Transport of spin polarization in a system of disperse and random walks in disordered media

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The derivation of a kinetic equation describing the transport of spin polarization through a system of impurity nuclei in fixed positions is examined. The problem of deriving a solution of this equation averaged over random positions of the impurity nuclei is also examined. A control equation is first derived. Its accuracy is evaluated by modeling the effect of the medium by a realistic random process. A simpler equation of the balance-equation type is then derived. The conditions for its applicability in real experiments on the magnetic resonance and relaxation of polarized β-active nuclei are studied. A solution [i.e., a propagator $P_{xy}(t)$] averaged over the positions of the impurity nuclei is derived for intermediate times satisfying $\beta t \lesssim 1$, where β is the Förster constant, which is proportional to the rate of transport over the average distance. This derivation is based on a concentration expansion. The problem of the long-term asymptotic behavior $P_{xy}(t\to\infty)$ is discussed in detail. A semiphenomenological theory is formulated for calculating the propagator at all t and all concentrations. A new method is found for calculating the diffusion coefficient $\mathscr{D}$. This new method is based on a restructing of the concentration expansion. The results calculated in the leading approximation for $\mathscr{D}$ and for the first correction agree well with the $\mathscr{D}$ value which has been measured by the method of optical four-wave mixing in a related exciton-transport problem. For contemporary experiments, it is important to consider the preasymptotic terms in $P_{00}(t\to\infty)$ and in the memory function $M(\mathbf{k}\to 0)$. A coherent-medium method is formulated for calculating P_{xy}. Explicit expressions are proposed for $P_{00}(t)$ for use in planning experiments on the delocalization of excitations in disordered systems by the β-NMR method and by the method of time-resolved fluorescence line narrowing. Criteria of importance for a critical comparison of theory and experiment are discussed.

1. INTRODUCTION

The problem of a random walk in a disordered system is one of the most interesting and most fundamental problems in physics today. It has attracted interest because of the extreme simplicity with which very complex problems can be formulated, because of the profound relationships with general problems of nonlinear statistics and field theory, because of the wide range of phenomena which can be described by this approach, and because of the opportunities which have opened up for extremely informative experiments.

The first reports of detailed experimental analyses of the problem of a random walk in a disordered system with a dipole-dipole interaction have recently appeared. The method of β-NMR spectroscopy (the magnetic resonance and relaxation of polarized β-active nuclei) has been used.[1–4] Our purpose in the present paper is to take a systematic look at the physics underlying these studies. In particular, we discuss a kinetic equation which describes the transport of polarization through a system of impurity nuclei in a crystal, the problem of deriving a solution of this equation averaged over the impurity distribution, and the basic theoretical results and hypotheses which can be brought before the court of experiment for judgment. Some of the results of a theory[5] originally derived for β-NMR have now found use in optical studies of exciton transport.[6,7] Accordingly, we will also take a look at the correspondence between the theoretical work in this field and the approach taken in Refs. 1–5.

For definiteness, we will consider a specific system[1–4] everywhere below in our discussion of the relaxation of polarized β-active nuclei. This specific system consists of a ^{6}Li isotopic impurity in a LiF crystal with one added β-active ^{8}Li nucleus with a spin $I_0 = 2$, a g-factor $g_0 = 0.8267$, and a decay half-life $T_{1/2} = 0.84$ s. In the initial state, only the β-active nucleus is polarized, and the spin density matrix of the crystal is

$$\rho(t=0)=\rho_0=\frac{1}{\mathrm{Sp}\,1}\left[1+\frac{3p_0}{I_0(I_0+1)}I_0^z\right],$$
$$p_0=\mathrm{Sp}\,I_0^z\rho_0\sim 1. \qquad (1)$$

Here P_0 is the polarization of the β-active nucleus, which is parallel to the external magnetic field $\vec{\mathscr{H}}_0 \| \mathbf{z}$. An important point is that over wide ranges of $\mathscr{H}_0$ and of the temperature the depolarization of the β-active ^{8}Li nucleus results from an interaction which simultaneously changes the z components of the two spins by $\Delta m = 1$ in different directions. These (flip-flop) transitions are effective only with the spins of the ^{6}Li nuclei ($I_k = 1, g_k = 0.8220, k \neq 0$), which are present in a concentration $c \ll 1$, because the g-factors of ^{8}Li and ^{6}Li are almost exactly the same. The measurable quantity is the anisotropy of the β emission of ^{8}Li. The theoretical problem is to calculate the polarization $\langle p_0(t)\rangle_c$ of the ^{8}Li nucleus, averaged over the random distribution of the ^{6}Li impurity in the crystal.

It was in this process, which was observed in Refs. 8 and

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9, that it was first found possible to apply nuclear-physics methods to the study of condensed media for the direct observation of the dynamics of the densities of additive integrals of motion. This is a fundamental problem in statistical mechanics.[10-12] No less important is the circumstance that the dynamics of the density of additive integrals of motion in disordered systems is being studied.

2. KINETIC EQUATION

The transport of nuclear polarization through a system of impurity spins is described by the equation

$$\dot{p}_i = -\sum_j (\nu_{ji}p_i - \nu_{ij}p_j), \quad p_i(t=0)=\delta_{i0}, \tag{2}$$

which was formulated in Ref. 13 (it was later proposed in Ref. 14 on the basis of the same arguments). Here $p_i(t) = \mathrm{Sp}\, I_i^z \rho(t)$ is the polarization of spin i in the ^{8}Li–^{6}Li system; $i=0$ corresponds to ^{8}Li; $i\neq 0$ corresponds to ^{6}Li; and ν_{ji} is the rate of the polarization transport from spin i to spin j, which is given by

$$\begin{aligned}
&\nu_{ij} = I_i(I_i+1)\, w_{ij},\\
&w_{ij} = \frac{\pi}{6}\left(\frac{g_i g_j \beta_n^2}{\hbar}\right)^2 g_{ij}(\Delta_{ij})\,(1-3\cos^2\vartheta_{ij})^2 r_{ij}^{-6},\\
&g_{ij}(\omega) = \int_{-\infty}^{\infty} \frac{dt}{2\pi}\cos\omega t\, \frac{\mathrm{Sp}[I_i^+ I_j^- I_i^-(t) I_j^+(t)]}{\mathrm{Sp}(I_i^+ I_j^- I_i^- I_j^+)}.
\end{aligned} \tag{3}$$

Here $\Delta_{ij} = \omega_i - \omega_j$, g_i and ω_i are respectively the g-factor and Larmor frequency of spin i, $\mathbf{r}_{ij}$ is the vector between spin i and spin j, ϑ_{ij} is the angle between $\vec{\mathscr{H}}_0 \| \mathbf{z}$ and $\mathbf{r}_{ij}$, β_n is the nuclear magneton, and $g_{ij}(\omega)$ is the normalized cross-relaxation form function. The time dependence in the definition of g_{ij} stems from the secular[15,16] Hamiltonian $H_0 = H_T + H_{IT}$ of the dipole-dipole interactions H_T of the spins of the host (^{7}Li^{19}F) and the interactions H_{IT} of the impurity spins and the host spins.

The total spin Hamiltonian of the system is

$$H = H_z + H_0 + H_l' + H_1 = H_z + H_s + H_1. \tag{4}$$

Here H_z is the Zeeman Hamiltonian, H_l' is the interaction of the z components of the impurity spins with each other, and H_1 is the flip-flop interaction in the ^{8}Li–^{6}Li system, given by

$$\begin{aligned}
&H_1 = {}^1/_2 \sum_{ij} a_{ij} I_i^+ I_j^-,\\
&a_{ij} = {}^1/_2 \frac{\beta_n^2 g_i g_j}{r_{ij}^3 \hbar}(3\cos^2\vartheta_{ij}-1), \quad a_{ii}=0.
\end{aligned} \tag{5}$$

We wish to stress that only the z components of the local fields determined by Hamiltonians H_{IT} and H_l' are nonzero.

The derivation of Eq. (2) which was proposed in Refs. 13 and 14 is based on the hypothesis that the density matrix $\rho(t)$ is in quasiequilibrium form:

$$\rho(t) = \rho^{(0)}(t) = \frac{1}{\mathrm{Sp}\,1}\left[1 + \sum_k \frac{3p_k(t)}{I_k(I_k+1)} I_k^z\right]. \tag{6}$$

A more systematic analysis is carried out in two steps.[17,18] The first step is to derive a control equation for ρ_D, which is the diagonal part of the impurity density matrix ($[I_k^z, \rho_D] = 0$ for all k). The next step is to work from this control equation to derive Eq. (2). An important point for this analysis is that the magnetic moments and g-factors of the impurity spins are considerably smaller than those of the host spins, while the characteristic differences between the impurity spins are considerably larger. The fluctuation rate of the local fields at the impurity spins and these fields themselves are therefore substantially higher than the fields created by the impurity spins at each other. From an analysis of elementary problems (equivalent to two-spin problems)[19] one would expect that Hamiltonian H_1 would be small. This suggestion and also the choice of parameters for the description (ρ_D) should be tested in some realistic model of the motion of local fields. If the choice is wrong, the terms which are the lowest-approximation corrections to the memory function will increase without bound as time elapses, despite the fact that they are of higher order in H_1.

We introduce the superoperator $\mathscr{P}$, which projects the total density matrix onto states which are diagonal for the impurity spins and equilibrium for the reservoir (i.e., for the host spins):

$$\mathscr{P}\rho = \frac{(\mathrm{Sp}_T\rho)_D}{\mathrm{Sp}_T 1} = \frac{\rho_D}{\mathrm{Sp}_T 1}, \tag{7}$$

where the subscript D means to take the diagonal part. In the Nakajima–Zwanzig projection technique,[16,20] the exact kinetic equation

$$\begin{aligned}
&\frac{\partial \rho_D}{\partial t} = -\int_0^t d\tau \langle \mathscr{L}_1 \bar{\mathscr{P}} \exp(-i\mathscr{L}\bar{\mathscr{P}}\tau)\mathscr{L}_1\rangle_T \rho_D(t-\tau),\\
&\mathscr{L}\rho = [H\rho], \quad \mathscr{L}_1\rho = [H_1\rho], \quad \bar{\mathscr{P}} = 1-\mathscr{P},\\
&\langle \ldots\rangle_T = \mathrm{Sp}_T(\ldots)/\mathrm{Sp}_T 1
\end{aligned} \tag{8}$$

is reduced in lowest order in H_1 to the control equation

$$\begin{aligned}
&\frac{\partial \rho_D}{\partial t} = -\int_0^t d\tau [H_1[\langle H_1(\tau)\rangle_T, \rho_D(t-\tau)]]_D,\\
&H_1(\tau) = e^{-iH_s\tau} H_1 e^{iH_s\tau},
\end{aligned} \tag{9}$$

and H_s can be replaced by H_0 at the same accuracy level. According to (5), the time dependence $H_1(\tau)$ is concentrated in the operators

$$I_k^{\pm}(\tau) = \exp(-iH_0\tau) I_k^{\pm} \exp(iH_0\tau) = (I_k^{\mp}(\tau))^+.$$

Here we have

$$\begin{aligned}
&I_k^+(t) = T\exp\left[-i\int_0^t d\tau \hat{\omega}_l^k(\tau)\right] I_k^+ = I_k^+ \bar{T}\exp\left[-i\int_0^t d\tau \hat{\omega}_l^k(\tau)\right],\\
&\hat{\omega}_l^k = \sum_{i(F)} f_{ki} F_i^z + \sum_{i(L)} l_{ki} L_i^z = \sum_{A=F,L} \hat{\omega}_l^k(A),\\
&f_{ki} = \frac{\beta_n^2 g_k g_F (1-3\cos^2\vartheta_{ki})}{\hbar r_{ki}^3},\\
&l_{ki} = \frac{\beta_n^2 g_k g_L (1-3\cos^2\vartheta_{ki})}{\hbar r_{ki}^3}.
\end{aligned} \tag{10}$$

We have used the usual chronological- and antichronologi-

cal-ordering operators here, and we have introduced a local-field operator $\hat{\omega}_l^k$ (more precisely, a local-frequency operator). The first sum in (10) is over all the fluorine sites; the second is over the lithium sublattice; and F, g_F and L, g_L are the spins and g-factors of the ^{19}F and ^{7}Li nuclei, respectively.

At present we do not have a microscopic theory of the reservoir in a form adequate for evaluating the accuracy of Eq. (9). The evolution of the local frequencies $\hat{\omega}_l^k(t)$ of the spins I_k—this evolution is determined by the behavior of the z components of the reservoir spins—would thus naturally be approximated as a steady-state normal random process. This method, which dates back to the Anderson–Weiss lineshape theory, has been used previously for similar purposes in single-spin problems (see Ref. 19, which also has basic information on the theory of random processes). In this approximation we have

$$\left\langle \exp\left[i \sum_i \int_0^t d\tau\, \xi_i(\tau)\, \hat{\omega}_l{}^i(\tau) \right] \right\rangle_T = \exp\left\{ -\frac{1}{2} \int_0^t d\tau_1\, d\tau_2 \sum_{ij} \xi_i(\tau_1) \times \xi_j(\tau_2) \langle \omega_l{}^i(\tau_1-\tau_2)\, \omega_l{}^j \rangle \right\}, \tag{11}$$

where $\xi_i(\tau)$ is some arbitrary function. Entities of specifically this sort arise when the memory function in (8) is expanded in a series in H_1. The method is based on the idea that $z_T \gtrsim 10$ host spins contribute substantially to the local field at spin I_k, so the distribution of local fields at any instant can be approximated well by a normal distribution. We are obviously ignoring the effect of the impurity on the reservoir. This simplification is justified since the time scale for energy and polarization transport in the reservoir is no greater in order of magnitude than the flip-flop time of the reservoir spins, τ_T, and it is such that the relation $R\tau_T \ll 1$ holds. Here R is the rate of the impurity flip-flop, found from Eq. (2). Another important point is that in the initial state only one spin—not a macroscopic number of spins—is driven from equilibrium.

