Statistical dynamics of spin systems and β-NMR spectroscopy

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Experimental studies of the spin dynamics of impurity nuclei in crystals using the technique of β-NMR spectroscopy are reviewed along with the associated theoretical studies. Special attention is paid to the spectrum of spin fluctuations, the effect of correlations of local fields on the impurity spins, processes like spin diffusion in disordered systems, and the effect on the latter of external alternating fields and thermal translational motion of the nuclei. © *1995 American Institute of Physics.*

1. INTRODUCTION

The kinetics of spin systems is one of the best studied areas of statistical mechanics. From the experimental side this is primarily due to the very high sensitivity of the technique of nuclear magnetic resonance (NMR) spectroscopy.[1,2] For the theoretical analysis it is important that the spin systems are often well separated from the other degrees of freedom, while the Hamiltonian of the dipole–dipole interactions is sufficiently complicated to ensure the ergodicity of multispin systems and at the same time simple enough that, for example, the three-spin problem can in many cases be solved analytically. It is also extremely important that under typical conditions the spin temperature is nearly infinite and equilibrium states are almost trivial. The kinetics of the nonequilibrium states is nevertheless very complicated and rich. As an illustration of the progress that has been made, we note the experimentally realized treatment of the evolution (rejuvenation) of nuclear dipole systems,[3,4] the ergodicity of which has been reliably demonstrated by other experiments.[5]

However, even standard NMR has some disadvantages. One of its main limitations is that the observed quantity is the total magnetization of some type of spin,[5] which under typical conditions reduces to a correlation function:

$$G_T^{\alpha\beta}(t)=\langle I_T^{\alpha}(t)I_T^{\beta}\rangle_0/\langle I_T^{\alpha}I_T^{\beta}\rangle_0, \quad I_T^{\alpha}=\sum_i^N I_i^{\alpha},$$

$$\alpha=x,y,z, \quad \langle\ldots\rangle_0=\mathrm{Tr}(\ldots)/\mathrm{Tr}\ 1,$$

where I_i^{α} is the α component of the ith spin of the subsystem in question, and the Heisenberg time dependence is determined, as usual, by the secular part of the dipole–dipole interaction. As a result, a fundamental process such as the redistribution of polarization between various nuclei of the subsystem is manifested very indirectly.

The method of β-NMR spectroscopy was the first to really allow this process to be studied in great detail. In addition, owing to the high sensitivity of the method, it proved possible to make a deeper study of such a standard and fundamental characteristic as the shape function of the NMR line.

The β-NMR method is based on study of the evolution of the polarization $\langle I_0^z(t)\rangle$ of an impurity nucleus (the β nucleus) on the basis of the anisotropy of its β emission.[6] As a rule, such processes proceed exponentially, because both the relaxation of the surrounding spins of the crystal and the phonon propagation occur much more quickly. It was therefore extremely important to find systems in which the nuclear probe has nearly the same g factor as one of the stable isotopes, so that the spin degrees of freedom of this system are well separated from the vibrational modes of the crystal. In the subsystem formed by the β nucleus and spins with similar g factor there is an additive integral of the motion which projects the total spin on the constant external field, $\Sigma_{i=0}^N I_i^z$. Under ordinary conditions it can be assumed that at first, when the probe nucleus is created, only it is polarized. Then the polarization passes to the nearest spins of the subsystem. This is followed by its transfer both back to the β nucleus and on to more distant impurity nuclei, which is typical of processes of the diffusion type. Such redistributions of the densities of additive integrals of the motion (DAIMs) are the most important processes in all of physical kinetics. This is a consequence both of the important role played by additive integrals of the motion in equilibrium theory,[6] and of the relative slowness of the evolution of DAIMs, as a result of which all faster processes are tuned to them.[7–9]

The first and so far only known spin subsystem of this type is that formed by the spins of the β nucleus ^{8}Li and the ^{6}Li impurity nuclei in the LiF crystal.[10–12] The most important feature of this subsystem is the random distribution of ^{6}Li nuclei in the crystal. As a result of polarization transfer among them, it is possible to have random walks in disordered media (RWDMs), which represents one of the newest areas in statistical mechanics (see, for example, Ref. 13). This is a statistical disorder of the medium, in contrast to the dynamical disorder characteristic of, for example, the canonical theory of gases and liquids. The transport within a single fixed impurity configuration is described by a theory using the idea of dynamical disorder of spin orientations in a crystal,[14] and relatively simple kinetic equations are obtained as a result. The RWDM problem is to obtain the solutions to these equations averaged over the ensemble of statistical impurity configurations. The analysis of Ref. 15 shows that this problem leads to very general nonlocal, nonlinear, and nonrenormalizable field and superfield theories. On the other hand, its propagators can be represented by functional integrals reminiscent of the Feynman theory of the polaron, but

with a considerably more complicated action. This relationship between RWDM problems and general, fundamental problems of theoretical physics provides a powerful stimulus for our studies.

Among the interesting results, we note that the initial stage of the depolarization kinetics of the nuclear probe in the ^{8}Li–^{6}Li system gives direct information about the Fourier transform of the correlation function $\langle I_j^+(t) I_0^-(t) I_0^+ I_j^- \rangle_0$, which cannot be studied in such detail by other experimental methods. Here it turned out that there is a particular correlation of local fields which, at first glance, appears to violate the central limit theorem of probability theory.

In this review we discuss the main physical principles and results of these studies. For more detailed information on particular aspects we refer the reader to the original studies.

2. β-NMR SPECTROSCOPY

The β radiation of polarized nuclei obeys the law

$$W(\vartheta) \sim 1 + \alpha \cdot p_0 \cdot \cos \vartheta,$$

where $W(\vartheta)$ is the probability of emission of a β particle at angle ϑ to the polarization $\vec{p}$ of the β-active nucleus, and α is the nuclear constant. Therefore, by creating polarized β-active nuclei (β nuclei) in matter and studying the dependence of the angular distribution of their β emission on time, external constant and rf magnetic fields, temperature, and pressure, one can study the evolution of the polarization of the β nuclei and thereby a number of processes arising from the hyperfine and dipole–dipole interactions.

The method was proposed by F. L. Shapiro[16] and realized in Refs. 17 and 18 for studying the spins and dipole and quadrupole moments of β nuclei. Recently the method has been used to study condensed media, where it has important advantages over both classical NMR spectroscopy and other nuclear techniques (perturbed angular γ correlations and the Mössbauer effect) in studying a number of phenomena.[19,20]

Our studies were performed at the reactors of the Institute of Theoretical and Experimental Physics and the Moscow Engineering Physics Institute. The reaction ^{7}Li $(\vec{n},\gamma)^8\vec{\text{Li}}$ on thermal polarized neutrons was used to obtain β nuclei. Immediately after this reaction the spin density matrix of the β nuclei has the form

$$\rho(t=0) = \frac{1}{2I_0+1}\left(1 + \frac{3p_0(t=0)}{I_0(I_0+1)} I_0^z\right), \quad \vec{p}_0 = \mathrm{Tr}(\vec{I}_0 \rho), \quad (1)$$

where $\vec{I}_0^z$ is the spin operator and $\vec{p}_0$ is the polarization. The external field $\mathscr{H}_0$ is assumed to point along the z axis.

The design of a β-NMR spectrometer is shown in Fig. 1. A polarized beam of neutrons with density $\sim 4\times 10^6$ neutrons/(cm^{2}sec) is obtained by reflection from a magnetized cobalt mirror. The β-decay electrons are recorded independently by two scintillation counters located on either side of the sample in the gap of the electromagnet.

The angular asymmetry of the β emission is usually calculated from the expression

$$\varepsilon = (N(0°) - N(180°))/(N(0°) + N(180°)),$$

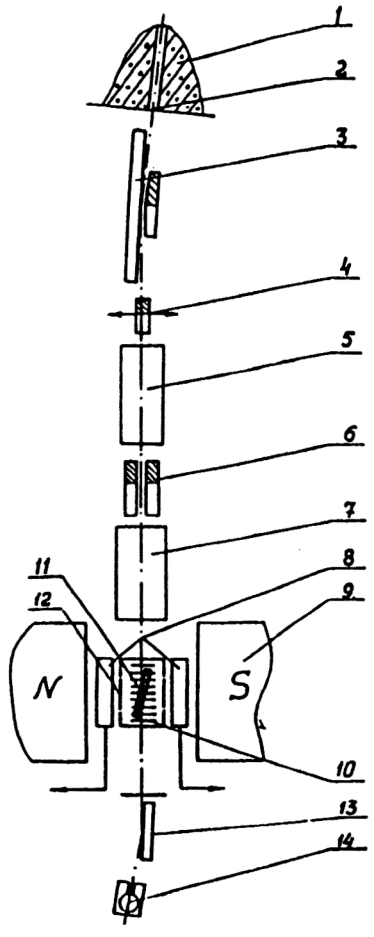

FIG. 1. Schematic diagram of a β-NMR spectrometer: (1) reactor shielding; (2) collimator; (3) cobalt mirror-polarizer; (4) beam chopper; (5) spin flipper (NS and N′S′ are magnets; φ is a current-carrying foil); (6) collimators; (7) guiding field magnet; (8) β counters; (9) NMR magnet; (10) rf coil; (11) sample; (12) cryostat or thermostat; (13) cobalt mirror-analyzer; (14) neutron counter.

where $N(0°)$ and $N(180°)$ are the numbers of β-decay electrons recorded by the β counter for two orientations of the neutron (and β-nucleus) polarization relative to the external stationary field $\mathscr{H}_0$. The asymmetry ε is proportional to the polarization $P_{00}(t) = \langle p_0(t) \rangle_{\text{ave}}$ of the ensemble of β nuclei averaged over their distribution in the sample. It is also necessary to take into account the averaging over the time τ_{irr} for which the sample is irradiated by neutrons and over the time $\Delta t = t_{n+1} - t_n$, $n = 0,1,2, \ldots$ of successive counting of β particles in the nth channel of the time analyzer:

$$\varepsilon\left(t_n + \frac{\Delta t}{2}\right) = \varepsilon_0 \cdot \int_{-\tau_{\text{irr}}}^{0} d\tau \int_{t_n}^{t_{n+1}} dt e^{-\lambda_0(t-\tau)} \times P_{00}(t-\tau) \Big/ \int_{-\tau_{\text{irr}}}^{0} d\tau \int_{t_n}^{t_{n+1}} dt e^{-\lambda_0(t-\tau)}. \quad (2)$$

