RAPID COMMUNICATIONS

PHYSICAL REVIEW A VOLUME 44, NUMBER 2 15 JULY 1991

Exact results on exponential screening in two-dimensional diffusion-limited aggregation

Robin C. Ball and Raphael Blumenfeld
Cavendish Laboratory, University of Cambridge, Madingley Road, Cambridge, CB3 0HE England
(Received 8 January 1991)

We present an exact expression for the rate of screening with time of an arbitrary point on a growing diffusion-limited aggregate, and use it to study the multifractal singularities α that correspond to strongly screened sites. We find that the time evolution of these singularities is controlled by the field at the lips (the outer corners) of the fjord. We show quantitatively that if the time evolution of the strongest singularity α_{max} is self-consistent with the growth process, the key issue is the process by which long fjords are generated. We analyze this process and find an asymptotic linear slope for the decreasing part of $f(\alpha)$. This form agrees with recent measurements [Barabasi and Vicsek, J. Phys. A **23**, L729 (1990)] and excludes an infinite value of α_{max}.

The growth of many structures in nature is limited by diffusive processes. Examples include electrodeposition, bacterial growth, colloidal aggregation, etc. It is a decade since the model of diffusion-limited aggregation (DLA) has been introduced,[1] but in spite of the intensive study that this model attracted, only limited analytical progress has been made towards a complete explanation of the resultant pattern. In DLA the harmonic measure, which is the distribution of flux $\partial_n\Phi$ at an equipotential surface $\Phi=0$, is of particular interest as it governs the growth of the structure. The first incisive discussion of the distribution of Φ was given in two complementing works[2] for two-dimensional (2D) DLA, using a formalism anticipated by Mandelbrot.[3] It was suggested that a number $n(p)\sim(R/a)^{f(\alpha)}$ of regions of size a have "measure" $p\sim(R/a)^{-\alpha}$, where the measure is the total integrated flux of particles into the region. This description enables one to characterize the moments of this distribution, $M_q\equiv\sum_p n(p)p^q$, by a single function $f(\alpha)$, termed the multifractal function. This multifractal behavior has been associated with the self-similar pattern of the equipotential boundary of the aggregate.[4]

Slow convergence to accurate scaling behavior has long fostered doubts about precise self-similarity of DLA clusters in 2D, and more recently the applicability of the $f(\alpha)$ description has been called into question.[5–9] Such a situation occurs in the context of the current distribution in percolative random resistor networks, where it was found that the multifractal description is inadequate in the negative moments ($q<0$) regime.[10] As pointed out in Ref. 6, while inspecting actual clusters it is relatively easy to envisage structures for which the distribution of harmonic measure is, for small values, not usefully described by $f(\alpha)$.

In this paper we first show that the change in flux at the bottom of a fjord *is governed by the growth at its "lips"* (the outer corners), extending analytic work of Halsey[11] and Ball and Blunt.[12] We discuss sustainability of growth in the sense that two different definitions of α should agree asymptotically: α_r is the rate of screening as a function of distance from the point in question at fixed time, and α_t is the rate of increase in screening of that point as the radius of the cluster grows with time. We also study the relation of this question to the competition between lips as the fjords grow and derive the behavior of the asymptotically decreasing part of $f(\alpha)$, thus clarifying this much debated issue of the most screened sites. We check our analytical predictions against recently published numerical results[13] and find excellent agreement.

All the discussions in the literature concerning the issue of exponential screening, except Ref. 8, have thus far disconnected the distribution of the measure from the actual growth process of the structure. This separation is possible for the current distribution in random resistor networks, where the geometry of the network that determines $n(p)$ is independent of the magnitude of the currents through the bonds, which determine p.[10] In DLA such a separation cannot be justified because the growth process that determines $n(p)$ is governed exactly by this very same distribution of p. Hence a consistent analysis should incorporate some closure condition that takes this fact into account. Here we introduce such a requirement and find its implications.