It is natural to require that the correlation function of the local frequencies $\omega_l^k(t)$ be equal to the correlation function of the local-frequency operators: $\langle \omega_l^k(t)\omega_l^n \rangle = \langle \hat{\omega}_l^k(t)\hat{\omega}_l^n \rangle_T$, where

$$\hat{\omega}_l{}^k(t) = \exp(-iH_T t)\, \hat{\omega}_l{}^k \exp(iH_T t).$$

A direct calculation and a comparison with the results of a numerical simulation[21] of correlation functions of the $\langle F_i^z F_j^z(t) \rangle_T$ type show that the following equation holds with an accuracy sufficient for the calculations below:[22]

$$\langle \omega_l{}^i(t)\, \omega_l{}^j \rangle = \sum_{A=F,L} \langle (\hat{\omega}_l{}^i(A))^2 \rangle_T \left(\frac{\tau_{cA}+T_{2A}}{(t^2+T_{2A}{}^2)^{1/2}+\tau_{cA}} \right)^{3/2} \times \exp\left\{ -\frac{(\mathbf{r}_i-\mathbf{r}_j)^2}{4D_A[(t^2+T_{2A}{}^2)^{1/2}+\tau_{cA}]} \right\}. \tag{12}$$

Here T_{2A}, τ_{cA}, and $D_A \sim zr_0^2/\tau_{cA}$ are respectively the phase relaxation time, the flip-flop time, and the spin diffusion coefficient for the spins of species A ($T_{2A} \lesssim T_{cA}$); z is the coordination number ($z = 12$ in LiF); and r_0 is the distance between the nearest spins of the same species. Expression (12) and also a satisfactory estimate of its parameters can be found by applying Eq. (2) to the host spins, rather than to the impurity. In this case we find

$$\frac{1}{\tau_{cA}} = \sum_{i(A)} w_{ij}{}^A, \quad \mathcal{D}_A \approx {}^1/_6 \sum_{i(A)} (\mathbf{r}_i-\mathbf{r}_j)^2 w_{ij}{}^A, \qquad T_{2A} = \int_0^\infty dt \frac{\langle A_i{}^+ A_i{}^-(t) \rangle_T}{\langle A_i{}^+ A_i{}^- \rangle_T}. \tag{13}$$

Here w_{ij}^A is defined in the way that ν_{ij} is defined in (3), but with $I \to A$ and $g_k \to g_A$. Expression (12) reflects (first) the circumstance that the correlation function $\langle A_i^z A_j^z(t) \rangle_T$ is an even and smooth function of the time; at $t \lesssim T_{2A}$, the t dependence is quadratic. Second, it reflects the circumstance that at $t \lesssim \tau_{cA}$ this correlation function is localized in a volume $\sim r_0^3$. Third, it reflects the circumstance that at $t \gg \tau_{cA}$ this correlation function satisfies a diffusion equation and thus falls off comparatively slowly.

We can now estimate the accuracy of Eq. (9) by evaluating the terms coming after the leading term in the expansion of the memory function of Eq. (8) in the series in H_1. A direct calculation shows that these terms do not grow with the time and that the corrections which they make to the memory function of Eq. (2), found by the procedure described below in the derivation of (2) from (9) [*sic*], do not exceed in order of magnitude a fraction $\varepsilon = \max(\varepsilon_1, \varepsilon_2, \varepsilon_3)$ of the value of the leading term. Here

$$\varepsilon_1 = R\tau_T, \quad \varepsilon_2 = RT_2, \quad \varepsilon_3 = R(T_{2T}\tau_T)^{1/2} z_e^{-1} \ln(1+\eta), \quad \eta = T_{2T}{}^3/\tau_T T_2{}^2,$$

T_2 is the phase relaxation time of the impurity spins,

$$T_2 = \int_0^\infty dt \left\langle T \exp\left(-i \int_0^t d\tau \hat{\omega}_l{}^k(\tau) \right) \right\rangle_T, \quad \tau_T = \max(\tau_{cF}, \tau_{cL}),$$

and $R \sim z_e a_e^2 T_2$ where z_e and a_e are the effective number of nearest neighbors and the interaction between them in the impurity system.

It follows from the results of Ref. 23 that at small t we have $z_e = 1$, and a_e is the interaction at a distance r_0. At large t, the quantity z_e may increase to $z_e \sim 10$ (Sec. 3), but in this case a_e corresponds to the average distance $\bar{r} = (\Omega/c)^{1/3} \sim r_0 c^{-1/3}$, where Ω is the volume of the unit cell. To estimate T_2, T_{2A}, and τ_{cA} in this system, we can assume that the correlation functions of the transverse components of the reservoir spins and the impurity spins are Gaussian. This conclusion follows from the results of Ref. 24 (from measurements of the ^{8}Li NMR lineshape in LiF) and Ref. 25 (from measurements of the spin diffusion coefficient in CaF_2). As a result we find $\eta < 1$ and that the error of Eq. (9) at an impurity concentration $c \leqslant 10\%$ is $\varepsilon < 0.01$.

The condition for the observability of a process is usually $Rt \sim 1$. Accordingly, using $RT_2 \ll 1$, we can put Eq. (9) in the following form at $t \gg T_2$:

$$\dot{\rho}_D = -\int_0^\infty d\tau [H_1[\langle H_1(\tau) \rangle_T, \rho_D(t)]]_D = -{}^3/_8 \sum_{km} w_{km}[I_k{}^- I_m{}^+ [I_k{}^+ I_m{}^-, \rho_D]]. \tag{14}$$

Certain questions concerning the conditions $\varepsilon_1 \ll 1$ and $\varepsilon_2 \ll 1$—conditions for the applicability of a perturbation theory in single-particle problems—are discussed in Ref. 19. Here we wish to stress that in the multiparticle case both of these conditions are required in order to keep the terms of higher order in H_1 small. In several elementary single-spin problems (and in two-spin problems which can be reduced to single-spin problems; see Refs. 26 and 27, for example), the corrections for H_1 are kept small by the condition $\varepsilon_1 \ll 1$. The condition $\varepsilon_2 \ll 1$ is required only to make the transition from a nonlocal equation as in (9) to a Markov equation as in (14). In the case $T_{2T}^3 > T_2^2 \tau_T$ we find yet another condition: $\varepsilon_3 \ll 1$. However, for these relations between parameters it appears that we cannot assume that the impurity has only a slight effect on the reservoir, so the method of estimates which has been used becomes incorrect. This analysis leans heavily on the condition

$$\int_0^\infty dt \langle \omega_l^i(t)\, \omega_l^k \rangle \lesssim \tau_T \langle (\omega_l^i)^2 \rangle < \infty,$$

which is valid for three-dimensional reservoirs.

Equation (14) is still too complicated for specific calculations, and it contains much unobservable information. To find a simpler equation we note that $p_k(t)$ is the density of an additive integral of motion. The time variation of the density of an additive integral of motion is the slowest process, and we would naturally expect that after a long time ρ_D should be a function of p_k, i.e., should reduce to $\rho^{(0)}(t)$ [see (6)]. According to (1), the initial state is also the same as $\rho^{(0)}$ at $p_{k\neq 0} = 0$. If we take $\rho^{(0)}(t)$ as a lowest approximation, we find Eq. (2) after we substitute $\rho_D = \rho^{(0)}$ into (14), multiply by I_i^z from the left, and take the trace.

If, on the other hand, we simply substitute (6) into (14), we find that with $I_k = 1/2$ we actually have $\rho_D(t) = \rho^{(0)}(t)$, and Eq. (2) follows exactly from (14). In the case $I_k \neq 1/2$, however, Eq. (14) takes ρ_D out of the space with the basis $(1, \{I_k^z\})$ and puts it in the space with the basis $(1, \{I_k^z\}, \{I_i^z U_j\})$, where

$$U_j = (I_j^z)^2 - {}^1/_3 I_j (I_j+1).$$

Taking

$$\rho_D(t) = \rho^{(1)}(t) = \frac{1}{\mathrm{Sp}\,1}\left[1 + \sum_k \frac{3p_k(t)}{I_k(I_k+1)} I_k^z + \sum_{k\neq m} \frac{s_{km}(t)}{V_{km}} I_k^z U_m\right], \tag{15}$$

$$s_{km}(t) = \mathrm{Sp}\, I_k^z U_m \rho^{(1)}(t), \quad V_{km} = \mathrm{Sp}\,(I_k^z U_m)^2/\mathrm{Sp}\,1,$$

as the next approximation, we can derive the system of equations[17]

$$\dot{p}_i = -\sum_j (\nu_{ji} p_i - \nu_{ij} p_j) - {}^3/_2 \sum_j w_{ij}(s_{ji} - s_{ij}), \quad p_i(0) = \delta_{i0}, \tag{16}$$

$$\dot{s}_{ij} = \frac{\varkappa_j}{30}(\nu_{ji} p_i - \nu_{ij} p_j) - 3\sum_{k\neq i} \nu_{kj} s_{ij} - \sum_{k\neq j} (\nu_{ki} s_{ij} - \nu_{ik} s_{kj}) + {}^1/_2 w_{ij} \varkappa_j s_{ji} - {}^3/_4 w_{ij} s_{ij} ({}^5/_{21}\varkappa_j + {}^3/_5 \varkappa_i - {}^6/_7), \quad s_{ij}(0) = 0.$$

Here $\varkappa_i = (2I_i - 1)(2I_i + 3)$. Eliminating s_{ij} from this system of equations, we find the equation

$$\dot{p}_i = -\int_0^t d\tau \sum_j K_{ij}(\tau)\, p_j(t-\tau). \tag{17}$$

Continuing this procedure, which might be called a "method of expanding projection operators," we can expand the Laplace transform of the memory function in a continued fraction:[17]

$$K(\lambda) = z_e - z_e(\lambda + z_e - z_e(\lambda + z_e - z_e(\lambda + z_e - \ldots)^{-1})^{-1})^{-1}. \tag{18}$$

Some operator coefficients on the order of unity have been omitted from this expression, and a dependence on only z_e and λ is indicated. It follows from Eqs. (16) that p_k and s_{km} depend on the time in different ways and that we have $s_{km}/p_k \to 0$ as $t \to \infty$ in a substantial spatial region. However, the p_k and s_{km} contributions to Eq. (17) are of the same order in t; this circumstance is reflected in (18). The hypothesis which led to (6) is thus by no means flawless; partial justification for it can be found only after a direct evaluation of the correction. We note that $K(\lambda \to 0)$ is not an analytic function of H_1.

A calculation of the correction from Eqs. (16) shows that its numerical value is substantially smaller than $1/z_e$ in order of magnitude and does not exceed 1% of the leading term in K, which is determined by Eq. (2) with $z_e \gtrsim 6$. Over a fairly large temporal region the quantities $z_e = 1$ and $z_e = 2$ are important (see Sec. 3 in Ref. 23). This case has been analyzed numerically. The error of Eq. (2) in two-spin problems ($z_e = 1$) is on the order of 1% if the spins are identical or approximately the same in magnitude. If there is instead a large difference, e.g., $I_0 = 1/2$ and $I_1 = 2$, Eq. (2) is not accurate, since it generates limiting values $p_k(t \to \infty)$ which are 100% off in comparison with those found from (14). Even in this case, however, the first term of the concentration expansion of $\langle p_0(t) \rangle_c$ calculated from (2) has an error of only 10%. The difference in the $p_k(t \to \infty)$ values stems from the existence of nonadditive integrals of motion J in addition to ΣI_k^z ($[JH_1] = 0, [JI_k^z] = 0$). In the two-spin problem with $I_0 = 1/2$ and $I_1 = 1$, for example, we have

$$J = I_0(I_0+1) I_1^z - I_1(I_1+1) I_0^z + {}^{33}/_2 I_0^z U_1,$$

while with $I_0 = I_1 = 1$ we have

$$J = U_0 I_1^z + U_1 I_0^z.$$

These integrals are important if $\langle J(t=0) \rangle \neq 0$. Table I reflects the overall situation with regard to two-spin problems. When we move to three-spin problems or, in general, as z_e increases, Eqs. (2) become more accurate.

In the expansion of the projection basis to $(1, \{I_k^z\}, \{I_k^z U_m\}, \{I_i^z I_j^z m^z\})$ which follows (15), the results given above remain valid if at large t we have $z_e \gtrsim 6$ (the correlation $I_i^z I_j^z I_k^z$ is important since, in contrast with $I_k^z U_m$, it contains the product of only densities of additive integrals of motion). The estimates become unsatisfactory, however, as z_e approaches $z_p = 2.7$, which is the percolation threshold of three-dimensional systems. We would naturally expect that $z_e(t \to \infty)$ would be related to the diffusion coefficient $\mathscr{D}$ for the polarization in a disordered system by

TABLE I. The limiting values $p_0(t\to\infty)$ for the solution of the problem with two spins, I_0, and I_1, with $p_0(t=0)=1$ and $p_1(0)=0$.

I_0	I_1 = 1/2		I_1 = 1		I_1 = 3/2		I_1 = 2	
1/2	1/2	1/2	1/3	3/11	1/4	1/6	1/5	1/9
1	3/4	8/11	1/2	1/2	3/8	8/23	3/10	1/4
3/2	17/20	5/6	2/3	15/23	1/2	1/2	2/5	5/13
2	9/10	9/11	23/30	3/4	5/8	8/13	1/2	1/2

Note. At an intersection of the spin values, the result of the exact solution of Eq. (14) with initial condition (1) is shown at the left, while the solution of approximate equation (2) is shown at the right.

$$\mathscr{D}=z_e\nu(\bar{r})\,\bar{r}^2/6,$$

where $\nu(\bar{r})$ is the transport rate over the average distance. Ordinarily, theoretical estimates yield $z_e \gtrsim 15$ (Refs. 5, 14, 28, and 29). The smallest of the known values of $\mathscr{D}$ was found for the diffusion of excitons in the experiments of Ref. 30 (see also Sec. 6), but again in this case we have $z_e > 8$.

In summary, we would expect that the error of Eqs. (2) and (3) in application to the ^{8}Li–^{6}Li system would be on the order of 1% at all t and that we would have $c\leqslant 10\%$.

