Nuclear spin-lattice relaxation and spin diffusion in an inhomogeneous field

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The theory of spin diffusion is extended to the case of spin-lattice relaxation and spin diffusion in an inhomogeneous magnetic field. Two coupled equations describing the mutual relaxation and the spin diffusion of nuclear magnetization and dipolar energy were obtained using the method of the nonequilibrium state operator. The equations were solved for short- and long-time approximations corresponding to the direct and diffusion relaxation regimes. It is shown that at the beginning of the relaxation process in the mixed state of the conventual superconductor the direct relaxation regime is dominant. The nuclear magnetization decays with a stretched exponential ($\alpha = 0.5$) while the dipolar energy decreases exponentially. Then the relaxation regime changes both for nuclear magnetization and dipolar energy, to the diffusion one described by the exponential time dependence which agrees with the experiment. The radii of the diffusion barrier and the spin-diffusion coefficient were estimated.

I. INTRODUCTION

Studies of the NMR of nuclei have demonstrated that spin diffusion plays an important role in the relaxation of nuclei in the presence of paramagnetic impurities.[1–3] However, most of them deal with the process of spin diffusion in homogeneous magnetic fields.[4,5] The spin-diffusion processes result from dipole-dipole interaction (DDI) between nuclear spins I and between nuclear spins I and paramagnetic impurity (PI) spins S.[4,5] The DDI between nuclear spins I and PI spins S leads to a direct spin-lattice relaxation of the nuclear-spin system. Due to the inverse sixth power dependence on the distance between nuclei and the PI, the local nuclear magnetization reaches its equilibrium value at a faster rate near the PI's.[4,5] Therefore the nuclear magnetization will be a function of the position. This induces the spatial diffusion of the nuclear Zeeman energy in homogeneous magnetic fields by flip-flop transitions due to the DDI's between neighboring nuclear spins. The diffusion coefficient D has values of the order of $10^{-12} \div 10^{-13}$ cm²/sec for inorganic solids and $10^{-14} \div 10^{-16}$ cm²/sec for organic solids.[5]

The role of the DDI between PI spins in the nuclear-spin diffusion and in the dynamics polarization of nuclei has been considered in detail.[5,6] It was shown that the relaxation process is described by sum of two exponentials. For the DDI between nuclear spins, most theories consider only the transfer function of the Zeeman energy.[1–3] Only a few papers tried to take into account other functions of the spin-spin interactions.[7–10] For example, the theory was extended to include a diffusion of the spin-spin energy in an inhomogeneous magnetic field,[7] an exchange energy in He³,[8] and the transport of spin-spin energy was calculated numerically for classical gyromagnets, coupled by truncated dipole-dipole and nearest-neighbor exchange interactions.[9]

However, as is well know,[11–14] the DDI between nuclear spins plays an important role in spin thermodynamics in solids. On the one hand, the secular part of the nuclear DDI's form an independent energy reservoir[11,13–15] with its own spin temperature, which can be different from the spin temperature of the Zeeman reservoir, and, on the other hand, the

nuclear DDI's stimulate the relaxation processes which bring a nuclear-spin system into a thermal equilibrium state.[13]

As was stressed by Genack and Redfield,[7] the spin-diffusion processes no longer exactly conserve Zeeman energy in an inhomogeneous field because the Zeeman interaction energy with applied field is not identical for neighboring nuclear spins. In order for the spin-diffusion process to materialize the Zeeman energy difference must be taken up by the dipole-dipole energy reservoir. A situation very similar to this was considered by Provotorov.[11,13] Using an intuitive approach, coupled equations were proposed for magnetization and dipolar energy, describing anomalous rapid relaxation in mixed-state superconducting vanadium.[7]

Recently a theory for spin-lattice relaxation and spin diffusion of nuclear dipolar order via PI's has been developed.[16,17] Nuclear dipolar order is characterized by a state with nuclear spins oriented along an internal local field generated by DDI and it is characterized by a dipolar temperature.[12–15] The Zeeman order is characterized by a state with nuclear spins oriented along an external magnetic field. Usually, the degree of Zeeman order is described by a magnetization of the sample or by a spin temperature.[13] Here we consider the phenomena of spin-lattice relaxation and spin diffusion both of Zeeman and dipolar orders of the nuclear spins due to their DDI in solids containing PI's in an inhomogeneous magnetic field. As an example of the application of the theory that was developed in this paper we consider spin-lattice relaxation and spin diffusion in type-II conventional superconductors.

II. THEORY

A. Diffusion equations

Let us consider a spin system consisting of nuclear and PI spins, localized in an inhomogeneous magnetic field $\vec{H}_0(\vec{r})$, at positions $\vec{r}_\mu$ and $\vec{r}_j$, respectively. Here the Greek indices indicate the nuclei and the Latin the impurities.

The dynamics of the system and its relaxation can be described by the solution of the equation for the density matrix $\sigma(t)(\hbar = 1)$,

$$i\frac{d\sigma(t)}{dt}=[\mathcal{H},\sigma(t)] \tag{1}$$

with the Hamiltonian

$$\mathcal{H}=\mathcal{H}_Z+\mathcal{H}_{dd}+\mathcal{H}_{IS}+\mathcal{H}_S, \tag{2}$$

where $\mathcal{H}_Z$ is the Hamiltonian of the Zeeman interaction,

$$\mathcal{H}_Z=\sum_\mu \omega_0^\mu I_\mu^z, \tag{3}$$

$\omega_0^\mu=\gamma_I H_0(\vec{r}_\mu)$, and γ_I is the gyromagnetic ratio of the nuclei. $\mathcal{H}_{dd}$ is a specific part of the nuclear DDI Hamiltonian

$$\mathcal{H}_{dd}=\sum_{\mu\neq\eta}\mathcal{H}_{\mu\eta}=\sum_{\mu\neq\eta}G_{\mu\eta}\left[I_\mu^z I_\eta^z-\frac{1}{4}(I_\mu^+ I_\eta^-+I_\mu^- I_\eta^+)\right], \tag{4}$$

where

$$G_{\mu\eta}=\gamma_I^2 r_{\mu\eta}^{-3}(1-3\cos^2\theta_{\mu\eta}), \tag{5}$$

and $r_{\mu\eta}$ and $\theta_{\mu\eta}$ are the spherical coordinates of the vector $\vec{r}_{\mu\eta}$ connecting the μth and ηth nuclei. In the impurity-nuclear DDI Hamiltonian, $\mathcal{H}_{IS}$, we retain only the term which gives the dominate contribution to the relaxation process,[5,13]

$$\mathcal{H}_{IS}\sim\sum_{\mu j}S_j^z(f_{\mu j}I_\mu^++f_{\mu j}^* I_\mu^-), \tag{6}$$

where $f_{\mu j}=-\frac{3}{4}\gamma_I\gamma_S r_{\mu j}^{-3}\sin^2\theta_{\mu j}e^{-i\varphi_{\mu j}}$, γ_S is the gyromagnetic ratio of the PI, and $r_{\mu j}$ is the distance between nuclear and PI spins. $\mathcal{H}_S$ describes the impurity spin system.