Here λ_0 is the β-decay probability, and ε_0 is the maximum (i.e., in the absence of depolarization) value of the asymmetry. Measurements differential in time were made for $\lambda_0 \Delta t \ll 1$ and $\lambda_0 \tau_{\text{irr}} \gtrsim 1$. The dependences on the external

magnetic fields and on the temperature were studied in the integral mode with counting of decay electrons in one time channel of duration τ_{obs}. Here the asymmetry $\tilde{\varepsilon}=\varepsilon(t_0+\Delta t/2)$ with $t_0=0$ and $\Delta t=\tau_{obs}\gg\lambda_0^{-1}$ was measured. In the case $\lambda_0\tau_{irr}\gg 1$ and $\lambda_0\tau_{obs}\gg 1$,

$$\tilde{\varepsilon}=-\varepsilon_0\lambda_0^2\partial P_{00}(\lambda_0)/\partial\lambda_0, \quad \text{where } P_{00}(\lambda)$$

$$=\int_0^\infty dt\cdot\exp(-\lambda t)P_{00}(t). \tag{3}$$

We studied single-crystal, oriented LiF samples of dimensions $60\times 40\times 2$ mm^3 with the (100), (110), and (111) crystallographic planes parallel to the large surface of the sample, which in turn was perpendicular to the direction of the stationary magnetic field. The concentration of the ^{6}Li isotope was 3.21(3)%. The samples were placed in a special thermostat for heating. The half-life $T_{1/2}$ of the ^{8}Li nuclei is 0.84 sec ($\lambda_0=\ln 2/T_{1/2}=0.825$ sec^{-1}). The spins of the nuclei ^{8}Li, ^{6}Li, ^{7}Li, and ^{19}F are, respectively, $I_0=2$, $I_{r\neq 0}=1$, $L=3/2$, and $F=1/2$, and their g factors are $g(^8\text{Li})=0.8267$, $g(^6\text{Li})=0.8220$, $g_L=2.171$, and $g_F=5.257$. Whenever possible, we shall neglect the unimportant difference between the g factors of ^{8}Li and ^{6}Li: $g(^8\text{Li})\approx g(^6\text{Li})\approx g_I=0.824$.

3. PHASE RELAXATION AND THE NMR OF β-ACTIVE ^{8}Li NUCLEI IN LiF

In paramagnetic nuclear spin systems the phase relaxation is almost completely determined by collective many-particle effects. Modern studies of phase relaxation fall into several categories. The first is the simple, pragmatic description of this phenomenon in terms of memory functions and the first few moments of the resonance line, that is, the first derivatives with respect to time of the correlation function $g(t)=\langle I_+(t)I_-\rangle_0/\langle I_+I_-\rangle_0$ at $t=0$ (see Ref. 21 and references therein). In the second, special attention is paid to the oscillations of this correlation function in homonuclear systems.[21,22] The third group of studies[23,24]focuses on explaining the exponential behavior of the spectral density of the spin fluctuations or, in other words, the shape functions of the NMR line,

$$g(\omega)=\int_{-\infty}^{\infty}g(t)\ \cos(\omega t)\ \frac{dt}{2\pi}, \tag{4}$$

for large ω found experimentally in Ref. 25 and in a smaller range of $g(\omega)$ in the studies listed in Sec 1.B.d.3 of the monograph in Ref. 21. The fourth, important, group of studies was begun in Refs. 26–28, which introduced into the theory of phase relaxation the model of a normal random process for describing the fluctuations of local fields. This model was first used to explain the effect of motional narrowing. Here the decrease of the free induction $g(t)=\langle\exp[i\int_0^t d\tau\cdot\hat{\omega}_1(\tau)]\rangle_0$ is written as $g(t)\approx\exp[-\int_0^t d\tau\cdot(t-\tau)\Lambda(\tau)]$, where $\Lambda(\tau)=\langle\hat{\omega}_1(\tau)\hat{\omega}_1(0)\rangle_0$, $\Lambda(0)=\langle\hat{\omega}_1^2\rangle_0=M_2$. Here M_2 is the second moment of the NMR shape function, and $\hat{\omega}_1(\tau)$ is the local-field operator at the nucleus. If the characteristic time $\tau_c\sim\int_0^\infty dt\cdot\Lambda(\tau)/M_2$ of the local-field fluctuations varies from $\tau_{c\infty}\gg 1/\sqrt{M_2}$ (static local fields) to $\tau_{c0}\ll 1/\sqrt{M_2}$ (rapid motion of the spins of the matrix), then $g(t)$ changes from a Gaussian $g_G(t)=\exp(-M_2t^2/2)$ to an exponential $g_L(t)=\exp(-\Gamma t)$, $\Gamma\sim M_2\tau_c$, falling off more slowly at large t. Usually $\Lambda(\tau)=M_2\exp(-|\tau|/\tau_c)$ is used for a qualitative description of the nuclear jumps, and $\Lambda(\tau)=M_2\exp(-\tau^2/2\tau_c^2)$ is used for studying the smooth evolution (such as flip–flop transitions) of local fields. If we take $\Lambda(\tau)=M_2\cosh^{-2}(\tau/\tau_c)$, we can obtain an analytic expression for $g(t)$ in terms of elementary functions.

In our opinion, the possibilities of the Anderson–Weiss–Kubo theory are far from exhausted at present, because: (1) so far there has been no satisfactory solution of the problem of the motion of a spin in a three-dimensional fluctuating field, and (2) the full potential of the general theory of irreversible processes has not been used in analyzing the spin correlators of impurity nuclei when the local field is one-dimensional. The first of these has been partially taken care of in the studies of Zobov,[24] who also showed that in the limit of a large number of neighbors (more precisely, for infinite size) the local-field fluctuations are a normal random process. Overcoming the second defect is the subject of Ref. 29, where the NMR shape function of impurity nuclei was measured for the first time in a wide range $g_0(\omega)\geq 10^{-5}\times g_0(\omega=0)$ for various orientations of the magnetic field. There it was also found that when the motion of the local fields at impurity nuclei is modeled as a realistic normal random process, a good description of the experimental data is obtained if in the calculation of the correlator of these fields $\Lambda(t)$ one takes into account the fact that it is determined by the evolution of the z components of the spins of the basic nuclei of the crystal (the matrix): at small t it is an analytic function of t^2, and then as t increases the collective nature of the spin evolution of the matrix nuclei becomes evident, where at first $\ln\Lambda(t)\propto -t$, and then the large-time asymptote of $\Lambda(t)$ turns out to be characteristic of diffusion: $\Lambda(t\to\infty)\propto t^{-3/2}$, since the total z component of each type of spin of the matrix is an additive integral of the motion.

The spin Hamiltonian of the ^{8}Li–^{6}Li system in LiF can be written as

$$H=H_0+H_I+H_{IF}+H_{IL}, \quad H_0=H_F+H_L+H_{FL},$$

$$H_A=\frac{1}{4}\sum_{\substack{\mathbf{x},\mathbf{q}\\ \mathbf{x}\neq\mathbf{q}}} b_{\mathbf{xq}}^A(3A_{\mathbf{x}}^zA_{\mathbf{q}}^z-\vec{A}_{\mathbf{x}}\vec{A}_{\mathbf{q}}),$$

$$H_{AB}=\sum_{\substack{\mathbf{x},\mathbf{q}\\ \mathbf{x}\neq\mathbf{q}}} b_{\mathbf{xq}}^{AB}\cdot A_{\mathbf{x}}^zB_{\mathbf{q}}^z. \tag{5}$$

Here and below, the vectors **r**, **q**, **x**, **y**, and **z** specify the allowed locations of the nuclei and take values from the corresponding fcc sublattices of the LiF crystal. The superscripts A and B label the type of sublattice and spin, taking the values F, L, and I, where $F=1/2$, $L=3/2$ for ^{19}F and ^{7}Li nuclei and $I=I_0=2$ and $I=I_{\mathbf{q}\neq 0}=1$ are the spins of ^{8}Li and ^{6}Li, respectively. The coefficients of the dipole–dipole interaction have the form[30]

$$b^{AB}_{\mathbf{xq}}=\omega_{AB}\tilde{b}_{\mathbf{xq}},\quad b^{A}_{\mathbf{xq}}=b^{AA}_{\mathbf{xq}},\quad \omega_{AB}=\frac{g_A g_B \beta_n^2}{\hbar d^3},$$

$$\tilde{b}_{\mathbf{xq}}=\frac{1-3\cos^2\vartheta_{\mathbf{xq}}}{|(\mathbf{x}-\mathbf{q})/d|^3}. \tag{6}$$

Here $\vartheta_{\mathbf{xq}}$ is the polar angle of the vector $\mathbf{x}-\mathbf{q}$, and $d=2.01$ Å is the minimum distance between Li and F in LiF [$\omega_{IF}=2\pi(2.05$ kHz), $\omega_{IL}=2\pi(0.85$ kHz), $\omega_{II}=2\pi(0.322$kHz)].

In the model of a normal random process the signal of the free induction of an impurity spin $g_0(t)$ is approximated by the first few terms of the cumulant expansion:

$$g_0(t)=\left\langle I_-I_+T\exp\left(-i\int_0^t\hat{\omega}_0(\tau)d\tau\right)\right\rangle_0\Big/\langle I_-I_+\rangle_0$$

$$\approx\exp\left(-\int_0^t(t-\tau)\Lambda_{00}(\tau)d\tau\right),$$

$$\Lambda_{00}(\tau)=\langle\hat{\omega}_0(\tau)\cdot\hat{\omega}_0\rangle_0, \tag{7}$$

where in calculating $\Lambda_{00}(\tau)$ the effect of the impurity spins on the spins of the matrix is neglected. Here and below in this section we neglect the effect of the ^{6}Li nuclei on the β nucleus because their concentration is small. Assuming that the β nucleus is located at the coordinate origin, the local field can be written in units of frequency as

$$\hat{\omega}_0=\sum_{A=F,L}\hat{\omega}_0^{(A)},\quad \hat{\omega}_0^{(A)}=\sum_{\mathbf{x}\in A}b^{IA}_{0\mathbf{x}}\cdot A^z_{\mathbf{x}}.$$

Accordingly,

$$\Lambda_{00}(t)=\sum_{A=F,L}\Lambda_{00}^{(A)}(t),$$

$$\Lambda_{00}^{(A)}(t)=\langle\hat{\omega}_0^{(A)}(t)\cdot\hat{\omega}_0^{(A)}(0)\rangle_0$$

$$=\frac{A(A+1)}{3}\sum_{\mathbf{x}\in A}\sum_{\mathbf{y}\in A}b^{IA}_{0\mathbf{x}}G^A_{\mathbf{xy}}(t)b^{AI}_{\mathbf{y}0}. \tag{8}$$

