Cross relaxation in a dilute paramagnetic system

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Cross relaxation in a dilute paramagnetic spin system is considered. The investigation is based on configuration averaging (over all possible spatial positions of the particles) of the iteration solution of the kinetic equations for the Z component of the spin momentum. The terms proportional to the square of the concentration c are retained in the obtained averaged exact solution. The actual calculations were made for a quantitative description of the cross relaxation of the ^{8}Li–^{6}Li nuclei; which determines the depolarization of the β-active nuclei ^{8}Li in an external constant magnetic field in an LiF single crystal. It is shown that the contribution of the first term, proportional to c^3, is negligible at the ^{8}Li half-lives ($T\approx 0.8$ sec) compared with the retained terms. Some common regularities of excitation migration in disordered lattices are investigated.

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The 1957 discovery of parity nonconservation in radioactive β decay,[1] besides being of general physical significance, has led to the development of new experimental spectroscopic methods (see, e.g., Refs. 2–6), initially used to determine the characteristics (g-factors, quadrupole moments, etc.) of β-active nuclei. Later, however, β-active nuclei came into extensive use as unique probes for the investigation of condensed bodies.[6–15] Interest in research of this nature has increased particularly of late, as is evidenced by the ever growing number of studies.

The methods developed are typically combinations of procedures used in traditional magnetic resonance and observation methods used in nuclear physics. The latter has greatly contributed to the unusually high sensitivity of the procedures. The purpose of the present paper is to describe quantitatively experiments on the observation of depolarization of β-active ^{8}Li nuclei in single-crystal LiF and to determine some general regularities of the process of migration of excitation in disordered systems. We shall dwell briefly on the experimental setup (a detailed description of the experiment and its detailed comparison with the theory will be made subsequently; see also Ref. 15). The LiF single crystal was bombarded by a beam of polarized thermal neutrons. Capture of a polarized neutron by the ^{7}Li nucleus produced a polarized beta-active ^{8}Li nucleus with a half-life 0.8 sec. At room temperature, the depolarization of ^{8}Li nuclei in a constant magnetic field applied parallel to the initial polarization and of strength $150\ \text{Oe} \leqslant H \leqslant 3000$ Oe is the result of cross relaxation with the stable isotope ^{6}Li, whose concentration in the sample was 3.5%. This relaxation mechanism was predominant in so wide a range of fields because the g-factors were close: $g(^8\text{Li}) = 0.8265$, $g(^6\text{Li}) = 0.8220$. The mutual influence of the ^{8}Li nuclei was negligible because of their small total number, $\sim 10^8$, in the sample. The dependence of the polarization of the ^{8}Li nuclei on the time was determined from the angular anisotropy of the β decay. Thus, to describe the experiment it is necessary to solve the problem of excitation transfer (cross relaxation) in a dilute paramagnetic system. Very similar problems are encountered in electron paramagnetic resonance[15–18] (except that they are made complicated there by spectral diffusion), in the study of the effect of resonant dipole-dipole interaction between the donors on the luminescence quenching process,[19–21] in problems of hopping conduction,[20–24] and others. From the mathematical point of view these problems consist of constructing a solution, averaged over all possible spatial positions of the particles, of kinetic equations that are perfectly analogous to the fundamental equation of our paper, which will be formulated below. Notwithstanding the dozens of papers dealing with this problem since Forster's pioneering work,[25] it is still far from solved, as noted, e.g., in Ref. 20.

In this paper we average the iteration solution of the kinetic equation at arbitrary concentration, and sum partially the obtained series. An estimate of the first discarded term has shown that at times of the order of the half-life of ^{8}Li it is negligible compared with the retained terms. The results of the theory agree with experiment. Some general properties of diffusion in a disordered lattice are discussed and the presented approach is compared with earlier studies.

When a polarized ^{8}Li nucleus is placed in a crystal it is subjected to a dipole-dipole interaction with the nuclei ^{19}F, ^{7}Li, and ^{6}Li. The Hamiltonian of the nuclear spin subsystem of the crystal is[26, 27]

$$\mathscr{H}=\mathscr{H}_z+\mathscr{H}_1+\mathscr{H}_2+\mathscr{H}_3+\mathscr{H}_4+\mathscr{H}_5, \tag{1}$$

$\mathscr{H}_z$ is the Zeeman interaction of the nuclei with the external magnetic field, $\mathscr{H}_1$ and $\mathscr{H}_2$ are the secular parts of the dipole-dipole interaction of the ^{8}Li nuclei with the ^{19}F and ^{7}Li nuclei; $\mathscr{H}_3$ and $\mathscr{H}_4$ are the interactions of the ^{8}Li and ^{6}Li nuclei and of the ^{6}Li nuclei with one another. Finally, $\mathscr{H}_5$ includes the secular part of the dipole-dipole interaction of the ^{19}F with each other and of the ^{7}Li with each other, as well as the interaction between ^{19}F and ^{7}Li. The presence of the terms $\mathscr{H}_1$, $\mathscr{H}_2$, and $\mathscr{H}_5$ in the Hamiltonian causes broadening of the NMR spectrum of ^{8}Li and ^{6}Li, whereas the terms $\mathscr{H}_3$ and $\mathscr{H}_4$ lead to cross relaxation of the nuclei ^{8}Li and ^{8}Li and to further dissipation of the polarization in the magnetically dilute system of the ^{6}Li spins. The nuclei ^{8}Li and ^{6}Li are located in fluctuating local magnetic fields determined by the Hamiltonian $\mathscr{H}_1+\mathscr{H}_2+\mathscr{H}_5$. The rate

