Concentration expansion in the theory of excitation migration along a disordered lattice

F. S. Dzheparov, V. S. Smelov, and V. E. Shestopal
Institute of Theoretical and Experimental Physics

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An exact expansion of the Green's function with respect to concentration is constructed. It is used to describe the cross-relaxation of β-active ^{8}Li nuclei with ^{6}Li nuclei in LiF-type crystals.

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1. A wide class of physical phenomena, such as the cross-relaxation of β-active nuclei,[1–3] luminescence depolarization,[4–7] and jump conductivity,[8–11] is described by the kinetic equation

$$\frac{\partial}{\partial t}\widetilde{P}_{xy} = -\sum_z \left(n_z \nu_{zx} n_x \widetilde{P}_{xy} - n_x \nu_{xz} n_z \widetilde{P}_{zy}\right), \quad \widetilde{P}_{xy}(0) = n_y \delta_{xy}. \tag{1}$$

Here $\widetilde{P}_{xy}$ is the probability of finding an excitation at the point x of a d-dimensional, regular lattice, if it is at y at $t = 0$, ν_{xz} is the probability of a transition per unit time from z to x (generally, $\nu_{xz} \neq \nu_{zx}$)[1,2], and $n_x = 0$ or unity is the occupation number of the point x by an object capable of excitation, which we call a donor. The nonvanishing occupation numbers form a disordered donor lattice along which the excitation migrates. The observed clues are described in terms of the Green's function $P_{xy} = \langle \widetilde{P}_{xy} \rangle$ (the angular brackets denote an averaging over the occupation numbers). For specificity, below we use a power-law dependence of ν_{xz} on the distance $\nu_{xz} \sim (1/|x - z|^s)$.

If the donors form a regular lattice, then Eq. (1) [for $\nu_{xz} = \nu(x - z)$ can be solved by a Fourier transformation. Disorder causes a severe complication of the problem. Exact information about its solution shows that the lowest approximation of the concentration $c = \langle n_x \rangle$ can be obtained[1,3,4]; it has been proved[1] that for $c \ll 1$

$$P_{yy}(t) = c f(\bar{\nu} t); \quad P_{xy}(t) = c^2 g((x - y)/\bar{r}, \bar{\nu} t), \quad x \neq y, \tag{2}$$

where $\bar{\nu}$ is the probability of a transition at an average distance $\bar{r}$ between the donors, and the letters f and g are independent of time, coordinates, and concentration (theorem for the singular point f); the estimates of P_{xy} on the low side[1] and of P_{yy} on the high side[4] are poor; the Laplace transform of P_{yy} has been calculated numerically.[5] The asymptotic form of $P_{yy}(t \to \infty)$ has been obtained for one-dimensional systems with transport to the nearest neighbor.[7] However, the accuracy of the approximations used in estimating P_{xy} $(t \to \infty)$ (see Refs. 3, 8, and 10 and the papers cited in Refs. 1, 3, 8, 10, and 11), is not known.

In our paper we obtained a general concentration expansion of P_{xy} and the nu-

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merical results for $P_{yy}(t)$, which are accurate to c^3 inclusively, for the cross-relaxation of β-active nuclei.

2. Taking into account that $n_x P_{xy} = P_{xy}$, we represent the solution of Eq. (1) in the form

$$P_{xy} = \langle n_y \exp\left(-\sum_z A^z n_z t\right)\rangle_{xy} = c\,\langle \exp\left(-\sum_{z\neq y} A^z n_z t - A^y t\right)\rangle_{xy},$$

where $A^z_{xq} = \delta_{xq}\nu_{zx} - \delta_{zx}\nu_{xq}$. We introduce the symmetrization operator S [12] that acts on the operator A^z. The operators commute after the symbol S; therefore,

$$P_{xy} = c\,\langle S \exp\left(-\sum_{z\neq y} A^z n_z t - A^y t\right)\rangle_{xy} = c\,\{S \prod_{z\neq y}[1 + c(e^{-A^z t} - 1)] \times e^{-A^y t}\}_{xy} = c\,\{S \exp[-A^y t + \sum_{z\neq y} \ln(1 + c e^{-A^z t} - c)]\}_{xy}. \quad (3)$$

3. If $c \ll 1$, then, disregarding the expressions such as $c^k\bar{\nu}t$ in contrasts to $\bar{\nu}t$, which corresponds to retaining the first term of the expansion in c of the logarithm in Eq. (3), we obtain

$$P_{xy} = c\,\{S \exp[-A^y t + c\sum_z (e^{-A^z t} - 1)]\}_{xy} = c\,e^{-cN} K_{xy}(N). \quad (4)$$