The simplest but qualitatively correct representation for the spin-diffusion propagator $G^A_{\mathbf{xy}}(t)=\langle A^z_{\mathbf{x}}(t)A^z_{\mathbf{y}}\rangle_0/\langle(A^z_{\mathbf{x}})^2\rangle_0$ can be obtained using the equations[14,29]

$$\partial G^A_{\mathbf{xy}}/\partial t^A_{\text{eff}}=-\sum_{\mathbf{z}}\mathscr{A}^A_{\mathbf{xz}}\cdot G^A_{\mathbf{zy}},\quad G^A_{\mathbf{xy}}(t=0)=\delta_{\mathbf{xy}},$$

$$\mathscr{A}^A_{\mathbf{xy}}=-\nu^A_{\mathbf{xy}}+\delta_{\mathbf{xy}}\cdot\sum_{\mathbf{z}}\nu^A_{\mathbf{zx}},$$

$\mathbf{x},\mathbf{y},\mathbf{z}\in$ the Ath sublattice, (9)

where $\nu^A_{\mathbf{xy}}$ is the probability for polarization transfer between two A spins:

$$\nu^A_{\mathbf{xy}}=\frac{\pi A(A+1)}{6}\cdot(\omega_{AA}\tilde{b}^A_{\mathbf{xy}})^2\cdot g_{cA}(\omega=0)=\frac{(\tilde{b}^A_{\mathbf{xy}})^2}{\tau_{cA}\sigma^F},$$

$$\sigma^F\equiv\sigma^L=\sum_{\mathbf{z}}(\tilde{b}^A_{\mathbf{xz}})^2,\quad\frac{1}{\tau_{cA}}=\sum_{\mathbf{z}}\nu^A_{\mathbf{xz}}. \tag{10}$$

Here τ_{cA} is the correlation time (flip–flop transitions) of the A spins. The effective time $t^{(A)}_{\text{eff}}(t)=\int_0^t g_{cA}(\tau)\,d\tau/T_{2A}$ smooths the spin-diffusion propagator at small times $t\leqslant T_{2A}$ [$T_{2A}=\int_0^\infty g_{cA}(\tau)d\tau$ is the phase-relaxation time of the A spins]. The choice of $g_{cA}(\tau)$ in the form $\exp(-M_{2A}\tau^2)$, $\cosh^{-2}\sqrt{M_{2A}}\tau$, or $(1+\frac{2}{3}M_{2A}\tau^2)^{-3/2}$, where M_{2A} is the "one-spin" second moment of the NMR shape function of the matrix nuclei,[12] has practically no effect on the final result. If $g_{cA}(\tau)$ is chosen in the form of a Gaussian, then $T_{2F}=\sqrt{\pi/4M_{2F}}=6.3$ μsec, $T_{2L}=\sqrt{\pi/4M_{2L}}=14.2$ μsec, $\tau_{cF}=(\sqrt{\pi}/12F(F+1)\omega^2_{FF}\sigma^{FF}/\sqrt{M_{2F}})^{-1}=175$ μsec, and $\tau_{cL}=530\mu$sec. These values correspond to the orientation $\mathscr{H}_0\|[001]$ of the crystal in the external magnetic field $\mathscr{H}_0$. The expressions for $g_{cA}(\tau)$ are most accurate at small $M_{2A}\tau^2$ and give a qualitatively correct representation of the damping of $g_{cA}(\tau\to\infty)$, and the spin-diffusion coefficient for the crystal CaF_2 calculated using them is in satisfactory agreement with experiment.[31] Actually, the authors of Ref. 29 used not the exact solution of Eqs. (8)–(10), but the much simpler and more convenient expressions

$$\Lambda_{00}^{(A)}(t)=\tilde{\Lambda}_{00}^{(A)}(t^{(A)}_{\text{eff}})=M_{2IA}\left[(1-\gamma_A)\cdot\exp\left(-\frac{\alpha_A t^{(A)}_{\text{eff}}}{\tau_{cA}}\right)\right.$$

$$\left.+\frac{\gamma_A}{(1+\beta_A t^{(A)}_{\text{eff}}/\tau_{cA})^{3/2}}\right],$$

$$M_{2IA}=\langle(\omega_0^{(A)})^2\rangle_0. \tag{11}$$

The coefficients α_A, β_A, and γ_A were determined from the conditions that the true values of $\partial\tilde{\Lambda}_{00}^{(A)}(t^{(A)}_{\text{eff}})/\partial t^{(A)}_{\text{eff}}|_{t=0}$, $\int_0^\infty dt\tilde{\Lambda}_{00}^{(A)}(t)$ and of the asymptote $\tilde{\Lambda}_{00}^{(A)}(t\to\infty)$ coincide with the corresponding values given by (11) (see Ref. 29).

The results of the calculations of $g_0(\omega)$ for various crystallographic orientations are shown in Fig. 2 along with the experimental data. Good agreement with experiment over a range of variation of $g_0(\omega)$ by 4–5 orders of magnitude was obtained without fitted parameters for the first time. This can be viewed as a convincing argument in favor of the modeling of the behavior of the local field of the matrix as a normal random process.

The use of various trial functions for $g_{cA}(\tau)$ leads to the practically indistinguishable curves for the NMR shape function of the ^{8}Li nucleus shown in Fig. 2. The high-frequency asymptotes $g_0(\omega\to\infty)$ corresponding to these functions are exponential and differ by their numerical parameters. However, these were not reached in our experiments, i.e., the preasymptotic terms are important. Regarding this, we note that the theory of Zobov[23] also gives a fairly correct picture of the slopes of the wings of $g_0(\omega)$ on a semilogarithmic scale.

Although the true asymptote of the NMR shape function $g(\omega\to\infty)$ has not yet been reached in current experiments, the opinion in the early 1970s was that it is exponential. It can be seen from the literature that theoreticians put a great deal of effort into proving this. Apparently, the main difficulties here are purely psychological and are related to the tradition of interpreting NMR results on the basis of comparison with Gaussian and Lorentzian lines. In our opinion, the exponential nature of the wings of $g(\omega)$ is completely natu-

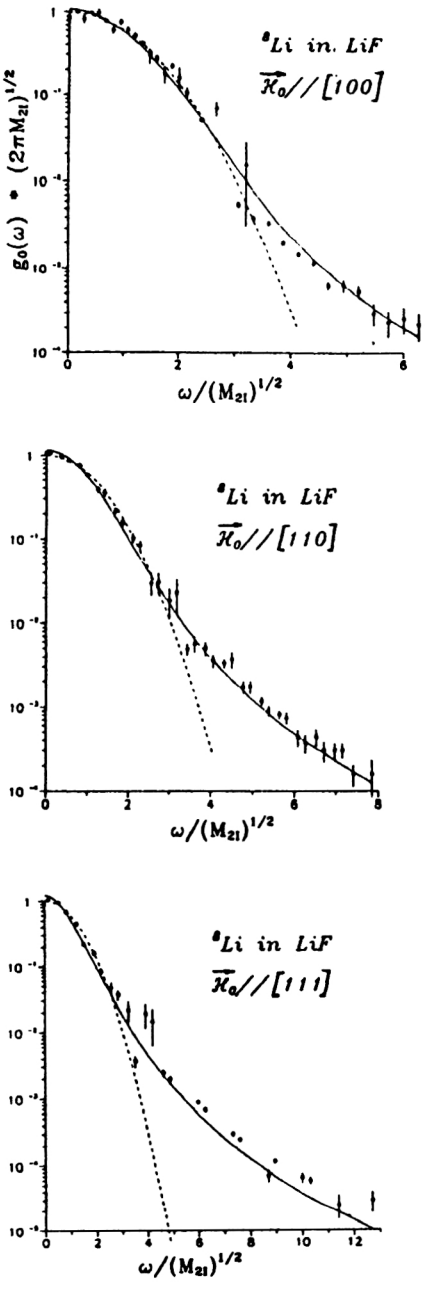

FIG. 2. NMR shape functions of β-active ^{8}Li nuclei in LiF crystals as a function of the frequency of the scanning rf field for various orientations of the crystals in a magnetic field $\mathcal{H}_0 = 2984.9(3)$ G at room temperature. The error in the value of the field was limited by its uniformity in the sample ($\approx 10^{-5}$). The concentration of paramagnetic centers was less than 10^{-5}. The solid lines are the result of calculating $g_0(\omega)$ using Eqs. (4) and (7)–(11). The dashed lines are Gaussians with theoretical second moments of the NMR shape function equal to $M_{21}/(2\pi)^2 = \langle \hat{\omega}_0^2 \rangle_0/(2\pi)^2 = 14$, 5.13, and 2.17kHz^2 for $\vec{\mathcal{H}}_0 \| [001]$, $\vec{\mathcal{H}}_0 \| [011]$, and $\vec{\mathcal{H}}_0 \| [111]$, respectively.

ral. It is well known that all the derivatives $d^n g(t)/dt^n$ are finite at $t=0$, as are all the moments $\int d\omega \cdot \omega^n g(\omega)$. There is no doubt that $g(t)$ is a smooth function. The NMR shape function $g(\omega)$ is non-negative. It therefore follows from the general properties of the Fourier transform[32] that $g(t)$ as a function of a complex variable can have singularities only for purely imaginary t. The one closest to $t=0$ determines the exponential asymptote of the NMR shape function. The Lorentzian function does not have finite moments, and the Gaussian $g(t)$ has a singularity only at infinity, so that both are excluded and are not suitable for interpretating the general case. Therefore, the exponential form of $g(\omega \to \infty)$ is quite natural, and the problem of the theory is not to prove this fact, but to determine the specific parameters of the asymptote.

The special feature of the problem of the line shape is that the spectrum of the Hamiltonian is not known and the time picture, where only the first few derivatives of the studied correlation functions at $t=0$ are actually known, is primary. Given these conditions, it is possible to state at least three important and general *a priori* principles which determine the advantage of using the model of a normal random process over the method of memory functions:[33]

(1) In the limit of slow fluctuations the model of a normal random process gives a Gaussian line in very good agreement with the exact solution. The same is true for the instantaneous distribution of the local field. To obtain similar results using the method of memory functions it is necessary to use considerably more complicated constructions.

(2) The model of a normal random process is the simplest generalization of a normal static distribution of local magnetic fields using a physically obvious hypothesis about the nature of their motion. There is no such clear picture in the method of memory functions.

(3) If the model of a normal random process itself is analyzed by the method of memory functions in lowest order in perturbation theory in the spin–field interaction and with realistic expressions for local-field correlators of the type (11), incorrect asymptotes of $g_0(\omega)$ are obtained both for high and for low frequencies, and also for $\langle I_0^+(t \to \infty) I_0^- \rangle_0$.