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of these fluctuations is of the order of the rate of the flip-flop processes in the ^{8}Li–^{6}Li subsystem. The ^{8}Li–^{6}Li and ^{6}Li–^{6}Li cross relaxations can therefore be described by the standard perturbation theory for the density matrix,[26–28] which leads to the kinetic equation

$$\frac{dP_k}{dt}=-\sum_{l\neq k}(\nu_{lk}P_k-\nu_{kl}P_l). \tag{2}$$

The external magnetic field H is assumed here applied along the Z axis. P_k is the average value of the Z component of the k-th spin from the ^{8}Li–^{6}Li subsystem, ν_{ik} is the probability of cross relaxation of spin k under the influence of spin l:

$$\nu_{lk}=\frac{8\pi}{3}S_l(S_l+1)\left(\frac{g_lg_k\beta_n^2}{4\hbar}\right)^2 g(\omega_l-\omega_k)\frac{(1-3\cos^2\theta_{lk})^2}{r_{lk}^6}, \tag{3}$$

S_l, g_l, and ω_l are respectively the magnitude, g-factor, and Zeeman frequency of the l-th spin, β_n is the nuclear magneton, $g(\omega)$ is the cross-relaxation form function normalized to unity, $\mathbf{r}_{lk}$ is the vector that separates the spins l and k, and θ_{lk} is the angle between H and $\mathbf{r}_{lk}$. At $S_l=1/2$ formula (3) coincides with the known Bloembergen formula.[29] For the sake of argument it can be assumed that $l=0$ corresponds to the spin $S_0=2$ of ^{8}Li, and $l\neq 0$ to the spin $S_l=1$ of ^{6}Li. We shall hereafter, however, not specify the value of the spin, and consider by the same token the more general case.

It must be noted that the function $g(\omega_l-\omega_k)$ depends not only on $\omega_l-\omega_k$ but also on the concrete arrangement of all the ^{8}Li–^{6}Li spins. At a low concentration of ^{6}Li, however, this dependence can be neglected, and at a high concentration a good approximation is obtained by substituting in (3) the value of $g(\omega_l-\omega_k)$ averaged over the configurations of the spins of ^{8}Li and ^{6}Li.

Equation (2) is exact within the framework of the assumptions formulated above but only if $S_k=1/2$. In the case of arbitrary spins it can be obtained by assuming the diagonal part ρ_D of the spin density matrix to have the so-called local-equilibrium form

$$\rho_D(t)=\Big[\prod_k(2S_k+1)\Big]^{-1}\Big[1+\sum_l\frac{3P_l(t)}{S_l(S_l+1)}\hat{S}_l^z\Big], \tag{2a}$$

where $\hat{S}_l^z$ is the operator of the Z-component of the l-th spin. This is precisely the form of ρ_D at $t=0$, $P_0\sim 1$, and $P_l=-\beta_n g_l(S_l+1)H/3T\approx 0$, where T is the temperature in energy units, since the nuclei produced in (n,γ) reactions on thermal polarized neutrons have only dipole polarization.[2] In addition, the representation (2a) usually serves as a basis for the description at large t (see, e.g., Ref. 27). Estimates show that dispensing with formula (2a) leads to corrections amounting to several per cent of the solution of (2). We note that Eq. (2) can lead to incorrect results in the description of systems for which the initial state can not be represented in the form (2a), or else call for allowance for effects due to the finite propagation velocity of the excitations in the dipole-dipole reservoir.

Introducing the angular-momentum density p_x and the density of the occupation number n_x

$$p_x=\sum_k\delta(\mathbf{x}-\mathbf{x}_k)P_k,\quad n_x=\sum_k\delta(\mathbf{x}-\mathbf{x}_k),$$

$$n_xp_x=p_x,\quad \delta(\mathbf{x})=\begin{cases}1, & \mathbf{x}=0\\ 0, & \mathbf{x}\neq 0\end{cases}$$

(the summation is over all the occupied sites), we can identically rewrite (2) in the form

$$\frac{dp_x}{dt}=-\sum_{z\neq x}(\nu_{zx}n_zp_x-\nu_{xz}n_xp_z). \tag{4}$$

The index z runs here over all the lattice sites, and n_z differs from zero if the site $\mathbf{z}$ is occupied.

