The generalized atomic hopping problem—particle correlation functions*

O. F. Sankey and Peter A. Fedders
Department of Physics, Washington University, St. Louis, Missouri 63130
(Received 30 November 1976)

We extend the formalism of the preceding paper to include a description of the hopping of specific particles. The formalism is used to calculate frequency- and wave-vector-dependent two-particle correlation functions for specific particles on lattice sites in sc, bcc, and fcc Bravais lattices and on octahedral interstitial sites in a bcc lattice.

I. INTRODUCTION

In the preceding paper[1] we developed a formalism for calculating two-point occupancy correlation functions for the generalized hopping problem. The purpose of this paper is to extend that formalism to include calculations of the correlation functions for specific particles and to exhibit the use of the formalism by including calculations for a number of simple cases.

The rate equation for the occupancy of a site by one specific particle is

$$\left(\frac{dp_\alpha}{dt}\right)_0 = -\sum_\beta \Gamma_{\alpha\beta} p_\alpha (1-n_\beta) + \sum_\beta \Gamma_{\beta\alpha} p_\beta (1-n_\alpha) , \tag{1}$$

where Greek letters denote lattice sites, $\Gamma_{\alpha\beta}$ is the hopping rate for a particle at the site α to jump to a vacant site β, n_α denotes the occupancy of the site α ($n_\alpha = 1$ if the site α is occupied by any particle and $n_\alpha = 0$ if the site α is vacant), and p_α denotes the occupancy of the site α by the one specific particle ($p_\alpha = 1$ if the specific particle is at the site α and $p_\alpha = 0$ otherwise). As discussed in Paper I, the notation $(d/dt)_0$ does not indicate a true time derivative but rather the change in probability of the quantity operated on in a small time Δt, divided by Δt. Since the right-hand side of Eq. (1) is quadratic in occupancy variables, the problem is nontrivial even if $\Gamma_{\alpha\beta} = \Gamma_{\beta\alpha}$. That is, the equation cannot be iterated analytically in a simple fashion.

Various aspects of specific particle-correlation functions have been extensively studied for the special case where there is a vanishingly small concentration of vacancies in a substance with only one type of site ($\Gamma_{\alpha\beta} = \Gamma_{\beta\alpha}$). The effects of correlations on vacancy hopping was first pointed out by Bardeen and Herring[2] and correlation factors or effective diffusion coefficients for vacancy or impurity migration have been worked out for many different lattices.[3] Because vacancy migration plays an important role in a number of NMR relaxation phenomena, there have been a number of calculations of specific particle-correlation functions convoluted with spin dipolar interactions.[4] Recent work on this subject has tended towards computer simulations of the motion of a single vacancy. On the other hand, our formalism is not restricted to small concentration of vacancies and is not restricted to cases where $\Gamma_{\alpha\beta} = \Gamma_{\beta\alpha}$.

However, in this paper we discuss only the correlation functions for a single specific particle while most NMR applications require the correlation functions for a pair of specific particles. While we believe that our methods can be generalized to this more complex problem, we have not yet attempted to do so. On the other hand there are some interesting physical phenomena which depend only on single-particle correlation functions. One example is the diffusion coefficients measured in tracer or NMR experiments. Another example is the nuclear spin relaxation of interstitial deuterium in metals due to the fluctuating electric field gradients seen by the nuclei as they hop from site to site.[5]

In Sec. II we shall develop a diagrammatic representation of all of the moments of the single pair-correlation functions and discuss methods of obtaining the time-dependent correlation functions from these moments. The arguments of this section are simple generalizations of the arguments of Paper I. In Sec. III we obtain some results for the special case of $\Gamma_{\alpha\beta} = \Gamma_{\beta\alpha}$ for sc, bcc, and fcc Bravais lattices and for octahedral interstitial sites in a bcc lattice.

