Three-dimensional diffusion in a two-component disordered medium

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A scaling approach developed previously[14] to describe the diffusion of particles in a two-dimensional disordered medium is generalized to the three-dimensional case. Three-dimensional lattice models are investigated for hopping diffusion on a regular cubic lattice consisting of two types of edges, which differ in their characteristic hopping times. A scaling transformation procedure is developed that allows us to derive renormalization group equations for the resulting spatial and temporal scaling transformations. We obtain analytic forms for the values of the diffusion coefficient in a three-dimensional heterogeneous medium and the time dependence of the mean square displacement of migrating particles in the limiting cases when the fraction of one component of the medium is small or the hopping times along edges of different types are very different. In the remaining cases the corresponding quantities are found numerically.

INTRODUCTION

Interest in the study of diffusion processes in disordered media has been unflagging because many physical phenomena in real heterogeneous condensed media are diffusive in nature or are associated with diffusion processes, and in a number of these cases the diffusion stage is the limiting factor. As examples we could cite descriptions of the properties of polymer macromolecules,[1] diffusion in amorphous systems and polymer solutions and in polymer gels,[2] the kinetics of diffusion-controlled chemical reactions,[3] dispersive transport,[4] etc.

Despite their relatively simple formulation, diffusion problems in disordered media can be difficult to solve mathematically due to the non-Markovian nature of random walk processes, which in many cases prevents us from finding exact solutions. Random migration processes in disordered systems are usually investigated using both analytic methods of various kinds[5–10] and mathematical simulations[11] for simple lattice models. The most widespread method is expansion in the impurity concentration[6–8] using an averaged generating functional.[9] The scaling approach[10] has been used successfully to describe the random walk process in a special case, random walks on fractals; this approach appears to us to be very promising. We are led to use the concept of scaling, which has been found to be an extraordinarily fruitful way to describe phase transitions and properties of polymer molecules, by the analogy with the block-spin (coarsening) lattice procedure introduced by Kadanoff in Ref. 12 and the renormalization group method for calculating thresholds in the percolation theory.[13]

In a previous paper[14] we proposed a rather simple, transparent, and approximately analytical method for solving the problem of diffusion of particles in a two-component two-dimensional disordered medium. In this paper, which is the direct continuation of our previous one, we have succeeded in generalizing the approach proposed in Ref. 14 to the case of a three-dimensional medium.

STATEMENT OF THE PROBLEM AND MODEL DESCRIPTION

The medium in which diffusive migration of particles occurs consists of regions of two kinds (I and II), which differ in the values of their diffusion coefficients (DC); in what follows we will refer to these as the "intrinsic" DC's $D_c^{\mathrm{I}} \geqslant D_c^{\mathrm{II}}$. For simplicity we will assume that these regions have the same characteristic linear size L_0, and that their random locations in space are distributed uniformly on the average and independently of one another. Let the volume fractions of regions with rapid (I) and slow (II) diffusion be p and $1-p$, respectively. According to the approach developed in Ref. 14, our problem consists of finding the average DC's D_n of media with corresponding scales L_n which in the limit allow us to calculate the average macroscopic DC of the medium $D_{\mathrm{av}} \equiv D_\infty$. (The definitions that we will use for these quantities will be made more precise below.)

For convenience in presenting the material that follows, let us briefly formulate the basic idea and conclusions of our previous paper,[14] generalizing it where necessary to the case of a three-dimensional medium. Our approach is based on the use of the scaling hypothesis (scale invariance), which when applied to the medium under discussion here has the following meaning: we propose that at any coarsened scale $L_n > L_0$ the original two-component medium can be treated once more if it were composed of random scaled (size L_n) regions of two types with new values of the intrinsic DC's $D_{\mathrm{cn}}^{\mathrm{I}}$ and $D_{\mathrm{cn}}^{\mathrm{I}}$ and volume fractions p_n and $1-p_n$, i.e., like the original medium with scale L_0. In other words, when we replace the original medium by a new medium consisting of coarsened regions, we should preserve (even if approximately) the macroscopic DC of the medium (D_{ac}) and also the average DC (D_m) for scales $L_m \geqslant L_n$. For this case the laws of transformation of the intrinsic DC of regions and their volume fractions should depend only on the ratio of the initial and final scales L_0/L_n.

The specific form of these scaling transformation laws in the case of a two-dimensional medium were found in Ref. 14 for a model of hopping diffusion on a square lattice. In that paper we found that it is possible to use the renormalization procedure, i.e., to apply the scaling transformations many times with renormalized quantities p_n, D_{cn}^{I}, and D_{cn}^{II} at each step, to find the values of the average macroscopic DC of the medium D_{av} as a limit of a sequence D_n as $L_n \to \infty$. The existence of such a limit is ensured by the fact that for infinitely large scales the medium is averaged and becomes macroscopically homogeneous.

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SCALING TRANSFORMATION ALGORITHM

In order to find the required scaling transformation laws in the three dimensional case we introduce the initial scale L_0 and approximate the two-component medium by a cubic lattice S_0 consisting of randomly located (uniformly and independently) edges of two kinds (I and II) corresponding to the "fast" and "slow" regions (the length of a region is L_0). For random migration of particles along such a lattice the jump time is $\tau_0^{\rm I}$ along a type I edge and $\tau_0^{\rm II}$ along a type II edge ($\tau_0^{\rm I} \leqslant \tau_0^{\rm II}$); the direction of a jump is equally probable in all directions, and does not depend on the properties of the neighboring edges. The intrinsic DC's of the edges are defined in the following way:

$$D_{c0}{}^{\rm I}=L_0{}^2/\tau_0{}^{\rm I},\ D_{c0}{}^{\rm II}=L_0{}^2/\tau_0{}^{\rm II}=h_0 D_{c0}{}^{\rm I}, \tag{1}$$

where $h_0 = \tau_0^{\rm I}/\tau_0^{\rm II}$, $0\leqslant h_0 \leqslant 1$.

The fractions of type I and type II edges equal the volume fractions of the corresponding regions, i.e., p_0 and $1-p_0$.

