Structure of noise generated on diffusion fronts

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We show in this paper the characteristic behavior of the noise generated by the fluctuation of diffusion fronts. We predict a $1/f^2$ noise at high frequency and a $1/f$ noise at low frequency. A crossover frequency f_c separates the two regimes. This crossover frequency f_c has a power-law behavior as a function of the diffusion length. To relate the static properties of the fronts represented by a problem of percolation in a gradient to the dynamical behavior, we assume that the probability of disconnecting a finite cluster is proportional to the number of red bonds present in a disk with a radius equal to the cluster radius. A scaling of the different fluctuation regimes as well as a scaling of the density of events of a given size is proposed. The various critical exponents are compared with those extracted from numerical simulations performed in the two-dimensional case. We also point out the close relation of these results to noise in invasion experiments in the presence of a gravity field.

I. INTRODUCTION

Diffusion naturally creates objects with a fractal geometry.[1-3] In the case where the diffusing particles do not interact, this geometry is closely related to the geometry of percolation clusters and the most general concept which allows a unique approach of these structures is percolation in a concentration gradient called "gradient percolation."[2-7] The purpose of this work is to study more deeply than in our preliminary approaches[8,9] the "geometrical noise" generated by the fluctuations of the diffusion front during the diffusion process.

A. Recall of the main results on noninteracting hard-core diffusing lattice gas

A detailed presentation of the model has been given elsewhere.[2,3] As we will compare our theoretical model to two-dimensional numerical calculations, we will refer here to $d=2$ values for the critical exponents. However, the scaling laws are general. The $d=3$ case will be discussed at the end of the paper.

The present study will be limited to the case of noninteracting hard-core diffusing particles: Particles are allowed to jump at random to first-neighbor sites in a d-dimensional lattice, provided these sites are empty. Under these conditions, the concentration is known[10] to follow an ordinary diffusion equation. Basically, we consider in this paper the most simple case of a stationary system with fixed concentrations on both sides of the finite sample of size $L^{d-1}\times L'$: The concentration profile presents a constant gradient ∇p along coordinate x, $0\le x\le L'$. Nevertheless, it is important to remark that the result should be applied to nonstationary cases provided that the evolution of ∇p with time is on a much larger time scale than the characteristic fluctuation time.

A number of physical properties are associated with a "connection" rule between the particles. For example, nonzero hopping conductivity supposes that an electron on an atom can find another atom close enough to present overlapping external orbitals. In our model, we will simply choose a first-neighbor connection.

We can define, with respect to this connection, different subsets of particles.[2-7] The set of particles connected to the high-concentration region corresponds, in the limit of an infinite sample and an evanescent gradient, to the "infinite cluster" in the percolation problem. Its external surface is located around p_c: it is precisely what we call the diffusion front. There also exist finite clusters with an average linear size (radius of gyration) given by the correlation length ξ which decreases with the distance from the average position x_c of the front, according to

$$\xi \propto |p(x)-p_c|^{-\nu} , \tag{1}$$

with $p(x)=p_c+\nabla p(x-x_c)$. ν is the correlation length critical exponent. At short distances, this front has a fractal behavior with a dimension D_f. It has been shown[2,11,12] that for $d=2$,

$$D_f=1+1/\nu=\tfrac{7}{4} . \tag{2}$$

The width of the front σ_f has a power-law dependence on the concentration gradient,[2]

$$\sigma_f \propto |\nabla p|^{-\alpha_\sigma} \quad \text{with } \alpha_\sigma=\nu/(1+\nu) . \tag{3}$$

In the concentration variable, this width is proportional to a "concentration width" $\delta p \propto |p(\sigma_f)-p_c|$ defined as

$$\delta p \equiv |\nabla p|^{\alpha_\sigma/\nu} . \tag{4}$$

As far as physical properties of the interface are concerned, another important quantity is the overall length of the front. The average number of particles on the front N_f is also a power-law function of the gradient[2,7]

$$N_f = \langle N_f(t) \rangle \propto |\nabla p|^{-\alpha_N} \tag{5a}$$

with

$$\alpha_N = (D_f - d + 1)\nu/(1+\nu) \tag{5b}$$

and (only in $d=2$) $\alpha_n = \alpha_\sigma/\nu = 1/(1+\nu) = \frac{3}{7}$. Let $p_f(x)$ be the probability to find a particle of the front at distance x. The variation of front concentration has the following scaling behavior:[7]

$$p'_f(x) = p_f(x)/p(x)$$
$$\approx |\nabla p|^{(d-D_f)\nu/(\nu+1)} \Pi_f[(p-p_c)/\delta p] , \tag{6a}$$

where Π_f is a scaling function which depends on the reduced variable $(p-p_c)/\delta p$. This scaling law is based on the assumption that there exists a unique scaling length in the problem which is σ_f (δp in concentration variable). The gradient-dependent prefactor is obtained by integrating (6a) and comparing the result with the total number of sites on the front given by Eqs. (5a) and (5b). Expression (6a) has been numerically checked in the $d=3$ case. We assume here that this scaling is valid also in two dimensions. We will discuss below the limit when $|\nabla p|$ goes to zero: then δp also goes to zero and Eq. (6a) could, in some cases, be conveniently approximated by a δ function,

$$p_f(x) \approx n_0 |\nabla p|^{-\alpha_N + 1} \delta(p - p_c) . \tag{6b}$$

Another crude approximation is

$$p_f(x) \approx n_0 |\nabla p|^{(d-D_f)\nu/(\nu+1)}$$
for $|x - x_c| < \sigma_f$, and zero otherwise .