II. METHOD

The arguments in this section of the paper are very similar to the arguments in Paper I and thus many details will be omitted. Following Paper I it is convenient to use variables whose average values are zero and that are in some sense orthonormal. Since $\langle n_\alpha \rangle = c_\alpha$ and $\langle p_\alpha \rangle = c_\alpha/N$, where c_α is the concentration of particles at site α and N is the total number of particles in the system, the

desired variables are

$$\xi_\alpha = (n_\alpha - c_\alpha)/a_\alpha ,$$
$$a_\alpha = [c_\alpha(1-c_\alpha)]^{1/2} , \tag{2}$$
$$\rho_\alpha = (N/c_\alpha)^{1/2} p_\alpha ,$$

where terms of order $1/\sqrt{N}$ compared to one have been neglected. Equation (1) can be rewritten in terms of these new variables as

$$\left(\frac{d\rho_\alpha}{dt}\right)_0 = -\sum_\beta \bar{\omega}_{\alpha\beta}\rho_\beta - \sum_{\beta\gamma} \bar{\Omega}_{\alpha\beta\gamma}\rho_\beta \xi_\gamma , \tag{3}$$

where

$$\bar{\omega}_{\alpha\beta} = \bar{\omega}_{\beta\alpha} = \delta_{\alpha\beta}\sum_\gamma \Gamma_{\alpha\gamma}(1-c_\gamma) - \Gamma_{\beta\alpha}(1-c_\alpha)\left(\frac{c_\beta}{c_\alpha}\right)^{1/2} ,$$
$$\bar{\Omega}_{\alpha\beta\gamma} = \delta_{\alpha\gamma}\Gamma_{\beta\alpha}a_\alpha(c_\beta/c_\alpha)^{1/2} - \delta_{\alpha\beta}\Gamma_{\beta\gamma}a_\gamma . \tag{4}$$

In deriving these equations we have used the equilibrium condition Eq. (I-10). Further, the meaning of $(d/dt)_0$ is literally defined only in terms of the p's and n's.

By neglecting the last term on the right-hand side of Eq. (3) we obtain a linear equation in the ρ's or p's which can be iterated indefinitely without complications. This is the mean-field or effective-field approximation which is the best approximation that one can expect from a linearized theory. Essentially the approximation consists of replacing all n_β's in Eq. (1) by c_β. In the special case where $\Gamma_{\alpha\beta} = \Gamma_{\beta\alpha}$, there is only one type of site and thus only one concentration c. In this case, we have

$$\bar{\omega}_{\alpha\beta} = (1-c)\omega_{\alpha\beta} , \tag{5}$$

where $\omega_{\alpha\beta}$ is given by Eq. (I-8). Thus, in the effective-field theory, the particle correlation functions can be obtained from occupancy correlation function by substituting $(1-c)\omega_{\alpha\beta}$. In fact this approximation has been used in the literature.[6] We note, however, that if $\Gamma_{\alpha\beta} \neq \Gamma_{\beta\alpha}$, the frequency spectra and diffusion coefficients associated with the particle and occupancy correlation functions are not so simply related even in the mean-field approximation.

In order to go beyond the mean-field approximation we define two-point particle correlation functions and their moments as in Paper I. The correlation functions are defined as

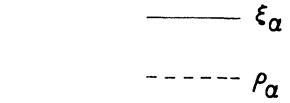

FIG. 1. Diagrammatic representations of the variables ρ_α and ξ_α.

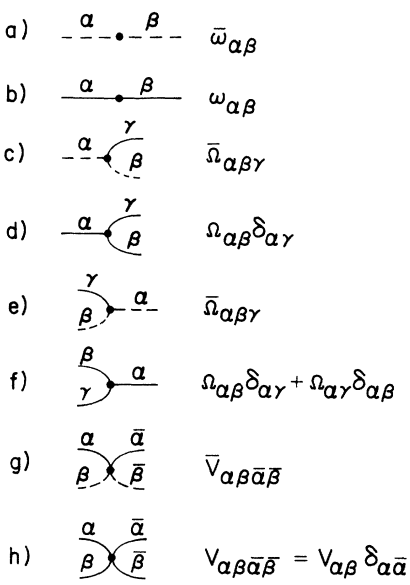

FIG. 2. Basic event diagrams and their corresponding analytical expressions in configuration space.