Defining the Green's function $\tilde{p}_{xy}(t)$ of Eq. (4) as the solution that satisfies at $t=0$ the condition

$$\tilde{p}_x(t=0)=n_y\delta_{xy} \tag{5}$$

(δ_{xy} is the Kronecker symbol), we rewrite (4) in integral form

$$\tilde{p}_{xy}(t)=n_y\delta_{xy}\exp\Big(-\sum_z n_z\nu_{zx}t\Big)$$

$$+\int_0^t dt_1\exp\Big[-\sum_z n_z\nu_{zx}(t-t_1)\Big]\sum_q n_x\nu_{xq}\tilde{p}_{qy}(t_1). \tag{6}$$

Here and below we put $\nu_{xx}=0$; $\tilde{p}_{xy}(t)$ is the Z-component of the angular momentum at the point $\mathbf{x}$ if the entire angular momentum of the system was concentrated at the site y. For the angular momentum to be located at the point y, this point must be occupied by the nucleus; this is taken into account by the factor n_y in (5). In the ^{8}Li–^{6}Li system the site y is occupied by the spin of ^{8}Li, and the spins of ^{6}Li can be situated in the remaining sites.

After introducing the matrix

$$G_{xq}=n_x\delta_{xq}G_x(t),\quad G_x(t)=\exp\Big[-\sum_z n_z\nu_{zx}t\Big]$$

Eq. (6) reduces to

$$\hat{p}(t)=\hat{G}(t)+\int_0^t\hat{G}(t-t_1)\hat{\nu}\hat{p}(t_1)dt_1. \tag{7}$$

The last equation is convenient for obtaining the iteration solution

$$\hat{p}(t)=\hat{G}(t)+\sum_{n=1}^{\infty}\int_0^t dt_1\ldots\int_0^{t_{n-1}}dt_n\hat{G}(t-t_1)\hat{\nu}\hat{G}(t_1-t_2)\ldots\hat{\nu}\hat{G}(t_n). \tag{8}$$

The problem consists now of finding the matrix element $p_{xy}(t)$ averaged over the random positions of the spins, i.e., over all possible sets of the values of the occupation numbers $\{n_x\}$ at arbitrary $\mathbf{x}$.

To illustrate the employed technique, let us examine in detail the averaging of the zeroth term in (8). We assume that the occupation numbers of the different sites do not correlate, i.e., the probability of filling the site $\mathbf{x}$ does not depend on whether or not the site $\mathbf{z}$ is occupied at $\mathbf{z}\neq\mathbf{x}$, but not more than one spin can be located in one site. Therefore, assuming that $\langle n_x\rangle=c$ (c is the concentration of the occupied sites), we have, for example $\langle n_xn_y\rangle=c^2$ at $\mathbf{x}\neq\mathbf{y}$ and $\langle n_xn_y\rangle=c$ if $\mathbf{x}=\mathbf{y}$. In addition, $\exp(-n_z\nu_{zx}t)\equiv 1+n_z\{\exp(-\nu_{zx}t)-1\}$. The function

$$G_x(t)=\exp\Big(-\sum_{z\neq x}n_z\nu_{zx}t\Big)$$

contains a sum only over different sites, whose occupations do not correlate, therefore

$$\left\langle \exp\left(-\sum_{z\neq x} n_z\nu_{zx}t\right)\right\rangle=\left\langle\prod_{z\neq x}\exp(-n_z\nu_{zx}t)\right\rangle=\prod_{z\neq x}\langle\exp(-n_z\nu_{zx}t)\rangle$$
$$=\prod_{z\neq x}\langle 1+n_z[\exp(-\nu_{zx}t)-1]\rangle=\prod_{z\neq x}\{1+c[\exp(-\nu_{zx}t)-1]\}$$
$$=\exp\left\{\sum_{z\neq x}\ln[1+c(\exp(-\nu_{zx}t)-1)]\right\}. \qquad (9)$$

The last equation, previously derived in Ref. 19, is convenient on going to the limit of low concentration; this is done by expanding the logarithm in powers of c. Introducing $\tau_k=t_k-t_{k+1}, t_0=t, t_{n+1}=0$, we obtain for the integrand $A^{(n)}$ in the n-th term of the series in (8)

$$A_{xy}^{(n)}=\langle\hat{G}(\tau_0)\hat{\nu}\hat{G}(\tau_1)\ldots\hat{\nu}\hat{G}(\tau_n)\rangle_{xy}$$
$$=\sum_{q_1\ldots q_{n-1}}\langle G_x(\tau_0)n_x\nu_{xq_1}G_{q_1}(\tau_1)n_{q_1}\nu_{q_1q_2}\ldots\nu_{q_{n-1}y}G_y(\tau_n)n_y\rangle$$
$$=\sum_{q_1\ldots q_{n-1}}\nu_{xq_1}\nu_{q_1q_2}\ldots\nu_{q_{n-1}y}\left\langle n_xn_{q_1}\ldots n_{q_{n-1}}n_y\right.$$
$$\left.\times\exp\left[-\sum_z(\nu_{zx}\tau_0+\nu_{zq_1}\tau_1+\ldots+\nu_{zy}\tau_n)n_z\right]\right\rangle. \qquad (10)$$

It follows from this, taking into account the equality $n_xf(n_x)=n_xf(1)$, which is valid for any bounded $f(n_x)$, that

$$A_{xy}^{(n)}=\sum_{q_1\ldots q_{n-1}}\left(\prod_{i=0}^{n-1}\nu_{q_iq_{i+1}}\right)\left\langle\prod_{j=0}^{n}n_{q_j}\right\rangle\exp\left(-\sum_{k,l=0}^{n}{}'\nu_{q_kq_l}\tau_l\right)$$
$$\times\left\langle\exp\left(-\sum_{z\neq q_0,q_1\ldots q_n}\sum_{l=0}^{n}n_z\nu_{zq_l}\tau_l\right)\right\rangle. \qquad (11)$$

