Theory of atomic diffusion in cubic crystals with impurities based on the correlated-walks theory

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A walker is allowed to move on the simple cubic lattice with the following rules: If it should arrive at any site, it may move in the same direction as that of the previous step with probability α, turn at right angles with probability γ, reverse with probability β, or remain with probability σ, normalized such that $\alpha + 4\gamma + \beta + \sigma = 1$. If it were at rest, it may move in any direction with the same probability μ or remain with probability σ', normalized such that $6\mu + \sigma' = 1$. An exact expression for the mean-square displacement $\langle r^2 \rangle$ after N units of time is derived. From this expression, the diffusion coefficient D is obtained as follows: $D = (1/6)(1+\alpha-\beta)(1-\alpha+\beta)^{-1} \times [1+\sigma/(1-\sigma')]^{-1} a_0^2 \tau^{-1}$, where a_0 is the step length and τ the unit of time. Similar results are obtained for the face-centered and body-centered cubic lattices. These results are used to discuss the atomic diffusion in cubic crystals with impurities, which act as traps. Comparison with previous experimental and theoretical results is made and discussed.

I. INTRODUCTION

Recent experiments using neutron scattering[1] and other techniques[2] showed that hydrogen atoms diffusing in a metallic crystal like niobium are influenced strongly by interstitial impurities such as nitrogen atoms, which act as traps. The effective diffusion coefficient D for such a system can be several times smaller than the diffusion coefficient D_0 for pure metal. Fedders[3] developed a correlation-function theory of atomic diffusion for a crystal with a random distribution of deep traps and obtained the relation

$$D = D_0(1 + c\tau_0\tau_1^{-1})^{-1}, \tag{1.1}$$

where c is the concentration of impurities and τ_1 and τ_0 are the average-jump and trap time, respectively. In a typical experiment analysis,[1] the trap time τ_0 is estimated to be several hundred times greater than the jump time τ_1, and the concentration c ranges between 0.004 and 0.007. Fedder's result is in qualitative agreement with the experiments.

The purpose of this paper is to present a theory of atomic diffusion on the basis of correlated walks.[4,5] Our model for a simple cubic (sc) lattice is set up as follows. A walker (model atom) is allowed to jump between nearest-neighbor lattice sites per unit time τ, or to stay at the same site with the following rules: If it should arrive at any site, it may move in the same direction as that of the previous step with probability α, turn at right angles with probability γ, reverse with probability β, or remain at that site with probability σ [see Fig. 1(a)]. The step probabilities are normalized such that

$$\alpha + 4\gamma + \beta + \sigma = 1, \tag{1.2a}$$

where the numerical factor 4 corresponds to the number of possibilities of turning at right angles. If the walker were at rest, it might move in any direction with the same probability μ or remain with probability σ' with the normalization

$$6\mu + \sigma' = 1. \tag{1.2b}$$

[See Fig. 1(b).]

The connection between the present model and the atomic diffusion is rather obvious. For definiteness let us assume that the atoms migrate on interstitial sites, which also form an sc lattice

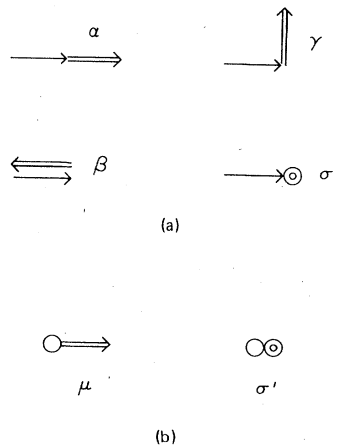

FIG. 1. The probabilities of possible moves with correlation for the sc lattice. They are normalized such that $\alpha + 4\gamma + \beta + \sigma = 1$ and $6\mu + \sigma' = 1$.

