II. Tracer diffusion in a system with randomly distributed traps

1997 J. Phys.: Condens. Matter 9 7949

(http://iopscience.iop.org/0953-8984/9/38/004)

J. Phys.: Condens. Matter **9** (1997) 7949–7961. Printed in the UK PII: S0953-8984(97)81198-8

II. Tracer diffusion in a system with randomly distributed traps

L F Perondi†§, R J Elliott†, and K Kaski‡

† University of Oxford, Department of Physics, 1 Keble Rd, Oxford OX1 3NP, UK
‡ Helsinki University of Technology, Laboratory of Computational Engineering, Miestentie 3, FIN-02150 Espoo, Finland

Received 22 January 1997

Abstract. An approximate theory for tracer diffusion on a lattice containing randomly distributed traps and in the presence of a finite concentration of diffusing particles which preclude double occupancy of any site is developed by extending earlier theories of diffusion in a many-particle system on perfect lattices using random-walk concepts. Both blocking and dynamic correlation effects are considered. The theoretical results are compared with computer simulation for a two-dimensional square lattice with two types of trap over the entire concentration range for particles and traps. The agreement between the simulation results and the theory is satisfactory and gives confidence that the approximation will be valid more widely in models of diffusion in disordered lattices.

1. Introduction

In this and the previous paper [1], we study two simple models of systems of classical particles undergoing activated hopping motion on a disordered lattice. These represent, in very idealized form, the physics of systems which display superionic conduction and are particularly relevant to the study of ionic motion in glassy materials such as lithium boracites used as solid-state electrolytes [2]. In particular we study the characteristics of motion of a single tagged tracer particle in the presence of a finite concentration of other particles where the interaction is taken into account by precluding the possibility that two particles can simultaneously occupy the same site. This avoidance of double occupancy gives rise to a dynamic disorder which causes correlations in the motion of the tracer particle. For a general introduction to these systems see the previous paper [1] and references therein.

Figure 1(b) of [1] represents the case treated here where wells of varying depth are distributed at random on a lattice. As is clear from this figure the potential barriers which must be surmounted differ from site to site and the hopping rates between wells of different depths are no longer symmetric. Our main objective is to consider a qualitative model for the diffusion coefficient of a tagged particle including the concepts of blocking and correlation. It is known from earlier work that a tagged particle in a many-particle system without static disorder shows both of these effects. However, as will be shown in section 2, even a one-particle system with random traps displays a reduction in its diffusivity. It is therefore to be expected that the blocking in a many-particle system in the presence of

§ Permanent address: Laboratório Associado de Sensores e Materiais, Instituto de Pesquisas Espaciais (INPE), São José dos Campos, São Paulo, Brazil.

0953-8984/97/387949+13$19.50

traps will have two contributions. The effect on correlations is more subtle. Although no correlations are induced by the direct interaction of the tagged particle with the traps, there is the possibility that the traps, by affecting the movement of the background particles, will induce changes in the dynamic correlation factor. The qualitative model considered in this paper allows us to elucidate these effects.

The theoretical predictions are checked against computer simulation results which are carried out for a system with two types of trap randomly distributed in a square lattice. The diffusion coefficient is computed as a function of the concentration of traps and of background particles. Other simulations of many-particle systems with random traps have been performed by Mak *et al* [3], whose studies were concerned with chemical or collective diffusion. A more complete treatment has been given by Kehr and Paetzold [4] including a simulation of tracer diffusion in a variety of lattices. Recent work by Wichmann and Kehr [5] discusses an effective-medium treatment, building on earlier treatments [6, 7], and applies this to collective diffusion for various distributions of site energies for comparison with simulations in one, two, three, four and five dimensions. Our work is more complete for a single case in order to allow detailed comparison with the approximate analytic theory, developed here.

The paper is organized as follows. In section 2 we discuss theoretical models. Section 3 is dedicated to the main aspects of the simulation. In section 4 we compare the simulation results with the prediction of the theory. Finally in section 5 we summarize the main results of this study.

2. Theory and model

We consider a system with only two types of site, A and B, with the corresponding hopping rates denoted by J_A and J_B. In this situation the elements of the hopping matrix are defined by

$$J_{il} = \begin{cases} J_A & \text{if } l \text{ is a site of type } A \\ J_B & \text{if } l \text{ is a site of type } B. \end{cases} \tag{1}$$

We will assume that $J_A > J_B$ and that sites A and B are randomly distributed in a square lattice. The formalism can easily be adapted to other dimensions and lattice geometries. We first consider the problem of a single particle moving in this system and leave discussion of the many-particle case to subsection 2.2.