3. OCCUPATION-NUMBER REPRESENTATION AND CONCENTRATION EXPANSION

To calculate observables averaged over the random impurity distribution in the crystal, it is convenient to introduce the occupation-number representation.[31,13] In this representation, Eq. (2) is replaced by

$$\dot{\tilde{P}}_{\mathbf{xy}}=-\sum_{\mathbf{z}} n_{\mathbf{x}}n_{\mathbf{z}}(\nu_{\mathbf{zx}}\tilde{P}_{\mathbf{xy}}-\nu_{\mathbf{xz}}\tilde{P}_{\mathbf{zy}}),\quad \tilde{P}_{\mathbf{xy}}(t=0)=\delta_{\mathbf{xy}},\quad (19)$$
$$n_{\mathbf{y}}=1,$$

or by the equivalent equation [13,23]

$$\dot{\tilde{P}}_{\mathbf{xy}}=-\sum_{\mathbf{z}}(n_{\mathbf{z}}\nu_{\mathbf{zx}}\tilde{P}_{\mathbf{xy}}-n_{\mathbf{x}}\nu_{\mathbf{xz}}\tilde{P}_{\mathbf{zy}})=-(\tilde{A}\tilde{P})_{\mathbf{xy}},$$
$$\tilde{A}=\sum_{\mathbf{z}} n_{\mathbf{z}}A^{\mathbf{z}},\quad A_{\mathbf{xq}}{}^{\mathbf{z}}=(\delta_{\mathbf{xq}}-\delta_{\mathbf{xz}})\nu_{\mathbf{zq}},\quad \tilde{P}_{\mathbf{xy}}(t=0)=\delta_{\mathbf{xy}}. \quad (20)$$

Here $\tilde{P}_{\mathbf{xy}}(t)$ is the polarization of the lattice site $\mathbf{x}$ at time t under the condition that site $\mathbf{y}$ is initially polarized; $\nu_{\mathbf{xz}}=\nu_{ij}$ ($\mathbf{r}_i=\mathbf{x}, \mathbf{r}_j=\mathbf{z}$) and $n_{\mathbf{x}}$ is the occupation number. Its value is $n_{\mathbf{x}}=1(0)$ when site $\mathbf{x}$ is (is not) occupied by an impurity spin. Site $\mathbf{y}$ may be polarized only if it is occupied by a spin, so we have set $n_{\mathbf{y}}=1$ in (19). At sites $\mathbf{x}\neq\mathbf{y}$ we have $\langle n_{\mathbf{x}}\rangle_c=c$, and the occupations of the different sites are not correlated. That Eqs. (2) and (19) are equivalent can be verified by noting that in unoccupied sites we have $\tilde{P}_{\mathbf{xy}}=0$, while in occupied sites, where $n_{\mathbf{x}}=1$, Eq. (19) reduces to (2) with $\mathbf{r}_0=\mathbf{y}$. Equation (20) follows from (19) when the identity $n_{\mathbf{x}}\tilde{P}_{\mathbf{xy}}=\tilde{P}_{\mathbf{xy}}$ is taken into account. We assume below that the transition rate is

$$\nu_{\mathbf{xz}}=\frac{\nu_0\chi(\vartheta_{\mathbf{xz}})r_0{}^s}{|\mathbf{x}-\mathbf{z}|^s},\quad \mathbf{x}\neq\mathbf{y}\neq\mathbf{z}, \quad (21)$$
$$\nu_{\mathbf{xy}}=\frac{\nu_1\chi(\vartheta_{\mathbf{xy}})r_0{}^s}{|\mathbf{x}-\mathbf{y}|^s},\quad \nu_{\mathbf{yx}}=\xi\nu_{\mathbf{xy}},\quad \nu_{\mathbf{xx}}=0,$$

where $\nu_1=\nu_1(\mathscr{H}_0)$, and $\nu_1(0)=\nu_0$. In the ^{8}Li–^{6}Li system we have $s=6, \xi=3=I_0(I_0+1)/I_k(I_k+1)$, $\delta_{\mathbf{xz}}$ is the angle between $\mathscr{H}_0$ and $\mathbf{x}-\mathbf{z}$, and $\chi(\vartheta)=(1-3\cos^2\vartheta)^2$. In descriptions of exciton transport it is customary to set $s=6$, $\chi=1$, and $\xi=1$ (Refs. 31, 28, and 29; see also Secs. 6 and 7). In the theory of hopping conductivity one uses an isotropic exponential dependence of $\nu_{\mathbf{xz}}$ on $|\mathbf{x}-\mathbf{z}|$ (Refs. 32 and 33).

The problem in the occupation-number representation is to calculate the propagator $P_{\mathbf{xy}}(t)=\langle\tilde{P}_{\mathbf{xy}}(t)\rangle_c$ averaged over occupation numbers, i.e., over impurity configurations. Corresponding to the polarization of the β-active ^{8}Li nucleus is $P_{\mathbf{yy}}(t)=P_{00}(t)=\langle p_0(t)\rangle_c$.

A natural time scale in this problem is the Förster constant $\beta=(256/243)(r_0^3/\Omega)^2\pi^3c^2\nu_0$ (Ω is the volume of the unit cell), which is defined by

$$Q_{\mathbf{x}}{}^{\mathbf{y}}(t)=\left\langle\exp\left(-\sum_{\mathbf{z}\neq\mathbf{y}} n_{\mathbf{z}}\nu_{\mathbf{zx}}t\right)\right\rangle_c=(1-\delta_{\mathbf{xy}})\exp(-(\beta t)^{1/2})$$
$$+\delta_{\mathbf{xy}}\exp(-(\beta_1 t)^{1/2}),\quad \beta_1=\frac{\nu_1}{\nu_0}\beta. \quad (22)$$

This expression is valid at small values of c, i.e., in the low-concentration limit, in which we have $c\to 0$, but βt remains nonzero [see Ref. 13 for a simple derivation of (22)].

A convenient method for calculating $P_{\mathbf{xy}}(t)$ under the condition $\beta t\lesssim 1$ is a concentration expansion[23] in cumulant form.[1-4] From Ref. 23 we have

$$P_{\mathbf{yy}}(t)=1+\sum_{m=1}^{\infty}\frac{c^m}{m!}\sum_{\mathbf{z}_1\ldots\mathbf{z}_m}{}'\sum_{k=0}^{m}(-1)^{m-k}C_m{}^k b_0^{(k)}(\mathbf{y},\mathbf{z}_1,\ldots,\mathbf{z}_k,t),$$
$$P_{\mathbf{x}\neq\mathbf{y}}(t)=cb_1^{(1)}(\mathbf{y},\mathbf{x},t)+\sum_{m=1}^{\infty}\frac{c^m}{m!}\sum_{\mathbf{z}_1\ldots\mathbf{z}_m}{}'\sum_{k=0}^{m}(-1)^{m-k}$$
$$\times C_m{}^k b_1^{(k+1)}(\mathbf{y},\mathbf{x},\mathbf{z}_1,\ldots,\mathbf{z}_k,t). \quad (23)$$

Here $b_i^{(k)}(\mathbf{y},\mathbf{z}_1,\ldots,\mathbf{z}_k,t)$ is the solution p_i of Eq. (2) for the case of a solitary $(k+1)$-spin cluster of impurity nuclei in sites $\mathbf{y},\mathbf{z}_1,\ldots,\mathbf{z}_k$. The prime on the summation over $\mathbf{z}_1,\ldots,\mathbf{z}_m$ means that all coincidences of summation variables are excluded and that they are bounded by the one common (large) volume V. Before we take the limit $V\to\infty$, the expression in the Σ' summation must be put in symmetric form.

If $\nu_0 t\gg 1$, then $b_i^{(k)}(\mathbf{y},\mathbf{z}_1,\ldots\mathbf{z}_k)$ is a smooth function of the coordinates $\mathbf{y}$ and $\mathbf{z}_i$. It varies slowly over a distance $\sim r_0$.

Under the condition $c \ll 1$ we can replace the sums over $\mathbf{z}_i$ by integrals. The shifts $\mathbf{z}_i \to \mathbf{z}_i + \mathbf{y}$ and the substitution $\mathbf{z}_i \to \mathbf{z}_i (\nu_0 t)^{1/3}$ lead to, for example,

$$P_{yy}(t) = 1 + \sum_{m=1}^{\infty} \frac{(c(\nu_0 t)^{d/s})^m}{m!} \times \int \frac{d^d z_1}{\Omega} \dots \int \frac{d^d z_m}{\Omega} \hat{S} \sum_{k=0}^{\infty} C_m^k (-1)^{m-k} \times b_0^{(k)}\left(0, \mathbf{z}_1 \dots \mathbf{z}_k, t = \frac{1}{\nu_0}\right),$$

where $d(=3)$ is the dimensionality of the space, and $\hat{S}$ means to put in a form symmetric with respect to the integration variables. The expression for P_{xy} transforms similarly. As a result we find[13]

$$P_{yy}(t) = f(\beta t), \quad P_{x \neq y}(t) = cg\left(\beta t, \frac{\mathbf{x}-\mathbf{y}}{\bar{r}}\right). \tag{24}$$

The functions f and g depend on the coordinates, the time, and the concentration only through the arguments specified in (24).

After a restructuring of expansion (23) into a cumulant expression we find the following result, which holds to within c^2 inclusively in the ^{8}Li–^{6}Li system:

$$P_{yy}(t) = \exp\left\{-\left(\frac{\beta_1 t}{\xi+1}\right)^{1/2} - \alpha\beta_1 t\right\}. \tag{25}$$

With $\mathscr{H}_0 = 0$ and $\xi = 3$, we have $\alpha = 0.013$, while with $\xi = 1$ we have $\alpha \lesssim 3 \cdot 10^{-3}$. The parameter α depends on $\mathscr{H}_0$ through $v = \nu_0/\nu_1(\mathscr{H}_0)$; values of this parameter are shown in Table II, which was constructed from the results of Ref. 23. For the transport of excitons we find the following from the results of Ref. 28, put in a different form ($d = 3, s = 6$):

$$P_{yy}(t) = \exp\{-(1/2\beta_0 t)^{1/2}\}(1 + \alpha_0\beta_0 t), \tag{26}$$

where $\beta_0 = (16/9)\pi^3 (r_0^3/\Omega)^2 c^2 \nu_0$ and $\alpha_0 = 0.0064$. Equation (23) of Ref. 31 leads to the same result in the leading order in c; explicit expression (26) (with $\alpha_0 = 0$) was proposed in Ref. 34.

The exceedingly small values of the constants α and α_0 show that (25) and (26) should hold well over a broad range of $p_{yy}(t)$. This expectation can be verified by comparing (26) in the case $\alpha_0 = 0$ with numerical calculations[34] and also with optical measurements of exciton detrapping.[6,7,35] Further support of (25) comes from a test in β-NMR (Refs. 2 and 4).

The cumulant version of the concentration expansion has proved extremely effective for describing a wide range of phenomena under the condition $P_{00}(t) > 0.1 P_{00}(0)$ (Refs. 1–4, 36, 37). We might note a generalization of this version for describing polarization transport in the case of translational motions of ^{8}Li–^{6}Li nuclei.[3,4,36–38] A corresponding generalization of the concentration expansion incorporating the motion of impurity nuclei was proposed in Ref. 39.

4. LONG-TERM ASYMPTOTIC BEHAVIOR

The concentration expansion gives us a solution of our problem under the condition $\beta t \lesssim 1$. The problem of determining the long-term asymptotic behavior, $P_{xy}(\beta t \to \infty)$, is considerably more complex and fundamental. In the case of a regular lattice (i.e., with $c = 1$) with a slight disorder,[40] a diffusive asymptotic behavior is realized in asymptotically exactly solvable models[41–43] with a moderate disorder and also in several mathematical models (Ref. 44, for example). In our system, the fluctuations of the operator $\tilde{A}$ from (20) are large (in fact, infinite in the small-c limit), but we have

$$Q_x^y(\lambda = 0) = \left\langle \left(\sum_{z \neq y} n_z \nu_{zx} \right)^{-1} \right\rangle_c < \infty .$$

The models of Refs. 41–43, which we just mentioned, have similar properties, so the ^{8}Li–^{6}Li system can naturally be classified as a system with a moderate disorder, and we can assume that it has a diffusive asymptotic behavior. To find a correct quantitative formulation of this hypothesis, we must allow for the circumstance that site $\mathbf{y}$ in (19) is reliably filled with the ^{8}Li spin, while the probability for finding a ^{6}Li spin at $\mathbf{x} \neq \mathbf{y}$ is $c \ll 1$ and that under the condition $(\mathscr{D}t)^{1/2} \gg \max(\bar{r}, |\mathbf{x} - \mathbf{y}|)$ the solution of Eqs. (19) and (20) is proportional to the steady-state solution

$$\tilde{P}_{xy}^{\,s} = \text{const}\, n_x (\xi\delta_{xy} + 1 - \delta_{xy}).$$

As a result we find

$$P_{yy}(\beta t \to \infty) = \frac{\xi\Omega}{c} \prod_{\alpha=1}^{d} (4\pi \mathscr{D}_\alpha t)\, \Pi^{1/2} \sim (\beta t)^{-d/2}, \tag{27}$$

$$P_{x \neq y}(\beta t \to \infty) = \Omega \prod_{\alpha=1}^{d} \{(4\pi \mathscr{D}_\alpha t)^{-1/2} \exp[-(x_\alpha - y_\alpha)^2/(4\mathscr{D}_\alpha t)], \tag{28}$$

where $\mathscr{D}_\alpha$ are the principal values of the diffusion tensor.[5,13] From (24) we have $\mathscr{D}_\alpha \sim \bar{r}^2\beta$.

It is extremely difficult to satisfy this simple relation when deriving a microscopic theory. Although expression (28) is repeated in all the papers on the theory of a random walk in a disordered system, starting with (for example) the paper by Scher and Lax,[32] the various authors have derived, in an equally systematic way, results other than (27) which

TABLE II. The parameter α in expression (25) as a function of $v = \nu_0/\nu_1(\mathscr{H}_0)$.

v	1	2	3	4	5	6	7	8	9	10	15	20
α	0,013	0,047	0,076	0,100	0,123	0,144	0,163	0,181	0,198	0,216	0,287	0,357

lead to the following expression in the case of dipole transport in three-dimensional space:

$$P_{yy}(t\to\infty)=\Omega\prod_{\alpha=1}^{3}(4\pi\mathcal{D}_\alpha t)^{-1/2}+be^{-\phi(t)}\sim c(\beta t)^{-3/2}+e^{-\phi(t)}, \tag{29}$$

where $\phi(t)\sim(\beta t)^{\varkappa}$, $1/2\leqslant\varkappa\leqslant 1$, and $b\sim 1$. In the low-c limit, we are left with only the exponential term here. That expression (29) is not correct was pointed out back in Ref. 13, but even in Ref. 29, which was an effort to introduce a diffusive tail on $P_{yy}(t)$ in the *GAF* theory,[28] it was only (29)—not (27)—which was found. This conclusion follows from, for example, Eq. (5.5) of Ref. 29. In those other papers, the memory function (more precisely, operator) $Z_{xq}(\lambda)$ was introduced in the following way:

$$P_{xy}(\lambda)=((\lambda+Z(\lambda))^{-1})_{xy}, \tag{30}$$

and Z was calculated approximately. According to (24) with $c\ll 1$, the relation $P_{00}(\lambda)=(\lambda+Z_{00}(\lambda))^{-1}$ should hold.[28,36] Using that relation along with (27), we find

$$Z_{00}(t\to\infty)=-P_{00}(t)\Big/\Big(\int_0^\infty dt\,P_{00}(t)\Big)^2, \quad d\geqslant 3. \tag{31}$$

In other words, the memory function $Z_{xq}(t)$ must have a long-term diffusion tail.[36] In Refs. 28, 29, and 32 this function fell off (in dipole transport) exponentially and was of a short-term nature.

A suitable short-term memory function can be introduced by

$$\lambda P_{xy}(\lambda)=\delta_{xy}-\sum_{q}\mathcal{M}_{xq}^{y}(\lambda)P_{qy}(\lambda),$$
$$\mathcal{M}^{y}=\langle\bar{A}\rangle_c^{y}-\langle\bar{A}\bar{C}^{y}(\lambda+\bar{A}\bar{C}^{y})^{-1}\bar{A}\rangle_c^{y}. \tag{32}$$

This function was proposed in Ref. 5 and later in Ref. 45 (see also Refs. 18 and 23). Here $\tilde{C}^{y}=1-C^{y}$, and C^{y} is a projection operator which performs a configurational averaging under the condition $n_y=1$: $C^{y}(...)=\langle...\rangle_c^{y}$. A memory function of this sort [a geometric function, in contrast with dynamic function (30)] explicitly incorporates the circumstance that the capacity of site **y** is larger by a factor of ξ/c than that of sites $\mathbf{x}\neq\mathbf{y}$. Equation (32) [see also (33) and (38)] converts at large t into the equation corresponding to the problem of heat transfer in a homogeneous medium with a specific heat $\zeta=c/\Omega$, to which a small object with a large specific heat $m=\xi\Omega\zeta/c$ and an initial temperature different from that of the medium has been added at point **y**.

