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The spin-diffusion effect in the dipolar ordering state

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Abstract. The theory of spin diffusion is extended to the case of nuclear dipolar order in solids containing paramagnetic impurities. We show that at the beginning of the relaxation process the relaxation function of the dipolar order is non-exponential, due to the direct interaction with paramagnetic impurities, and proceeds to become an exponential function of time, when spin diffusion of the dipolar order takes place. Using experimental data on these two relaxation regimes in CaF_2 doped with Mn^{2+} , the diffusion coefficient and the radius of the diffusion barrier were estimated.

1. Introduction

The dipole–dipole interaction (DDI) plays the dominant role in the spin diffusion and spin–lattice relaxation in solids containing paramagnetic impurities (PIs) [1, 2]. The DDIs between nuclear spins I and PI spins S lead to a direct spin–lattice relaxation of the nuclear spin system. Due to the inverse-sixth-power dependence on the distance between the nuclei and the PI, the local nuclear magnetizations reach their equilibrium values at a faster rate near the PIs [1–3]. Therefore the nuclear magnetization will be a function of the position. This induces the spatial diffusion of the nuclear Zeeman energy by flip-flop transitions due to DDIs between nuclear spins. For the diffusion coefficient, D , values in the interval 10^{-12} – 10^{-13} cm s^{-1} for inorganic solids and 10^{-14} – 10^{-16} cm s^{-1} for organic solids are obtained [4].

The role of the DDI between PI spins in nuclear spin diffusion has been considered in detail [4, 5]. It was shown that the relaxation process is described by the sum of two exponentials. As regards the DDI between nuclear spins, in most theories only the transfer function of the Zeeman energy was considered [1–3]. Only a few papers tried to take into account other functions of the spin–spin energy [6–8]. For example, the theory was extended to include a diffusion of the spin–spin energy in an inhomogeneous magnetic field [6] and an exchange energy in ^3He [7], and the transport of spin–spin energy was calculated numerically for classical gyromagnets, coupled by truncated dipole–dipole and nearest-neighbour exchange interactions [8].

However, as is well known [9–11], the DDI between nuclear spins plays an important part in the spin thermodynamics in solids. On the one hand, the secular parts of the nuclear DDIs form an independent energy reservoir [9–12] with its own spin temperature, which can be different from the spin temperature of the Zeeman reservoir, and, on the other hand, the nuclear DDIs turn on the relaxation processes which bring a nuclear spin system into a thermal equilibrium state [10].

Recently a theory for the spin–lattice relaxation of the nuclear dipolar order via PIs has been developed [13] without taking into account any diffusion processes. Nuclear dipolar

order is characterized by a state with nuclear spins oriented along an internal local field [10] caused by DDIs. Here we discuss the phenomena of spin diffusion of dipolar order of the nuclear spins due to their DDI in solids containing PIs, and the spin-diffusion coefficient of the dipolar order will be obtained from experimental data.

2. Theory

The pure state of the spin system in the form of dipolar order can be reached by using the method of adiabatic demagnetization in a rotating frame (ADRF) [10, 14] or by applying a pair of phase-shifted radio-frequency pulses (the JB method) [15]. The Hamiltonian of the spin system in the frame rotating with the Larmor frequency has the following form:

$$\mathcal{H} = \mathcal{H}_d + \mathcal{H}_{IS} + \mathcal{H}_S \quad (1)$$

where \mathcal{H}_d is secular part of the nuclear DDI Hamiltonian:

$$\mathcal{H}_d = \sum_{\mu \neq \eta} \mathcal{H}_{\mu\eta} = \sum_{\mu \neq \eta} G_{\mu\eta} \left[I_\mu^z I_\eta^z - \frac{1}{4} (I_\mu^+ I_\eta^- + I_\mu^- I_\eta^+) \right] \quad (2)$$

where

$$G_{\mu\eta} = \gamma_I^2 r_{\mu\eta}^{-3} (1 - 3 \cos^2 \theta_{\mu\eta}). \quad (3)$$

γ_I is the gyromagnetic ratio of the nuclei, and $r_{\mu\eta}$ and $\theta_{\mu\eta}$ are the spherical coordinates of the vector $\vec{r}_{\mu\eta}$ connecting the μ th and η th nuclei in a coordinate system with the z -axis along the external magnetic field direction. In the impurity–nuclear DDI Hamiltonian, \mathcal{H}_{IS} , we retain only the term which gives the dominant contribution to the relaxation process:

$$\mathcal{H}_{IS} = \sum_{\mu j} F_{\mu j} I_\mu^z S_j^z \quad (4)$$

where $F_{\mu j} = \gamma_I \gamma_S r_{\mu j}^{-3} (1 - 3 \cos^2 \theta_{\mu j})$, γ_S is the gyromagnetic ratio of the PIs, and $r_{\mu j}$ is the distance between the nuclear and PI spins. Here the Greek indices indicate the position of the nuclei and the italic ones those of the impurities. \mathcal{H}_S describes the impurity spin system.