with the same lattice constant a_0. This lattice can be used as the reference lattice in the model. If the concentration of diffusing atoms is small, atoms should move independently of each other. This allows us to describe the diffusion in terms of a *single particle* migrating on the lattice. Let us first consider a perfect lattice without impurities. The jump rate should be the same everywhere on the lattice, and the average jump time τ_1 from one site to the next will be taken to be the unit time τ in the model. In order to jump, the atom must overcome a certain energy barrier which is formed by the surrounding lattice molecules. Once the atom moves to a new site, it faces another barrier at that site. Since the atom carries momentum, the jump probabilities will be *correlated*. Once the atom starts to move, it may jump over several sites in succession. Colloquially speaking, the atom acquires a "momentum," and this carries it over several sites. Such correlated motion can, to some degree, be described in terms of the step probabilities α, β, and γ. If, on the other hand, the atom were stationary, it might move in any direction without preference; this possibility is represented by the start-to-move probability μ. The atom may also remain at the same site, and it may come to a stop temporarily. To account for these possibilities we use the probabilities σ' and σ.

In the main body of the paper we will derive an exact expression for the mean-square displacement $\langle r^2\rangle$ of the walker after N units of time. The result is given by (3.9). By using the known relation[6] between the mean-square displacement and the diffusion coefficient D, we obtain

$$D=\frac{1+\alpha-\beta}{6(1-\alpha+\beta)[1+\sigma/(1-\sigma')]}\,\frac{a_0^2}{\tau}, \tag{1.3}$$

where a_0 is the nearest-neighbor distance, which is equal to the lattice constant in this case.

Our theory can be extended simply to other cubic lattices, face-centered cubic (fcc), and body-centered cubic (bcc) lattices. The results for the diffusion coefficient are similar to (1.3).

So far we have considered a perfect lattice with no impurities. Let us now assume that interstitial impurities are introduced and distributed uniformly over the lattice with a concentration c. These impurities in general should cause the change in jump probabilities not only at the particular interstitial sites where they are located, but also at the surrounding sites. But the most significant change should occur in the come-to-stop probability σ and the stay-stationary probability σ', both at the very same impurity sites. Let (σ_0, σ_0') $[(\sigma_1, \sigma_1')]$ be the probabilities when the interstitial site is occupied [unoccupied] by impurity. The probability of the occupation is equal to the concentration c. The moving probabilities (α, β, γ) will be chosen to be the same as before. (This assumption will be reasonable if the concentration c of impurities is low.) We can then obtain the following expression for the effective diffusion coefficient:

$$D=D_0\Big/\left(1-c+c\,\frac{1+\sigma_0/(1-\sigma_0')}{1+\sigma_1/(1-\sigma_1')}\right), \tag{1.4}$$

where D_0 represents the diffusion coefficient for a perfect crystal, that is, expression (1.3) with $(\sigma, \sigma')=(\sigma_1, \sigma_1')$. In the limit of low concentration ($c \ll 1$), our expression (1.4) agrees with the result of Fedders (1.1). A more detailed discussion of our results is given in Sec. V.

In Sec. II correlated walks on an auxiliary lattice are formulated and solved by means of generating-function techniques. The solutions are used in Secs. III and IV to obtain the mean-square displacements $\langle r^2\rangle$ for the sc, fcc, and bcc lattices.

II. CORRELATED WALKS ON AN AUXILIARY LATTICE

Let us consider a simple orthogonal lattice in four dimensions. The object (walker) starts at the origin and moves one step per unit time along the *positive* X, Y, Z, or U axes. The four *directions* of steps will be designated by 1, 2, 3, and 4, respectively. If the last step has the direction a, the probabilities of stepping along the positive X, Y, Z, and U axes will be denoted by p_a, q_a, r_a, and s_a, respectively, with the normalization

$$p_a+q_a+r_a+s_a=1\,. \tag{2.1}$$

After N units of time, the object will arrive at a site whose coordinates (X, Y, Z, U) satisfy

$$X+Y+Z+U=N\,. \tag{2.2}$$

The site of arrival may therefore be specified by (X, Y, Z, N). Let the probability of the object arriving at (X, Y, Z, N) with direction a be $P_a(X, Y, Z, N)$. Consideration of two successive steps yields the following relations for P_a:

$$\begin{aligned}
P_1(X,Y,Z,N)&=\sum_{a=1}^{4} p_a P_a(X-1,Y,Z,N-1)\,,\\
P_2(X,Y,Z,N)&=\sum_{a} q_a P_a(X,Y-1,Z,N-1)\,,\\
P_3(X,Y,Z,N)&=\sum_{a} r_a P_a(X,Y,Z-1,N-1)\,,\\
P_4(X,Y,Z,N)&=\sum_{a} s_a P_a(X,Y,Z,N-1)\,,
\end{aligned} \tag{2.3}$$

where all arguments for P's are non-negative.