It is convenient to analyze the harmonic measure within the framework of electrostatics; we seek to calculate the change in E_n at a point s' on the growing perimeter of the aggregate due to an infinitesimal advance $\delta x(s)$ at another perimeter point s. The cluster is assumed at potential $\Phi=V_0$, while the boundary at a large radius R_∞ is kept at $\Phi=0$. To shift the $\Phi=V_0$ equipotential outwards from $x(s)$ to $x+\delta x(s)$ requires an increment of $\delta\Phi=\delta x(s)E(s)$ to the potential just outside the growth. Alternatively, this can be achieved by placing a density of dipoles $\delta x(s)E(s)$ at the surface. Hence, if the field at s' due to a unit dipole (just outside the original surface) at s is denoted $H(s',s)$ we have[11]

$$\delta E(s')=\int H(s',s)\delta x(s)E(s)ds,\quad s'\neq s \tag{1}$$

for points s' where $\delta x(s)=0$. For points at which $\delta x(s)\neq0$ there are some subtleties which need to be explained.[14] but which do not change the global result obtained here. Elementary considerations show that the quantity $H(s',s)/[E(s)E(s')]$ is conformally invariant. Therefore applying a simple conformal mapping leads to the exact result[15]

$$H(s',s)=-\frac{(2\pi/Q^2)E(s)E(s')}{4\sin^2(\theta/2)},\quad s'\neq s$$
$$Q\equiv\oint E(s)ds,\quad \theta\equiv(2\pi/Q)\int_s^{s'}E(s'')ds'' , \tag{2}$$

where Q is the total charge on the growth and θ is the charge in units of angle between points s' and s.

If we now use the (constitutive) growth rule $\delta x(s) = E(s)\delta t$ [it is trivial to extend the present analysis to the dielectric breakdown model where $\delta x(s)=|E(s)|^{\eta}\delta t$], we can write down the temporal equation for the evolution of $E(s')$ as

$$\frac{\partial E(s')}{\partial t}=\oint H(s',s)[E(s)]^2 ds . \tag{3}$$

We now proceed to apply this result by first considering the change in the overall charge

$$\frac{dQ}{dt}=\oint ds[E(s)]^2\oint ds' H(s',s) . \tag{4}$$

Employing the symmetry of H under an interchange of s and s', the inner integral has a very simple interpretation: It is the change in $E(s)$ due to a unit dipole density (which is equivalent to a uniform increase in the potential) over the entire growth surface. Hence the inner integral equals $E(s)/V_0$ exactly, and $dQ/dt=(1/V_0)\int E(s)^3 ds$, as first noted by Halsey.[11]

Next we look at $\partial E/\partial t$ for a region s' where $E(s)\rightarrow 0$ as $s\rightarrow s'$ sufficiently strong so that the region from s to s' does not dominate the integral in (3) (see below). Substituting (2) into (3) yields an expression for the rate of change of $E(s')$, which exactifies the approximation of Ball and Blunt.[12] If we now represent the overall size of the cluster in terms of a radius $R(t)$ of a circle which has the same capacitance as the cluster, with respect to the outer earth [this definition of $R(t)$ should agree with the radius of gyration up to a constant of order one], i.e.,

$$\oint ds\, E(s)\ln[r(s)/R(t)]=0 ,$$

then we have $V_0=(Q/2\pi)\ln[R_{\text{earth}}/R(t)]$ and we can eliminate Q, V_0, and time from the above equations to obtain

$$\alpha_t\equiv-\frac{\partial\ln[E(s')]}{\partial\ln[R(t)]}=\frac{\oint ds[E(s)]^3/[4\sin^2(\theta/2)]}{\oint ds[E(s)]^3} . \tag{5}$$

This is the primary bare result of this paper—an explicit expression for the rate of screening with time of an arbitrary (negligibly growing) point. Its numerator could only be dominated by the field near s' when $E(s)$ increases outwards slower than $|s-s'|$. By contrast, therefore, we must conclude that for exponential screening [where $E(s)$ should increase faster than any power of $|s-s'|$], *the value of α_t at the bottom must be dominated by the action at the lips of the fjord.*

