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Spin–Spin Interactions and Nuclear Magnetic Relaxation in Magnetic Solids

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Abstract. The influence of the orientational fluctuations of the electronic magnetization, which modulate nuclear spin–spin interactions (Suhl–Nakamura and dipole–dipole), on the spin-lattice relaxation of magnetic nuclei with spin $I = 1/2$ in the magnetically ordered solids has been investigated. It has been shown that this mechanism of the spin-lattice relaxation is less effective in comparison with the process of spin-lattice relaxation caused by the direct fluctuations of hyperfine fields, which appear when there are the fluctuations of electronic magnetization direction.

Key Words: spin-lattice relaxation, Suhl–Nakamura and dipole–dipole interactions.

At present for analysis of the influence of nuclear spin–spin (Suhl–Nakamura and dipole–dipole [1]) interactions on the nuclear spin-relaxation of magnetic nuclei in the magnetic materials is used, mainly, the model, which can be named as the model of 'rigid' lattice [1]. In this model it is assumed that the direction of the electron magnetization $\vec{M}_e$ does not depend on time. Within the framework of this model nuclear spin–spin interactions give the contribution only to the transverse (spin–spin) relaxation rate (T_2^{-1}) [1]. On the other hand, from NMR experiments, executed on the YIG films with the different concentration of magnetic nuclei ^{57}Fe, it follows that the longitudinal relaxation rate (T_1^{-1}) depends on the concentration of magnetic nuclei [2]. The influence of nuclear dipole–dipole interactions on the longitudinal relaxation rate is well known for the diamagnetic materials and this phenomenon is caused by thermal fluctuation of nuclear interactions [3]. At present it is well established that in the magnetic materials there are thermal fluctuations of the direction of electronic magnetization $\vec{M}_e$ [1, 4–6]. These thermal fluctuations in the direction of $\vec{M}_e$ induce the thermal fluctuations of nuclear spin–spin interactions and this can lead to the fact that the nuclear spin–spin interactions can give the contribution to the longitudinal relaxation rate (T_1^{-1}).

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The purpose of the present note is the consideration of the possibility of this new channel of spin-lattice relaxation of nuclei with spin $I = 1/2$ in magnetically ordered solids.

The interaction Hamiltonian of the two magnetic nuclei with spin $I = 1/2$ in magnetic solids can be written as [1]

$$H_0 = H_{HF} + H_{S-N} + H_{d-d}, \tag{1}$$

where the first term is the hyperfine magnetic interaction (HF) Hamiltonian, H_{S-N} is the Suhl–Nakamura interaction (*SN*) Hamiltonian and H_{d-d} is dipole–dipole interaction (*DD*) Hamiltonian.

For simplicity we shall assume that the *HF* Hamiltonian is isotropic and has the form ($\hbar = 1$)

$$H_{HF} = -\omega_0 I_Z. \tag{2}$$

Here $I_Z = I_{1Z} + I_{2Z}$ and ω_0 is the NMR frequency of nuclei.

Usually $\|H_{HF}\| >> \|H_{S-N}\|, \|H_{d-d}\|$ and the full interaction Hamiltonian can be written as [1, 3]

$$H_0 = -\omega_0 I_Z + V_{12}(I_{1+}I_{2-} + I_{1-}I_{2+}) - D_{12}[4I_{1Z}I_{2Z} - (I_{1+}I_{2-} + I_{1-}I_{2+}). \tag{3}$$

In Equation (3) V_{12} is the constant of the *SN*-interaction between nuclear magnetic moments. The dipolar coupling constant D_{12} is given by

$$D_{12} = \frac{\gamma^2\hbar}{8r_{12}^3}\left(3\cos^2\vartheta_{12} - 1\right). \tag{4}$$

It should be noted that in the interaction Hamiltonian (3) the Z-axis is the quantization axis for the nuclear spins and in magnetically ordered materials this axis coincides with the direction of the electron magnetization $\vec{M}_e$ [1].

Now we assume that there are thermal fluctuations in the electron magnetization vector $\vec{M}_e$. We want to show that the thermal fluctuations in the direction of the quantization axis for the nuclear spins lead to the temporal fluctuations of *SN* and *DD* interaction Hamiltonians.

Consider two coordinate system X, Y, Z and X', Y', Z'. In the 'rigid' coordinate system X, Y, Z the Z-axis coincides with the equilibrium direction of an electron magnetization $\vec{M}_{e0}$. The 'fluctuating' coordinate system X', Y', Z' is obtained by the rotation of the coordinate system X, Y, Z by an angle φ about the Z-axis and by an angle θ about new Y' axis. In this 'fluctuating' coordinate system the Z'-axis coincides with the direction of an electron magnetization $\vec{M}_e(t)$ at the time t. The transformation from the coordinate system X, Y, Z to the coordinate system X', Y', Z' is defined by the rotation operator [7, 8]

$$R(\theta, \varphi) = \exp(-i\varphi I_Z)\exp(-i\theta I_Y)\exp(i\varphi I_Z). \tag{5}$$

Here θ is the angle between Z-axis and Z'-axis.