Introducing a nuclear-spin density operator

$$\vec{I}(\vec{r})=\sum_\mu \delta(\vec{r}-\vec{r}_\mu)\vec{I}_\mu, \tag{7}$$

the density of the Zeeman and dipole-dipole Hamiltonians can be written in the following form:

$$\mathcal{H}_Z(\vec{r})=\sum_\mu \delta(\vec{r}-\vec{r}_\mu)\omega_0^\mu I_\mu^z=\omega_0(\vec{r})I^z(\vec{r}), \tag{8}$$

$$\mathcal{H}_{dd}(\vec{r})=\int d\vec{r}'G(\vec{r}-\vec{r}')\left\{I^z(\vec{r})I^z(\vec{r}')-\frac{1}{4}[I^+(\vec{r})I^-(\vec{r}')\right.$$
$$\left.+I^-(\vec{r})I^+(\vec{r}')]\right\}, \tag{9}$$

$$\mathcal{H}_{IS}(\vec{r})=\int d\vec{r}'S^z(\vec{r}')[f(\vec{r}-\vec{r}')I^+(\vec{r})+f^*(\vec{r}-\vec{r}')I^-(\vec{r})]. \tag{10}$$

To obtain the equation describing the spin diffusion and spin-lattice relaxation of both the Zeeman and dipolar orders we use the method of the nonequilibrium state operator,[18] which has been applied to obtain the diffusion equation in cases of the Zeeman order spin diffusion[19] and dipolar order[17] spin diffusion. We assume that the density matrix can be written as

$$\sigma=Z^{-1}\exp\left\{-\int d\vec{r}[\beta_Z(\vec{r},t)\mathcal{H}_Z(\vec{r})+\beta_d(\vec{r},t)\mathcal{H}_{dd}(\vec{r})]\right.$$
$$-\beta_S\mathcal{H}_S+\int_{-\infty}^0 dt\,e^{\epsilon t}\left[\int d\vec{r}\left(\beta_Z(\vec{r},t)\frac{\partial\mathcal{H}_Z(\vec{r})}{\partial t}\right.\right.$$
$$\left.\left.\left.+\beta_d(\vec{r},t)\frac{\partial\mathcal{H}_d(\vec{r})}{\partial t}\right)+\beta_S\frac{\partial\mathcal{H}_S}{\partial t}\right]\right\}, \tag{11}$$

where $Z=\mathrm{Tr}\exp\{\dots\}$ and the transition to the limit $\epsilon\rightarrow+0$ should be made after the calculation of the integral. $\beta_Z(\vec{r},t)$ and $\beta_d(\vec{r},t)$ are the local inverse temperatures of the Zeeman and nuclear dipole reservoirs. In Eq. (11) all operators are taken in the Heisenberg representation to be

$$Q(t)=e^{i\mathcal{H}t}Qe^{-i\mathcal{H}t}$$

with

$$Q=\mathcal{H}_Z(\vec{r}),\quad \mathcal{H}_{dd}(\vec{r}),\quad \mathcal{H}_S \tag{12}$$

and they are time dependent. Taking into account that the heat capacity of PI, despite the fact that the concentration C_P is small, $P_S=-\delta/\delta\beta_S\langle\mathcal{H}_S\rangle$, is large in comparison with nuclear-spin heat capacity, both the Zeeman P_Z and dipolar P_d, $(P_S/P_d\ll P_S/P_Z\sim\gamma_S^2/\gamma_I^2\sim10^6)$, and the spin-lattice relaxation time of the PI, T_{1S}, are very short $(T_{1S}/T_{1d},T_{1S}/T_{1Z}\sim10^{-3})$ (Ref. 5), a case which is justified experimentally, it is reasonable to consider only the relaxation process with constant inverse spin temperature of PI, β_S, equal to that of the lattice: $\beta_S=\beta_L$. Therefore, the PI reservoir is in thermal equilibrium with the lattice and β_S is independent of the position and time.

Using the commutation rules between the components of the spin-density operator (7) $[I_x(\vec{r}),I_y(\vec{r}')]=i\delta(\vec{r}-\vec{r}')I_z(\vec{r})$, we can obtain the following equations in the form of localized laws of conservation of the spin energy densities:

$$\frac{\partial\mathcal{H}_Z(\vec{r})}{\partial t}+\omega_0(\vec{r})\mathrm{div}\,\vec{j}_Z(\vec{r})=K_{ZS}(\vec{r}), \tag{13}$$

$$\frac{\partial\mathcal{H}_d(\vec{r})}{\partial t}+\mathrm{div}\,\vec{j}_d(\vec{r})+\vec{j}_Z(\vec{r})\frac{\partial\omega_0(\vec{r})}{\partial\vec{r}}=K_{dS}(\vec{r}), \tag{14}$$

$$\frac{\partial\mathcal{H}_S}{\partial t}=-\int d\vec{r}\left[\frac{\partial\mathcal{H}_Z(\vec{r})}{\partial t}+\frac{\partial\mathcal{H}_d(\vec{r})}{\partial t}\right]. \tag{15}$$

The last equation is the result of the energy-conservation law. In Eq. (13) $\vec{j}_Z(\vec{r})$ is the operator of the flux of nuclear spin,

$$\vec{j}_Z(\vec{r}) = \frac{i}{4} \int d\vec{r}'(\vec{r}-\vec{r}')G(\vec{r}-\vec{r}')[I_+(\vec{r})I_-(\vec{r}')$$
$$-I_-(\vec{r})I_+(\vec{r}')] \quad (16)$$

and in Eq. (14) $\vec{j}_d(\vec{r})$ is the operator of the flux of nuclear dipolar energy,

$$\vec{j}_d(\vec{r}) = \frac{i}{4} \int d\vec{r}' \int d\vec{r}''(\vec{r}-\vec{r}'')G(\vec{r}-\vec{r}')G(\vec{r}-\vec{r}'')$$
$$\times \{[I_+(\vec{r})I_-(\vec{r}')-I_-(\vec{r})I_+(\vec{r}'),I_z(\vec{r}')]$$
$$+[I_+(\vec{r}')I_-(\vec{r})-I_-(\vec{r}')I_+(\vec{r}),I_z(\vec{r}'')]$$
$$+I_z(\vec{r})[I_+(\vec{r}')I_-(\vec{r}'')-I_-(\vec{r}')I_+(\vec{r}'')]\}, \quad (17)$$

where $[A,B]_+ = AB+BA$ is an anticommutator. $K_{ZS}(\vec{r})$ in Eq. (13) is the change of the nuclear Zeeman energy density due to the interaction with the PI,

$$K_{ZS}(\vec{r}) = -i\omega_0(\vec{r}) \int d\vec{r}' S_z(\vec{r}')[f(\vec{r}-\vec{r}')I_+(\vec{r})$$
$$-f^*(\vec{r}-\vec{r}')I_-(\vec{r})], \quad (18)$$

and $K_{dS}(\vec{r})$ in Eq. (14) is the change of the nuclear dipolar energy density due to the interaction with the PI,

$$K_{dS}(\vec{r}) = -\frac{3i}{2} \int d\vec{r}' \int d\vec{r}'' S_z(\vec{r}'')G(\vec{r}-\vec{r}')\{[f(\vec{r}''-\vec{r})I_+(\vec{r})$$
$$-f^*(\vec{r}''-\vec{r})I_-(\vec{r})]I_z(\vec{r}')+[f(\vec{r}''-\vec{r}')I_+(\vec{r}')$$
$$-f^*(\vec{r}''-\vec{r}')I_-(\vec{r}')]I_z(\vec{r})\}. \quad (19)$$

Note that in the case with a homogenous magnetic field, $\partial\omega_0(\vec{r})/\partial\vec{r}=0$, from the system of Eqs. (13) and (14), we have two separate equations: Equation (13) leads to the localized law of conservation of the Zeeman energy densities[19] and Eq. (14) leads to a conservation law of the dipolar energy.[17] In the high-temperature approximation we can write the density matrix (11) in the following form:[18]

$$\sigma = \left\{1 - \int_0^1 d\lambda[\mathcal{B}(t+i\lambda)-\langle\mathcal{B}(t+i\lambda)\rangle]\right\}\sigma_{eq}, \quad (20)$$

where the thermodynamic average $\langle\,\ldots\,\rangle$ corresponds to an average with the quasiequilibrium operator $\sigma_{eq} = e^{-\mathcal{A}}/\mathrm{Tr}\,e^{-\mathcal{A}}$, and

$$\mathcal{A} = \int d\vec{r}[\beta_Z(\vec{r},t)\mathcal{H}_Z(\vec{r})+\beta_d(\vec{r},t)\mathcal{H}_{dd}(\vec{r})]+\beta_S\mathcal{H}_S, \quad (21)$$

$$\mathcal{B}(t+i\lambda) = e^{-\lambda\mathcal{A}} \int_{-\infty}^0 dt\,e^{\varepsilon t} \int d\vec{r}\left\{\omega_0(\vec{r})\vec{j}_Z(\vec{r},t)\nabla\beta_Z(\vec{r},t)\right.$$
$$+\vec{j}_Z(\vec{r},t)[\beta_Z(\vec{r},t)-\beta_d(\vec{r},t)]\frac{\partial\omega_0(\vec{r})}{\partial\vec{r}}$$
$$+\vec{j}_d(\vec{r},t)\nabla\beta_d(\vec{r},t)+[\beta_Z(\vec{r},t)-\beta_L]K_{ZS}(\vec{r},t)$$
$$\left.+[\beta_d(\vec{r},t)-\beta_L]K_{dS}(\vec{r},t)\right\}e^{\lambda\mathcal{A}}. \quad (22)$$

