

Diffusion in disordered media with long-range correlations: Anomalous, Fickian, and superdiffusive transport and log-periodic oscillations

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We present the results of extensive Monte Carlo simulation of diffusion in disordered media with long-range correlations, a problem which is relevant to transport of contaminants in field-scale porous media, such as aquifers, gas transport in soils, and transport in composite materials. The correlations are generated by a fractional Brownian motion characterized by a Hurst exponent H . For $H > 1/2$ the correlations appear to have no effect, and the transport process is diffusive. However, for $H < 1/2$ and depending on the morphology of the medium, three distinct types of transport processes, namely, anomalous, Fickian, and superdiffusive transport may emerge. Moreover, if the medium is anisotropic and stratified, biased diffusion in it is characterized by power-law growth of the mean square displacements with the time in which the effective exponents characterizing the power-law oscillates log periodically with the time. This result cannot be predicted by any of the currently available continuum theories of transport in disordered media.

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I. INTRODUCTION

Transport processes in disordered media constitute an important class of problems, in view of their relevance to the modeling of a wide variety of phenomena in natural and industrial processes. A partial list of their applications includes flow and transport phenomena in porous media, diffusion in soil and through biological tissues, conduction through composite solids, and many more. Many transport processes in heterogeneous media are nonlocal in the sense that, they involve long-range correlations. By long range we mean the correlation length is comparable with the linear extent of the medium. Such correlations either exist in the morphology of the heterogeneous media, or are induced by the transport processes themselves. A well-known example of the first type of long-range correlations is those that exist in geological formations [1]. An example of the second type of long-range correlations is those that arise in vector transport in rigidity percolation, or in mechanical fracture of materials [2,3]. Nonlocal transport processes are highly complex and often, because of the long-range correlations, a continuum formulation of them is not possible or even meaningful.

However, despite their significance, most of the transport processes in disordered media that have been studied over the past several decades involve no correlations, or at most short-range correlations. Geometrical fractals, such as the sample-spanning percolation cluster (SSPC) at the percolation threshold [4,5] p_c do induce long-range correlations, and transport in such systems has been studied extensively [6]. However, long-range correlations in the SSPC at p_c are due to the poor connectivity of the cluster that gives rise to tor-

tuos transport paths. Diffusion and other forms of molecular transport in a well-connected disordered medium (i.e., one far from the percolation threshold) in which there exist long-range correlations in the distribution of the local or *microscopic* transport properties, have received much less attention. The interest in such problems is more than academic: They are in fact relevant to modeling of a variety of phenomena of practical importance. For example, as mentioned earlier, geological formations, such as soil, oil reservoirs, and groundwater aquifers have been shown to contain such correlations [1]. Thus, for example, understanding of diffusion of gases in soil is an important problem that is essential to minimizing the potential hazards to the environment that arise as a result of migration of polluting gases in soil. Groundwater aquifers are usually contaminated by transport of pollutants that spreads the hazardous materials in the system. Many other systems of scientific and industrial importance involve long-range correlations [7], and thus study of diffusion and other types of transport processes in them is essential.

The purpose of this paper is to study diffusion in a disordered medium that is characterized by a distribution of local conductances that contains long-range correlations. Our main interest in this problem is to understand whether diffusion of pollutants in soil can be modeled by the classical diffusion equation with a constant diffusivity, or whether the presence of the correlations gives rise to a nonlocal transport process at the macroscopic level that cannot be represented by the diffusion equation. In the latter case, one must develop the appropriate transport equation.

The plan of this paper is as follows. In the following section, we describe the model of the disordered medium and its conductance distribution. We then describe the details of Monte Carlo simulations that we utilize to study diffusion in the medium. In Sec. IV, we present the results and discuss their implications. Section V contains a summary of the results.

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II. GENERATION OF LONG-RANGE CORRELATIONS

We represent the disordered medium by a square lattice in which the bonds represent the transport paths. Each bond is assigned a conductance g . To assign the bond conductances and generate long-range correlations between them, we use the following two methods.

(1) In the first method, we assume that the distribution of the conductances is a fractional Brownian motion (FBM). This assumption is based on the discovery that the statistics of the permeabilities and hydraulic conductances of oil reservoirs [8], groundwater aquifers [9], and soils [1] follow a FBM. Briefly, the FBM is a stochastic process $B_H(r)$ [10] with the properties that $\langle B_H(\mathbf{r}) - B_H(\mathbf{r}_0) \rangle = 0$, and

$$\langle [B_H(\mathbf{r}) - B_H(\mathbf{r}_0)]^2 \rangle \sim |\mathbf{r} - \mathbf{r}_0|^{2H}, \quad (1)$$