Introducing a nuclear spin-density operator

$$\vec{I}(\vec{r}) = \sum_{\mu} \delta(\vec{r} - \vec{r}_\mu) \vec{I}_\mu \quad (5)$$

the density of the nuclear dipolar Hamiltonian can be written down in the form

$$\mathcal{H}_d(\vec{r}) = \int d\vec{r}' \sum_{\mu \neq \eta} \delta(\vec{r} - \vec{r}_\mu) \delta(\vec{r}' - \vec{r}_\eta) \mathcal{H}_{\mu\eta}. \quad (6)$$

To obtain the equation describing the spin diffusion and spin–lattice relaxation of the dipolar order we will use the method of non-equilibrium state operators [16], which has been applied to obtain the diffusion equation in the case of the Zeeman-order spin diffusion [17]. Assuming that after the transformation of the Zeeman order to a dipolar one a quasi-equilibrium state is established [10, 14], the density matrix can be written as

$$\rho = Z^{-1} \exp \left\{ - \int d\vec{r} \beta_d(\vec{r}, t) \mathcal{H}_d(\vec{r}) - \beta_S \mathcal{H}_S + \int_{-\infty}^0 dt e^{\varepsilon t} \left[\int d\vec{r} \beta_d(\vec{r}, t) \frac{\partial \mathcal{H}_d(\vec{r})}{\partial t} + \beta_S \frac{\partial \mathcal{H}_S}{\partial t} \right] \right\} \quad (7)$$

where $Z = \text{Tr} \exp\{\dots\}$ and the transition to the limit $\varepsilon \rightarrow +0$ should be made after the calculation of the integral. $\beta_d(\vec{r}, t)$ is the local inverse temperature of the nuclear dipole

reservoir. In equation (7) all operators are taken in the Heisenberg representation. Taking into account that the heat capacity of the PI

$$P_S = -\frac{\delta}{\delta\beta_S} \langle \mathcal{H}_S \rangle$$

is large in comparison with the nuclear spin heat capacity ($P_S/P_I \sim \gamma_S^2/\gamma_I^2 \sim 10^6$) and that the spin–lattice relaxation time of the PI, T_{1S} , is very short ($T_{1S}/T_{1I} \sim 10^{-3}$) [4], a case which is experimentally realizable, it is reasonable to consider only the relaxation process with a constant inverse spin temperature of the 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.

Using the commutation rules relating the components of the spin-density operator (5), we can obtain the following equations in the form of localized laws of conservation of the spin energy densities:

$$\frac{\partial \mathcal{H}_d(\vec{r})}{\partial t} + \text{div } \vec{j}_d(\vec{r}) = K_{dS}(\vec{r}) \quad (8)$$

$$\frac{\partial \mathcal{H}_S}{\partial t} = - \int d\vec{r} K_{dS}(\vec{r}). \quad (9)$$

The last equation is the result of the energy conservation law. In equation (8) $\vec{j}_d(\vec{r})$ is the operator of the flux of the nuclear dipolar energy:

$$\begin{aligned} \vec{j}_d(\vec{r}) = & -\frac{i}{4} \sum_{\mu \neq \eta \neq \nu} \delta(\vec{r} - \vec{r}_\mu) (\vec{r}_\mu - \vec{r}_\nu) G_{\mu\eta} G_{\eta\nu} \left\{ I_\nu^z [I_\mu^+ I_\eta^- - I_\mu^- I_\eta^+] + I_\mu^z [I_\eta^+ I_\nu^- - I_\eta^- I_\nu^+] \right. \\ & \left. + \frac{1}{4} I_\eta^z [I_\nu^- I_\mu^+ - I_\nu^+ I_\mu^- + I_\mu^+ I_\nu^- - I_\mu^- I_\nu^+] \right\} \end{aligned} \quad (10)$$

and $K_{dS}(\vec{r})$ in equation (8) is the change of the nuclear dipolar energy density due to the interaction with the PI:

$$K_{dS}(\vec{r}) = -\frac{i}{4} \sum_{j, \mu \neq \eta} \delta(\vec{r} - \vec{r}_\mu) S_j^z (F_{\mu j} - F_{\eta j}) G_{\mu\eta} (I_\mu^+ I_\eta^- - I_\mu^- I_\eta^+). \quad (11)$$