5. SEMIPHENOMENOLOGICAL THEORY

An approach proposed in Ref. 5 for an approximate calculation of $\mathcal{M}^{y}$ corrects and substantially simplifies the semiphenomenological theory of Ref. 32, which was intended for calculating Z (the terminology was different). Scher and Lax[32] apparently regarded their analysis to be microscopic. In our approach, the conservation law

$$\sum_{x}P_{xy}(t)=1$$

is taken into account by putting Eq. (32) in the form

$$\dot{P}_{xy}=-\sum_{q}\Big[\int_0^t d\tau N_{qx}^{y}(\tau)P_{xy}(t-\tau)-\int_0^t d\tau N_{xq}^{y}(\tau)P_{qy}(t-\tau)\Big], \tag{33}$$

and by making the following assumptions: First, the first term in (square) brackets is the rate of polarization outflow from **x** to **q**, while the second term is the rate of inflow from **q** to **x**. Second, the polarization inflow rate from **q** to **x** does not depend on whether the polarization reached **q**. The processes of inflow to the site and outflow from it are now separated, and to determine the kernels N_{xq}^{y} we can treat the simple case in which an exact averaging can be carrried out. For this purpose we choose the process of the outflow of polarization from an arbitrary site:

$$\dot{\tilde{F}}_{xx}=-\sum_{z}n_z\nu_{zx}\tilde{F}_{xx}, \quad \dot{\tilde{F}}_{z\neq x}=n_z\nu_{zx}\tilde{F}_{xx},$$
$$\tilde{F}_{zx}(t=t_0)=n_z\delta_{zx}, \quad \tilde{F}_{zx}(t<t_0)=0. \tag{34}$$

In accordance with the assumptions above, the average $F_{zx}=\langle\tilde{F}_{zx}\rangle_c^{y}$ must satisfy the equations

$$\dot{F}_{xx}=-\int_0^t d\tau\sum_{z}N_{zx}^{y}(\tau)F_{xx}(t-\tau), \quad \dot{F}_{z\neq x}$$
$$=\int_0^t d\tau N_{zx}^{y}(\tau)F_{xx}(t-\tau),$$
$$F_{zx}(t=t_0)=\delta_{zx}(c+(1-c)\delta_{xy}), \quad F_{zx}(t<t_0)=0. \tag{35}$$

Using (34), we then find equations for the kernels,

$$\int_0^t d\tau\,N_{zx}^{y}(\tau)\left\langle\exp\Big(-\sum_{q}n_q\nu_{qx}(t-\tau)\Big)\right\rangle_c^{y}$$
$$=\left\langle n_z\nu_{zx}\exp\Big(-\sum_{q}n_q\nu_{qx}t\Big)\right\rangle_c^{y}, \tag{36}$$

from which the time t_0 has dropped out, as it should for a steady-state process. The averages in (36) can be calculated easily by the technique of Ref. 13. If we omit the condition $n_y=1$ from (36), we find that in the limit $c\ll 1$ Eqs. (33) and (36) are equivalent to the Scher–Lax equations.[32] Noting that we have $n_y=1$, we see that Eqs. (33) and (36) have a solution which is the same as (27), but—in contrast with (28)—it vanishes as $\mathbf{x}\to\mathbf{y}$. In this region, these equations can be refined by working from expansions (23) and the exactly solvable problem of Ref. 46. In the occupation-number representation number, that problem is

$$\dot{\tilde{\Phi}}_{yy}=-\sum_{z}(n_z\nu_{zy}\tilde{\Phi}_{yy}-n_y\nu_{yz}\tilde{\Phi}_{zy}), \quad \tilde{\Phi}_{xy}(0)=\delta_{xy},$$
$$\dot{\tilde{\Phi}}_{x\neq y}=-n_y\nu_{yx}\tilde{\Phi}_{xy}+n_x\nu_{xy}\tilde{\Phi}_{yy}, \quad n_y=1. \tag{37}$$

As a result we find the equations[5]

$$\int_0^t d\tau\,N_{zx}^{y}(\tau)Q_x^{y}(t-\tau)=\frac{c\nu_{zx}\exp(-\nu_{zx}t)}{1+c(\exp(-\nu_{zx}t)-1)}Q_x^{y}(t),$$
$$\mathbf{z}\neq\mathbf{y}, \tag{38}$$

$$N_{yx}{}^{y}=\frac{\nu_{yx}}{c\nu_{xy}}N_{xy}{}^{y},\qquad Q_{x}{}^{y}(t)=\left\langle \exp\left(-\sum_{z\neq y}n_{z}\nu_{zx}t\right)\right\rangle_{c}$$

$$=\prod_{z\neq y}[1+c(\exp(-\nu_{zx}t)-1)].$$

Equations (36) and (38) and their solutions are the same at $|\mathbf{x}-\mathbf{y}|\gg\bar{r}$. A more formal derivation of Eqs. (33) and (38), as the lowest approximation in the projection technique, is given in Ref. 36.

Equations (33) and (38) are exact in the case $c=1$. Their solutions are the same as the exact solutions up to terms of order c^{1} inclusively in the expansion in c. These equations given the exact asymptotic behavior at small and large values of βt when applied to problem (37). These equations are also exact in the limit in which the transport between sites $\mathbf{x}\neq\mathbf{y}$ is considerably more rapid than the exchange with site $\mathbf{y}$. In the ^{8}Li–^{6}Li system, this limit corresponds to high fields $\mathscr{H}_{0}$, with $\nu_{0}\gg\nu_{1}$.

At low concentrations, a solution of Eqs. (38) is

$$N_{zx}{}^{y}(\lambda)=\frac{(1-\delta_{zy})c\nu_{zx}Q_{x}{}^{y}(\lambda+\nu_{zx})+\delta_{zy}\nu_{yx}Q_{x}{}^{y}(\lambda+\nu_{yx})}{Q_{x}{}^{y}(\lambda)}. \quad (39)$$

The kernels $N_{zx}^{y}(t)$ are localized at $|\mathbf{z}-\mathbf{x}|\leqslant\bar{r}$ and fall off more rapidly than any power $(\beta t)^{-n}$ at $\beta t\gg 1$. The latter result is obvious from the fact that with $\lambda=0$ all the derivatives

$$(-\partial/\partial\lambda)^{n}Q_{x}{}^{y}(\lambda)=\int_{0}^{\infty}dt\,t^{n}Q_{x}{}^{y}(t)\exp(-\lambda t)$$

along with $(\partial/\partial\lambda)^{n}N_{zx}^{y}(\lambda)$, are finite.

To analyze the long-term asymptotic behavior of the solution of Eqs. (33) and (39), we introduce the auxiliary propagator $\hat{G}_{xy}(t)$, which is the solution of the problem

$$\dot{\hat{G}}_{xq}=-\sum_{z}\int_{0}^{t}d\tau\hat{N}_{zx}(\tau)[\hat{G}_{xq}(t-\tau)-\hat{G}_{zq}(t-\tau)],$$

$$\hat{G}_{xq}(t=0)=\delta_{xq},$$

$$\hat{N}_{zx}(\lambda)=\frac{c\hat{\nu}_{zx}Q(\lambda+\hat{\nu}_{zx})}{Q(\lambda)}, \quad (40)$$

$$Q(t)=\prod_{z}[1+c(e^{-\hat{\nu}_{zx}t}-1)].$$

The functions $\hat{\nu}_{zx}$ here are defined by the first equation in (21) for all $\mathbf{z}$ and $\mathbf{x}$. In integral form, Eq. (40) is

$$\hat{G}_{xq}(t)=\delta_{xq}Q(t)+\sum_{z}\int_{0}^{t}dt_{1}\int_{0}^{t_{1}}dt_{2}Q(t-t_{1})\hat{N}_{xz}(t_{1}-t_{2})\hat{G}_{zq}(t_{2}). \quad (41)$$

It follows from this representation that we have

$$\hat{G}_{xq}(t)=\delta_{xq}Q(t)+\hat{G}_{xq}{}^{0}(t). \quad (42)$$

At $\beta t\gtrsim 1$, $\hat{G}_{xq}^{0}(t)$ is a smooth function of $\mathbf{x}-\mathbf{q}$, and we have $\hat{G}_{xq}^{0}\sim c$, while we have $Q(t)\sim c^{0}$ according to the discussion accompanying Eqs. (29)–(31).

Since the kernels $\hat{N}_{zx}(\tau\to\infty)$ fall off rapidly at large βt, we have

$$\dot{\hat{G}}_{xq}=-\sum_{z}V_{zx}(\hat{G}_{xq}-\hat{G}_{zq})$$

$$-\sum_{z}V_{zx}^{(1)}\frac{\partial}{\partial t}(\hat{G}_{xq}-\hat{G}_{zq})-\ldots, \quad (43)$$

where

$$V_{xz}=\hat{N}_{xz}(\lambda=0),\qquad V_{xz}^{(1)}=\frac{\partial\hat{N}_{xz}(\lambda)}{\partial\lambda}\Big|_{\lambda=0}.$$

Solving Eq. (43) by Fourier transforms, we find

$$\hat{G}_{xq}(\beta t\gg 1)=\frac{\Omega}{(2\pi)^{3}}\int_{B}d^{3}k\exp[i\mathbf{k}(\mathbf{x}-\mathbf{q})$$

$$-(V(0)-V(\mathbf{k}))t](1+O(1/\beta t)), \quad (44)$$

where the integration is over a unit cell of the reciprocal lattice,[47] and

$$V(\mathbf{k})=\sum_{x}e^{-i\mathbf{k}x}\hat{N}_{x0}(\lambda=0)=\hat{N}(\mathbf{k},\lambda=0). \quad (45)$$

The asymptotic behavior $\hat{G}(\beta t\to\infty)$ is determined by the asymptotic behavior $V(\mathbf{k}\to 0)$; this point has already been taken into account, in the estimate of the correction term from (44). In this region we have

$$V(0)-V(\mathbf{k})=\mathscr{D}_{\parallel}k_{\parallel}{}^{2}+\mathscr{D}_{\perp}k_{\perp}{}^{2}-\sigma(\vartheta_{\mathbf{k}})|\mathbf{k}|^{3}+O(k^{4}), \quad (46)$$

$$\mathscr{D}_{\parallel}=\tfrac{1}{2}\sum_{x}V_{x0}x_{3}{}^{2},\qquad \mathscr{D}_{\perp}=\tfrac{1}{2}\sum_{x}V_{x0}x_{1}{}^{2}=\tfrac{1}{4}\sum_{x}V_{x0}(\mathbf{x}^{2}-x_{3}{}^{2}),$$

$$\sigma(\vartheta_{\mathbf{k}})=\frac{\pi^{2}}{12}\frac{c}{\Omega}\nu_{0}r_{0}{}^{6}\int_{0}^{2\pi}\frac{d\varphi_{\mathbf{n}}}{\pi}\int_{0}^{\pi}d\vartheta_{\mathbf{n}}\sin\vartheta_{\mathbf{n}}\chi(\vartheta_{\mathbf{n}})|\hat{\mathbf{k}}\mathbf{n}|^{3},$$

$$\hat{\mathbf{k}}=\frac{\mathbf{k}}{k}.$$

Here $\hat{\mathbf{k}}$ and $\mathbf{n}$ are unit vectors with polar angles $\vartheta_{\mathbf{k}},\varphi_{\mathbf{k}}$ and $\vartheta_{\mathbf{n}},\varphi_{\mathbf{n}}$, respectively. The calculation of expressions like (45) and (46) is presented in more detail in Ref. 43. In this manner we find

$$\hat{G}_{x0}{}^{0}(\beta t\gg 1, x^{2}\sim\bar{r}^{2})$$

$$=\Omega\left(\prod_{\alpha=1}^{3}(4\pi\mathscr{D}_{\alpha}t)^{-1/2}\right)\left(1+\frac{\varphi}{(\mu\beta t)^{1/2}}+O(1/\beta t)\right),$$

$$\hat{G}_{x0}{}^{0}(\beta t\gg 1, x^{2}\leqslant\mathscr{D}t) \quad (47)$$

$$=\Omega\left(\prod_{\alpha=1}^{3}\frac{\exp(-x_{\alpha}{}^{2}/(4\mathscr{D}_{\alpha}t))}{(4\pi\mathscr{D}_{\alpha}t)^{1/2}}\right)(1+O(1/(\beta t)^{1/2})),$$

where

$$\prod_{\alpha=1}^{3}(4\pi\mathscr{D}_{\alpha})=(\mu\beta)^{3}/c^{2},$$

$\mu=0.889$, and $\varphi=2.09$.

From this point on, the analysis is based on the idea that in this transport process the propagator $P_{xy}(t)$ adjusts rapidly (exponentially in the time) to the slowly varying solution in (27), (28) (this solution varies as a power law in t). From Eqs. (33) we find

$$P_{x\neq y}(t)=-\sum_z\int_0^t dt_1\,\hat{G}_{xz}(t-t_1)\int_0^{t_1}dt_2[N_{yz}{}^y(t_2)P_{zy}(t_1-t_2)$$

$$-N_{zy}{}^y(t_2)P_{yy}(t_1-t_2)]=P_{xy}^{(1)}+P_{xy}^{(2)}, \qquad (48)$$

where

$$P_{x\neq y}^{(1)}=-\sum_z\int_0^{t/2}dt_1\,\hat{G}_{xz}(t-t_1)\int_0^{t_1}dt_2[N_{yz}{}^y(t_2)P_{zy}(t_1-t_2)$$

$$-N_{zy}{}^y(t_2)P_{yy}(t_1-t_2)]=-\int_0^{t/2}dt_1\,\hat{G}_{xy}{}^0(t-t_1)\sum_z\int_0^{t_1}dt_2[N_{yz}{}^y(t_2)\cdot$$

$$\cdot P_{xy}(t_1-t_2)-N_{zy}{}^y(t_2)P_{yy}(t_1-t_2)](1+O(1/\beta t))$$

$$=-\int_0^{t/2}dt_1\,\hat{G}_{xy}{}^0(t-t_1)\dot{P}_{yy}(t_1)(1+O(1/\beta t))$$

$$=\hat{G}_{xy}{}^0(t)(1+O(1/\beta t)). \qquad (49)$$

Correspondingly,

$$P_{x\neq y}^{(2)}=-\int_{t/2}^t dt_1\sum_z\hat{G}_{xz}(t-t_1)\int_0^{t_1}dt_2[N_{yz}{}^y(t_2)P_{zy}(t_1-t_2)$$

$$-N_{zy}{}^y(t_2)P_{yy}(t_1-t_2)]=-\int_0^{t/2}dt_1\sum_z\hat{G}_{xz}(t_1)[\bar{N}_{yz}{}^yP_{zy}(t-t_1)$$

$$-\bar{N}_{zy}{}^yP_{yy}(t-t_1)](1+O(1/\beta t)). \qquad (50)$$

Here $\bar{N}_{xq}^y=N_{xq}^y(\lambda=0)$. It is clear from Eq. (33) that in the limit $\beta t\to\infty$ we have

$$\dot{P}_{yy}=-\sum_z(\bar{N}_{zy}{}^yP_{yy}-\bar{N}_{yz}{}^yP_{zy})(1+O(1/\beta t)). \qquad (51)$$