In order to introduce a new coarsened scale L_n, we consider the family $\{\Omega_n\}$ of all possible 2^n-link paths (including self-intersecting paths) ω_n for particle migration along the initial lattice S_0. Each path from this family has length $2^n L_0$ (where $n = 0,1,2,..$), while the squared displacement of a migrating particle averaged over the family $\{\Omega_n\}$, $\{\Omega_n\}$ equals

$$\langle R^2_{n,\omega}\rangle_{\Omega_n}=2^n L_0{}^2=L_n{}^2, \tag{2}$$

here $R_{n,\omega}$ is the distance between the beginning and end of a given path ω_n from the family $\{\Omega_n\}$ while the angle brackets denote averaging over all the paths from the family, $\{\Omega_n\}$.

The mean DC D_n for a scale L_n is defined in the following way:

$$D_n=\langle R^2_{n,\omega}/t_{n,\omega}\rangle_{\Omega_n}, \tag{3}$$

where $t_{n,\omega}$ is the time spent by a particle in transversing the path $\omega_n \in \{\Omega_n\}$. Then the average macroscopic DC of the medium $D_{\rm av} \equiv D_\infty$ is

$$D_\infty=\lim_{n\to\infty} D_n. \tag{4}$$

Introducing a new consolidated scale allows to introduce a new cubic lattice as well, with edges equal to the new scale L_n. In order to define the required scaling law, it is necessary to average over all possible paths with 2^n steps in keeping with (3) and (4), i.e., to consider all possible paths; this is realistic only for $n = 1$. Thus, the scale transformation $L_0 \to L_1 = 2^{1/2} L_0$ is the one of interest to us. Whereas in the two-dimensional case the sites of lattice S_1 with edges L_1 coincided with the corresponding sites of the initial lattice S_0, and the edges of the lattice S_1 were diagonals of the unit cells of the initial lattice S_0 so that a coarsened square lattice arose in a natural fashion, in the three-dimensional case this simple geometric connection fails. However, by virtue of the properties of the random migration $\langle \mathbf{R}_n \mathbf{R}_n' \rangle = 0$ we are permitted to introduce a lattice with mutually orthogonal edges [Eq. (2) is also based on this property].

DISCRETE SCALING TRANSFORMATION

For single-link paths the average DC equals

$$D_0=p_0 D_{c0}{}^{\rm I}+(1-p_0)D_{c0}{}^{\rm II}=D_0{}^{\rm I}+D_0{}^{\rm II}=[p_0+(1-p_0)h_0]D_{c0}{}^{\rm I}. \tag{5}$$

Consideration of all possible two-link paths on a cubic lattice leads to the same expression as for the square lattice:

$$D_1=[p_0{}^2+4p_0(1-p_0)h_0/(1+h_0)+(1-p_0)^2 h_0]D_{c0}{}^{\rm I}, \tag{6}$$

where $D_0^{\rm I} = p_0 D_{c0}^{\rm I}$ and $D_0^{\rm II} = (1-p_0) D_{c0}^{\rm II}$ are the "effective" DC's along the type I and type II edges, respectively, and $h_0 = \tau_0^{\rm I}/\tau_0^{\rm II} = D_{c0}^{\rm II}/D_{c0}^{\rm I}$.

In Eq. (6) the first and third terms correspond to paths consisting of two edges of the same kind (types I or II, respectively). The second term corresponds to a "mixed-type" path, i.e., consisting of two edges of different types. The fractions of "fast," "slow," and "mixed" edges are, respectively

$$p_1{}^{(1)}=p_0{}^2,\ p_1{}^{(2)}=(1-p_0)^2,\ p_1{}^{(3)}=2p_0(1-p_0). \tag{7}$$

Let us specify the jump time along each of these three types of edges in such a way that it coincides with the time for traversing a two-link path of the corresponding type on the lattice S_0. Then for the fast, slow, and mixed edges of the lattice S_1 we have

$$\tau_1{}^{(1)}=2\tau_0{}^{\rm I},\ \tau_1{}^{(2)}=2\tau_0{}^{\rm II},\ \tau_1{}^{(3)}=\tau_0{}^{\rm I}+\tau_0{}^{\rm II}=[(1+h_0)/h_0]\tau_0{}^{\rm I}.$$

In order to construct a consolidated lattice S_1 with the same average DC D_1 (6) but, like the initial lattice S_0, consisting only of fast and slow edges (i.e., only of edges of two types), we break up the overall number of mixed edges for the lattice S_1, whose fraction is $p_1^{(3)}$, into two parts:

$$p_1{}^{(3)}=\alpha_0 p_1{}^{(3)}+(1-\alpha_0)p_1{}^{(3)},\ 0\leqslant\alpha_0\leqslant 1. \tag{8}$$

Let us combine (only formally for the moment) the first part with the fast edges to form a single group, and do the same with the second part and the slow edges. Then in place of the three types of edges we obtain only two types, with fractions that are, respectively [see (7)]

$$\begin{aligned} p_1{}^{\rm I}&\equiv p_1=p_1{}^{(1)}+\alpha_0 p_1{}^{(3)},\\ p_1{}^{\rm II}&\equiv 1-p_1=p_1{}^{(2)}+(1-\alpha_0)p_1{}^{(3)}. \end{aligned} \tag{9}$$

In accordance with this decomposition, the contribution D_1 to the DC corresponding to mixed edges [i.e., the second term in Eq. (6)] is also divided into two parts. The first of these, which is proportional to α_0, is combined with the first term, and the second term, which is proportional to $(1-\alpha_0)$, is combined with the third. As a result, we obtain from (6) an expression that is structurally analogous to (5):

$$\begin{aligned} D_1{}^{\rm I}&=p_0[p_0+4\alpha_0(1-p_0)h_0/(1+h_0)]D_{c0}{}^{\rm I}\\ &=[p_0+4\alpha_0(1-p_0)h_0/(1+h_0)]D_0{}^{\rm I}, \end{aligned} \tag{10a}$$

$$\begin{aligned} D_1{}^{\rm II}&=(1-p_0)[(1-p_0)+4(1-\alpha_0)p_0/(1+h_0)]h_0 D_{c0}{}^{\rm I}\\ &=[(1-p_0)+4(1-\alpha_0)p_0/(1+h_0)]D_0{}^{\rm II}, \end{aligned} \tag{10b}$$

$$D_1=D_1{}^{\rm I}+D_1{}^{\rm II}. \tag{10c}$$

The average intrinsic DC's of the new edges in the first and second groups equal

$$D_{c1}{}^{\rm I}=D_1{}^{\rm I}/p_1,\ D_{c1}{}^{\rm II}=D_1{}^{\rm II}/(1-p_1). \tag{11}$$

Let us also note that the following recursion relation is implied by (7) and (9):

$$p_1-p_0=p_0(1-p_0)(2\alpha_0-1). \tag{12}$$

Note that up to this point in our discussion it has nowhere been necessary to take into account the specific structure of three-dimensional space, and that all the expressions (5) to (12) coincide formally with those obtained for the two-dimensional case. However, in order to find the form of the parameter $\alpha_0 \equiv \alpha_0(h_0, p_0)$ used to split the mixed paths into two groups, it is necessary to include the specific features of the three-dimensional situation.