By using Eqs. (11) and (13)–(15), and taking into account that for a single-crystal sample of cubic symmetry, the diffusion coefficients, both for Zeeman and dipole-dipole reservoirs, which in the general case of noncubic symmetry is a symmetrical tensor of second rank,[5] reduce to a scalar quantity. In this case, the diffusion equations can be obtained to be

$$\frac{\partial\beta_Z(\vec{r},t)}{\partial t} = \frac{1}{\omega_0(\vec{r})}\nabla(D_Z(\vec{r})\{\omega_0(\vec{r})\nabla\beta_Z(\vec{r},t)+[\beta_Z(\vec{r},t)$$
$$-\beta_d(\vec{r},t)]\nabla\omega_0(\vec{r})\})-W_Z(\vec{r})[\beta_Z(\vec{r},t)-\beta_L], \quad (23)$$

$$\frac{\partial\beta_d(\vec{r},t)}{\partial t} = \frac{D_Z(\vec{r})\nabla\omega_0(\vec{r})}{M_2}\{\omega_0(\vec{r})\nabla\beta_Z(\vec{r},t)+[\beta_Z(\vec{r},t)$$
$$-\beta_d(\vec{r},t)]\nabla\omega_0(\vec{r})\}+\nabla[D_d(\vec{r})\nabla\beta_d(\vec{r},t)]$$
$$-W_d(\vec{r})[\beta_d(\vec{r},t)-\beta_L], \quad (24)$$

where $M_2 = \int d\vec{r}'G^2(\vec{r}-\vec{r}')\langle I_Z^2\rangle$. The boundary conditions can be introduced by defining a sphere with radius l about each PI, called the spin-diffusion barrier radius. Inside this sphere the spin-diffusion process of Zeeman inverse temperature, $\beta_Z(\vec{r},t)$ and dipolar inverse temperature, $\beta_d(\vec{r},t)$, goes to zero:

$$\nabla\beta_Z(\vec{r},t)|_{|\vec{r}|=l}=0, \quad \nabla\beta_d(\vec{r},t)|_{|\vec{r}|=l}=0. \quad (25)$$

The diffusion barrier radius[1,2,20,21] can be found by solving the equation

$$\frac{3\gamma_S}{l^3}\langle S_z\rangle\left[1\pm\left(1-\frac{|\vec{r}_0|}{l}\right)\right]+\vec{r}\frac{\partial H_0(\vec{r})}{\partial\vec{r}}\bigg|_{|\vec{r}|=l}=\frac{6\gamma_I}{|\vec{r}_0|^3}, \quad (26)$$

where r_0 is distance between neighboring nuclei. Here it is worthwhile to mention that in a homogenous magnetic field, $\partial H_0(\vec{r})/\partial\vec{r}=0$, Eq. (26) leads to result obtained early.[1,2,20,21]

The first term in the curly brackets of the right side of Eq. (23) describes the time dependence of the inverse Zeeman spin temperature $\beta_Z(\vec{r},t)$ as a result of the spin diffusion with a diffusion coefficient of

$$D_Z(\vec{r}) = \frac{1}{2}\int d\vec{r}'(\vec{r}-\vec{r}')^2G^2(\vec{r}-\vec{r}'). \quad (27)$$

The second term gives the variation of $\beta_Z(\vec{r},t)$ as a result of interaction with the dipolar reservoir in the inhomogeneous field. The last term in the right side of Eq. (23) gives the relaxation of $\beta_Z(\vec{r},t)$ toward the inverse lattice temperature with density of the transition probability per unit time, $W_Z(\vec{r})$, which for a cubic crystal is given by

$$W_Z(\vec{r})=4\int d\vec{r}'|f(\vec{r}-\vec{r}')|^2\langle S_z(\vec{r}')S_z(\vec{r}')\rangle. \quad (28)$$

The first term in the curly brackets of the right side of Eq. (24) describes the time variation of the dipolar energy due to the spin diffusion with the diffusion coefficient,

$$D_d(\vec{r})=\frac{\mathrm{Tr}(I_Z^2)}{3M_2}\int d\vec{r}'\int d\vec{r}''(\vec{r}'-\vec{r}'')G(\vec{r}-\vec{r}')G^2(\vec{r}-\vec{r}'')$$
$$\times\left[(\vec{r}-\vec{r}')G(\vec{r}'-\vec{r}'')+\frac{5}{4}(\vec{r}'-\vec{r}'')G(\vec{r}-\vec{r}')\right]. \quad (29)$$

The second term gives the variation of $\beta_d(\vec{r},t)$ as a result of the interaction with the Zeeman reservoir in an inhomogeneous field. The last term in the right side of Eq. (24) gives the relaxation with the density of the transition probability per unit time, $W_d(\vec{r})$, which for a cubic crystal, is given by

$$W_d(\vec{r})=3\int d\vec{r}'|f(\vec{r}-\vec{r}')|^2\left\{1+\frac{\mathrm{Tr}(I_Z^2)}{M_2}\int d\vec{r}'''|f(\vec{r}'-\vec{r}'')|^2\right\}$$
$$\times\langle S_z(\vec{r}')S_z(\vec{r}')\rangle. \quad (30)$$

In Eq. (30) the first term describes direct interaction of a given nuclear spin with the PI and the second term corresponds to indirect interaction via neighboring nuclear spins. It should be noted that the second term in Eq. (30) has no diffusional character.

By introduction the quantities $[\beta_Z(\vec{r},t)-\beta_L]=\xi(\vec{r},t)$ and $[\beta_d(\vec{r},t)-\beta_L]=\zeta(\vec{r},t)$ the equations can be rewritten as

$$\frac{\partial\xi(\vec{r},t)}{\partial t}=\frac{1}{\omega_0(\vec{r})}\nabla(D_Z(\vec{r})\{\omega_0(\vec{r})\nabla\xi(\vec{r},t)+[\xi(\vec{r},t)$$
$$-\zeta(\vec{r},t)]\nabla\omega_0(\vec{r})\})-W_Z(\vec{r})\xi(\vec{r},t), \quad (31)$$

$$\frac{\partial\zeta(\vec{r},t)}{\partial t}=\frac{D_Z(\vec{r})\nabla\omega_0(\vec{r})}{M_2}\{\omega_0(\vec{r})\nabla\xi(\vec{r},t)+[\xi(\vec{r},t)$$
$$-\zeta(\vec{r},t)]\nabla\omega_0(\vec{r})\}+\nabla[D_d(\vec{r})\nabla\zeta(\vec{r},t)]$$
$$-W_d(\vec{r})\zeta(\vec{r},t) \quad (32)$$

with the boundary conditions

$$\nabla\xi|_{|\vec{r}|=l}=0, \quad \nabla\zeta|_{|\vec{r}|=l}=0. \quad (33)$$

In the case of a homogeneous magnetic field, $\nabla\omega_0(\vec{r})=0$, Eqs. (23) and (24) give the results obtain earlier for the spin diffusion of the Zeeman[5] and of the dipolar energy.[17] From Eqs. (31) and (32) we get that the dissipation of the density of the Zeeman and dipolar energies are driven by (i) the exchange between them, the (ii) spin-diffusion process, and (iii) direct relaxation to the PI.