The results of Ref. 29 extend these arguments to arbitrary values of the frequencies and show that the hypothesis of normal fluctuations of the local field together with the general principles of the theory of irreversible processes gives a good and relatively elementary description of the phase relaxation of impurity nuclei, which is useful for interpreting the experimental data. This method is practically insensitive to details introduced in a model-dependent manner.

4. SHAPE FUNCTIONS OF THE CROSS-RELAXATION OF ^{8}Li AND ^{6}Li NUCLEI IN LiF

Multi-spin correlation functions are usually related indirectly to observables, via the equations of motion. For example, the transfer of polarization in the system of impurity nuclei is described by the equations[14]

$$\dot{p}_{\mathbf{r}0} = -\sum_{\mathbf{q}} (\nu_{\mathbf{qr}} p_{\mathbf{r}0} - \nu_{\mathbf{rq}} p_{\mathbf{q}0}), \quad p_{\mathbf{r}0}(t=0) = \delta_{\mathbf{r}0}, \qquad (12)$$

where $p_{\mathbf{r}0}(t) = \mathrm{Tr}(I_{\mathbf{r}}^z(t) I_0^z)/\mathrm{Tr}(I_0^z)^2$ is the polarization of an impurity nucleus located at the site $\mathbf{r}$ with the condition that the nucleus 0 was initially polarized; $\nu_{\mathbf{rq}}$ is the probability for polarization transfer between impurity nuclei located at sites $\mathbf{r}$ and $\mathbf{q}$ of the lattice [the summation in (12) runs only over impurity nuclei]. It is important that the $\nu_{\mathbf{r}0}$ are proportional to the cross-relaxation shape functions [see also Eq. (22)]

$$g_{\mathbf{r}0}^{-}(\Delta_{\mathbf{r}0})=\int_{-\infty}^{+\infty}\frac{dt}{2\pi}\cdot\exp(i\Delta_{\mathbf{r}0}t)\cdot g_{\mathbf{r}0}^{-}(t),$$

$$g_{\mathbf{r}0}^{-}(t)=\langle I_{\mathbf{r}}^{+}(t)I_{0}^{-}(t)I_{0}^{+}I_{\mathbf{r}}^{-}\rangle_{0}/\langle I_{\mathbf{r}}^{+}I_{0}^{-}I_{0}^{+}I_{\mathbf{r}}^{-}\rangle_{0}. \tag{13}$$

Disordered spin systems are more amenable to experimental study because the functions $g_{\mathbf{r}0}^{-}(\Delta_{\mathbf{r}0})$ depend on the distance $\mathbf{r}$ between the nuclei and the frequency difference $\Delta_{\mathbf{r}0}=\Omega_{\mathbf{r}}-\Omega_{0}$, where $\Omega_{\mathbf{r}}$ is the Larmor frequency of the nucleus located at site $\mathbf{r}$. In fact, if the impurity nuclei were ordered on a regular sublattice, the polarization of the ensemble of β nuclei for $p_{00}(t)\geq 1/e$ would fall off exponentially:

$$p_{00}(t)\approx\exp\left(-\sum_{\mathbf{r}}\nu_{r0}t\right). \tag{14}$$

In this case the depolarization kinetics of $p_{00}(t)$ is determined by a single number $\Sigma_{\mathbf{r}}\nu_{\mathbf{r}0}$ and it is difficult to study the $\mathbf{r}$ dependence of $g_{\mathbf{r}0}^{-}$. On the other hand, if the impurity nuclei are randomly distributed and their concentration $c\ll 1$ is small, the polarization of the β nuclei after their averaging $\langle\ldots\rangle_{\text{ave}}$ over position in the crystal has the form[14]

$$\langle p_{00}(t)\rangle_{c}=\exp(-M_{0}(t)-M_{1}(t)),$$

$$M_{0}(t)=c\cdot\sum_{\mathbf{r}}\left(\frac{1-\exp[-(\xi_{\mathbf{r}}+1)\nu_{\mathbf{r}0}t]}{\xi_{\mathbf{r}}+1}\right),$$

$$\xi_{\mathbf{r}}=\frac{I_{0}(I_{0}+1)}{I_{\mathbf{r}}(I_{\mathbf{r}}+1)}. \tag{15}$$

It turns out that $M_{1}(t)/M_{0}(t)<0.01-0.1$ for $M_{0}(t)\leq 1$ and $g_{\mathbf{r}0}^{-}(\Delta_{\mathbf{r}0})\approx g_{\mathbf{r}0}^{-}(0)$ (Refs. 12 and 14). Actually, the ratio $M_{1}(t)/M_{0}(t)$ depends on the magnetic field.[14] Equation (15) is much more complicated than (14), and more information can be obtained by comparing it with experiment.

In addition to the functions $g_{\mathbf{r}0}^{-}(\omega)$, β-NMR spectroscopy offers the possibility of experimentally studying the correlation functions

$$g_{\mathbf{r}0}^{+}(t)=\langle I_{\mathbf{r}}^{+}(t)I_{0}^{+}(t)I_{0}^{-}I_{\mathbf{r}}^{-}\rangle_{0}/\langle I_{\mathbf{r}}^{+}I_{0}^{+}I_{0}^{-}I_{\mathbf{r}}^{-}\rangle_{0}, \tag{16}$$

since they determine the intensity of the two-spin flip–flip resonance at frequency equal to the sum of the Larmor frequencies of ^{8}Li and ^{6}Li (Ref. 12). As a rule, two-spin correlation functions are approximated as

$$g_{\mathbf{rq}}^{\pm}(t)\approx g_{\mathbf{r}}(t)\cdot g_{\mathbf{q}}(t),$$

$$g_{\mathbf{r}}(t)=\langle I_{\mathbf{r}}^{+}(t)I_{\mathbf{r}}^{-}\rangle_{0}/\langle I_{\mathbf{r}}^{+}I_{\mathbf{r}}^{-}\rangle=g_{\mathbf{r}}(-t), \tag{17}$$

which is exact for $\mathbf{r}-\mathbf{q}\to\infty$.

In the Anderson–Weiss–Kubo model the evolution of the local fields is modeled as a normal random process, and for $g_{\mathbf{r}0}^{\pm}(t)$ we therefore obtain

$$g_{\mathbf{r}0}^{\pm}(t)=\exp\left(-\int_{0}^{t}d\tau(t-\tau)[\Lambda_{00}(\tau)\mp 2\Lambda_{\mathbf{r}0}(\tau)+\Lambda_{\mathbf{rr}}(\tau)]\right)$$

$$\cong\exp\left(-2\cdot\int_{0}^{t}d\tau(t-\tau)[\Lambda_{00}(\tau)\mp\Lambda_{\mathbf{r}0}(\tau)]\right). \tag{18}$$

The effect of the correlation of the local magnetic fields [i.e., violation of the approximation (17)] is expressed in the fact that the correlation function of the fields produced by the matrix spins at the impurity nuclei, $\Lambda_{\mathbf{r}0}(t)=\langle\hat{\omega}_{\mathbf{r}}(t)\,\hat{\omega}_{0}(0)\rangle_{0}$, is nonzero for $\mathbf{r}\neq 0$.

By analogy with (8), the expression for $\Lambda_{\mathbf{r}0}(t)$ can be written as

$$\Lambda_{\mathbf{r}0}(t)=\sum_{A=F,L}\Lambda_{\mathbf{r}0}^{(A)}(t),$$

$$\Lambda_{\mathbf{r}0}^{(A)}(t)=\langle\hat{\omega}_{\mathbf{r}}^{(A)}(t)\cdot\hat{\omega}_{0}^{(A)}(0)\rangle_{0}$$

$$=\frac{A(A+1)}{3}\sum_{\mathbf{x}\in A}\sum_{\mathbf{y}\in A}b_{\mathbf{rx}}^{IA}G_{\mathbf{xy}}^{A}(t)b_{\mathbf{y}0}^{AI}. \tag{19}$$

Using the Fourier transform on the lattice to find $\Lambda_{\mathbf{r}0}^{(A)}(t)$, we obtain the expression

$$\Lambda_{\mathbf{r}0}^{(A)}(t)=\frac{A(A+1)}{3}\int_{B}\frac{\Omega\cdot d^{3}k}{(2\pi)^{3}}[b^{IA}(\mathbf{k})]^{2}$$

$$\times\exp[i\mathbf{kr}-\mathscr{A}^{A}(\mathbf{k})t_{\text{eff}}^{(A)}],$$

$$\mathscr{A}^{A}(\mathbf{k})=\sum_{\mathbf{x}\in A}e^{-i\mathbf{ky}-x}\cdot\mathscr{A}_{\mathbf{yx}}^{A},\quad b^{IA}(\mathbf{k})=\sum_{\mathbf{x}\in A}e^{-i\mathbf{kx}}\cdot b_{0\mathbf{x}}^{IA}. \tag{20}$$

Here B is the Brillouin zone and $\Omega=2d^{3}$ is the volume of the elementary cell in the LiF crystal.

The strongest correlation of the local fields [or difference of the function $\Lambda_{\mathbf{r}0}^{(A)}(t)$ from zero] occurs at short distances between the cross-relaxing spins. In the opposite limit, for $\mathbf{r}\to\infty$, the cross-relaxation shape function reduces to the convolution of two NMR shape functions:

$$g_{\mathbf{r}0}^{-}(\omega)\to g_{\text{CR}}(\omega)=\int d\omega_{1}g_{0}(\omega-\omega_{1})\cdot g_{0}(\omega_{1}). \tag{17a}$$

The first of the experimental data that we obtained on the local-field correlation are shown in Fig. 3 for $\vec{\mathscr{H}}_{0}\|[001]$. We see that they correspond to a line significantly narrower than the function $g_{\text{CR}}(\omega)$ calculated as the convolution of two NMR lines as in (17) and (17a). A similar effect of line narrowing is also seen in Figs. 3 and 4 for other orientations of the crystal in the magnetic field and follows from the agreement between theory (Fig. 3c) and the experimental data (Fig. 5) in fields of 153.3 and 643 G.

