PHYSICAL REVIEW B VOLUME 25, NUMBER 7 1 APRIL 1982

Long-time tail effects on particle diffusion in a disordered system

Joseph W. Haus
Fachbereich Physik, Universität Essen GHS, 4300 Essen 1, Federal Republic of Germany

Klaus W. Kehr
Institut für Festkörperforschung der Kernforschungsanlage Jülich, 5170 Jülich, Federal Republic of Germany

Kazuo Kitahara
Department of Liberal Arts, Shizuoka University, Shizuoka 422, Japan
(Received 21 January 1982)

We present a simple formulation of an effective-medium approximation to a model of particle transport in a random medium. The small- and large-time asymptotic properties of the effective transition rate in one dimension are computed. We also derive the first three corrections to the leading asymptotic terms in the expansion. The results for the mean-square displacement and fourth moment are in excellent agreement with a Monte Carlo simulation.

Classical diffusion in a disordered system has received a revival of interest in the literature.[1–6] In a single dimension, Alexander *et al.*[1] calculated some properties of single-particle diffusion using a set of master equations with distributions of transition rates, a special class of singular distributions were analyzed in detail. A class of one-dimensional hopping models with random lengths between sites was exactly solved by van Beijeren[2]; he discusses the long-time tail in the velocity autocorrelation function.

The problem of random transition rates has been treated more recently by Odagaki and Lax[3] and Webman,[4] using a effective-medium theory. They applied their approach to the bond percolation problem in higher dimensions. Machta[5] has derived a long-time tail of the velocity autocorrelation function in one dimension by a novel renormalization-group approach. One class of these models corresponds to the model of van Beijeren.[2] Another class of trap models cannot exhibit long-time tails, when equilibrium conditions prevail, as has been shown recently by two of the present authors.[6]

In this Communication we present our results for the frequency dependence of the particle's mobility within an effective-medium approximation. We have a simpler formulation than given in previous work,[3,4] one which can be easily extended to clusters of random transition rates.

Consider a master equation with nearest-neighbor transition rates $W_{\vec{n}',\vec{n}}$ from the $\vec{n}'$ to site $\vec{n}$; $W_{\vec{n}',\vec{n}}$ is a random function of the site variables $\vec{n}'$ and $\vec{n}$ and is symmetric $W_{\vec{n}',\vec{n}} = W_{\vec{n},\vec{n}'}$,

$$\frac{dP(\vec{n}t|\vec{0}0)}{dt} = \sum_{\vec{n}'} W_{\vec{n}',\vec{n}}[P(\vec{n}'t|\vec{0}0) - P(\vec{n}t|\vec{0}0)] \ . \tag{1}$$

$P(\vec{n}t|\vec{0}0)$ is the conditional probability that the particle is at site $\vec{n}$ at time t given that it was at site $\vec{0}$ at $t=0$. We explicitly choose a cluster of N sites, the transition rates outside this cluster are replaced by a time convolution of an effective nearest-neighbor transition $W(t)$ with the conditional probability. We suppress the subscripts, since $W(t)$ is understood to extend only to nearest neighbors.

We define a function $\Delta_{\vec{n}',\vec{n}}$ with the following property:

$$\Delta_{\vec{n}',\vec{n}} = \begin{cases} 1 & \text{if } \vec{n}' \text{ and } \vec{n} \text{ are nearest neighbors connected by stochastic transition rates} \\ 0 & \text{otherwise;} \end{cases} \tag{2}$$

then using the shorthand notation $P_{\vec{n}}(t) = P(\vec{n}t|\vec{0}0)$, we have the set of equations

$$\frac{dP_{\vec{n}}(t)}{dt} = -\sum_{\vec{n}'}^{\mathrm{NN}} \left\{ \int_0^t dt'\, W(t-t')(1-\Delta_{\vec{n}',\vec{n}})[P_{\vec{n}}(t') - P_{\vec{n}'}(t')] + W_{\vec{n}',\vec{n}}\Delta_{\vec{n}',\vec{n}}[P_{\vec{n}}(t) - P_{\vec{n}'}(t)] \right\} \ , \tag{3}$$

where the sum over $\vec{n}'$ extends only to nearest neighbors (NN).