The interaction Hamiltonian in the 'fluctuating' coordinate system has the form

$$H^{/} = R(\theta, \varphi) H_0 R^{-1}(\theta, \varphi). \tag{6}$$

Using Equation (5) and assuming that fluctuations in the angle θ is small ($\cos\theta \approx 1$, $\sin\theta \approx \theta$) we obtain

$$H^{/}(t) = H_0 + H_1(t). \tag{7}$$

The static part of interaction Hamiltonian (7) coincides with Hamiltonian (3). The time-dependent part in Hamiltonian (7) contains three terms

$$H_1(t) = H_{HF}(t) + H_{S-N}(t) + H_{d-d}(t). \tag{8}$$

Here the term

$$H_{HF}(t) = \frac{\omega_0}{\sqrt{2}} \cdot \theta(t) \cdot \left(e^{-i\varphi(t)} \cdot T_{1,+1} - e^{i\varphi(t)} \cdot T_{1,-1} \right) \tag{9}$$

describes the fluctuations in the hyperfine interaction Hamiltonian. In Equation (9)

$$T_{1,\pm1} = \mp \frac{1}{\sqrt{2}} I_{\pm} = \mp \frac{1}{\sqrt{2}} (I_{1\pm} + I_{2\pm}) \tag{10}$$

are components of irreducible tensor operator of the first rank [7–9].

The term

$$H_{S-N}(t) = 2V_{12} \cdot \theta(t) \cdot \left(e^{-i\varphi(t)} \cdot T_{2,+1} - e^{i\varphi(t)} \cdot T_{2,-1} \right) \tag{11}$$

is the fluctuating part of the Suhl–Nakamura interaction Hamiltonian. In Equation (11) the $T_{2,\pm1}$ are components of irreducible tensor operator of the second rank [7–9]:

$$T_{2,\pm1} = \mp \frac{1}{2} (I_{1Z} I_{2\pm} + I_{1\pm} I_{2Z}). \tag{12}$$

In Equation (8) the term

$$H_{d-d}(t) = 6D_{12} \cdot \theta(t) \cdot \left(e^{-i\varphi(t)} \cdot T_{2,+1} - e^{i\varphi(t)} \cdot T_{2,-1} \right) \tag{13}$$

describes the fluctuations in the dipolar interaction Hamiltonian.

From Equations (11) to (13) we see that these fluctuating Hamiltonians have the same forms. So we will use the new Hamiltonian

$$H_{\text{int}}(t) = A_{12} \cdot \theta(t) \cdot \left(e^{-i\varphi(t)} \cdot T_{2,+1} - e^{i\varphi(t)} \cdot T_{2,-1} \right) \tag{14}$$

where

$$A_{12} = 2 \cdot (V_{12} + 3D_{12}). \tag{15}$$

In order to derive the expression for the spin-lattice relaxation rate we use the well-known expression for the reduced spin density matrix [3, 9]

$$\frac{d\tilde{\rho}}{dt} = -\int_0^\infty d\tau \cdot \overline{\left[\widetilde{H}_1(t), \left[\widetilde{H}_1(t-\tau), \tilde{\rho}\right]\right]}. \tag{16}$$

Here

$$\widetilde{\rho}(t) = e^{i\omega_0 t I_Z} \rho(t) e^{-i\omega_0 t I_Z} \tag{17}$$

and

$$\widetilde{H}_1(t) = e^{i\omega_0 t I_Z} H_1(t) e^{-i\omega_0 t I_Z}. \tag{18}$$

In Equation (16) the upper bar denotes the average on the random fluctuations of the Hamiltonian $H_1(t)$.

Using Equation (18) we obtain from Equations (9) to (14)

$$\widetilde{H}_1(t) = F(t) \cdot T_+ \cdot e^{i\omega_0 t} + F^*(t) \cdot T_- \cdot e^{-i\omega_0 t}, \tag{19}$$

where

$$F(t) = \frac{\omega_0}{\sqrt{2}} \cdot \theta(t) \cdot e^{-i\varphi(t)}, \tag{20}$$

and

$$T_\pm = \pm\left(T_{1,\pm 1} + \alpha \cdot T_{2,\pm 1}\right). \tag{21}$$

In Equation (21)

$$\alpha = \frac{\sqrt{2} \cdot A_{12}}{\omega_0}. \tag{22}$$

Using the definition

$$J(\omega_0) = \int_0^\infty \overline{F(t)F^*(t-\tau)} \cdot e^{i\omega_0 \tau} d\tau \tag{23}$$

and retaining in Equation (16) only time independent terms (secular terms [3, 9]) we obtain

$$\frac{d\rho}{dt} = -J(\omega_0) \cdot [T_+, [T_-, \rho]] - J^*(\omega_0) \cdot [T_-, [T_+, \rho]]. \tag{24}$$

Multiplying both sides of Equation (24) by $I_Z = I_{1Z} + I_{2Z}$ and taking the trace, we obtain the equations of motions for the expectation values of I_Z:

$$\left\langle \frac{dI_Z}{dt} \right\rangle = -J(\omega_0) \cdot \langle [T_+, [T_-, I_Z]] \rangle - J^*(\omega_0) \cdot \langle [T_-, [T_+, I_Z]] \rangle, \tag{25}$$

Here $\langle C \rangle = Tr(C\rho)$.