It is important that for all three crystallographic orientations the NMR shape function $g_{0}(\omega)$ is nearly Gaussian in a fairly wide range near the peak $g_{0}(\omega)>10^{-2}g_{0}(\omega=0)$. This can be viewed as an illustration of the central limit theorem of probability theory: the local field at the β nucleus is made up of a large number (≥ 6) of roughly identically distributed contributions from the closest spins of the matrix. On the basis of (17) it might be expected that the cross-relaxation shape function should be "even more Gaussian." In fact, we

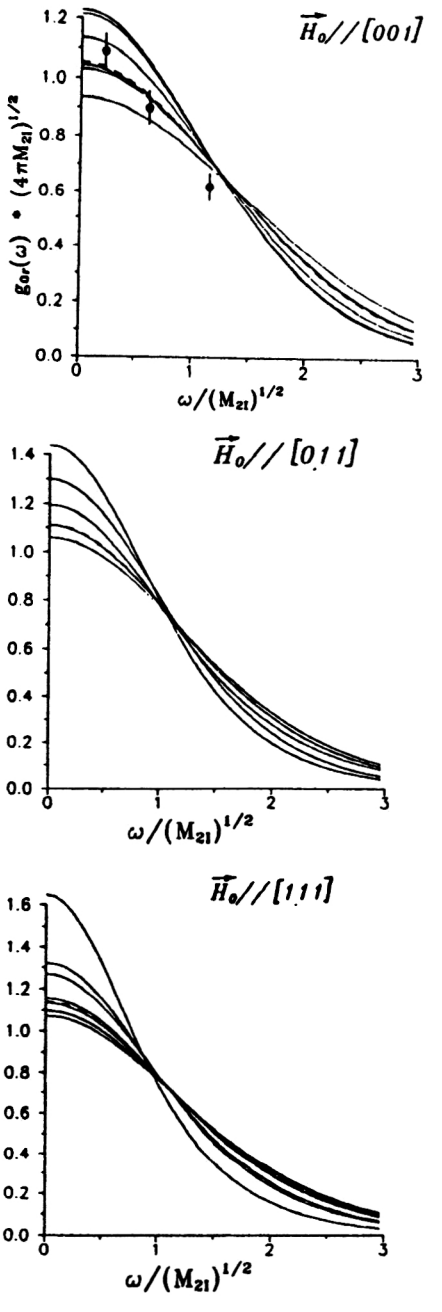

FIG. 3. Shape functions of the cross-relaxation of the ^{8}Li and ^{6}Li spins in LiF. The curves correspond to different relative locations of pairs of ^{8}Li and ^{6}Li nuclei in the crystal. The vector **r** connects the cross-relaxing nuclei. The dashed lines correspond to dilution of the nuclei at infinite distance, where $g_{r\to\infty,0}(\omega)$ is the convolution of two NMR shape functions. The points show the results of the fit of the experimental data on the depolarization kinetics of ^{8}Li nuclei in the ^{8}Li–^{6}Li system in fields of 218 G, 643 G, and 1200 G. The analysis was performed using Eq. (15), but neglecting the effect of local-field correlations, i.e., all the ν_{r0} were assumed to be proportional to the single parameter $g_{CR}(\omega)$, which was assumed to be fitted. The error of g_{CR} in the figure includes both the statistical uncertainty and the error in measuring the concentration of the isotope ^{6}Li.

have shown that the combination of Eqs. (18) and (19) leads to the same result, just as, when attempting to replace all the

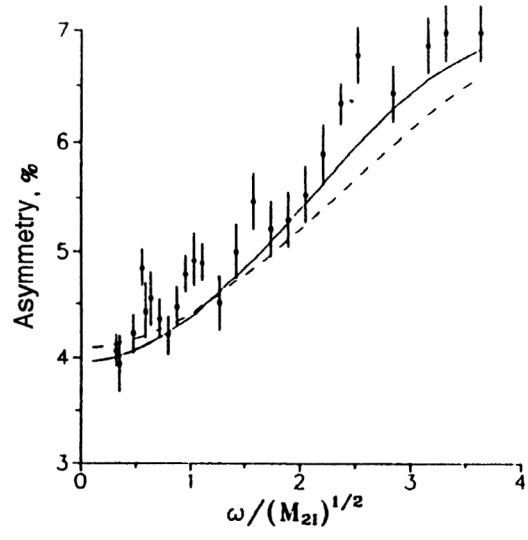

FIG. 4. Dependence of the integrated asymmetry of the β emission of ^{8}Li nuclei on the frequency difference (magnetic field) in LiF for $\vec{\mathscr{H}}_0 \| [110]$, $\tau_{irr} = \tau_{obs} = 4.1$ sec, and $\varepsilon_0 = 7.36(12)\%$. The solid line was obtained by taking into account the effect of the local-field correlation, and the dashed line was obtained without it.

cross-relaxation shape functions $g_{r0}(\omega)$ by one shape function independent of **r**, one finds significant narrowing and distortion of the cross-relaxation shape function compared with the estimate (17), so that it appears as violation of the central limit theorem. On the whole, it follows from the results of this section that our version of the model of a random process gives not only a better description of the NMR line shape, but also a good description of the more complicated effect of the local-field correlation on impurity spins.

5. DELOCALIZATION OF THE POLARIZATION IN A SPATIALLY DISORDERED SYSTEM OF ^{8}Li–^{6}Li NUCLEI

It was shown experimentally in Refs. 10 and 20 that in LiF crystals in fields $\mathscr{H}_0 > 150$ G depolarization (more precisely, delocalization of the polarization) of the ^{8}Li nuclei occurs as a result of cross-relaxation with ^{6}Li nuclei. The kinetics of this process is described by the equations[14]

$$\frac{\partial p_i}{\partial t} = -\sum_j (\nu_{ji} \cdot p_i - \nu_{ij} \cdot p_j), \quad p_i(t=0) = \delta_{i0}, \qquad (21)$$

where p_i is the polarization of the ith impurity nucleus. The transition rates ν_{ij} have the form

$$\nu_{ij} = \frac{\nu_{ij}^0 r_0^6}{r_{ij}^6} (1 - 3 \cdot \cos^2 \vartheta_{ij})^2, \quad i \neq 0 \neq j, \quad \nu_{ii} = 0,$$

$$\nu_{j0} = \frac{\nu_{ij}^1 r_0^6}{r_{j0}^6} (1 - 3 \cdot \cos^2 \vartheta_{j0})^2, \quad \nu_{0j} = \xi \cdot \nu_{j0},$$

$$\xi = \left. \frac{I_0(I_0+1)}{I_j(I_j+1)} \right|_{j \neq 0} = 3,$$

$$\nu_{ij}^0 = \frac{\pi}{6} \cdot (g_I^2 \beta_n^2 / \hbar r_0^3)^2 \cdot S(S+1) \cdot g_{ij}^-(0),$$

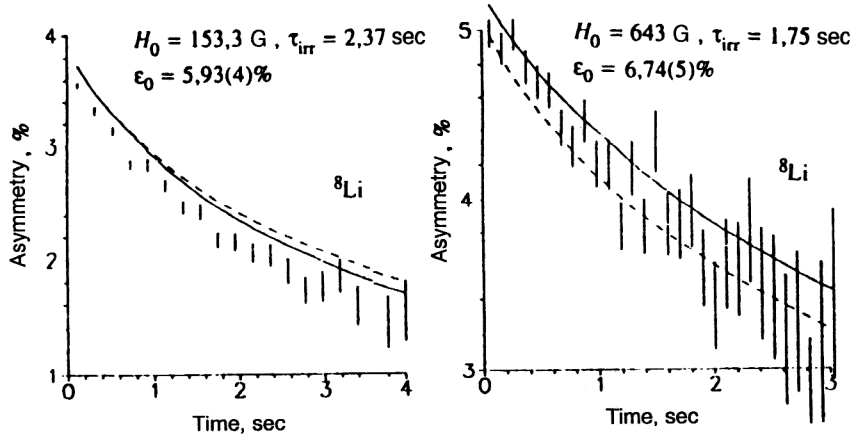

FIG. 5. Time dependence of the asymmetry of the β emission of ^{8}Li nuclei in LiF in various magnetic fields for $\vec{\mathscr{H}}_0 \| [111]$. The solid lines were obtained by taking into account the effect of the local-field correlation, and the dashed lines were obtained without it.

$$\nu_{ij}^1 = \frac{\pi}{6} \cdot (g_I^2 \beta_n^2 / \hbar r_0^3)^2 \cdot S(S+1) \cdot g_{ij}^-(\Delta). \tag{22}$$

Here Δ is the difference of the Larmor frequencies of the nuclei ^{8}Li and ^{6}Li.

We shall neglect the local-field correlations, setting $\nu_{ij}^0 = \nu_0$ and $\nu_{ij}^1 = \nu_1$. In the occupation-number representation,[14,34] in which one introduces the quantity $\tilde{P}_{xy}(t)$, the polarization of site x of the crystal at time t when at time $t=0$ only the site y was polarized, Eq. (21) becomes

$$\frac{\partial \tilde{P}_{xy}}{\partial t} = -\sum_z (n_z \nu_{zx} \tilde{P}_{xy} - n_x \nu_{xz} \tilde{P}_{zy}),$$

$$\tilde{P}_{xy}(t=0) = \frac{n_y}{c} \cdot \delta_{xy}. \tag{23}$$

Here $\nu_{xz} = \nu_{ij}$ ($\mathbf{r}_i = \mathbf{x}, \mathbf{r}_j = \mathbf{z}$); n_x is the occupation number of the site x, equal to 1 or 0 if the site x is or is not occupied by a ^{6}Li spin; ^{8}Li is located at the site y. The average over configurations is $\langle n_z \rangle = c$, where c is the dimensionless concentration of the isotope ^{6}Li. From the theoretical point of view this problem consists of calculating the polarization $P_{00}(t)$ of the β nuclei ^{8}Li averaged over the random distribution of ^{8}Li and ^{6}Li spins in the crystal. The problems related to the calculation of such quantities are reviewed in Refs. 13 and 14. The natural time scale in this problem is given by the Förster constant β_0 defined as

$$\left\langle \exp\left(-\sum_i \nu_{ij} t \right) \right\rangle_c = \exp(-\sqrt{\beta_0 t}), \quad j \neq 0,$$

$$\beta_0 = \frac{512}{243} \pi^3 c^2 \nu_0, \tag{24}$$

which is valid for an fcc lattice in the limit of small concentration, in which $c \to 0$ but $\beta_0 t$ is finite. For $\beta_1 t \leq 1$ and $\beta_1 \sim \beta_0$, a satisfactory solution of the problem can be obtained on the basis of the concentration expansion,[14,34] which leads to the expression

$$P_{00}(t) = \exp(-\sqrt{\beta_1 t/(\xi+1)} - \alpha \beta_1 t), \quad \beta_1 = \beta_0 \nu_1 / \nu_0. \tag{25}$$

The parameter α depends on $\mathscr{H}_0$ and can be calculated using the expressions and graphs given in Refs. 14 and 34. For $\mathscr{H}_0 = 0$, $\nu_1 = \nu_0$, and $\xi = 3$ we obtain $\alpha = 0.013$. Equation (25) is exact up to terms $\sim c^2$ inclusive.