2.1. The single-particle system

Particles moving in a lattice with randomly distributed traps are known to display normal diffusive properties [8]. It can be shown by a variety of methods [9] that the diffusion coefficient can be written as

$$D = \frac{ZJ_m}{2d} \tag{2}$$

where Z is the number of nearest neighbours, d is the space dimension and

$$J_m{}^{-1} = \left\langle \frac{1}{J} \right\rangle = \frac{1}{N_s} \sum_i \frac{1}{J_i}. \tag{3}$$

In this expression N_s is the number of sites in the lattice and J_i designates the rate of hopping out of site i in a given direction. For a system with two types of site we have

$$J_m^{-1} = \frac{1-x}{J_A} + \frac{x}{J_B} = \frac{1}{J_A f_m} \tag{4}$$

where x is the concentration of type B sites and $f_m = (1 - x + xJ_A/J_B)^{-1}$. In the following we show that the random walk of a particle in this system is not correlated, but only affected by a factor which is caused by the different timescales involved.

The rate equation for a particle moving in the system is given by

$$\frac{\mathrm{d}\langle p_i\rangle}{\mathrm{d}t} = \sum_l (J_{il}\langle p_l\rangle - J_{li}\langle p_i\rangle) \tag{5}$$

where $\langle p_i\rangle$ is the probability that the particle is occupying site i. In the steady-state regime, the equilibrium probabilities for the particle occupying a specific type A or type B site are then related as

$$J_A\langle p_A\rangle = J_B\langle p_B\rangle. \tag{6}$$

From conservation of probability we have that

$$(1-x)\langle p_A\rangle + x\langle p_B\rangle = 1/N_s. \tag{7}$$

From equations (6) and (7) we obtain the following probabilities for occupying either a type A or type B site at equilibrium:

$$\langle P_A\rangle = (1-x)N_s\langle p_A\rangle = \frac{(1-x)/J_A}{(1-x)/J_A + x/J_B} \tag{8}$$

$$\langle P_B\rangle = xN_s\langle p_B\rangle = \frac{x/J_B}{(1-x)/J_A + x/J_B}. \tag{9}$$

In the general many-particle case, using the discrete picture of the random walk, the mean square displacement after an average number of N attempted hops can be written as

$$\langle R^2\rangle = f_b f_c N \tag{10}$$

where f_b and f_c are the 'effective blocking' and correlation factors, respectively. The factor f_b is defined as the fraction of successful jumps out of a given number of attempted hops, i.e.,

$$f_b = N'/N \tag{11}$$

where N' is the number of successful jumps in N attempts. This factor can easily be calculated by considering all possible environments experienced by the random walker, calculating the probabilities of a successful hop in each one, multiplying the latter probabilities by the occurrence probabilities of the respective environments, and adding the results. It contains two effects; one due to the different hopping rates leads to an averaging given by f_m in equation (4) while the second is a true 'blocking' due to the hard-core repulsion between atoms. In the general case these two effects are interrelated so we use the single term *effective blocking factor* to include both.

Returning to the single-particle case and defining ε_0 as the rate of the Poisson process relating the discrete- and continuous-time pictures, the probabilities of a successful jump if the particle is occupying a type A or type B site are given by ZJ_A/ε_0 and ZJ_B/ε_0, respectively. Making use of equations (8) and (9) we then obtain

$$f_b = Z\langle P_A\rangle J_A/\varepsilon_0 + Z\langle P_B\rangle J_B/\varepsilon_0 = \frac{1}{(1-x)/J_A + x/J_B}\frac{Z}{\varepsilon_0}. \tag{12}$$

Noting that $N = \varepsilon_0 t$, we obtain

$$\langle R^2 \rangle = Z J_m f_c t \tag{13}$$

leading to the following expression for the diffusion coefficient:

$$D = \frac{Z J_m f_c}{2d}. \tag{14}$$

Comparing equations (2) and (14) we conclude that in this case $f_c = 1$, showing that the direct interaction of the particle with the traps does not induce correlations in its motion while $f_b = f_m$ since there is no true 'blocking'. This result could have been anticipated by noting that the traps do not cause any spatial imbalance in the probabilities for jumps of the particle.

2.2. The many-particle system

In treating the tracer diffusion coefficient in the present system we follow ideas similar to the ones developed in reference [1], based on a straightforward application of effective-medium results in conjunction with the Tahir-Kheli and Elliott (TKE) [11] approximation for tracer diffusion in systems without static disorder.

As a starting approximation we consider that the tagged particle and the background particles move in an effective medium with a hopping rate, given by equation (4), derived for the problem of a single particle moving in the disordered lattice. In this approximation and in the framework of the TKE interpolating scheme, the diffusion coefficient would be

$$D = (1 - c) f_m f_0 J_A \tag{15}$$

where c is the concentration of the background particles and

$$f_0 = \left(1 - \frac{2c \ \cos\theta_0}{(1 + (1 - c) f_0)(1 + \cos\theta_0)} \right)^{-1}. \tag{16}$$

From equation (15) we recognize the resulting effective blocking factor as being the combination of rate averaging f_m and true blocking by other atoms to give a vacancy concentration $1 - c$:

$$f_b = (1 - c) f_m \tag{17}$$

while the correlation factor is given solely by the dynamical factor f_0. Since the direct interaction of the tagged particle with the traps does not induce any additional correlation effects, it is plausible that the resulting correlation factor involves only the dynamical term. There is the possibility, however, that the traps influence the relative motion of the background particles with respect to the tagged particle causing changes in way in which these particles interact. More specifically, there is the possibility that this effect results in changes in the probabilities for a forward and a backward jump of the tagged particle. We will pursue this point further later on.