where $\mathbf{r} = (x, y, z)$ and $\mathbf{r}_0 = (x_0, y_0, z_0)$ are two arbitrary points in space, and H is the Hurst exponent. The main property of the FBM is that it generates correlations with an extent that is *infinite* (i.e., the extent of the correlations is as large as the linear size of the system). Moreover, the type of the correlations can be tuned by varying H . For $H > 1/2$ the FBM displays persistence or positive correlations, i.e., a trend (for example, a high or low value of the conductances) at \mathbf{r} is likely to be followed by a similar trend at $\mathbf{r} + \Delta\mathbf{r}$, whereas for $H < 1/2$ the FBM generates antipersistence or negative correlations, i.e., a trend at \mathbf{r} is likely to be followed by its opposite at $\mathbf{r} + \Delta\mathbf{r}$. For $H = 1/2$ the trace of the FBM is similar to that of a random walk and the increments are uncorrelated. A convenient way of representing a stochastic function is through its spectral density $S(\boldsymbol{\omega})$, the Fourier transform of its variance. For a d -dimensional FBM it can be shown that [10]

$$S(\boldsymbol{\omega}) = \frac{a_0}{\left(\sum_{i=1}^d \omega_i^2 \right)^{H+d/2}}, \quad (2)$$

where $\boldsymbol{\omega} = (\omega_1, \dots, \omega_d)$, with ω_i being the Fourier component in the i th direction, and a_0 is a constant. The variance of the FBM depends on the size of the system, and diverges for a large enough system. Thus, the broadness of the conductance distribution with FBM-type correlations increases with the size of the system. Note that the correlation function $C(r)$ of the FBM is given by

$$C(r) - C(0) \sim r^{2H}, \quad (3)$$

so that as long as $H > 0$, which is physically the case, the correlation function *increases* with increasing r .

(2) In the second case, we assume that $\log g$ follows the statistics of the FBM. The reason for this assumption is that, it has been shown [11] that the distribution of the permeabilities and hydraulic conductances of some porous media is so broad that cannot be described by a FBM, rather the logarithm of the conductances seem to follow the FBM. Therefore, this case represents a disordered medium with a very broad distribution of the conductances which, however, contains long-range correlations.

III. MONTE CARLO SIMULATION

We used the power spectrum method to generate the FBM distribution. All of the results presented in this paper were obtained with 1024×1024 lattices. This size of the lattice gives rise to a conductance distribution the broadness of which is about 2–3 orders of magnitude variations in the bonds' conductances. Periodic boundary conditions were used in all the directions. The diffusion process was simulated by the random walk of a particle that is initially (at time $t = 0$) inserted into the lattice at a randomly selected site. The particle executes a random walk between the nearest neighbor sites of the lattice. Each step of the walk from one site to another is taken with a transition probability proportional to the conductance of the bond between the two sites. After each step, the time t is increased by one unit. The mean square displacements (MSD) $\langle R^2(t) \rangle$ of the walkers at time t are computed, where the averaging is taken over the initial positions of the walkers, and the different realizations of the lattice. Typically, we used 4000 walkers (i.e., 4000 initial positions) and 40 realizations of each of the conductance distributions. All the random walkers took 2×10^6 steps (i.e., the MSD were computed up to time $t = 2 \times 10^6$). Moreover, to study the effect of the nature of the correlations, we used several values of the Hurst exponent H .

To characterize the diffusion process, we write the MSD of the particles as

$$\langle R^2(t) \rangle \sim t^\alpha, \quad (4)$$

where $D_w = 2/\alpha$ is the fractal dimension of the walk [6]. Normally, depending on how $\langle R^2(t) \rangle$ grows with t , one may have three distinct transport regimes. (1) $D_w = 2$, i.e., $\langle R(t)^2 \rangle$ grows linearly with t , and therefore diffusion is Fickian, i.e., the effective diffusivity D , defined by $\langle R^2(t) \rangle = 2Ddt$ (d is the dimensionality of the system), is a constant. (2) $D_w > 2$, which implies that $\langle R^2(t) \rangle$ grows with t slower than linearly. In this case diffusion is called anomalous [6,12] or fractal [13]. In this regime, $D \rightarrow 0$ as $t \rightarrow \infty$. Diffusion in geometrical fractals, such as the SSPC, is anomalous. (3) If $D_w < 2$, then $\langle R^2(t) \rangle$ grows with t faster than linearly. This type of transport process is called *superdiffusion* [14]; in this case, $D \rightarrow \infty$ as $t \rightarrow \infty$. The results presented below indicate that disordered media of the type that we study in this paper not only can give rise to these three types of diffusion processes, but also to a new type in which the random walk fractal dimension D_w *varies with the time*.

IV. RESULTS AND DISCUSSIONS

We have carried out extensive simulations of diffusion in disordered media of the type described above with a variety of conductance distributions and morphologies. In what follows we present the results and discuss their implications.