When the ^{6}Li spins form a regular lattice, the problem of transport among them can be solved in quadratures, with the result that the long-time asymptote has diffusion form: $P_{00}(t) \sim t^{-3/2}$. Random systems are vastly more complicated, and the exact asymptote is unknown. However, it is natural to expect that it is also of the diffusion form. This result was first obtained analytically in Ref. 35 (see also Ref. 14), using the semiphenomenological theory formulated in that study. This theory is based on the fact that the averaged propagator $P_{xy}(\lambda)$ satisfies the equation

$$\lambda P_{xy} = \delta_{xy} - \sum_z (N_{zx} P_{xy} - N_{xz} P_{zy}),$$

$$N_{xz} = N_{xz}(\lambda), \quad P_{xy} = P_{xy}(\lambda), \tag{26}$$

which is very similar to the Laplace transform of Eq. (23):

$$\lambda \tilde{P}_{xy} = \frac{n_y}{c} \delta_{xy} - \sum_z (n_z \nu_{zx} \tilde{P}_{xy} - n_x \nu_{xz} \tilde{P}_{zy}). \tag{27}$$

After introducing the hypothesis that the kernels $N_{zx}(\lambda)$ must also describe processes simpler than the basic one (27), the process for which the configuration averaging is exact is selected. From this the kernels $N_{zx}(\lambda)$ are determined, and then P_{xy} is calculated from Eq. (26). In this analysis it is necessary to systematically take into account (1) the probability that a node can be reliably assumed to be occupied by the ^{8}Li spin, when in the other medium a single impurity spin is associated on the average with $1/c$ lattice sites, and (2) the detailed analytic characteristics of the memory functions associated with the long-range behavior of the dipole–dipole interaction. As a result of matching the first two terms of the asymptotes at large and intermediate times, the authors of Refs. 14 and 35 obtained

$$P_{00}(t) = Q_0(t) + \xi \cdot \frac{1 - Q_0(t)}{(\mu \beta_0 (t+\tau))^{3/2}}$$

$$\times\left(1+\frac{\varphi}{(\mu\beta_0(t+\tau))^{1/2}}\right), \qquad (28)$$

where $Q_0(t)=\exp(-\sqrt{\beta_1 t})$, $\varphi=2.09$, $\mu=0.889$, and $\mu\beta_0\tau=5.11$. We refer the reader to Refs. 14 and 42 for a discussion of the later development of the theory.

The ^{8}Li lifetime is limited ($T_{1/2}=0.84$ sec), so that study of the time development of the process of delocalization of the ^{8}Li polarization in the ^{8}LiL–^{6}Li system in LiF and, consequently, the possibility of measuring the asymptotic (diffusion) regime essentially depends on the cross-relaxation rate, which is proportional to the cross-relaxation shape function. This rate can be varied in fields $\mathscr{H}_0\approx 150$ G by choosing the crystallographic orientation of the sample, changing the ^{6}Li spin density, or artificially changing the widths of the resonance lines of ^{8}Li and ^{6}Li by exposing the LiF crystal to strong (i.e., with amplitude greater than that of the local fields in the crystal) rf fields (Bloch narrowing). The spin motion induced by these fields narrows the NMR shape function $g_0(\omega)$ and the cross-relaxation shape function $g_{CR}(\omega)$, which is the convolution of the NMR shape functions of ^{8}Li and ^{6}Li. The narrowing is effective (i.e., all the rates ν_{ij} increase) as long as $g_{CR}(\Delta)\approx g_{CR}(0)$, where Δ is the difference of the Larmor frequencies of the ^{8}Li and ^{6}Li nuclei. For very narrow lines $g_{CR}(\Delta)\ll g_{CR}(0)$ and the spin dynamics of these nuclei are independent, since for $\mathbf{r}_i \sim |\mathbf{r}_k - \mathbf{r}_l|$ we have $\nu_{i0}\ll\nu_{k1}$, $i\neq 0, k\neq 0\neq 1$. In the absence of narrowing the NMR and cross-relaxation shape functions are nearly Gaussian, because the phase-relaxation time of the ^{8}Li and ^{6}Li nuclei is smaller than the time for flip–flop processes of the neighboring nuclei. We note that when narrowing is present in the experiments described above the difference of these times is increased even more, and therefore we shall always assume that the NMR and cross-relaxation shape functions are Gaussians with second moments related as $M_{2CR}=M_{2l}(^8\text{Li})+M_{2l}(^6\text{Li})\approx 2M_{2l}(^8\text{Li})$.

The narrowing of the NMR shape function is studied by putting the sample in a weak scanning rf field with amplitude $\mathscr{H}_1$ and frequency ω, varied near the Larmor frequencies of ^{8}Li and ^{6}Li.

The experimental data on the measurement of the NMR shape function of ^{8}Li nuclei and their depolarization kinetics in LiF crystals in the natural and the accelerated cross-relaxation processes are shown in Fig. 6. In fitting them by the dependences (25) and (28) we took into account the relation between β_1 and β_0: $\beta_1=\beta_0\exp(-\Delta^2/2M_{2CR})$, where Δ is the difference of the Larmor frequencies of ^{8}Li and ^{6}Li [$\Delta=2\pi(0.55$ kHz, in a field of 153.4 G]. Therefore, the fit was actually made in a single parameter, β_0. The dependences $\varepsilon(t)$ and $\tilde{\varepsilon}(\nu)$ were processed together.

The fact that the experimental values of β_0 and β_1 for the orientation $\vec{\mathscr{H}}_0\|[111]$ in the absence of acceleration turned out to be slightly larger than the theoretical values is related to the local-field correlations (see Sec. 4). When the lines are narrowed, the discrepancy between the calculated and measured values of β_0 and β_1 increases slightly (see the table in Ref. 12 and the accompanying discussion). Nevertheless, the agreement between theory and experiment can be deemed satisfactory over the entire range of times studied,

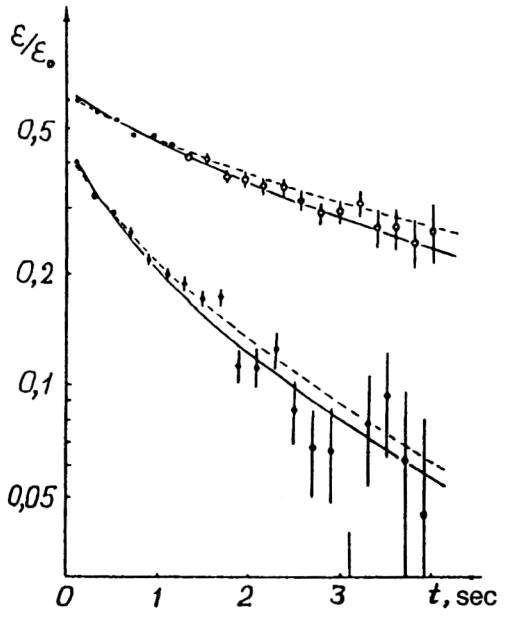

FIG. 6. Time dependence of the asymmetry of the β emission of ^{8}Li nuclei in LiF. The points ○ are for the natural cross-relaxation process for $\vec{\mathscr{H}}_0\|[111]$, $\mathscr{H}_0=153.4$ G, $\tau_{irr}=2.37$ sec, and $\varepsilon_0=5.93(4)\%$, and the points ● are for the accelerated cross-relaxation process for $\vec{\mathscr{H}}_0\|[111]$, $\mathscr{H}_0=153.4$ G, $\tau_{irr}=2.4$ sec, and $\varepsilon_0=6.19(4)\%$. The solid lines were calculated using Eq. (28), and the dashed lines using (25).

i.e., for $\beta_0 t\leq 15$. It is important that for $\beta_0 t\leq 5$ Eq. (25) and its analog from Sec. 4 give such close agreement with experiment, which can apparently be used to obtain structural information about matter.

6. DELOCALIZATION AND DESTRUCTION OF POLARIZATION UNDER THE INFLUENCE OF THE TWO-SPIN RESONANCE

Depolarization as a result of cross-relaxation under the conditions of rf exposure at a frequency equal to the sum of the Larmor frequencies of ^{8}Li and ^{6}Li is described by the equations[12]

$$\frac{\partial p_i}{\partial t}=-\sum_j(\nu_{ji}p_i-\nu_{ij}p_j)-\sum_j(\mu_{ji}p_i+\mu_{ij}p_j),$$

$$p_i(t=0)=\delta_{i0}, \qquad (29)$$

where the rates of elementary resonance two-spin transitions are given by

$$\mu_{ij}=\mu_{ij}^0\cdot(r_0/r_{ij})^6\cdot(3\sin 2\vartheta_{ij})^2,$$

$$\mu_{ij}^0=\frac{\pi}{6}\left(\frac{\mathscr{H}_1}{\mathscr{H}_0}\right)^2\left(\frac{g_i g_j\beta_n^2}{\hbar\cdot r_0^3}\right)^2\cdot I_i(I_i+1)\cdot g_{ij}^+(\omega-\omega_i-\omega_j). \qquad (30)$$

Here $g_{ij}^+(\Delta)$ is the shape function of the two-spin resonance line, approximated as a convolution of Gaussian NMR shape functions, g_i and ω_i are the g factor and the Larmor frequency of the ith spin of the ^{8}Li–^{6}Li system, and $\mathscr{H}_1$ is the amplitude of the rotating rf field. Analysis of the first few terms of the concentration expansion gives[12]

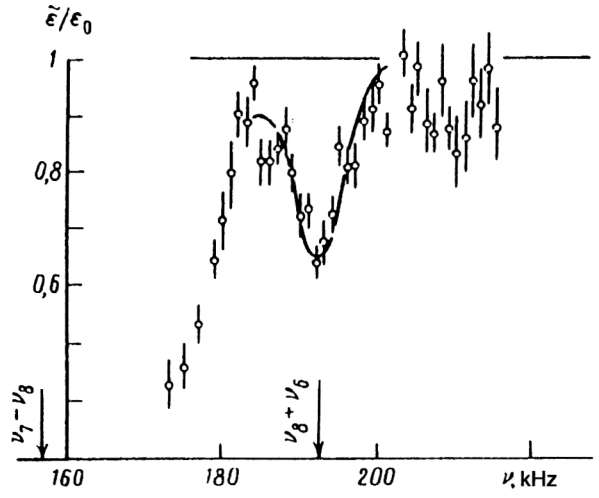

FIG. 7. β-NMR spectrum of ^{8}Li nuclei in LiF in the vicinity of the two-spin resonance $\omega_I+\omega_S$. Here $\vec{\mathscr{H}}_0\|[111]$, $\mathscr{H}_0=153.4$ G, ν is the frequency of the scanning rf field with amplitude $\mathscr{H}_1=10.4$ G, $\tau_{irr}=2.4$ sec, $\tau_{obs}=4.1$ sec, and $\varepsilon_0=6.19(4)\%$. The curves obtained using Eqs. (25) and (28) practically coincide.

