Anomalous diffusion and drift in a comb model of percolation clusters

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The example of a comb structure is used in a study of the influence of "dead ends" on the diffusion and drift of particles along percolation clusters. It is shown that the equation for the determination of the particle density on the axis of a structure is integrodifferential. When the number of particles is not conserved, the first derivative with respect to time in this equation is replaced with a fractional derivative of order 1/2. A correspondence is established with the problem of diffusion characterized by a continuous distribution of delay times. The results are given of a numerical modeling of the diffusion and drift in a two-dimensional percolation system. An analysis is made of the relationship between these results and the general problem of the drift and diffusion along fractals and in systems with traps.

1. INTRODUCTION

The anomalous diffusion in fractal structures is a subject of great topical interest. The anomaly consists in the usual time dependence of the mean-square displacement:

$$\langle x^2(t)\rangle \propto t^{2/(2+\theta)}. \tag{1}$$

Here, θ is the critical exponent of the anomalous diffusion process. The law (1) had been established by numerical modeling of percolation clusters (random fractals) and by the renormalization group method for regular fractals of the Sierpiński gasket type.[1] However, there is as yet no rigorous description of this effect: in fact, the anomalous diffusion equation has not yet been derived.

In our earlier paper[2] we suggested a phenomenological description of the anomalous diffusion in a homogeneous isotropic system. We demonstrated that in this case the generalized diffusion equation has fractional spatial derivatives. As a result, we established in Ref. 2 a nonlinear relationship between the current and the electric field due to anomalous diffusion:

$$\mathbf{j} \propto \mathbf{E}|\mathbf{E}|^{\theta}. \tag{2}$$

In other words, under the conventional diffusion conditions ($\theta = 0$) we can expect Ohm's law to be satisfied; in the anomalous diffusion case a more general expression (2) replaces Ohm's law (linear response). However, it is not clear to what extent the model of a homogeneous medium with anomalous diffusion can be used to describe real percolation systems.

A model allowing for the presence of "dead ends" in percolation systems (Fig. 1) was proposed in Ref. 3. The technique of generating functions was used in Ref. 3 to establish asymptotic behavior of the mean-square displacement along the axis of a structure and to show that it depends anomalously on time [Eq. (1)] with the exponent $\theta = 2$. However, the diffusion equation was not derived and incorrect extrapolation to the Gaussian form was proposed for the Green's function.

We shall derive below an equation describing the diffusion and drift along the axis of a structure. It can be represented as a continuity equation in which the first derivative with respect to time is replaced with a fractional derivative of order 1/2. An exact expression is also obtained for the Green's function used in the anomalous diffusion problem. It is shown that the mobility of this center of gravity in an external static electric field decreases with time in accordance with the following law:

$$\mu(t) \propto \mu_0/t^{1/2}. \tag{3}$$

It therefore follows that different types of behavior of the mobility in an external field are possible: we can either have the nonlinear dependence (2) in the case of constant mobility when $\mu \propto \mu_0|E|^{\theta}$ or the mobility may decrease with time in accordance with Eq. (3). We therefore carried out numerical modeling of the drift of a particle in a percolation cluster. We discovered power-law relaxation over short time intervals and a strongly nonlinear (exponential) behavior of the mobility for long times (10^5 steps) and in strong fields, when the capture by field-induced traps becomes important. The present paper deals with these topics.

2. MODEL OF A COMB STRUCTURE

2.1. Anomalous diffusion equation

A special feature of the diffusion in our model is that displacement in the x direction is possible only along the structure axis. In other words, the diffusion coefficient D_{xx} differs from zero only for $y = 0$: $D_{xx} = D_1\delta(y)$. The diffusion coefficient along the ribs is of the conventional type: $D_{yy} = D_2$. Therefore, a random walk on a comb structure is described by the diffusion tensor

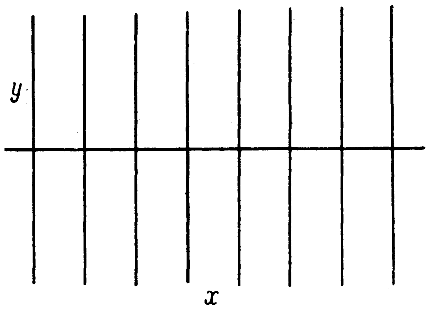

FIG. 1. Comb structure: the conducting axis ($y = 0$) has ribs going to infinity.