B. Direct relaxation regime

Exact solutions of Eqs. (31) and (32) are extremely difficult problems even for simple model situations. That is why we consider evolution of the spin system in time by using the next considerations. Immediately after a disturbance of the nuclear-spin system, the gradients of $\xi(\vec{r},t)$ and $\zeta(\vec{r},t)$ are sufficiently small and diffusion cannot be of importance at the start of the relaxation process;[20] this is the so-called diffusion vanishing regime.[22] To describe the relaxation at that time interval we can use Eqs. (31) and (32) putting all inverse temperature gradient terms equal to zero, $\nabla\xi(\vec{r},t)=0$ and $\nabla\zeta(\vec{r},t)=0$. We also accept the approximation that at distances larger than the radius of the diffusion barrier the diffusion coefficient is independent of $\vec{r}^4$. Under these approximations Eqs. (31) and (32) arrive at

$$\frac{\partial\xi(\vec{r},t)}{\partial t}=\frac{D_Z\Delta\omega_0(\vec{r})}{\omega_0(\vec{r})}[\xi(\vec{r},t)-\zeta(\vec{r},t)]-W_Z(\vec{r})\xi(\vec{r},t), \quad (34)$$

$$\frac{\partial\zeta(\vec{r},t)}{\partial t}=\frac{D_Z[\nabla\omega_0(\vec{r})]^2}{M_2}[\xi(\vec{r},t)-\zeta(\vec{r},t)]-W_d(\vec{r})\zeta(\vec{r},t). \quad (35)$$

The coupled Eqs. (34) and (35) describe the relaxation processes of the Zeeman and dipolar reservoirs as a result of the exchange between them [first terms in Eqs. (34) and (35)] and direct relaxation to the PI [last terms in Eqs. (34) and (35)]. Equations (34) and (35) are similar to the Provotorov equitations.[13] In a homogeneous magnetic field, Eqs. (34) and (35) give the results obtained earlier.[4,17] The evolutions of $\xi(\vec{r},t)$ and $\zeta(\vec{r},t)$ toward their steady-state values is a linear combination of two exponents.[11] At very low temperatures, where the rates of the direct relaxation $W_Z(\vec{r})$ and $W_d(\vec{r})$ as a result of the PI are very slow, the relaxation rates as a result of the PI are

$$\tau_+^{-1}(\vec{r})=D_Z\left\{\frac{\Delta\omega_0(\vec{r})}{\omega_0(\vec{r})}-\frac{[\nabla\omega_0(\vec{r})]^2}{M_2}\right\}$$
$$-\frac{\Delta\omega_0(\vec{r})}{\Delta\omega_0(\vec{r})-\dfrac{[\nabla\omega_0(\vec{r})]^2\omega_0(\vec{r})}{M_2}}W_Z(\vec{r})$$
$$-\frac{[\nabla\omega_0(\vec{r})]^2}{\dfrac{M_2\Delta\omega_0(\vec{r})}{\omega_0}-[\nabla\omega_0(\vec{r})]^2}W_d(\vec{r}), \quad (36)$$

$$\tau_-^{-1}(\vec{r}) = -\frac{[\nabla\omega_0(\vec{r})]^2}{\dfrac{M_2\Delta\omega_0(\vec{r})}{\omega_0}-[\nabla\omega_0(\vec{r})]^2}W_Z(\vec{r})$$

$$-\frac{\Delta\omega_0(\vec{r})}{\Delta\omega_0(\vec{r})-\dfrac{[\nabla\omega_0(\vec{r})]^2\omega_0(\vec{r})}{M_2}}W_d(\vec{r}). \quad (37)$$

These relaxation times $\tau_+(\vec{r})$ and $\tau_-(\vec{r})$ are the functions of the position $\vec{r}$. In order to obtain the observed magnetization, the solutions of Eq. (34) and (35) must be averaged over the sample. For this averaging procedure one needs to know the field distribution. We consider below an example of the internal distribution of the field in the case of a conventional superconductor.

As a result of the diffusion vanishing relaxation regime the local inverse temperatures, $\xi(\vec{r},t)$ and $\zeta(\vec{r},t)$, become spatially distributed over the sample with a distribution which is not the equilibrium one. In this case we have to take into account also the gradient terms, $\nabla\xi(\vec{r},t)$ and $\nabla\zeta(\vec{r},t)$, in Eqs. (31) and (32). In the next section we consider the influence of the spin-diffusion process.

C. Diffusion relaxation regime

Here we obtain the time dependence of experimentally observable values—the nuclear magnetization and the dipolar energy. We consider a spin system with a sufficiently low concentration C_P of the PI.

For this the sample may be divided into regions, each including only one PI. These regions are assumed to be spherically centered on the PI with radius R equal to the average separation of the PI's (Ref. 22) $R=(3/4\pi C_P)^{-1/3}$.

In order to compare the theoretical results with experimentally observed quantities of the nuclear magnetization and the dipolar energy we use Eqs. (31) and (32) for inverse temperatures, the equations describing time behavior of the local nuclear magnetization and local dipolar energy. The first equation is multiplied by $\gamma_I\omega_0(\vec{r})\mathrm{Tr}(I_Z^2)$ and the second by $M_2\mathrm{Tr}(I_Z^2)$ which leads to the following equations:

$$\frac{\partial m_Z(\vec{r},t)}{\partial t}=D_Z\nabla\left[\nabla m_Z(\vec{r},t)-\frac{\gamma_I}{M_2}\varepsilon_d(\vec{r},t)\nabla\omega_0(\vec{r})\right]$$

$$-W_Z(\vec{r})[m_Z(\vec{r},t)-m_L], \quad (38)$$

$$\frac{\partial\varepsilon_d(\vec{r},t)}{\partial t}=\frac{\gamma_I D_Z\nabla\omega_0(\vec{r})}{M_2}\left[\nabla m_Z(\vec{r},t)-\frac{\gamma_I}{M_2}\varepsilon_d(\vec{r},t)\nabla\omega_0(\vec{r})\right]$$

$$+D_d\Delta\varepsilon_d(\vec{r},t)-W_d(\vec{r})[\varepsilon_d(\vec{r},t)-\varepsilon_L]. \quad (39)$$

By introducing the quantities $[m_Z(\vec{r},t)-m_L]=m(\vec{r},t)$ and $[\varepsilon_d(\vec{r},t)-\varepsilon_L]=\varepsilon(\vec{r},t)$ Eqs. (38) and (39) can be rewritten as

$$\frac{\partial m(\vec{r},t)}{\partial t}=D_Z\nabla\left[\nabla m(\vec{r},t)-\frac{\gamma_I}{M_2}\varepsilon(\vec{r},t)\nabla\omega_0(\vec{r})\right]$$

$$-W_Z(\vec{r})m(\vec{r},t), \quad (40)$$

$$\frac{\partial\varepsilon(\vec{r},t)}{\partial t}=\frac{\gamma_I D_Z\nabla\omega_0(\vec{r})}{M_2}\left[\nabla m(\vec{r},t)-\frac{\gamma_I}{M_2}\varepsilon(\vec{r},t)\nabla\omega_0(\vec{r})\right]$$

$$+D_d\Delta\varepsilon(\vec{r},t)-W_d(\vec{r})\varepsilon(\vec{r},t). \quad (41)$$

Without the terms describing the spin-lattice relaxation, Eqs. (40) and (41) are similar to the two coupled equations obtained earlier using an intuitive approach.[7] Equations (40) and (41) describe the time behavior of the local nuclear magnetization $m(\vec{r},t)$ and the local dipolar energy $\varepsilon(\vec{r},t)$. However, they cannot be observed directly.

The experimentally observed values, the total nuclear magnetization $M(t)=\int d\vec{r}m(\vec{r},t)$ and total dipolar energy $E_d(t)=\int d\vec{r}\varepsilon(\vec{r},t)$, can be obtain from Eqs. (40) and (41) by integrating in limits from l to R both equations. Taking into account the boundary conditions and neglecting surface effects at $r=l$ and R, we have

$$\frac{\partial M(t)}{\partial t}=-\frac{\gamma_I D_Z}{M_2}\int_l^R d\vec{r}\varepsilon(\vec{r},t)\Delta\omega_0(\vec{r})-\int_l^R d\vec{r}W_Z(\vec{r})m(\vec{r},t), \quad (42)$$

$$\frac{\partial E_d(t)}{\partial t}=-\frac{\gamma_I D_Z}{M_2}\int_l^R d\vec{r}m(\vec{r},t)\Delta\omega_0(\vec{r})$$

$$-\int_l^R d\vec{r}\tau_{1d}^{-1}(\vec{r})\varepsilon(\vec{r},t). \quad (43)$$

It follows from Eqs. (42) and (43) that for a short time after the excitation of the spin system, the relaxation of the total nuclear magnetization and the dissipation of the total dipolar energy are governed by the direct relaxation to the PI and by the flow in a inhomogeneous magnetic field. To obtain the relaxation times, both in case of the magnetization and in the case of the dipolar energy, we need to know the internal distribution function of the field, $\omega_0(\vec{r})$, in a sample. Below we consider an example of the internal distribution of the field in the case of the conventional superconductor vanadium compounds.[23]