$$P_{00}(t)=\langle p_0(t)\rangle_c=\exp(-\sqrt{\beta_e t}/2-\alpha_e\beta_e t),$$

$$\beta_e=\beta_1\cdot\left(1+\frac{27}{4}\cdot\sqrt{\gamma}\right)^2,\quad \alpha_e=0.014\cdot(1+21.5\cdot\sqrt{\gamma}),$$

$$\gamma=\mu_{i0}^0/\nu_0\propto(\mathscr{H}_1/\mathscr{H}_0)^2\ll 1. \tag{31}$$

Here we have used the facts that $\xi=3$ and $\beta_0\approx\beta_1$, and the value of α_e is quoted for $\mathscr{H}_0=153.4$ G. The calculation was carried out in the leading order in γ. Equation (31) indicates that the relative change $\delta\beta/\beta_1=(\beta_e-\beta_1)/\beta_1=\text{const}\sqrt{\gamma}$ of the effective depolarization rate in a disordered system is significantly larger than the ratio of the elementary rates $\mu_{i0}^0/\nu_0\sim\gamma\ll 1$. The physical essence of this effect of enhancement of the depolarization process is that the two-spin resonance violating the conservation law $\Sigma_i p_i=1$ of cross-relaxation makes it possible to achieve complete depolarization in closely spaced pairs of ^{8}Li–^{6}Li nuclei [it is such pairs which determine the behavior of $P_{00}(t)$ for $\beta_e t\leqslant 1$].

We note that this reason and even the enhancement effect itself, which arise from the violation of the conservation laws of a stronger process by a weaker process, must be rather general in the physics of relaxation processes.

The measured line shape of the two-spin resonance $\omega_I+\omega_S$ and the dependence of the β-emission asymmetry ε on time and on the amplitude of $\mathscr{H}_1$ (for $\omega=\omega_I+\omega_S$) are given in Figs. 7–9. The solid lines in the figures correspond to the value $\beta_1=1.32(2)$ sec^{-1}. The theoretical value is $\beta_1=1.06$ sec^{-1}. A possible reason for the difference between these quantities is given in Sec. 4. The data on $\varepsilon(t)$ are satisfactorily described by (31) up to $\beta_1 t\approx 5$. The numerical coefficient in front of $\sqrt{\gamma}$ in the expression for β_e obtained as a result of the fit is equal to 6.0(3), while its theoretical value is 6.75. This excess is apparently related to neglect in the calculation of the "forbidden" volume ($\sim r_0^3$) for ^{6}Li around the β nucleus ^{8}Li.

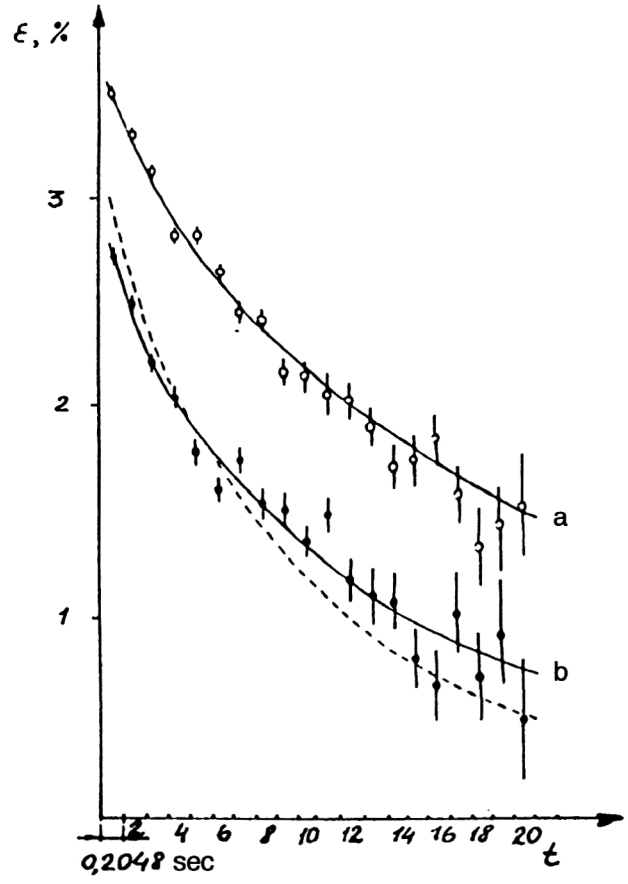

FIG. 8. Time dependence of the asymmetry of the β emission of ^{8}Li nuclei in LiF. Here $\vec{\mathscr{H}}_0\|[111]$, $\mathscr{H}_0=153.4$ G, $\tau_{irr}=2.4$ sec, and $\varepsilon_0=5.93(4)\%$. The points O are for the natural cross-relaxation process in the ^{8}Li–^{6}Li system (without an rf field), and the points ● are the same with an rf field of frequency $(\omega_I+\omega_S)/2\pi=192.8$ kHz and amplitude $\mathscr{H}_1=10.4$ G. The solid lines were calculated using Eq. (31), and the dashed line is the result of the fit assuming that the enhancement effect is absent, i.e., that the resonance depolarization is monoexponential. The experimental estimate of the second moment of the resonance $\omega_I+\omega_S$ gives 5.5 ± 1.5 kHz^2, which is consistent with the theoretical value of 4.3 kHz^2. If the depolarization process were a consequence of quadrupole interactions of ^{8}Li with crystal defects at frequency $2\omega_I\approx\omega_I+\omega_S$, the value should have been about 8.6 kHz^2.

7. DEPOLARIZATION AS A CONSEQUENCE OF CROSS-RELAXATION IN THE PRESENCE OF TRANSLATIONAL DIFFUSION OF LITHIUM NUCLEI

The effect of the spatial motion of the lithium ions on the process of polarization delocalization in the ^{8}Li–^{6}Li system of the LiF crystal becomes important at temperatures $T>500$ K. As T increases the effectiveness of cross-relaxation grows, because the spatial diffusion of lithium nuclei begins to outstrip the spin–spin delocalization of the polarization. The main relaxation mechanism is the transfer of polarization to ^{6}Li spins close to ^{8}Li and the subsequent transfer of the polarization to large distances owing to the spatial dispersal of these nuclei.[12]

To describe this process quantitatively, the concentration expansion developed for the case of static disorder was generalized to include the translational, spatially uncorrelated motion of the nuclei. In the leading order in the concentration of ^{6}Li nuclei,

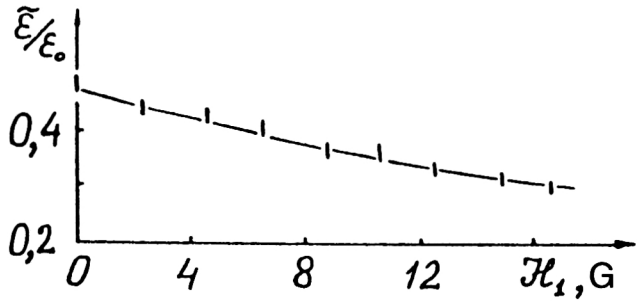

FIG. 9. Dependence of the asymmetry of the β emission of ^{8}Li nuclei in the LiF crystal on the amplitude $\mathcal{H}_1$ of the rf field at the frequency 204 kHz of the two-spin resonance $\omega_I+\omega_S$ in the field $\mathcal{H}_0=162.7$ G, $\vec{\mathcal{H}}_0\|[111]$, $\tau_{irr}=2.45$ sec, $\tau_{obs}=3$ sec, and $\varepsilon_0=5.69(4)\%$. The data were processed using Eqs. (31).

$$\langle p_0(t)\rangle = e^{-M(t)}, \quad M(t)=c\cdot\sum_{\mathbf{r}}[1-p_0^{(1)}(\mathbf{r},t)]. \tag{31}$$

Here $p_0^{(1)}(\mathbf{r},t)$ is the polarization of the ^{8}Li nucleus interacting only with a single ^{6}Li spin, which at time t is separated from ^{8}Li by the vector $\mathbf{r}$, and taking into account the preceding translational motion of the two spins.[12] In the Laplace representation $p_0^{(1)}(\mathbf{r},\lambda)$ is calculated using the equations

$$\lambda p_0^{(1)}(\mathbf{r},\lambda)=1-\nu_{r0}p_0^{(1)}+\xi\nu_{r0}p_0^{(1)}-2\hat{\kappa}p_0^{(1)},$$
$$\lambda p_1^{(1)}(\mathbf{r},t)=-\xi\nu_{r0}p_1^{(1)}+\nu_{r0}p_0^{(1)}-2\hat{\kappa}p_1^{(1)}$$
$$\hat{\kappa}f(\mathbf{r})=\kappa f(\mathbf{r})-\frac{\kappa}{Z}\cdot\sum_{\mathbf{r}_1}f(\mathbf{r}+\mathbf{r}_1). \tag{32}$$

Here the definition of the hopping-transport operator $\hat{\kappa}$ involves the sum over the $Z=12$ nearest neighbors in the lithium sublattice, and the coefficient of 2 in front of $\hat{\kappa}$ arises because both the ^{8}Li and the ^{6}Li spins move.

In the case of slow motion, when the hopping frequency κ of the lithium ions is small compared with the cross-relaxation rate ν_1, in the limit of a continuous medium the leading correction in κ to the static limit $[M(t,\kappa=0)=\sqrt{\beta_1 t}/2]$ coincides with the semiclassical approximation for the propagator close to its stationary point $\cos^2\vartheta_{r0}=1/3$, at which the cross-relaxation probability [see (22)] vanishes.[12] Finally, for $\xi=3$ we obtain

$$M(t)=\sqrt{\beta_1 t}/2+1.69c\kappa^{3/4}\nu_1^{1/4}t. \tag{32a}$$

The derivation of this expression is valid as long as the second term is small compared with the first, that is, for $\kappa^{3/4}\nu_1^{-1/4}t^{1/2}\ll 1$. In the case where $\kappa^{3/4}\nu_1^{-1/4}t^{1/2}\gg 1$ and $\kappa/\nu_1\ll 1$, $M(t)$ can be calculated by the method developed in the theory of the scattering of slow particles (scattering-length theory).[36] Here it turns out that $M(t)$ has the same form as in (32a), but with a somewhat larger numerical coefficient (equal to 1.76) in the second term. The difference between these coefficients was neglected when comparing theory with experiment.