A. Isotropic media

Figure 1 presents the MSD vs the time t for three values of H . In these systems, the conductance distribution is represented by the FBM. Analysis of the results based on Eq. (4)

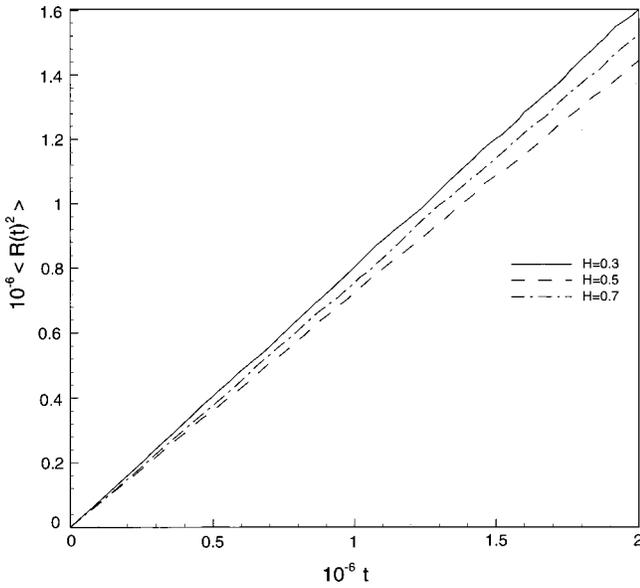


FIG. 1. Mean square displacements $\langle R^2(t) \rangle$ vs the time t for three values of the Hurst exponent H . The conductances are distributed according to the FBM.

indicates that for all values of H , even at relatively short times, diffusion is Fickian, i.e., $\alpha = 1$ ($D_w = 2$). Consider next the case in which the logarithm of the bonds' conductances follows the FBM. Figure 2 presents the results. At short times, diffusion is anomalous for all values of H , as expected. However, while for $H > 0.5$ the long time behavior of the diffusion process approaches the Fickian regime, for $H < 0.5$, it is not; instead it is anomalous with $\alpha < 1$. To obtain the true asymptotic values of $\alpha(H)$, we fit the MSD data to Eq. (4) using the data with $t \geq t_0 = 5 \times 10^5$ and obtain an estimate of α . We then fit the data to Eq. (4) again but with $t \geq t_0 = 10^6$ and obtain a second estimate of α . By increasing

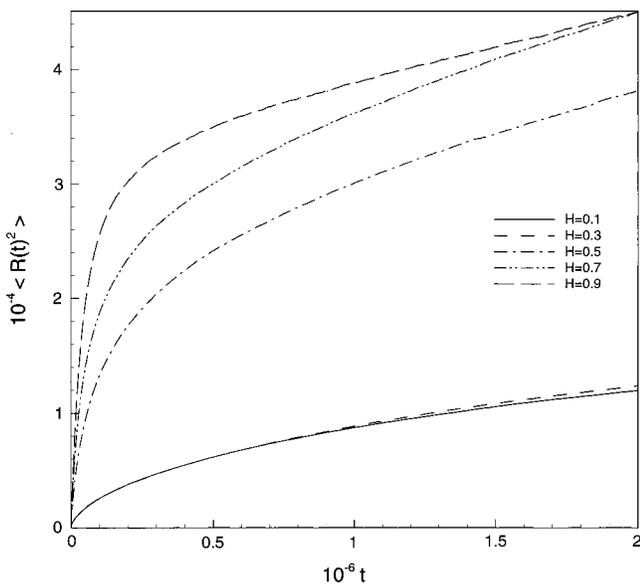


FIG. 2. Same as in FIG. 1, except that the logarithm of the conductances follows the FBM statistics.

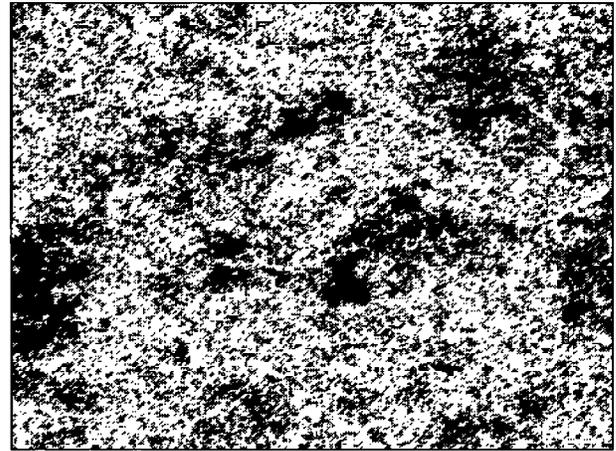


FIG. 3. The map of the conductance distribution with $H=0.3$. All the low conductances (smaller than a fixed threshold) are black.

t_0 we obtain a series of α , which when extrapolated to $t_0 \rightarrow \infty$, yields the true asymptotic value of α . When this was done for all the data obtained for $H > 1/2$, we always obtained $\alpha = 1$, whereas we obtained $\alpha \approx 0.81, 0.89,$ and 0.97 for $H = 0.1, 0.3,$ and 0.5 , respectively. The case $H = 1/2$ represents a borderline limit, indicating that diffusion is almost Fickian.

