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# A Mechanism of Spin-Lattice Relaxation in Ferromagnetic Substances\*

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## Synopsis

A mechanism of the line broadening in a ferromagnetic resonance absorption was explained on the basis of the relaxation of spin orientation due to the interaction between spin wave and phonon fields in the crystal lattice. It was theoretically found that, at low temperatures, the line width was roughly proportional to  $3/2$ th power of the absolute temperature. The experimental relationship between the line width and the temperature was explained by the theory. It was also found by this consideration that the relaxation time of spin reorientation was greatly influenced by the mean free path of Debye wave in the crystal.

## I. Introduction

The mechanism of the line broadening in a ferromagnetic resonance absorption remains an unsolved problem from a theoretical point of view. As is well known,<sup>(1)</sup> the usual spin-spin interaction causes a line broadening, whereas, the exchange interaction makes the line width very narrow. Both factors, however, can not give a right order of magnitude of the line broadening. All other mechanisms of the line broadening in a ferromagnetic substance hitherto considered seem to be hopeless of explaining the observed line broadening. Spin-lattice relaxation mechanism<sup>(2)</sup> has also been shown to be insufficient for the explanation of the phenomenon, and the methods hitherto adopted for its estimation do not seem to be strictly correct. In the following pages a theory of line broadening due to spin-lattice relaxation is proposed, based on the spin wave picture of ferromagnet by taking into account the interaction between the spin wave and the Debye phonon fields.\*\*

When a ferromagnetic substance is subjected to the microwave field for a sufficiently long time and then the field is suddenly removed, the system will be reduced to the initial thermal equilibrium state with a relaxation time  $\tau$ . In the microwave resonance absorption, it is known that the line width contains the spin-lattice relaxation frequency  $1/\tau$  additively.<sup>(3)</sup> For the purpose of finding the behavior of  $\tau$ , we considered first the transition probability of the system between

\* The 675th report of the Research Institute for Iron, Steel and Other Metals.

\*\* Strictly speaking, only transversal waves are of importance for the following discussions.

(1) J. H. Van Vleck, *Phys. Rev.* **73** (1950), 266.

(2) D. Polder, *Phil. Mag.* **11** (1949), 99.

(3) N. Bloembergen, *Phys. Rev.* **73** (1950), 572.

the two states, in which the difference in  $z$ -component of the total spin eigen value is  $\mathcal{K}$ , the interaction between the field of the spin wave and that of the Debye-phonon being taken into account. In this case, the spin wave having the wave number  $\vec{k}$  is created or annihilated and, corresponding to this Debye wave is annihilated, in such a way that both the energy and momentum of the system are conserved. Using the above-mentioned transition probability, the relaxation time  $\tau$  for the change of distribution function of the spin wave field due to the sudden removal of microwave field could be obtained.

In the following calculation, the deformable ion model<sup>(4)</sup> was used for simplicity, and the temperature of the system was assumed to be sufficiently low, so that the state of the substance might be describable by the free spin wave.<sup>(5)</sup>

## II. Hamiltonian of the system and the transition probability

Total Hamiltonian  $H$  of the system contains three terms  $H_e$ ,  $H_l$ , and  $H'$ :  $H_e$  is the electronic term,  $H_l$  represents that of the lattice vibration and  $H'$  implies the interaction between the electrons and the lattice.

$$H = H_e + H_l + H'. \quad (1)$$

The electronic term  $H_e$  is further divided into two parts  $H_{e1}$  and  $H_{e2}$ :  $H_{e1}$  is the usual exchange term<sup>(6)</sup> and  $H_{e2}$  is the small term due to the spin-spin and spin-orbit interactions;

$$H_e = H_{e1} + H_{e2}, \quad (2)$$

$$H_{e1} = \frac{\mathcal{K}^2}{2m} \sum_i \Delta_i + \sum_{i,m} V_n(\mathbf{r}_i - \mathbf{R}_{n0}) + \sum_{i>k} \frac{e^2}{r_{ik}} + \sum_{m>n} V_{mn}(\mathbf{R}_m - \mathbf{R}_n) - \frac{eH}{2mc} \sum_i S_{zi}, \quad (3)$$