The effective blocking factor in this problem due to the traps should depend on the steady-state probabilities of the tagged particle occupying either a type A or a type B site, which in turn depend on the probabilities of the background particles occupying either a type A or a type B site, so it is clearly not given correctly by equation (17). Therefore, the first correction to the effective-medium formula is to improve on this result. As seen in subsection 2.1, the effective blocking factor can be calculated by considering all possible environments experienced by the tagged particle and the probabilities of a successful hop in each one. Figure 1 shows all possible static environments when the tagged particle is

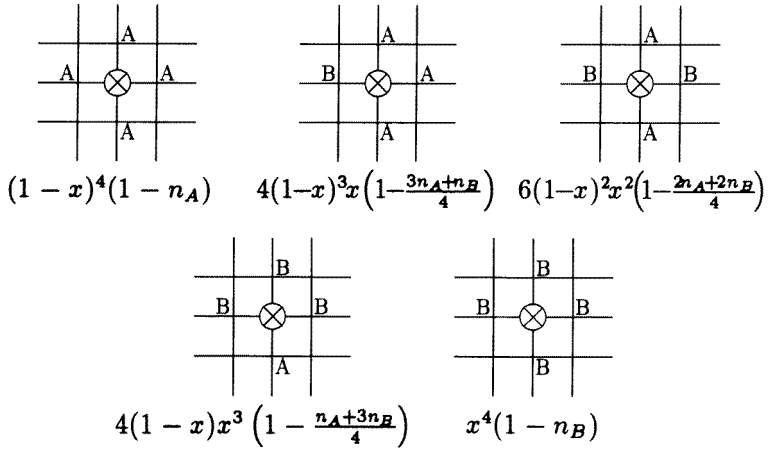

Figure 1. Possible static environments experienced by the tagged particle. Here n_A and n_B have the same meaning as $\langle n_A\rangle$ and $\langle n_B\rangle$ in the text.

occupying either a type A or a type B site. The quantities $\langle n_A\rangle$ and $\langle n_B\rangle$ designate the probabilities that a type A or a type B site is occupied by a background particle. When one considers all possible dynamical arrangements and the probability of a successful hop in each one, the weights shown in figure 1 are obtained. Denoting by P_A and P_B the probabilities that the tagged particle occupies a type A site or a type B site, respectively, it can be shown that the effective blocking factor assumes the form

$$f_b = (1-c)(P_A + P_B J_B/J_A) \tag{18}$$

noting that if the tagged particle is occupying a type B site the relative probability of a successful hop is J_B/J_A, and using the relation

$$(1-x)\langle n_A\rangle + x\langle n_B\rangle = c. \tag{19}$$

To obtain P_A and P_B we make use of the rate equations for a many-particle system. The rate equation for the site occupation probabilities of background particles is given by

$$\frac{\mathrm{d}\langle n_i\rangle}{\mathrm{d}t} = \sum_l (J_{il}\langle n_l(1-n_i-p_i)\rangle - J_{li}\langle n_i(1-n_l-p_l)\rangle). \tag{20}$$

In equilibrium with $\mathrm{d}\langle n_i\rangle/\mathrm{d}t = 0$ we shall assume that the particle averages decouple [10], giving the following condition:

$$J_B\langle n_B\rangle(1-\langle n_A\rangle - \langle p_A\rangle) = J_A\langle n_A\rangle(1-\langle n_B\rangle - \langle p_B\rangle) \tag{21}$$

where $\langle p_A\rangle$ and $\langle p_B\rangle$ are now the probabilities that the tagged particle is occupying a type A or type B site, respectively. Since these are of the order of $1/N_s$ they can be disregarded in equation (21). The resulting equation together with equation (19) form a system of equations the solution of which gives the probabilities $\langle n_A\rangle$ and $\langle n_B\rangle$:

$$\langle n_B\rangle = \frac{K - \sqrt{K^2 - 4cx(1-J_B/J_A)}}{2x(1-J_B/J_A)} \tag{22}$$

$$\langle n_A\rangle = \frac{c - x\langle n_B\rangle}{1-x} \tag{23}$$

where

$$K = (1 - c - x)J_B/J_A + c + x. \tag{24}$$

The probabilities that the tagged particle is occupying either a type A or type B site can be inferred from equation (19) to give

$$P_A = (1 - x)\langle p_A\rangle N_s \tag{25}$$

$$P_B = x\langle p_B\rangle N_s \tag{26}$$

from which we obtain

$$P_A = \frac{1 - x}{1 - x + x\langle n_A\rangle/\langle n_B\rangle} \tag{27}$$

$$P_B = \frac{x\langle n_A\rangle/\langle n_B\rangle}{1 - x + x\langle n_A\rangle/\langle n_B\rangle}. \tag{28}$$