To understand these results better, we define a conductance threshold ϵ , where ϵ is defined as a fraction of the largest bond conductance in the system, and prepare a map of the local conductances. Figure 3 presents the results for $H = 0.3$, while Fig. 4 shows the same for $H = 0.8$, where the same ϵ was used in both cases. In these figures, all the conductances that are less than ϵ are shown as the black areas, while those that are larger than ϵ represent the white regions. In the case of the $H = 0.8$ system, there are well-connected, but separated, regions of the low- and high-conductance regions. Therefore, if the number of the particles and the simulation times are both large enough, one expects asymptotically Fickian diffusion, consistent with our numerical estimate of $\alpha = 1$. In contrast, in the case of $H = 0.3$ (Fig. 3) most of the medium consists of islands of very low conductances dispersed in the high-conductance patches. Thus, the



FIG. 4. Same as in FIG. 3, but for $H=0.8$.

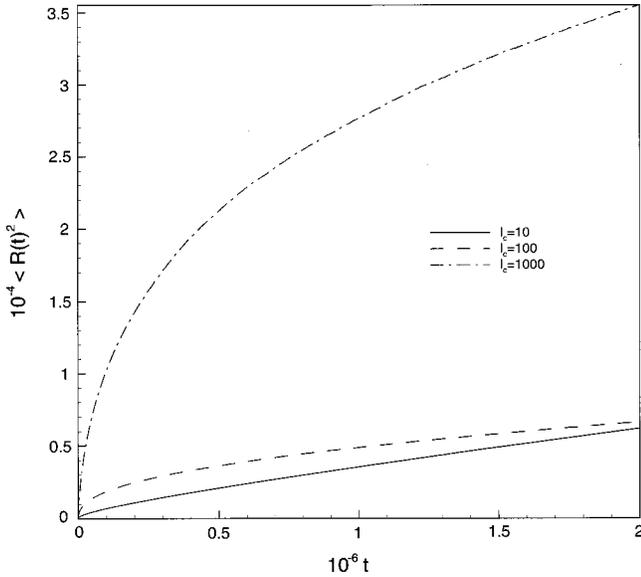


FIG. 5. Mean square displacements vs the time t for three cutoff lengths l_c for the correlations. The logarithm of the conductances follows the FBM statistics with $H=0.3$.

vast majority of the particles begin their diffusion in the low-conductance islands, and can hardly escape from them for long periods of time, as a result of which diffusion is slow and anomalous.

Next, we introduce a cutoff length scale l_c for the extent of the correlations, and study its effect. To include l_c in the conductance distribution, we rewrite the spectral density of FBM as

$$S(\omega) = \frac{a_0}{\left(\omega_c + \sum_{i=1}^d \omega_i^2 \right)^{H+d/2}}, \quad (5)$$

where $\omega_c = 1/l_c^2$. Thus, for length scales $L < l_c$ the conduc-

tances are correlated and follow the statistics of FBM, while for $L > l_c$ they are uncorrelated and randomly distributed. We may expect to have Fickian diffusion when $\langle R^2(t) \rangle^{1/2} \gg l_c$, while in the opposite case we should have anomalous diffusion. Figure 5 presents the resulting MSD vs time t for $H=0.3$ and three values of the cutoff length scale (measured in units of the lattice bonds), when the logarithm of the conductances follow FBM. Analysis of the data for the MSD indicates that in all the cases the asymptotic value of α is unity, confirming our expectations. The main difference between the three cases studied is the rate of convergence of the transport process to the asymptotic Fickian regime: While for small values of l_c diffusion of the particles quickly becomes Fickian, in the case of large values of l_c it takes a long time to reach the Fickian regime. The crossover between the Fickian and anomalous diffusion regime should take place at a crossover time t_c such that

$$t_c \sim l_c^\beta. \quad (6)$$

Figure 6 presents a plot of $\log t_c$ vs $\log l_c$, which indicates that $t_c \sim l_c^\beta$ with $\beta \approx 1.91 \pm 0.10$, consistent with Eq. (6). The significance of Eq. (6) is that, if measurements of the transport properties of such disordered media are done at time $t < t_c$, the resulting transport coefficients would be time dependent, and therefore, it is critical to identify the extent of the correlations in heterogeneous media in order to ensure that the measurements are carried out at sufficiently long times after the diffusants have been introduced into them.