$$H_{e2} = \sum_{n,i} \frac{1}{2m^2 c^2} \left( -\frac{1}{r} \frac{\partial V}{\partial \mathbf{r}} \right)_{r=r_{ni0}} \cdot ((\mathbf{r}_{ni0}, \mathbf{p}_i], \mathbf{S}_i) + \sum_{i>k} \frac{e^2}{2m^2 c^2} r_{in}^{-3} ((\mathbf{r}_{ik}, 2\mathbf{p}_k - \mathbf{p}_i], \mathbf{S}_i) \\ + \sum_{i>k} \frac{e^2}{2m^2 c^2} r_{ik}^{-3} \left\{ (\mathbf{S}_i, \mathbf{S}_k) - 3r_{ik}^{-2} (\mathbf{S}_i, \mathbf{r}_{ik})(\mathbf{S}_k, \mathbf{r}_{ik}) \right\}. \quad (4)$$

And  $H_l$  is given by

$$H_l = \sum_{q,j} \mathcal{K} \omega_{qj} (N_{qj} + 1/2), \quad (5)$$

with  $N_{q,j} = a_{qj}^* a_{qj}$  and  $a_{qj} a_{q'j'}^* - a_{q'j'}^* a_{qj} = \delta_{qj q'j'}$ . In these expressions  $\Delta_i$  is the Laplacian operator with respect to the coordinate  $\mathbf{r}_i$  of the  $i$ -th electron,  $V_n(\mathbf{r}_i - \mathbf{R}_{n0})$  the potential energy of the  $i$ -th electron due to the  $n$ -th ion at the lattice point  $\mathbf{R}_{n0}$ ,  $\mathbf{r}_{ij}$  the distance vector between  $i$ -th and  $j$ -th electrons,  $e$  and  $m$  respectively the charge and mass of the electron,  $c$  the light velocity,  $H$  the external magnetic field,  $\mathbf{S}_i$  the spin operator of the  $i$ -th electron,  $\mathbf{p}_i$  the momentum operator of it, and  $\omega_{qj}$  the circular frequency of Debye wave having the wave number  $q$  with  $j$ -th polarization.  $a_{qj}$  and  $a_{qj}^*$  are respectively the annihilation and creation operator of the Debye wave mentioned above.

(4) A. Sommerfeld and H. Bethe, *Handb. der Phys.* XXIV, II (1933), 505.

(5) F. Bloch, *ZS. f. Phys.* 61 (1930), 206.

(6) A. Sommerfeld and H. Bethe, *Handb. der Phys.* XXIV, II (1933), 599.

The interaction term  $H'$  is also divided into two parts,  $H_1'$  and  $H_2'$ .  $H_1'$  is the part responsible for the spin reorientation now in question and  $H_2'$  represents the part which is independent of these transitions. They are given by the following expressions:

$$H_1' = \sum_{n,i} \frac{1}{2m^2c^2} \left\{ r^{-3} \left( \frac{\partial}{\partial r} - r \frac{\partial^2}{\partial r^2} \right) V_n \right\}_{r=r_{ni0}} \cdot ([\mathbf{r}_{ni0}, \mathbf{p}_i] \mathbf{S}_i) \cdot (\mathbf{r}_{ni0} \mathbf{u}_i), \quad (6)$$

$$H_2' = -\sum_{n,i} (\mathbf{u}_n, \mathbf{grad}_i V_n(\mathbf{r}_{ni} - \mathbf{R}_n)) + \sum_{n>m} (\mathbf{u}_n - \mathbf{u}_m, \mathbf{grad}_n V_{nm}(\mathbf{R}_{n0} - \mathbf{R}_{m0})) + \sum_{n,i} \frac{1}{2m^2c^2} \left( \frac{1}{r} \frac{\partial V_n}{\partial r} \right)_{r=r_{ni0}} \cdot ([\mathbf{u}_n, \mathbf{p}_i], \mathbf{S}_i), \quad (7)$$

where  $\mathbf{u}_n$  is given by the following form

$$\mathbf{u}_n = N^{-1/2} \sum_{q,j} \mathbf{e}_{qj} (a_{qj}^* \exp[-i(\mathbf{q}, \mathbf{R}_{n0})] + a_{qj} \exp[i(\mathbf{q}, \mathbf{R}_{n0})]). \quad (8)$$

In the above formulas,  $N$  is the total number of ions and  $\mathbf{e}_{qj}$  the unit vector indicating the direction of polarization of Debye wave.