As in the two-dimensional case, the parameter α_0 is defined by comparison with the percolation problem. The procedure for consolidating the lattices then reduces to investigating certain effective paths that join sites of a new "consolidated" lattice. Since the percolation problem (i.e., the problem of connectivity) corresponds to successively traversing paths in our algorithm (in contrast to diffusion processes), it is the time a particle takes to travel along a given path that is averaged. Therefore, if we initially replace the mixed edges by edges of two types in a proportion corresponding to the percolation problem (a fraction μ_0 for the fast paths, and a fraction $1-\mu_0$ for the slow paths) and compare it to the diffusion-equivalent partition (fractions α_0 and $1-\alpha_0$, respectively), then the following relation should hold:

$$\frac{\Delta^2}{\mu_0\tau_1+(1-\mu_0)\tau_2} = \alpha_0\frac{\Delta^2}{\tau_1} + (1-\alpha_0)\frac{\Delta^2}{\tau_2} \qquad (13)$$

(where Δ is the hopping scale), from which we immediately find the required form of α_0:

$$\alpha_0 = \frac{\mu_0 h_0}{(1-\mu_0+\mu_0 h_0)}. \qquad (14)$$

The form of the function $\mu_0(p_0)$ is determined by the following features of the percolation problem in the three-dimensional case. First of all, there exist two percolation thresholds in the problem of bonds on a cubic lattice: $p_{c1} = 1/4$, and $p_{c2} = 3/4$, corresponding to the appearance of connectivity along type I edges and loss of connectivity along type II edges. In the concentration interval $1/4 < p_0 < 3/4$, connectivity exists for edges of both types (the region of combined connectivity). Secondly, in the scale transformation equations for the probability of connectivity along edges of type I in analogy with (12):

$$p_1 - p_0 = p_0(1-p_0)(2\mu_0 - 1). \qquad (15)$$

we find that, in addition to the unstable stationary points at $p_0 = 1/4$ and $p_0 = 3/4$, corresponding to the percolation thresholds, and the stable points at $p_0 = 0$ and $p_0 = 1$, the stationary point at $p_0 = 1/2$ should also be stable, corresponding to a macroscopic state of the system with combined connectivity along edges of both types. In order to satisfy these conditions, the expression for $\mu_0(p)$ $\equiv \alpha_0(p, h_0 = 1)$ should be a polynomial of degree no more than three and have zeroes at the points 1/4, 1/2, and 3/4. Consequently, the simplest possible form of the function $\mu_0(p)$ is given by the relation

$$\mu_0(p) = k(p-1/4)(p-1/2)(p-3/4) + 1/2. \qquad (16)$$

This expression has the required symmetry with respect to the replacement $p \to 1-p$: $\mu_0(1-p) = 1-\mu_0(p)$, i.e., $\mu_0(1/2) = 1/2$. The coefficient of proportionality k in (16) is determined from the condition $\mu_0(1) = 1$, which by virtue of symmetry is equivalent to $\mu_0(0) = 0$. As a result, we find that $k = 16/3$ and

$$\mu_0(p_0) = 1/3\, p_0(16p_0^2 - 24p_0 + 11). \qquad (17)$$

Thus, taking into account (14), we obtain the required expression for α_0:

$$\alpha_0(p_0, h_0) = \frac{h_0 p_0 \beta(p_0)}{h_0 p_0 \beta(p_0) + (1-p_0)\gamma(p_0)}, \qquad (18)$$

where $\beta(p_0) = 1/3(16p_0^2 - 24p_0 + 11)$ and $\gamma(p_0)$ $\equiv \beta(1-p_0) = 1/3(16p_0^2 - 8p_0 + 3)$.

Since we have limited ourselves to two-link paths in order to derive the scaling transformation law, we must include (if only in an average way) those correlations in the mutual positions of same-type edges that effectively arise from returning portions of the migration paths of particles for long migration path lengths. As in the two-dimensional case, we include these correlations by altering Eq. (7) in the following way:

$$p_1^{(1)} = p_0 Q(p_0), \quad p_1^{(2)} = (1-p_0)Q(1-p_0), \qquad (19)$$

$$p_1^{(3)} = 1 - [p_0 Q(p_0) + (1-p_0)Q(1-p_0)].$$

In the case where correlation is absent, $Q(p_0) \equiv p_0$, and (19) coincides with (7).

Let us pick the correlation function $Q(p_0)$ in the simplest form

$$Q(p_0) = \frac{p_0 + a}{1+a}. \qquad (20)$$

For the function $Q(p_0)$ defined in this way, at $a = 0$ there is no correlation at all, while for $a = 1$ the correlation corresponds to the two-dimensional case. It is clear that in the three-dimensional case the effect of correlation will be smaller than in the two-dimensional case (this is due to a well-known property of random walks the probability of return to an initial point in the two-dimensional case is finite, while in the three-dimensional case it equals zero[1]). Therefore, in the three-dimensional problem the parameter a should take on a value between zero and one. It is found that by choosing $a = 2/d$ (where d is the dimension of the space) we obtain the correct time dependence in the asymptotic expression for calculated quantities in those cases where the answer is known[6-8] (in the corresponding expressions t is replaced by $t^{d/(d+2)}$), both for the two-dimensional ($d = 2$) and three-dimensional ($d = 3$) cases.