This set of difference equations may be solved subject to a given initial condition. We will assume that the object arrived at the origin with direction 4, which may be expressed by

$$P_a(X, Y, Z, 0) = \delta_{X,0}\delta_{Y,0}\delta_{Z,0}\delta_{a,4}, \tag{2.4}$$

where the symbol δ denotes the Kronecker delta.

From the dynamics of the walker, it is clear that

$$P_a(X, Y, Z, N) = 0 \quad \text{if} \quad X + Y + Z > N. \tag{2.5}$$

Let us introduce *generating functions*:

$$\psi(\xi, \eta, \zeta, \nu) \equiv \sum_{N=0}^{\infty} \sum_{X}^{X+Y+Z\le N} \sum_{Y} \sum_{Z} \xi^X \eta^Y \zeta^Z \nu^N P_a(X, Y, Z, N)$$
$$= \sum_{N=0}^{\infty} \sum_{X=0}^{\infty} \sum_{Y=0}^{\infty} \sum_{Z=0}^{\infty} \xi^X \eta^Y \zeta^Z \nu^N P_a(X, Y, Z, N), \tag{2.6}$$

where the last member was obtained with the aid of (2.5). Multiplying Eqs. (2.3) by $\xi^X \eta^Y \zeta^Z \nu^N$, summing with respect to X, Y, Z, and N, and using (2.4) and (2.6), we obtain

$$\begin{pmatrix} 1-p_1\xi\nu & -p_2\xi\nu & -p_3\xi\nu & -p_4\xi\nu \\ -q_1\eta\nu & 1-q_2\eta\nu & -q_3\eta\nu & -q_4\eta\nu \\ -r_1\zeta\nu & -r_2\zeta\nu & 1-r_3\zeta\nu & -r_4\zeta\nu \\ -s_1\nu & -s_2\nu & -s_3\nu & 1-s_4\nu \end{pmatrix} \begin{pmatrix} \psi_1 \\ \psi_2 \\ \psi_3 \\ \psi_4 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ 0 \\ 1 \end{pmatrix}. \tag{2.7}$$

These equations can be solved in an elementary manner, yielding

$$\psi_a = \frac{(t)_{4,a}}{|T|}, \tag{2.8}$$

where $T \equiv (T_{ij})$ represents the 4×4 matrix appearing in (2.7), $|T|$ is its determinant, and $(t)_{4,a}$ is the cofactor of the element $T_{4,a}$.

The probability that the walker arrives at (X, Y, Z, N) from any direction is given by

$$\sum_a P_a(X, Y, Z, N) \equiv P(X, Y, Z, N). \tag{2.9}$$

The generating function of this quantity is provided by

$$\sum_a \psi_a(\xi, \eta, \zeta, \nu) \equiv \psi(\xi, \eta, \zeta, \nu). \tag{2.10}$$

The theory and results presented here will be used in the following two sections.

III. CORRELATED WALKS ON THE sc LATTICE

Let us consider an sc lattice. A walker is allowed to jump between nearest-neighbor sites per unit time τ or to stay at the same site with the rules given in the Introduction. We assume that the walker was stationary at the origin O up to the initial time $N=0$. We wish to find the mean square of the displacement r of the walker after N units of time:

$$\langle r^2 \rangle \equiv \langle x^2 + y^2 + z^2 \rangle$$
$$= \langle x^2 \rangle + \langle y^2 \rangle + \langle z^2 \rangle. \tag{3.1}$$