III. SPIN-LATTICE RELAXATION AND SPIN DIFFUSION IN THE MIXED STATE OF CONVENTIONAL TYPE-II SUPERCONDUCTORS

As an example of an application of the theory, we consider type-II superconductors in a magnetic field. In the type-II superconductors, an applied magnetic field $\vec{H}_0$, in the range between the lower and upper critical field, $H_{c1}<H_0<H_{c2}$, penetrates into the bulk sample in the form of filaments (vortices), each with a quantum flux of $\Phi_0=c\hbar/2e$, which form a two-dimensional structure in the plane perpendicular to $\vec{H}_0$.[24] An internal magnetic field $\vec{H}(\vec{r})$ is generated by vortices, which are spatially distributed over the sample[25]

with the vortex lattice constant $a \approx (\Phi_0/H_0)^{1/2}$. The thermal motion of the vortices in this mixed state gives rise to a time-varying magnetic field $\vec{H}(\vec{r},t)$. The component of this time-varying field perpendicular to the applied magnetic field $\vec{H}_0$ may induce relaxation of the Zeeman[23,26–28] and dipolar energies of the nuclear-spin system. Because of the spatial dependence of the transition probability, the local nuclear magnetization $m(\vec{r},t)$ and the local dipolar energy reach their equilibrium value at a faster rate in the vicinity of the core of vortex. Therefore, the nuclear magnetization and the dipolar energy are functions of the distance, and their spatial diffusion may be induced. This process is important only when the difference of the internal local magnetic field (in units of a frequency) at sites occupied by neighboring nuclei is of the order (or lower than) of the NMR linewidth in order to fulfill the energy-conservation law.[7] We consider a spin system in a sufficiently low external magnetic field, i.e., a concentration $C_v \approx B_0/\Phi_0$ of small vortices, and the sample may be divided into regions with radius R_v equal to the average separation of the vortex core $R_v = (\pi C_v)^{-1/2} \sim a$. For $H_0 \sim 2000$ G, the order is $R_v \sim 500$ Å. Each region includes only one vortex.

In type-II conventional superconductor the distribution of the magnetic field with[29]

$$H(\rho) = \frac{\phi_0}{2\pi\lambda^2} \ln \frac{\rho}{\lambda} \quad \text{for } r < \lambda, \qquad (44)$$

where λ is the London penetration length and ρ is the distance from the core of the vortex in the cylindrical coordinate, $r^2 = \rho^2 + z^2$. In the ideal situation the vortex fields do not have the transversal components, which can cause relaxation. In this case the thermal motion of the vortices in this mixed state is not the reason for the rapid relaxation in type-II conventional superconductors. Another relaxation mechanism must be proposed, such as spin-lattice relaxation as a result of the PI or if there are pinning centers, a result of the thermal fluctuations of the vortex section between the pinning centers.[26] We consider only the relaxation produced by the PI. The thermal fluctuations of vortices will be described in a future paper.

At the first step of the relaxation, immediately after excitation, $\xi(\vec{r},t)$ and $\zeta(\vec{r},t)$ will relax according to Eqs. (34) and (35). From Eqs. (31) and (32) for inverse temperatures we take into account that

$$\nabla\omega_0(\rho) = -\frac{\gamma_I\phi_0}{2\pi\lambda^2\rho}, \quad \Delta\omega_0(\rho) = 0. \qquad (45)$$

We then obtain the time behavior of the local nuclear magnetization and dipolar energy to be

$$\frac{\partial m(\vec{r},t)}{\partial t} = -W_Z(\vec{r})m(\vec{r},t), \qquad (46)$$

$$\frac{\partial\varepsilon(\vec{r},t)}{\partial t} = \frac{D_Z}{M_2}\left(\frac{\gamma_I\phi_0}{2\pi\lambda^2\rho}\right)^2 [m(\vec{r},t) - \varepsilon(\vec{r},t)] - W_d(\vec{r})\varepsilon(\vec{r},t). \qquad (47)$$

The solutions for Eqs. (46) and (47) are

$$\frac{m(\vec{r},t)}{m(r,0)} = \exp\left[-\frac{t}{\tau_{1z}(\vec{r})}\right], \qquad (48)$$

$$\frac{\varepsilon(\vec{r},t)}{\varepsilon(r,0)} = \frac{m(r,0)}{\varepsilon(r,0)}\frac{1}{1 + \frac{\tau_{1z}^{-1}(\vec{r})}{\tau_{1d}^{-1}(\vec{r})}}\left\{\exp\left[-\frac{t}{\tau_{1z}(\vec{r})}\right] - \exp\left[-\frac{t}{\tau_{1d}(\vec{r})}\right]\right\} + \exp\left[-\frac{t}{\tau_{1d}(\vec{r})}\right], \quad (49)$$

where $m(0)$ and $\varepsilon(0)$ are the initial values of $m(\vec{r},t)$ and $\varepsilon(\vec{r},t)$, and $\tau_{1z}^{-1}(\vec{r})$ and $\tau_{1d}^{-1}(\vec{r})$ are the relaxation rates of the local nuclear magnetization and local dipolar energy, respectively,

$$\tau_{1z}^{-1}(\vec{r}) = \tau_{-}^{-1}(\vec{r}) = W_Z(\vec{r}), \qquad (50)$$

$$\tau_{1d}^{-1}(\vec{r}) = \tau_{+}^{-1}(\vec{r}) = D_Z\left(\frac{\gamma_I\phi_0}{2\pi\lambda^2\rho M_2}\right)^2 + W_d(\vec{r}). \qquad (51)$$

At very low temperatures, where the rates of the direct relaxation $W_d(\vec{r})$ to the PI are very slow, the second term can be ignored relative to the first one:

$$\tau_{1d}^{-1}(\vec{r}) = D_Z\left(\frac{\gamma_I\phi_0}{2\pi\lambda^2\rho M_2}\right)^2. \qquad (52)$$

Moreover, the experiments of the measurement of dipolar energy relaxation starting with converting the Zeeman order to a dipolar one by adiabatic demagnetization in the rotating frame[7] result in initial conditions at which $m(r,0) = 0$. Then the time variation of the local dipolar energy can be described by the following equation:

$$\frac{\varepsilon(\vec{r},t)}{\varepsilon(r,0)} = \exp\left[-D_Z\left(\frac{\gamma_I\phi_0}{2\pi\lambda^2\rho M_2}\right)^2 t\right]. \qquad (53)$$

In order to obtain the observed value of the total magnetization $M(t)$ and the total dipolar energy $E_d(t)$, the solution of Eqs. (48) and (53) must be averaged over the sample. To obtain the time dependence of $M(t)$ and $E_d(t)$, the quantities $G_Z(t) = m(\vec{r},t)/m(r,0)$ and $G_d(t) = \varepsilon(\vec{r},t)/\varepsilon(r,0)$ are the normalized relaxation functions of the total magnetization, and the total dipolar energy must be averaged. Taking into account the spherical symmetry of the problem, let us neglect the detailed angular dependence of the transition probability $W_Z(\vec{r})$, whose dependence on the distance from the PI can be presented as[4] $W_Z(\vec{r}) = A/r^6$. In the limit of the number of PI's and of vortices, $N_P \to \infty$, a volume of the

sample, $V_L \to \infty$, and $N_P/V_L = C_P$, the PI concentration, and as a result of the averaging procedure of Eq. (51), we obtain[20,30]

$$\langle G_Z(t) \rangle_V = \exp\left[-\left(\frac{t}{T_{1Z}^{dir}} \right)^\alpha \right] \qquad (54)$$

with the relaxation times for the direct relaxation process written as

$$T_{1Z}^{dir} = \frac{9}{16\pi^3 A C_P^2} \qquad (55)$$

and $\alpha = 1/2$. A similar averaging procedure of Eq. (53) can be performed: in the limit $\lim_{N_v \to \infty, A_v \to \infty}(N_v/A_v) = C_v$, and for a short-time interval after an excitation of the nuclear-spin system, we obtain

$$\langle G_d \rangle_V = \exp\left(-\frac{t}{T_{1d}^{dir}} \right), \qquad (56)$$

where

$$T_{1d}^{dir} = \frac{2\pi}{D_Z C_v \ln \frac{R_v}{l}} \left(\frac{\lambda^2 M_2}{\gamma_I \phi_0} \right)^2. \qquad (57)$$

Consequently, we obtain that for a short time after excitation of the spin system, the decay of the total magnetization is described by the stretched exponential (54) with $\alpha = 0.5$, while the dipolar energy decrease is completely exponential (56).