As a result, we have completely specified the procedure for scale transformations in these systems from the initial scale L_0 to the coarsened scale L_1. Repeating this procedure many times, we can obtain a scale transformation law from an arbitrary scale L_n to a scale L_{n+1}:

$$p_{n+1} = p_n\left[1 + \frac{d}{d+2}(1-p_n)(2\alpha_n - 1)\right], \qquad (21)$$

$$D_{n+1}^{\mathrm{I}} = \left\{1 - \frac{d}{d+2}(1-p_n)\left(1 - \frac{4\alpha_n h_n}{1+h_n}\right)\right\} D_n^{\mathrm{I}}, \qquad (22)$$

$$D_{n+1}^{\mathrm{II}} = \left\{1 - \frac{d}{d+2} p_n\left[1 - 4\frac{(1-\alpha_n)}{1+h_n}\right]\right\} D_n^{\mathrm{II}}. \qquad (23)$$

In the three-dimensional case, $d = 3$, and the factor $d/(d+2)$ is 3/5.

INFINITESIMAL SCALE TRANSFORMATIONS AND THE RENORMALIZATION GROUP EQUATIONS

The right-hand portions of the recursion relations (21)–(23) are in a form that allows us to pass from a discrete scale transformation $L_n \to L_{n+1} = 2L_n$ to an infinitesimal one $L_n \to L_{n+\delta n} = 2^{\delta n} L_n$ $(\delta n \to 0)$. In fact, Eqs. (21)–(23) can be written in the form of difference equations; from there, we can pass to the differential equations:

$$\frac{dp(n)}{dn} = \frac{3}{5} p(n)[1-p(n)][2\alpha(p,h)-1], \tag{24}$$

$$\frac{dD^{\mathrm{I}}(n)}{dn} = -\frac{3}{5}[1-p(n)]\left[1-\frac{4\alpha(p,h)h(n)}{1+h(n)}\right]D^{\mathrm{I}}(n), \tag{25}$$

$$\frac{dD^{\mathrm{II}}(n)}{dn} = -\frac{3}{5} p(n)\left[1-\frac{4[1-\alpha(p,h)]}{1+h(n)}\right]D^{\mathrm{II}}(n). \tag{26}$$

Integrating (25) and (26) we obtain

$$D^{\mathrm{I}}(n) = p_0 D_{c0}{}^{\mathrm{I}} \exp\left\{-\frac{3}{5}\int_0^n [1-p(n)] \times \left[1-\frac{4\alpha(p,h)h(n)}{1+h(n)}\right]dn\right\}, \tag{27}$$

$$D^{\mathrm{II}}(n) = (1-p_0) D_{c0}{}^{\mathrm{II}} \exp\left\{-\frac{3}{5}\int_0^n p(n) \times \left[1-\frac{4[1-\alpha(p,h)]}{1+h(n)}\right]dn\right\}. \tag{28}$$

After differentiating the function $h(n) = D_c^{\mathrm{II}}(n)/D_c^{\mathrm{I}}(n)$, taking into account relations (25)–(28) we obtain a differential equation which, along with Eq. (24), determines the functions $p(n)$ and $h(n)$:

$$\frac{dh(n)}{dn} = \frac{6}{5} p(n)[1-p(n)] \times \frac{h(n)[1-h(n)][h(n)\beta(p)+\gamma(p)]}{[1+h(n)][h(n)p(n)\beta(p)+(1-p)\gamma(p)]}. \tag{29}$$

A very simple analysis of the system of kinetic equations (24) and (29) shows that the point $(p=1, h=0)$ is an unstable node, the point $(p=1/2, h=1)$ is a stable node, and the points $(p=1/4, h=1)$ and $(p=3/4, h=1)$ are saddle points. Furthermore, the lines $p=0$ and $p=1$ are lines of stationary equilibrium. The phase portrait of the system of kinetic equations (24) and (29) in the quadrant $(0 \leqslant p, h \leqslant 1)$ is shown in Fig. 1. The unstable node $(p=1, h=0)$ is connected with the saddle points $(p=1/4, h=1)$ and $(p=3/4, h=1)$ by two separatrices, which divide the physical region of the phase plane (the square $0 \leqslant p, h \leqslant 1$) into three parts. If an initial point (p_0, h_0) lies below the lowest separatrix (region I, then $p_\infty = 0$, which corresponds to complete disappearance at large scales of regions with rapid diffusion, while all the space will consist only of regions of small diffusion, whose rate is increased due to absorption of regions with rapid diffusion. In the opposite case, if the initial point (p_0, h_0) lies above the higher separatrix (region III), then $p_\infty = 1$, which conversely corresponds to complete disappearance at large scales of regions with slow diffusion, and the entire space will consist only of regions of rapid diffusion whose rate is decreased due to absorption of regions with slow diffusion. If the initial point (p_0, h_0) lies between the separatrices (region II), then $p_\infty = 1/2$ and

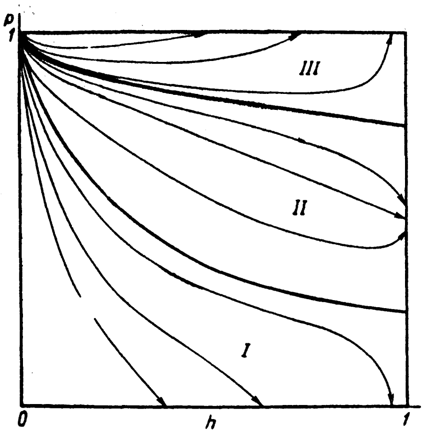

FIG. 1. Phase portrait of the system of equations (36) and (37): the separatrices are pointed out by boldface curves.

$h_\infty = 1$, i.e., at large scales the fractions of regions with rapid and slow diffusion become equal, while the properties of these regions due to the mutual mixing become equal. If, however, the initial point (p_0, h_0) lies on one of the separatrices, then $p_\infty = 1/4$ or $p_\infty = 3/4$; however, in both cases $h_\infty = 1$, i.e., the intrinsic DC of regions of both types are equal. We note that in region I the connectivity takes place along the slow edges, in region III along the fast edges, while in region II it is sometimes along one, sometimes along the other. The presence of region II is a distinctive feature of three-dimensional space.