It follows that

$$P_{yy}(\beta t\to\infty)=\sum_z\bar{N}_{yz}{}^yP_{zy}\Big/\sum_z\bar{N}_{zy}{}^y(1+O(1/\beta t)). \qquad (52)$$

If we now substitute (52) into (50), then substitute (49) and (50) into (48), solve the resulting equation for $P_{x\neq y}(\beta t\to\infty)$ by an iterative method, and then substitute the result into (52), we find

$$P_{00}(\beta t\to\infty)=\frac{\xi}{c}\hat{G}_{00}{}^0(\beta t)(1+O(1/\beta t)), \qquad (53)$$

$$P_{x\neq 0}(\beta t\to\infty)=\hat{G}_{x0}{}^0(\beta t)(1+O(1/\beta t)). \qquad (54)$$

To find an approximation of P_{x0} for all t we consider the expressions

$$P_{00}(t)=Q_0{}^0(t)+(1-Q_0{}^0(t))\frac{\xi}{c}\Psi_{00}(t+\tau), \qquad (55)$$

$$P_{x0}(t)=(1-P_{00}(t))\Psi_{x0}(t+\tau_1),\quad x\neq 0, \qquad (56)$$

where $\Psi_{x0}(t)$ is defined in the same way as $\hat{G}_{x0}^0$ by (47), but without the correction terms $O(...)$. Here we have made use of the normalization condition (to within small quantities $\sim c$) and the condition that P_{x0} adjusts to a quasisteady asymptotic behavior. We have introduced delays τ and τ_1, which eliminate the nonphysical singularity of $\Psi_{x0}(t)$ at small t. It is natural to choose the delay τ in such a way that Eq. (55) is the same as the exact equation

$$P_{00}(t)=1-(\beta_1 t/(\xi+1))^{1/2}+O(\beta t)$$

at small values of βt. As a result we find[5]

$$P_{00}(t)=Q_0{}^0(t)+(1-Q_0{}^0(t))$$

$$\times\frac{\xi}{(\mu\beta(t+\tau))^{1/2}}\left(1+\frac{\varphi}{(\mu\beta(t+\tau))^{1/2}}\right). \qquad (57)$$

Corresponding to the ^{8}Li–^{6}Li system is the set of values[5]

$$\xi=3,\ \mu=0{,}889,\ \varphi=2{,}09,\ \mu\beta\tau=5{,}11,\ Q_0{}^0(t)=\exp(-(\beta_1 t)^{1/2}), \qquad (58)$$

while for the detrapping of excitons with isotropic ν_{xq} (Ref. 6) we would have

$$\xi=1,\ \mu=0{,}780,\ \varphi=1{,}93,\ \mu\beta\tau=3{,}61,\ \beta_1=\beta=\beta_0. \qquad (59)$$

The most important property of (57) is that at large βt we have

$$P_{00}(t)>\xi(\mu\beta t)^{-1/2},$$

while at small values of βt the opposite inequality obviously holds. The diffusive asymptotic behavior is thus reached through a damped oscillation, as was verified in the experiments of Ref. 6. The importance of this effect is that it indicates that we are approaching an asymptotic regime which can be used in experimental and numerical studies. The damped oscillation is a consequence of the long-range nature $\nu_{xz}\sim|\mathbf{x}-\mathbf{z}|^{-6}$ [which generates a term $-\sigma|\mathbf{k}|^3$ in expansion (46)], the sign variation of this expansion, and the fact that the expansion of $\hat{N}(\mathbf{k},\lambda)$ in small values of λ has no nonanalytic terms [it is actually sufficient that such terms be $O(\lambda\ln\lambda)$]. If the transition rates fall off more rapidly with the distance, e.g., with $s\geqslant 8$, a damped oscillation is again possible. For example, it exists in the case $c=1$, although in this case the preasymptotic term is on the order of $t^{-5/2}$ rather than t^{-2} as in (47) (Ref. 43).

Note that the coefficient σ in (46) is the same as the coefficient of $|\mathbf{k}|^3$ in the expansion of the Fourier transform of the average operator

$$\langle\bar{A}(\mathbf{k})\rangle_c=c(\nu(0)-\nu(\mathbf{k})),$$

$$\nu(\mathbf{k})=\sum_{\mathbf{x}}\exp(-i\mathbf{k}(\mathbf{x}-\mathbf{q}))\nu_{xq},\qquad \mathbf{q}\neq\mathbf{y}, \qquad (60)$$

and the same as the corresponding coefficient in the expansion in $\mathbf{k}$ of the quantity

$$\bar{A}(\mathbf{k},\mathbf{q})=\sum_{\mathbf{x}}\exp(-i\mathbf{k}(\mathbf{x}-\mathbf{q}))\bar{A}_{xq}. \qquad (61)$$

This coefficient stems from ν_{xq} as $|\mathbf{x}-\mathbf{q}|\to\infty$. It is the sum of a large number of small terms and accordingly undergoes a self-averaging. In Sec. 7 below we will show that σ, in contrast with $\mathscr{D}$, is not renormalized in the exact theory.

6. DIFFUSION COEFFICIENT

Gochanour *et al.*[28] have proposed a method for calculating the diffusion coefficient $\mathscr{D}$. In that method, they calculate not only the leading approximation $\mathscr{D}_{OGAF}$ but also (in contrast with other approaches, including some taken later) the first correction to it, $\Delta\mathscr{D}_{OGAF}$. This correction turns out to be small: $\Delta\mathscr{D}_{OGAF}/\mathscr{D}_{OGAF} = -0.11$.

The study by Gochanour *et al.*[28] is based on a calculation of the autocorrelation function of P_{00}, but Eq. (27) of Ref. 28 was not satisfied, and an exponential asymptotic behavior $P_{00}(t\to\infty)$ was found instead of a diffusion behavior.[6,48,49] For one- and two-dimensional systems, that method incorrectly predicts both $P_{00}(t\to\infty)$ and the Laplace transform of $P_{00}(\lambda\to 0)$, in terms of which $\mathscr{D}_{GAF}$ is expressed [in Ref. 28, the expression was $P_{00}(\lambda\to 0)\sim\lambda^{0}$, while in the diffusive behavior we would have $P_{00}(\lambda\to 0)\sim\lambda^{-1/2}$ with $d=1$ and $P_{00}(\lambda\to 0)\sim|\ln\lambda|$ with $d=2$]. Direct measurement of the exciton diffusion coefficient by Gomez-Jahn *et al.*[30] at $\beta t\sim 10^3$ by optical four-wave mixing, in which the observable was the Fourier transform of the propagator $P(\mathbf{k},t)$, led to a value smaller than the prediction of Ref. 28 by a factor of nearly 2. All these flaws of Ref. 28 were carried over to the later analysis in Ref. 29. The value of the diffusion coefficient in the semiphenomenological theory of Sec. 5 is the same as the value of $\mathscr{D}_{SL}$ in the Scher–Lax theory.[32] It is larger than the value found in Ref. 30, again by a factor of nearly 2.

Accordingly, we present below a new method for calculating the diffusion coefficient. This new method does not suffer from these flaws, and it leads to satisfactory agreement with the experiments of Ref. 30. We also show that a correct comparison of theory with those experiments requires consideration of the preasymptotic ($\sim|\mathbf{k}|^3$) term in an expansion of the type in (46).

We restrict the discussion to the limit of low concentrations, and we first consider the case of isotropic $\nu_{xz}=\nu_0 r_0^6/|\mathbf{x}-\mathbf{z}|^6$.

If we omit the condition $n_y=1$ in Eq. (20), we find the equation[36,50]

$$\dot{\tilde{G}}_{xy}=-\sum_z (n_z\nu_{zx}\tilde{G}_{xy}-n_x\nu_{xz}\tilde{G}_{zy}),\quad \tilde{G}_{xy}(t=0)=\delta_{xy}. \tag{62}$$

This equation describes a process in which the excitation is initially not at an impurity nucleus, as in (20) (where this circumstance was caused by the equality $n_y=1$), but at some arbitrary lattice site. The excitation subsequently moves to (nearest) donors and then migrates exclusively along them. The diffusion coefficients for the propagators $G=\langle\tilde{G}\rangle_c$ and P are therefore identical. Introducing a memory operator, and using the conservation law $\Sigma_x G_{xy}(t)=1$, we find

$$G(\lambda)=[\lambda+M(\lambda)]^{-1},\quad M(\lambda)=D(\lambda)-N(\lambda),\quad D_{xz}=\delta_{xz}D_0,$$
$$D_0=\sum_x N_{xz}. \tag{63}$$

The propagator $\hat{G}$, which we used back in Sec. 5, is a semiphenomenological approximation of G. This conclusion follows from a comparison of representation (32) with the corresponding representation for G.

From Eq. (62) it follows directly that[36,50]

$$n_y\tilde{G}_{xy}=n_y\tilde{P}_{xy}=n_y\tilde{P}_{xy}(n_y=1), \tag{64}$$
$$(1-n_x)\tilde{G}_{xy}=(1-n_x)\delta_{xy}\exp(-\sum_z n_z\nu_{zy}t). \tag{65}$$

In the small-c limit we find thus, as in (22),

$$G_{yy}(t)=G_{00}(t)=\left\langle\exp\left(-\sum_z n_z\nu_{zy}t\right)\right\rangle_c$$
$$=\exp(-(\beta_0 t)^{d/s}). \tag{66}$$

In our isotropic case, with $d=3$ and $s=6$, we find

$$\beta_0=\frac{16}{9}\pi^3\nu_0 r_0^6\frac{c^2}{\Omega^2}.$$

We evidently have $G_{00}(t)\sim c^0$ and $G_{x\neq y}(t)<c$, so we find, as in (30),

$$G_{00}(\lambda)=[\lambda+D_0(\lambda)]^{-1}. \tag{67}$$

The diffusion coefficient is $\mathscr{D}=\lim_{\lambda\to 0}\mathscr{D}(\lambda)$, where

$$\mathscr{D}(\lambda)=\frac{1}{2d}\sum_x \mathbf{x}^2 N_{x0}=\frac{\lambda^2}{2d}\sum_x \mathbf{x}^2 G_{x0}(\lambda). \tag{68}$$

Let us examine a concentration expansion of $G_{00}(\lambda)$ and $\mathscr{D}(\lambda)$. Within terms $\sim c^3$ inclusively, with $d=3$ and $s=6$, we find

$$G_{00}(\lambda)=\frac{1}{\lambda}(1-{}^1/_2\pi^{1/2}u+{}^1/_2u^2-{}^1/_8\pi^{1/2}u^3), \tag{69}$$
$$\mathscr{D}(\lambda)=\frac{\lambda}{6}\left(\frac{\beta_0}{\lambda}\right)^{5/6}d_0\bar{r}^2(1+d_1u+d_2u^2), \tag{70}$$

where $(u=(\beta_0/\lambda)^{1/2}$, $d_0=6^{2/3}/(4\pi^{1/2})=0.46573$, $d^1=0.01521$ and $d_2=-0.2036(8)$.

In deriving (69) we used explicit expression (66). Expression (70) was derived by the method described in Sec. 3. In the case at hand, it leads to the expansion

$$G_{x\neq y}(\lambda)=c\bar{b}_1^{(1)}(\mathbf{y},\mathbf{x},\lambda)+\sum_{m=1}^{\infty}\frac{c^{m+1}}{m!}\sum_{\mathbf{z}_1\ldots\mathbf{z}_m}\sum_{k=0}^{m}(-1)^{m-k}$$
$$\times C_m^k\bar{b}_1^{(k+1)}(\mathbf{y},\mathbf{x},\mathbf{z}_1\ldots\mathbf{z}_k,\lambda), \tag{71}$$

but now, in contrast with (23), $\bar{b}_1^{(k+1)}(\mathbf{y},\mathbf{x},\mathbf{z}_1,\ldots\mathbf{z}_k,\lambda)$ is the solution of Eq. (62) for an isolated $(k+1)$ cluster of impurity nuclei, at sites $\mathbf{x},\mathbf{z}_1,\ldots,\mathbf{z}_k$. The error in d_2 reflects the error in the calculation of the corresponding sextuple integral [after (71) is substituted into (68), after the sums are replaced by integrals, and after the spherical symmetry is taken into account, the first term reduces to a single integral, the second reduces to a triple integral, and $m=2$ corresponds to a sixtuple integral].

Working from expansion (69), using (67), we find an expansion for D_0:

$$D_0(\lambda)=\lambda(b_1u+b_2u^2+b_3u^3), \tag{72}$$

where $b_1=(1/2)\pi^{1/2}$, $b_2=\pi/4-1/2$, and $b_3=(1/8)\pi^{1/2}(\pi-3)$. When we now solve Eq. (69) for λ and find a relation $\lambda=\lambda_0(G_{00})$, again as an expansion in c[i.e., in $(\beta_0G_{00})^{1/2}$], substitute the result into (72), and expand it in a power series in $(\beta_0G_{00})^{n/2}$, we find, using (67),

an equation for the function $G_{00}(\lambda)$:

$$G_{00}=[\lambda+D_0(\lambda_0(G_{00}))]^{-1}. \tag{73}$$

Solving Eq. (73), we find that $\beta_0 G_{00}(\lambda=0)$ is equal to 1.273, 1.819, and 1.918 when one, two, and three terms, respectively, are retianed in the expansion of D_0 in $(\beta_0 G_{00})^{1/2}$. The exact value is 2. A corresponding calculation for $G_{00}(t)=\exp(-(\beta_0 t)^{\alpha})$ with $\alpha=3/10$ (i.e., for a quadrupole-quadrupole transport) leads to values 1.434, 3.821, and 8.467, respectively, for $\beta_0 G_{00}(\lambda=0)$. The exact value is $\beta_0 G_{00}(\lambda=0)=\Gamma(1+1/\alpha)=9.261$. In this case, however, it was necessary to carry out an additional restructuring of the expansion of D_0 in a continued fraction in $(\beta_0 G_{00})^{\alpha}$; otherwise, G_{00} would have turned out to be complex in the second step (more on this below). It can be seen from these examples that there is promise in the self-consistent method which was proposed in Ref. 28 for restructuring series in $u^n=(\beta_0/\lambda)^{n/2}$ in an expression having a finite limit as $\lambda\to 0$. We see that this method (or a simple modification of it) has a good numerical convergence, which can be monitored by observing the behavior of the successive approximations found through the use of the first terms of the concentration expansions.

The primary distinction from Ref. 28 so far is that a concentration expansion has been used (in accordance with the suggestions of Refs. 51, 52, and 29) instead of a diagram technique to derive expansions (69), (70), and (72). In addition, we have applied the method not to P_{00} as in Ref. 28 but to G_{00}, which is known exactly.