Here $q_0=x$ and $q_n=y$. The quantity

$$\left\langle\prod_{j=0}n_{q_j}\right\rangle=c^N,$$

is the number of different vectors in the set $\{\mathbf{q}_0, \mathbf{q}_1\ldots\mathbf{q}_n\}$ (the calculation of mean values of this type is described in greater detail in Ref. 30), $\sum'_{k,l=0}$ means that the sum over k includes only these different $\mathbf{q}_k$, whereas the sum over l is taken in the usual manner. The exponential in (11) is calculated in exactly the same way as in (9). As a result, the averaged solution of (4) takes the form

$$p_{xy}(t)=c\delta_{xy}\prod_z\{1+c[\exp(-\nu_{zx}t)-1]\}$$
$$+\sum_{n=1}^{\infty}\int_0^t dt_1\ldots\int_0^{t_{n-1}}dt_n\sum_{q_1\ldots q_{n-1}}\left(\prod_{i=0}^{n-1}\nu_{q_iq_{i+1}}\right)\left\langle\prod_{j=0}^{n}n_{q_j}\right\rangle$$
$$\times\exp\left(-\sum_{k,l=0}^{n}{}'\nu_{q_kq_l}\tau_l\right)\prod_{z\neq q_0,q_1\ldots q_n}\left\{1+c\left[\exp\left(-\sum_{l=0}^{n}\nu_{zq_l}\tau_l\right)-1\right]\right\}. \qquad (12)$$
$$q_0=x,\ q_n=y,\ \tau_k=t_k-t_{k+1},\ t_0=t,\ t_{n+1}=0.$$

We have denoted here by p_{xy} the quantity $\langle\tilde{p}_{xy}\rangle$. Formula (12) is exact. We call attention to the normalization $p_{xy}(0)=\delta_{yx}c$. The factor c stems here from the fact that we have averaged expression (8) with respect to n_y on a par with all other $n_{x\neq y}$. The calculation could have been made under the assumption $n_y\equiv 1$, which would correspond more accuracy to the gist of the process in the ^{8}Li-^{6}Li system, but we would then obviously obtain a result that differs from (12) only by an inessential common factor c, but would have lost greatly in the symmetry of the exposition.

The experimentally observed quantity is p_{xy}, i.e., the Z-component of the angular momentum of the β-active nucleus ^{8}Li. Let us continue its calculation, separating from the series (12) all the terms proportional to c^2 (only two unequal indices, q and y, for any n). In this approximation,

$$p_{yy}(t)=c\prod_z\{1+c[\exp(-\nu_{zy}t)-1]\}$$
$$+c^2\sum_{n=1}^{\infty}\int_0^t dt_1\ldots\int_0^{t_{2n-1}}dt_{2n}\sum_q(\nu_{yq}\nu_{qy})^n\exp[-\nu_{yq}\tau-\nu_{qy}(t-\tau)]$$
$$\times\prod_{z\neq q,y}\{1+c[\exp(-\nu_{zq}\tau-\nu_{zy}(t-\tau))-1]\}, \qquad (13)$$
$$\tau=t_1-t_2+t_3-t_4+\ldots-t_{2n}.$$

The integrand in (13) depends, in any order in n, only on two temporal arguments, t and τ, and there is no integration with respect to t. Rearranging the integration limits, we have after obvious intermediate transformations

$$p_{yy}(t)=c\prod_z\{1+c[\exp(-\nu_{zy}t)-1]\}+c^2\sum_{n=1}^{\infty}\int_0^t dt_1\sum_q(\nu_{yq}\nu_{qy})^n$$
$$\times\frac{t_1^{n-1}(t-t_1)^n}{n!(n-1)!}\exp[-\nu_{yq}t_1-\nu_{qy}(t-t_1)]\prod_{z\neq q,y}\{1+c[\exp(-\nu_{zq}t_1-\nu_{zy}(t-t_1))-1]\}.$$

Putting $\varepsilon=\nu_{yq}\nu_{qy}t_1(t-t_1)$ we obtain ultimately

$$p_{yy}(t)=c\prod_z\{1+c[\exp(-\nu_{zy}t)-1]\}$$
$$+c^2\int_0^t dt_1\sum_q\frac{\varepsilon^{1/2}}{t_1}\exp[-\nu_{yq}t_1-\nu_{qy}(t-t_1)]I_1(2\varepsilon^{1/2})$$
$$\times\prod_{z\neq q,y}\{1+c[\exp(-\nu_{zq}t_1-\nu_{zy}(t-t_1))-1]\}. \qquad (14)$$

In the same approximation,

$$p_{x\neq y}(t)=c^2\int_0^t dt_1\nu_{xy}I_0(2\varepsilon^{1/2})\exp[-\nu_{xy}t_1-\nu_{yx}(t-t_1)]$$
$$\times\prod_{z\neq x,y}\{1+c[\exp(-\nu_{zy}t_1-\nu_{zx}(t-t_1))-1]\}. \qquad (14a)$$

Here $I_0(x)$ and $I_1(x)$ are Bessel functions of imaginary argument. Formula (14) was used by us for numerical calculations with the BÉSM-6 computer.

