RAPID COMMUNICATIONS

PHYSICAL REVIEW E VOLUME 49, NUMBER 1 JANUARY 1994

Conductance noise in electrodeposition

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(Received 8 October 1993)

We present a theoretical study of the conductance fluctuations of an electrode where a cluster is grown though electrodeposition, in a diffusion-limited regime. We show that the distribution of conductance jumps $\mathcal{N}(\delta g)$ measured during the deposition process from a pointlike seed can be analyzed using the multifractal nature of the current flowing at the surface of the cluster. The distribution $\mathcal{N}$ consists of a power law for large jumps. We derive the relation between the exponent of this distribution and the multifractal spectrum.

PACS number(s): 81.15.Pq, 64.60.Ak

INTRODUCTION

We present in this paper an analysis of the conductance noise which occurs in electrodeposition in the regime where it can be modeled by the diffusion-limited aggregation (DLA) model [1] or its generalization, the dielectric-breakdown (DB) model [2]. Many reviews present the essential results on these models and their application to a variety of physical situations [3]. In particular, the multifractal properties of the growth probabilities that result from these models have received much attention. However, no easily accessible properties connected with these growth processes depend crucially on the multifractal growth properties. It is the aim of this paper to present just such a property. As we will show, the statistical distribution of noise in the conductance during the process of electrodeposition follows a power law whose exponent is a direct reflection of an underlying multifractal distribution.

The DB model can be briefly presented as a stochastic model of fractal cluster growth. At any stage of its growth the cluster is modeled as a perfect electrode (with a zero resistivity) and it is immersed in a very large (ideally infinite) Ohmic medium with a finite conductivity. The cluster is set to zero potential, whereas at large distance, a unit potential is applied. The cluster grows according to the following probabilistic rule: a new particle is added to the cluster with a probability proportional to the potential at this point raised to the power η. This is the free parameter of the model. DLA is recovered for $\eta=1$. As the cluster grows, the conductance of the medium between the cluster and the external electrode increases each time a new particle is added. This increase, however, exhibits strong short-time fluctuations. It is these fluctuations we analyze in the following.

Earlier studies have focused on various fluctuations occurring in DLA or DB models with a particular emphasis on "$1/f$" noise. In particular, Evertz and Mandelbrot [4] have reported numerical results on the fluctuations of some geometrical features—such as the location of the longest branch—of a cluster grown in a strip geometry. Van Damme [5] has experimentally studied the growth rate along the boundary of the cluster in viscous fingering experiments. The power spectrum of this growth rate as a function of the curvilinear abscissa has been shown to reveal some original power-law behavior. Finally, Louis and Guinea [6], while studying a closely related fracture model, have studied numerically the time evolution of the stresses along the cluster (i.e., the crack in this case) and the growth probability of active sites. The theoretical analysis we present below can be simply adapted to this problem. However, the data were not quantitatively discussed in this reference and thus no comparison can be made.

Analogous results can also be used in the context of the invasion with a nonviscous fluid of a porous media saturated by a more viscous one. DLA has been proposed as a model for viscous fingering in some flow regimes [7]. Inasmuch as this modeling is correct, the fluctuations of permeability during the invasion can also be described by the formalism developed below. A somewhat similar treatment can also be performed in a very different framework, e.g., percolation [8,9]. Extension of the concepts presented here can also be used in the field of fracture of brittle heterogeneous materials for the analysis of acoustic emission [10].

MULTIFRACTALITY OF JUMPS AND SCALING

It is now well known that the distribution of growth probabilities at the surface of DLA or DB clusters is multifractal; see the review by Meakin in Ref. [3]. If r is the radius of the cluster, and p a growth probability, $n(p,r)dp$ is the number of sites whose growth probability is in the range $[p,p+dp]$. $n(p,r)$ is explicitly dependent on r. This is where the multifractal properties of $n(p,r)$ become important: By a change of variable, we may isolate the r dependence by writing

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$$\alpha_p = -\frac{\log(p)}{\log(r)} ,$$
$$f_p(\alpha_p) = \frac{\log[pn(p,r)]}{\log(r)} . \tag{1}$$

As r approaches infinity, $f_p(\alpha_p)$ approaches a universal function which is independent of r (provided the outer boundary is far enough from the cluster). The number of sites $n(p,r)$ is multiplied by p in the above expression as the natural density to use is the distribution of $\log(p)$ rather than simply p. Another equivalent way of introducing the multifractal formalism is to consider the scaling of different moments of the growth probability, $M_p(q)=\sum_i p_i^1$ (*not* normalized by the number of terms in the sum). Those moments scale with r as a power law with exponent $\tau_p(q)$. Both functions $\tau_p(q)$ and $f_P(\alpha)$ contain the same information and are related through the Legendre transform,

$$q = \frac{\partial f_p(\alpha)}{\partial \alpha} ,$$
$$\tau_p(q) = f_p(\alpha) - q\alpha . \tag{2}$$

The local growth probability is by definition a simple power law of the voltage at a given site. Thus the multifractal nature of the probability implies in turn a multifractal character of the distribution of local voltages v at the surface of the cluster. Computing the voltages using a constant total current condition makes it possible to relate $v_i = p_i^{1/\eta}/M_p(1/\eta)$.