We have reported a test of this method in some exactly solvable samples, since neither the general conditions for the applicability of the method nor the results of any test of it were reported in either Ref. 28 or later papers.[29,52] Incidentally, it has been pointed out that there is an obvious limitation: If Eq. (69) is to have a single-valued solution for λ, the function $G_{00}(\lambda)$ must be monotonic.[29,52]

In actual calculations it is convenient to formulate this method in a different way. Assuming $\lambda=(1-y)/G_{00}$, and introducing the new variable y, we have $D_0=y/g_{00}$, and the equation

$$D_0(\lambda)=\lambda\sum_{k=1}^{\infty}b_k u^k, \qquad u=(\beta_0/\lambda)^{\alpha}, \tag{72a}$$

becomes

$$y=\sum_{k=1}^{\infty}b_k\zeta^k(1-y)^{1-\alpha k}, \qquad \zeta=(\beta_0 G_{00})^{\alpha}. \tag{74}$$

The *GAF* method[28] is now equivalent to the following procedure. We first solve Eq. (74) for y in the form

$$y=y_0(\zeta)=\sum_{k=1}^{\infty}g_k\zeta^k. \tag{75}$$

We can do this by iterating Eq. (74) and re-expanding the right side in a power series in ζ. Using $y=1-\lambda G_{00}$, we find then the equation

$$1-\lambda G_{00}=y_0(\zeta), \qquad \zeta=(\beta_0 G_{00})^{\alpha} \tag{76}$$

for the autocorrelation function G_{00}. In the aforementioned restructuring of the expansion into a continued fraction in $(\beta_0 G_{00})^{\alpha}$ the transformation under consideration was $(\lambda=0)$:

$$1=g_1\zeta+g_2\zeta^2+g_3\zeta^3\to g_1\zeta\left[1-\frac{g_2}{g_1}\zeta\left[1-\left(\frac{g_3}{g_2}-\frac{g_2}{g_1}\right)\zeta\right]^{-1}\right]^{-1}, \tag{77}$$

with $\alpha=3/10$, $g_1=0.8975$, $g_2=-0.2051$, and $g_3=0.0055$.

In the *GAF* method,[28] to calculate $\mathscr{D}$ we must now replace λ by $\lambda_0(G_{00})$ in expansion (70) and in this manner obtain an expansion of $\mathscr{D}(\lambda)$ in powers of ζ. The result can be put in the form

$$\mathscr{D}=\varkappa\mathscr{D}_0, \quad \mathscr{D}_0={}^1/_6\beta_0\bar{r}^2, \quad \bar{r}=(\Omega/c)^{1/3}. \tag{78}$$

When terms of up to order c, c^2, and c^3 are retained, the following respective values of $\varkappa$ are found: $\varkappa_1'=0.447$, $\varkappa_2'=0.346$, and $\varkappa_3'=0.512(1)$. The second correction turns out to be significantly larger than the first:

$$\frac{|\varkappa_3'-\varkappa_2'|}{\varkappa_3'}=1{,}28\gg\frac{|\varkappa_1'-\varkappa_2'|}{\varkappa_2'}=0{,}29.$$

We recall that the values $\varkappa_{1GAF}=0.355$ and $\varkappa_{2GAF}=0.315$ were found in Ref. 28 from an equation equivalent to Eq. (19), with terms on the order of c and c^2 in the expansions for $P_{\mathbf{x}0}$ and P_{00}. The difference $|\varkappa_{2GAF}-\varkappa_{1GAF}|/\varkappa_{2GAF}=0.13$ was small, but $\varkappa_{2GAF}$ was found to be larger by a factor of nearly 2 than the number corresponding to the experiments of Ref. 30 (more on this below).

A small value of the first correction to $\mathscr{D}$ in the *GAF* method thus does not lead to a rapid convergence of the results based on the first terms of the concentration expansion, either in the original version[28] or after a switch to Eq. (62) which eliminates some of the flaws mentioned above. The essential reason for this circumstance is that the functions $D_0(\lambda)$ and $\mathscr{D}(\lambda)$ are extremely different. The *GAF* method actually involves replacing λ by the function $\lambda_0(G_{00})$ in $\mathscr{D}(\lambda)$. That substitution is effective in a calculation of $D_0(\lambda)$, since $\lambda_0(G_{00})$ is calculated along with D_0. However, there are no grounds for believing that this substitution would be effective in a calculation of the entirely different function $\mathscr{D}(\lambda)$.

It is thus natural to attempt to calculate $\mathscr{D}$ by simply making use of the method which proved successful in the calculation of G_{00}. That method cannot be applied directly to expansion (70), since the latter is not in the standard form (69) that begins with a $1/\lambda$ term. Comparing (70) with the expression for $\mathscr{D}(\lambda)$ in the Scher–Lax theory[32] [cf. (46)],

$$\mathscr{D}_{SL}(\lambda)={}^1/_6\sum_{\mathbf{x}}\mathbf{x}^2\nu_{\mathbf{x}0}G_{00}(\lambda+\nu_{\mathbf{x}0})/G_{00}(\lambda), \tag{79}$$

and with the analogous representation for $\mathscr{D}$ in lowest order in the version of the outlined *GAF* theory,

$$\mathscr{D}'(\lambda)={}^1/_6\sum_{\mathbf{x}}\mathbf{x}^2\nu_{\mathbf{x}0}[\lambda+D_0(\lambda)+\nu_{\mathbf{x}0}]^{-1}/G_{00}(\lambda), \tag{80}$$

we can assume that the common factor of λ in (70) is the beginning of an expansion in c for $1/G_{00}(\lambda)$. We therefore rewrite $\mathscr{D}(\lambda)$ as

$$\mathscr{D}(\lambda)=\frac{d_0\bar{r}^2}{6G_{00}(\lambda)}\Phi(\lambda),$$

$$\Phi(\lambda)=\lambda G_{00}(\lambda)u^{5/6}(1+d_1u+d_2u+\ldots)=(\beta_0F)^{5/6}. \quad (81)$$

The expansion of F in c, i.e., in $u=(\beta_0/\lambda)^{1/2}$, is

$$F=\frac{1}{\lambda}(1+f_1u+f_2u^2+\ldots)=[\lambda+\lambda(h_1u+h_2u^2+\ldots)]^{-1}, \quad (82)$$

where $h_1=1.0452$ and $h_2=0.6620(10)$, and where we can apply to this expansion a method based on Eqs. (74)–(76). As a result, when we retain two and three terms from (82), we find $\varkappa_2=0.2163$ and $\varkappa_3=0.186(1)$, respectively. Using the same method to calculate $\mathscr{D}_{SL}$, we find values which are 1.318, 1.077, and 1.046 times the exact value $\varkappa_{SL}=0.3725$, when terms up to c^2,c^3, and c^4, respectively, are retained. These results demonstrate the good convergence of the method again in this case. From this comparison we can estimate the error of the method in the calculation of $\varkappa_3$ to be 10%, since in the case of $\varkappa_{SL}$ it is even smaller, although the relative change in $\varkappa_{SL}$ as we go from c^2 to c^3 is larger than the corresponding change in $\varkappa$.

The value found in this manner, $\varkappa=0.186$, is half the value of $\varkappa_{SL}$ and smaller by a factor of nearly 3 than the number $\varkappa_{GJ}=0.49$ which follows from the method proposed in Ref. 14 for describing spin diffusion.

To compare the number which we found with the experiments of Ref. 30, we need to allow for the circumstance that the function $\varkappa$ from (21) for the case of electric-dipole interactions is

$$\chi_{ij}={}^3/_2(\mathbf{d}_i\mathbf{d}_j-3(\mathbf{n}_{ij}\mathbf{d}_i)(\mathbf{n}_{ij}\mathbf{d}_j))^2,$$

where $\mathbf{d}_i$ are unit vectors along the directions of the transition dipole moments, $\mathbf{n}_{ij}=\mathbf{r}_{ij}/r_{ij}$, and an average $\langle\ldots\rangle_d$ must be carried out over the random isotropic distribution of vectors $\mathbf{d}_i$ (Refs. 53 and 54). This anisotropy was incorporated in Refs. 28, 30, and 53 by means of the substitution $c\to c\langle\chi_{ij}^{1/2}\rangle_d=0.845c$, as in the calculation of the Förster constant $\beta=\langle\chi^{1/2}\rangle_d^2\beta_0$. Here[30] $\mathscr{D}\to R\mathscr{D}=0.845^{4/3}\mathscr{D}=0.799\mathscr{D}$. Analysis of (79) and (80) shows, however, that the renormalization factor in these cases is $R=\langle\chi^{1/2}\rangle_d^{1/6}\langle\chi^{5/6}\rangle_d=0.899$. When we take this factor into account, we find that the value $\varkappa_3=0.186(1)$ above becomes $\varkappa_{theor}=0.167(1)$.

In the next section of this paper we show that

$$P(\mathbf{k},t)=\exp[-(D\mathbf{k}^2-\sigma|\mathbf{k}|^3)t](1+O(1/\beta t)),$$
$$\beta t\to\infty,\quad \mathscr{D}\mathbf{k}^2t\sim 1, \quad (83)$$

as in the semiphenomenological theory. In an analysis of the experimental results in Ref. 30, the value $\sigma=0$ was assumed, and the value $\varkappa_{exp}=0.147(15)$ was found as a result. Taking the correct value

$$\sigma=\frac{\pi^2}{12}\frac{c}{\Omega}v_0r_0^6=\frac{3}{64\pi}\beta_0\bar{r}^3$$

into account, we find $\varkappa_{exp}=0.168(17)$, which is the same as $\varkappa_{theor}$.

The results presented in this section of the paper seem to mean that this study has yielded the first regular method for calculating the diffusion coefficient for multipole transport in a disordered system, and a value has been found for $\varkappa$ which agrees with experimental results within the experimental error and the methodological error of the calculation.

7. PREASYMPTOTIC BEHAVIOR FOR DIPOLE TRANSPORT; THE COHERENT-MEDIUM METHOD

In this section of the paper we show that $G_{\mathbf{x}\neq\mathbf{y}}$ and $P_{\mathbf{x}\neq\mathbf{y}}$ are the same as $\mathbf{x}\to\mathbf{y}$, we show that the coefficient σ of $|\mathbf{k}|^3$ in (46) is exact, we find the relationship between the asymptotic behavior of $G(k,t)$ and $P(\mathbf{k},t)$ as $\beta t\to\infty$ with $\mathscr{D}k^2t\sim 1$, we find $G(k,t)$ for this case, and we refine the parameters of (57) using the value found for $\mathscr{D}$ in Sec. 6. As in Sec. 6, we consider only the small-c limit in this case.

Using the identity

$$\bar{G}(\lambda)=\bar{Q}(\lambda)+\bar{G}(\lambda)\bar{N}\bar{Q}(\lambda)=\bar{Q}(\lambda)+\bar{P}(\lambda)\bar{N}\bar{Q}(\lambda),$$
$$\bar{Q}_{\mathbf{xz}}=\delta_{\mathbf{xz}}\bar{Q}_{\mathbf{z}},\quad \bar{Q}_{\mathbf{z}}(\lambda)=\left(\lambda+\sum_{\mathbf{q}}\bar{N}_{\mathbf{qz}}\right)^{-1}, \quad (84)$$
$$\bar{N}_{\mathbf{xz}}=n_{\mathbf{x}}v_{\mathbf{xz}},$$

where we have used $\tilde{G}_{\mathbf{xz}}n_{\mathbf{z}}=\tilde{P}_{\mathbf{xz}}n_{\mathbf{z}}$, we introduce $\tilde{G}^0=\tilde{G}\tilde{N}\tilde{Q}$. We obviously have

$$G_{\mathbf{xy}}=\delta_{\mathbf{xy}}Q+G_{\mathbf{xy}}^0,\quad Q=\langle\bar{Q}_{\mathbf{x}}\rangle_c,\quad G^0=\langle\bar{G}^0\rangle_c.$$

From (64) and (65) we find $G^0_{\mathbf{yy}}=cP_{\mathbf{yy}}$. With $\mathbf{x}\neq\mathbf{y}$ we find

$$G_{\mathbf{xy}}^0(\lambda)=\langle\bar{G}_{\mathbf{xx}}n_{\mathbf{x}}v_{\mathbf{xy}}\bar{Q}_{\mathbf{y}}\rangle_c+\sum_{\mathbf{z}\neq\mathbf{x}}\langle\bar{G}_{\mathbf{xz}}n_{\mathbf{z}}v_{\mathbf{zy}}\bar{Q}_{\mathbf{y}}\rangle_c.$$

If $\mathbf{x}\to\mathbf{y}$ but $\mathbf{x}\neq\mathbf{y}$, then

$$\langle\bar{G}_{\mathbf{xx}}n_{\mathbf{x}}v_{\mathbf{xy}}\bar{Q}_{\mathbf{y}}\rangle_c=\left\langle\bar{G}_{\mathbf{xx}}n_{\mathbf{x}}v_{\mathbf{xy}}\left(\lambda+v_{\mathbf{xy}}+\sum_{\mathbf{q}\neq\mathbf{x}}n_{\mathbf{q}}v_{\mathbf{qy}}\right)^{-1}\right\rangle_c$$
$$\to\langle\bar{G}_{\mathbf{xx}}n_{\mathbf{x}}\rangle_c=cP_{00}(\lambda).$$

Using $\tilde{G}_{\mathbf{xz}}n_{\mathbf{z}}=\tilde{P}_{\mathbf{xz}}n_{\mathbf{z}}=n_{\mathbf{x}}\tilde{P}_{\mathbf{xz}}n_{\mathbf{z}}$, we find

$$\sum_{\mathbf{z}\neq\mathbf{x}}\langle\bar{G}_{\mathbf{xz}}n_{\mathbf{z}}v_{\mathbf{zy}}\bar{Q}_{\mathbf{y}}\rangle_c$$
$$=\sum_{\mathbf{z}\neq\mathbf{x}}\left\langle\bar{G}_{\mathbf{xz}}n_{\mathbf{z}}v_{\mathbf{zy}}\left(\lambda+v_{\mathbf{xy}}+v_{\mathbf{zy}}+\sum_{\mathbf{q}\neq\mathbf{x},\mathbf{z}}n_{\mathbf{q}}v_{\mathbf{qy}}\right)^{-1}\right\rangle_c.$$

As $\mathbf{x}\to\mathbf{y}$, this expression approaches z because of the term $v_{\mathbf{xy}}\to\infty$ in the denominator on the right. Consequently, in the low-concentration limit we find, as $\mathbf{x}\to\mathbf{y}$,

$$G_{\mathbf{x}\neq\mathbf{y}}(\lambda)=G_{\mathbf{yy}}^0(\lambda)=cP_{\mathbf{yy}}(\lambda). \quad (85)$$