Since the elements of dynamics given by step probabilities and the initial condition are both symmetric with respect to all directions, the three averages on the right-hand side must have the same value. We therefore have

$$\langle r^2 \rangle = 3\langle x^2 \rangle. \tag{3.2}$$

The average $\langle x^2 \rangle$ can be calculated by considering the *projected motion* of the walker along the x axis. The walker may appear to move right, left, or to be stationary; the last alternative arises either from the motion in the y and z directions, or from no real motion. The projection may now be represented in terms of the motion of the object on the auxiliary four-dimensional lattice introduced in Sec. II with the correspondence

$$\begin{aligned} &\text{right step} \leftrightarrow \text{positive step in } X, \\ &\text{left step} \leftrightarrow \text{positive step in } Y, \\ &\text{stationary (sideways move)} \leftrightarrow \text{positive step in } Z, \\ &\text{stationary (no move)} \leftrightarrow \text{positive step in } U, \end{aligned} \tag{3.3}$$

and

$$\begin{pmatrix} p_1 & q_1 & r_1 & s_1 \\ p_2 & q_2 & r_2 & s_2 \\ p_3 & q_3 & r_3 & s_3 \\ p_4 & q_4 & r_4 & s_4 \end{pmatrix} = \begin{pmatrix} \alpha & \beta & 4\gamma & \sigma \\ \beta & \alpha & 4\gamma & \sigma \\ \gamma & \gamma & \alpha+2\gamma+\beta & \sigma \\ \mu & \mu & 4\mu & \sigma' \end{pmatrix}. \tag{3.4}$$

The displacement x on the sc lattice corresponds to the difference $X - Y$ on the auxiliary lattice. We therefore have

$$\langle x^2 \rangle / a_0^2 = \langle (X-Y)^2 \rangle = \langle X^2 \rangle + \langle Y^2 \rangle - 2\langle XY \rangle, \tag{3.5}$$

where a_0 represents the step length, that is, the nearest-neighbor distance.

The average $\langle X^2 \rangle$ depends on the time $N\tau$. From (2.6), (2.9), and (2.10), this quantity can be obtained from

$$\frac{\partial}{\partial \xi} \xi \frac{\partial}{\partial \xi} \psi(\xi, \eta, \zeta, \nu)\Big|_{\xi=\eta=\zeta=1} = \sum_{N=0}^{\infty} \nu^N \langle X^2 \rangle_N. \tag{3.6}$$

In a similar manner, the averages $\langle Y^2 \rangle$ and $\langle XY \rangle$ can be computed. It then follows that $\langle r^2 \rangle = 3\langle x^2 \rangle$ can be calculated from

$$G \equiv 3\left(\frac{\partial}{\partial \xi}\xi\frac{\partial}{\partial \xi}+\frac{\partial}{\partial \eta}\eta\frac{\partial}{\partial \eta}-2\frac{\partial^2}{\partial \xi \partial \eta}\right)\psi(\xi,\eta,\zeta,\nu)\Big|_{\xi=\eta=\zeta=1} = \sum_{N=0}^{\infty}\nu^N\langle r^2\rangle_N . \tag{3.7}$$

After straightforward calculations using (2.7), (2.8), and (2.10), we obtain

$$G=\frac{(1-\sigma')[1+(\alpha-\beta)\nu]\nu}{(1-\nu)^2[1-(\alpha-\beta)\nu][1-(\sigma'-\sigma)\nu]} . \tag{3.8}$$

After further extensive calculations, we obtain

$$\begin{aligned}\langle r^2\rangle/a_0^2 = {} & \frac{(1-\sigma')(1+\alpha-\beta)}{(1-\alpha+\beta)(1-\sigma'+\sigma)}N-(1-\sigma')(1-\alpha+\beta)^{-2}(1-\sigma'+\sigma)^{-2}(\alpha-\beta-\sigma'+\sigma)^{-1}\\ & \times\{2(\alpha-\beta)(1-\sigma'+\sigma)[\alpha-\beta-\sigma'+\sigma-(1-\sigma'+\sigma)(\alpha-\beta)^{N+1}]\\ & +(\sigma'-\sigma)(1-\alpha+\beta)[(1+\alpha-\beta)(\alpha-\beta-\sigma'+\sigma)\\ & +(1-\alpha+\beta)(\alpha-\beta+\sigma'-\sigma)(\sigma'-\sigma)^N]\} .\end{aligned} \tag{3.9}$$

The significance of this result will be discussed in Sec. V.