Our analysis shows that (as we assumed) at large scales the medium becomes macroscopically homogeneous. Thus, in order to solve the problem as posed—i.e., to determine the effective DC of a disordered medium—it is necessary for us to solve the system of equations (24) and (29), substitute the resulting solutions into (27) and (28), and determine the required quantities using the expression

$$D_{\mathrm{av}} = D^{\mathrm{I}}(\infty) + D^{\mathrm{II}}(\infty). \tag{30}$$

After some uncomplicated mathematical transformations taking into account (27)–(29), Eq. (30) reduces to a rather simple form

$$D_{\mathrm{av}} = [p_\infty + (1-p_\infty)h_\infty]\exp\left\{-\int_{h_0}^{h_\infty} \frac{dh}{h+W(h)}\right\} D_{c0}{}^{\mathrm{I}}, \tag{31}$$

where $h_\infty \equiv \lim_{n\to\infty} [h(n)]$ is the value of h when $p_\infty \equiv \lim_{n\to m} [p(n)]$, equaling 0, 1, or 1/2 depending on the initial condition,

$$W(h) \equiv W(p(h)) = \gamma(p(h))/\beta(p(h)),$$

and the function $p(h)$ is determined from an ordinary differential equation obtained by dividing Eq. (24) by Eq. (29):

$$\frac{dp(h)}{dh} = \frac{[1+h][p(h)h-(1-p)W(p)]}{2h[1-h][h+W(p)]}. \tag{32}$$

COMPUTATION OF THE AVERAGE DIFFUSION COEFFICIENT

Unfortunately, Eq. (32) cannot be integrated in general. Analysis of the phase portrait (see Fig. 1) shows that the asymptotic solutions to Eq. (32) can be obtained for the following limiting types of initial conditions:

1) $h_0 \to 0$, p_0 such that $h_0/(1-p_0)^2 \ll 1$;

2) $p_0 \to$ for any h_0;

3) $p_0 \to 1$, h_0 such that $h_0/(1-p_0)^2 \gg 1$.

In case 1) Eq. (32) simplifies greatly

$$dp/dh=(p-1)/2h, \tag{32a}$$

which allows us to immediately write the functional relation

$$h/h_0=[(1-p)/(1-p_0)]^2, \tag{33}$$

while the inequality $h/(1-p)^2 \ll 1$ holds on all the phase trajectories.

Since $p_\infty = 0$, from (33) we immediately find that $h_\infty = h_0/(1-p_0)^2$. Substituting the expression so obtained into (31) gives [to second order in the parameter $h_0/(1-p)^2$] the following value for the average DC:

$$D_{\text{av}} = \frac{D_{c0}{}^1 h_0}{(1-p_0)^2}\left\{1 - \frac{h_0}{(1-p_0)^2}\left[p_0\left(\frac{20}{3}-p_0\right)\right.\right.$$

$$\left.\left.+2^{-1/2}\operatorname{arctg}\left(\frac{2^{1/2}p_0}{3/4-p_0}\right)\right]\right\}. \tag{34}$$

In case 2), Eq. (32) takes the form

$$\frac{dp}{dh}=-\frac{3(1+h)}{2h(1-h)(11h+3)}, \tag{32b}$$

from which we have

$$p=p_0-\frac{1}{2}\ln\left(\frac{h}{h_0}\right)+\frac{3}{14}\ln\left(\frac{1-h}{1-h_0}\right)+\frac{2}{7}\ln\left(\frac{11h+3}{11h_0+3}\right), \tag{35}$$

while the value h_∞ is determined from (35) taking into account the condition $p_\infty = 0$:

$$\frac{1}{2}\ln\left(\frac{h_\infty}{h_0}\right)-\frac{3}{14}\ln\left(\frac{1-h_\infty}{1-h_0}\right)-\frac{2}{7}\ln\left(\frac{11h_\infty+3}{11h_0+3}\right)=p_0, \tag{36}$$

which to first order in p_0 gives

$$h_\infty=h_0\left[1+\frac{2p_0(1-h_0)(11h_0+3)}{3(1+h_0)}\right], \tag{37}$$

Substituting (37) into (31) and assuming that as $p \to 0$, $W(p) \approx 3/11$, we obtain to second order

$$D_{\text{av}}=D_{c0}{}^1 h_0\left\{1+\frac{2p_0(1-h_0)}{1+h_0}\right.$$

$$\left.-\frac{11}{9}\left[\frac{2p_0(1-h_0)}{1+h_0}\right]^2 h_0(11h_0+3)\right\}. \tag{38}$$

In case 3), Eq. (32) takes the form

$$\frac{dp}{dh}=\frac{3(1+h)}{2(1-h)(11+3h)}, \tag{32c}$$

from which

$$p=p_0-\frac{3}{14}\ln\left(\frac{1-h}{1-h_0}\right)-\frac{2}{7}\ln\left(\frac{11+3h}{11+3h_0}\right), \tag{39}$$

while because $dp/dh>0$ by virtue of (32c), the inequality $dp/dh>0$ holds on all the phase trajectories. The value of h_∞ is determined from (39) taking into account the initial condition $p_\infty = 1$:

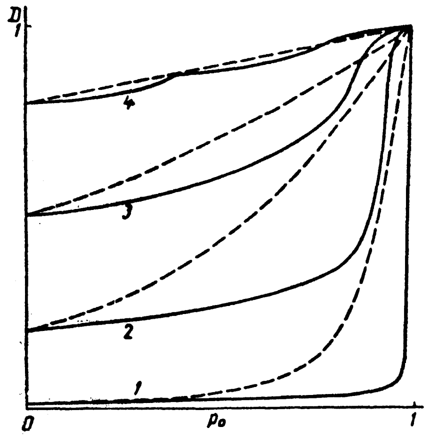

FIG. 2. Concentration dependence $D_{\text{av}}(p_0)$ (in units of D_{c0}^1) for various values of h_0: *1*—0.01; *2*—0.2; *3*—0.5; *4*—0.8. The solid curves correspond to the three-dimensional problem, the dashed curves to the two-dimensional problem.

$$1-p_0=\frac{3}{14}\ln\left(\frac{1-h_0}{1-h_\infty}\right)+\frac{2}{7}\ln\left(\frac{11+3h_0}{11+3h_\infty}\right), \tag{40}$$

which to first order in $(1-p_0)$ gives

$$h_\infty=h_0+\frac{2(1-p_0)(1-h_0)(11+3h_0)}{3(1+h_0)}. \tag{41}$$

Substituting (41) into (31) and taking into account that as $p \to 1$, $W(p) \approx 1/3$, we obtain

$$D_{\text{av}}=D_{c0}{}^1\left\{1+\frac{2(1-p_0)(1-h_0)}{(1+h_0)}\right\}^{-1}. \tag{42}$$

In the remaining cases, Eq. (32) is integrated numerically, and the exponent in Eq. (31) is also calculated by numerical integration. The results of these calculations are shown in Fig. 2 as solid curves (for comparison, the corresponding functions for the two-dimensional case are plotted as dashed curves). The function $D_{\text{av}}(p_0)$ so obtained is monotonic for fixed h_0, and has two inflection points, corresponding to the separatrices on the phase portrait.