To analyze the preasymptotic behavior of the autocorrelation function $P_{00}(t\to\infty)$, it is thus sufficient to analyze $G_{\mathbf{xy}}(t\to\infty)$. At small $\mathbf{k}$ and large t we find from (84) that the Fourier transform is

$$G(\mathbf{k},t)=Q(t)+\sum_{\mathbf{xz}}\int_0^t d\tau\exp(-i\mathbf{kx})\langle\tilde{P}_{\mathbf{xz}}(t-\tau)\bar{N}_{\mathbf{z}0}\bar{Q}_0(\tau)\rangle_c$$
$$=Q(t)+\sum_{\mathbf{xz}}\exp(-i\mathbf{kx})\left[\langle\tilde{P}_{\mathbf{xz}}(t)\bar{N}_{\mathbf{z}0}\tilde{Q}_{\mathbf{r}}(\lambda=0)\rangle_c\right.$$
$$\left.+\left\langle\dot{\tilde{P}}_{\mathbf{xz}}\bar{N}_{\mathbf{z}0}\frac{\partial\bar{Q}_0}{\partial\lambda}\Big|_{\lambda=0}\right\rangle_c+\ldots\right],$$

where we have made use of the circumstance that as t increases the function $\bar{Q}_0(t)$ falls off considerably more rapidly than $\tilde{P}_{\mathbf{xz}}(t)$ does. In the sum over $\mathbf{z}$ here, values $z\sim\bar{r}$ are important. The contribution of this region to the sum over $\mathbf{x}$ can be estimated by using $\tilde{P}_{\mathbf{xz}}<n_{\mathbf{x}}\tilde{P}_{zz}$. At large values of

$|\mathbf{x}-\mathbf{z}|$, the propagator $\tilde{P}_{\mathbf{xz}}$ depends only weakly on $\mathbf{z}$. Taking all these circumstances into account, we can write

$$G(\mathbf{k}, t)=P(\mathbf{k}, t)+O(P_{00}(t))+O(\mathscr{D}k^2\beta^{-1}P(\mathbf{k}, t)),$$
$$\beta t\to\infty, \quad \mathscr{D}k^2t\sim 1,$$

or

$$P(\mathbf{k}, t)=G(\mathbf{k}, t)+O((\beta t)^{-d/2})+O(\mathscr{D}k^2\beta^{-1}G(\mathbf{k}, t)), \quad (86)$$

where

$$G(\mathbf{k}, t)=\exp(-M(\mathbf{k}, \lambda=0)t)\,(1+O(1/\beta t)). \quad (87)$$

This relation will be proved below.

We now consider the asymptotic behavior of $G_{\mathbf{xy}}(\lambda)$ as $|\mathbf{x}-\mathbf{y}|\to\infty$. It follows directly from Eq. (62) that

$$\lambda\bar{G}_{\mathbf{xy}}(\lambda)=\delta_{\mathbf{xy}}-\sum_{\mathbf{z}}(n_{\mathbf{z}}\nu_{\mathbf{zx}}\bar{G}_{\mathbf{xy}}-n_{\mathbf{x}}\nu_{\mathbf{xz}}\bar{G}_{\mathbf{zy}})$$
$$=\delta_{\mathbf{xy}}-\sum_{\mathbf{z}}n_{\mathbf{z}}\nu_{\mathbf{zx}}\bar{G}_{\mathbf{xy}}+n_{\mathbf{x}}\nu_{\mathbf{xy}}\bar{G}_{\mathbf{yy}}+\sum_{\mathbf{z}\neq\mathbf{y}}n_{\mathbf{x}}n_{\mathbf{z}}\nu_{\mathbf{xz}}\bar{G}_{\mathbf{zy}}.$$

As $\mathbf{x}-\mathbf{y}\to\infty$, the region $\mathbf{z}\approx\mathbf{x}$, in which $G_{\mathbf{zy}}$ ($n_{\mathbf{z}}=1$) varies slowly, and also the region $\mathbf{z}\approx\mathbf{y}$, in which $\nu_{\mathbf{xz}}$ varies slowly, contribute substantially to the last sum. Taking these points into account, we write

$$\sum_{\mathbf{z}\neq\mathbf{y}}n_{\mathbf{x}}n_{\mathbf{z}}\nu_{\mathbf{xz}}\bar{G}_{\mathbf{zy}}=\sum_{\mathbf{z}\neq\mathbf{y}}n_{\mathbf{x}}n_{\mathbf{z}}\{\nu_{\mathbf{xz}}[\bar{G}_{\mathbf{xy}}+(\mathbf{z}-\mathbf{x})\nabla_{\mathbf{x}}\bar{G}_{\mathbf{xy}}(n_{\mathbf{x}}=1)+\ldots]$$
$$+\bar{G}_{\mathbf{zy}}[\nu_{\mathbf{xy}}+(\mathbf{z}-\mathbf{y})\nabla_{\mathbf{y}}\nu_{\mathbf{xy}}+\ldots]\},$$
$$\nabla_{\mathbf{x}}=\partial/\partial\mathbf{x}.$$

Taking the symmetry $\nu_{\mathbf{xz}}=\nu_{\mathbf{zx}}$ into account, we finally find

$$\lambda\bar{G}_{\mathbf{xy}}(|\mathbf{x}-\mathbf{y}|\to\infty)=n_{\mathbf{x}}\nu_{\mathbf{xy}}/\lambda-n_{\mathbf{y}}\nu_{\mathbf{xy}}\bar{G}_{\mathbf{xy}}+n_{\mathbf{x}}\nabla_{\mathbf{x}}\bar{G}_{\mathbf{xy}}(n_{\mathbf{x}}=1)$$
$$\times\sum_{\mathbf{z}\neq\mathbf{y}}n_{\mathbf{z}}\nu_{\mathbf{zx}}(\mathbf{z}-\mathbf{x})+n_{\mathbf{x}}\nabla_{\mathbf{y}}\nu_{\mathbf{xy}}\sum_{\mathbf{z}\neq\mathbf{y}}(\mathbf{z}-\mathbf{y})\bar{G}_{\mathbf{zy}}+\ldots. \quad (88)$$

The first term here is obviously the leading term, so we can write

$$G_{\mathbf{xy}}(\lambda)=c\lambda^{-2}\nu_{\mathbf{xy}}, \quad |\mathbf{x}-\mathbf{y}|\to\infty. \quad (89)$$

On the other hand, it follows in a completely analogous way from representation (63) that we have

$$G_{\mathbf{xy}}(\lambda)=\lambda^{-2}N_{\mathbf{xy}}(\lambda), \quad |\mathbf{x}-\mathbf{y}|\to\infty. \quad (90)$$

Consequently, for all λ we have

$$N_{\mathbf{xz}}(\lambda)=c\nu_{\mathbf{xz}}, \quad |\mathbf{x}-\mathbf{z}|\to\infty. \quad (91)$$

As was mentioned in Sec. 5, the term $\sim|\mathbf{k}|^3$ in the expansion of memory operator (46) is due entirely to the asymptotic behavior $\hat{N}_{\mathbf{x}0}\propto|\mathbf{x}|^{-6}$ as $\mathbf{x}\to\infty$. It thus follows from (91) that the coefficient σ is given exactly by the representation (46), while the leading term ($\sim k^2$) in the expansion is renormalized in the more accurate theory. In the isotropic case we thus have

$$M(\mathbf{k}, \lambda=0)=\mathscr{D}k^2-\sigma|\mathbf{k}|^3+o(k^3), \quad \mathscr{D}=\varkappa\mathscr{D}_0, \quad (92)$$

and from (46) we find

$$\sigma=\frac{\pi^2}{12}\frac{c}{\Omega}\nu_0 r_0^6=\frac{3}{64\pi}\beta_0\bar{r}^3.$$

A simple formula for joining the expansions of P_{00} at large and small values of βt was proposed in Sec. 5. Using the new value of the diffusion coefficient found in Sec. 6 and carrying out a refinement which makes the formula valid for terms up to $\sim\beta t$ under the condition $\beta t\ll 1$, we can write this formula in the form

$$P_{00}(t)=Q_1+(1-Q_1)\frac{\xi}{(\mu\beta(t+\tau))^{3/2}}\left(1+\frac{\varphi}{(\mu\beta(t+\tau))^{1/2}}\right),$$
$$Q_1=\exp(-(\beta_1 t)^{1/2}-\varepsilon\beta_1 t). \quad (93)$$

The parameters $z=\mu\beta\tau$ and ε are determined by the equations

$$\frac{\xi}{\Pi^{3/2}}\left(1+\frac{\varphi}{z^{1/2}}\right)=1-\frac{\varphi}{(\xi+1)^{1/2}},$$
$$\varepsilon=\frac{1}{2}\left(1-\frac{1}{(\xi+1)^{1/2}}+2\alpha(\xi+1)^{1/2}\right), \quad (94)$$

which follow from the circumstance that at small values of βt we have [see (25) and (26)]

$$P_{00}(t)=1-\left(\frac{\beta_1 t}{\xi+1}\right)^{1/2}+\left(\frac{1}{2(\xi+1)}-\alpha\right)\beta_1 t+O((\beta_1 t)^{3/2}). \quad (95)$$

In the isotropic model of exciton transport we would have $\alpha=\alpha_0=0.0064$. Using the value $\varkappa=0.186(1)=0.5\varkappa_{\mathrm{SL}}$ found in Sec. 6, and noting that μ is proportional to $\mathscr{D}$, we find

$$\beta_1=\beta=\beta_0, \quad \xi=1, \quad \mu=0{,}390,$$
$$\varphi=1{,}93, \quad \mu\beta\tau=3{,}61, \quad \varepsilon=0{,}155. \quad (96)$$

When the anisotropy in the exciton transport is taken into account through the introduction of a renormalization factor $R=0.899$ (Sec. 6), we find the new values $\beta=\beta_1=0.714\beta_0$ and $\mu=R\cdot 0.390/0.714=0.491$, without any change in φ or $\mu\beta\tau$.

New parameters of polarization transport in the ^{8}Li–^{6}Li spin system are found from (58) through a similar replacement of the old value $\mu=0.889$ by $\mu=0.5\cdot 0.889=0.445$. This replacement corresponds to the assumption that, as in the isotropic model, a more accurate value of the diffusion tensor is half its value in the Scher–Lax theory. As a result we find

$$\xi=3, \quad \mu=0{,}445, \quad \varphi=2{,}09, \quad \mu\beta\tau=5{,}11, \quad \varepsilon(\alpha=0{,}013)=0{,}276. \quad (97)$$

A previous attempt has been made[3] to use expression (93). With $\varepsilon=0.276$ and the phenomenological value $\mu=0.889$ which was used in Ref. 3, however, one degrades rather than improves the agreement with experiment. The parameter values in (97) lead to a more satisfactory agreement.

If expression (93) is to be accurate in the first two terms of the expansion of $P_{00}(\beta t\to\infty)$ (so a damped oscillation will exist), it is sufficient (as mentioned in Sec. 5) that the terms in $M(\mathbf{k},\lambda\to 0)$ which are not analytic in λ be $o(\lambda\ln\lambda)$. Alternatively, in the time domain, it is sufficient that $M(\mathbf{k},t\to\infty)=o(t^{-2})$. These conditions follow directly from the expression

$$G_{\mathbf{x}_0}(\lambda)=\frac{\Omega}{(2\pi)^3}\int_B d^3k\exp(i\mathbf{kx})\,[\lambda+M(\mathbf{k},\lambda)]^{-1},$$

and from the Tauber theorems which relate the asymptotic behavior as $\lambda\to 0$ and $t\to\infty$ (Ref. 43). Alternatively, they follow from the analysis corresponding to Eqs. (40)–(47). It is not a trivial matter to test these conditions.

In the analysis developed in Sec. 6, no terms which are not analytic in λ arise in $M(\mathbf{k},\lambda\to 0)$ at all, as in the original *GAF* theory. So far, however, that method is still in a primitive stage, and invoking it to answer this question would seem to be premature.

Another possible approach is to study the T-matrix expansions and the method of a coherent (or effective) medium for the propagator G. This method is also interesting as an alternative method for deriving G or P. An approach of this sort, which was developed by Kirkpatrick,[55] is a regular basis for solving random-walk problems in systems with disordered links between the nearest sites of a regular lattice (link problems). The conditions for the applicability of that approach are not yet clear. It is usually assumed that the coherent-medium method is good (i.e., yields satisfactory values of $\mathscr{D}$) if there are a large number z_e of effective nearest neighbors. As it turns out, however, the coherent-medium method leads to the exact value of the diffusion coefficient in one-dimensional systems which can be solved exactly in asymptotic limits, with $z_e=2$ (Ref. 41). After some modification, it is equally successful in the model of isotropic random hops[42] and in the version of that model[43] which simulates dipole transport. In these systems, the coherent-medium method leads to the correct long-term asymptotic behavior of the memory functions,[42] $M(t\to\infty)\sim t^{-1-d/2}$. Several attempts have been undertaken to generalize that approach to our problem (a site problem). Many of these attempts are reflected in a recent paper,[56] but they do not involve a construction based on T-matrix expansions, and they do not contain a regular approach.