IV. CORRELATED WALKS ON fcc AND bcc LATTICES

A. fcc lattice

Correlated walks on the fcc lattice can be set up, and the mean-square displacement can be calculated in a similar manner. A walker is allowed to move on the lattice with the following rules: If it should arrive at any site, it may move in the same direction as that of the previous step with probability α_f (see Fig. 2), turn at 60° with probability δ_f (there are four possibilities), turn at 90° with probability γ_f (two possibilities), turn at 120° with probability ε_f (four possibilities), turn at 180° with probability β_f, or it may remain with probability σ. The step probabilities are normalized such that

$$\alpha_f+4\delta_f+2\gamma_f+4\varepsilon_f+\beta_f+\sigma=1 , \tag{4.1}$$

where various numerical factors correspond to the possibilities of turning at each angle.

If the walker should happen to be at rest, it may move out in any direction with the same probability μ_f or remain with probability σ', with the normalization

$$12\mu_f+\sigma'=0 . \tag{4.2}$$

The staying probabilities are denoted by σ and σ' without subscripts as they do not depend on the lattice type, by assumption. The walker starts at the origin O from rest as before.

Let us choose the Cartesian axes along the cubic axes. Then, the mean-square displacement can again be equal to three times the mean-square displacement in one direction:

$$\langle r^2\rangle = 3\langle x^2\rangle .$$

The average $\langle x^2\rangle$ can now be calculated by looking at the projection of the motion along the x axis, which can be represented in terms of the correlated walks on the four-dimensional lattice with the following correspondence:

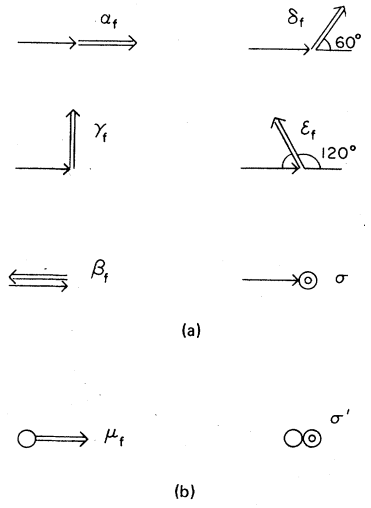

FIG. 2. The probabilities of possible moves for the fcc lattice. They are normalized such that $\alpha_f+4\delta_f+2\gamma_f+4\varepsilon_f+\beta_f+\sigma=1$ and $12\,\mu_f+\sigma'=1$.

$$\begin{pmatrix} p_1 & q_1 & r_1 & s_1 \\ p_2 & q_2 & r_2 & s_2 \\ p_3 & q_3 & r_3 & s_3 \\ p_4 & q_4 & r_4 & s_4 \end{pmatrix} = \begin{pmatrix} \alpha_f+2\delta_f+\gamma_f & \beta_f+\gamma_f+2\varepsilon_f & 2\delta_f+2\varepsilon_f & \sigma \\ \beta_f+2\varepsilon_f+\gamma_f & \alpha_f+2\delta_f+\gamma_f & 2\delta_f+2\varepsilon_f & \sigma \\ 2\delta_f+2\varepsilon_f & 2\delta_f+2\varepsilon_f & \alpha_f+2\gamma_f+\beta_f & \sigma \\ 4\mu_f & 4\mu_f & 4\mu_f & \sigma' \end{pmatrix} . \tag{4.3}$$

The projection reduces the step length by the factor $\frac{1}{2}$. We may now compute $\langle x^2\rangle$ in the same manner as before. The results obtained for $\langle r^2\rangle$ are found to be expressed as in the form of (3.9) with the following substitutions:

$$\begin{aligned} \alpha &= \alpha_f + 2\delta_f + \gamma_f , \\ \beta &= \beta_f + 2\varepsilon_f + \gamma_f . \end{aligned} \tag{4.4}$$

B. bcc lattice

Correlated walks on the bcc lattice can be treated in a similar manner. Only main steps will be indicated below. The step probabilities with correlation are shown in Fig. 3. They are normalized such that

$$\begin{aligned} &\alpha_b + 3\delta_b + 3\varepsilon_b + \beta_b + \sigma = 1 , \\ &8\mu_b + \sigma' = 1 . \end{aligned} \tag{4.5}$$