Applying these results in equation (18), the final expression for the effective blocking factor now assumes the form

$$f_b = (1 - c)\left(\frac{1 - x + x(1 - \langle n_A\rangle)/(1 - \langle n_B\rangle)}{1 - x + x\langle n_A\rangle/\langle n_B\rangle}\right) \tag{29}$$

and in our approximation the diffusion coefficient is expressed as

$$D = f_b f_0 J_A. \tag{30}$$

It is easily checked that equation (30) reproduces correctly the single-particle limit. We expect a good agreement with the simulations in the regime of low concentrations of background particles. In the opposite limit, i.e., the single-vacancy limit, the correlation factor predicted by equation (30) is given by $f_0 = (1 + \cos\theta_0)/(1 - \cos\theta_0)$, which is the same result as for a perfect lattice. It is not difficult to see, however, that the dynamical correlation factor in this limit should depend on the relative concentration of traps. For instance, if just after an exchange with the vacancy the tagged particle is occupying a type A site, a return jump will be more probable than if the tagged particle were occupying a type B site. This is because in the first case the vacancy moves on average with a hopping rate lower than that of the tagged particle, J_A. The opposite situation holds if the tagged particle is occupying a type B site. Therefore, in the limit of large particle concentration we expect that the traps will induce changes in the dynamical correlation factor. In what follows, based on the above considerations, we will attempt to derive an expression for the dynamical correlation factor appropriate to the large-c limit.

We will denote by $\cos\theta_1$ the difference between the probabilities for a forward and a backward jump of the tagged particle just after an exchange with the vacancy. Note that $\cos\theta_0$ denotes the value of $\cos\theta_1$ for a perfect lattice. We begin by assuming that the vacancy moves in an effective medium with an effective hopping rate J_v which interpolates between the limits J_A at $x = 0$ and J_B at $x = 1$. Then it is possible to show that

$$(\langle\cos\theta_1\rangle)_\alpha = \frac{J_\alpha}{\varepsilon_0}(U_{mi} - U_{ii}) \qquad (\alpha = A, B) \tag{31}$$

where U designates the usual generating functions associated with the random walk of a single particle.

Following the procedures outlined in section 3 and the appendix of the previous paper we find that in this case

$$(\langle\cos\theta_1\rangle)_A = \frac{J_A\cos\theta_0}{J_v(1 + \cos\theta_0) - J_A\cos\theta_0} \tag{32}$$

$$(\langle\cos\theta_1\rangle)_B = \frac{J_B\cos\theta_0}{J_v(1 + \cos\theta_0) - J_B\cos\theta_0}. \tag{33}$$

The final value of $\langle \cos\theta_1 \rangle$ is obtained by averaging over these two values weighted with the probabilities of the two situations. The probability that, just after an exchange with the vacancy, the tagged particle is occupying a type A site is given by the probability that the vacancy was occupying a type A site just before the exchange. The same principle applies in the case of type B sites. Now if $\langle v_\alpha \rangle$ is the probability that the vacancy occupies a specific type α site, by conservation of probability we have

$$(1-x)\langle v_A \rangle + x\langle v_B \rangle = \frac{1}{N_s}. \tag{34}$$

On the other hand, in the single-vacancy limit the equilibrium condition given by equation (18) may be written as

$$J_B\langle v_A \rangle = J_A\langle v_B \rangle \tag{35}$$

since in this limit $\langle n_A \rangle \sim \langle n_B \rangle \sim 1$. The probabilities that the vacancy occupies any one of the type A or type B sites are given by

$$V_A = (1-x)N_s\langle v_A \rangle \tag{36}$$

$$V_B = xN_s\langle v_B \rangle. \tag{37}$$

Combining equations (34)–(37) we obtain

$$V_A = \frac{1-x}{1-x+xJ_B/J_A} \tag{38}$$

$$V_B = \frac{xJ_B/J_A}{1-x+xJ_B/J_A}. \tag{39}$$

The average value of $\cos\theta_1$ then assumes the form

$$\begin{aligned}\langle \cos\theta_1 \rangle &= V_A(\langle \cos\theta_1 \rangle)_A + V_B(\langle \cos\theta_1 \rangle)_B \\ &= \left(\frac{(1-x)J_A}{J_m(1+\cos\theta_0) - J_A\cos\theta_0}\right)\frac{\cos\theta_0}{1-x+x(J_B/J_A)} \\ &\quad + \left(\frac{xJ_B(J_B/J_A)}{J_m(1+\cos\theta_0) - J_B\cos\theta_0}\right)\frac{\cos\theta_0}{1-x+x(J_B/J_A)}.\end{aligned} \tag{40}$$

Here, since the vacancy does not move on its own, but because of the movement of the particles in the system, it is reasonable to suppose that its effective hopping rate is the same as the one ascribed to the particles (see equation (4)), i.e. $J_v = J_m$. Our final expression for the dynamical correlation factor in the limit of high concentration of background particles is then given by equation (16) with $\cos\theta_0$ replaced by $\cos\theta_1$ given by equation (40).