$$\bar{D}(\alpha,\beta;t) = \langle \rho_\alpha(t)\rho_\beta(0)\rangle\theta(t) , \tag{6}$$

where the angular brackets $\langle x\rangle$ denote the ensemble average of x, and θ is the step function. Since we expect exponential-like solutions, the moments of $\bar{D}$ are defined as

$$\bar{M}_n(\alpha,\beta) = \left(\frac{-d}{dt}\right)^n \bar{D}(\alpha,\beta;t)\Big|_{t=0} , \tag{7a}$$

$$\bar{D}(\alpha,\beta;t) = \sum_{n=0}^{\infty} \bar{M}_n(\alpha,\beta)\frac{(-t)^n}{n!}\theta(t) . \tag{7b}$$

Since all of the moments of the correlation functions are defined in the limit as $t \to 0$, one can relate the nth moment to the nth-order hopping probability

$$\bar{M}_n(\alpha,\beta) = \left\langle \left[\left(\frac{-d}{dt}\right)_0^n \rho_\alpha\right]\rho_\beta\right\rangle . \tag{8}$$

However since $(d/dt)_0^n\,\rho$ involves products of one ρ and up to n of the ξ's, we must be able to calculate d/dt_0 acting on products of ρ and arbitrary numbers of ξ's.

The derivation of the rules for calculating d/dt_0 on products of ρ's and ξ's and thus the derivation of the rules for calculating the moments of $\bar{D}$ is so similar to the analogous derivation in Paper I that it is superfluous to repeat it here. Again the rules for the moments are best described diagrammatically. As shown in Fig. 1, we associate solid lines with ξ_α and dashed lines with ρ_α. Further, consider the basic event diagrams displayed in Fig. 2 and the analytical expressions associated with them. The quantities $\omega_{\alpha\beta}$, $\Omega_{\alpha\beta}$,

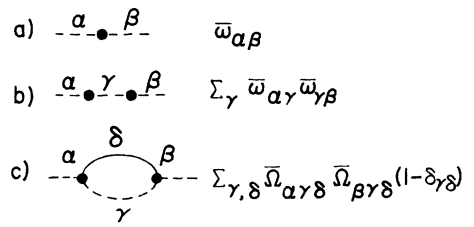

FIG. 3. Diagrammatic representations and analytical expressions for (a) the one diagram that contributes to $\bar{M}_1(\alpha,\beta)$ and (b) and (c), the two diagrams that contribute to $\bar{M}_2(\alpha,\beta)$.

and $V_{\alpha\beta}$ are defined in Eqs. (I-8), (I-9), and (I-22), respectively, and

$$\bar{V}_{\alpha\beta\bar{\alpha}\bar{\beta}} = -\delta_{\alpha\bar{\alpha}}\delta_{\beta\bar{\beta}}[\Gamma_{\alpha\beta}(1-c_\beta)+\Gamma_{\beta\alpha}(1+c_\beta-2c_\alpha)] - \delta_{\alpha\bar{\beta}}\delta_{\bar{\alpha}\beta}\Gamma_{\alpha\beta}c_\alpha(c_\alpha/c_\beta)^{1/2}\alpha_\beta/a_\alpha . \quad (9)$$

The rules for calculating $\bar{M}_n(\alpha,\beta)$ are as follows: From the set of basic event diagrams, (i) form all distinct connected diagrams with n events that start on the left with a single dashed line labeled α and end on the right with a single dashed line labeled β. These diagrams will be made up of n basic event diagrams and will contain n dots since each dot signifies one event. (ii) Label all internal lines with dummy site indices and associate the proper analytical expression with each diagram that is the product of the analytical expressions associated with each event. (iii) Sum over internal dummy site indices with the restriction that no two sites at the same "time" (or horizontal position) can be equal. In Fig. 3 we have displayed the only diagram that contributes to $\bar{M}_1(\alpha,\beta)$ and the two diagrams that contribute to $\bar{M}_2(\alpha,\beta)$ along with the appropriate analytical expressions. At this point essentially all of Sec. III of Paper I can be taken over and applied to the present problem since the results of that section depend only on the diagrammatic expansion of the moments. Thus we define a self-energy or memory function $\bar{K}(\alpha,\beta;t)$ with moments $\bar{L}_n(\alpha,\beta)$ and all equations in Sec. III of Paper I apply if we replace D, K, and L by $\bar{D}$, $\bar{K}$, and $\bar{L}$, respectively.