The projected motion along one of the cubic axes will be right, left, or at rest. The last alternative can arise only from no real move. Thus, for this lattice, calculation of the average $\langle x^2\rangle$ is a little simpler and can be carried out in terms of the solutions of correlated walks on a three-dimensional auxiliary lattice. The resulting expression for $\langle r^2\rangle$ can again be written in the form of (3.9) with the following substitutions:

$$\begin{aligned} \alpha &= \alpha_b + 2\delta_b + \varepsilon_b , \\ \beta &= \beta_b + 2\varepsilon_b + \delta_b . \end{aligned} \tag{4.6}$$

In obtaining this result, we used the fact that the projection reduces the step length a_0 by the factor $1/\sqrt{3}$.

V. DISCUSSIONS

In the last two sections we have studied correlated walks on the three cubic lattices and obtained the mean-square displacement $\langle r^2\rangle$ after N units of time. The results are similar and can be expressed in a unified form:

$$\begin{aligned} \langle R^2\rangle/a_0^2 = \frac{(1-\sigma')(1+\Delta)}{(1-\sigma'+\sigma)(1-\Delta)} N &- (1-\sigma')(1-\sigma'+\sigma)^{-2}(1-\Delta)^{-2}(\Delta-\sigma'+\sigma)^{-1} \\ &\times\{2\Delta(1-\sigma'+\sigma)[\Delta-\sigma'+\sigma-(1-\sigma'+\sigma)\Delta^{N+1}] \\ &+(\sigma'-\sigma)(1-\Delta)[(1+\Delta)(\Delta-\sigma'+\sigma)+(1-\Delta)(\Delta+\sigma'-\sigma)(\sigma'-\sigma)^N]\} , \end{aligned} \tag{5.1}$$

$$\Delta = \begin{cases} \alpha-\beta & \text{for sc} \\ \alpha_f+2\delta_f-2\varepsilon_f-\beta_f & \text{for fcc} \\ \alpha_b+\delta_b-\varepsilon_b-\beta_b & \text{for bcc} , \end{cases} \tag{5.2}$$

where (3.9), (4.4), and (4.6) were used. Since all cases can be treated in a similar manner, we will discuss the case of the sc lattice for definiteness.

In the limit $\alpha\to 1$, which corresponds to the particle moving always in the same direction, the mean-square displacement $\langle r^2\rangle$ approaches $N^2a_0^2$, as it should. Except for this case, the first term on the right-hand side of (5.1) is proportional to N and dominates for large N.

It is known that the long time limit of the mean-square displacement $\langle r^2\rangle$ is connected with the diffusion coefficient by[6]

$$\langle r^2\rangle = 6DN\tau \quad \text{for large } N . \tag{5.3}$$

Thus, we find that except for the free-motion limit ($\alpha=1$) there *exists* a diffusion coefficient D, which is given by

$$D = \frac{(1-\sigma')(1+\Delta)}{6(1-\sigma'+\sigma)(1-\Delta)} \frac{a_0^2}{\tau} . \tag{5.4}$$

Using this result, we will now discuss the atomic diffusion.

A. Perfect crystals

Let us consider the case in which the particle moves on the lattice without stopping. In mathematical terms, $\sigma=\sigma'=0$. The diffusion coefficient D is given by

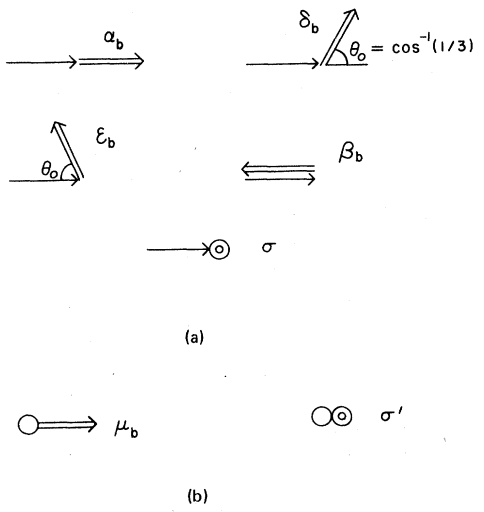

FIG. 3. The probabilities of possible moves for the bcc lattice. They are normalized such that $\alpha_b+3\delta_b+3\varepsilon_b+\beta_b+\sigma=1$ and $8\mu_b+\sigma'=1$.