It is not obvious that $\vec{j}_d(\vec{r}) \neq 0$, because $\partial \vec{j}_d / \partial \vec{r}$ is proportional to the commutator $[\mathcal{H}_d, \mathcal{H}_d(\vec{r})]$ and at first sight it appears that this has to be equal to zero. But in fact only the integral of the commutator over a sample is equal to zero:

$$\int d\vec{r} [\mathcal{H}_d, \mathcal{H}_d(\vec{r})] = 0$$

as it has to be according to the energy conservation law. Due to the fact that the dipolar term is bilinear with respect to the spin operators, the local dipolar energy from a position given by vector \vec{r}_μ can be transferred to the position \vec{r}_ν by using a direct method (the last two terms in the curly brackets in equation (10)) and by means of a spin at position \vec{r}_η (the first two terms in curly brackets in equation (10)). The changes of the local dipolar energy of the μ th nuclear spin at position r_μ via interaction with a PI at the position \vec{r}_j can be made direct (the term $F_{\mu j}$) or indirect via a nuclear spin at position \vec{r}_η (the term $F_{\eta j}$).

In the high-temperature approximation, we can write the density matrix (7) in the following form [16]:

$$\rho = \left\{ 1 - \int_0^1 d\lambda [\mathcal{B}(t + i\lambda) - \langle \mathcal{B}(t + i\lambda) \rangle] \right\} \rho_{eq} \quad (12)$$

where the thermodynamic average $\langle \dots \rangle$ corresponds to an averaging with the quasi-equilibrium operator $\rho_{eq} = e^{-A}/\text{Tr} e^{-A}$, where

$$A = \int d\vec{r} \beta_d(\vec{r}, t) \mathcal{H}_d(\vec{r}) + \beta_L \mathcal{H}_S \quad (13)$$

$$\begin{aligned} B(t + i\lambda) = & \int_{-\infty}^0 dt e^{\epsilon t} \int d\vec{r} \{ \nabla \beta_d(\vec{r}, t) \cdot [e^{-\lambda A} \vec{j}_d(\vec{r}, t) e^{\lambda A}] \\ & + [\beta_d(\vec{r}, t) - \beta_L] e^{-\lambda A} K_{dS}(\vec{r}, t) e^{\lambda A} \}. \end{aligned} \quad (14)$$

By using equations (10) and (12)–(14) and taking into account that for a cubic single-crystal sample, the diffusion coefficient D —which in the general case of non-cubic symmetry is a symmetrical tensor of second rank [4]—reduces to a scalar quantity, the diffusion equation can be obtained:

$$\frac{\partial \beta_d(\vec{r}, t)}{\partial t} = D \Delta \beta_d(\vec{r}, t) - W(\vec{r}) [\beta_d(\vec{r}, t) - \beta_L] \quad (15)$$

with the boundary condition

$$\nabla \beta_d(r)|_{r=l} = 0 \quad (16)$$

where $l \sim (\gamma_S/\gamma_I)^{1/3} r_0$ is the radius of the diffusion barrier [1, 2] inside which the spin-diffusion process is quenched; r_0 is the distance between neighbouring nuclei. The first term on the right-hand side of equation (15) describes the variation (over time) of the dipolar order due to the spin diffusion with the diffusion coefficient

$$\begin{aligned} D = & \left(\frac{3}{4} \right)^2 \int_{-\infty}^{\infty} dt \sum_{\mu \neq \eta \neq \nu} \delta(\vec{r} - \vec{r}_\mu) (\vec{r} - \vec{r}_\nu) (\vec{r}_\mu - \vec{r}_\nu) \langle I_\nu^z I_\nu^z(t) I_\mu^+ I_\mu^-(t) I_\eta^- I_\eta^+(t) \rangle \\ & \times (G_{\mu\eta} G_{\eta\nu})^2 / \langle \mathcal{H}_d \mathcal{H}_d(\vec{r}) \rangle \end{aligned} \quad (17)$$

and the second term gives the variation of $\beta_d(\vec{r})$ due to the relaxation with density of the transition probability per unit time, $W(\vec{r})$, which is given for a cubic crystal by