Here the summation domain is restricted to a volume that contains N points (correspondingly $\Sigma_z \to \Sigma'_z$), and

$$K_{xy} = S\,[\exp(-A^y t + c\sum_z{}' e^{-A^z t})]_{xy} = \sum_{m=0}^{\infty} \frac{c^m}{m!} P^{(m)}_{xy}(N), \quad (5)$$

$$P^{(m)}_{xy}(N) = \sum_{z_1 \ldots z_m}{}' [\exp(-\sum_{i=1}^{m} A^{z_i} t - A^y t)]_{xy} = \sum_{z_1 \ldots z_m}{}' B^{(m)}_{xy}(y, z_1, \ldots, z_m).$$

It follows from the definition of the operators A^z that

$$B^{(m)}_{xy}(y, z_1, \ldots, z_m) = \sum_{i=0}^{m} b^{(m)}_i(z_0, z_1, \ldots, z_m)\,\delta_{x z_i},\ z_0 = y, \quad (6)$$

and $b^{(m)}_i$ satisfy the equation

$$\frac{\partial}{\partial t} b^{(m)}_i = -\sum_{j=0}^{m} (\nu_{ji} b^m_i - \nu_{ij} b^{(m)}_j),\ b^{(m)}_i(t=0) = \delta_{i0},\ \nu_{ij} = \nu_{z_i z_j}, \quad (7)$$

that describes the excitation migration along a system of $m+1$ donors located at the points $y, z_1, \ldots, z_m$. Substitution of Eq. (6) in Eq. (5) gives

$$P_{yy} = c\,e^{-cN} \sum_{m=0}^{\infty} \frac{c^m}{m!} \sum_{z_1 \ldots z_m} b^{(m)}_0(y, z_1, \ldots, z_m), \quad (8a)$$

$$P_{x \neq y} = c^2 e^{-cN} \sum_{m=0}^{\infty} \frac{c^m}{m!} \sum_{z_2 \ldots z_{m+1}}' b_1^{(m+1)}(y, x, z_2, \ldots, z_{m+1}).$$

(8b)

Finally, a complete expansion in c gives

$$P_{yy} = c \sum_{m=0}^{\infty} \frac{c^m}{m!} \sum_{z_1 \ldots z_m} \sum_{k=0}^{m} C_m^k \, b_0^{(k)}(y, z_1, \ldots, z_k)(-1)^{m-k},$$

(9a)

$$P_{x \neq y} = c^2 \sum_{m=0}^{\infty} \frac{c^m}{m!} \sum_{z_1 \ldots z_m} \sum_{k=0}^{m} C_m^k \, b_1^{(k+1)}(y, x, z_1, \ldots z_{(k)})(-1)^{m-k}$$

(9b)

These equations give a solution of the stated problem. Equation (7) in the Laplace representation $[\phi/\lambda = \int_0^\infty dt\, e^{-\lambda t}\phi(t)]$ can be converted into algebraic equations and $b_i^{(k)}(\lambda)$ are expressed in terms of the ratio of the determinants.

Equations (9) contain summations over all lattice points. But it is clear from the physical considerations that all identical summation variables z_i must be excluded, since each point can have only one donor.

4. For simplicity, Eqs. (9) were derived on the assumption that $c \ll 1$. In fact they are valid for all c. This can be proved by expanding in c the product in Eq. (3).

5. Our derivation was based on the kinetic equation (1); however, Eqs. (8) and (9) which are much more general, are valid for processes described by Hamiltonians such as $H = \Sigma_{xz} n_x n_z h(x,z)$, if $\tilde{P}_{xy} = n_x n_y \mathrm{Sp}\, Q_x Q_y(t)/\mathrm{Sp}1$, where h and Q_x are operators that are independent of n. But, in this case

$$b_i^{(k)} = \mathrm{Sp}\, Q_{z_i} U_{(k)}^{+} Q_y U_{(k)} / \mathrm{Sp}1, \quad U_{(k)} = \exp\left[-i \sum_{j,m=0}^{k} h(z_j, z_m)\, t\right].$$

The calculation of correlators in the theory of disordered paramagnets can be formulated in this way. To prove this, we need only to mention that any function $\phi(n_x) = \phi(0) + n_x[\phi(1) - \phi(0)]$. This makes it possible to represent, for example, P_{yy} in the form of a series in $n_y n_{z_1} \ldots n_{z_k} b_0^{(k)}$. All other actions are independent of the type of dynamics.

6. The method of moments[13] consists of expanding the correlators in a Taylor series in t. In the case $c \ll 1$ the first terms of this expansion describe the negligibly small part of $(\sim c)$ of the total change of P_{xy}. The expansions (8) and (9) gives an alternative method, in which the first terms describe changes of the correlators by an amount of the order of unity. We are reminded that if the process is described by Eq. (1), then the expansion actually occurs with respect to $ct^{d/s}$ [compare with Eq. (2)]. A calculation of the coefficients of c^n is only slightly more complicated that that of the moments. But the formulation of expansions (8) and (9) in the Fourier representation is

complicated. Therefore, a comparison of it with the experiment must be done in the time representation.