TIME DEPENDENCE OF THE MEAN SQUARE DISPLACEMENT

The quantity D_{av} calculated above is asymptotic when the scale of particle migration is large, and analysis of the phase portrait shows that complete averaging occurs in the system. For finite scales or after a finite migration time, fluctuations in the positions of the regions can turn out to have a considerable influence of the character of the motion of the migrating particles, as a result of which the time dependence of the observable turns out to be nontrivial. In principle, we can determine the time dependence of the quantity $D_{\text{av}}(n)$ that we have introduced for finite values of n; however, it is more interesting to construct the time dependence of the mean square displacement $R^2_{\text{av}}(t)$.

In order to obtain the required function, we make use of the scaling transformation algorithm obtained above. However, in order to obtain the time dependence, by analogy with what we did in the two dimensional case we consider variation of the temporal rather than the spatial scale as we did above.

Let us modify the discrete algorithm for the scaling transformation in the following way. Each step of the scaling transformation corresponds to a doubling of the time

$$t_{n+1}=2t_n. \tag{43}$$

During motion along a type I edge, the mean square displacement after a time t_n equals the square of the edge, L_n^2, for the lattice at the corresponding scale S_n, while after the doubled time $2t_n$ $-2L_n^2$. During motion along the "slow" edges, the squared displacement equals $h_n L_n^2$ and $2h_n L_n^2$ after times t_n and $2t_n$, respectively [for the original lattice S_0, the mean square displacement along a type II edge equals $(\tau_0^{\rm I}/\tau_0^{\rm II})L_0^2$]. If after a time $2t_n$ a particle has traversed edges of both types (analogous to the mixed edge of the consolidated lattice), the mean square displacement will equal $4h_n L_n^2/(1+h_n)$. As a result, in place of the discrete relations (22) and (23) we obtain

$$R_{{\rm I},n+1}^2=2\left\{1-\frac{d}{d+2}(1-p_n)\left[1-\frac{4\alpha_n h_n}{1+h_n}\right]\right\}R_{{\rm I},n}^2, \tag{44}$$

$$R_{{\rm II},n+1}^2=2\left\{1-\frac{d}{d+2}p_n\left[1-\frac{4(1-\alpha_n)}{1+h_n}\right]\right\}R_{{\rm II},n}^2, \tag{45}$$

where $d=3$ is the dimension of the space. When we pass to the infinitesimal transformation, in place of (25) and (26) we obtain the differential equations

$$\frac{dR_{\rm I}^2}{dn}=\left\{1-\frac{3}{5}(1-p_n)\left[1-\frac{4\alpha_n h_n}{1+h_n}\right]\right\}R_{\rm I}^2, \tag{46}$$

$$\frac{dR_{\rm II}^2}{dn}=\left\{1-\frac{3}{5}p_n\left[1-\frac{4(1-\alpha_n)}{1+h_n}\right]\right\}R_{\rm II}^2. \tag{47}$$

Further transformations give

$$R_{\rm I}^2(n)=p_0L_0^2\exp\left\{n-\frac{3}{5}\int_0^n[1-p(k)]\times\left[1-\frac{4\alpha(k)h(k)}{1+h(k)}\right]dk\right\}. \tag{48}$$

$$R_{\rm II}^2(n)=(1-p_0)h_0L_0^2\exp\left\{n-\frac{3}{5}\int_0^n p(k)\times\left[1-\frac{4(1-\alpha(k))}{1+h(k)}\right]dk\right\}, \tag{49}$$

where $\alpha(n)\equiv\alpha[p(n),h(n)]$ is given by Eq. (18).

In addition to the equations for the functions p and h, which retain the forms (24) and (29), we obtain from (43) an equation for the function $t(n)$: $dt(n)/dn=t(n)$, which relates the scaling transformation parameter n to the time t:

$$t(n)=\tau_0^{\rm I}\exp(n). \tag{50}$$

The average value of the mean square displacement of diffusing particles as a function of time $R_{\rm av}^2(t)$ is expressed in terms of $R_{\rm I}^2(t)$ and $R_2^{\rm II}(t)$ from (48) and (49), in which the dependence on n is replaced by a dependence on t by using Eq. (5):

$$R_{\rm av}^2(t)=R_{\rm I}^2(t)+R_{\rm II}^2(t). \tag{51}$$

As a result of some simple transformations, we obtain an expression analogous to (31):

$$R_{\rm av}^2=\{p(t)+[1-p(t)]h(t)\}\exp\left[-\int_{h_0}^{h(t)}\frac{dh}{h+W(h)}\right]D_{\rm c0}^{\rm I}t. \tag{52}$$

Note that Eq. (52) can be written in the form $R_{\rm av}^2(t)=D_{\rm av}(t)t$, while $\lim_{t\to\infty}D_{\rm av}(t)$ equals the quantity $D_{\rm av}$ defined in (31); this demonstrates the correctness of the renormalization group when time scales replace spatial scales.

In order to compute the integral in (52), it is necessary to have an explicit expression for the function $W(h)\equiv W[p(h)]$, i.e., to have an explicit solution to Eq. (32). Furthermore, the functions $p(t)$ and $h(t)$ enter in (32). Their explicit form is determined from this system of equations (24), (29), which also requires the solution of Eq. (32). However, since Eq. (32) cannot be integrated in general, we can obtain the function $R_{\rm av}^2(t)$ in analytic form only in a few limiting cases, for which we can construct the solution to Eq. (32).