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$$\hat{D}=\begin{pmatrix} D_1\delta(y) & 0 \\ 0 & D_2 \end{pmatrix}. \qquad (4)$$

The diffusion equation is therefore

$$\frac{\partial G}{\partial t}-D_1\delta(y)\frac{\partial^2 G}{\partial x^2}-D_2\frac{\partial^2 G}{\partial y^2}=\delta(x)\delta(y)\delta(t). \qquad (5)$$

Here, $G(x,y,t)$ is the Green function for the diffusion problem. Applying the Laplace approximation with respect to time and the Fourier transformation along the coordinate x, we obtain a mixed (p,k,y) representation:

$$\left(p+D_1k^2\delta(y)-D_2\frac{\partial^2}{\partial y^2}\right)G(p,k,y)=\delta(y). \qquad (6)$$

The solution of Eq. (6) is

$$G(p,k,y)=\frac{\exp[-(p/D_2)^{1/2}|y|]}{2(D_2p)^{1/2}+D_1k^2}. \qquad (7)$$

As pointed out above, the diffusion along the structure axis, i.e., for $y=0$, is anomalous. The corresponding Green's function is

$$G(x,0,t)=\frac{D_2^{1/2}}{2\pi(D_1t^3)^{1/2}}\int_0^\infty \exp\left(-\frac{x^2}{4D_1\tau}-\frac{D_2\tau^2}{t}\right)\tau^{1/2}\,d\tau. \qquad (8)$$

Equation (8) is derived assuming the identity

$$\int_0^\infty \exp(-\alpha\tau)\,d\tau=\frac{1}{\alpha}.$$

The total number of particles on the structure axis decreases with time:

$$\langle G\rangle=\int_{-\infty}^{+\infty} G(x,0,t)\,dx=\frac{1}{2(\pi D_2t)^{1/2}}. \qquad (9)$$

Therefore, the Green's function $G(x,0,t)$ describes the diffusion when the number of particles is not conserved. Bearing this point in mind, we can calculate the displacement along the structure axis:

$$\langle x^2(t)\rangle=\frac{\langle x^2G\rangle}{\langle G\rangle}=D_1\left(\frac{\pi t}{D_2}\right)^{1/2}. \qquad (10)$$

It must be stressed that the results given by Eqs. (9) and (10) are exact and they follow from the integral representation of Eq. (8). The asymptotic form is found by the steepest-descent method:

$$G(x,0,t)\propto\frac{1}{(D_2t)^{1/4}}\left[\frac{x^2}{\langle x^2(t)\rangle}\right]^{1/4}\exp\left[-3\pi^{1/3}\left(\frac{x^2}{\langle x^2(t)\rangle}\right)^{2/3}\right]. \qquad (11)$$

One should also mention the unusual statistics of a random walk along the axis, the example, the probability of returning to the starting point of a random walk is

$$G(0,0,t)\propto(D_1^{1/2}D_2^{1/4}t^{3/4})^{-1}. \qquad (12)$$

We now consider the equation for $G(x,0,t)$. It follows from Eq. (7) that in the (p,k) representation, this equation is

$$[2(D_2p)^{1/2}+D_1k^2]G(p,k)=1. \qquad (13)$$

Using the definition of the fractional derivative with respect to time

$$\left(\frac{\partial^\alpha f(t)}{\partial t^\alpha}\right)_p=p^\alpha f(p)$$

(see Ref. 4), we obtain the following diffusion equation for the particle number on the structure axis:

$$\left(\frac{\partial^{1/2}}{\partial t^{1/2}}-\frac{D_1}{2D_2^{1/2}}\frac{\partial^2}{\partial x^2}\right)n(x,0,t)=0, \qquad (14)$$

where

$$\frac{\partial^{1/2}n}{\partial t^{1/2}}=\frac{1}{\Gamma(1/2)}\int_0^t\frac{\partial n(x,\tau)}{\partial\tau}\frac{d\tau}{|t-\tau|^{1/2}}.$$

The integrodifferential nature of the diffusion equation (14) is a consequence of the random disappearance and subsequent creation of a diffusing particle (disappearance from the axis and return to it) in the course of a random walk along the structure axis.