We are interested in the change of conductance that will occur when a new site is added to the cluster. If the conductance increases from g to g', we define the conductance jump as $\delta g = (g'-g)$. It is intuitive that δg will be related to the local voltage v at the growth site. More precisely, using Cohn's theorem it is possible to establish that δg will be proportional to the square of the voltage, v^2, when the latter is small. It seems that this proportionality is valid even when the voltage is not infinitesimal. Thus we can relate the conductance jump δg to the local growth probability

$$\delta g \propto p^{2/\eta}/M_p(1/\eta)^2 . \tag{3}$$

As a simple consequence of the above equation, the distribution of conductance jumps that would occur if we add any surface site to the cluster is also multifractal. Thus this distribution can be characterized through a spectrum $f_g(\alpha_g)$, by using the following definition of these variables:

$$\alpha_g = -\frac{\log(\delta g)}{\log(r)} ,$$
$$f_g(\alpha_g) = \frac{\log[\delta g n(\delta g, r)]}{\log(r)} . \tag{4}$$

The connection with the previous multifractal spectrum is through the following relations:

$$\alpha_g = (2/\eta)\alpha_p + 2\tau_p(1/\eta) ,$$
$$f_g(\alpha_g) = f_p((\eta/2)\alpha_g + \tfrac{1}{2}\tau_p(1/\eta)) , \tag{5}$$

i.e., the α scale is simply dilated by a factor $2/\eta$ and translated by $2\tau_p(1/\eta)$ whereas the f scale is unchanged. This is a simple consequence of Eq. (3).

RESCALING OF THE SPECTRUM FOR INTERMEDIATE STAGES

It is important to note that the overall conductance of the medium depends on its geometry, and hence the size of the system L, as well as the radius of gyration at a given stage r. The difference δg does not depend on L, as long as $r/L \ll 1$. This can be argued for in the following way: If we increase L to L', the system can be seen as consisting of two parts in series, from the origin to L, and from L to L'. The approximation of having an equipotential at radius L is expected to be increasingly fulfilled as L increases. The resistance jump occurring during one elementary growth event will thus be independent of the resistance of the L to L' part, the latter being canceled by the difference. Using again the limit $r/L \ll 1$ we expect that the total change in conductance from the initial seed to the developed cluster will be small. As a result, the conductance jump δg is simply proportional to the resistance jump, with a proportionality constant equal to the square of the initial conductance of the system.

We will have to consider the evolution of the conductance at different cluster radii r. Let us introduce a reference radius R which in practice will be the final stage of the cluster, with the constraint $R \ll L$. Although the natural variable to use for a fixed r is the expression for α and f introduced in Eqs. (1) and (4), the variability of r in the range 1—for the initial seed — to R requires that a fixed reference radius, e.g., R, should be chosen. We thus introduce as before the reduced variable $\alpha = -\log(\delta g)/\log(R)$, and the intermediate notation $\beta = -\log(\delta g)/\log(r)$. We can simply express

$$\beta = \alpha/\lambda , \tag{6}$$

where $\lambda = \log(r)/\log(R)$ is a scaling factor. Similarly, the number of growth sites which give rise to a jump δg for a radius r is

$$\delta g n(\delta g, r) \propto r^{f_g(\beta)} = R^{\lambda f_g(\alpha/\lambda)} . \tag{6'}$$

It is possible to interpret the transformation of Eqs. (6) and (6') in simple geometrical terms. Being given the spectrum for a radius R, the spectrum relative to a radius r can be obtained by a simple dilation of the previous function in the f-α plane. The center of the dilation is the origin and the amplitude of the dilation is λ. This construction is illustrated in Fig. 1.

