# SPIN-LATTICE RELAXATION OF NUCLEI DURING NUCLEAR RESONANCE IN FERROMAGNETIC SUBSTANCES

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A mechanism for spin-lattice relaxation of nuclei during nuclear resonance in ferromagnetics is proposed. The results are in satisfactory agreement with the experimental data.

NUCLEAR magnetic resonance (NMR) in ferromagnetic cobalt has been studied by Portis and Gossard.[1] From the absorption characteristics, it turned out that the exchange field acting on the nuclei at the resonance frequency is a thousand times greater in amplitude than the external radio frequency field.

In order to explain this phenomenon, Portis and Gossard proposed that NMR in a ferromagnet is determined by the domain structure of the latter. It is known that a strongly inhomogeneous magnetic field exists in the boundary layer between two domains, i.e., in the domain wall. The exciting field brings about forced vibrations of the domain wall; as a result an additional alternating field is produced in the boundary layer. Estimates indicate[1] that this field is approximately $10^3$ times greater than the original one. On the basis of this Portis and Gossard concluded that during NMR in a ferromagnet the nuclei that are situated in the domain walls play the predominant role. This is further confirmed by the fact that when the domain structure is destroyed (by the action of a constant external field) the intensity of absorption drops sharply.

By taking spin diffusion into account, the effective spin-lattice relaxation time $T_1$ was then calculated from the relaxation time $\tau_l$ associated with the direct interaction with the lattice. It turned out that the observed $\tau_l$ was one order of magnitude less than the value obtained by assuming that the relaxation is accomplished by the indirect mechanism of Korringa,[2,1] i.e., via the hyperfine interaction between the nuclei and the conduction electrons. This suggests the existence of another mechanism predominating over Korringa's mechanism, at least in the boundary layer.

As was mentioned above, an internal, inhomogeneous magnetic field acts on the nuclei in the domain walls, as distinct from the other nuclei. Together with this, the thermal motion of the nuclei leads to the development of a time-varying magnetic field, which, on its part, evokes transitions between nuclear levels.

Let us consider the case of a 180° boundary. As the z direction we choose the direction of the constant internal field $\mathbf{H_n}$. The x axis is chosen in the plane of the boundary. Then the angle through which the field is rotated by a given displacement of the nucleus from its equilibrium position will be[1]

$$\vartheta = \pi u_y / \delta, \tag{1}$$

where $u_y$ is the displacement of the nucleus in the y direction, and $\delta$ is the thickness of the domain wall. The corresponding perturbing magnetic field is obtained in the form

$$H_x \approx \pi u_y H_n / \delta. \tag{2}$$

The displacement $u_y$ can be expanded in plane waves[3]:

$$u_y = \sum_{\mathbf{k}} (a_{\mathbf{k}} e^{i\mathbf{kr}} + a_{\mathbf{k}}^{+} e^{-i\mathbf{kr}}), \tag{3}$$

where $\mathbf{k}$ is a wave vector and $a_{\mathbf{k}}$ and $a_{\mathbf{k}}^{\dagger}$ are the annihilation and creation operators, respectively. The non-zero matrix elements of these operators, as is known,[3] have the form

$$\begin{aligned} (n_k - 1 \,|\, a_{\mathbf{k}} \,|\, n_k) &= \sqrt{\hbar n_k / 2m_0 N\omega_{\mathbf{k}}}\, e^{-i\omega_{\mathbf{k}} t}, \\ (n_k + 1 \,|\, a_{\mathbf{k}}^{+} \,|\, n_k) &= \sqrt{\hbar (n_k + 1)/2m_0 N\omega_{\mathbf{k}}}\, e^{i\omega_{\mathbf{k}} t}, \end{aligned} \tag{4}$$

where $n_{\mathbf{k}}$ is the occupation number of the k-th state, $\omega_{\mathbf{k}}$ is the frequency corresponding to the wave vector $\mathbf{k}$, $m_0$ is the mass of the nucleus, and N is the number of particles per unit volume.

The perturbation energy associated with the alternating field $H_x$ is determined from the relation

$$H' = \pi \frac{\hbar\omega_0}{2\delta} (I^{+} + I^{-}) \sum_{\mathbf{k}} (a_{\mathbf{k}} e^{i\mathbf{kr}} + a_{\mathbf{k}}^{+} e^{-i\mathbf{kr}}), \tag{5}$$

where $\omega_0 = \gamma H_n$ is the nuclear resonance frequency, and $I^{\pm} = I_x \pm iI_y$.

Finally, in agreement with [4], the transition probability between nuclear energy levels

$$W_{m,m-1} \approx (I+m)(I-m+1) \times (3\pi^3\hbar/4\delta^2 m_0)(\omega_0/\omega_m)^3 (e^{\hbar\omega_0/\Theta}-1)^{-1} \quad (6)$$

is easily obtained, where $\omega_m$ is the maximum Debye frequency and $\Theta$ is the temperature in energy units.