III. RESULTS

In Sec. III A and III B we use our formalism to calculate frequency-wave-vector-dependent particle-correlation functions for the special case where there is only one type of site and thus $\Gamma_{\alpha\beta} = \Gamma_{\beta\alpha}$. In particular we consider simple cubic (sc), bcc, and fcc Bravais lattices and octahedral interstitial sites in a bcc lattice. In Sec. III A we develop the necessary notation for these specific lattices. In Sec. III B we investigate methods of obtaining solutions for the specific particle-correlation functions.

A. Notation

As discussed in Sec. III of Paper I, it is much easier to work in reciprocal space or $\vec{q}$ space than in configuration space. For lattice structures with more than one site per unit cell we denote basis indices by Latin letters which go from 1 to b where b is the number of lattice sites in a unit cell. Then all functions $f(\alpha,\beta)$ can be transformed as

$$f(i,j;\vec{q}) = \sum_{\vec{l}} e^{-i\vec{q}\cdot\vec{R}_{\vec{l}}} f(\alpha,\beta) , \quad (10)$$

where α is the site i in the cell $\vec{l}$, β is the site j in the site $\vec{l}=0$, and $\vec{R}_{\vec{l}}$ is the lattice vector of the site $\vec{l}$. For the three Bravais lattices with a hopping rate Γ_0 to a single nearest neighbor, the transforms of $\Gamma_{\alpha\beta}$ for sc, bcc, and fcc lattices are, respectively,

$$\text{sc:}\quad \Gamma(\vec{q}) = 2\Gamma_0(\cos q_x a + \cos q_y a + \cos q_z a) , \quad (11a)$$

$$\text{bcc:}\quad \Gamma(\vec{q}) = 8\Gamma_0 \cos Q_x \cos Q_y \cos Q_z , \quad (11b)$$

$$\text{fcc:}\quad \Gamma(\vec{q}) = 4\Gamma_0(\cos Q_x \cos Q_y + \cos Q_y \cos Q_z + \cos Q_z \cos Q_x) \quad (11c)$$

where a is the length of a cube edge and the shorthand notation

$$Q_i = \tfrac{1}{2} q_i a \quad (12)$$

has been used for the bcc and fcc lattices. The three octahedral (oct) sites in a bcc unit cell can be chosen to lie at the points $(\frac{1}{2},0,0)a$, $(0,\frac{1}{2},0)a$, and $(0,0,\frac{1}{2})a$ with respect to the origin. With this choice, we get

$$\Gamma(i,i,\vec{q}) = 0 ,$$

$$\text{oct:}\quad \Gamma(x,y,\vec{q}) = \Gamma(y,x,\vec{q})^* = 2\Gamma_0 \cos Q_z\, e^{i(Q_x - Q_y)} , \quad (11d)$$

with cyclic variations in x, y, and z. In this case the basis indices $i = 1,2,3$ can be associated with the cartesian directions x,y,z, respectively.

It would be extremely convenient to express all of the moment diagrams in $\vec{q}$ space. The reason that this cannot be done trivially is because of the restrictions that no two sites at the same "time" or horizontal position can be equal. For the present we shall ignore this restriction and Fourier transform the diagrams. The error that this introduces will be discussed in Sec. III B. The whole diagrammatic expansion scheme can then be put into reciprocal space. In Fig. 4 the basic event diagrams and their corresponding analytical expressions in $\vec{q}$ space are displayed. Since we con-

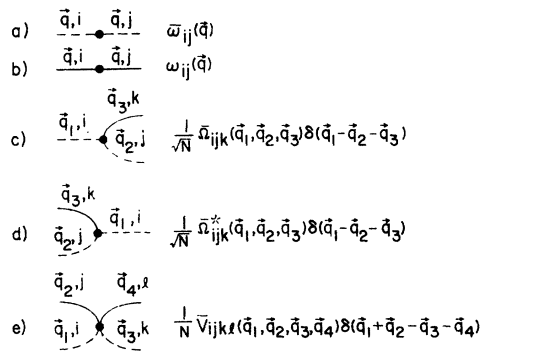

FIG. 4. Basic event diagrams and their corresponding analytical expressions in $\vec{q}$ space.