We can now relate α_t to the geometry of the fjord. The denominator on the right-hand side of (5) is simply the third moment which behaves as

$$m_3(R)=\oint ds[E(s)]^3\sim R^{-\tau_3} , \tag{6}$$

where τ_3 is independent of the size and we choose the normalization $Q=\int E(s)ds=2\pi$ so that $E=|\partial\theta/\partial s|$. The numerator $\mathcal{N}$ we handle as follows: It is plausible to assume that locally the most screened point is at the bottom of a fjord, of width $w(r)$ where r is a distance variable that increases from the site outwards. Then $\mathcal{N}$ can be broken into successive boxes of size $w\times w$ giving

$$\mathcal{N}=\sum_{\text{boxes}}\frac{q_{\text{box}}^3}{\theta_{\text{box}}^2}\int_{\text{box}}ds[E(s)/q_{\text{box}}]^3 , \tag{7}$$

where the integral amounts to the third moment of the growth probability, normalized to one within the box, namely, $m_3(w)$. Elementary considerations of screening down channels give[16] $d\theta/dr\approx\theta/w$ so that $q_{\text{box}}\approx\theta_{\text{box}}$, and we have

$$\mathcal{N}=\int_{\text{fjord}}dr[\theta(r)/w(r)]m_3(w)=\int_{\text{fjord}}d\theta(r)m_3(w(r)) . \tag{8}$$

We expect the (positive) local moment m_3 to behave as $m_3\sim w^{-\tau_3}$, and take a general case where on the average $w(r)\sim r^z$. If $\theta(r)$ increases faster than a power of r, the integral is dominated by large r as argued above, and the field at the bottom of the fjord is irrelevant. The same applies to $\theta(r)\sim r^{\alpha}$ for $\alpha>z\tau_3+z-1$, so in all interesting cases the numerator is dominated by the lips of the fjord (i.e., θ_L and w_L) and

$$\alpha_t\approx\frac{m_{3,L}\theta_L}{m_{3,\text{cluster}}}\approx\theta_L(R/w_L)^{\tau_3} . \tag{9}$$

Using the above equation, we now examine whether there is a bound on the strength of screening which can be consistently maintained under growth. We write the charge on the lip as $\theta_L\approx\pi(w_L/R)^{\alpha_L}$ to obtain

$$\alpha_t\approx C\alpha_r^{\tau_3-\alpha_L} , \tag{10}$$

where C is a constant and we used $\alpha_r\approx R/w_L$. If $\alpha_L>\tau_3-1$, this puts an upper bound on the sustainable screening through the relation $\alpha_t=\alpha_r=C^{1/(1+\alpha_L-\tau_3)}$. Using established results, $\tau_3=D$ and $\alpha_{\min}=D-1$,[11,12,17] we see that an anomalously large value of α can be sustained only in the extreme case where a long narrow fjord opens directly onto one of the strongest singularities ($\alpha_{\min}$) of the growth. Another way to interpret these results is to regard the rate of increase in screening as a function of growth of the fjord rather than the growth of the entire cluster. θ_L can be written as $(\partial_\tau L/\partial_\tau R)(w_L/R)^{D-1}$ whereupon

$$\alpha_{t,L}\equiv-\frac{\partial\ln E}{\partial\ln L(t)}=\frac{L\partial_t R}{R\partial_t L}\alpha_t=L/w_L , \tag{11}$$

where $\tau_3=D$ and the above results have been used. So we have a consistent picture where $\alpha_{t,L}=\alpha_L$.

Thus the next relevant question, which we address below, concerns the probability P that such a process occurs. The above discussion establishes that the dominating case corresponds to two nearby lips growing at the same rate to generate a long fjord. Such a process is inherently unstable because falling behind by either lip impairs its growth further. It follows that the natural candidates to promote large values of α evolve by maintaining a close race between the lips for comparatively long times, yielding long and narrow fjords.