B. Anisotropic media

We also investigated diffusion in anisotropic disordered media that contain long-range correlations. There are several ways of generating an anisotropic medium. For example, the distribution of the bonds' conductances may be direction dependent. Alternatively, one may consider disordered media that are stratified and contain a distribution of layers with

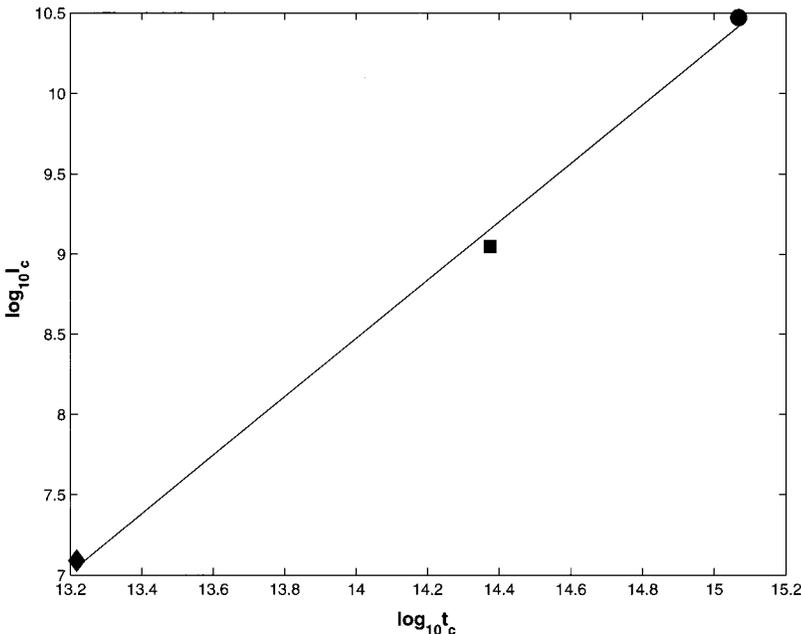


FIG. 6. Scaling of the crossover time t_c with the cutoff length scale l_c . The results are for $l_c = 10$ (\diamond), $l_c = 100$ (\square), and $l_c = 250$ (\circ).

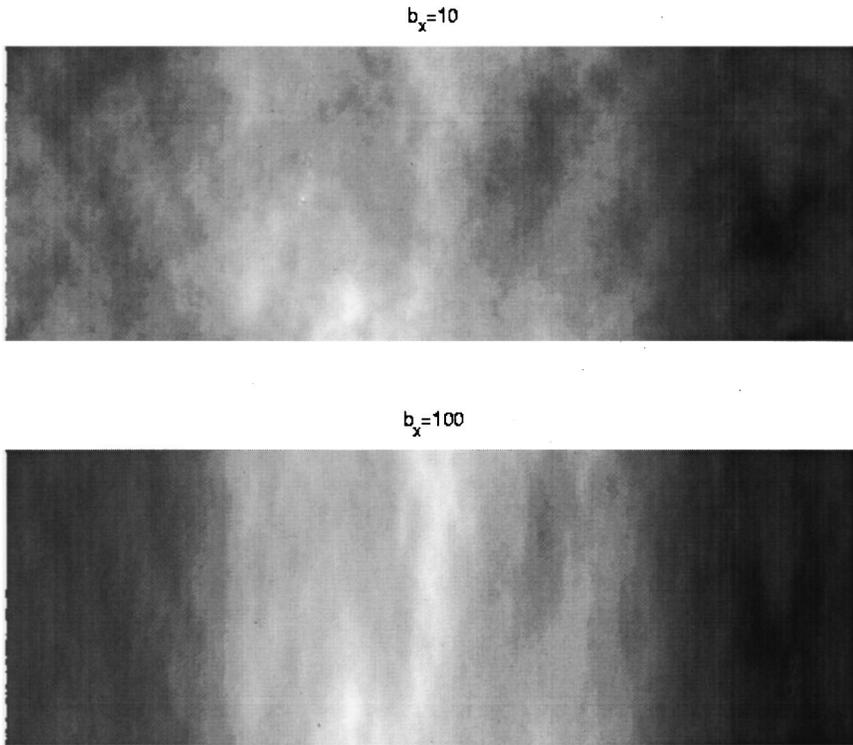


FIG. 7. Two typical anisotropic media with two anisotropy factors b_x and $H=0.7$. Note that increasing b_x increases stratification.

large conductance contrasts between the layers. Such disordered media, which are anisotropic, are used in this study. Our use of such disordered media is motivated by the fact that many natural disordered media, such as rock, oil reservoirs, and groundwater aquifers are typically layered. In addition, many semiconducting materials, such as the family of dichalcogenides of transition metals TX_2 have a layered morphology, and therefore are highly anisotropic in that, they contain a preferred direction for conduction and transport. To generate a stratified medium, we use the spectral representation of FBM, and rewrite it as

$$S(\omega) = \frac{a_d}{(b_x \omega_x^2 + \omega_y^2)^{H+d/2}}, \quad (7)$$

where b_x is a constant, such that $b_x > 1$ generates strata that are essentially parallel to each other in the direction perpendicular to the x direction. Figure 7 presents two examples of such anisotropic disordered media, where the x direction is the horizontal direction of the figure, and the logarithm of the conductances follows the statistics of FBM with $H=0.3$. Note that increasing b_x increases the number of strata of the medium. Because of the anisotropy of the medium, we must calculate MSD separately for each principal direction, so that we write

$$\langle R_x^2(t) \rangle \sim t^{\alpha_x}, \quad \langle R_y^2(t) \rangle \sim t^{\alpha_y}. \quad (8)$$