Using (33), (35), and (39)—the solutions to Eqs. (32a), (32b), and (32c)—we calculate the integral in (52). Then, substituting (33), (35), and (39) into Eq. (29), which we simplify in a corresponding fashion, we obtain ordinary differential equations for the function $h(n)$ whose solution taking (50) into account gives the functions $h(t)$ and $p(t)$. As a result, we obtain the function $R_{\rm av}^2(t)$ for finite times (i.e., as a result of a finite number of scaling transformation steps) in the limiting cases considered above:

1) $h_0\to 0$, p_0 such that $h_0/(1-p_0)^2\ll 1$:

$$R_{\rm av}^2(t)=\frac{p_0D_{\rm c0}^{\rm I}t}{p_0+(1-p_0)(t/\tau_0^{\rm I})^{3/5}}\times\left\{1+\frac{h_0(1-p_0)(t/\tau_0^{\rm I})^{8/5}}{p_0[p_0+(1-p_0)(t/\tau_0^{\rm I})^{3/5}]^2}\right\}, \tag{53a}$$

2) $p_0\to 0$ for any h_0:

$$R_{\rm av}^2(t)=D_{\rm c0}^{\rm I}t\left\{p_0\left(\frac{\tau_0^{\rm I}}{t}\right)^{3/5}+h_0\left[1-p_0\left(\frac{\tau_0^{\rm I}}{t}\right)^{3/5}\right]\left[1+p_0\left[1-\left(\frac{\tau_0^{\rm I}}{t}\right)^{3/5}\right]\times\frac{2(1-h_0)(11h_0+3)}{3(1+h_0)}\right]\right\}\times\left\{1+\frac{22}{3}p_0\left[1-\left(\frac{\tau_0^{\rm I}}{t}\right)^{3/5}\right]\frac{h_0(1-h_0)}{1+h_0}\right\}^{-1}$$
$$\approx D_{\rm c0}^{\rm I}h_0t\left\{1+2p_0\frac{1-h_0}{1+h_0}\left[1-\left(\frac{\tau_0^{\rm I}}{t}\right)^{3/5}\right]\right\}, \tag{53b}$$

3) $p_0\to 1$, h_0 such that $h_0/(1-p_0)^2\gg 1$:

$$R_{\rm av}^2(t)=\frac{D_{\rm c0}^{\rm I}t[1-(1-p_0)(1-h_0)(\tau_0^{\rm I}/t)^{3/5}]}{1+2(1-p_0)[1-(\tau_0^{\rm I}/t)^{3/5}](1-h_0)/(1+h_0)}$$
$$\approx D_{\rm c0}^{\rm I}t\left\{1-(1-p_0)\frac{1-h_0}{1+h_0}\left[2-(1-h_0)\left(\frac{\tau_0^{\rm I}}{t}\right)^{3/5}\right]\right\}. \tag{53c}$$

To sum up, we have found that for small times the function $R_{\rm av}^2(t)$ is nonlinear, but that for $h_0\neq 0$ its asymptotic form when $t\gg\tau_0^{\rm I}[p_0(1-p_0)/h_0]^{5/3}$ becomes linear for all the limiting cases, while the coefficient of proportionality coincides precisely with the average DC corresponding to the given limiting cases.

For $h_0 \equiv 0$, as occurs in the two-dimensional situation, the function $R^2_{av}(t)$ never enters the linear regime:

$$R^2_{av}(t) = \frac{p_0 D_{c0}{}^{I} t}{p_0 + (1-p_0)(t/\tau_0{}^{I})^{3/5}} = \frac{p_0 L_0{}^2 (t/\tau_0{}^{I})}{p_0 + (1-p_0)(t/\tau_0{}^{I})^{3/5}}. \quad (54)$$

We note that in three-dimensional space the nonlinearity is weaker than in the two-dimensional case; this is easily understood if we recall that in three-dimensional space the probability that the path of a diffusing particle will return to its point of origin equals zero, while in the two-dimensional case it is finite.[1] For this reason, the influence of fluctuations on the course of the diffusion process in three-dimensional space is found to be weaker than in two-dimensional space. As a result, for $h_0 \equiv 0$ we have $R^2_{av} \propto t^{2/5}$ in the three dimensional case, and not $R^2_{av} \propto t^{1/2}$ as occurs in a plane (see Refs. 4, 6, 14).

DISCUSSION OF RESULTS

Let us investigate the questions of accuracy and region of applicability of the results obtained here separately. In percolation theory it is well known that the renormalization group equations of type (24)–(26) correctly describe the scaling properties of systems up to scales on the order of the correlation length, and also allow us to determine the limiting values of the quantities under study to a high degree of accuracy. However, the rate at which these quantities reach their limiting values is usually slowed. In the scaling approach, the quantities under study reach their limiting values by way of a power law, while in other approaches, and in numerical experiments, this rate is exponential. Consequently, the reliability of the limiting values we have obtained for the average diffusion coefficients D_{av} (34), (38), and (42), and also the results of numerical integration shown in Fig. 2, should not be doubted.

However, the calculated results (53) for $R^2_{av}(t)$ are valid only up to times for which $(D^{I}_{0} t)^{1/2}$ is less than or on the order of the correlation length, i.e., the Eqs. (53) we have obtained are intermediate asymptotic forms. On the other hand, for $h_0 \neq 0$, as $t \to \infty$ the functions (53) reach the limiting asymptotic behavior $R^2_{av}(t) \propto t$, which by virtue of the considerations presented above is also correct. However, the passage from intermediate asymptotic behavior to the diffusion regime apparently should not take place by way of a power law (as occurs in this paper), but rather should be exponential. For $h_0 \neq 0$, our results also have a finite range of applicability. For scaling times corresponding to displacements larger than the correlation length, Eqs. (24) and (46) are poor descriptions of the variation of the quantities $p(n)$ and $R^2_{av}(t)$. We note in particular that changing the correlation properties [the value of the parameter a in the expression for the correlation function (20)] does not affect the limiting value D_{av}.

Unfortunately, we have not been successful in devising a universal recipe for correctly modifying these equations. To begin with, this is connected with the fact that we cannot in general identity the interval of time over which the intermediate asymptotic form is valid, since we have been unable to accurately determine the correlation length quantitatively for diffusing systems when it differs greatly from the percolation correlation length. However, as $h_0 \to 0$ (i.e., limiting case (1)), we have succeeded in making the corresponding estimates and modifications.