To construct a T-matrix expansion of memory operator $M(\lambda)$, we rewrite Eq. (62) in the form

$$\lambda\bar{G}(\lambda)=1-\bar{A}\bar{G}(\lambda)=1-\Big[B(\lambda)-\sum_z F^z(\lambda)\Big]\bar{G}(\lambda),$$
$$F^z(\lambda)=B^z(\lambda)-n_zA^z,\qquad \sum_z B^z=B, \tag{98}$$
$$A_{\mathbf{xq}}{}^z=(\delta_{\mathbf{xq}}-\delta_{\mathbf{xz}})v_{\mathbf{zq}}.$$

Here we have introduced translationally invariant operators B and B^z which do not depend on the occupation numbers but which have not yet been determined. Multiplying the identity

$$\bar{G}(\lambda)=G_0{}^z+G_0{}^z\sum_{\mathbf{q}\neq\mathbf{z}}F^{\mathbf{q}}\bar{G},\qquad G_0{}^z=(\lambda+B-F^z)^{-1}, \tag{99}$$

by F^z from the left, and introducing

$$T^z=F^z\bar{G}=T_0{}^zG_0,\quad t^z=F^zG_0{}^z=t_0{}^zG_0,\quad G_0=(\lambda+B)^{-1}, \tag{100}$$

we find the equation

$$T^z=t^z+t^z\sum_{\mathbf{q}\neq\mathbf{z}}T^{\mathbf{q}}. \tag{101}$$

The equation is conveniently written and solved in matrix form:

$$\hat{T}=\hat{t}+\hat{t}\hat{\varphi}\hat{T},\quad \hat{T}=(1-\hat{t}\hat{\varphi})^{-1}\hat{t},$$
$$T^z=\sum_{\mathbf{q}}\hat{T}^{\mathbf{zq}}\qquad \hat{t}^{\mathbf{xq}}=\delta^{\mathbf{xq}}t^{\mathbf{q}},\qquad \hat{\varphi}^{\mathbf{xq}}=1-\delta^{\mathbf{xq}}. \tag{102}$$

We use

$$\bar{G}=G_0+G_0\sum_{\mathbf{z}}F^z\bar{G}=G_0+G_0\sum_{\mathbf{z}}T^z=G_0\Big(1+\sum_{\mathbf{zq}}\hat{T}^{\mathbf{zq}}\Big)$$
$$=G_0\Big[1+\frac{1}{N_s}\sum_{\mathbf{zq}}((1-\hat{t}\hat{\varphi})^{-1}\hat{t}\hat{\varphi})^{\mathbf{zq}}\Big]$$
$$=\frac{1}{N_s}G_0\sum_{\mathbf{zq}}[(1-\hat{t}\hat{\varphi})^{-1}]^{\mathbf{zq}}, \tag{103}$$

where N_s is the total number of sites in the crystal. Introducing the projection operator C of configurational averaging, we then find

$$G=\langle\bar{G}\rangle_c=\frac{1}{N_s}G_0\sum_{\mathbf{xz}}\{[1-(\bar{t}+v)\hat{\varphi}]^{-1}\}^{\mathbf{xz}}$$
$$=G_0(1-m)^{-1}=(\lambda+M(\lambda))^{-1},$$
$$\bar{t}=\langle\hat{t}\rangle_c,\quad v=\langle\hat{t}\hat{\varphi}\bar{C}(1-\hat{t}\hat{\varphi}\bar{C})^{-1}\hat{t}\rangle_c, \tag{104}$$
$$\bar{C}=1-C,\qquad m=\sum_{\mathbf{xz}}\Big(\frac{\bar{t}+v}{1+\bar{t}+v}\Big)^{\mathbf{xz}}.$$

In the limit of low concentrations we naturally have $B^z\sim c$, with

$$\sum_{\mathbf{z}}v^{\mathbf{xz}}\sim\langle t^{\mathbf{x}}\rangle_c\sim c,$$

and we are left with

$$m=\sum_{\mathbf{z}}\langle t^z\rangle+\sum_{\mathbf{xz}}v^{\mathbf{xz}},\qquad M=M_0+M_1, \tag{105}$$
$$M_0=c\sum_{\mathbf{z}}A^z(1+G_0A^z)^{-1},\qquad M_1=-\sum_{\mathbf{xz}}v^{\mathbf{xz}}G_0{}^{-1}.$$

Kirkpatrick's theory[55] is structurally equivalent to the substitution of an iterative solution of Eq. (101) into the second or third equation in (103). The construction of the T-matrix $t_0^z=t^zG_0^{-1}$, which describes a scattering by one site $\mathbf{z}$, not by one link as in Ref. 55, is based on our analysis, and we have written all the important equations in closed operator form.

The coherent-medium method corresponds to the condition $\langle t_0^r\rangle=0$ (or $B=M_0$), which reduces in the case $c\ll 1$ to the following equation for B:

$$B(\lambda)=c\sum_{\mathbf{z}}A^z[1+(\lambda+B)^{-1}A^z]=c\sum_{\mathbf{z}}\bar{A}^z(1+R_0\bar{A}^z)^{-1}, \tag{106}$$

$$\bar{A}^z=A^z[1+(\lambda+D_0)^{-1}A^z]^{-1},\quad R_0=(\lambda+D_0)^{-1}N_0(\lambda+B)^{-1},$$
$$B=D_0-N_0,$$

$$\bar{A}_{xq}{}^z=(\delta_{xq}-\delta_{xz})\bar{\nu}_{zq},\quad \bar{\nu}_{zq}=\nu_{zq}(1+\nu_{zq}(\lambda+D_0)^{-1})^{-1}.$$

Here we have introduced regularized transition rates $\bar{\nu}_{xq}$ ("regularized" here means smoothed over short distances), and $\mathbf{R}_{0xq}$ is the regular part (smooth at small values of $\mathbf{x}-\mathbf{q}$) of the propagator G_{0xq}.

Under the condition $\langle t_0^z\rangle=0$ the first nonvanishing contribution to M_1 is

$$M_1=-\sum_{z\neq q}\langle t_0{}^z G_0 t_0{}^q G_0 t_0{}^z G_0 t_0{}^q\rangle_c. \tag{107}$$

It is on the order of t^4, like the first correction in Kirkpatrick's method.

An advantage of the coherent-medium method is that its leading approximation can be found from the following graphic arguments, which bring out the relationship between the coherent-medium method and mean-field theories. If the operator

$$G_0=(\lambda+B)^{-1}=\left(\lambda+\sum_z B^z\right)^{-1}$$

is a good approximation of the exact propagator, then we would naturally expect that it would not change, on the average, when one of the "representatives of scatterers" B^z is replaced by an actual one, $n_z A^z$. The result is the equation

$$(\lambda+B)^{-1}=\langle(\lambda+B-B^z+n_zA^z)^{-1}\rangle_c, \tag{108}$$

which is equivalent to the equation $\langle t^z\rangle=0$.

Going back to the analysis of the asymptotic behavior $M(\mathbf{k},\lambda\to\infty)$, we find that the factor $(\lambda+B)^{-1}$ or R_0 on the right in (106) might be a source of dangerous terms in M_0 ("dangerous" in that they cannot be expanded in integer powers of λ and are no smaller than $\lambda\ln\lambda$ in order of magnitude). The reason is that the hypothesis that there are no dangerous terms in B leads to the presence of such terms in $((\lambda+B)^{-1})_{xq}$. However, it follows from the asymptotic behavior in (47), the localization of the functions $\bar{\nu}_{xq}$ in terms of $\mathbf{x}-\mathbf{q}$, and the presence of a conservation law, which can be expressed as

$$\sum_x A_{xq}{}^z=0,$$

that there are no dangerous terms in the product $R_0(\lambda)\bar{A}^z(\lambda)$. More precisely, the hypothesis that there are no such terms in B turns out to be noncontradictory. A term-by-term analysis of the expansion of M in $\hat{t}$ leads to a similar conclusion. That analysis is conveniently carried out in the Fourier representation with the help of the expression

$$(t_0{}^z)_{kp}=\exp(i(\mathbf{p}-\mathbf{k})\mathbf{z})\,[\mathbf{kp}f_1(\mathbf{k}^2,\mathbf{p}^2)+(\mathbf{kp})^2f_2(\mathbf{k}^2,\mathbf{p}^2,\mathbf{kp})$$
$$+\mathbf{k}^2f_3(\mathbf{k}^2,\mathbf{p}^2,\mathbf{kp})], \tag{109}$$

where $\mathbf{k}$ and $\mathbf{p}$ are quasimomenta, and the functions f_1, f_2 and f_3 are nonzero at vanishing values of their arguments. This analysis has also incorporated the circumstance that the range of the quasimomenta is finite and that the only source of damgerous terms is the divergence of the propagator $G_0\sim(\lambda+\mathscr{D}_0k^2)^{-1}$ at small $\mathbf{k}$ and λ, since in $t_0^z(\lambda)$ (as in M_0) there are no dangerous terms.

8. CONCLUSION

The basic content of this study consists of the theoretical arguments which can be made to support the assertion that a fundamental law of nature-the diffusion law-is also valid in disordered systems with dipole interactions. We have restricted the discussion to a comparison of simple systems describable by kinetic equation (2).

The derivation of Eq. (2) in Sec. 2 is based to a large extent on the modeling of the effect of the medium as an admixture of a stochastic process. Even this extremely limited problem does not yet have a completely microscopic solution, in agreement with Kubo's comment[57] regarding the unavoidability of stochastic elements in physical kinetics.

Aleksandrov's study[19] of the conditions for the applicability of a perturbation theory has been improved here, through a reformulation of that theory in a projection technique, through a generalization to multispin problems, and through the use of a realistic random process with smooth local-field trajectories and a slow diffusive decay of correlations. The model[22] used makes it possible to compare the form functions of the various important correlation functions with the results of a moment analysis of these functions. This model leads to qualitatively correct high-frequency asymptotic results, which are exponential according to the present understanding.[16,58] These properties are also of importance for finding a competent estimate of the relative roles played by the various multispin and multiquantum processes, a theory for which was derived in Refs. 59 and 60, among other places.

The slow decay of the correlations gives rise to a nonanalytic H_1 dependence of the Markov approximation for the memory operator of Eq. (8), but this event occurs at orders higher than H_1^4, and it does not affect our analysis of the conditions for the applicability of a perturbation theory. Yet another reason for the appearance of a nonanalytic H_1 dependence [at the order H_1^4, but for Eq. (17)] was pointed out in our discussion of Eqs. (17) and (18).

Analysis of the transformation of kinetic equation (14) to the considerably simpler form in (2) shows that Eq. (2), which describes the transport of the density of additive integrals of motion, and which is essentially a hydrodynamic equation, is also highly accurate in the kinetic stage of the evolution. One might expect that in the kinetic stage, with $p_0\sim 1$, a nonlinear equation for $\rho_i(m)$-the population of the state of spin i with magnetic quantum number m-would be preferable. At large t, that equation can be approximated well by a linear equation of the type in (2). Equations of this sort have been suggested in several places for describing processes of a spin-diffusion type, starting with the pioneering paper by Bloembergen[61] (see, for example, Refs. 15, 19, 62, and 63), but the accuracy of those equations appears to be unknown. Our deviation of balance equation (2) from control equation (14) is similar to Mori's method,[64] but it differs from the latter in that it is applied to a kinetic equation rather than to a Liouville equation, and it also differs in the type of projection operators used. Mori's method apparently does not lead to an expansion of memory function (18) in a continued fraction $1/z_e$.

The problem of deriving a solution of Eq. (2) averaged over a random impurity distribution is exceedingly complicated, no matter how simple it may look superficially. The

actual status of this problem, including its relationship with field theories and superfield theories, is seen when it is formulated in functional-integration terms.[50,51] One can find a corresponding analysis of some simpler but related problems in Refs. 65 and 66, among other places. In particular, it is only the major conceptual complexity of this problem which can explain the fact that papers have previously appeared with the assertion that the long-term asymptotic behavior (the diffusion coefficient or the conductivity, which is proportional to the diffusion coefficient, in problems of the hopping transport of electrons) is determined by the average operator $\langle \hat{A} \rangle_c$ (Refs. 67 and 68). That conclusion is erroneous, since it is reached without the use of a kinetic equation; it is reached by formally taking the limit of a weak impurity-impurity interaction, without a genuine evaluation of the dimensionless parameters or a selection of the important slow modes, for which a kinetic equation is usually formulated. The complexity of the problem is also seen in the circumstance that none of the existing methods for calculating the propagator in the limit $\beta t \to \infty$ and the diffusion tensor are beyond question, and experiments play a really major role at the present stage of development.

The concentration expansion presented in Sec. 3 is so far the only source of reliable information for realistic systems at moderate times. The result of Sec. 6, which is based on Ref. 69, shows that this expansion can also be used for a quantitative analyusis of the long-time asymptotic behavior. The methods used here are similar in spirit to the general ideas of the Padé approximations and to expansions in continued fractions.[70] They are effective when diverging or poorly converging series are restructured in such a way that the analytic properties of sums of these series can be taken into account correctly. An important point is that the theory of Sec. 6 can be generalized in an obvious way to the case of anisotropic $\nu_{\mathbf{xq}}$. The only difficulties which arise here stem fromthe increase in computer time required in the numerical calculations of the concentration expansion. An important advantage of the theory is that there is an *a posteriori* estimate of the acuracy of the diffusion-coefficient calculation on the basis of the first two approximations. However, this circumstance does not rule out the need for a theoretical proof (which we do not have) that the long-term asymptotic behavior is diffusive. In Sec. 4 we dwelt at length on a correct formulation of this hypothesis, since errors on this matter continue to be published to this day.

The semiphenomenological theory of Sec. 5 is based on the fact that by introducing an irreducible operator in the form in (30) it is not possible to make the memory a short-term memory. Here we are essentially contradicting the purpose for which this operator is usually introduced in many-body theory.[20,71] The derivation in Sec. 5 makes it possible to circumvent this problem and to derive equations which have a simple physical meaning. This theory, which was originally derived in Ref. 5, remains today the only theory which gives a uniform, qualitatively correct approximation of the propagator $p_{\mathbf{xy}}(t)$ for all c and t. Equations (38) and (46) generalize the Scher–Lax diffusion tensor $\mathscr{D}_{\mathrm{SL}}$ with small c to arbitrary c. Again in a more recent paper[29] we read the claim that a description has been found for all c and t. That paper presents a diffusion coefficient which is close in magnitude but does not satisfy condition (27). The agreement between the experimental results of Ref. 30 and the theory in Sec. 6 indicates that the diffusion tensor may have a value half that of $\mathscr{D}_{\mathrm{SL}}$ in the case $c \ll 1$ and also that we would also like the theory to be more accurate for all c.

We have paid close attention to the preasymptotic behavior of the functions $P_{00}(t)$ and $M(\mathbf{k},\lambda)$ and also Eqs. (57) and (93), since they are necessary for planning and analyzing experiments and also because the corresponding experimental results are important to the theory. At the moment we known of no approaches along which the memory function $M(k,t)$ would fall off as t^{-2} or more slowly. The absence of a damped oscillation would mean specifically that there is a long-term memory of this sort. Accordingly, measurements of damped oscillations would provide important information about possible directions for developing the theory further.

A contradictory picture is evolved at present. In the case of transport on a regular lattice, the damped oscillation is large,[43] larger in fact than the effect predicted by (57) and (59). In disordered systems, both experiment[6] and study of a system which can be solved exactly in an asymptotic limit[43] point to a completely significant effect. In the model of Ref. 43 the damped oscillation should grow with increasing disorder. This circumstance is seen in the fact that in the expansion

$$P_{00}(t\to\infty)=\frac{a_0}{t^{3/2}}\left(1+\frac{a_1}{t^{1/2}}-\frac{a_2}{t}+\frac{a_3}{t^{3/2}}+\ldots\right) \tag{110}$$

the quantity a_2 goes negative with increasing disorder, while a_3 increases. Analysis of the results of Ref. 6 in the region $\beta t \sim 25$ again reveals no contradiction of the possibility of a damped oscillation greater than that predicted by (57) and (59).

On the other hand, according to (93) and (96) (after we use a more realistic $\mathscr{D}$) the damped vibration is small, but in the theory presented above the coefficients a_2 and a_3 were not calculated. If they were known, we could refine Eq. (93), through the substitution

$$\tau \to \frac{\tau(\tau^{1/2}-\alpha_1 t^{1/2})}{(\tau^{1/2}+\alpha_2 t^{1/2})}$$

(for example), followed by a determination of ε, α_1, and α_2 from a_2 and a_3. There are certain indications in the calculations of Ref. 7 against the presence of a damped oscillation, but those indications are contradictory. On the whole, it is clear from the derivation of (57) and (93) that at present no claim can be made that these equations give more than a qualitative description of the damped oscillation and a formulation of questions which need to be solved experimentally or in a suitable numerical simulation. Experiments and numerical simulation will apparently have the last word on the matter. Experimental studies of the transport of nuclear polarization in the ^{8}Li–^{6}Li system will play a unique role in testing predictions of the microscopic theory, since calculations of the microscopic parameters in exciton systems are considerably less accurate.

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