Our simulations indicate that for $H > 1/2$ diffusion in these anisotropic media is Fickian. However, for $H < 1/2$, the extent of stratification greatly influences the nature of the transport process. When the number of strata is small, i.e., when the system is almost isotropic, diffusion in both directions is not Fickian, in agreement with the results for isotropic

media described above. Figure 8 presents the typical MSD in the x and y directions, and analysis of these data indicated that $\alpha_x < 1$ and $\alpha_y < 1$. However, as the number of the strata increases, the nature of transport in the two directions becomes very different. In the y direction, i.e., parallel to the strata, diffusion is not Fickian, since when the number of the strata is large, each stratum is a narrow, essentially one-dimensional and highly disordered channel, and therefore, one expects to have non-Fickian diffusion [7]. For example, for $H=0.3$ we obtain $\alpha_y \approx 0.88$. However, diffusion

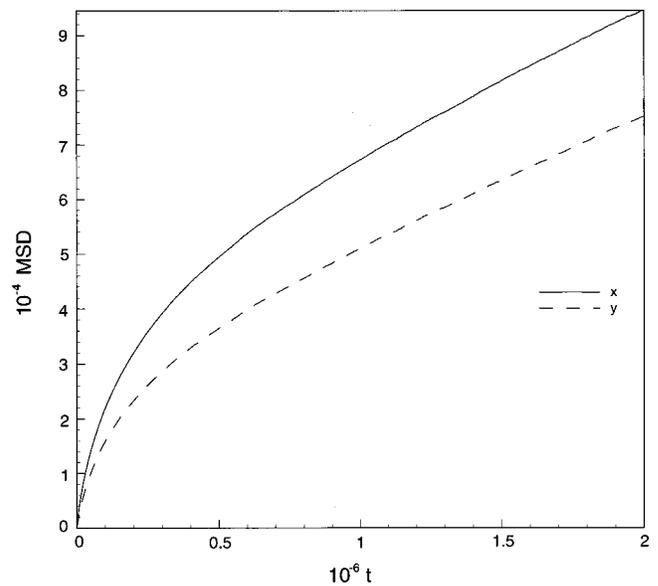


FIG. 8. Mean square displacements vs time in the direction perpendicular to the strata (the x direction) and parallel to the strata (the y direction) with anisotropy factor $b_x=10$.

in the x direction, i.e., in the direction perpendicular to the strata is still Fickian.

C. Biased diffusion

We now consider the effect of an external field on the diffusion process. The external field can be an electric field, as in composite materials, or a pressure gradient, as in flow in a porous medium. The external field induces bias in the motion of the diffusants and generates a preferred direction for the transport process. In principle, the preferred direction changes locally (dynamically), but at the macroscopic scale, it is parallel to the direction of the external field. Thus, the simplest way of investigating the effect of the external field is to bias the motion of the particles in a particular direction [15], say the x direction, so that the probability that the particles move in this direction is higher than the other directions. Hence, we introduce a bias B so that in the square lattice the probability of moving in the direction parallel to the external field (i.e., the positive x direction) is $B + \frac{1}{4}(1 - B)$, while in the direction opposite to the external field and also in the transverse direction (i.e., perpendicular to the external field) the probability of jump from one node to another is $\frac{1}{4}(1 - B)$.

We carried out extensive simulations using $0.1 \leq B \leq 0.99$, and computed $\langle R_x^2(t) \rangle$ and $\langle R_y^2(t) \rangle$. We also calculated the overall MSD $\langle R^2(t) \rangle$. When the bias B is very small, we expect to recover the behavior for the unbiased case, and our simulations indicated that this is indeed the case. Moreover, if there were no correlations in the system, then one would expect [15] to have $\alpha = 2$, i.e., the motion of the particles is superdiffusive. Our simulations indicated, once again, that there is a qualitative difference between $H < 1/2$ and $H > 1/2$. In the latter case, the behavior of the system is similar to the case of no correlations, i.e., the transport process is superdiffusive with $\alpha = 2$. However, interesting results emerge for $H < 1/2$. For $0.1 \leq B \leq 0.99$ the MSD $\langle R_y^2(t) \rangle$ in the transverse direction (perpendicular to the direction of the bias) is asymptotically diffusive, i.e., $\alpha_y = 1$. Figure 9 presents typical results for $\langle R_y^2(t) \rangle$ obtained with various values of B and $H = 0.3$. On the other hand, the asymptotic behavior of $\langle R_x^2(t) \rangle$, the MSD in the longitudinal (bias) direction, is superdiffusive. For example, for $0.1 \leq B \leq 0.99$ and $H = 0.3$ we find $\alpha \approx 5/3$; see Fig. 10 for the typical results.

