest harmonic density [2,4,9,10,20–23] in N-particle off-off-lattice DL aggregates. The typical maximum Hölder, $\alpha^{+}_{\max}(n)=\max_n \alpha_{\max}$, which is the value one expects to occur once in an ensemble of size n of eight-atom clusters, is given by $\mathrm{Prob}(\alpha \geq \alpha^{+}_{\max}(n))=1/n$ [9]. Assuming the validity of equation $P(\alpha)\sim\alpha^{-x}$, we find $\alpha^{+}_{\max}(n)\sim n^{1/(x-1)}$. Therefore the $\alpha_{\max}$ is infinite, if taken over an infinitely large ensemble of eight-atom clusters.

Let the "typical" behavior of $\alpha_{\max}$ refer to the behavior of the largest value $\alpha_{\max,\mathrm{typ}}(N)$ of α, expected to occur at least once in any N-particle cluster. A lower bound on $\alpha_{\max,\mathrm{typ}}(N)$ is easily obtained by assuming the N-particle cluster to be an ensemble of $n_8 \approx N/8 \sim N$ *uncorrelated* eight-particle clusters. Then, the smallest harmonic density is expected to be at least smaller than

$$d\mu_{\min} < \exp(-\alpha^{+}_{\max}(n_8)) \sim \exp(-cN^{1/(x-1)}) ,$$

where c is a finite positive constant. This is a lower bound, because the eight-particle clusters forming the N-particle cluster are in fact strongly correlated; otherwise, the large cluster could not possibly be a fractal. This correlation manifests itself in the nesting of fjords within fjords [13], which gives rise to multiple screening, and consequently, lower probabilities.

This lower bound on the "typical" behavior of $\alpha_{\max,\mathrm{typ}}(N)$ corresponds to a stretched exponential decay for the smallest probability. It is also apparent that this exponential decay has nothing to do with the existence of some "typical" fjord shape. It appears in this crude lower bound because of constraints imposed by the cluster size N on the size of the ensemble of smaller clusters. So, in an experiment with one sample of each cluster size, one would expect to see the "typical" behavior. (It should, however, be kept in mind that such an experiment would be very unreliable, because of large sample fluctuations. An example of these fluctuations was given in Sec. III.) However, in the case of infinite ensemble sizes ($n=\infty$), it makes no sense to talk about the behavior of $\alpha_{\max}(N)$ as a function of cluster size N, since the infinite ensemble value of $\alpha_{\max}(N)$ is infinite for all N larger than ≈ 8.

VII. SUMMARY AND CONCLUSIONS

Both very short and very deep fjords contribute to the small growth probabilities in DLA. The geometry of the smaller fjords is severely limited, not only in lattice formulations of DLA, but also in the case where off-lattice grown DLA is forced on lattice for the sake of estimating the Laplacian potential and the harmonic measure. In order to study the effects of the lower cutoff imposed by the lattice, we studied a more careful model of DLA called off-off-lattice DLA, which is based on the continuum Laplacian.

The study of the self-similarity and multifractality of the harmonic measure on clusters grown with this model is based on the density of the harmonic measure, and requires some form of regularization in order to get rid of irrelevant divergences due to folds on the atomic scale. Unfortunately, actual numerical simulations of this model require the introduction of an ultraviolet cutoff, namely, the lattice constant δ. In practice, δ is taken to be smaller than or equal to the diameter of the atoms.

In the study of the scaling properties of the harmonic measure, the principal quantity is the density $d\mu$ of the measure, and the normalized Hölder $\alpha=-\log_N d\mu$. Since the density is, in principle, independent of the lattice constant, it is, in theory, clear how one should compare data obtained with different values of δ, for the same number of particles. However, for DLA, our results suggest that the right-hand tail of the distribution of α strongly depends on the spatial precision δ of the Laplacian. This by no means implies that the behavior for different cluster sizes of the right-hand tail of the distribution of α is ambiguous and ill defined. As we have shown here, it is indeed true that the exact shape of the tail depends on the value of the ultraviolet cutoff. For that matter, one should avoid drawing conclusions from the shape of the tail. Instead one should concentrate on the scaling properties of the distribution; that is, the rules that collapse the distributions on clusters of different sizes. For those rules, the dependence of the right-hand tail on δ does not seem to matter much. As reported in [19], the collapse rules seem to apply simultaneously to data for different values of δ.

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