What is the probability $p(\delta g)$ to have a conductance jump equal to δg, at a given stage where the radius of the cluster is r? This probability is given by the probability to pick a bond with the correct voltage times the number of these bonds; thus, using the definition of f and the relation (3), we can write

$$\rho(\delta g)\delta g \propto r^{f_g(\beta)}(\delta g)^{\eta/2} . \tag{7}$$

We have introduced the probability $\rho(\delta g)\delta g$ which corresponds to the distribution of $\log(\delta g)$ rather than of δg, so

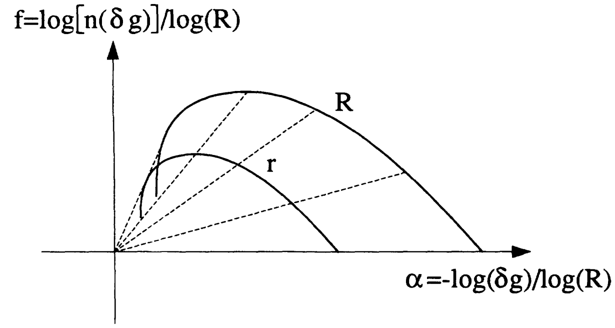

FIG. 1. Rescaling of the multifractal spectrum of the conductance jumps at a given stage of growth with radius r, compared to that measured at the reference radius R.

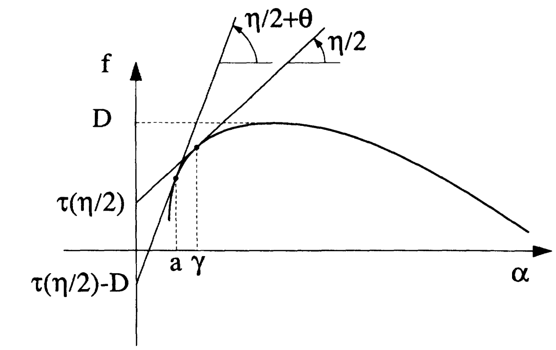

FIG. 2. Geometrical construction of the exponent θ of the conductance jump distribution integrated during the entire growth process. By drawing the tangent to the spectrum with a slope $\eta/2$ we deduce γ and $\tau(\eta/2)$. From the f value of the apex we get the fractal dimension D. The difference $\tau(\eta/2)-D$ being known, we construct the tangent to the spectrum which goes by the point $(\alpha=0, f=\tau(\eta/2)-D)$. The difference of slopes between the two tangents gives the conductance jump distribution exponent θ.

as to be consistent with the usual definition of f, e.g., in Eq. (1). We choose a normalization ρ so that the exponent $\tau(1)=0$. The first moment is governed by the maximum of the probability distribution $\rho(\delta g)\delta g$. This maximum is reached for a value of β such that

$$f_g'(\beta)=\eta/2 , \quad (8)$$

where f' denotes the derivative of f. We thus introduce γ as the root of $f_g'(\gamma)=\eta/2$, so as to simplify the notation. We note that the parameter γ is uniquely defined for a given η parameter. Thus expression (7) can be rewritten as

$$\rho(\delta g)\delta g = R^{\varphi(\alpha,\lambda)-\varphi(\gamma\lambda,\lambda)} , \quad (9)$$

where we have introduced the function

$$\varphi(\alpha,\lambda)=\lambda f_g(\alpha/\lambda)-(\eta/2)\alpha . \quad (10)$$

With the normalization introduced in Eq. (9), the maximum of $\rho(\delta g)\delta g$ is 1.

Let us note that in the case of $\eta=1$, i.e., for the DLA case, we can make use of an exact result concerning γ. It has been shown rigorously by Makarov [11] that the first moment of the growth probability (for which $\tau=1$ by normalization) is characterized by $\alpha_p=1$ and $f_p=1$. Using the correspondence between p and δg given in Eq. (5), we deduce that $\gamma=2$. Moreover, still for $\eta=1$, the expression $\varphi(\gamma\lambda,\lambda)$ in Eq. (9) which has been added for normalization is identically 0.

INTEGRATION THROUGH THE ENTIRE GROWTH PROCESS

In order to obtain the distribution of the conductance jumps during the growth process, it suffices to integrate the probability given in Eq. (9). The cluster is fractal, and thus there is a power-law relation between the number of sites in the cluster $m(r)$, and its radius r

$$m(r)\propto r^D . \quad (11)$$

The measure needed to integrate p is dm. Using the variables introduced above, the measure can be written

$$dm = DR^{D\lambda}\log(R)d\lambda . \quad (12)$$

The number of conductance jumps δg recorded throughout the entire growth is characterized by the distribution, $\mathcal{N}(\delta g)d\delta g$, which can be written

$$\mathcal{N}(\delta g)\delta g = D\log(R)\int_0^1 R^{\varphi(\alpha,\lambda)-\varphi(\gamma\lambda,\lambda)+D\lambda}d\lambda . \quad (13)$$