It has the great advantage of leading to the correct results using very simple arguments, and has been used in the preliminary approach.[9] A general presentation of this approach is in print.[13]

B. Fluctuation noise

The dynamical behavior of the diffusion front was revealed by simulation calculations in two-dimensional systems.[8] "Catastrophic" events appear through connection or disconnection of very large clusters after the jump of a single particle.

We observe that during the diffusion process, there are some microscopic events (motion of a single particle) that may induce semimacroscopic changes of the front. A typical time evolution of the number N_f of particles on the front is shown in Fig. 1. where we see a succession of rare large events and frequent small events. One of the most important characteristics of the diffusion fronts is their instability.

Another remarkable point is that the time scale for a fluctuation is enormously reduced as compared to the hopping time. The reason is that for a fluctuation to appear it is sufficient, for example, one of the particles of a large cluster near the front moves to a position such that this cluster becomes a part of the front itself. This motion corresponds to a frequency much higher than the inverse of the average jump time for individual particles. Suppose that the diffusion is an activated process with an average jump time θ. If the system contains N particles, then $\tau_0 = \theta/N$ is the elementary fluctuation time of the system. With N_f particles on the front, then $\tau_f = \theta/N_f$ is the elementary fluctuation time of this front. For macroscopic systems we have $\tau_0 \ll \tau_f \ll \theta$. We will also show below that there exists a crossover time t_c (called t^* in Ref. 14) between a region with a $1/f^2$ noise and a region with a $1/f$ noise. This time t_c will be shown to decrease with a power law of the gradient of concentration $|\nabla p|$. As an example, in the calculations presented here, for a gradient $|\nabla p| = \frac{1}{800}$ and a sample with $N \cong 56\,000$ particles, we found $N_f \cong 8500$ on the front so that $\tau_0 \cong 210^{-5}\theta$, $\tau_f \cong 10^{-4}\theta$, and the simulation gives $t_c \cong 10^{-1}\theta$.

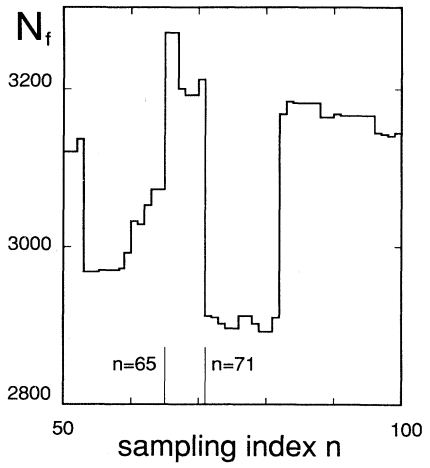

FIG. 1. Time evolution of the number N_f of particles on the diffusion front (from Ref. 8).

Hence, even if θ is extremely large (apparently quenched systems), fluctuations will be observable if the number of particles in the system (and in the front) is large enough. This is a very important property with practical physical implications.[8]

We would like to point out here that the fluctuations appearing in slow invasion by nonwetting fluids[15] can be treated in a rather similar way. This is, in particular, the case for the fluctuations of flux during the invasion process in the presence of a gravity field in which the pressure is very slowly increased.[16,17]

During a diffusion process different quantities can be measured: the fluctuation of the frontier length (or frontier surface in $d=3$); the fluctuation of the total number of particles connected to the source; and any other more subtle physical quantities related to the cluster parameters. These will all be treated following the same way.

We have chosen here to investigate the fluctuations with time of the frontier length $N_f(t)$. They are intended to have a power-law dependence in t:

$$\langle \Delta N_f(t)^2 \rangle = \langle [N_f(t) - N_f(0)]^2 \rangle \propto t^{2H} , \tag{7}$$

where H is an exponent (to be determined) called the Hurst exponent. As indicated above, the fluctuations are produced either by successive connections and disconnections of finite clusters of particles, or by successive opening or closure of finite clusters of empty sites. The corresponding variation of length of $N_f(t)$ for an event at time t is $\pm h$, where h is the perimeter size of the island (or the lake).

C. Preliminary approach of the problem

A zeroth-order approach[9] consists in considering that the frontier is everywhere at $p = p_c$ [Eq. (6c)] and that major contributions in the fluctuations come from the clusters of typical radii close to σ_f (from now on we will suppose that the width σ_f is much smaller than the size L of the sample). The average perimeter size h_M of a large event is then of order $(\sigma_f)^{D_f}$. Moreover, clusters separated by a distance larger than σ_f can be considered independent (uncorrelated). As a first approximation, the fluctuations are then essentially a connection of independent clusters of size $(\sigma_f)^{D_f}$. We expect their number to vary as $n = (L/\sigma_f)^{d-1}$. At short times when only a small part of the n clusters have been probed, the mean-square fluctuation $\langle \Delta N_f(t)^2 \rangle$ is a random Brownian process and the Hurst exponent is $H = \frac{1}{2}$:

$$\langle \Delta N_f(t)^2 \rangle \cong \mathcal{A} t .$$

When all the clusters have been explored, $\langle \Delta N_f(t)^2 \rangle$ saturates to a time-independent value. This appears for times larger than a crossover time t_c. The power-law dependence of $\langle \Delta N_f(t)^2 \rangle$ is approximately n times the square of a typical cluster fluctuation,

$$\begin{aligned} \langle \Delta N_f(t \gg t_c)^2 \rangle &\propto \left[\frac{L}{\sigma_f} \right]^{d-1} (\sigma_f)^{2D_f} \\ &\propto (\sigma_f)^{2D_f - d + 1} \\ &\propto |\nabla p|^{-(\nu/1+\nu)(2D_f - d + 1)} , \end{aligned}$$

or explicitly,

$$\langle \Delta N_f(t)^2 \rangle \propto |\nabla p|^{-10/7} .$$

The prefactor $\mathcal{A}$ in the short-time regime is slightly more difficult to determine, as it requires the knowledge of the probability per unit time to connect or disconnect a typical cluster. The result has been outlined in a preliminary presentation[9] using the same approximation. As we need to go further in the details to understand the meaning of this crude approximation, we will study the fluctuations of a system in which the concentration is slowly varying, so slowly that the system can be represented by the juxtaposition of uniform concentration subsystems.