sider only the special case of $\Gamma_{\alpha\beta}=\Gamma_{\beta\alpha}$ in this section, $V_{\alpha\beta}$ and $\Omega_{\alpha\beta}$ are zero. The expressions in Fig. 4 are defined as

$$\begin{aligned}
&\omega_{ij}(\vec{q})=z\Gamma_0\delta_{ij}-\Gamma(i,j,\vec{q}),\\
&\bar{\omega}_{ij}(\vec{q})=(1-c)\omega_{ij}(\vec{q}),\\
&\bar{\Omega}_{ijk}(\vec{q}_1,\vec{q}_2,\vec{q}_3)=a[\delta_{i,k}\Gamma(i,j,q_2)\\
&\qquad\qquad -\delta_{i,j}\Gamma(i,k;q_3)],\\
&\bar{V}_{ijkl}(\vec{q}_1,\vec{q}_2,\vec{q}_3,\vec{q}_4)=-c\delta_{i,l}\delta_{j,k}\Gamma(j,i,\vec{q}_2-\vec{q}_3)\\
&\qquad\qquad -2(1-c)\delta_{i,k}\delta_{j,l}\Gamma(j,i,\vec{q}_2-\vec{q}_4),
\end{aligned}\tag{13}$$

where z is the number of nearest neighbors.

In q space the rules for forming $\bar{L}_n(i,j,\vec{q})$ are as follows: (i) Form all distinct irreducible diagrams with n events from the set of basic event diagrams that start on the left with a single dashed line labeled $(\vec{q},i)$ and end on the right with a single dashed line labeled $(\vec{q},j)$. (ii) Label all internal lines with dummy indices $\vec{q}',k$ and associate the analytical expression with each diagram that is the product of the analytical expressions associated with each event. (iii) Sum over all internal wave vectors and basis indices.

B. Examples

In this subsection we shall investigate some approximations for the particle-correlation functions. First we consider the method of self-consistent integral equations. This method is described in Paper I and is derived in detail in Ref. 7. With this method $\bar{K}(i,j,\vec{q};t)$ is expanded in a series, each term of which is a functional of $\bar{D}(i,j,\vec{q};t)$. Thus, we get

$$\bar{K}(i,j,\vec{q};t)=\sum_{n=1}^{\infty}\bar{K}_n(i,j,\vec{q};t).\tag{13a}$$

The rules for forming $\bar{K}_n(i,j,\vec{q};t)$ are as follows: (i) Draw all distinct skeleton diagrams made up of the basic event diagrams, starting on the left with a single dashed line labeled $\vec{q},i$, ending on the right with a single line labeled $\vec{q}$, j, and containing n events. (ii) Label the first dot on the left with the time t, the last dot on the right by time 0, and all internal dots by t_l. Label all internal lines with dummy wave vectors $\vec{q}'$ and label all lines where they go into and out of dots by dummy basis indices k. Thus each internal line has a distinct basis index at each end. (iii) Associate the appropriate analytical expression from Fig. 4 with each dot. Replace every line going from (k,t_l) to (k',t_m) along a line labeled $\vec{q}'$ by $D(k,k',\vec{q}';t_l-t_m)$ if the line is solid and by $\bar{D}(k,k',\vec{q}';t_l-t_m)$ if the line is dashed. (iv) Integrate over all internal times, sum over all internal wave vectors, sum over all internal basis indices, and multiply by $(-1)^{n+1}$. If the diagram has only one event, multiply by $\delta(t)$.

In Fig. 5 the skeleton diagrams for $\bar{D}$ up to third order for the case $\Gamma_{\alpha\beta}=\Gamma_{\beta\alpha}$ are displayed using the stated rules we obtain

$$\bar{K}_1(i,j,\vec{q};t)=\bar{\omega}(i,j,\vec{q})\delta(t),\tag{14a}$$

$$\bar{K}_2(i,j,\vec{q};t)=-\frac{1}{N}\sum\bar{\Omega}_{ikl}(\vec{q},\vec{q}-\vec{q}',\vec{q}')\bar{\Omega}_{jmn}(\vec{q},\vec{q}-\vec{q}',\vec{q}')D(ln,\vec{q}',t)\bar{D}(km,\vec{q}-\vec{q}',t),\tag{14b}$$