Schroedinger equation of the system with  $r$  antiparallel spins with the eigenvalue  $E_{r, \dots, N_{qj}, \dots}$  and eigenfunction  $\Psi_{r, \dots, N_{qj}, \dots}$  is given in the first approximation by

$$(H_{01} + H_1) \Psi_{r, \dots, N_{qj}, \dots} = E_{r, \dots, N_{qj}, \dots} \Psi_{r, \dots, N_{qj}, \dots}, \quad (9)$$

where

$$E_{r, \dots, N_{qj}, \dots} = E_0 - NJ + 2J \sum_{j=1}^r \left\{ 3 - \cos\left(\frac{2\pi}{N} k_{jx}\right) - \cos\left(\frac{2\pi}{N} k_{jy}\right) - \cos\left(\frac{2\pi}{N} k_{jz}\right) \right\} + g\beta r H + \sum_{q,j} \mathcal{K} \omega_{qj} (N_{qj} + 1/2), \quad (10)$$

and

$$\Psi_{r, \dots, N_{qj}, \dots} = (r! N^r N!)^{-1/2} \sum_p^r \mathbf{p} \exp \left\{ \frac{2\pi i}{N} (n_1 k_{p1} + n_2 k_{p2} + \dots + n_r k_{pr}) \right\} \times \sum_{p'} (-)^{p'} \mathbf{p}' \psi(1) \alpha(1), \psi_2(2) \alpha(2), \dots, \psi(n_r) \beta(n_r) \dots \psi_N(N) \alpha(N) \cdot \Pi_{qj} \Phi(N_{qj}). \quad (11)$$

Hereafter we will use the following assumption concerning the form of the potential function:

$$V = -ze^2/r. \quad (12)$$

In this equation  $z$  means the effective charge of the ions. Thence, the perturbation term  $H_1'$ , which is of importance to the transition now under consideration, becomes\*

$$H_1' = N^{-1/2} \frac{3}{2i} \frac{ze^2 \mathcal{K}}{m^2 c^2} \sum_{n,i} \sum_{q,j} \left\{ a_{qj} r_{ni0}^{-5} (\mathbf{r}_{ni0}, \mathbf{e}_{qj}) ([\mathbf{r}_{ni0}, \mathbf{grad}_i] \mathbf{S}_i) e^{i(\mathbf{q}, \mathbf{R}_{ni})} + \text{Comp. Conj.} \right\}. \quad (13)$$

Using this this relation, the matrix elements of  $H_1'$ , which are different from zero, will be obtained as follows:

$$(r+1, N_{qj} + 1 | H_1' | r, N_{qj}) = i \frac{3}{4} \frac{ze^2 \mathcal{K}}{m^2 c^2} \pi \mathcal{K} \sqrt{N(r+1)} \sqrt{\frac{\mathcal{K}(N_{qj}+1)}{2M\omega_{qj}N}} F_{qj}, \quad (14)$$

\* For this calculation following relation is used:

$$\langle r \frac{d}{dr} \rangle = \int_0^\infty f(r) r \frac{d}{dr} f(r) r^2 dr = - \int_0^\infty f \left\{ r^3 \frac{df}{dr} + 3r^2 f \right\} dr = - \int_0^\infty f r \frac{df}{dr} r^2 dr - 3 \int_0^\infty f^2 r^2 dr = -3/2$$

$$(r-1, N_{qj}-1|H_1'|r, N_{qj}) = i \frac{3}{4} \frac{ze^2 \hbar}{m^2 c^2} \pi \hbar \sqrt{Nr} \sqrt{\frac{\hbar N_{qj}}{2M\omega_{qj}N}} F_{qj}^*, \quad (15)$$

In this equation  $F_{qj}$  represents the following expression:

$$\begin{aligned} F_{qj} &= \sum_n e^{i(qR_{n0})} (c_{qj}, [\mathbf{r}, \mathbf{R}_{n0}]) R_{n0}^{-5} = \frac{1}{|q|^2} (\mathbf{r}[\mathbf{q}, \mathbf{e}_{qj}]) \sum_n (qR_{n0}) e^{i(qR_{n0})} R_{n0}^{-5} \\ &\equiv i \frac{4\pi}{3} f(qd)(\mathbf{r}, [\mathbf{q}, \mathbf{e}_{qj}]) d^{-3}. \end{aligned} \quad (16)$$

and  $\mathbf{r}$  means a vector  $(1, i, 0)$  and  $d$  is lattice spacing.