II. A MODEL FOR THE FLUCTUATIONS

To describe the mechanism of appearance of the fluctuations we need to know the scaling structure of various necessary ingredients. We need, in particular, to know the distribution of the clusters which are close enough to the front to contribute to the fluctuations; we also need to know the probability for a fluctuation of size h to appear.

A. Distribution and cutoff of the finite clusters with external perimeter h

In a region of concentration p, the number of islands with h sites on their external surface, or hull, follows a scaling law similar to that of the number of clusters of s sites.[18] For the clusters of particles, this may be written[19,20] as

$$n_h = h^{-\tau_h} f[(p - p_c) h^{\sigma_h}] \tag{8a}$$

with

$$\tau_h = 1 + d/D_f$$

and

$$\sigma_h = 1/\nu D_f . \tag{8b}$$

A similar expression may be written for the clusters of empty sites (lakes). However, in the three-dimensional case, attention shall be paid to the location of the critical concentration (see the end of the paper). In two dimensions, $\tau_h = 1 + 2\nu/(1+\nu)$ and $\sigma_h = 1/(1+\nu)$.

To study the system in the presence of a concentration gradient, we will suppose that the scaling behavior of the finite-cluster distribution is essentially preserved. But due to this gradient, there exists an overlap between regions of different concentrations. This fact has already been used to determine the width of the front,[2] writing that the correlation length ξ at a distance σ_f of the percolation threshold is equal to σ_f.

Here we will say very generally that the radius of gyration R_h of a cluster with h sites on its hull cannot be much larger than the distance $x - x_c$ of the center of mass of this cluster from the region of concentration p_c. The radius of gyration has also a scaling behavior which can be written

$$R_h \cong h^{1/D_f} g[(p - p_c) h^{\sigma_h}] . \tag{9}$$

This expresses, in particular, that the perimeter is fractal at p_c with dimension D_f and that the scaling function g depends only on the dimensionless quantity $(p - p_c) h^{\sigma_h}$. Hence, on the average, we cannot find finite clusters with perimeter h such that

$$R_h > x - x_c = (p - p_c)/\nabla p . \tag{10}$$

The same arguments can be used for the lakes in the region $p > p_c$. Thus there exists a cutoff length $h_{\max}(p)$ in

the perimeter distribution defined by

$$R_{h_{\max}}(p)=(p-p_c)/\nabla p \ , \tag{11}$$

or from Eq. (9),

$$(h_{\max}|\nabla p|^{\alpha_\sigma D_f})^{1/D_f} g\{[(p-p_c)/\delta p](h_{\max}|\nabla p|^{\alpha_\sigma D_f})^{\sigma_h}\}=(p-p_c)/\delta p \ ,$$

which is solved to give the largest possible cluster perimeters in a region where the center of mass of the clusters is at concentration p,

$$h_{\max}(p)=h_M\phi[(p-p_c)/\delta p] \tag{13}$$

with

$$h_M=|\nabla p|^{-\alpha_\sigma D_f}=(\delta p)^{-1/\sigma_h} \ .$$

ϕ is an unknown scaling function.

To resume, the perimeters h of the potentially connected clusters have an upper bound $h_{\max}(p)$. This upper bound depends on the average concentration p at the cluster location and is proportional to h_M. h_M is also proportional to the typical perimeter of clusters with radius σ_f. If we consider that the entire front is at p_c, we have, from Eq. (3),

$$h_{\max}(p=p_c)=h_M\phi(0)\propto|\nabla p|^{-\alpha_\sigma D_f}\propto\sigma_f^{D_f}$$

as mentioned in the Introduction. This relation was used in the crude approach.[9] Here $d=2$, the exponent $\alpha_\sigma D_f=1$, and

$$h_{\max}=|\nabla p|^{-1}\phi[(p-p_c)/\delta p] \ .$$

Result (13) will be important relative to the saturation of the fluctuations of $N_f(t)$.

B. The model for the dynamics

During the diffusion process, a certain number of islands (lakes) are close enough to the frontier to allow connection (opening) after the jump of one particle (Fig. 2). The reverse jump will disconnect (close) this island (lake). To simplify the presentation, taking account of the similarity of the scaling laws, we will consider only connection and disconnection of islands. We assume that this connection-disconnection process is such that there is *no correlation* between two successive fluctuations of the length of the front. We consider then the number $\mathcal{M}_h(p)$ per surface unit (i.e., per site) of clusters with h sites on their hulls. These clusters are centered in a region of concentration p and are able to be connected or disconnected by *only one particle jump*. $\mathcal{M}_h(p)$ does not vary with time in a stationary situation. At a given time t, and in a region of concentration p, $m_h(p,t)$ of $\mathcal{M}_h(p)$ are connected, and

$$0\le m_h(p,t)\le\mathcal{M}_h(p) \ .$$

Over the total concentration range the number of connected clusters m_h is

$$h_{\max}^{1/D_f} g[(p-p_c)h_{\max}^{\sigma_h}]=(p-p_c)/\nabla p \ . \tag{12}$$

Equation (12) can be rewritten with the two reduced variables $h_{\max}/h_M=h_{\max}|\nabla p|^{\alpha_\sigma D_f}$ and $(p-p_c)/\delta p$,