An estimate of the upper bound of the first term, proportional to c^3, has shown that the contribution made to $p_{yy}(t)$ in times of the order of the half-life of ^{8}Li is negligible compared with the contributions of the retained terms. The figure shows plots of $p_{yy}(t)$ and of

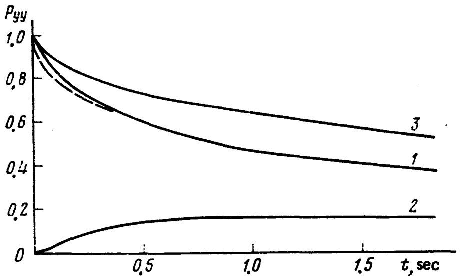

FIG. 1. Plot of the function $p_{yy}(t)$ calculated by formula (14). Line 1 represents the contribution of the first term of (14), the dashed-line shows this term calculated in the Forster approximation. Line 2 is the contribution of the second term of (14). Line 3 represents p_{yy}. An external field $H=200$ Oe is applied along [100].

both terms of (14), as well as plots of the first term of (14) as calculated in Forster's approximation, i.e., in the limit of low concentration and with the summation replaced by integration. We used in the calculation the numerical parameters corresponding to the system ^{8}Li–^{6}Li at small H. We note that for this system the error due to the low-concentration approximation is about 1%, whereas the discrepancy between the Forster approximation of the first term of (14) and its exact value is due at low values of the time to the replacement of the summation by the integration:

$$\exp\left\{\sum_{z\neq x}\ln[1+c(\exp(-\nu_{zx}t)-1]\right\}\approx\exp\left\{c\sum_{z\neq x}[\exp(-\nu_{zx}t)-1]\right\}$$

$$\approx\exp\left\{c\int\frac{d^3z}{\Omega}[\exp(-\nu_{zx}t)-1]=\exp[-(\beta t)^{1/2}],\right.$$

$$\beta=\left(\frac{16\pi}{9}\sqrt{\frac{\pi}{3}}\right)^2\frac{\alpha}{\Omega^2}.$$

Here Ω is the volume of the unit cell, and α is defined by the relation

$$\nu_{xz}=\alpha(1-3\cos^2\theta_{xz})^2/(|\mathbf{x}-\mathbf{z}|)^6.$$

We consider now some general properties of the series (12) in the low-concentration limit. In this case

$$p_{xy}(t)=c\delta_{xy}\exp\left\{c\sum_z[\exp(-\nu_{zy}t)-1]\right\}$$

$$+\sum_{n=1}^{\infty}\int_0^t dt_1\ldots\int_0^{t_{n-1}}dt_n\sum_{q_1\ldots q_{n-1}}\left(\prod_{i=0}^{n-1}\nu_{q_iq_{i+1}}\right)\left\langle\prod_{j=0}^{n}n_{q_j}\right\rangle$$

$$\times\exp\left\{-\sum_{k,l=0}^{n}{}'\nu_{q_kq_l}\tau_l\right)\exp\left\{c\sum_z\left[\exp\left(-\sum_{l=0}^{n}\nu_{zq_l}\tau_l\right)-1\right]\right\}. \qquad (15)$$

We introduce new integration variables: $x_k=t_k/t$. We rearrange (15) into a series in powers of the concentration, in the same manner as when the lower terms in c were separated in (13). We replace the summation over the lattice by integration:

$$\sum_q\to\int\frac{d^3q}{\Omega}.$$

As a result we get

$$p_{yy}(t)=c\exp\left\{c\int\frac{d^3z}{\Omega}[\exp(-\nu_{zy}t)-1]\right\}$$

$$+\sum_{n=2}^{\infty}c^n\int\frac{d^2q_1}{\Omega}\ldots\int\frac{d^3q_{n-1}}{\Omega}A_n, \qquad (16)$$

$$p_{x\neq y}(t)=\sum_{n=1}^{\infty}c^{n+1}\int\frac{d^3q_1}{\Omega}\ldots\int\frac{d^3q_{n-1}}{\Omega}B_n.$$

Here

$$A_n=\sum_{k=n}^{\infty}\int_0^1 dx_1\ldots\int_0^{x_{k-1}}dx_ka_k(\{x_i\},\{t\nu_{q_iq_j}\})$$

$$\times\exp\left\{c\int\frac{d^3z}{\Omega}\left[\exp\left(-\sum_{l=0}^{k}t\nu_{zq_l}(x_l-x_{l+1})\right)-1\right]\right\}, \qquad (17)$$

$$B_n=\sum_{k=n}^{\infty}\int_0^1 dx_1\ldots\int_0^{x_{k-1}}dx_kb_k(\{x_i\},\{t\nu_{q_iq_j}\})$$

$$\times\exp\left\{c\int\frac{d^3z}{\Omega}\left[\exp\left(-\sum_{l=0}^{k}t\nu_{zq_l}(x_l-x_{l+1})\right)-1\right]\right\},$$