$$D=\frac{1}{6}\frac{1+\Delta}{1-\Delta}\frac{a_0^2}{\tau}. \tag{5.5}$$

This case roughly simulates the case of a Lorentz gas in which the independently moving particles (electrons) are scattered elastically by static scatterers (impurities) distributed at random. In fact, expression (5.5) is in essential agreement with the result obtained based on the Boltzmann equation,[7] except for the fact that the directions of the motion are restricted in our model.[5]

Let us consider another extreme case where the particle is allowed to jump just one step at a time. This case is characterized by

$$\alpha=\beta=\gamma=0$$

From (5.4) we obtain

$$D=\frac{1}{6[1+\sigma/(1-\sigma')]}\frac{a_0^2}{\tau}. \tag{5.6}$$

This result is similar to that of the usual random walks[8]:

$$D_R=\frac{1}{6}\frac{a_0^2}{\tau}. \tag{5.7}$$

The difference arises from the fact that the particle may stay at the same site and restart from rest in our model. This allows the particle to leave the site immediately or a number of rest-times after arrival with probability weights, which accounts for the "correction" factor:

$$1+\sigma+\sigma\sigma'+\sigma\sigma'^2+\cdots=1+\sigma(1-\sigma')^{-1}. \tag{5.8}$$

The come-to-stop probability σ and the remain-stationary probability σ' should have different values for different crystals. Equation (5.8) therefore represents the effect of the difference in the "trap" time.

The case of actual atomic diffusion should fall between these two limits. The full expression (5.4) should yield a good description of the diffusion coefficient D.

B. Crystals with impurities

We have briefly discussed this case in the Introduction. If the impurities are distributed with concentration c and generate significant change in the come-to-stop (σ) and stay-stationary (σ') probabilities only, we may replace the correction factor (5.8) by

$$1+c\frac{\sigma_0}{1-\sigma_0'}+(1-c)\frac{\sigma_1}{1-\sigma_1'}. \tag{5.9}$$

The three terms here correspond to the following processes: (i) The particle arrives and leaves with no intermediate stop, (ii) it arrives at an impurity site, which occurs with the probability c, and eventually leaves, and (iii) it arrives at a regular site and leaves. Introducing (5.9) in (5.6) and rearranging terms, we obtain the expression (1.4) quoted earlier,

$$D=D_0\Big/\left(1-c+c\frac{1+\sigma_0/(1-\sigma_0')}{1+\sigma_1/(1-\sigma_1')}\right).$$

This new expression is different from Fedders' expression (1.1) only in the additional term $-c$ in the denominator. If $c\ll 1$, the difference is negligible. The validity of (1.4), however, should be considerably greater. In fact, with inclusion of the term $-c$, our expression yields a formally correct result even in the high-concentration limit $c=1$. This, however, may be deceptive because high concentrations of impurities will change the dynamics of hopping characterized by the step probabilities (α,β,γ).

The present theory can be extended simply to the case in which there exist impurities of several kinds. If the concentrations of different sites are denoted by $c_{\mathrm{I}}, c_{\mathrm{II}}, \ldots$, with the normalization

$$c_{\mathrm{I}}+c_{\mathrm{II}}+\cdots=1, \tag{5.10}$$

the correction factor should take the form

$$1+c_{\mathrm{I}}\frac{\sigma_{\mathrm{I}}}{1-\sigma_{\mathrm{I}}'}+c_{\mathrm{II}}\frac{\sigma_{\mathrm{II}}}{1-\sigma_{\mathrm{II}}'}+\cdots, \tag{5.11}$$

where σ_α and σ_α' denote, respectively, the come-to-stop and stay-stationary probabilities at the site of type α, $\alpha=\mathrm{I}, \mathrm{II}, \ldots$.

It is stressed that our expression (1.4) should be valid for any type of lattice. We also note that our main results (5.4) and (1.4) are obtained in an

elementary manner without sophistications associated with the correlation-function techniques.

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