These equations may be treated by Fourier-Laplace transformation methods. In this manner, the equations are diagonalized, except for a set of N coupled equations for the sites connected by random transition rates. However, these are linear equations.

In order to determine the function $W(t)$, we use a self-consistency condition; namely, the average over the remaining random transition rates must yield conditional probabilities consistent with that given by Eq. (2) when there are *no* random transition rates.[7]

In the Laplace transformed time variable this condition is

$$\langle P_{\vec{n}}(s)\rangle = \frac{1}{\|\Omega_0\|}\int_{\Omega_0} \frac{d^d k \exp(i\vec{k}\cdot\vec{n})}{s+W(s)\sum_{\vec{n}'}^{\mathrm{NN}}[1-\exp(i\vec{k}\cdot\vec{\delta}_{\vec{n}'})]} . \tag{4}$$

The brackets denote an average over the remaining fluctuating transition rates and the integral extends over a d-dimensional unit cell in the reciprocal space; $\|\Omega_0\|$ denotes the volume of this unit cell and $\vec{\delta}_{\vec{n}'}$ is the vector from the lattice site $\vec{n}$ to one of the nearest-neighbor sites $\vec{n}'$.

In the rest of this work we shall restrict ourselves to a one-dimensional lattice of sites separated by a distance a. Equation (4) is now ($\vec{n}=\vec{0}$)

$$\langle P_0(s)\rangle = E_0(s) , \tag{5}$$

where

$$E_n(s) = \frac{a}{\pi}\int_0^{\pi/a} \frac{dk \cos(nka)}{s+2W(s)[1-\cos(ka)]} . \tag{6}$$

For $N=2$ we have one fluctuating bond, say $W_{0,1}$. The solution of Eq. (3) with the self-consistency condition,[8] yields the same results as in Refs. 2 and 3

$$\left\langle \frac{W(s)-W_{0,1}}{W_{0,1}+sE_0(s)[W(s)-W_{0,1}]}\right\rangle = 0 . \tag{7}$$

For a cluster of three sites, $N=3$, the solution Eq. (3) and the self-consistency requirement yields

$$\left\langle \frac{[W(s)-W_{0,1}]\{1+[E_1(s)-E_0(s)][W(s)-W_{0,-1}]\}}{Q(s)}\right\rangle = 0 , \tag{8a}$$

where

$$Q(s) = \{1+2[E_1(s)-E_0(s)][W(s)-W_{0,1}]\}\{1+2[E_1(s)-E_0(s)][W(s)-W_{0,-1}]\}$$
$$-[2E_1(s)-E_2(s)-E_0(s)][W(s)-W_{0,1}][W(s)-W_{0,-1}] . \tag{8b}$$

In one dimension both Eqs. (7) and (8) yield the following asymptotic results up to second order in the expansion variable:

(a) $s\rightarrow\infty$,

$$W(s) = D_\infty\left(1+\frac{\delta_1}{s}+\frac{\delta_2}{s^2}\right) , \tag{9}$$

where

$$D_\infty = \langle W_{0,1}\rangle ,$$

$$\delta_1 = -2\langle (W_{0,1}-D_\infty)^2\rangle / D_\infty ,$$

and

$$\delta_2 = 4\langle (W_{0,1}-D_\infty)^3\rangle / D_\infty + 6\langle (W_{0,1}-D_\infty)^2\rangle .$$

(b) $s\rightarrow 0$,

$$W(s) = D_0(1+\theta_1\sqrt{s}+\theta_2 s) , \tag{10}$$

where

$$D_0^{-1} = \left\langle \frac{1}{W_{0,1}}\right\rangle ,$$

$$\theta_1 = \tfrac{1}{2}D_0^{3/2}\left\langle\left(\frac{1}{W_{0,1}}-\frac{1}{D_0}\right)^2\right\rangle ,$$

and

$$\theta_2 = -\tfrac{1}{4}D_0^2\left\langle\left(\frac{1}{W_{0,1}}-\frac{1}{D_0}\right)^3\right\rangle + \tfrac{3}{2}\theta_1^2 .$$