3. Simulations

In section 4 of the previous paper [1] we discussed the relationship between the transition probability per unit time ($W_{\alpha\beta}$) of the continuous-time master equation and the transition matrix element ($A_{\alpha\beta}$) for the discrete-time Poisson process. In order to define the simulation procedure we can rewrite the matrix elements of the previous paper [1] as follows:

$$A_{\alpha\beta} = \begin{cases} \dfrac{J_B}{J_A}\dfrac{1}{N_pZ} & \text{if the transition occurs 'through' } J_B \\ \dfrac{1}{N_pZ} & \text{if the transition occurs 'through' } J_A \end{cases} \tag{41}$$

and

$$A_{\alpha\alpha} = 1 - \sum_{\beta \neq \alpha} A_{\beta\alpha} \tag{42}$$

where N_p is the total number of particles and $N_p Z J_A$ stands for the maximum rate of hopping out of the possible configurations.

The simulations were performed in square lattices of $N_s \times N_s$ sites ($N_s = 100$) using periodic boundary conditions. A randomly chosen fraction x of the sites were labelled as type B, while the rest of them were labelled as type A. Then $N_p = cN_s^2$ labelled particles were randomly distributed on the lattice, where each site was assigned a label to indicate whether it is empty or not. In the innermost simulation loop a particle and one of its nearest-neighbour sites are selected randomly. If the nearest-neighbour site is already occupied, the configuration of the system is kept unchanged, but if it is empty, the site occupied by the selected particle is identified. If this site is of type A, the particle is moved to the nearest-neighbour site. On the other hand if it is a type B site, a random number between zero and one is generated and the move is made if the random number is less than J_B/J_A, otherwise the configuration is kept unchanged. Note that we assumed $J_A > J_B$, in particular we set $J_A = 2J_B$ throughout the simulations. This procedure reproduces correctly the probabilities given by equations (41) and (42).

In these simulations the time was measured by Monte Carlo steps per particle (MCS/p), with the unit now being $1/J_A Z$. Like in the previous paper [1] the mean square displacement of a given particle has been studied using the lattice repetition scheme for enabling particle movement up to arbitrarily large displacement. Also the time history of any one of the particles in the system was considered as a tracer particle history. The tracer mean square displacement $\langle R^2 \rangle$ and quantity $\langle R^4 \rangle$ were calculated at each MCS/p by averaging them over all particles. The diffusion coefficient was obtained from the $\langle R^2 \rangle$ data with a weighted linear least-squares fitting, using $\langle R^4 \rangle - \langle R^2 \rangle^2$ as weighting factors, as described in detail in the previous paper [1]. The average occupation of type A sites, $\langle n_A \rangle$, and of type B sites, $\langle n_B \rangle$, were also calculated at each time step. In order to calculate the effective blocking factor, the number of successful hops of each particle was recorded. At each MCS/p, the average number of successful hops was obtained by averaging over all of the particles.

In terms of the simulation time span, averaging over tracer particle histories and averaging over disorder the simulation procedure followed that of the previous paper [1]. We therefore expect the error estimates to be equivalent with those given there. Although we did not check for the finite-size effects, we note that similar simulations carried out using comparable lattice sizes suggest that finite-size corrections are of the same order of magnitude as the statistical fluctuations [1, 12].

To check the model for high concentrations, we also performed a simulation to calculate directly the values of $\langle \cos\theta_1 \rangle$, $(\langle \cos\theta_1 \rangle)_A$ and $(\langle \cos\theta_1 \rangle)_B$. The procedure and its justification are quite simple. Since the maximum rate of transition out of the possible configurations in the limit of only one vacancy is $4J_A$, we can perform a simulation which makes use of this value for the total rate of the Poisson process. A procedure which reproduces these transition probabilities is as follows. Select at random a particle neighbouring the vacancy. If the site occupied by the selected particle is of type A, the particle is moved to the vacancy position. If the particle is occupying a type B site, a random number between zero and one is generated and tested against the value of J_B/J_A. If the random number is less than J_B/J_A, the particle is moved to the vacancy position. Otherwise the system configuration is kept unchanged. We calculate $\langle \cos\theta_1 \rangle$ using its definition, i.e., giving a value -1 for two successive steps of a particle in opposite directions and 1 for two successive steps in the

same direction. The final value is obtained by averaging over the history of each particle and then averaging over all particles. $(\langle\cos\theta_1\rangle)_A$ and $(\langle\cos\theta_1\rangle)_B$ were calculated in the same way. These simulations were also performed in a lattice with $N_s = 100$, for a total of 2×10^7 vacancy steps and 10 ensembles over the disorder.