$$x_0=1,\ x_{k+1}=0.$$

Here $\{x_i\}$ is the set of x_i with $0\leqslant i\leqslant k$, $\{t\nu_{q_iq_j}\}$ is the set $t\nu_{q_iq_j}$ at $0\leqslant i\leqslant k$, $0\leqslant j\leqslant k$; a_k and b_k are certain functions of $\{x_i\}$, $\{t\nu_{q_iq_j}\}$, independent of the concentration and dependent on t and c only via the indicated arguments. The explicit form of these functions is immaterial in what follows. The difference between the powers of the concentration (c^n in p_{yy} and c^{n+1} in p_{xy}) is due to the fact that for p_{yy} we have

$$\prod_{j=0}^{n}n_{q_j}=\prod_{j=1}^{n}n_{q_j},$$

since $\mathbf{q}_0\equiv\mathbf{q}_n\equiv\mathbf{y}$, whereas for p_{xy} we have $\mathbf{x}\equiv\mathbf{q}_0\neq\mathbf{q}_n\equiv\mathbf{y}$.

Introducing now in (16) and (17) new spatial integration variables $\tilde{\mathbf{q}}_i=\mathbf{q}_i/r_a$, $\tilde{\mathbf{z}}=\mathbf{z}/r_a$, where $r_a=(\Omega/c)^{1/3}$ is the average distance between spins, we get

$$p_{yy}(t)=cf(\nu_at),\quad p_{x\neq y}(t)=c^2g\left(\frac{\mathbf{x}-\mathbf{y}}{r},\nu_at\right). \qquad (18)$$

Here ν_a is the average (over the orientations) rate of cross relaxation between two spins separated by a distance r_a. The functions f and g themselves do not depend on the time, coordinates, or concentration, and are dimensionless.

We examine now the physical meaning of (18). In it are separated the natural parameters that determine the evolution of $p_{xy}(t)$: the average distance between the spins, and the cross-relaxation rate at this distance.

On the other hand, the difference between the powers of the concentration in f and g of (18) is due to the following. The initial condition for (14) was chosen in the form $p_x(t=0)=n_y\delta_{xy}$. This corresponds to the condition that the site y is reliably occupied by a spin. Actually, n_y can take on two values, $n_y=1$ with probability c, and $n_y=0$ with probability $1-c$. But at $n_y=0$ we we have $p_x(t)=0$. Thus, only realizations of the random spin distributions for which the site y is occupied contribute to $p_{xy}(t)$. Any other angle in these realizations is occupied with probability c. Consequently, in the rest of space there is only one spin for $N_0=1/c$ sites. Cross relaxation equalizes, in order of magnitude, the angular-momentum projections per spin in various points of space. Consequently, after the lapse of a sufficient time we can write

$$p_{yy}\sim\int_V\frac{d^3x}{\Omega}p_{x\neq y}.$$

Here V is a volume containing N_0 sites. Therefore $p_{x\neq y}\sim cp_{yy}$, a fact indeed reflected in (18). From the derivation of (18) it follows that these relations are valid regardless of the actual dependence of ν_{xz} on $|\mathbf{x}-\mathbf{z}|$.

It is now easy to ascertain which is the true expansion parameter for p_{yy} in (16). Since $\nu_a\sim c^2$, this parameter is, apart from a numerical factor, $T=(\nu_at)^{1/2}$.

It follows from (18), in particular that at $c\ll 1$ the value of p_{xy} does not tend with increasing t to the solution of the macroscopic diffusion equation:

$$\partial p/\partial t=D\Delta p,\quad p(t=0)=\delta(x-y). \qquad (19)$$

Let us assume that the opposite is true. Relations (18) are asymptotically exact (for time at which the summation over the lattice indices can be replaced by integration, at low concentrations, but at arbitrary values of T) solutions of the initial equation (2) under the condition that at the initial instant of time the polarization source is located at the point y. Since $p_{xy}(t)$ is the Green's function of (19) we get

$$c^2 g\left(\frac{\mathbf{x}-\mathbf{y}}{r_a}, \nu_a t\right) = \frac{\Omega c}{(4\pi D t)^{3/2}} \exp\left[-\frac{(\mathbf{x}-\mathbf{y})^2}{4Dt}\right].$$

We thus obtain

$$D \sim c^{4/3}.$$

A similar analysis for $p_{yy}(t)$ (we equate $p_{yy}(t)$ to the polarization in the source) leads to the estimate

$$D \sim c^2.$$

Thus, the assumption that the diffusion equation is valid is untenable. For no value of the time does the averaged solution of Eq. (2) coincide with the solution of the diffusion equation, nor can it be represented by a smooth function: there is a singularity at the point y. This circumstance highlights the fundamental difference between the migration of an excitation over ordered and disordered lattices. In the former case, Eq. (19) is valid at large enough t.

We note that relations (18) are violated at small t. The reason is that in the derivation of (18) the lattice sums were replaced by integrals over all of space, including the points where the functions $\nu_{q_i q_j}(|\mathbf{r}_{q_i} - \mathbf{r}_{q_j}|)$ have singularities. At sufficiently large t the contribution of these points becomes insignificant, because these functions enter the integrand in the form of either $\nu_e^{-\nu t}$ or simply $e^{-\nu t}$. At low values of t, however, the difference can be appreciable from the point of view of the reduction of precision experiments of the type described in Ref. 15.