The transition probability  ${}_A P_B$  from the state  $B$  to  $A$  will be obtained by usual way<sup>(7)</sup> as

$$\begin{aligned} {}_A P_B &= \frac{\partial}{\partial t} \sum_q |(A|H_1'|B)|^2 \frac{2\{1 - \cos(\Delta E/\hbar)t\}}{(\Delta E)^2} \\ &= \frac{\partial}{\partial t} \left( \frac{Gd}{2\pi} \right)^3 \int |(A|H_1'|B)|^2 \frac{2\{1 - \cos(\Delta E/\hbar)t\}}{(\Delta E)^2} q^3 dq \sin\theta d\theta d\varphi, \end{aligned} \quad (17)$$

where

$$\Delta E = g\beta H + 2J(1 - \cos(qa)) - \hbar\omega_{qj} \doteq g\beta H + Jg^2 d^2 - \hbar\omega_{qj}. \quad (18)$$

The integration in eq. (17) can be carried out in the following manner: In eq. (15),  $(\mathbf{r}[\mathbf{q}, \mathbf{e}_{q2}])$  and  $(\mathbf{r}, [\mathbf{q}, \mathbf{e}_{q3}])$  are found to be equal to  $iqe^{i\varphi}$  and  $\cos\theta e^{-i\varphi} q$ , respectively. Since  $\omega_{q2}$  and  $\omega_{q3}$  are both the circular frequencies of transversal waves, being equal to each other, the integration over  $\theta$  and  $\varphi$  yields\*

$$\begin{aligned} &\frac{\partial}{\partial t} \left( \frac{Gd}{2\pi} \right)^3 6\pi \left( \frac{4\pi}{3} \right)^2 \frac{qz^2 e^2 \hbar^4 \pi^2}{16m^4 c^4 d^6} \frac{\hbar N}{2NM} \\ &\quad \cdot \int_0^{\omega_m} [f(qd)]^2 \left\{ \begin{matrix} r+1 \\ r \end{matrix} \right\} \left\{ \begin{matrix} N_q+1 \\ N_q \end{matrix} \right\} \frac{4\sin^2(\Delta Et/2\hbar)}{(\Delta E)^2} \frac{q^4}{\omega_q} \left( \frac{\hbar\omega}{\hbar\omega} \right) d\omega \\ &= \frac{3\pi^3 G^3 z^2 e^4 \hbar^3}{4m^4 c^4 d^6 M u^2} \left[ (qd)^3 [f(qd)]^2 \left\{ \begin{matrix} r+1 \\ r \end{matrix} \right\} \left\{ \begin{matrix} (N_q+1) \\ N_q \end{matrix} \right\} \right]_{\Delta E=0}. \end{aligned}$$

Hence,

$${}_{r+1} P_r = W(N_q+1), \quad {}_{r-1} P_r = W N_q, \quad (19)$$

where

$$W \doteq r G^3 \frac{3\pi^3 z^2 e^4 \hbar^3}{4m^4 c^4 d^6 M u^2} (qd)^3 |f(qd)|^2 \quad (20)$$

### III. Relaxation time

Let us now consider the equilibrium of the system exposed to the microwave field. Let  $N_r$  be the relative probability with which the system occupies the states indicated by the number  $r$  of antiparallel spins, then the thermal equilibrium conditions under the onset of microwave field with the spin-lattice relaxation mechanism will be given by

$$N_r \{ W N_q + P_r \} = N_{r+1} \{ W(N_q+1) + P_{r+1} \}, \quad (21)$$

\*  $\int |(ie^{i\varphi} + \cos\theta e^{-i\varphi})|^2 d(\cos\theta) d\varphi = 6\pi$