Table 1. Simulation results for the effective blocking factor as a function of the concentration of type B sites (x) and the concentration of background particles (c).

	Concentration of type B sites (x)								
c	0.10	0.20	0.30	0.40	0.50	0.60	0.70	0.80	0.90
0.05	0.8669	0.7961	0.7354	0.6831	0.6371	0.5968	0.5610	0.5293	0.5007
0.10	0.8240	0.7582	0.7010	0.6509	0.6068	0.5680	0.5335	0.5024	0.4749
0.20	0.7370	0.6807	0.6305	0.5860	0.5458	0.5102	0.4781	0.4493	0.4234
0.30	0.6485	0.6012	0.5581	0.5190	0.4833	0.4510	0.4220	0.3956	0.3717
0.40	0.5586	0.5198	0.4836	0.4501	0.4192	0.3909	0.3651	0.3413	0.3198
0.50	0.4675	0.4365	0.4072	0.3796	0.3536	0.3294	0.3071	0.2865	0.2675
0.60	0.3756	0.3516	0.3290	0.3071	0.2863	0.2666	0.2483	0.2311	0.2148
0.70	0.2825	0.2556	0.2489	0.2328	0.2174	0.2024	0.1882	0.1746	0.1620
0.80	0.1889	0.1781	0.1674	0.1570	0.1466	0.1367	0.1269	0.1175	0.1086
0.90	0.0948	0.0895	0.0844	0.0793	0.0741	0.0692	0.0642	0.0594	0.0546
0.95	0.0475	0.0449	0.0424	0.0398	0.0373	0.0348	0.0323	0.0298	0.0274

Table 2. Simulation results for the correlation factor computed through equation (30) using the simulation data for f_b and D. The results are displayed as a function of the concentration of type B sites (x) and the concentration of background particles (c).

	Concentration of type B sites (x)								
c	0.10	0.20	0.30	0.40	0.50	0.60	0.70	0.80	0.90
0.05	0.97	0.97	0.97	0.97	0.97	0.97	0.97	0.97	0.98
0.10	0.93	0.94	0.94	0.95	0.94	0.94	0.94	0.95	0.95
0.20	0.89	0.88	0.88	0.89	0.88	0.89	0.88	0.89	0.89
0.30	0.82	0.82	0.81	0.82	0.81	0.82	0.82	0.82	0.82
0.40	0.76	0.75	0.75	0.76	0.76	0.75	0.75	0.76	0.77
0.50	0.71	0.70	0.70	0.68	0.68	0.69	0.69	0.70	0.71
0.60	0.65	0.64	0.63	0.63	0.63	0.63	0.64	0.64	0.65
0.70	0.59	0.58	0.58	0.57	0.57	0.57	0.57	0.58	0.59
0.80	0.54	0.53	0.52	0.52	0.51	0.52	0.52	0.53	0.54
0.90	0.49	0.48	0.47	0.47	0.46	0.46	0.46	0.48	0.48
0.95	0.47	0.46	0.45	0.44	0.43	0.44	0.44	0.45	0.47

4. Results

Since the relation between the numbers of successful and attempted hops is linear with time for all times, we took as the effective blocking factor f_b the result obtained at $t = 2000$ MCS/p. The average occupations of type A and type B sites were obtained by averaging over the results of the last 50 MCS/p. Tables 1 and 2 display the results for f_b and the correlation factor f_c, respectively.

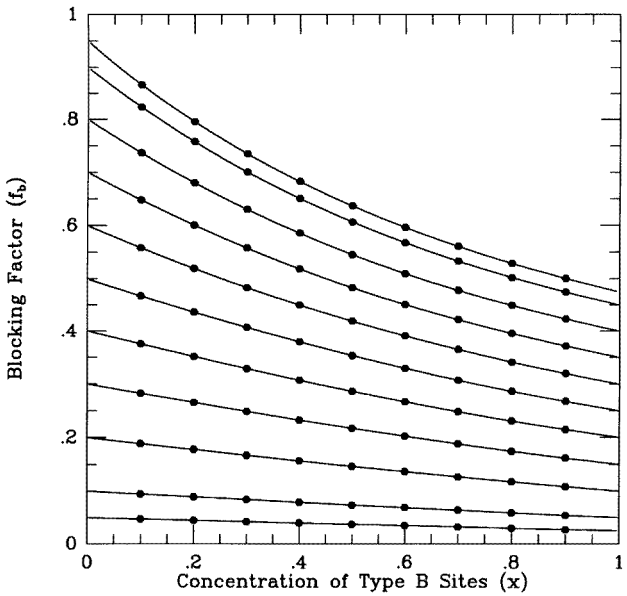

Figure 2. Comparison between theoretical (curves) and simulation results for the effective blocking factor. The curves are arranged from top to bottom by the concentration of background particles in the same order as is given in table 1.