$$\begin{aligned}\bar{K}_3(i,j,\vec{q};t)=\frac{1}{N^2}\sum\int dt'\,\bar{\Omega}_{ilk}(\vec{q},\vec{q}-\vec{q}',\vec{q}')D(k,m,\vec{q}',t-t')\bar{D}(l,n,\vec{q}-\vec{q}',t-t')\\ \times\bar{V}_{nmpo}(\vec{q}-\vec{q}',\vec{q}',\vec{q}-\vec{q}'',\vec{q}'')D(O,r,\vec{q}'',t')\bar{D}(p,s,\vec{q}-q'',t')\bar{\Omega}_{jsr}(\vec{q},\vec{q}-\vec{q}'',\vec{q}'').\end{aligned}\tag{14c}$$

All internal wave vectors and basis indices are summed over in these equations. In this case $(\Gamma_{\alpha\beta}=\Gamma_{\beta\alpha})$, D is trivial and in frequency space is given by

$$\sum_j[\omega\delta_{ij}+i\omega(i,j,\vec{q})]D(j,k,\vec{q},\omega)=i\delta_{ik}.\tag{15}$$

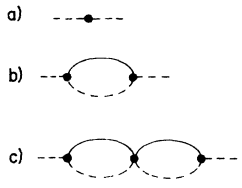

FIG. 5. Skeleton diagrams for $\bar{D}$ up to third order for $\Gamma_{\alpha\beta}=\Gamma_{\beta\alpha}$.

However, Eq. (14) must be solved self-consistently with the equation

$$\omega\bar{D}(i,k,\vec{q},\omega)+i\sum_j \bar{K}(i,j,\vec{q},\omega)\bar{D}(j,k,\vec{q},\omega)=i\delta_{ik}. \tag{16}$$

We have not solved Eqs. (14) and (16) self-consistently. We have solved them for various values of $\vec{q}$ and ω by using the effective field or first-order solution for $\bar{D}$ in Eqs. (14). This is rigorously correct only in the limits $c\to 0$ and $c\to 1$. It is correct when $c\to 0$ because if $c=0$, then $\bar{D}=D$. Thus the procedure gives $\bar{D}$ correctly to lowest order in c. If $c\to 1$, $\bar{K}(i,j,\vec{q},\omega)$ is proportional to $\bar{c}=1-c$ and thus $\bar{D}(t)$ is essentially constant over the time it takes $D(t)$ to decay to zero. Thus in this case we obtain $\bar{D}$ correctly to lowest order in $\bar{c}$.

In particular, we have calculated the diffusion coefficients for the four lattices under consideration. In the effective-field or first-order approximation we obtain

$$D_1(\mathrm{sc})=\bar{c}\Gamma_0 a^2,\quad D_1(\mathrm{bcc})=\bar{c}\Gamma_0 a^2, \tag{17}$$
$$D_1(\mathrm{fcc})=\bar{c}\Gamma_0 a^2,\quad D_1(\mathrm{oct})=\tfrac{1}{6}\,\bar{c}\Gamma_0 a^2,\quad \bar{c}=1-c.$$

For the three Bravais lattices we have calculated the D_2, the second-order approximation solving the equations non-self-consistently. The results to second order are given by

$$D_2=[1-\alpha c/(2-c)]D_1, \tag{18}$$

where $\alpha=0.419$, 0.318, and 0.245 for the sc, bcc, and fcc lattices, respectively. To third order, the results are

$$D_3=[1-\alpha c/(2-c)+\beta c(3c-2)/(2-c)^2]D_1, \tag{19}$$

where $\beta=0.088$, 0.050, and 0.030 for the sc, bcc, and fcc lattices, respectively. From these results it would appear that the procedure converges rapidly. In fact, as $c\to 1$ or $\bar{c}\to 0$ (the concentration of vacancies approaches zero), we obtain $D_3/D_1=0.669$, 0.732, and 0.785 for the sc, bcc, and fcc lattices, respectively. This is very close to the accepted values[3] of 0.653, 0.727, and 0.781, respectively.