We proceed to estimate P by considering a fjord of length L and lip separation $w_L \ll L$, whose lips advance a distance w_L outwards, $L \rightarrow L' = L + w_L$. The field inside is reduced by a factor $\zeta = (1+1/y)^{\pi y}$, where $y \equiv L/w_L$ (for $1 < y < \infty$, ζ is between 2^π and e^π). The only mechanism that can counterfeit the instability is the structural fluctuations. Specifically, if a mismatch Δr (in the growth direction) appears between the lips of a fjord, and Δr is smaller than the noise amplitude, this mismatch may be spontaneously closed. The amplification of the mismatch under diffusion-controlled growth would lead to $\Delta r(L')/\Delta r(L) = \gamma > 1$. We expect the spontaneous fluctuation to be of order $\Delta r_{\text{rms}} \approx w_L \sqrt{\epsilon/D}$, where ϵ is the asymptotic relative variance of mass added discussed in Ref. 18. An unstable runaway occurs only if the amplification by γ takes Δr outside the noise range, while when $\Delta r < \sqrt{\epsilon/D}\, w_L/\gamma$ the fjord will keep growing as such. It follows that the probability that the two lips keep abreast at L' is the probability of retaining the balance until L multiplied by $1/\gamma$, or $\delta(\ln P) = -\ln\gamma$. If this process dominates occurrence of smallest growth probabilities, we can conclude that $P \sim L^{f-A}$. The field inside obeys $E_{\text{inside}}/E_{\text{lip}} \sim L^{-\alpha-B}$, where A and B are constants. It follows that

$$\frac{\delta \ln P}{\delta \ln(E_{\text{inside}}/E_{\text{lip}})} = \frac{\delta[(f-A)\ln L]}{-\delta[(\alpha+B)\ln L]} = \frac{\ln\gamma}{\ln\zeta}. \tag{12}$$

Integrating over this equation one finds

$$f(\alpha) = -\frac{\ln\gamma}{\ln\zeta}\alpha + K + O(1/\ln L), \tag{13}$$

where K is a constant. This implies that if old fjords dominate the large-α regime, the slope of the $f(\alpha)$ curve approaches a limiting *finite* value. This bounds the value of $f(\alpha)$ for large enough α, which excludes exponential screening (i.e., exponentially small growth probabilities leading to infinitely large values of $\alpha_{\max}$) even under noisy growth. However, it should be stressed that (i) the $f(\alpha)$ curve may achieve a steeper slope for intermediate values of α, and (ii) that finite-size effects of order $1/\ln L$ [as appears in (13)] may be large for the present sizes of simulated aggregates. Unfortunately the analysis presented here cannot yield a value for $\alpha_{\max}$ itself.

Returning to the survival probability of old fjords P, we note that when the fjords grow by w_L, their y value in-