D. Log-periodic oscillations

Simulation of diffusion in some disordered media [16–18] have indicated that, under certain conditions, the effective value of the exponent α varies smoothly with time, so that no unique value of this exponent can be defined. More specifically, if one defines, through Eq. (4), an effective value α_e by

$$\alpha_e = \frac{d[\log \langle R^2(t) \rangle]}{d \log t}, \quad (9)$$

then α_e is found to be a function of the time, never reaching an asymptotic constant value. In particular, Bernasconi and

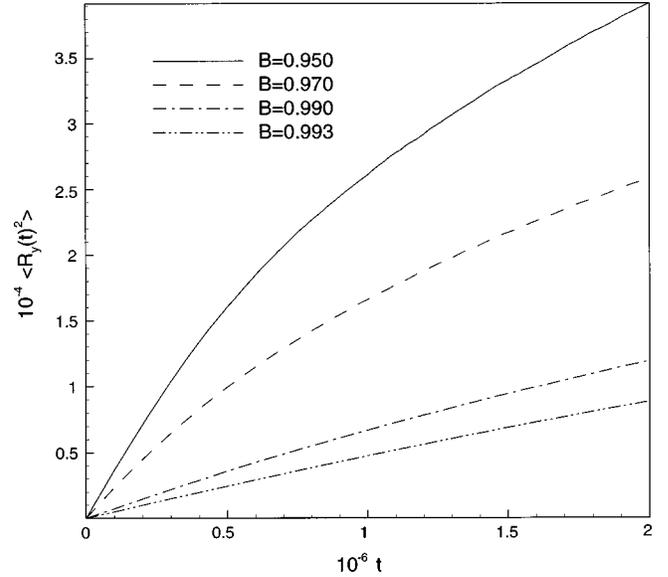


FIG. 9. Mean square displacements vs the time t in the transverse direction (perpendicular to the bias), as a function of the bias B , for $H = 0.3$.

Schneider [16] found that for a random walk on a one-dimensional lattice with a special type of the distribution of the transition rates (i.e., the probabilities of jumping from one node to another), the exponent α_e varies *log periodically* with the time t . Seifert and Suessenbach [17], who studied biased diffusion on percolation clusters, found that α_e varies smoothly with $\log t$. More recently, Stauffer and Sornette [18] found that biased diffusion on three-dimensional percolation lattices far from the percolation threshold and with $B \geq 0.95$ exhibits the same type of log-periodic variations of the effective exponent α_e with the time t . Such log-periodic oscillations have also been observed in many other phenom-

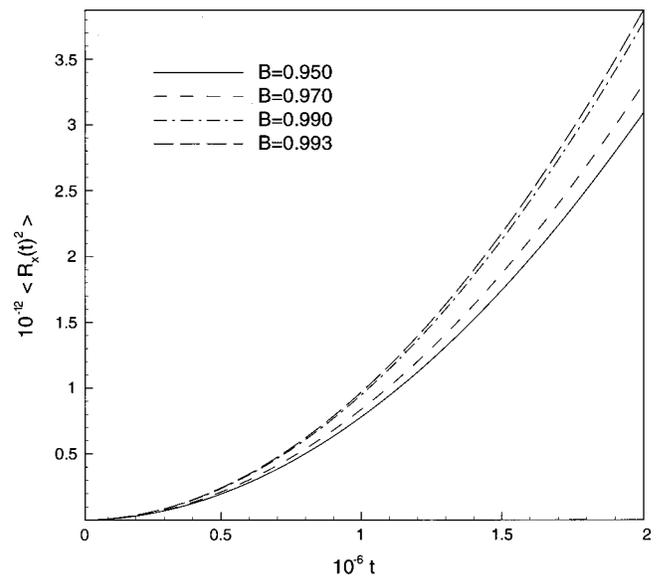


FIG. 10. Same as in FIG. 9, but for the longitudinal direction (parallel to the bias).

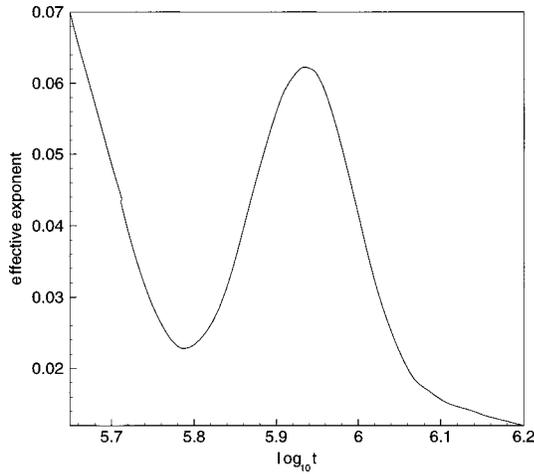


FIG. 11. The effective exponent α_e [see Eq. (9)] vs the logarithm of the time t in a medium in which the logarithm of the conductances follows a FBM with $H=0.3$, with 10% of the lowest conductances removed. The bias is $B=0.930$.

ena, ranging from fracture of rock [19] to stock market crashes; see Sornette [20] for a comprehensive review of the subject.