However, this fast convergence hold only for $qa\ll 1$ and does not persist over the whole Brillouin zone. In fact, in the limit $c\to 1$, the convergence is so bad that the approximation gives unphysical results for the sc lattice for large q along a [111] direction. In the limit $c\to 1$ one can talk in the language of the hopping of a single vacancy. In this limit the only diagrams that contribute to $\bar{K}$ are those like Figs. 5(b) and 5(c) but with an arbitrary number of $\bar{V}$ events. Each term corresponds to a term in a Born scattering series. Clearly, what happens here is that the Born series converges rapidly for small q but very slowly for large q. Thus, in order to obtain good results, when c is near 1, one has to sum the entire Born series. This program is now under way.

For the octahedral problem, we obtain

$$D_2=0.32D_1, \tag{20}$$

$$D_3=0.55D_1, \tag{21}$$

in the limit $\bar{c}\to 0$. Here even the convergence at small wave vector is poor and the entire Born series will have to be summed for even an accurate diffusion coefficient. At lower concentrations the convergence is much better, scaling approximately as c.

We have briefly investigated the effects of removing the restrictions and Fourier transforming the diagrams. Analytically we have been able to include the effects of the restrictions by computing the change in $\bar{K}$ if the restriction was taken into account once. The effects are zero at zero and infinite frequencies and the effects were at most 8% of K_2 at any frequency or wave vector for the sc lattice. The effects on the other Bravais lattices were even less.

Finally, we have briefly investigated the results obtained with Padé approximates as described in Eq. (I-35). The lowest order or [1,0] Padé approximant yields

$$\bar{K}(i,j,\omega,\vec{q})=\bar{c}\omega(i,j,\vec{q})[1-b_1/(1-ib_2\omega)],$$
$$b_1=2c/[z(2-c)+(3c-2)-2(2-c)\delta(\mathrm{fcc})], \tag{22}$$
$$b_2=1/\Gamma_0[z(2-c)+(3c-2)-2(2-c)\delta(\mathrm{fcc})],$$

where z is the number of nearest neighbors and $\delta(\mathrm{fcc})$ equals one for the fcc Bravais lattice and zero for the other three lattices. The reason for the extra term with the fcc lattice is that three sites can be mutual nearest neighbors.

In the limit of small wave vectors, Eqs. (22) give a reasonably good approximation to the results obtained from the self-consistent integral equations. For example, the diffusion coefficients obtained are $D/D_1=0.714$, 0.778, 0.818, and 0.600 for the sc, bcc, fcc, and octahedral cases, respectively, in the limit $c\to 1$. The results for finite frequencies and/or lower concentrations were even closer to the results obtained from the self-consistent integral equations. At larger wave vectors the results obtained from Eq. (22) diverge considerably from the results obtained from Eqs. (14).

We have also tried the second order or [2,1] Padé approximant in the limit $c\to 1$ and for small wave vectors. With all four lattices the diffusion coefficients obtained were almost exactly midway between the values obtained from Eq. (22) and the values obtained from the self-consistent integral equations.

*Supported in part by the NSF under Grant No. DMS 75-18100.

[1]Peter A. Fedders and O. F. Sankey, preceding paper, Phys. Rev. B 15, 3580 (1977), hereafter referred to as I. Equation (n) of this paper will be referred to as Eq. (I-n).

[2]J. Bardeen and C. Herring, in *Imperfections in Nearly Perfect Crystals* (Wiley, New York, 1950).

[3]For a review and further references, see C. P. Flynn, *Point Defects and Diffusion* (Clarendon, Oxford, 1972), Chap. 6.

[4]See, for example, H. C. Torrey, Phys. Rev. 92, 962 (1953); M. Eisenstadt and A. G. Redfield, *ibid.* 132, 635 (1963); D. Wolf, Z. Naturforsch, A 26, 1816 (1971); D. Wolf, Phys. Rev. B 10, 2710 (1974); D. Wolf, J. Magn. Resonance 17, 1 (1975).

[5]P. A. Fedders (unpublished).

[6]E. F. W. Seymour, R. M. Cotts, and David Williams, Phys. Rev. Lett. 35, 165 (1975).

[7]C. W. Myles and P. A. Fedders, Phys. Rev. B 9, 4872 (1974).