(7) W. Pauli, Handb. d. Phys. XXIV, 1 (1933), 154. From (16), it will be evident that only the transversal waves are effective for the said transition.

where  $P_r$  is the probability for the unit spin change of the system with  $r$  antiparallel spins due to the microwave field: here we assume that  $P_{r+1}$  is nearly equal to  $P_r \approx P_{r+1}$ , since  $r \gg 1$ ). Indicating the system in the equilibrium states without microwave field by naught, the relation

$$N_r^0/N_{r+1}^0 = (N_q + 1)/N_q \tag{22}$$

holds good. Hence, the initial and final conditions of the system mentioned in I will be determined. Eased on these distributions, the following equation of relaxation is obtained at the sudden removal of microwave field:

$$\frac{d}{dt} N_r = [N_{r-1} W N_q - N_r W (N_q + 1)] - [N_r W N_q - N_{r+1} W (N_q + 1)]. \tag{23}$$

After some approximate calculations, this equation can be reduced to the following simple form:

$$df_r/dt = -Wf_r, \tag{24}$$

in which  $f_r$  means the difference between  $N_r$  and  $N_r^0$ . Hence, the relaxation time  $\tau$  of the spin reorientation is given by  $1/W$ . The expression  $W$  contains one undetermined parameter  $G^3$  which is the number of atoms contained in the present system and the order of magnitude of  $\tau$  will be given by an appropriate value of  $G$ , by using the relation

$$N_q = [e^{\hbar\omega_q/kT} - 1]^{-1}. \tag{25}$$

The results are shown in Table 1.

Table 1.

	$G$	$rG^{-3} = (M_0 - M_s)/M_s$	$\tau^{-1}$ (line width)
Calculated	100	0.1 %	$3.10^7$
		1.0 %	$3.10^8$
		10.0 %	$3.10^9$
		30.0 %	$1.10^{10}$
		5 %	$1.10^7$
Observed		Several percent	$10^9$

#### IV. Temperature dependence of line width

The order of magnitude of relaxation time is thus obtained; hence, that of line width estimated theoretically from these data may be seen to be in fairly good agreement with those estimated from the experiment.<sup>(3)</sup> If one assumes for the value of  $Gd$  the mean free path of phonon (several times the lattice spacing) and for  $rG^{-3}$  several percent, then the order of magnitude of  $\tau$  will be found to be  $10^{-9}$ , which gives a correct order of relaxation time obtained experimentally. Since the above expression of line width contains the factor  $r$ , which represents the number of antiparallel spin in a domain, this parameter is proportional to  $T^{3/2}$ , as is well known<sup>(5)</sup>; hence, for the temperature dependence of the line width, the  $T^{3/2}$ -law also holds good, assuming another parameter such as  $G^3$  and  $u$  to be nearly constant or to be a slowly varying function of temperature. If the origin of the line width

in the ferromagnetic absorption is due to the above mentioned mechanism alone, then the line width will become very narrow when the temperature falls to the

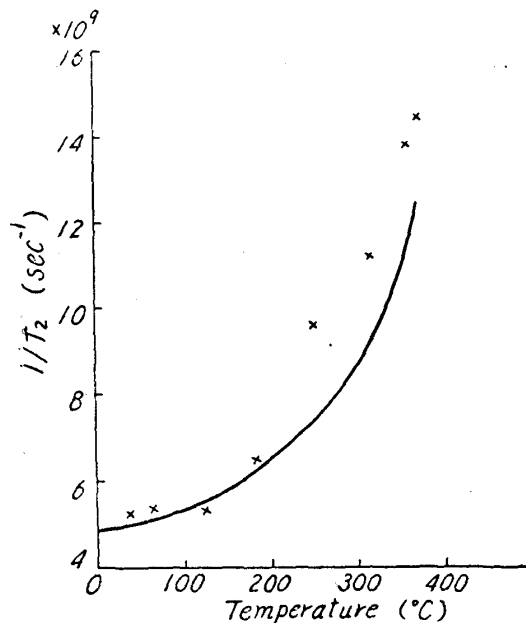


Fig. 1. The calculated line width for nickel as a function of temperature.  
× : Bloembergen's data.