On comparing the predictions of equations (22) and (23) for $\langle n_A \rangle$ and $\langle n_B \rangle$ with the simulation results we find almost exact agreement for the entire range of values of x. Figure 2 displays the theoretical and simulation results for the effective blocking factor. The agreement is remarkably good. One of the reasons for this good agreement is that the relation between the numbers of successful and attempted hops is linear for all times. A comparison between the correlation factor of TKE theory given by equation (16) and the simulation results show that, from low to intermediate concentrations c, the agreement is quite good. For values of c in the range $c \geqslant 0.50$, the simulation results begin to show an increasing variation as a function of the relative concentration of traps. This can be best appreciated in figure 3 and is an indication that towards higher concentrations of background particles there is an increasing influence of the disorder on the motion of the background particles. This influence is sufficiently strong to cause alterations in the dynamical correlation factor. We note that the largest deviation from the predicted values occurs at $x \simeq 0.50$ when the disorder is greatest. These deviations in the limit of high concentrations of background particles are in accordance with our expectations as discussed at the end of section 2. Figure 4 displays a comparison between theoretical and simulation results for the diffusion coefficient. Despite the discrepancies in the correlation factor for intermediate to high concentrations of background particles, we note that a model incorporating the correct effective blocking factor gives a fair account of the simulation results. In order to check the qualitative model for high concentrations of background particles developed at the end of section 2, we display in figure 5 the computed values of $\langle \cos\theta_1 \rangle$, $(\langle \cos\theta_1 \rangle)_A$ and $(\langle \cos\theta_1 \rangle)_B$ in conjunction with the values predicted by equations (32), (33) and (40). The general agreement between theory and simulation is again quite good, and gives the expected limit at $x = 0$. The maximum deviation from the perfect-lattice value occurs at $x \simeq 0.50$ when the amount of disorder is maximal. In figure 6 we

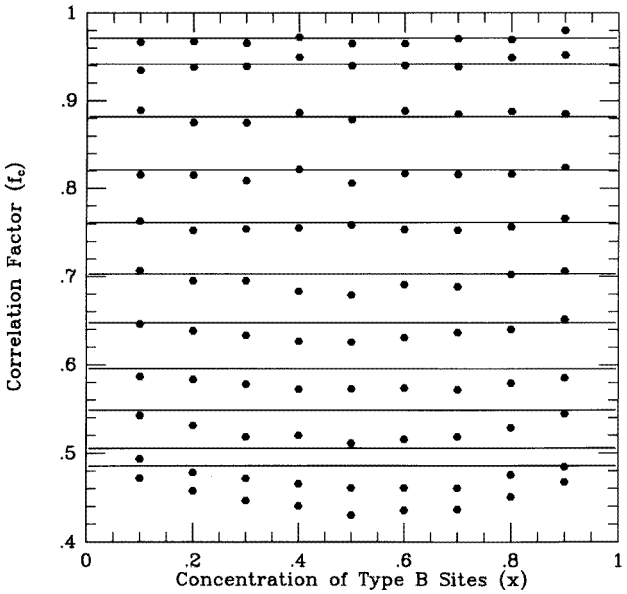

Figure 3. Comparison between theoretical and simulation results for the correlation factor. The theoretical results (curves) were calculated for $\cos\theta_1 = \cos\theta_0$. The results are expressed as a function of the concentration of type B sites. The curves are arranged from top to bottom by the concentration of background particles in the same order as is given in table 1.

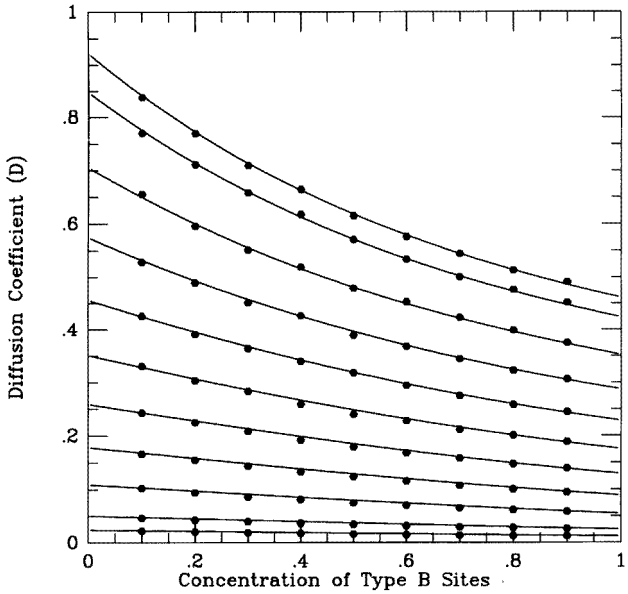

Figure 4. Comparison between theoretical (curves) and simulation results for the diffusion coefficient. The theoretical results are for $\cos\theta_1 = \cos\theta_0$. The results are shown as a function of the concentration of type B sites. The curves are arranged from top to bottom by the concentration of background particles in the same order as is given in table 1.