$$W(\vec{r}) = \sum_{\mu j} \delta(\vec{r} - \vec{r}_\mu) \left[A_{\mu j} + \sum_{\eta} B_{\mu\eta j} \right] \quad (18)$$

where

$$A_{\mu j} = \frac{1}{2\pi} F_{\mu j}^2 \int_{-\infty}^{\infty} dt \sum_{\eta} f_{\mu\eta}^j(t) \quad (19)$$

$$B_{\mu\eta j} = \frac{1}{2\pi} F_{\eta j}^2 \int_{-\infty}^{\infty} dt f_{\mu\eta}^j(t) \quad (20)$$

$$f_{\mu\eta}^j(t) = \frac{1}{4} \delta(\vec{r} - \vec{r}_\mu) \langle I_\mu^+ I_\mu^-(t) I_\eta^- I_\eta^+(t) S_j^z S_j^z(t) \rangle G_{\mu\eta}^2 / \langle \mathcal{H}_d \mathcal{H}_d(\vec{r}) \rangle. \quad (21)$$

In equation (18) the first term, $A_{\mu j}$, describes the direct interaction of a given nuclear spin with the PI and the second, $\sum_{\eta} B_{\mu\eta j}$, corresponds to indirect interaction via neighbouring nuclear spins. It should be noted that the second term in equation (18) has no diffusional character.

3. Results and discussion

Immediately after a disturbance of the nuclear spin system, there is no gradient of $\beta_d(\vec{r}, 0)$ and diffusion cannot be of importance at the start of the relaxation process [18]. To describe

the relaxation we can use equation (15) without the first term; this is the so-called direct-relaxation regime [19]. Since the local inverse temperature, $\beta_d(\vec{r}, t)$, is distributed throughout the sample and depends on position, in order to obtain a quantity which is connected with the experimentally observable component of the magnetization, a suitable averaging procedure must be performed [13]. After averaging, the normalized relaxation function

$$R(t) = \left\langle \frac{\beta_d(\vec{r}, t) - \beta_d(\vec{r}, \infty)}{\beta_d(\vec{r}, 0) - \beta_d(\vec{r}, \infty)} \right\rangle$$

takes the form [13]

$$R(t) = \exp\left[-\left(\frac{t}{T_{1d}^A}\right)^\alpha\right] \exp\left[-\left(\frac{t}{T_{1d}^B}\right)\right] \quad (22)$$

where $T_{1d}^A \sim C_p^{-1/\alpha}$ and T_{1d}^B are the spin-lattice relaxation times due to direct and indirect interaction with the PI; C_p is the impurity concentration. For a homogeneous distribution of paramagnetic centres and nuclear spins, $\alpha = Q/6$ where Q is the sample dimensionality; for an inhomogeneous distribution, the sample is divided into q -dimensional subsystems, each containing one paramagnetic centre, yielding $\alpha = (Q + q)/6$. In the two limiting cases, when the radius of the diffusion barrier, l , is large or small enough relative to size of the subsystems, only one exponent from equation (22) plays an important role [13]. This result coincides with experimental data for CaF_2 doped with 0.8×10^{-3} wt% of Mn^{2+} , where non-exponential decay of the dipolar signal with $\alpha = 0.83$ has been observed [20]. In this case, as a result of the adiabatic demagnetization, the local magnetization is equal to zero and it is reasonable

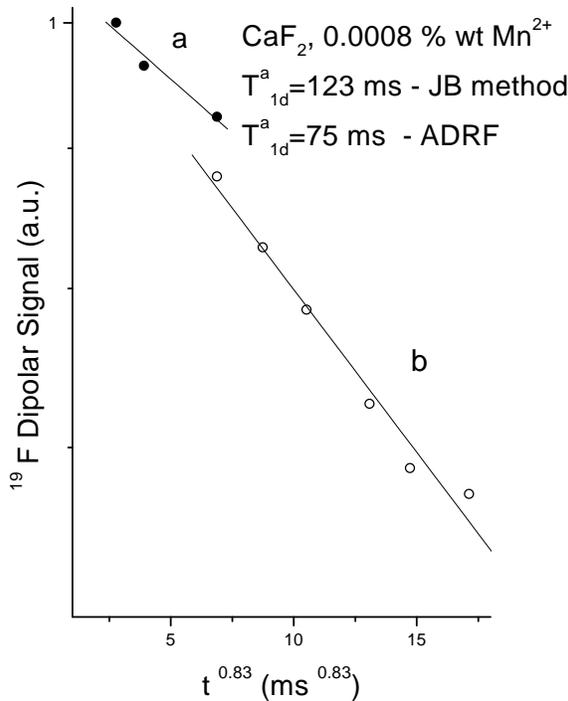


Figure 1. The log of the ^{19}F dipolar signal for CaF_2 doped with Mn^{2+} as a function of $t^{0.83}$ for the diffusion-vanishing regime of relaxation: (a) solid circles: JB method; (b) open circles: ADRF method. The solid lines show apparent fits to expression (23).