absolute zero. This situation is contradictory to the experimental evidence.<sup>(8)</sup> There are, however, other mechanisms of line broadening which are independent of temperature such as due to internal stresses in the absorbing material, and, accordingly, the above-mentioned mechanism together with that considered here may give very similar temperature dependence to those found experimentally. The calculated results for nickel are shown in Fig. 1. The calculated curve in this figure was obtained by adjusting two constants in the two above-mentioned mechanisms. Thus we may reach the following conclusion: the temperature-dependent part of the line width in ferromagnetic resonance absorption is probably caused by the spin-lattice relaxation mechanism.\*

### V. Discussion of the results

In the above calculation, the spin-lattice relaxation process was treated on the assumption that the wave number vectors of excited spin waves due to the perturbation of the microwave field are distributed through wide range of the reciprocal lattice space. In other words, it was assumed that the excited spin waves by the microwave field with the wave number vector of zero were decomposed into those with various kinds of wave numbers. The occurrence of this processes may be due to the collisions between spin waves with each other or those of spin waves with the lattice imperfections. Hence the relaxation time of the latter process will influence that of microwave resonance if the former time is longer compared with the latter. In this calculation, only the latter relaxation time was considered.

The assumption of the deformable ion model mentioned above was adopted for the simplicity of calculation of the matrix elements. Since we are concerned only with the order of magnitude of the line width, the treatment of ionic and electronic fields in the crystal lattice like Bardeen's self-consistent method<sup>(9)</sup> is not needed, because the latter treatment can not give the different order of magnitude. Waller's<sup>(10)</sup> rigid ion model can not be used also from the same point of view.

\* Partly due to the eddy current damping, see reference (1).

(8) For example, see N. Bloembergen loc. cit.

(9) J. Bardeen, Phys. Rev. **52** (1937), 688.

(10) I. Waller, ZS. f. Phys. **79** (1932), 370.

The model of free spin waves is a fairly good one for the ferromagnet at low temperatures, because, in this case, the concentration of antiparallel spin is considered to be very small. If the temperature is not so low and, accordingly, the eigenfunction (11) becomes only an approximate one, the magnitude of relaxation time may be somewhat different from that calculated above except its order. The neglect of a part of Hamiltonian (4) causes i) the variation of  $H$  (demagnetization field) and ii) that of Lande's factor from 2; but these variations do not influence the value of matrix element in eq. (7). The reasons for these facts are as follows: the condition  $\Delta E=0$  in (17) gives two values of  $q$ ; one has long wave length and is dependent of the value of the Zeeman energy and the other has short wave length and is little dependent on it. The expression (20) of  $W$  contains the factor  $(aq)^3 f^2(aq)$  and, accordingly, the Debye waves with short wave length will be effective in this case. Thus the neglect of  $H_{0z}$  is justified.

In the calculation of the relaxation time, the reduced decay function (24) does not hold exactly, but it can be shown that the relaxation time obtained from this equation does not exceedingly diverge from the exact one. Here we shall consider the undetermined parameter  $(Gd)^3$ , the volume of domain. Based on the eq. (17), we may conclude that  $Gd$  seems to be equal to the path of coherent Debye wave  $l_D$  in the crystal and that this parameter will, then, be determined from the mechanism of the collision of the Debye waves with each other. The mean free pass  $l_D$  of Debye wave<sup>(11)</sup> is proportional to the thermal conductivity and inversely proportional to the heat capacity and to the velocity  $u$  of Debye wave. Since the thermal conductivity is nearly inversely proportional to the temperature, the heat capacity is a slowly increasing function of temperature and the velocity of sound wave  $u$  is a decreasing function of it and accordingly,  $l_D$  increases monotonously with the fall of temperature. The order of magnitude of  $G$  is 10~100 as cited in Table 1. If the estimated value for  $G$  is correct, then the line width may vary with temperature more slowly than  $T^{3/2}$  and these tendencies may be more accentuated by the factor  $u^{-2}$  contained in the expression of  $W$ .

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(11) P. Debye, *Vorlesungen über die kinetische Theorie usw.*, Leipzig.