$$m_h(t)=L^{d-1}\int dx\, m_h[p(x),t] \tag{14}$$

among M_h defined by

$$M_h=L^{d-1}\int dx\,\mathcal{M}_h[p(x)] \tag{15}$$

and

$$0\le m_h(t)\le M_h \ .$$

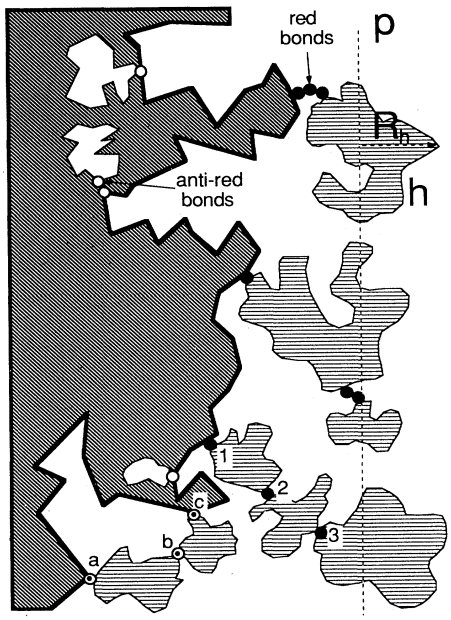

FIG. 2. The diffusion fronts fluctuates by connecting or disconnecting islands (finite clusters in light grey) or by opening or closing lakes (in white). This appears when one "red bond" represented by black or white points is open or closed. A cluster with a concentration p at its center-of-mass location is characterized by its perimeter h and its gyration radius $R_h(p)$. The dark grey region represents the set of particles which remains connected to the source when all the red bonds are open. Its frontier (heavy solid line) is the bare diffusion front (with length $N_{f_{\min}}$). The active clusters can possibly be in series, connected in the figure by the red bonds 0, 1, and 2. In $d=2$, the red bonds can even be not well defined as black or white points. If a, b, and c are closed, a lake is created and a, b, and c would be white points, but if only one of them is open the two others would be black points as they now connect islands. Hence, there are many possibilities of connections or disconnections; however, being in an equilibrium situation, in average the distribution of clusters of size h must satisfy Eqs. (16)–(19).

The total length $N_f(t)$ is given by

$$N_f(t)=N_{f_{\min}}+\sum_h h m_h(t) , \tag{16}$$

$N_{f_{\min}}$ corresponding to all $m_h=0$, and can be considered to be generated by a subset of the front such that at least two jumps are necessary to disconnect a part of it. Here we will make the following assumption: The mass of the front which comes from the contribution of the "one-link" clusters is dominant in such a way that one can neglect the contribution from $N_{f_{\min}}$ (this assumption is based on the fact that when all the "one-link" clusters have been disconnected the $N_{f_{\min}}$ remaining particles can be compared to the accessible perimeter of Aharony-Grossman;[21] this subset has in $d=2$ a fractal dimension $D_e=\frac{4}{3}$).

From Eq. (16) we obtain the equilibrium length of the front,

$$\langle N_f(t)\rangle=N_{f_{\min}}+\sum_h h\langle m_h\rangle_{t=\infty} . \tag{17}$$

The probability $P(m_h,t)$ to have m_h clusters connected at time t is given by a master equation,

$$\begin{aligned}\partial P(m_h,t)/\partial t=&w(m_h+1\to m_h)P(m_h+1,t)\\&+w(m_h-1\to m_h)P(m_h-1,t)\\&-[w(m_h\to m_h+1)\\&\quad+w(m_h\to m_h-1)]P(m_h,t) .\end{aligned} \tag{18}$$

The events of the set of $\mathcal{M}_h$ clusters are assumed to be equally likely, so that the probability

$$\begin{aligned}w(m_h\to m_h-1)&=m_h\pi_h ,\\ w(m_h\to m_h+1)&=(\mathcal{M}_h-m_h)\pi_h ,\end{aligned} \tag{19}$$

where π_h is the elementary probability of connection-disconnection of one particular cluster among the $\mathcal{M}_h$ (for simplification, the connection and the disconnection probabilities are taken to be equal; they are actually expected to be only proportional but this detail is irrelevant for the scaling behavior). Equation (18) can be written in the continuous limit,

$$\frac{\partial P}{\partial t}=2\pi_h P+(2m_h-\mathcal{M}_h)\pi_h\frac{\partial P}{\partial m_h}+\frac{\mathcal{M}_h\pi_h}{2}\frac{\partial^2 P}{\partial m_h^2} . \tag{20}$$

This classical statistical problem is easily solved knowing the propagator,

$$P(m_h,t_0+\tau|m_h^0,t_0)=\frac{1}{\mu(\tau)\sqrt{2\pi}}\exp-\frac{(\xi-\bar{\xi})^2}{2\mu(\tau)^2} , \tag{21}$$

with

$$\mu(\tau)^2=\frac{\mathcal{M}_h}{4}[1-\exp(-2\pi_h\tau)]$$

and

$$\xi=m_h-\frac{\mathcal{M}_h}{2} ,$$

$$\xi^0=m_h^0-\frac{\mathcal{M}_h}{2} ,$$

$$\bar{\xi}=\bar{\xi}(\tau)=\xi^0\exp-2\pi_h\tau=\xi^0\exp-\frac{\tau}{\tau_r} .$$