This formulation of the random-walk problem differs from the case of a continuous distribution of the delay times (continuous-time random walk—CTRW), first discussed in Refs. 5 and 6. The difference is this: in the latter problem the particle does not disappear but is delayed at each site with some probability. The total number of particles is then conserved. In the case of a comb structure the formulation with a continuous distribution of the delay times is equivalent to a study of

$$\tilde{G}(x,t)=\int_{-\infty}^{\infty}G(x,y,t)\,dy.$$

According to Eq. (7), the function $\tilde{G}$ is described by the equation

$$\left[p+\frac{D_1}{2}\left(\frac{p}{D_2}\right)^{1/2}k^2\right]\tilde{G}(p,k)=1. \qquad (15)$$

Consequently, in the CTRW case the diffusion equation represents the continuity equation for a medium with time dispersion:

$$\frac{\partial n}{\partial t}+\operatorname{div}\mathbf{j}=0, \qquad (16)$$

where

$$\mathbf{j}=-\frac{D_1}{2D_2^{1/2}\Gamma(1/2)}\frac{\partial}{\partial x}\int_0^t\frac{\partial n(x,\tau)}{\partial\tau}\frac{d\tau}{|t-\tau|^{1/2}}.$$

The diffusion is still anomalous to the same exponent $\theta=2$. The Green's function for this problem is different:

$$\tilde{G}(x,t)=\frac{D_2^{1/2}}{\pi(D_1t)^{1/2}}\int_0^\infty\exp\left(-\frac{x^2}{4D_1\tau}-\frac{D_2\tau^2}{t}\right)\frac{d\tau}{\tau^{1/2}}. \qquad (17)$$

The results obtained are readily generalized. For example, it is interesting to consider the transition from the anomalous to the conventional diffusion. For this transition to occur it is sufficient to immerse a comb structure in a poorly conducting medium with the diffusion coefficient D_0 $(D_0\ll D_2)$. In this case the diffusion coefficient is described by

$$\hat{D}=\begin{pmatrix} D_1\delta(y)+D_0 & 0 \\ 0 & D_2 \end{pmatrix}. \qquad (18)$$

For short time intervals, namely when $t \ll D_1^2/D_2 D_0^2$, the anomalous diffusion of Eq. (10) applies, whereas for long times we have the conventional diffusion obeying the $D_0 t$ law.

The transition to the conventional diffusion also occurs when the average length of the ribs L is finite. Then, for long times $t \gg L^2/D_2$, we obtain $\langle x^2(t) \rangle \propto D_1 t/L$.

2.2. Diffusion in an electric field

The application of an electric field E gives rise to anisotropy of the random walks. In weak fields the anisotropy parameter is small $[\alpha(E) \ll 1]$ and is proportional to the field. Consequently. in the case of the field current we have $j = n\mu E$. The mobility tensor for a comb structure is

$$\hat{\mu} = \begin{pmatrix} \mu_1 \delta(y) & 0 \\ 0 & \mu_2 \end{pmatrix}. \quad (19)$$

The Green's function for the problem of diffusion in an electric field directed along the structure axis is

$$G(p, k; \mathbf{E}) = [2(D_2 p)^{1/2} + D_1 k^2 + ik\mu_1 \mathbf{E}]^{-1}. \quad (20)$$

When the method described above is applied to Eq. (20), we obtain the following expression for the Green's function describing the diffusion and drift along the structure axis:

$$G(x, t; \mathbf{E}) = \frac{D_2^{1/2}}{2\pi D_1^{1/2} t^{3/2}} \int_0^\infty \exp\left[-\frac{(x-\mu_1 \mathbf{E}\tau)^2}{4D_1 \tau} - \frac{D_2 \tau^2}{t}\right] \tau^{1/2}\, d\tau. \quad (21)$$

We now find the first moment of the Green's function in the field

$$\langle \mathbf{x}(t) \rangle = \frac{\mu_1 \mathbf{E}}{2} \left(\frac{\pi t}{D_2}\right)^{1/2}. \quad (22)$$

It should be stressed that the response to a static electric field is time-dependent, i.e., the velocity of the center of gravity decreases with time in accordance with the law

$$\mathbf{V} = \frac{\mu_1 \mathbf{E} \pi^{1/2}}{2(D_2 t)^{1/2}}. \quad (23)$$

It is clear from Eq. (23) that in the anomalous diffusion case we cannot select a suitable inertial reference system moving at a constant velocity in which only diffusion would take place.