We note finally that merely the asymptotic accuracy of (18) leads to a conservation of the normalization

$$\sum_x p_{xy}(t) = p_{yy}(0) = c, \tag{20}$$

only under the condition $\lim_{x\to y} p_{xy}(t) = 0$. In fact,

$$\sum_x p_{xy} = p_{yy} + \sum_{x \neq y} p_{xy} \approx p_{yy} + \int \frac{d^3x}{\Omega} p_{xy} - \lim_{x \to y} p_{xy}.$$

But

$$\int \frac{d^3x}{\Omega} p_{xy}(t) = c^2 \int \frac{d^3x}{\Omega} g\left(\frac{\mathbf{x}-\mathbf{y}}{ra}, \nu_a t\right)$$
$$= c \int d^3x \, g(\mathbf{x}-\mathbf{y}, \nu_a t) = c\varphi(\nu_a t).$$

From the equality

$$cf(\nu_a t) + c\varphi(\nu_a t) + c^2 g(0; \nu_a t) = c \tag{21}$$

it follows that to ensure the normalization we must have $g(0, \nu_a t) = \lim_{x\to y} p_{xy}(t) = 0$. It is easy to show, however (assuming in 14a) for simplicity $\nu_{xy} = \nu_{yx}$), that

$$\lim_{x \to y} p_{\bar{x}y}(t>0) \geqslant \frac{c^2}{2} \exp\left\{c \sum_z [\exp(-\nu_{zy} t) - 1]\right\},$$

and thus, the condition (20) has in fact been violated. It must be emphasized, however, that the normalization-violating term in (21) is proportional to the small factor c^2 and its inclusion in (21) exaggerates the accuracy of (18).

In conclusion, we compare the presented approach to the description of diffusion in a disordered lattice with the results obtained in earlier studies that are closest to ours in their scope.

To solve Eq. (4), Sakun[19] introduced an occupation-number formalism and developed a diagram method with the aid of which he obtained the exact formula (9), which generalizes the Forster decay law. To describe diffusion by the same method, on the other hand, an irreducible operator was introduced and defined by an expansion in powers of the concentration c, in the lowest order in c. It follows from our results that such an expansion is in reality an expansion in $ct^{1/2}$ and can be valid, just as our formula (14), only for the start of the process. Sakun[19] separated correctly the natural scales of the variation of p_{xy}, but the solutions obtained there still does not satisfy relations (18), since it does not account for the special nature of the site y to which the excitation is initially applied.

Scher and Lax[22] developed a general phenomenologial concept of random walk in a disordered lattice, in which, just as in Ref. 19, the natural units of distance and time were correctly separated in the case of small c. Butcher[23] interpreted this approach as a partial summation of an iteration series, a summation replaced in Refs. 17 and 18 by an uncontrollable decoupling procedure. In the sense of the described constructions the method developed in these papers should yield an averaged solution of Eq. (4). The singular character of the initial point was not noted in these papers, however, so that their results do not satisfy the relations (18). This manifests itself quantitatively, for example, in the fact that the iteration solution of Eq. (6) of Ref. 17, which should be close in its meaning to our series (15), accounts correctly for the first term in p_{yy}, but yields a coefficient c^3 instead of c^2 for the second term.

An approach similar to that proposed in Ref. 22 is treated in Ref. 24 as a gross-structure averaging. This method is presently under intensive study (see, e.g., Ref. 31). The results of the gross-structure averaging should no longer satisfy formulas (18), but they cannot yield information on the function p_{yy}, which is essential for experiments with β-active nuclei.

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[1] C. S. Wu, E. Ambler, R. Hayward, D. Hoppes, and R. Hudson, Phys. Rev. **105**, 1413 (1957); **106**, 1361 (1957).
[2] F. L. Shapiro, Usp. Fiz. Nauk **65**, 133 (1958).
[3] D. Connor, Phys. Rev. Lett. **3**, 429 (1959).
[4] Y. G. Abov, O. N. Yermakov, A. D. Gulko, P. A. Krupchitsky, and S. S. Trostin, Nucl. Phys. **34**, 505 (1962).
[5] Perturbed Angular Correlations, eds. E. Karlsson *et al.*, North-Holland, 1964.
[6] A. D. Gul'ko, S. S. Trostin, and A. Khudoklin, Zh. Eksp. Teor. Fiz. **52**, 1504 (1967) [Sov. Phys. JETP **25**, 998 (1967)]; Yad. Fiz. **6**, 657 (1967) [Sov. J. Nucl. Phys. **6**, 477 (1967)].
[7] M. I. Bulgakov, A. D. Gul'ko, Yu. A. Oratovskii, and S. S. Trostin, Zh. Eksp. Teor. Fiz. **61**, 667 (1971) [Sov. Phys.