to assume that the subsystem dimensionality is equal to 2 with the plane perpendicular to the external magnetic field in accordance with the symmetry of the system. So $q = 2$ and, according to the structure of CaF_2 , $Q = 3$. For an inhomogeneous distribution we have $\alpha = \frac{5}{6} \simeq 0.83$. Fitting the experimental data [20] yields a good agreement with $T_{1d}^A = 123$ ms for the JB method; $T_{1d}^A = 75$ ms for the ADRF at the beginning of the relaxation process (figure 1). This means that only the first exponent in equation (22) plays an important role, and using this fact we can neglect the second term in equation (18), so $W \sim A/r^6$, where $A/r^6 = \langle A_{\mu j} \rangle_{\theta_{\mu j}}$ is an average over the spherical coordinates of the vector $\vec{r}_{\mu j}$. Another independent check of the results obtained is the dependence of the relaxation time on the impurity concentration. In accordance with $1/\alpha = 1.2$, we have $T_{1d}^A \sim C_p^{-1.2}$. Exactly this dependence on the impurity concentration of the relaxation time has been found in the experiment [20]. The difference between the relaxation times T_{1d}^A found after using JB and ADRF preparation methods can be explained by taking into account the fact that the ADRF method is isentropic, whereas the JB method is not [10]. Therefore the dipolar ordering states of the spin system achieved by these two preparation methods have some differences [20].

The direct-relaxation regime of the dipolar order should be valid for a short time, $t \ll A^{1/2}D^{-3/2}$, after a disturbance of the nuclear spin system [18]. Then it is expected that β_d starts as a non-exponential function that is time dependent:

$$R(t) \sim \exp\left[-\left(\frac{t}{T_{1d}^A}\right)^\alpha\right] \quad (23)$$

and proceeds asymptotically to become an exponential function of time, the so-called diffusion-limited relaxation regime [19]. In this case we have to take into account also the first term in equation (15). One of the ways to solve this equation and extract the time dependence is by introducing the eigenfunctions $\varphi_n(\vec{r})$ of the operator $D\Delta - W(\vec{r})$ [21]. Using a spherical symmetry approximation, the general solution of the equation (15) can be written as an expansion in terms of the orthogonal functions $\varphi_n(\vec{r})$:

$$\beta_d(r, t) = \int dr_1 \sum_n \exp(-k_n^2 D t) \varphi_n(r) \tilde{\varphi}_n^*(r_1) \quad (24)$$

where the functions $\varphi_n(r)$ satisfy the equation

$$\Delta \varphi_n(r) - (W(r)/D) \varphi_n(r) = -k_n^2 \varphi_n(r) \quad (25)$$

with the boundary condition

$$\nabla \varphi_n(r)|_{r=l} = 0. \quad (26)$$

Equation (25) is well known in the theory of scattering as a low-energy limit [22], and has an asymptotic solution for $r \gg l$: $\varphi_n(r) \sim \sin(k_n r + \delta_n)/r$, where $\delta_n \sim k_n \xi$ is the phase shift and ξ is the scattering length [22]. According to the spherical symmetry, only s-wave scattering with $n = 0$ is taken into account. Using the boundary condition (26) and the fact that $W \sim A/r^6$ for $(A/D)^{1/4} \ll r \ll 1/k_0$, we obtain [22]

$$\xi = \left(\frac{A}{D}\right)^{1/4} \frac{\Gamma(3/4)}{2\Gamma(5/4)}. \quad (27)$$

Using the last result, for the long-time approximation, $t \gg l^2/D$, we obtain the normalized relaxation function for the diffusion-limited regime:

$$R(t) = \exp(-t/T_{1d}^D) \quad (28)$$

where

$$T_{1d}^D = \frac{1}{4\pi C_p D \xi}.$$

So we obtain an expression that describes the exponential time dependence of $\beta_d(t)$ (figure 2) with the relaxation time

$$T_{1d}^D = \left[2\pi C_p D^{3/4} A^{1/4} \frac{\Gamma(3/4)}{\Gamma(5/4)} \right]^{-1} \quad (29)$$

which is inversely dependent on the impurity concentration and has the same form as for the Zeeman spin-diffusion case [4].