The quantities $\tau_r=1/2\pi_h(p)$ are the characteristic relaxation times of the system in a region of concentration p. The equilibrium distribution is given simply by

$$P_{\rm eq}(m_h)=[1/\mu_{\rm eq}\sqrt{(2\pi)}]\exp[-\xi^2/(2\mu_{\rm eq}^2)] \tag{22}$$

with

$$\mu_{\rm eq}=\sqrt{\mathcal{M}_h}/2$$

and

$$\xi=m_h-\mathcal{M}_h/2 .$$

The equilibrium value of m_h is $\mathcal{M}_h/2$ so that from Eqs. (14) and (15), the average equilibrium value of m_h will be $M_h/2$. From this, we obtain the equilibrium length of the front,

$$\begin{aligned}\langle N_f(t)\rangle&=\langle N_{f_{\min}}\rangle+\sum_h h\langle m_h\rangle_{t=\infty}\\&=\langle N_{f_{\min}}\rangle+\sum_h hM_h/2 .\end{aligned} \tag{23a}$$

The dominant contribution is given by the last sum, $\sum_h hM_h/2$, which will have the same gradient dependence as

$$\langle N_f(t)\rangle\propto|\nabla p|^{-\alpha_N} . \tag{23b}$$

We suppose that $\mathcal{M}_h(p)$ is proportional to the front density p_f because we can consider that the larger the front density, the larger the number of single jump connected clusters. $\mathcal{M}_h$ must also be proportional to the density $\tilde{n}_h$ of clusters ($\tilde{n}_h$ is a subset of the clusters having a perimeter h) which are close enough to the front to be connected in a single jump. $\tilde{n}_h$ will have a power-law dependence in h with an exponent $-\chi$, times a reduced function depending on $(p-p_c)h^{\sigma_h}$ as for n_h [Eq. (8a)].

$\mathcal{M}_h(p)$ must finally contain a cutoff function C which takes into account limitation on the size h (this cutoff could have equally been included in $\tilde{n}_h$). We will check the correctness of this assumption by calculating $\langle N_f(t)\rangle$, recovering its power-law behavior, and finding the correct average value for the number of large fluctuating clusters. We have

$$\mathcal{M}_h\cong h^{-\chi}\mathcal{F}[(p-p_c)h^{\sigma_h}]p_f(x)C(h/h_{\max}) . \tag{24}$$

Exponent χ will be determined using the result of Eqs. (23a) and (23b). Replacing $\sum_h hM_h/2$ in expression (23) by integrating over h and x leads to

$$\langle N_f(t)\rangle \cong L^{d-1}\int_0^\infty dh\, h^{1-\chi}\int dx\, \mathcal{F}[(p-p_c)h^{\sigma_h}] \times C\left[\frac{h}{h_{\max}}\right] p_f(x)$$
$$\propto |\nabla p|^{-\alpha_N}(h_M)^{2-\chi} \equiv |\nabla p|^{-\alpha_N}, \quad (25)$$

which implies $\chi=2$.

(i) The above approach supposes that the concentration varies slowly on a size R_h of the clusters contributing to N_f.

(ii) The average value for the number of large fluctuating clusters is correctly recovered as expected in the preliminary approach (see Sec. I C),

$$\langle \mathcal{M}_h(t)\rangle \cong L^{d-1}\int_{kh_M}^\infty dh\, h^{-\chi}\int dx\, \mathcal{F}[(p-p_c)h^{\sigma_h}] \times C\left[\frac{h}{h_{\max}}\right] p_f(x)$$
$$\propto L^{d-1}|\nabla_p|^{-\alpha_N}(h_M)^{-1} \propto (L/\sigma_f)^{d-1}.$$

C. Determination of the connection frequency π_h

We conjecture that the probability π_h is proportional to the density of red bonds in the region occupied by a cluster of size R_h. In doing so, we consider that the number of red bonds connecting a cluster of perimeter h to the front follows the same power law as the red bonds present in this region:[22,12]

$$\pi_h \propto (R_h)^{1/\nu} \quad (26a)$$

or from (9) (and as $\sigma_h \nu D_f = 1$),

$$\pi_h = h^{\sigma_h} g[(p-p_c)h^{\sigma_h}]^{1/\nu}. \quad (26b)$$

The probability π_h is per unit time θ.

III. CALCULATION OF THE AUTOCORRELATION FUNCTION $\langle \Delta N_f(t)^2\rangle$

It is now easy to calculate the autocorrelation function

$$\langle \Delta N_f(t)^2\rangle = \langle [N_f(t)-N_f(0)]^2\rangle$$

using relations (14) and (15),

$$\langle (\Delta N_f(t)^2\rangle = \sum_h \sum_{m_h^0} \sum_{m_h} h^2 P(\{m_h\}t|\{m_h^0\}0) m_h m_h^0 P_{\rm eq}(\{m_h^0\})$$
$$= L^{d-1}\int dx \sum_h \sum_{m_h^0} \sum_{m_h} h^2 P(m_h t|m_h^0 0) m_h m_h^0 P_{\rm eq}(m_h^0). \quad (27)$$

From Eqs. (21) and (22) this becomes

$$\langle [\Delta N_f(t)]^2\rangle = 2L^{d-1}\int dx \sum_h h^2 \mu_{\rm eq}^2 [1-\exp(-2\pi_h t)]. \quad (28)$$

Expression (28) is the basic relation for the noise behavior.