We find similar log-periodic oscillations of the exponent α_e when the diffusion process is simulated in two completely different media. In one case, we generated the disordered medium by assuming that the logarithms of the conductances follow the statistics of FBM. Then, in the spirit of the work of Stauffer and Sornette, we removed a small fraction (10%) of the bonds, except that, in order to preserve the correlations, we did not remove the bonds at random, rather we removed 10% of the bonds with the lowest conductances. We then simulated biased diffusion in this medium using various values of the bias B . We found that for $B \geq 0.93$ the effective value α_e varies log-periodically with the time. Figure 11 presents the results obtained with $H=0.3$. Similar results were obtained when we removed about 20% of the bonds with the lowest conductances; see Fig. 12.

A much more realistic model is one in which the system is

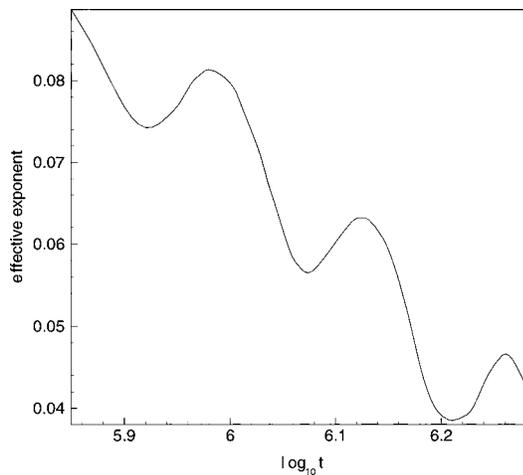


FIG. 12. Same as in FIG. 11, but with about 20% of the lowest conductances removed.

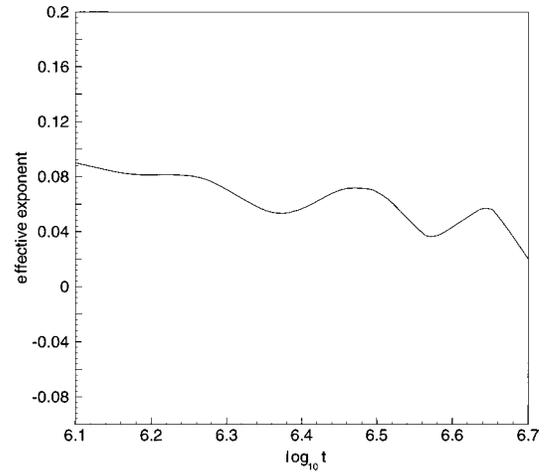


FIG. 13. Same as in FIG. 11, but in a stratified medium with an anisotropy factor $b_x=500$. The bias is $B=0.970$.

stratified. Biased diffusion in such a medium represents a realistic model of transport in many natural disordered media, such as soil and groundwater aquifers. We thus generated a stratified medium by the same method that was described above, and carried out simulation of biased diffusion in the medium. Figure 13 presents the results obtained with $B=0.97$ and $H=0.3$. Once again, the effective value of the exponent α exhibits log-periodic oscillations with the time. We believe that this is an important result in that, to the extent that this model simulates transport processes in disordered and stratified (anisotropic) media under the influence of an external field, *none* of the present continuum models of transport in such media can predict such a behavior, and therefore, one must develop new a theoretical framework for explaining and modeling these results.

V. SUMMARY

We have carried out extensive simulations of diffusion in disordered media with long-range correlations. The correlations are generated by a fractional Brownian motion and are characterized by a Hurst exponent H . Our two most important results are as follows.

(1) There is a qualitative difference between the behavior of the transport process with $H > 1/2$ and $H < 1/2$. While in the former case diffusion is Fickian, the later case represents a system that gives rise to non-Fickian diffusion. In a sense, this is consistent with the simulation of percolation in systems with the type of long-range correlations that is used here. Knackstedt, Sahimi, and Sheppard [21] showed that for $H > 1/2$ the percolation clusters are compact and nonfractal, whereas for $H < 1/2$ one obtains fractal structures with fractal dimensions that depend on the Hurst exponent H .

(2) Depending on the morphology of the medium, one may have anomalous or superdiffusive transport. Previous simulations of diffusion in fractal systems always yielded anomalous diffusion. In addition, biased diffusion in anisotropic media with long-range correlations is characterized by an effective value of the exponent α , that characterizes the power-law growth of the mean square displacements with the

time, which itself varies log periodically with the time. This type of transport process is important from a practical point of view, as it models diffusion of gases in soils and transport of tracers in flow through groundwater aquifers. However, log periodicity of the effective exponent α cannot be predicted by any of the currently available continuum models, and therefore, new theoretical frameworks must be developed in order to explain and predict their properties.

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