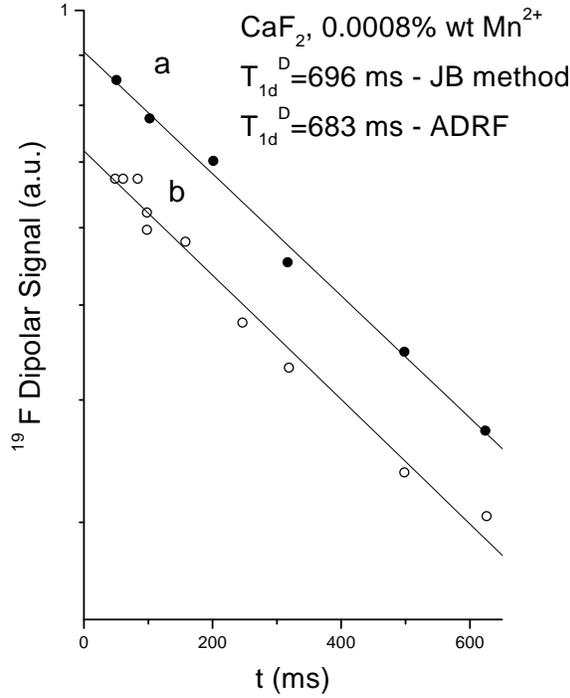


Figure 2. The log of the ^{19}F dipolar signal for CaF_2 doped with Mn^{2+} as a function of t for the diffusion-limited regime of relaxation: (a) solid circles: JB method; (b) open circles: ADRF method. The solid lines show apparent fits to expression (28).

We do not evaluate the relaxation times T_{1d}^A and T_{1d}^D . Instead we will obtain these quantities from experimental data [20] and then we will use them to calculate the diffusion coefficient of the dipolar order, D . Fitting the experimental data [20] yields a good agreement with $T_{1d}^D = 696$ ms for the JB method and $T_{1d}^D = 683$ ms for the ADRF obtained for the second part of the relaxation process. At times longer than 50 ms the direct-relaxation regime is changed to a diffusion-limited one for both JB and ADRF preparation methods. Using this information, the interaction constant, A , and the radius of the dipolar diffusion barrier, l , can be estimated

Table 1. Diffusion coefficients of dipolar order for Mn^{2+} -doped CaF_2 crystal.

Method	T (K)	C_p (cm^{-3}) [20]	T_{1d}^A (ms)	T_{1d}^D (ms)	D (cm s^{-1})
JB	4.2	1.2×10^{19}	123	696	1.8×10^{-13}
ADRF	4.2	1.2×10^{19}	75	683	1.5×10^{-13}

to be $A \sim 10^{-41} \text{ cm}^6 \text{ s}^{-1}$ and $l \sim 8.6 \times 10^{-8} \text{ cm}$. The values for the diffusion coefficient, D , for both preparation methods, are given in table 1.

4. Conclusions

We found that the spin-diffusion process of the dipolar order takes place in solids containing paramagnetic impurities. As mentioned above, at the beginning of the relaxation process the direct-relaxation regime is realized with a non-exponential time dependence and T_{1d}^A is inversely proportional to the $6/(Q+q)$ power of C_p . Then the relaxation regime is changed to a diffusion-limited one in accordance with $T_{1d}^A \ll T_{1d}^D$. The latter type of relaxation behaviour of the dipolar order is seen in the experiments [20]. Using experimental results [20] for these two regimes, we can estimate the diffusion coefficient of the nuclear dipolar order in CaF_2 doped with paramagnetic Mn^{2+} ; we obtain, for typical values of the impurity concentration $C_p \sim 10^{19} \text{ cm}^{-3}$, the diffusion coefficient of the dipolar order as $D \sim 10^{-13} \text{ cm}^2 \text{ s}^{-1}$ and the radius of the diffusion barrier as $l \sim 10^{-7} \text{ cm}$ for both JB and ADRF preparation methods; these are close to the values for the cases of Zeeman-energy spin diffusion for inorganic solids [4,23] and of a spin-spin diffusion in an inhomogeneous magnetic field [6].

Acknowledgments

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