For short times $t \ll \tau_r = 1/2\pi_h$, we write

$$\langle [\Delta N_f(t)]^2\rangle = \left[2L^{d-1}\int dx \sum_{h=1}^{h_{\max}} h^2 \mathcal{M}_h \pi_h\right] t = \mathcal{A} t. \quad (29)$$

This expression shows that the short-time behavior corresponds in Eq. (7) to $H=\frac{1}{2}$ (as in the Brownian motion). The linear dependence in t is verified by numerical calculations. We find $2H=1.00\pm0.02$ [Fig. 3(a)]. This confirms the independence hypothesis of the connection-disconnection process. In numerical simulation, the time t is the number of jump trials per occupied site in the diffusion process. The coefficient $\mathcal{A}$ of t in Eq. (29) has been determined *numerically* in two dimensions [Fig. 3(c)],

$$\mathcal{A} \cong 183.4|\nabla p|^{-1.83}. \quad (30)$$

If we put the scaling structure of π_h given by [26(b)] into Eq. (29) we obtain

$$\mathcal{A} \cong L^{d-1}\int_0^\infty dh\, h^{\sigma_h}\int dx\, \mathcal{G}((p-p_c)h^{\sigma_h}) \times C\left[\frac{h}{h_{\max}}\right] p_f(x),$$

where the function $\mathcal{G}$ is simply $\mathcal{F}g^{-1/\nu}$. Setting $u=(p-p_c)/\delta p$ and $v=h/h_M$, we obtain from (13), admitting that the integral on the right-hand sides converges,

$$\mathcal{A} \propto L^{d-1}|\nabla p|^{-(2D_f-d)\nu/(1+\nu)-1}$$
$$\underset{d=2}{\propto} |\nabla p|^{-13/7}. \quad (31)$$

The exponent $\frac{13}{7}=1.857\ldots$ is very close to the numerical value $1.83\ldots$ of Eq. (30).

For large times Eq. (28) gives

$$\langle [\Delta N_f(t)]^2\rangle = 2L^{d-1}\int dx \sum_{h=1}^{h_{\max}} h^2 \mu_{\rm eq}^2, \quad (32)$$

which corresponds to a plateau in the variation of $\langle \Delta N_f(t)^2\rangle$ as a function of time and $H=0$ [see Fig. 3(a)]. The gradient dependence of the height of this plateau is determined using (22) and (24),

$$\langle [\Delta N_f(t)]^2\rangle \propto L^{d-1}|\nabla p|^{-(2D_f-d+1)\nu/(1+\nu)}$$
$$\underset{d=2}{\propto} L|\nabla p|^{-10/7}. \quad (33)$$

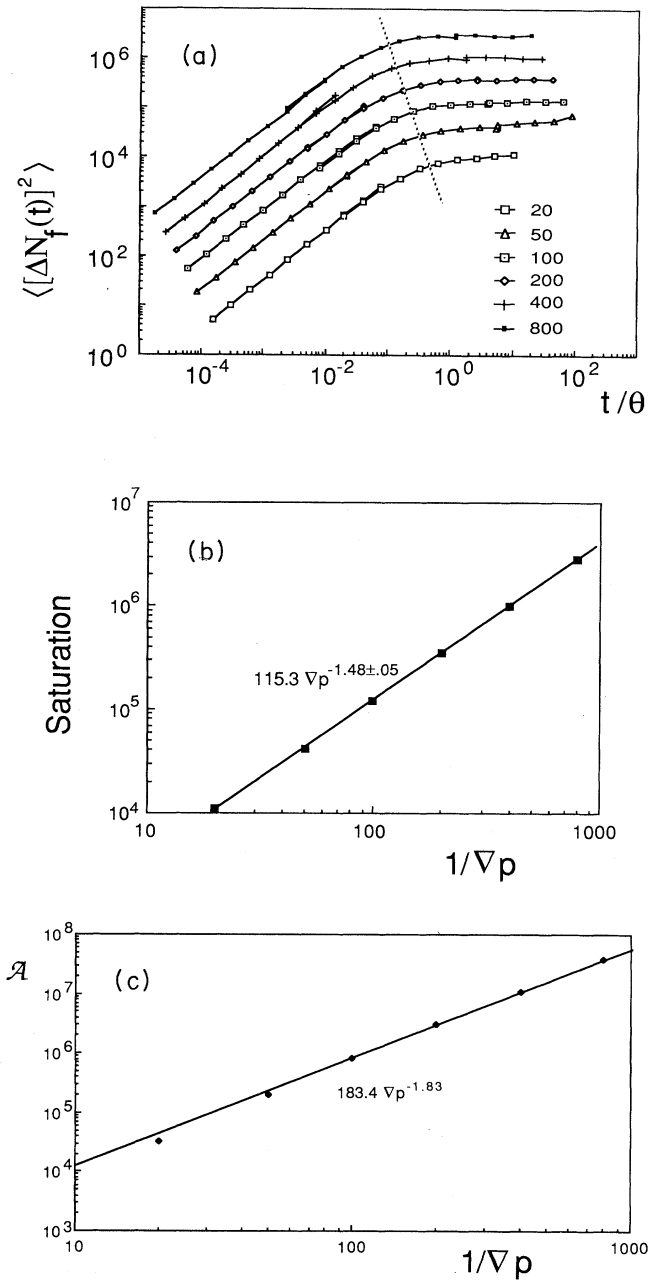

FIG. 3. (a) The graph shows the fluctuation $\langle \Delta N_f(t)^2 \rangle$ for different values of the inverse of the gradient $|\nabla p|^{-1}=20$, 50, 100, 200, 400 and 800. Times t are measured in Monte Carlo steps (MCS) per particle (taking $\theta=1$). The curves are obtained using three time interval scales, namely 1, 128, 2^{16} steps (MCS/sample). The saturation value of $\langle \Delta N_f(t)^2 \rangle$ as a function of the inverse of the concentration gradient has an exponent 1.48 close to the theoretical value $\frac{10}{7}$ obtained in $d=2$. The exponent for the coefficient $\mathcal{A}$ of the linear time variation is found 1.83 also reasonably close to the theoretical value $\frac{13}{7}$. (b) Saturation value of $\langle \Delta N_f(t)^2 \rangle$ as a function of the inverse of the concentration gradient. The slope 1.48 obtained is close to the theoretical value $\frac{10}{7}$. (c) Coefficient $\mathcal{A}$ of the linear time variation as a function of the inverse of the gradient. The slope 1.83 is reasonable close to the theoretical value $\frac{13}{7}$.