We now estimate the integral (13) using the steepest-descent method. The maximum of the exponent of R in the integral is reached for a value of λ which satisfies the equation

$$f_g\left[\frac{\alpha}{\lambda}\right]-\frac{\alpha}{\lambda}f_g'\left[\frac{\alpha}{\lambda}\right]=f_g(\gamma)-\gamma f_g'(\gamma)-D . \quad (14)$$

Using the expression for $\tau(q(\alpha))$ given in Eq. (2), we can simplify the above expression to

$$\tau_g(q(\alpha/\lambda))=\tau_g(q(\gamma))-D . \quad (14')$$

Equation (14′) allows a simple geometrical construction which is illustrated in Fig. 2. From Eq. (2), it can be seen that $\tau(q(\alpha))$ is the f coordinate of the intersection between the tangent to the spectrum at the point $(\alpha, f(\alpha))$ with the f axis. The slope of the tangent is $q(\alpha)$. As an example $\tau(0)$ is the fractal dimension of the cluster, and it corresponds to the f value of the apex of the spectrum. The γ parameter can be obtained by looking for the abscissa of the tangent point with a slope $\eta/2$ [see Eq. (8)]. The quantity $\tau_g(q(\gamma))=\tau_g(\eta/2)$ can therefore easily be read off the graph. Knowing $\tau_g(\eta/2)$ and D we can search for the root a of

$$\tau_g(q(a))=\tau_g(q(\gamma))-D , \quad (15)$$

by drawing the tangent to the spectrum knowing the intersection of the tangent with the f axis.

We have introduced the root a in Eq. (15), from which we deduce the value of λ^* which maximizes the expression to integrate, $\lambda^*=\alpha/a$. We now substitute this expression for $\mathcal{N}$,

$$\mathcal{N}(\delta g)\delta g \propto \sqrt{\log(\delta g)}(\delta g)^{\theta}\ , \tag{16}$$

where we have neglected the constant prefactor, with the expression for the exponent

$$\theta = \frac{f_g(a)+D-\tau_g(\eta/2)}{a} - \eta/2$$
$$= f_g'(a) - \eta/2\ . \tag{17}$$

Equation (16) is the main result of this paper. Apart from a weak $\log(\delta g)^{1/2}$ correction, the distribution of conductance jumps follows a power-law behavior with a universal exponent which can be obtained from the multifractal spectrum of the growth probability at a fixed stage of growth. The final expression of θ given in Eq. (17) can again be simply interpreted in Fig. 2. The difference of slopes between the two tangents at $\alpha = a$ and $\alpha = \gamma$ gives θ.

It should also be noted that Eq. (15) may have no solution. Such will be the case whenever the origin lies below the multifractal spectrum $f_g(\alpha)$, or equivalently when $f_g(0) > 0$. For DLA, the minimum value of α is strictly positive, and hence there exist a solution. The result discussed above is therefore valid. However, for large η it is conceivable that the largest potential drop at the border of the cluster *increases* with the radius of gyration r. In this case, the steepest-descent method obviously fails because Eq. (15) has no root, and a simple analysis shows that the integration through the entire growth is dominated by the latest stage. Therefore, in this case, $\mathcal{N}(\delta g)$ can be identified directly with $n(\delta g, r)$.

CONCLUSION

Equations (16) and (17) constitute our main results: The histogram of the conductance jumps occurring during the growth of the electrode through an electrodeposition process consists essentially in a power law, the exponent of which can be computed from the multifractal spectrum of growth probabilities occurring at a reference radius. This law is universal in the same sense as the multifractal spectrum of the local probabilities is. The exponent θ is expected to vary with the parameter η, as most geometrical properties do. The fact that the distribution of the jumps is multifractal has the practical consequence that the domain of validity of the power-law regimes is increasing with the size of the cluster.

It would be of interest to measure this distribution experimentally, and thus to have access to some information on the multifractal spectrum of the growth probabilities without having to resort to local measurement of voltages or currents.

ACKNOWLEDGMENTS

We acknowledge useful discussions with E. Guyon, H. J. Herrmann, and E. L. Hinrichsen. We furthermore thank the CNRS Groupement de Recherche "Milieux Hétérogènes Complexes" for support. The Laboratoire de Physique et Mécanique des Milieux Hétérogènes is "Unité de recherche associée au CNRS No. 857." The Groupe Matière Condensée et Matériaux is "Unité de recherche associée au CNRS No. 804."

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