

# Dynamics of electron-nuclear spin systems in solids

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A new mechanism is proposed for the electron-nuclear cross relaxation caused by nuclear spin-spin flips. Equations connecting the polarizations of the spin packets in an inhomogeneously broadened ESR line with the reciprocal temperature of the nuclear dipole-dipole pool (NDDP) are derived. A method of identifying the proposed mechanism is proposed, based on the change of the ESR line shape when the NDDP is cooled.

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Electron and nuclear spin systems that are fixed in solids are coupled by dipole-dipole interactions, and the dynamic processes in these systems are related as a result.<sup>[1]</sup> Electron spin echo (ESO) investigations<sup>[2,3]</sup> have shown that in samples with sufficiently low concentrations of paramagnetic centers the electron magnetic phase relaxation is due to nuclear spin-spin transition. It is shown in<sup>[4]</sup> that these transitions form spin packets in inhomogeneous electron spin systems. When the electron spin-spin interactions are taken into account, the overlap of such spin packets leads to thermal contact between the electron Zeeman pools and the nuclear dipole-dipole pool (NDDP).

The Hamiltonian of the system in question is given by

$$\begin{aligned} \mathcal{H} &= \sum_j \omega_j S_j^z + \sum_j \mathcal{H}_j + V + \omega_I \sum_k I_k^z = \mathcal{H}_S + \omega_I \sum_k I_k^z, \\ \mathcal{H}_j &= \mathcal{H}_{II} + \sum_k a_{kj} S_j^z I_k^z, \\ V &= \frac{1}{2} \sum_{j,k} B_{jk} (S_j^+ S_k^- + S_j^- S_k^+). \end{aligned} \quad (1)$$

Here  $\mathcal{H}_{II}$  is the Hamiltonian of the dipole-dipole interactions of the nuclear spins. It is assumed that the scatter of the Zeeman frequencies of the electrons exceeds the dipole-dipole broadening. Therefore the spin-spin transitions due to the operator  $V$  are predominantly of the cross-relaxation type. In intervals exceeding the lifetimes of the nuclear spin states, the described spin system can be described by a quasi-equilibrium density matrix<sup>[4]</sup>

$$\rho_{eq} = Z^{-1} \exp \left\{ - \sum_j (P_j^s S_j^z + \beta_j \mathcal{H}_j) - P^I \sum_k I_k^z \right\}. \quad (2)$$

Following the standard procedure<sup>[5,6]</sup> we find that the quantities  $P_j^s$  and  $\beta_j$  are connected in the low-temperature approximation by the equations

$$\begin{aligned} \frac{\partial}{\partial t} P_j^s &= \sum_i W_{ji} \left\{ P_i^s - P_j^s + \frac{\Delta_{ji}}{2} (\beta_j + \beta_i) \right\}, \\ \frac{\partial}{\partial t} \beta_j &= \frac{2 \langle S_j^z \rangle^2}{\langle \mathcal{H}_j^2 \rangle} \sum_i \Delta_{ji} W_{ji} (P_i^s - P_j^s - \Delta_{ji} \beta_i) + V_{ji} (\beta_i - \beta_j), \end{aligned} \quad (3)$$

where

$$\begin{aligned} W_{ji} &= 2 \frac{B_{ji}^2}{\langle S_j^z \rangle} \int_0^\infty G^2(t) \cos \Delta_{ji} t dt, \quad \Delta_{ji} = \omega_j - \omega_i, \\ V_{ji} &= -2 \frac{B_{ji}^2}{\langle \mathcal{H}_j^2 \rangle} \int_0^\infty G(t) \frac{\partial^2}{\partial t^2} G(t) \cos \Delta_{ji} t dt, \\ G(t) &= \langle \exp(i\mathcal{H}_j t) S_j^- \exp(-i\mathcal{H}_j t) S_j^+ \rangle, \quad \langle \dots \rangle = \text{Sp}(\dots) / \text{Sp} 1. \end{aligned}$$

In the derivation of (3) it was assumed that the energy of the interaction of the electrons with the constant magnetic field greatly exceeds the energy of the magnetic interaction between the electrons and the holes. In the opposite case, it is necessary to include in the Hamiltonian (1) of our system the term

$$\sum_k (C_{kj} I_k^+ + C_{kj}' I_k^-) S_j^z,$$

which is responsible for the so-called forbidden electron-nuclear transitions.