JETP 34, 356 (1972)].
[8] J. Mertens, H. Ackermann, D. Dubbers, P. Heitjams, A. Winnacker, and P. von Blanckenhagen, Z. Phys. 262, 189 (1973).
[9] T. K. McNab and R. E. McDonald, Phys. Rev. B 13, 34 (1976).
[10] T. Minamisonsi, J. W. Hugg, D. G. Mavis, T. K. Saylor, S. M. Lasarus, H. F. Slavish, and S. S. Hanna, Phys. Rev. Lett. 34, 1465 (1975).
[11] R. C. Haskell, F. D. Correll, and L. Madansky, Phys. Rev. B 11, 3268 (1975).
[12] H. Ackermann, D. Dubbers, H. Grupp, M. Grupp, P. Heitjams, and H. J. Stockmann, Phys. Lett. A 54, 399 (1975).
[13] R. E. McDonald and T. K. McNab, Phys. Lett. A 63, 177 (1977).
[14] D. Dubbers, K. Dorr, H. Ackermann, F. Fujara, H. Grupp, M. Grupp, P. Heitjans, A. Korblein, and H. J. Stockmann, Z. Phys. 282A, 343 (1977).
[15] M. I. Bulgakov, S.P. Borovlev, A. D. Gul'ko, F. S. Dzheparov, I. G. Ivanter, and S. S. Trostin, Preprint ITEF-150, 1976.
[16] V. A. Atsarkin and M. I. Rodak, Usp. Fiz. Nauk 107, 3 (1972) [Sov. Phys. Usp. 15, 251 (1972)].
[17] B. E. Vugmeĭster, Phys. Status Solidi B 76, 161 (1976).
[18] B. E. Vugmeĭster, Fiz. Tverd. Tela (Leningrad) 18, 819 (1976) [Sov. Phys. Solid State 18, 469 (1976)].
[19] V. P. Sakun, Fiz. Tverd. Tela (Leningrad) 14, 2199 (1972) [Sov. Phys. Solid State 14, 1906 (1973)].
[20] Yu. K. Voron'ko, T. G. Mamedov, V. V. Osiko, A. M. Prokhorov, V. P. Sakun, and I. A. Shcherbakov, Zh. Eksp. Teor. Fiz. 71, 478 (1976) [Sov. Phys. JETP 44, 251 (1976)].
[21] A. I. Burshteĭn, Zh. Eksp. Teor. Fiz. 62, 1695 (1972) [Sov. Phys. JETP 35, 882 (1972)].
[22] H. Scher and M. Lax, Phys. Rev. B 7, 4491 (1973); 7, 4502 (1973).
[23] P. N. Butcher, J. Phys. C 7, 879 (1974); 7, 3533 (1974).
[24] H. Scher and E. W. Montroll, Phys. Rev. B 12, 2455 (1975).
[25] T. Forster, Z. Naturforsch. Teil A 4, 321 (1949).
[26] A. Abragam, Principles of Nuclear Magnetism, Oxford, 1961.
[27] I. V. Aleksandrov, Teoriya magnitnoĭ relaksatsii (Theory of Magnetic Relaxation), Nauka, 1975.
[28] V. M. Faĭn, Fotony i nelineĭnye sredy (Photons and Nonlinear Media), Sov. Radio, 1972.
[29] N. Bloembergen, S. Shapiro, P. S. Pershan, and J. O. Artman, Phys. Rev. 114, 445 (1959).
[30] Yu. A. Izyumov and M. V. Medvedev, Teoriya magnitouporyadochennykh kristallov s primesyami (Theory of Magnetically Ordered Crystals with Impurities), Nauka, 1970, §21.
[31] D. T. Gillespie, Phys. Lett. A 64, 22 (1977).

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Dynamic damping of dislocations by magnetizations in ferromagnets

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The dynamic damping of dislocations by magnetizations in ferromagnets is considered. The temperature dependence of the damping force is investigated in the vicinity of the Curie point within the framework of the molecular-field approximation. The relative efficiencies of the phonon, electron, and magnetic contributions to the damping force are compared. It is shown that the magnetic contribution is decisive only in ferromagnets with low Curie points.

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It is by now reliably established that the plastic properties of metals are connected in one way or another with the motions of dislocations that are either pinned by static obstacles (impurities, complexes or impurities, other dislocations, long-range fields or elastic stresses, and others), or are dynamically damped by phonons, electrons, and other elementary excitations. Dynamic damping is decisive at high dislocation velocities, when the motion is above the barrier. It must be taken into account also in the case of low velocities when dislocations glide between obstacles. The most effective among the dynamic mechanisms is a rule damping by phonons. Contributions of other elementary excitations manifest themselves only in some distinct situations: thus, dislocation damping by electrons is significant at sufficiently low temperatures, and damping by magnons takes place in ferromagnets below the Curie point.

Dynamic damping of dislocations by phonons and electrons has been studied quite thoroughly, and is the subject of exhaustive reviews.[1,2] There is still no corresponding description of dislocation damping by magnons in ferromagnets. Among the latest papers connected with this problem mention should be made of the article by Bar'yakhtar and Druinskiĭ, who investigated dislocation damping by magnons and obtained the temperature dependence of the magnon damping

$$F_s = B_s^{(2)} (T/\Theta_c)^{5/2} V, \tag{1}$$

where V is the dislocation, $B_s^{(2)}$ is a coefficient independent of the temperature T, and Θ_c is the Curie temperature. Bar'yakhtar and Druinskiĭ[3] used the spin-wave approximation, which describes ferromagnets at relatively low temperatures. Equation (1) therefore describes correctly the temperature dependence of the magnetic-damping force at low values of T, but is not

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