This behavior is relatively well verified by the *numerical* calculations where ($L=524$) [Fig. 3(b)],

$$\langle [\Delta N_f(t)]^2 \rangle \cong 115.3 |\nabla p|^{-1.48} . \tag{34}$$

The theoretical value is $\frac{10}{7} \cong 1.43$.

IV. THE CROSSOVER TIME t_c

The crossover time t_c is the time at which the fluctuations saturate, i.e., the time at which we observe a crossover from $1/f^2$ to $1/f$ noise. t_c is defined by the relation

$$\mathcal{A} t_c = \langle \Delta N_f(t=\infty)^2 \rangle .$$

Comparing the power-law relations (31) and (33), we find the remarkable power-law behavior

$$t_c \propto |\nabla p|^{1/(1+\nu)} . \tag{35}$$

We observe that the crossover time is also the average (over h and p) of the relaxation times τ_r of the system,

$$t_c \cong \langle \tau_r \rangle = \langle (2\pi_h)^{-1} \rangle \propto (h_M)^{\sigma_h} \propto |\nabla p|^{1/(1+\nu)} .$$

The crossover time varies as the inverse of the number of red bonds in a volume of linear size σ_f. The larger the diffusion time, the smaller the crossover time. The crossover time is expected to be independent on L because two large identical samples and their union must have the same t_c.

V. THE DENSITY OF EVENTS OF SIZE h

The density of events $N_{\rm ev}(h)$ is defined as follows: $N_{\rm ev}(h)dh$ is the average number of occurrences per unit time when the frontier varies by a length between h and $h+dh$. In the numerical calculation the frontier length is determined after each particle jump. $N_{\rm ev}(h)$ represents the number of events of size h after each particle has made (in average) one jump.

The density of events in a region of concentration p is

$$N_{\rm ev}(h) = \frac{L^{d-1}}{2} \int dx \, \mathcal{M}_h(p) \pi_h(p) . \tag{36}$$

From Eqs. (24) and (26b),

$$N_{\rm ev}(h) \cong \frac{L^{d-1}}{2} h^{\sigma_h - 2} \int dx \, \mathcal{G}((p-p_c)h^{\sigma_h}) \times C\left[\frac{h}{h_{\max}}\right] p_f(x)$$

or from Eqs. (6a) and (13),

$$N_{\rm ev}(h) = \frac{L^{d-1}}{2} h^{\sigma_h - 2} |\nabla p|^{-\alpha_N} \times \int du \, C\left[\frac{h}{h_M \phi(u)}\right] \mathcal{G}\left[u \left[\frac{h}{h_M}\right]^{\sigma_h}\right] \Pi_f(u) ,$$

which takes the general form

$$N_{\rm ev}(h) = \frac{L^{d-1}}{2} h^{\sigma_h - 2} |\nabla p|^{-\alpha_N} K\left[\frac{h}{h_M}\right] , \tag{37}$$

where $\alpha_n \cong 0.428$ and $2-\sigma_h = \frac{11}{7} \simeq 1.57$. K is a cutoff function resulting from the integration of the above expression. We notice that for $d=2$, the exponent $2-\sigma_h$ of h is precisely the exponent Υ_2 of polymers at the θ point:[12] $\Upsilon_2 = 2 - d_{\text{red}}/D_f$ where $d_{\text{red}} = 1/\nu$, in agreement with expression (8b). This approach here shows that this expression also holds in $d=3$ when h is a perimeter surface.

Numerical calculations show that $N_{\text{ev}}(h)$ is the product of power-law behaviors in $|\nabla p|$ and h, and a cutoff function $K(h|\nabla p|)$ (Fig. 4),

$$N_{\text{ev}}(h) = 73.5|\nabla p|^{-0.41} h^{-1.40} K(h|\nabla p|) .$$

This is in reasonable agreement with Eq. (37).

VI. SUMMARY AND CONCLUSION

The noise generated during a diffusion process presents two main behaviors separated by a crossover time t_c such that

$$t_c \propto |\nabla p|^{1/(1+\nu)} .$$

For short times, $t < t_c$, the noise varies linearly with time,

$$\langle [\Delta N_f(t)]^2 \rangle = \mathcal{A} t , \tag{38}$$

where

$$\mathcal{A} \propto L^{d-1} |\nabla p|^{-1-(2D_f-d)\nu/(1+\nu)} \underset{d=2}{\propto} L|\nabla p|^{-13/7} . \tag{39}$$