The next coefficient in Eq. (9), as derived from Eq. (7), is

$$\delta_3 = -8\langle (W_{0,1}-D_\infty)^4\rangle / D_\infty - 24\langle (W_{0,1}-D_\infty)^3\rangle$$
$$+4\langle (W_{0,1}-D_\infty)^2\rangle^2 / D_\infty - 20 D_\infty \langle (W_{0,1}-D_\infty)^2\rangle ; \tag{11}$$

while in the same approximation, the coefficient of $s^{3/2}$ in Eq. (10) is

$$\theta_3 = \tfrac{1}{8} D_0^{5/2} \left\langle \left(\frac{1}{W_{0,1}} - \frac{1}{D_0} \right)^4 \right\rangle + \tfrac{21}{64} D_0^{9/2} \left\langle \left(\frac{1}{W_{0,1}} - \frac{1}{D_0} \right)^2 \right\rangle^3 - \tfrac{7}{16} D_0^{7/2} \left\langle \left(\frac{1}{W_{0,1}} - \frac{1}{D_0} \right)^3 \right\rangle \left\langle \left(\frac{1}{W_{0,1}} - \frac{1}{D_0} \right)^2 \right\rangle - \tfrac{1}{4} D_0^{5/2} \left\langle \left(\frac{1}{W_{0,1}} - \frac{1}{D_0} \right)^2 \right\rangle^2 - \tfrac{1}{16} D_0^{1/2} \left\langle \left(\frac{1}{W_{0,1}} - \frac{1}{D_0} \right)^2 \right\rangle . \tag{12}$$

The corresponding coefficients obtained from Eq. (8) differ from the above results only by a factor $-\langle (W_{0,1} - D_\infty)^2 \rangle^2 / 8 D_\infty$, and $-\frac{1}{8} D_0^{5/2} \times \langle (1/W_{0,1} - 1/D_0)^2 \rangle^2$, respectively.

Also, we note that the diffusion coefficients D_0 and D_∞ are exact. The term θ_1 presents a long-time anomaly in the velocity autocorrelation function, which is the inverse Laplace transform of $W(s)$. This function has a long-time tail proportional to $t^{-3/2}$, a typical behavior for one-dimension disordered systems.[2,5] The mean-square displacement in Laplace transformed form is

$$\langle x^2 \rangle (s) = 2 W(s) a^2 / s^2 , \tag{13}$$

it has correction terms proportional to $t^{1/2}$, t^0, and $t^{-1/2}$ for long times, which are obtained from Eqs. (10) and (12).

A quantitative comparison of the above results was made with a Monte Carlo computer simulation of the model. We consider random transition probabilities with two values Γ and $\Gamma^<$ (for the present work we chose $\Gamma^< = \frac{1}{20}\Gamma$ and equal numbers of both species of transition rates). In Fig. 1, a log-log plot of the mean-square displacement over four decades of time is presented. The short-time behavior following from Eq. (9) is given by the full line; it yields a good fit to the data up to about $t \simeq 1$. For times greater than $t \gtrsim 5$ the long-time asymptotic approximation from Eq. (10) is an excellent representation of the data. In view of the agreement with the numerical simulation where θ_1 and θ_2 represents significant corrections and because θ_1 and θ_2 are unchanged by including larger clusters, we conjecture that they represent exact results.

The fourth moment is more sensitive to the functional form of $W(t)$, since it is a nonlinear functional of this rate

$$\langle x^4 \rangle (s) = \left(\frac{2 W(s)}{s^2} + \frac{24 W^2(s)}{s^3} \right) a^4 . \tag{14}$$

Figure 2 shows the agreement between the effective-medium theory and the computer simulation.