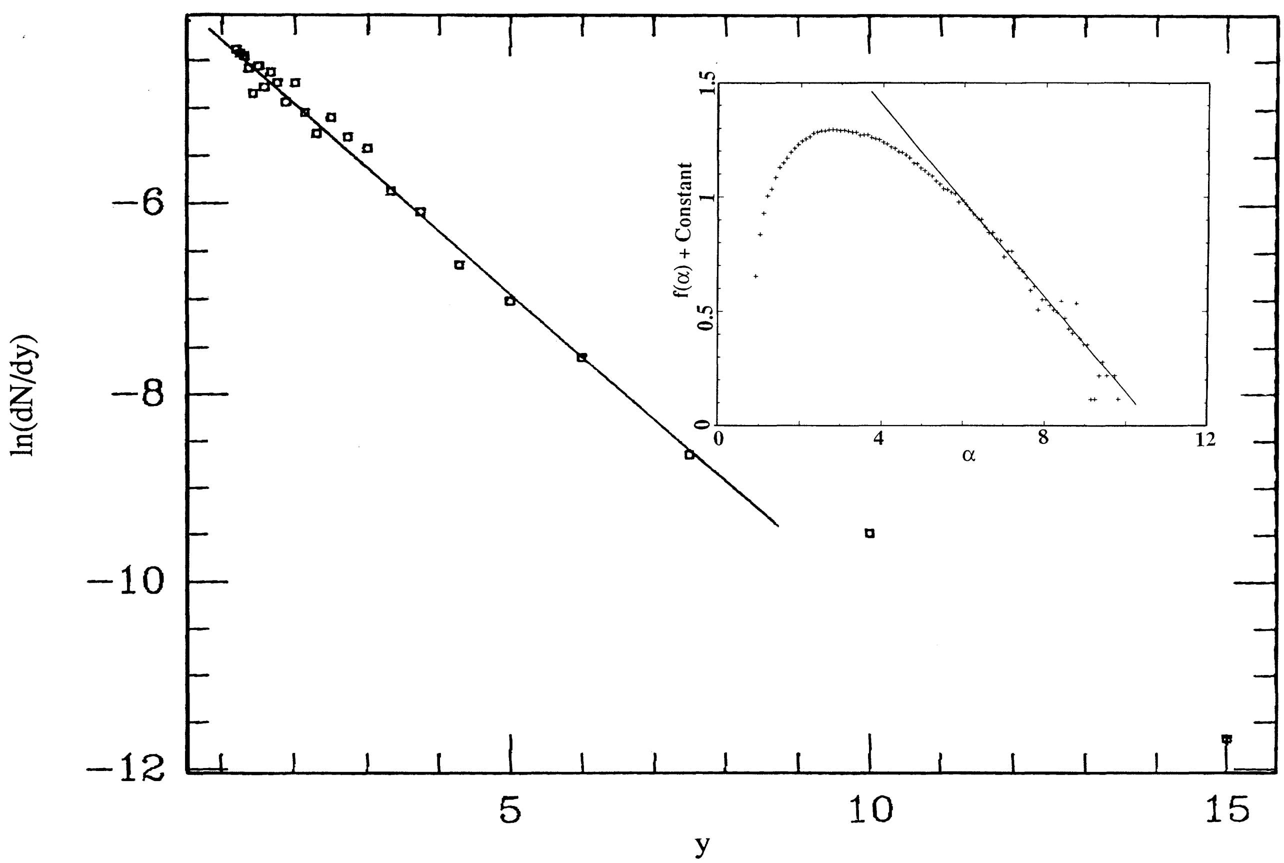

FIG. 1. The logarithm of the distribution dN/dy of the aspect ratio y (length divided by outer width) for fjords of DLA clusters from the data of Ref. 13. The behavior is linear, as predicted in the text, with slope 0.67 ± 0.05. The corresponding asymptotic linear decrease of $f(\alpha)$ predicted is compared in the inset with $f(\alpha)$ data (Ref. 19).

creases by unit, $\delta y=1$. Hence $\delta \ln P/\delta y=-\ln\gamma$, which yields $P\sim\exp(-\ln\gamma y)$ for large y. Thus we expect the distribution of fjord aspect ratios F to behave as

$$\frac{dF}{dy}\sim\exp(-y\ln\gamma)$$

for large y. Figure 1 shows the data in Ref. 13 (originally published in the form dF/dx, where $x=1/y$) plotted on a semilog scale, indicating good agreement with our form, with a slope of $\ln\gamma=0.67\pm0.05$. For the narrow fjords that dominate this regime, we thus have to a good accuracy $\ln\zeta=\pi$ and hence the asymptotic value of the slope of the $f(\alpha)$ in DLA is predicted to be 0.21 ± 0.02. The inset in Fig. 1 shows this slope compared to $f(\alpha)$ obtained from simulations on clusters of 100000 sites,[19] indicating reasonably good agreement.