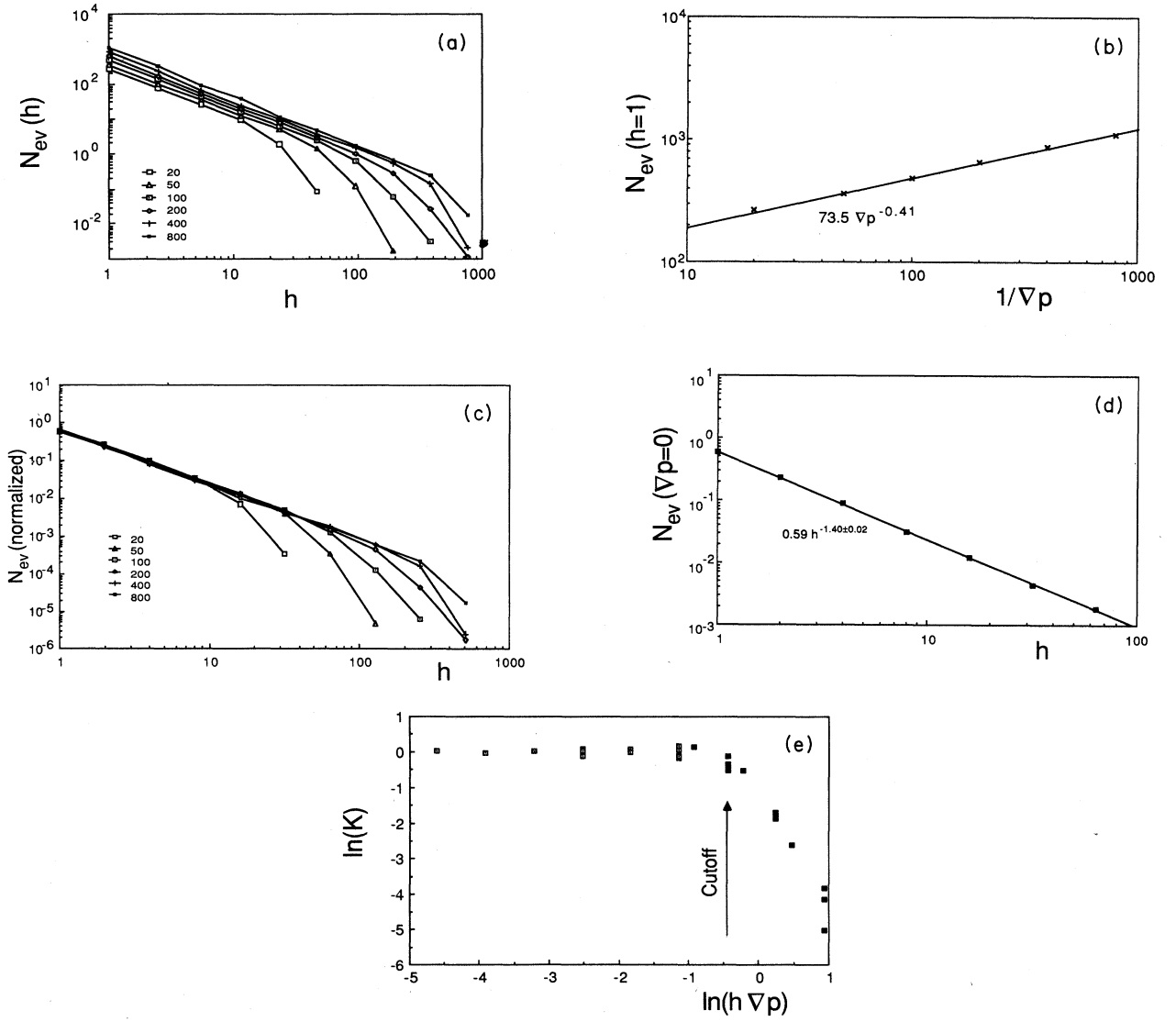

FIG. 4. (a) Density of events N_{ev} of size h for different values of the gradient. N_{ev} contains three factors. A gradient-dependent factor with a power-law dependence is shown in (b). The slope 0.41 is closed to the theoretical value $\alpha_N = \frac{3}{7}$. A scaling structure (c) also with a power-law dependence is shown in (d). The corresponding exponent is around 1.40 while the theoretical value is found to be $\frac{11}{7} \cong 1.57$. A cutoff function $K(h|\nabla p|)$ is represented in (e).

For large times, $t > t_c$, the noise saturates at a value which has again a power-law dependence in $|\nabla p|$,

$$\langle [\Delta N_f(t)]^2 \rangle \propto L^{d-1} |\nabla p|^{-(2D_f - d + 1)\nu/(1+\nu)} \underset{d=2}{\propto} L |\nabla p|^{-10/7} . \quad (33)$$

The noise is generated by events whose density is h dependent and $|\nabla p|$ dependent,

$$N_{\rm ev}(h) = \frac{L^{d-1}}{2} h^{\sigma_h - 2} |\nabla p|^{-\alpha_N} K\left(\frac{h}{h_M}\right) . \quad (37)$$

In these expressions $\sigma_h = 1/(\nu D_f)$, $\alpha_N = (D_f - d + 1)\nu/(1+\nu)$, and $h_M = |\nabla p|^{-(\nu D_f/1+\nu)}$. The cutoff function K cuts the sizes $h > h_M$.

We expect that the above expressions are not restricted to two-dimensional (2D) systems. The scaling hypothesis represented here appears to also be valid in three dimensions. Differences exist[6,7] between the 2D and 3D systems. In three dimensions, the front is extended between two different concentration limits [$0.312 \leq p \leq 0.9$, for the simple-cubic lattice (SC)] so that the fluctuating regions are located around the front tail corresponding to the percolation threshold p_c ($\cong 0.312$ for the SC lattice) and around the back of the front corresponding to a concentration $1 - p_c'$ ($\cong 0.9$ for the SC lattice, p_c' being the percolation threshold of the complementary 1,2,3-SC lattice). Only clusters of occupied particles with scaling behavior [Eq. (8a)] particles are located in the back of the front (in $d = 2$ this happens in the same concentration region). For $d = 3$, the fluctuating regions have a width σ_{ft} given by the same expression (3) as in $d = 2$. Also for $d = 3$, the number of particles situated in these fluctuating regions is given by Eqs. (5a) and (5b), with the critical exponents

$$\nu \simeq 0.88 \quad \text{and} \quad D_f \cong 2.52 .$$

The present approach could be applied to the study of the fluctuations of the geometry of a diffused contact[8] which should play a role in the noise generated by such a contact. It is also interesting to remark that a similar physical situation occurs in the slow invasion of a porous medium by a nonwetting fluid in the presence of gravity.[16,17] When the fluid pressure is very slowly increased, we observe a devil staircase evolution of the flow, in which case the fluctuations involve the *size* of the connected clusters. Such an evolution has also been studied in the absence of gravity.[15] A detailed study is in progress.

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