For the case of fast motion ($\kappa\gg\nu_1$) it is possible to construct an expansion for $M(t)$ as a continued fraction in ν_1/κ. Here in leading order in ν_1/κ we obtain $M(t)=Zc\nu_1 t$. More accurate expressions for the effective coordination number Z are given in Ref. 12.

In Figs. 10–12 we show the experimental and calculated data on the effect of the temperature of LiF crystals on the depolarization of impurity β-active ^{8}Li nuclei. This effect becomes important at sample temperatures $T>500$ K. The dependences $\varepsilon(t)$ (Fig. 11) confirm the predicted change of the depolarization kinetics of ^{8}Li from $\ln P_{00}\sim-\sqrt{\beta_1 t}$ occurring in the absence of motion at room temperature to $\ln P_{00}\sim-\beta_1 t/c$ realized in the case of sufficiently rapid motion of the lithium nuclei at $T>600$ K. All the data were processed together using the Arrhenius approximation for the frequency of translational migrations of lithium:

$$\kappa=c_v\tau_v^{-1}, \quad c_v=\exp(S_f/k-h_f/kT),$$
$$\tau_v^{-1}=\nu_D\exp(S_m/k-h_m/kT), \tag{33}$$

where c_v is the lithium vacancy concentration, τ_v^{-1} is the vacancy hopping frequency, ν_D is the Debye frequency in LiF, S_f and S_m are the entropies and h_f and h_m are the enthalpies of lithium vacancy creation and migration, k is the Boltzmann constant, and T is the temperature.

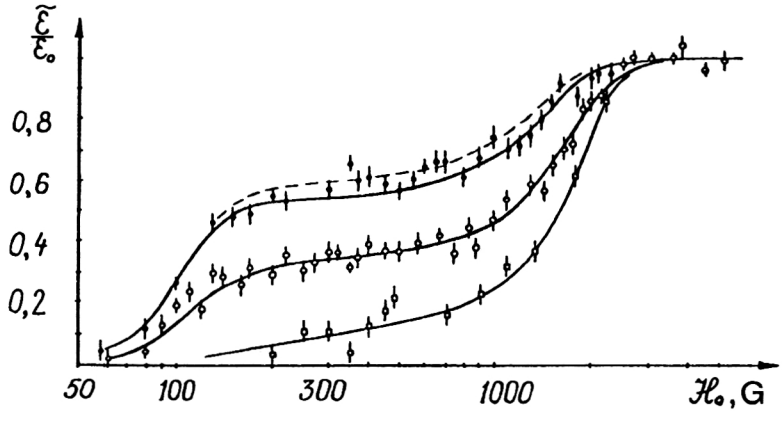

FIG. 10. Dependence of the asymmetry of the β emission of ^{8}Li nuclei in LiF on the magnetic field $\vec{\mathcal{H}}_0\|[110]$ for various sample temperatures, $\tau_{irr}=\tau_{obs}=4.1$ sec, $\varepsilon_0=7.36(12)\%$. The points ● are for $T=296$ K and $M_{2CR}=6.4(4)$ kHz^2; ○ are for $T=540$ K and $M_{2CR}=7.5(4)$ kHz^2; and □ are for $T=577$ K and $M_{2CR}=7.1(4)$ kHz^2. The theoretical value of M_{2CR} for the orientation [110] is 10.2 kHz^2.

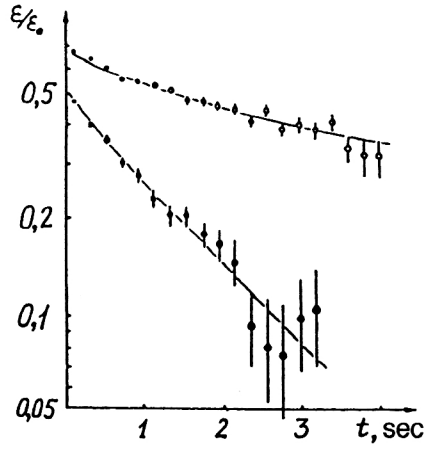

FIG. 11. Time dependence of the asymmetry of the β emission of ^{8}Li nuclei of LiF crystals for $\mathscr{H}_0=200$ G, $\vec{\mathscr{H}}_0\|[110]$, $\tau_{irr} = \tau_{obs}=4.1$ sec, and $\varepsilon_0=7.36(12)\%$. The points O are for 296 K, and the points ● are for 540 K.

We have also taken into account the quadrupole interaction of ^{8}Li with the diffusing lithium vacancies, and the interaction constant at minimum impact parameter was assumed to be $\beta_Q=3\omega_Q=2\pi\cdot 11.7(4)$ kHz.[20,37] In fields <150 G the depolarization associated with cross relaxation of ^{8}Li with ^{7}Li and ^{19}F was taken into account.

The processing of $\tilde{\varepsilon}(\mathscr{H}_0)$ for $g_{CR}(\Delta)\approx g_{CR}(0)$ and $\kappa=0$ can be done in the continuous-medium approximation, where the sum $M^{(1)}(t)=[c/(\xi+1)]\Sigma_r \times (1-\exp(-(\xi+1)\nu_{r0}t))$ is replaced by an integral. Here $M^{(1)}(t)=\sqrt{\beta_1 t/(\xi+1)}$ in (31). The continuous-medium approximation is valid when $c\to 0$ and $\beta_1 t$ is finite or, equivalently, when $c\ll 1$ and $\exp(-(\xi+1)\nu_1 t)\ll 1$. The latter inequality is violated in strong fields, and the calculation using the exact expression for $M^{(1)}(t)$ leads to a stronger dependence of the polarization on $\mathscr{H}_0$. The result of processing $\tilde{\varepsilon}(\mathscr{H}_0)$ at room temperature taking into account this improvement and assuming that the cross-relaxation shape function is Gaussian with variable second moment M_{2CR} is shown by the solid line in Fig. 10 and gives a value $M_{2CR}=6.4(4)$ kHz^2 somewhat larger than that from the fit using the continuous-medium approximation [5.9(4) kHz^2]. The theoretical value of M_{2CR} for a rigid lattice neglecting local-field correlations is 10.2 kHz^2. The fit of the data in the parameters β_0 and M_{2CR} [with the constraint $\beta_1=\beta_0\exp(-\Delta^2/2M_2^c)$, where Δ is the difference of the Larmor frequencies of ^{8}Li and ^{6}Li] apparently leads to better agreement with experiment [dashed line in Fig. 10, $M_{2CR}=5.9(4)$ kHz^2, $\beta_0=0.68(2)$ sec^{-1} for $\beta_0^{theor}=0.71$ sec^{-1}], but the discrepancy in M_{2CR} remains significant. The curve for $\tilde{\varepsilon}(\mathscr{H}_0)$ at room temperature was processed separately without using the continuous-medium approximation and including the effect of the local-field correlation. It has been shown in Sec. 4 that this leads to considerably better agreement between the shapes of the theoretical and experimental lines, but on the whole the calculated relaxation rates are somewhat larger than the measured ones.

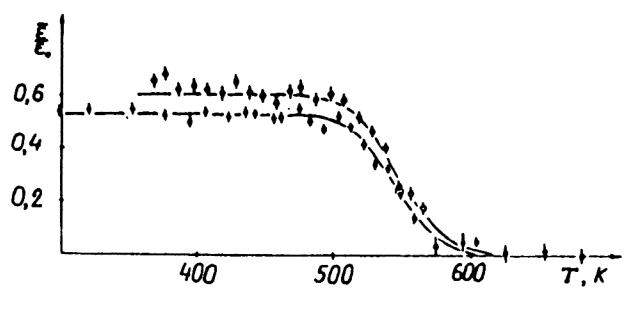

FIG. 12. Temperature dependence of the asymmetry of the β emission of ^{8}Li nuclei in the crystal LiF for $\mathscr{H}_0=200$ G, $\tau_{irr}=2.4$ sec, and $\varepsilon_0=7.36(12)\%$. The points O are for $\vec{\mathscr{H}}_0\|[100]$, and the points ● are for $\vec{\mathscr{H}}_0\|[110]$.

It is impossible to independently determine κ_0 and h from the experimental data, but the ratio $h/\ln\kappa_0=0.104(1)$ is determined, where h is measured in eV and κ_0 in sec^{-1}. Physically, at temperature $T_0=h/(\kappa\ln\kappa_0)\approx 540$ K the time $(\kappa c^{2/3})^{-1}$ for lithium to move a distance equal to the average distance ($\sim r_0 c^{1/3}$) between the ^{6}Li nuclei in LiF is set equal to the inverse cross-relaxation rate $(\nu_1 c^2)^{-1}$ of the two ^{8}Li and ^{6}Li spins at average separation. This leads to the expression $\kappa c^{2/3}\sim\nu_0 c^2$ (or in this case to $\kappa\sim 1$ sec^{-1}), which determines the region of maximum sensitivity of the β-NMR method for nuclear translational motion. Using the value $\nu_D=1.5\times 10^{13}$ sec^{-1} (Ref. 38) and the sum $S_f+S_m=2.3k$ (Ref. 39), we obtain $\kappa_0=1.8\times 10^{15}$ sec^{-1} and $h=1.58$ eV, in agreement with the values obtained by other methods.

8. CONCLUSION

Let us discuss some of the natural ways in which this research can be extended.

The NMR shape function has a very complicated dependence on the various parameters of the spin system of the matrix. Nevertheless, we think that in our experiments the theory has been verified fairly completely for the case in which the magnetic moments of the impurity nuclei are smaller than those of the matrix nuclei. Clearly, in the opposite limit it is necessary to take into account the effect of the impurity on the evolution of the matrix spins, which considerably complicates the theory. It would therefore be interesting to find how far in this direction our theory is applicable.

The study of the cross-relaxation shape function and the local-field correlation is considerably more complicated, primarily because Eq. (25) has a very limited range of applicability in the ratio β_0/β_1 and, therefore, in the magnitude of the external magnetic field. In particular, it is insufficient for studying the line wings. Therefore, in the near future it would be wise to make a more detailed check of the field and time dependences of the theory for various orientations of the crystals in moderate magnetic fields. It would be interesting to understand the extent to which such dependences can be used to solve the inverse problem of studying the correlations in the spatial location of the ^{6}Li spins.

The field of research in the long-time asymptotics of random walks in disordered media appears to be extremely broad. First of all, experimental progress in this area is necessary. From the theoretical side, further study of asymptotically exactly solvable models (see, for example, Refs. 40 and

41) may be helpful for making them more similar to realistic systems. It would also be useful to analyze the field[15] and T-matrix[14] representations for the propagators and, finally, to perform direct numerical modeling of such processes.

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