Using the following commutation relations

$$[T_{\pm}, I_Z] = \mp T_{\pm},$$

we have from Equation (25)

$$\begin{aligned}\left\langle \frac{dI_Z}{dt} \right\rangle &= -2\mathrm{Re}[J(\omega_0)]\cdot\langle [T_+, T_-]\rangle \\ &= -2J(\omega_0)\cdot\left\{\left\langle \left[T_{1,-1}, T_{1,+1}\right]\right\rangle + \alpha\cdot\left(\left\langle \left[T_{1,-1}, T_{2,+1}\right]\right\rangle,\right.\right. \\ &\quad \left.\left. +\left\langle \left[T_{2,-1}, T_{1,+1}\right]\right\rangle\right) + \alpha^2\cdot\left\langle \left[T_{2,-1}, T_{2,1}\right]\right\rangle\right\} \end{aligned} \tag{26}$$

Using the relation [10]

$$\left\langle \left[T_{1,-1}, T_{1,+1}\right]\right\rangle = \langle I_z\rangle,$$

$$\left\langle \left[T_{1,-1}, T_{2,+1}\right]\right\rangle = \left\langle \left[T_{2,-1}, T_{1,+1}\right]\right\rangle = 0,$$

$$\left\langle \left[T_{2,-1}, T_{2,1}\right]\right\rangle = \frac{2}{3} I(I+1)\cdot\langle I_Z\rangle,$$

we obtain

$$\left\langle \frac{dI_Z}{dt} \right\rangle = -2\mathrm{Re}[J(\omega_0)]\cdot\left[1 + \frac{2}{3} I(I+1)\alpha^2\right]\cdot\langle I_Z\rangle\}. \tag{27}$$

From Equation (27) it follows that the spin-lattice relaxation rate is defined by equation

$$T_1^{-1} = \left(T_1^{-1}\right)_{HF} + \left(T_1^{-1}\right)_{\mathrm{int}}, \tag{28}$$

where

$$\left(T_1^{-1}\right)_{HF} = 2\mathrm{Re}[J(\omega_0)] \tag{29}$$

is the spin-lattice relaxation rate defined by the fluctuations in the hyperfine interaction Hamiltonian;

$$\left(T_1^{-1}\right)_{\mathrm{int}} = \mathrm{Re}[J(\omega_0)]\cdot\frac{4}{3} I(I+1)\cdot\alpha^2 \tag{30}$$

is the spin-lattice relaxation rate defined by the fluctuations in the Suhl–Nakamura and dipolar interaction Hamiltonians.

From Equations (29) to (30) we have

$$\frac{\left(T_1^{-1}\right)_{\mathrm{int}}}{\left(T_1^{-1}\right)_{HF}} = \frac{1}{\omega_0^2}\left[\frac{4}{3} I(I+1)A_{12}^2\right] = \frac{4M_2}{\omega_0^2}, \tag{31}$$

where

$$M_2 = \frac{4}{3} I(I+1) \cdot (V_{12} + 3D_{12})^2. \tag{32}$$

Usually the NMR resonance frequency for nuclei ^{57}Fe is $\omega_0 \approx 10^8\ rad \cdot \sec^{-1}$ [1]. Assuming that $\sqrt{M_2} \approx \left(10^3 \div 10^6\right) rad \cdot \sec^{-1}$ [1], we have from Equation (31)

$$\frac{\left(T_1^{-1}\right)_{\text{int}}}{\left(T_1^{-1}\right)_{HF}} = \frac{4M_2}{\omega_0^2} \approx 4 \cdot 10^{-4} \div 4 \cdot 10^{-10}. \tag{33}$$

It follows from the obtained results (Equation (33)) that the main contribution to the nuclear spin-lattice relaxation rate of magnetic nuclei with spin $I = 1/2$ in the magnetically ordered solids is caused by thermal fluctuations of hyperfine magnetic field. The nuclear spin–spin interactions, modulated by fluctuations in electron magnetization direction, give negligibly small contribution to nuclear spin-lattice relaxation. As a result, the proposed mechanism cannot be used to explain the dependence of longitudinal relaxation time on the concentration of magnetic nuclei, which was observed experimentally [2].

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