The steepest-descent method yields the asymptotic form of the Green's function in an electric field:

$$G(x, t; \mathbf{E}) \propto \begin{cases} \exp\left\{-\frac{x}{2L_E} - \frac{3}{4}\left[\left(\frac{x}{L_E}\right)^4 \frac{t_E}{t}\right]^{1/3}\right\}, & \frac{t}{t_E} \ll \frac{x}{L_E} \\ \exp\left\{-\frac{x-|x|}{2L_E} - \left[\left(\frac{x}{L_E}\right)^2 \frac{t_E}{t}\right]\right\}, & \frac{t}{t_E} \gg \frac{x}{L_E} \end{cases} \quad (24)$$

The quantities introduced above have a clear physical meaning: $L_E = D_1/\mu_1$ is the length governed by the external electric field, while

$$t_E \sim \frac{D_1^2 D_2}{\mu_1^4 E^4} \sim L_E^{2+\theta}$$

is the diffusion time over a distance L_E.

According to Eq. (24), the probability distribution function for short distances is governed primarily by the drift; in the case of long distances it is governed by the diffusion. In the conventional diffusion case the motion of the center of gravity and of the maximum of a packet coincide, whereas in the anomalous diffusion case the spreading of a packet is more complex. For short times $t \ll t_E$, the center of gravity of a packet travels faster than the point corresponding to the maximum of the function $G(x, t; E)$. For $t \gg t_E$, a definite front is established and the velocity of the packet maximum is higher than the velocity of the center of gravity:

$$V_{max}(E) \propto \frac{L_E}{t_E} \propto \frac{\mu_1^3 E^3}{D_1 D_2}. \quad (25)$$

We also consider the influence of an electric field on the probability of returning to the starting point of a random walk. The sideways drift in the conventional diffusion case gives rise to an exponential reduction in the probability of return:

$$G(0, t; \mathbf{E}) = G(0, t; \mathbf{E}=0) \exp(-t/t_E). \quad (26)$$

Here, $t_E \sim L_E^2/D_2$ $(\theta = 0)$. In our case, it follows from Eqs. (12) and (21) that for long times we can expect a power-law reduction:

$$G(0, t; \mathbf{E}) \sim G(0, t; \mathbf{E}=0)\, (t_E/t)^{1/4}. \quad (27)$$

The result of Eq. (27) is readily understood. The electric field acts on the particles only when they reach the structure axis; most of the time they are at the ribs and, therefore, a weaker (power-law) dependence is obtained. This accounts for the lag of the center of gravity of a packet behind its maximum.

3. NUMERICAL MODELING OF DRIFT IN PERCOLATION CLUSTERS

The models discussed above do not allow fully for the complex geometry of the percolation clusters. It is not possible to say *a priori* which of the expressions given by Eqs. (2) and (3) satisfactorily describes the drift of a particle in a percolation cluster. Therefore, we carried out numerical modeling of the drift in percolation clusters.

We describe briefly the modeling procedure. A random-number generator produced a quadratic lattice of size $L \times L$ (L = 400 or 500) and with a given fraction of knocked-out sites near the percolation threshold. This is the familiar problem of sites in percolation theory.[7] We then modeled a random walk on this lattice with periodic boundary conditions. The selection of the starting point of the walk was arbitrary. The probability of a step along one of four directions was determined by selecting a random number within the interval [0, 1] split into four equal parts. When a site in the selected direction was knocked out, a random selection of the direction was made until a transition to the nearest site took place. The number of failed attempts was ignored in the total number of steps. Configurations in which the starting point of a random walk corresponded to an empty site or when there were no nearest intact sites were ignored in calculating the mean-square displacement.

Averaging was carried out over randomly selected starting points of random walks and over realizations of the random percolation lattice. The total number of averaging procedures was of the order of 250. The anisotropy of a jump (in an electric field) was modeled by a nonuniform splitting of the interval [0, 1]. The length representing the motion along the field (or opposite to the field) was increased (or reduced) by an amount equal to the anisotropy α.

The calculation program was checked by applying it to a homogeneous medium and a comb structure. The results of the modeling of the diffusion itself in clusters agreed with those given in Ref. 8.