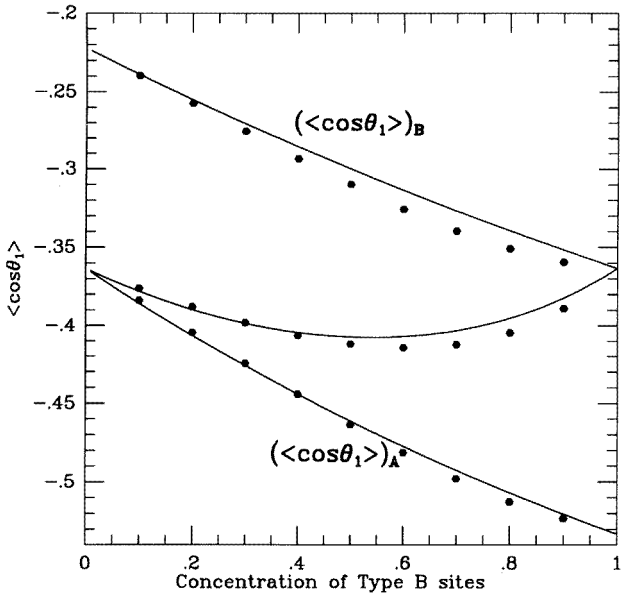

Figure 5. Comparison between theoretical (curves) and simulation results for the average cosine between two successive displacements of the tagged particle.

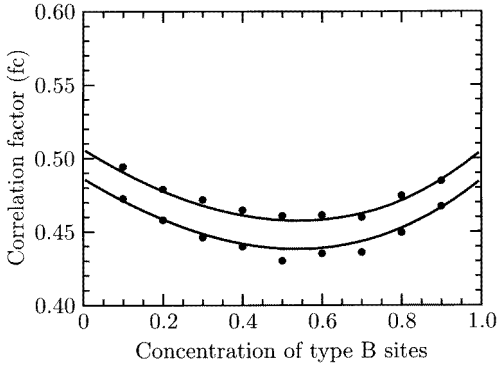

Figure 6. Comparison between theoretical results (curves) obtained using $\cos\theta_1$ from equation (40) and simulation results (points) for the correlation factor. The upper curve is for $c = 0.90$, while the lower curve corresponds to $c = 0.95$. The theoretical results obtained using $\cos\theta_0$ are horizontal lines as shown in figure 4.

compare the theoretical result for f_0 with the simulation results in the limit of large c. It is seen that the model with corrections to $\cos\theta_1$ greatly improves the description of the high-concentration limit.

5. Summary

A qualitative model for explaining the diffusive behaviour of a tagged particle in a system with two types of trap randomly distributed in a lattice has been proposed. The model

relies on the concepts of blocking and correlation and is readily generalized to the situation in which there are several different types of trap. It has been shown that the direct interaction of the tagged particle with the traps does not introduce correlations in the motion of the former. This interaction, however, in combination with the interaction between tagged and background particles, is responsible for a blocking effect which gives the dominant contribution to the structure of the diffusion coefficient. From low to intermediate concentrations of background particles, the correlation factor is well reproduced by the dynamical correlation factor corresponding to a system of particles in a perfect lattice. For high concentrations of background particles the traps considerably affect the movement of the background particles inducing observable changes in the dynamical correlation factor. Such changes can be thought of as being a consequence of the fact that the traps induce changes in the relative velocity between tagged and background particles. A simple model, based on the random walk of a vacancy in a disordered lattice, accounts well for the observed behaviour of the dynamical correlation factor.

Acknowledgments

LFP thanks the Brazilian Research Council (CNPq) and the Brazilian Space Research Institute (INPE) for financial support. KK gratefully acknowledges the hospitality of the Department of Physics in Oxford where this work was begun.

References

[1] Perondi L F, Elliott R J and Kaski K 1997 *J. Phys.: Condens. Matter* **9** 7933
[2] Balkanski M and Massot M 1989 *Disorder in Condensed Matter Physics* ed J A Blackman and J Tagueña (Oxford: Oxford University Press) p 74
[3] Mak C H, Andersen H C and George S M 1988 *J. Chem. Phys.* **88** 4052
[4] Kehr J W and Paetzold O 1992 *Physica* A **190** 1
[5] Wichmann T and Kehr K W 1995 *J. Phys.: Condens. Matter* **7** 717
[6] Brouwer R C, Salomons E and Griessen R 1988 *Phys. Rev.* B **38** 10 217
[7] Salomons E 1988 *J. Phys. C: Solid State Phys.* **21** 5953
[8] Haus J W, Kehr K W and Lyklema J W 1983 *Phys. Rev.* B **25** 2095
[9] Haus J W and Kehr K W 1987 *Phys. Rep.* **150** 265
[10] Spohn H 1983 *J. Phys. A: Math. Gen.* **16** 4275
[11] Tahir-Kheli R A and Elliott R J 1983 *Phys. Rev.* B **27** 844
[12] Tahir-Kheli R A and El-Meshad N 1985 *Phys. Rev.* B **32** 6166