The effective-medium theory yields a quantitative representation of the data in one dimension, and it is not restricted to a single dimension. Future work will be devoted to a more detailed analysis of the above problem and especially, the contributions of the long-time tails to the physical moments and mobility in higher dimensions.

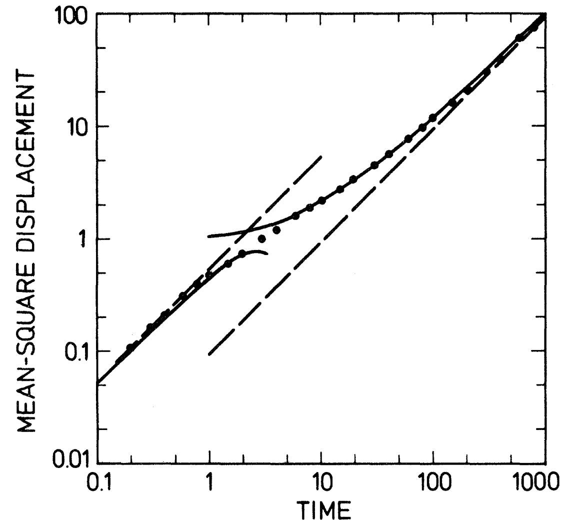

FIG. 1. The log-log plot of the mean-square displacement vs time. The time is scaled to units $1/2\Gamma$ and the mean-square displacement is scaled to units of a^2. The dashed lines represent the long- and short-time asymptotic behavior.

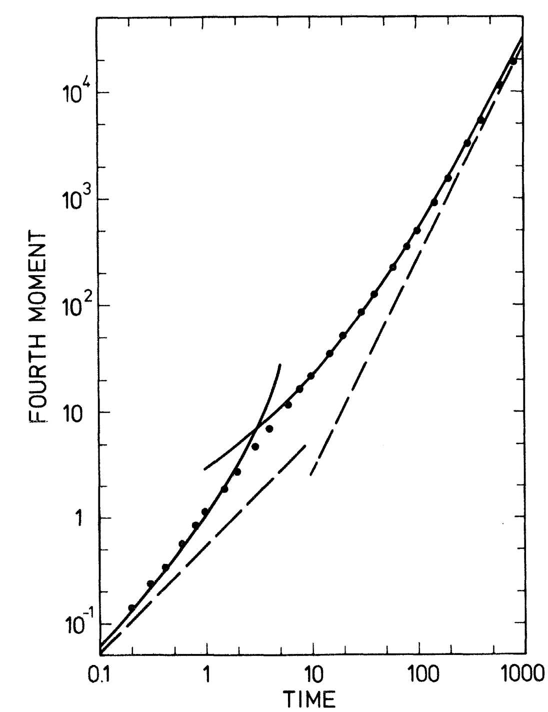

FIG. 2. The log-log plot of the fourth moment of the displacements vs time.

ACKNOWLEDGMENTS

We thank K. Mika and K. Wingerath for help in programming the symbolic formulas and H. van Beijeren for discussions concerning the work. This work is financially supported by the Grant-in-Aid for Scientific Research of the Ministry of Education, Science and Culture of the Japanese Government.

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[1]S. Alexander, J. Bernasconi, W. R. Schneider, and R. Orbach, Rev. Mod. Phys. 53, 175 (1981).

[2]H. van Beijeren, Rev. Mod. Phys. 54, 195 (1982).

[3]T. Odagaki and M. Lax, Phys. Rev. B 24, 5284 (1981).

[4]I. Webman, Phys. Rev. Lett. 47, 1496 (1981).

[5]J. Machta, Phys. Rev. B 24, 5260 (1981).

[6]J. W. Haus, K. W. Kehr, and J. W. Lyklema, Phys. Rev. B (in press).

[7]H. Shiba, Prog. Theor. Phys. 46, 77 (1971).

[8]Webman's expression must be evaluated for $\epsilon = s/2W(s)$. Our equation agrees with Ref. 3.