This direct relaxation process of the Zeeman and dipolar reservoirs should be valid for a short time after a disturbance of the nuclear-spin system. Then it is expected that the nuclear magnetization and the dipolar order relaxation processes are determined by spin diffusion, described by Eqs. (42) and (43). Taking into account that $\Delta\omega_0(r) = 0$ (45), the observed values of the total nuclear magnetization $M(t)$ and the dipolar energy $E_d(t)$ are given by

$$\frac{\partial M(t)}{\partial t} = -\int_l^R d\vec{r} \left[\frac{m(\vec{r},t)}{\tau_{1z}(\vec{r})} \right], \qquad (58)$$

$$\frac{\partial E_d(t)}{\partial t} = -\int_l^R d\vec{r} \left[\frac{\varepsilon(\vec{r},t)}{\tau_{1d}(\vec{r})} \right], \qquad (59)$$

with $\tau_{1z}(\vec{r})$ and $\tau_{1d}(\vec{r})$ given by Eqs. (50) and (52), respectively. The solution of Eqs. (58) and (59) can be obtained by using the expansion of $m(\vec{r},t)$ and $\varepsilon(\vec{r},t)$ in the following forms:[17]

$$m(\vec{r},t) = \int d\vec{r}' \sum_n \exp(-k_n^2 D_Z t) \Phi_n(\vec{r}) \Phi_n^*(\vec{r}') \qquad (60)$$

and

$$\varepsilon(\vec{r},t) = \int d\vec{r}' \sum_n \exp(-p_n^2 D_d t) \Psi_n(\vec{r}) \Psi_n^*(\vec{r}'), \qquad (61)$$

where k_n and $\Phi_n(\vec{r})$ are the eigenvalue and eigenfunction of the operator

$$D_Z \Delta - W_Z(\vec{r}) \qquad (62)$$

and p_n and $\Psi_n(\vec{r})$ are the eigenvalue and eigenfunction of the operator

$$D_d \Delta - D_Z \left[\frac{\gamma \nabla \omega_0(\vec{r})}{M_2} \right]^2. \qquad (63)$$

Taking into account the spherical symmetry of Eq. (62) and the circular symmetry of Eq. (63), just the terms with $n = 0$ contribute to the solutions (58) and (59). This approximation is well known in the quantum theory of scattering[31] and gives the following equations:

$$\frac{\partial M(t)}{\partial t} = -\frac{M(t)}{T_{1Z}^{dif}} \qquad (64)$$

with the relaxation time

$$T_{1Z}^{dif} = \frac{\int_l^R d\vec{r} \, m(\vec{r},t)}{\int_l^R d\vec{r} \, \frac{m(\vec{r},t)}{\tau_{1z}(\vec{r})}} \qquad (65)$$

and the total dipolar energy

$$\frac{\partial E_d(t)}{\partial t} = -\frac{E_d(t)}{T_{1d}^{dif}} \qquad (66)$$

with the relaxation time

$$T_{1d}^{dif} = \frac{\int_l^R d\vec{r} \, \varepsilon(\vec{r},t)}{\int_l^R d\vec{r} \, \frac{\varepsilon(\vec{r},t)}{\tau_{1d}(\vec{r})}}. \qquad (67)$$

For the calculation of the relaxation times in this relaxation regime, T_{1Z}^{dif} and T_{1d}^{dif}, we note that the relaxation processes both for total nuclear magnetization $M(t)$ and the total dipolar energy $E_d(t)$ are not sensitive to the detailed distribution of the local quantities $m(\vec{r},t)$ and $\varepsilon(\vec{r},t)$ and reflect just an integral character of the shape of them. While $m(\vec{r})$ and $\varepsilon(\vec{r})$ do not look like $m(\vec{r},t)$ and $\varepsilon(\vec{r},t)$ in detail these differences cannot produce major errors in the calculations of T_{1Z}^{dif} and T_{1d}^{dif}. Using this fact and the spherical symmetry of the magnetization problem and the circular symmetry of the dipolar energy problem we use the approximation[22] that both $m(\vec{r},t)$ and $\varepsilon(\vec{r},t)$ can be substituted by the time-independent solutions $m(\vec{r})$ and $\varepsilon(\vec{r})$ of the stationary diffusion equations

$$D_Z \Delta_r m(r) - W_Z(r) m(r) = 0, \qquad (68)$$

$$D_d \Delta_\rho \varepsilon(\rho) - D_Z \left[\frac{\gamma \nabla \omega_0(\rho)}{M_2} \right]^2 \varepsilon(\rho) = 0, \qquad (69)$$

where Δ_r and Δ_ρ are the Laplacian in the Cartesian and cylindrical coordinate frames, respectively. Substituting the solution of Eq. (68), which was obtained earlier,[4] into Eq. (65) leads to the relaxation time for the total nuclear magnetization,[4]

$$T_{1Z}^{dif} = (8.5 C_p D_Z^{3/4} A^{1/4})^{-1}. \qquad (70)$$

Equation (69) can be solved using two boundary conditions. For the first boundary condition we adopt an assumption that the local dipolar energy at $\rho = R_v$ has the fixed value $\varepsilon(R_v) \neq 0$ whose actual value is unimportant, because it is be canceled in the expression for the relaxation time T_{1d}^{dif} (67). The second one can be found by integrating Eq. (69) from l to R_v and taking into account that $D_d = 0$ for $\rho < l$, which results in $d\varepsilon(\rho)/d\rho|_{\rho=l} = 0$. Taking into account these boundary conditions the solution of Eq. (69) can be presented as

$$\varepsilon(\rho) = \varepsilon(R_v) \frac{\left(\frac{R_v}{l} \right)^\chi}{1 + \left(\frac{R_v}{l} \right)^{2\chi}} \left[\left(\frac{\rho}{l} \right)^\chi + \left(\frac{\rho}{l} \right)^{-\chi} \right], \qquad (71)$$

where $\chi = (D_Z/D_d)^{1/2} (\gamma_I \phi_0 / 2\pi \lambda^2 M_2)$. Performing the integration in Eq. (67) with the obtained solution of Eq. (71) yields the relaxation time T_{1d}^{dif} of the total dipolar energy,

$$T_{1d}^{dif} = \frac{R_v^2}{D_d} \frac{1}{(\chi^2 - 4)} \left[1 + 2 \frac{y^{-2} - y^\chi - y^{-\chi}}{\chi(y^\chi - y^{-\chi})} \right], \qquad (72)$$

where $y = R_v/l$. In the limit $\chi \gg 1$ $(D_Z/D_d \gg 1)$ from Eq. (72) we obtain

$$T_{1d}^{dif} = \frac{4\pi}{D_Z C_v} \left(\frac{\lambda^2 M_2}{\gamma_I \phi_0} \right)^2. \qquad (73)$$

In the other limit $\chi \ll 1$ $(D_Z/D_d \ll 1)$ the relaxation time $T_{1d}^{dif} \to \infty$. In this case the term describing the relaxation to PI, $W_d(r) \sim B/r^6$, must be taken into account, which leads to a result similar to that of Eq. (70): $T_{1d}^{dif} = (8.5 C_p D_d^{3/4} B^{1/4})^{-1}$. In this case the relaxation times for the total magnetization and for the total dipolar energy are at the same order: $T_{1Z}^{dif} \approx T_{1d}^{dif}$.