We found that for short times the mobility of a particle decreased with time in accordance with a power law

$$\mu(t) \propto \mu_0/t^{\gamma}. \qquad (28)$$

The results are presented in Fig. 2. Relaxation of the mobility with time is a consequence of the loss of a particle from a current-carrying path to "dead ends" and of the twisted nature of the percolation paths. It follows from these numerical calculations that the value of the exponent γ differs from the 1/2 typical of a comb structure and is approximately equal to 1/3. Using a scale analysis we can relate the power exponent γ to the anomalous diffusion exponent θ:

$$\gamma=\theta/(2+\theta).$$

If we represent the mobility of a particle in a percolation cluster in the scaling form

$$\mu(t)=\mu_0 f(t/t_c)/t_c^{\theta/(2+\theta)}$$

and repeat the familiar procedure in which it is assumed that for $t_c \to \infty$ the mobility should be independent of the proximity to the percolation threshold, we can determine the asymptotic behavior of the function f and the relationship between the power exponents γ and θ given above.

We also found the steady-state mobility after a long time (10^5 steps) and in strong electric fields ($eEL_c/T \gg 1$, where L_c is the correlation radius of an infinite cluster). We found that the mobility is a nonlinear function of the electric field (Fig. 3). It can be extrapolated using the following exponential dependence:

$$\mu(E) \propto \mu_0 \exp(-eEL_c/T). \qquad (29)$$

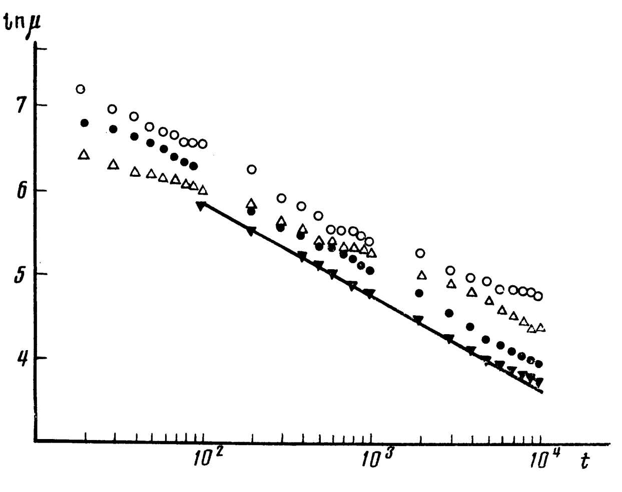

FIG. 2. Time dependence of the mobility μ of a particle in percolation clusters plotted for $\alpha = eEa/T = 0.05$ (○), $\alpha = 0.10$ (△), and $\alpha = 0.15$ (●). The excess above the percolation threshold is 0.03. The straight line and the points are obtained for a comb structure.

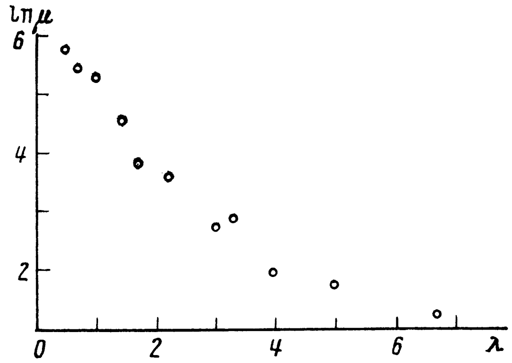

FIG. 3. Dependence of the particle mobility on the electric field $\lambda = eEL_c/T$ for long times ($t = 10^5$).

Therefore, in place of the possible law (2), we found a different nonlinear behavior of Eq. (29). This was because the electric field plays a dual role in inhomogeneous percolation systems. On the one hand, the field causes the drift of particles and, on the other, it creates particle traps. The electric-field-induced traps may be the "dead ends" and parts of the percolation paths directed against the field. Equation (2) was derived using a model description of the diffusion of the effective-medium type without any allowance for the capture by traps. Equation (29) can be explained as follows. The probability of crossing a trap of length L is exponentially small: it is proportional to $\exp(-eEL/T)$; the characteristic size of traps is of the order of the correlation radius L_c.