A consequence of Eqs. (3) is the predicted possibility of observing the NDDP by ESR methods. It is known<sup>[7]</sup> that in a nuclear spin system situated initially in a state of thermodynamic equilibrium with reciprocal temperature  $\beta_L$  it is possible to transform the Zeeman ordering of the nuclear spins into dipole-dipole ordering. The spin system considered by us, with a dipole-dipole pool cooled in this manner, is described by a density matrix of the type

$$\rho = Z^{-1} \exp \left\{ - \sum_j P_j^s S_j^z - \beta \mathcal{H}_D \right\}. \quad (4)$$

Here  $\mathcal{H}_D = \sum_j \mathcal{H}_j$ , while  $\beta$  is the maximum value of the reciprocal temperature of the dipole-dipole pool and is given by

$$\beta = 1/2 \beta_L \omega_I (\text{Sp } S^2 / \text{Sp } \mathcal{H}_D^2)^{1/2}.$$

An investigation of equations such as (3), carried out in in<sup>[8]</sup>, has shown that the effective cross relaxation establishes a new quasi-equilibrium, given by a density matrix

$$\rho_n = Z^{-1} \exp \left\{ - P_0^s \sum_j S_j^z - \beta_n \left( \mathcal{H}_S - \omega_S \sum_j S_j^z \right) \right\}. \quad (5)$$

We obtain the value of  $\beta_n$  from the energy conservation condition in the cross-relaxation process:

$$\text{Sp } \rho \left\{ \sum_j (\omega_j - \omega_s) S_j^z + \mathcal{H}_D \right\} = \text{Sp } \rho_k \left\{ \sum_j (\omega_j - \omega_s) S_j^z + \mathcal{H}_D \right\}. \quad (6)$$

We assume that the time of the restructuring of the Zeeman ordering into dipole-dipole ordering is shorter than the cross-relaxation time. We can therefore put  $P_j = \omega_j \beta_L$ . For the case of a continuous distribution of the frequencies  $\omega_j$ , we have

$$\beta_k = \frac{\beta_L}{1+\epsilon} \left( 1 + \frac{\epsilon \omega_r}{2D} \right), \quad \epsilon = \frac{D^2}{M_2}, \quad D^2 = \frac{\text{Sp } \mathcal{H}_D^2}{\text{Sp } S_j^2}. \quad (7)$$

Here  $M_2$  is the second moment of the ESR line shape. Under conditions when the ESR line center is saturated<sup>[9]</sup> the coefficient of absorption of a control microwave field of frequency  $\Omega$  by a quasi-homogeneous spin system is given by

$$\chi = A (\Omega - \omega_s) g (\Omega - \omega_s) \beta_L \left( \frac{1 + \epsilon \omega_r / 2D}{1 + \epsilon} \right), \quad (8)$$

where  $A$  is a proportionality coefficient. An estimate of  $\chi$  shows that the effect of the thermal contact between the NDDP and the low-frequency electron pool<sup>[8]</sup> can be easily observed because of the appreciable distortion of the ESR line shape when the NDDP is cooled under conditions of effective electron-nuclear cross relaxation. This distortion is predicted by formula (8).

Another consequence of the equations in (3) is the predicted existence of a channel for spin-lattice relaxation of the NDDP energy via the low-frequency electron pool. Let us consider the limiting case of a simple two-step relaxation. Let  $\tau$  be the time required for the quantity

$$\sum_{j_i} \Delta_{j_i} W_{j_i} (P_j^s - P_{j_i}^s - \Delta_{j_i} \beta_{j_i})$$

to vanish, if the spin-lattice relaxation time  $T_1$  of the

polarizations of the electron spins is much longer than the time  $\tau$ , we can assume that the following equality holds over time intervals on the order of  $T_1$ :

$$\sum_j \beta_j = \sum_{j_i} \Delta_{j_i} W_{j_i} (P_j^s - P_{j_i}^s) / \sum_{j_i} \Delta_{j_i}^2 W_{j_i}.$$

Thus, in this case the spin-lattice relaxation of the quantity  $\sum_j \beta_j$  proceeds at a rate  $T_1^{-1}$ . Notice must be taken of the heuristic character of the last corollary, inasmuch as in nuclear spin-lattice relaxation via a paramagnetic impurity an important role is played by nuclear spin diffusion, a discussion of which is beyond the scope of the present note.

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## Investigation of the properties of a rotating He<sup>3</sup>-He<sup>4</sup> solution by the oscillating-disk method

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An experimental study was made of the increase of the radius of a vortex core as a result of the concentration of the light isotope in it. The critical velocities and their relaxation times in rotating solutions of He<sup>3</sup> in He<sup>4</sup> with different concentrations were also investigated.

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1. We report here the results of experiments that can be divided into two groups. In the first, the presence of the admixture of the light isotope plays the principal role. In the second are investigated phenomena

whose physical interpretation is so far insufficiently clear, despite of the presence of a large amount of experimental material. In these investigations, any additional information (in our case, information on the in-