IV. RESULTS AND DISCUSSION

We compare the results obtained here with the relaxation processes of nuclear magnetization[23] and dipolar energy[7] in mixed-state superconducting vanadium. First, as is clearly seen from Fig. 2 of Ref. 23 and from Fig. 1 of Ref. 7 at long times the nuclear magnetization[23] and dipolar energy[7] decrease to equilibrium exponentially. This is the spin-diffusion regime described by the solutions of Eqs. (64) and (66):

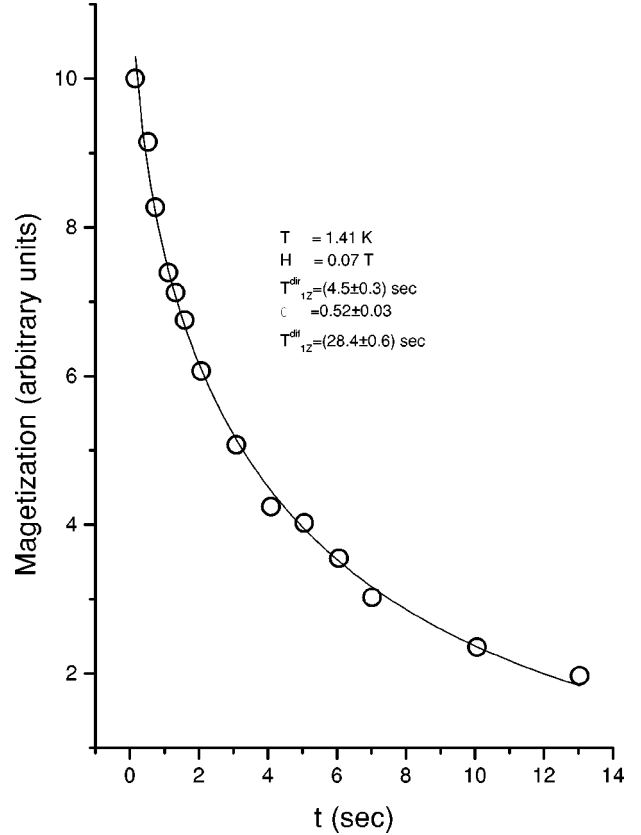$$\frac{M(t)}{M(0)} = \exp\left(-\frac{t}{T_{1Z}^{dif}} \right), \qquad (74)$$

FIG. 1. Relaxation of the total nuclear magnetization in the superconducting state at $T = 1.41$ K. The open circles are experimental data (Ref. 23). The solid line is a least-squares fit to expressions (54) and (74).

$$\frac{E_d(t)}{E_d(0)} = \exp\left(-\frac{t}{T_{1d}^{dif}} \right). \qquad (75)$$

However, such an exponential time dependence for the nuclear magnetization is not valid for shorter times following an excitation of the spin system. The relaxation of the total nuclear magnetization is described by the stretched exponential (54). A least-squares fitting (Figs. 1 and 2) to the sum of a stretched exponential [Eq. (54)] and an exponential (74) yields results very close to $\alpha = 0.5$ for the relaxation of the total nuclear magnetization, in agreement with the conclusion of the above-described theory. Comparing the relaxation times of the direct and the diffusion regimes and assuming $C_P \sim 10^{16}$ cm^{-3}, we conclude that $D_Z \sim 5.3 \times 10^{-13}$ cm^2/sec, which is very closed to the result obtained in Ref. 7.

The relaxation of the total dipolar energy remains exponential (56) also for the diffusion regime, but with the different relaxation time T_{1d}^{dir} (57), which is shorter (note that $R_v/l > 1$) than the T_{1d}^{dif}:

$$T_{1Z}^{dif} = 2 \ln \frac{R_v}{l} T_{1d}^{dir}. \qquad (76)$$

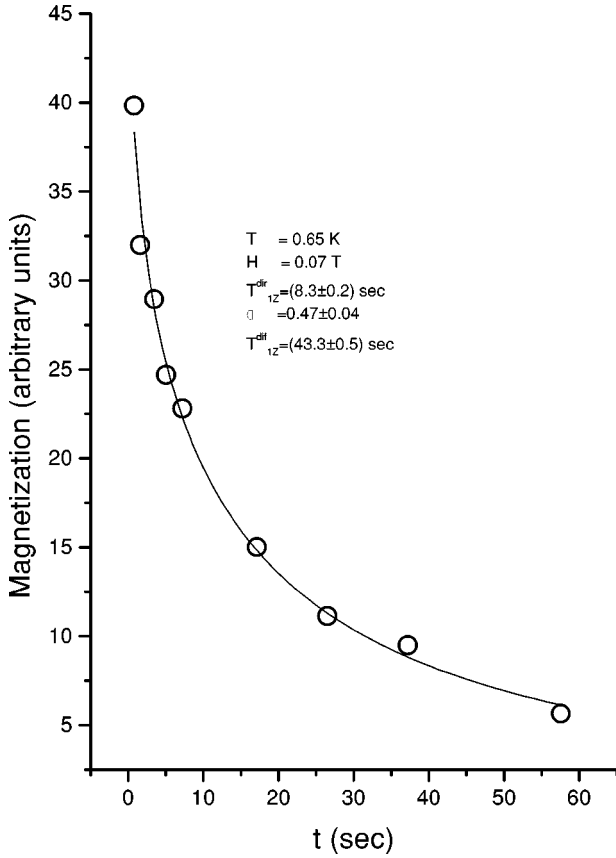A least-squares fitting (Fig. 3) to the sum of two exponents [Eqs. (56) and (75)] yields the result

FIG. 2. Relaxation of the total nuclear magnetization in the superconducting state at $T=0.65$ K. The open circles are experimental data (Ref. 23). The solid line is a least-squares fit to expressions (54) and (74).
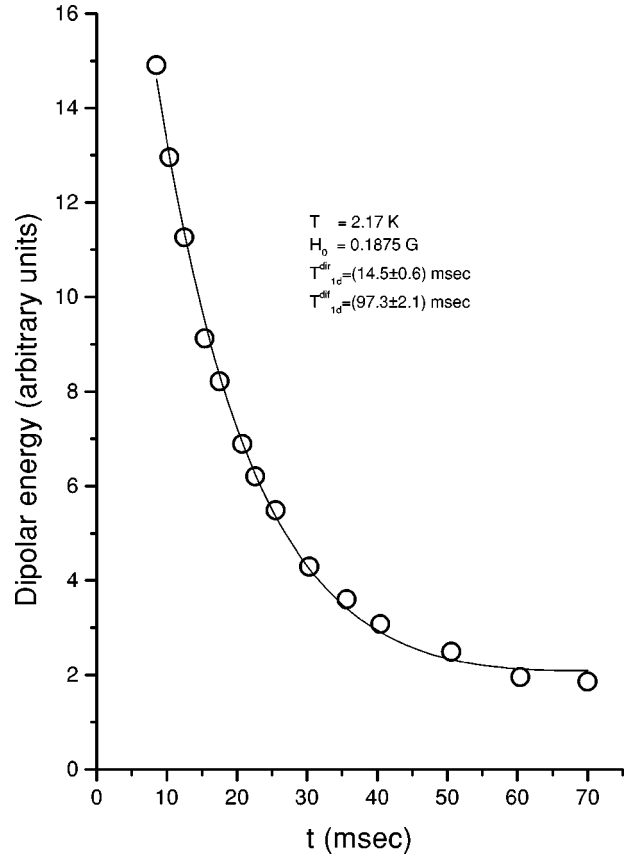

FIG. 3. Relaxation of dipolar energy after adiabatic demagnetization in the superconducting state at $T=2.17$ K. The open circles are experimental data (Ref. 7). The solid line is a least-squares fit expressions (56) and (75).

$$\frac{T_{1Z}^{dif}}{T_{1d}^{dir}} \approx 6.80. \qquad (77)$$

In order to estimate the radius of the diffusion barrier l, we calculate the quantity $R_v = (\pi C_v)^{-1/2}$ by using the value of the magnetic field used in the experiment, 1875 G. As a result we have $R_v \simeq 5.90 \times 10^{-6}$ cm, which is larger than the estimated[32] penetration length for pure vanadium (4.5×10^{-6} cm). Using experimental data (Fig. 3), Eq. (77) and R_v, the radius of the diffusion barrier l, can be estimated to be

$$l = \frac{R_v}{\exp\left(\dfrac{T_{1Z}^{dif}}{2T_{1d}^{dir}}\right)} = 1.96 \times 10^{-7} \quad \text{cm.} \qquad (78)$$

For theoretical estimation of the radius of the diffusion barrier l Eq. (26) can be used. Taking into account that $\vec{r}[\partial H_0(\vec{r})/\partial \vec{r}]|_{|\vec{r}|=l} = -(\phi_0/2\pi\lambda^2)$, the gyromagnetic ratio for vanadium nuclei is $\gamma_I/2\pi = 1119.3$ Hz/G, the distance between neighboring vanadium nuclei $r_0 = 2.63 \times 10^{-8}$ cm,[23] and we assume that for PI it is of the order $\gamma_S/2\pi \sim 3 \times 10^6$ Hz/G.[5] As a result of the calculation

with Eq. (26) for the radius of diffusion barrier l we obtain

$$l = \left[\left(\frac{\gamma_I}{\gamma_S} \right) \frac{1}{r_0^3} + \frac{\phi_0}{12\pi\lambda^2\gamma_S} \right]^{-1/3} = 2.07 \times 10^{-7} \text{ cm,} \quad (79)$$

which is in very good agreement with the result obtained from experimental data (78). Note that in a homogeneous magnetic field, for the radius of the diffusion barrier we have $l = 3.6 \times 10^{-7}$ cm.