4. CONCLUSIONS

We have discussed the diffusion and drift in a comb structure and in percolation clusters. The main problem in describing the anomalous diffusion is that of the nature of the generalized diffusion equation. For a homogeneous isotropic model with anomalous diffusion it is an equation with fractional spatial derivatives.[3] In the present paper the example of a comb structure is used to show that the presence of "dead ends" leads to fractional time derivatives. If the anomalous nature of the diffusion in percolation clusters were entirely due to the "dead ends," then it would have sufficied to replace the derivative $\partial^{1/2}/\partial t^{1/2}$ in Eq. (14) with $\partial^{2/(2+\theta)}/\partial t^{2/(2+\theta)}$. In general, of course, we have to allow also for the spatial nonlocalization. Therefore, the generalized diffusion equation for percolation clusters can be represented in the form

$$(\partial^S/\partial t^S - \Delta^{\nu})n(x, t)=0. \qquad (30)$$

The power exponents S and ν are related by

$$S(2+\theta)=\nu. \qquad (31)$$

When Eq. (31) is obeyed, the diffusion problems are characterized by a specific type of self-similar behavior in which the anomalous dependence of the mean-square displacement on time given by Eq. (1) is always reproduced. The explicit form of the self-similar variable is selected:

$$z \propto x^{(2+\theta)}/t.$$

Another important topic is the explicit form of the Green's function of the problem with anomalous diffusion.

It follows from Eq. (8) that the Green's function can be described asymptotically by

$$G(x,t) \propto \exp\left(-\text{const}\frac{x^{4/3}}{t^{1/3}}\right). \qquad (32)$$

A similar expression is obtained also in the model of a homogeneous isotropic medium. It should be stressed that Eq. (32) is rigorous, in contrast to the extrapolation expressions given in Ref. 9.

We studied the relationship between the diffusion and conductivity in the anomalous diffusion case. The problem is that the diffusion coefficient defined in the usual manner

$$D = \lim_{t\to\infty}\frac{\langle x^2(t)\rangle}{t}$$

vanishes. Consequently, in the linear approximation we can also use the Einstein relationship to obtain zero mobility. On the other hand, a classical particle does not become localized:

$$\langle x^2(t)\rangle_{t\to\infty} \to \infty.$$

This contradiction can be resolved by at least two methods, namely by showing that in the anomalous diffusion case 1) there is not linear response, i.e., the Einstein relationship is invalid [see Eq. (2)]; 2) there is no constant mobility in a static electric field, in other words, the average displacement increases with time more slowly than the first power of t.

In a comb structure we have both cases described by Eqs. (2) and (3). The constant and nonlinear mobility $\mu(E) \propto \mu_0 E^2$ characterizes the motion of the maximum of a packet, whereas the mobility of the center of gravity of the packet decreases with time: $\mu(t) \propto \mu_0/t^{1/2}$. This difference is easily understood. The contribution to the motion of the packet maximum is made only by the mobile particles, i.e., the particles on the structure axis, which migrate by the anomalous diffusion in a homogeneous one-dimensional medium. The anomalous nature of the diffusion is ensured by the presence of creation and annihilation centers at each point on the axis. It follows from earlier work[2] that the mobility in such media should be a nonlinear function of the electric field. The position of the center of gravity of a packet is determined by all the particles. In their case the anomalous diffusion is due to random delay at each site and the consequence of this is also a reduction in the mobility of the center of gravity with time.

The power-law relaxation of the mobility of a particle in percolation clusters discovered by the numerical modeling method suggests the need to use fractional time derivatives in the generalized diffusion equation of Eq. (30).

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[1]Y. Gefen, A. Aharony, B. B. Mandelbrot, and S. Kirkpatrick, Phys. Rev. Lett. **47**, 1771 (1981); Ya. B. Zel'dovich, S. A. Molchanov, A. A. Ruzmaĭkin, and D. D. Sokolov, Usp. Fiz. Nauk **152**, 3 (1987) [Sov. Phys. Usp. **30**, 353 (1987)].
[2]V. E. Arkhincheev, É. M. Baskin, and É. G. Batyev, J. Non-Cryst. Solids **90**, 21 (1987).
[3]G. H. Weiss and S. Havlin, Physica A (Utrecht) **134**, 474 (1986).
[4]M. M. Dzhrbashyan, *Integral Transformations and Representation of Functions in the Plane of the Complex Variable* [in Russia], Nauka, Moscow (1966).
[5]E. W. Montroll and G. H. Weiss, J. Math. Phys. (N. Y.) **6**, 167 (1965).
[6]M. F. Shlesinger, J. Stat. Phys. **10**, 421 (1974).
[7]B. I. Shklovskii and A. L. Efros, *Electronic Theory of Doped Semiconductors*, Springer Verlag, Berlin (1984).
[8]R. B. Pandey, D. Stauffer, A. Margolina, and J. Zabolitzky, J. Stat. Phys. **34**, 427 (1984).
[9]A. B. Harris and A. Aharony, Europhys. Lett. **4**, 1335 (1987).

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