As $h_0 \to 0$, we determine the consolidation scale that corresponds to the correlation scale from Eq. (24) and the condition $d^2 p(n)/dn^2 = 0$:

$$n_{cor} \sim -5/3 \ln[(1-p_0)/p_0]. \quad (55)$$

Consequently, the time scale up to which our scaling approach is valid is in order of magnitude equal to

$$t_{cor} \sim \tau_0{}^{I} [p_0/(1-p_0)]^{5/3}. \quad (56)$$

Furthermore, the rate of decrease of the volume fraction (or probability) of the fast regions for scales larger than the correlation scale (when $h_0 \equiv 0$ this is the rate of destruction of the diffusing particles) should be proportional to their number (the probability p) and the number of regions with irreversible traps, which is proportional to $(R^2_{av})^{3/2} \propto (t/\tau^{I}_0)^{3/5}$ (i.e., to the volume occupied by the diffusing particles). Consequently, when we have

$$[(1-p_0)/p_0](t/\tau_0{}^{I})^{3/5} \sim 1$$

and $1-p \sim 1$, Eq. (24) for the correlation scale is modified in the following way:

$$t\frac{dp(n)}{dt} \approx -\frac{3}{5} p(t)(1-p)[1-2\alpha(p,h)]\frac{1-p_0}{p_0}\left(\frac{t}{\tau_0{}^{I}}\right)^{3/5}, \quad (57)$$

where we have changed from the variable n to the variable t, taking into account (50).

Then, using the solution to Eq. (57), which is matched for $t \approx t_{corr}$ with the solution to Eq. (24), we find that the intermediate asymptotic form (53a) becomes the asymptotic expression

$$R^2_{av}(t) \approx D_{c0}{}^{I} t \left\{ \frac{h_0}{(1-p_0)^2} + \frac{p_0}{1-p_0} \exp\left[-\frac{1-p_0}{p_0}\left(\frac{t}{\tau_0{}^{I}}\right)^{3/5}\right]\right\}, \quad (58)$$

while for $h_0 \equiv 0$ the intermediate asymptotic form (54) becomes the asymptotic form

$$R^2_{av}(t) \approx \frac{p_0 D_{c0}{}^{I} t}{1-p_0} \exp\left[-\frac{1-p_0}{p_0}\left(\frac{t}{\tau_0{}^{I}}\right)^{3/5}\right]. \quad (59)$$

Note that the asymptotic form (59) precisely coincides with the results of Ref. 6. The character of the behavior of $R^2_{av}(t)$ for $h_0 \equiv 0$ is easy to explain. At the initial stage, the primary effect is one of diffusive smearing, and the destruction of migrating particles (including that due to the influence of fluctuations) is insignificant. As time passes, the destruction of particles becomes more effective and the average squared displacement begins to decrease, since in our method of defining R^2_{av} particles that are annihilated do not contribute to the mean square displacement.

Incidentally, this condition of matching of asymptotic forms can be used to justify our choice of the value of the coefficient $a = 2/d$ in the correlation function (20). Suppose that the coefficient in the corresponding equations for scale transformations is not $d/(d+2)$, but rather some arbitrary constant ν. Then the time corresponding to the correlation scale is the quantity $t_{corr} \sim (1-p_0)^{-1/\nu}$. On the other hand, from (54) we find that $R^2_{av}(t) \propto t^{1-\nu}$, from which it follows that the volume occupied by the diffusing particles is of order $t^{d(1-\nu)/2}$. Setting the exponents equal, we obtain a relation determining the constant ν:

$$d(1-\nu)/2=\nu, \tag{60}$$

from which it also follows that $\nu = d/(d+2)$ (for $d=3$, $\nu = 3/5$).

Thus, we can assert that the results we have obtained are correct for finite times, and also in those cases where the system reaches the diffusion regime, and for very long times. Furthermore, the considerations presented above regarding the limits of applicability of the scaling approach we have given here are valid for both three-dimensional and two-dimensional spaces.[14]

CONCLUSION

Our procedure for calculating the effective DC of a heterogeneous medium using scaling transformations shows that a unified approach is possible in both the three-dimensional and two-dimensional situations: the law of scale transformations is defined as the smallest possible coarsening of the lattice (by a factor of two). In this case specifics connected with the dimensionality of the space arise only at the stage of partitioning the mixed paths consisting of links of various types into two groups, fast and slow. For the percolation problem in three-dimensional space (whose results we used at this stage) there exists a region of combined connectivity that is absent in two-dimensional space, which corresponds to region II on the phase portrait of the system of renormalization group equations (see Fig. 1). As a result, the rate of growth of the effective DC in the medium for small values of the parameter p_0 is small and increases sharply after the initial point of the path passes from region II to region I (for fixed values of h_0). For comparison we show the corresponding concentration dependence of $D_{av}(p_0)$ in Fig. 2, using dashed curves for the two-dimensional system. Furthermore, if the parameters p_0 and h_0 belong to region II, then averaging in the three-dimensional case takes place much more slowly than in the two-dimensional case (if the parameters p_0 and h_0 belong to regions I or III, then, as we have already noted above, the nonlinearity in the three-dimensional case is weaker), while the contribution corresponding to fluctuations falls off like $t^{-3/20}$.

We note one other important difference between the two-dimensional and three-dimensional situations. As we have already pointed out above, the probability that the random walk of a diffusing particle will return equals zero in three-dimensional space, in contrast to two-dimensional space; this leads to a weakening of correlations in the properties of various portions of the path.

In conclusion, we point out several real physical processes whose description could benefit from the use of our model. One example is the diffusion of small molecules in dilute polymer solutions, in which case the regions of type I correspond to diffusion in the pure solute and regions in type II to diffusion through the polymer coils. This situation was studied in Ref. 2. We may assume that regions of type II correspond to reversible traps with a lifetime $\tau_2^{II} - \tau_1^{I}$; for $\tau_2^{II} \to \infty$, i.e., $h_0 \to 0$, these correspond to true trapping centers. A similar situation was studied in Refs. 7 and 8; as we have already noted above, in these papers changes in the character of the time dependence were observed analogous to those obtained in our paper (in the corresponding expressions t is replaced by $t^{d/(d+2)}$). The asymptotic time dependences we have obtained are closest to the power law dependences that arise in describing dispersive transport, although the exponents in them are determined not only by the dimension of the space but also by other factors.[4] The case $h_0 = 0$ describes the kinetics of molecular chemical reactions with randomly located initiation centers as well.[6]

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