To conclude, we analyzed the behavior of the multifractal function $f(\alpha)$ for large α. We found that by ignoring structural fluctuations we could employ a self-consistency argument that leads to an upper finite bound on $\alpha_{\max}$. Including the effect of noise, we have found that we can bound the slope of $f(\alpha)$ for large α, thus effectively bounding again $\alpha_{\max}$. The asymptotic slope has been estimated from numerical results in Ref. 13, to be of order -0.2. We suggest that our results indicate that $\alpha_{\max}$ is bounded by a finite value for asymptotic DLA. We should mention that these results also lead to a second-order phase transition at some value of α_c where the slope reaches its asymptotic value.

We thank Professor T. Vicsek and A.-L. Barabasi for providing the data shown in Fig. 1 and for helpful discussions. This work was supported by a grant of the Science and Engineering Research Council, United Kingdom.

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

[2] T. C. Halsey, P. Meakin, and I. Procaccia, Phys. Rev. Lett. **56**, 854 (1986); T. C. Halsey, M. H. Jensen, L. P. Kadanoff, I. Procaccia, and B. I. Shraiman, Phys. Rev. A **33**, 1141 (1986).

[3] B. B. Mandelbrot, J. Fluid Mech. **62**, 331 (1974).

[4] See, e.g., T. Vicsek, *Fractal Growth Phenomena* (World Scientific, Singapore, 1989).

[5] J. Lee and H. E. Stanley, Phys. Rev. Lett. **61**, 2945 (1988); J. Lee, P. Alstrøm, and H. E. Stanley, Phys. Rev. A **39**, 6545 (1989); Phys. Rev. Lett. **62**, 3013 (1989); P. Trufino and P. Alstrøm, Phys. Rev. B **41**, 896 (1990).

[6] R. Blumenfeld and A. Aharony, Phys. Rev. Lett. **62**, 2977 (1989).

[7] B. Fourcade and A.-M.S. Tremblay, Phys. Rev. Lett. **64**, 1842 (1990); R. Blumenfeld and A. Aharony, Phys. Rev. Lett. **64**, 1843 (1990).

[8] A. B. Harris and M. Cohen, Phys. Rev. A **41**, 971 (1990).

[9] C. J. G. Evertsz (unpublished); C. J. G. Evertsz and B. B. Mandelbrot (unpublished).

[10] R. Blumenfeld, Y. Meir, A. Aharony, and A. B. Harris, Phys. Rev. B **35**, 3524 (1987).

[11] T. C. Halsey, Phys. Rev. Lett. **59**, 2067 (1987).

[12] R. C. Ball and M. J. Blunt, Phys. Rev. A **39**, 6545 (1989).

[13] A. L. Barabasi and T. Vicsek, J. Phys. A **23**, L729 (1990).

[14] When $\delta x(s)\neq 0$ there are two complications: (i) There is a contribution to the integral from the motion of the boundary itself, but this drops if the field E is considered just before the advance occurs. Self-consistently the flux reduces proportionally to the local stretch of the surface. The contribution of this effect is irrelevant for highly screened points, which are our main concern here: (ii) The value of $H(s',s)$ should be considered just outside the dipole layer. This results in $\int H(s',s)ds$ being positive and finite for s' inside the integration region despite H itself being negative for $s'\neq s$, as we shall note below. A detailed discussion will be given elsewhere.

[15] For the small-θ regime, this result coincides with the analysis of T. C. Halsey, Phys. Rev. A **38**, 4749 (1988).

[16] This known result is analogous to the one recently cited as "Buerling equality" in Ref. 9.

[17] L Turkevich and H. Scher, Phys. Rev. Lett. **55**, 1026 (1985); Phys. Rev. A **33**, 786 (1986).

[18] P. W. H. Barker and R. C. Ball, Phys. Rev. A **42**, 6289 (1990).

[19] This curve is generated by applying the method of R. C. Ball, M. J. Blunt, and O. Rath Spivak, Proc. R. Soc. London Ser. A **423**, 123 (1989) to off-lattice DLA clusters of 100000 particles supplied by P. Meakin.