As follows from above-presented theory the relaxation mechanisms of the total magnetization and the dipolar energy are different, both for the direct and diffusion relaxation regimes. For a short time after the excitation of the spin system the relaxation of the nuclear magnetization is driven by a direct relaxation to the PI with the relaxation time T_{1Z}^{dir} (55), and the relaxation process is described by the stretched exponential low, while the dissipation of the dipolar energy is determined by its flow into an inhomogeneous magnetic field with a relaxation time T_{1Z}^{dir} (57), and it is completely exponential. Note that the direct relaxation to the PI is not an effective mechanism at a very low temperature. This conclusion is confirmed by experimental data. The relaxation of the nuclear magnetization is nonexponential with the relaxation times $T_{1Z}^{dir} = 4.5$ sec at $T = 1.41$ K and $T_{1Z}^{dir} = 8.3$ sec at T

$=0.65$ K, but the dipolar energy decreases exponentially by the anomalously short (as compared to both the nuclear magnetization time, T_{1Z}^{dir}, and the dipolar energy relaxation time in the normal state, $T_{1d}=120$ msec) relaxation time, $T_{1d}^{dir}=14.5$ msec at $T=2.17$ K. At long times the relaxation of the nuclear magnetization is determined mostly by the spin diffusion since it flows in an homogeneous magnetic field and it is exponential. The inhomogeneity of the local magnetic field does not influence the relaxation of nuclear magnetization due to the specific vortex field distribution in the conventional superconductor which leads to $\Delta\omega_0(\vec{r})=0$. In other type-II superconductors, for example, those of high temperature, with a different vortex field distribution, the influence of the inhomogeneity cannot be ignored. The dipolar energy is determined by the spin diffusion in an inhomogeneous magnetic field. The inhomogeneity of the local magnetic field plays an important role in the relaxation of the dipolar energy. The character of the relaxation remains exponential, but with a different relaxation time compared with that of the direct relaxation regime. Experimental results are in a good agreement with this conclusion. Both the nuclear magnetization and the dipolar energy relax exponentially, but with sufficiently different relaxation times: $T_{1Z}^{dif}=28.4$ sec at $T=1.41$ K and $T_{1Z}^{dif}=43.3$ sec at $T=0.65$ K for the nuclear magnetization and $T_{1d}^{dif}=97.3$ msec for the dipolar energy.

V. CONCLUSIONS

In conclusion, we obtained two coupled equations describing mutual relaxation and spin diffusion of the nuclear magnetization and dipolar energy using the method of the nonequilibrium state operator.[18] The equations were solved at short- and long-time approximations corresponding to the direct and diffusion relaxation regimes. We showed that in the mixed state of the conventional high-T_c superconductor, at the beginning of the relaxation process, the direct relaxation regime is realized. The nuclear magnetization relaxes with a nonexponential time dependence with $\alpha=0.5$ and dipolar energy decreases exponentially. Then the relaxation regime changes both for nuclear magnetization and the dipolar energy, to the diffusion one described by the exponential time dependence, which coincides with the experiment.[23] The radii of the diffusion barrier and the spin-diffusion coefficient were estimated and are in good agreement with experimental data. The obtained analytic expressions can be useful for extracting important information about the vortex dynamics and structure as well as on the internal magnetic-field distribution.

[1] N. Bloembergen, Physica (Amsterdam) **15**, 386 (1949).

[2] G.R. Khutsishvili, Zh. Éksp. Teor. Fiz. **31**, 424 (1956) [Sov. Phys. JETP **4**, 382 (1957)].

[3] P.G. de Gennes, J. Phys. Chem. Solids **3**, 345 (1958).

[4] G.R. Khutsishvili, Usp. Fiz. Nauk **87**, 211 (1965) [Sov. Phys. Usp. **8**, 743 (1966)].

[5] G.R. Khutsishvili, Usp. Fiz. Nauk **96**, 441 (1968) [Sov. Phys. Usp. **11**, 802 (1969)].

[6] L.L. Buishvili, Zh. Éksp. Teor. Fiz. **49**, 1868 (1965) [Sov. Phys. JETP **22**, 1277 (1966)].

[7] A.Z. Genack and A.G. Redfield, Phys. Rev. Lett. **31**, 1204 (1973).

[8] A.G. Redfield and W.N. Yu, Phys. Rev. **169**, 443 (1968); **177**, 1018 (1968).

[9] C. Tang and J.S. Waugh, Phys. Rev. B **45**, 748 (1992).

[10] L.J. Humphries and S.D. Day, Phys. Rev. B **12**, 2601 (1975).

[11] B.N. Provotorov, Zh. Éksp. Teor. Fiz. **41**, 1582 (1961) [Sov. Phys. JETP **14**, 1126 (1962)].

[12] J. Jeneer and P. Broekaert, Phys. Rev. **157**, 232 (1967).

[13] M. Goldman, *Spin Temperature and Nuclear Resonance in Solids* (Clarendon, Oxford, 1970).

[14] A. Abragam and M. Goldman, *Nuclear Magnetism: Order and Disorder* (Clarendon, Oxford, 1982).

[15] C. P. Slichter, *Principles of Magnetic Resonance* (Springer-Verlag, Berlin, 1980).

[16] G.B. Furman, A.M. Panich, A. Yochelis, E.M. Kunoff, and S.D. Goren, Phys. Rev. B **55**, 439 (1997).

[17] G.B. Furman and S.D. Goren, J. Phys.: Condens. Matter **11**, 4045 (1999).

[18] D. N. Zubarev, *Nonequilibrium Statistical Thermodynamics* (Consultants Bureau, New York, 1974).

[19] L.L. Buishvili and D.N. Zubarev, Fiz. Tverd. Tela (Leningrad) **7**, 722 (1965) [Sov. Phys. Solid State **3**, 580 (1965)].

[20] W.E. Blumberg, Phys. Rev. **119**, 79 (1960).

[21] H.E. Rorschach, Jr., Physica (Amsterdam) **30**, 38 (1964).

[22] I.J. Lowe and D. Tse, Phys. Rev. **166**, 279 (1968).

[23] W. Fite, II and A.G. Redfield, Phys. Rev. **162**, 358 (1967).

[24] A.A. Abrikosov, Zh. Éksp. Teor. Fiz. **32**, 1442 (1957) [Sov. Phys. JETP **5**, 1174 (1957)].

[25] D. E. MacLaughlin, in *Solid State Physics*, edited by H. Ehrenreich, F. Seitz, and D. Turnbull (Academic, New York, 1976), Vol. 31, p. 46.

[26] B.G. Silbernagel, M. Weger, and J.E. Wernick, Phys. Rev. Lett. **17**, 384 (1966).

[27] B.G. Silbernagel, M. Weger, W. Clark, and J.E. Wernick, Phys. Rev. **153**, 535 (1967).

[28] I.B. Goldberg and M. Weger, Phys. Rev. Lett. **24**, 1279 (1968).

[29] E. M. Lifshits and L. P. Pitaevskii, *Statistical Physics* (Nauka, Moscow, 1978), Chap. 2.

[30] G.B. Furman, E.M. Kunoff, and S.D. Goren, Phys. Rev. B **52**, 10 182 (1995).

[31] L. D. Landau and E. M. Lifshitz, *Quantum Mechanics Non-Relativistic Theory* (Pergamon, Oxford, 1989).

[32] R. Radebaugh and P.H. Keesom, Phys. Rev. **149**, 217 (1966).