7. In the case of cross-relaxation of the β-active ^{8}Li nuclei with the ^{6}Li nuclei[1–3] in the LiF-type crystals

$$\nu_{xz} = \nu_0 r_0^6 (1 - 3\cos^2\theta_{xz})^2 |x - z|^{-6}, \quad x \neq y \neq z,$$

$$\nu_{xy} = \nu_{yx}/\xi = \nu_1 r_0^6 (1 - 3\cos^2\theta_{xy})^2 |x - y|^{-6}, \quad \xi = 3.$$

In the limit of small c the summation over z can be replaced by an integration, and to an accuracy of c^3

$$P_{yy}(t) = c\left\{1 - \left(\frac{\beta_1 t}{\xi + 1}\right)^{1/2} + \beta_1 t\left[\frac{\pi}{4(\xi + 1)} - \alpha f_1(\mu) - \mu\xi\alpha f_2(\mu)\right]\right\}. \tag{10}$$

Here, $\beta_1 = c^2\nu_1\pi^3 [16r_0^3/(9\Omega\sqrt{3})]^2$, $\alpha = \{\pi^3[16/(9\sqrt{3})]^2\}^{-1}$, $\mu = \nu_0/\nu_1$, and Ω is the volume of a unit cell. The positive functions $f_1(\mu)$ and $f_2(\mu)$ are shown in Fig. 1. The relation $P_{yy}(\mu)$ represents the dependence of cross-relaxation on the external magnetic field (Fig. 2).

8. In conclusion, we shall examine the methods of introducing the mass operator. If we assume (in the Laplace representation) that $\lambda P/c = 1 - M(\lambda)P/c$ and determine M from the known expansions (8) and (9), then M_{xz} will depend only on $x - z$, but it very difficult to give it a clear meaning. If, however, we use the Schwanzig projector method to formally define M and employ the averaging operator as the projector, then M_{xz} obtained from Eq. (1) will also depend on y.[3] In this case we can construct obvious approximation, like in Ref. 3, in order to introduce the singularity of the point y in the

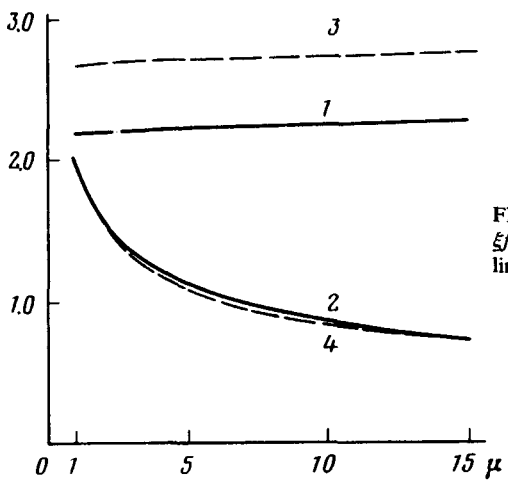

FIG. 1. Dependence of ξf_1 function (lines 1 and 3) and ξf_2 function (lines 2 and 4) on $\mu = \nu_0/\nu_1$. For solid lines $\xi = 1$.

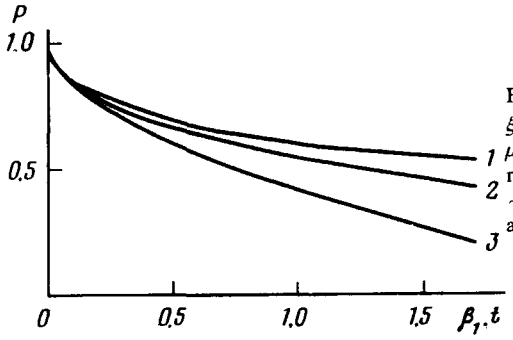

FIG. 2. Dependence of $P = P_{yy}(\beta_1 t)/P_{yy}(0)$ on $\beta_1 t$ for $\xi = 3$: 1 corresponds to $\mu = 1$; 2 corresponds to $\mu = 3$; and 3 corresponds to $\mu = 9$. At $\beta_1 t = 1.5$ the ratio of the term proportional to $\beta_1 t$ to the term $\sim\sqrt{\beta_1 t}$ in the equation is equal to -0.27, -0.12, and 0.20 for $\mu = 1$, 3, and 9, respectively.

Scher and Lax theory,[8] but the difficulties in describing P_{xy} for x close to y can be eliminated by obtaining information about the exact behavior of P_{xy} at small βt. Note that the initial condition for Eq. (1) in Ref. 11, which is specially devoted to obtaining the mass operator by using the Schwanzig projector method, is stated incorrectly and all the derived equations are incorrect. However, they are of some use since the relation $P_{yy} = \overline{P}_{yy} - (1 - c)$ is valid for P_{yy}, which is an experimentally observable value in many problems, and $P_{x\neq y} = \overline{P}_{xy}$, where $\overline{P}_{xy}$ is the average solution of Eq. (1) with the initial condition $\widetilde{P}_{xy}(0) = \delta_{xy}$, which was used in Ref. 11.

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