For $\hbar\omega_0/\Theta \ll 1$, which occurs at temperatures higher than about $10^{-2}$ °K,

$$W_{m,m-1} \approx (I+m)(I-m+1)(3\pi^3\omega_0^2/4\delta^2\omega_m^3 m_0)\Theta. \quad (7)$$

For metallic $Co^{59}$ ($I = 7/2$) the relaxation time $\tau_1 \approx 3 \times 10^{-3}$ sec (if we take $\omega_0/2\pi \approx 217 \times 10^6$ sec$^{-1}$, $\delta \approx 10^{-7}$ cm, [1] $m_0 \approx 10^{-22}$ g, and $\omega_m \approx 4 \times 10^{13}$ sec$^{-1}$), which is one order of magnitude less than the value calculated for the Korringa relaxation mechanism.

If it is assumed that outside the wall the Korringa mechanism remains dominant, the diffusion equations of Portis and Gossard [1] need to be generalized slightly. In accordance with this we have

$$\frac{dS_1}{dt} = wS_1 - \frac{S_1 - S_0}{\tau_1} + D_1\frac{d^2S_1}{dy^2} = 0 \quad (8a)$$

in the boundary layer and

$$\frac{dS_2}{dt} = -\frac{S_2 - S_0}{\tau_l} + D_2\frac{d^2S_2}{dy^2} = 0 \quad (8b)$$

in the domains. Here S is the nuclear spin polarization density, $S_0$ is its equilibrium value, w is the transition probability in the radiofrequency field, D is the nuclear spin diffusion coefficient, and the indices 1 and 2 refer respectively to the wall and the domains.

Selecting a solution that satisfies the condition of continuity of S and dS/dy in the boundaries, [1] and locating the mean value of the polarization $\langle S_1 \rangle$ in the boundary layer, we can determine the corresponding effective relaxation time $T_1$ from the relation

$$\langle S_1 \rangle = S_0/(1 + wT_1). \quad (9)$$

This gives

$$T_1 = \left[1 - \frac{(2/K_1\delta)\,\mathrm{sh}\,(K_1\delta/2)}{\mathrm{ch}\,(K_1\delta/2) + (\tau_1/\tau_l)(K_2/K_1)\,\mathrm{sh}\,(K_1\delta/2)}\right]\tau_1, \quad (10)^*$$

where $K_1^2 = 1/D_1\tau_1$, $K_2^2 = 1/D_2\tau l$, and $D_1$ and $D_2$ are the diffusion coefficients inside and outside the wall, respectively. [1] An estimate gives $K_1\delta/2 < 1$, as a consequence of which we obtain from (10)

$$T_1 \cong (3\pi^2/2)^{1/2}(\tau_1^4 T_2/\tau_l^3)^{1/2}, \quad (11)$$

where $T_2$ is the spin-spin relaxation time.

For $T_2 \approx 25 \times 10^{-6}$ sec, $\tau_l \approx 11.4 \times 10^{-3}$ sec, and $\tau_1 \approx 3 \times 10^{-3}$ sec, we find $T_1 \approx 140 \times 10^{-6}$ sec, which is close to the observed value ($T_1 \approx 280 \times 10^{-6}$ sec).

Furthermore, according to Eq. (7), $\tau_1$ is inversely proportional to $\Theta$. Since, as is well known, $\tau_l$ is also inversely proportional to $\Theta$, it follows from Eq. (11) that

$$T_1 \sim \Theta^{-1/2} \quad (11')$$

which is the dependence observed by Portis and Gossard. [1]

A three-phonon process was also considered; however, in view of the extremely large relaxation time the corresponding calculations are not presented here.

We note that the varying magnetic field acting on the nuclei situated in the boundary layer can also excite thermal vibrations of the domain wall as a whole. Since the characteristic frequency of domain wall vibrations lies in the same region as the nuclear resonance frequency in ferromagnets, [5] the corresponding relaxation mechanism will play the dominant role in some cases (in particular, when the difference in the frequencies is less than the nuclear resonance width).

Let us consider this question in detail. If we denote the amplitude of the thermal vibrations of the domain wall by $y_0$, then the amplitude of the exciting alternating field is determined by Eq. (2), where in place of $u_y$ it is necessary to substitute $y_0$. The amplitude of the thermal vibrations can be determined from the expression for the energy of an oscillator at thermal equilibrium, which gives

$$y_0 = (2\Theta/M\omega_0^2)^{1/2}, \quad (12)$$

where M is the mass of the wall ($M \sim 10^{-10}$ g). It is assumed here that $\hbar\omega_0 \ll \Theta$ and that the characteristic frequency of the domain wall coincides with the nuclear resonance frequency.

Following Khutsishvili, [6] we obtain for the transition probability

$$W_{m,m-1} \approx (I+m)(I-m+1)(\pi^2/4)(\Theta/M\delta^2)T_2^*, \quad (13)$$

where $1/T_2^*$ is the absorption line width. From this we have for the relaxation time in metallic $Co^{59}$ (taking $T_2^* = 2.5 \times 10^{-6}$ sec) $\tau_1 \sim 10^{-6}$ sec. An estimate shows that in this case $K_1\delta/2 > 1$; consequently we obtain from Eq. (10) $T_1 \sim \tau_1 \sim 10^{-6}$ sec.

Finally, we observe that since the proposed nu-

*sh = sinh, ch = cosh

clear relaxation mechanisms do not involve the conduction electrons, they will also play a definite role in ferroelectrics.

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