

Application of the Diffusion-Modified Bloch Equation to Electron Spin Resonance in Ordinary and Ferromagnetic Metals

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A new solution to the electron spin resonance absorption in metals is given by using Bloch's equation modified to include diffusion. It is shown that the coefficient for a similar diffusion term added to the Landau-Lifshitz equation for ferromagnetic resonance must be of order 10^{-5} times smaller than that for nonmagnetic metals. A restricted formula for saturation in nonmagnetic metals is given.

I. INTRODUCTION

RECENTLY a modification of the Bloch equations has been proposed which includes the effects of the diffusion of carriers.¹ It occurred to the author that the modified Bloch equations with Maxwell's equations should describe electron spin resonance of the conduction electrons in metals. This problem has already been solved by a different approach.² The first-order time-dependent solution of a spin $\frac{1}{2}$ in a static field H_0 and an rf field H_1 was averaged over both a spin-lattice relaxation probability and a diffusion probability. This integral representation of the magnetization was then combined with Maxwell's equations to effect a solution. Dyson's method of solution is so different from the main stream of resonance calculations that a further important application of diffusion was overlooked, i.e., what effect does diffusion have on ferromagnetic resonance in metals? If the magnetic electrons in ferromagnetic metals are free to move around, they will contribute to line shape distortions. Recently a theory has been given to account for the observed line shapes of ferromagnetic resonance in metals with a skin depth smaller than the sample size.³ This theory, without allowing for any diffusion effects, appears to account for the experiments. From this result we conclude that the diffusion constant for the magnetic electrons must be of order 10^{-5} times less than that in nonferromagnetic metals. This point will be discussed later in the paper.

A crude calculation of the initial effects of saturation will also be given.

II. CALCULATION OF ELECTRON SPIN RESONANCE IN METALS

The equations we need are the Bloch equations for the transverse components of magnetization modified to include diffusion¹:

$$\partial \mathbf{M} / \partial t = \gamma (\mathbf{M} \times \mathbf{H}) - \mathbf{M} / T_1 + D \nabla^2 \mathbf{M}, \quad (1)$$

and Maxwell's equations, neglecting the displacement

current:

$$\nabla \times \mathbf{E} = - (1/c) (\partial \mathbf{H} / \partial t + 4\pi \partial \mathbf{M} / \partial t), \quad (2)$$

$$\nabla \times \mathbf{H} = (4\pi \sigma / c) \mathbf{E}.$$

In terms of the velocity v and relaxation time τ , $D = v^2 \tau / 3$.

Assume the metal occupies the positive half of z space. The magnetic field is taken along the z axis and the rf field is assumed circularly polarized. The x and y components of M , H , and E will vary as $e^{i\omega t - \kappa z}$. Substituting in Eqs. (1) and (2), we obtain the equations for the case of no saturation

$$\begin{aligned} i\omega M_x &= \gamma [M_y H_0 - M_0 H_y] - M_x / T_1 + \kappa^2 D M_x, \\ i\omega M_y &= \gamma [M_0 H_x - M_x H_0] - M_y / T_1 + \kappa^2 D M_y, \\ [1 + \frac{1}{2} i \delta^2 \kappa^2] H_x + 4\pi M_x &= 0, \\ [1 + \frac{1}{2} i \delta^2 \kappa^2] H_y + 4\pi M_y &= 0, \end{aligned} \quad (3)$$

where

$$\delta^2 = c^2 / 4\pi \sigma \omega.$$

Defining $\mathfrak{M} = M_x - iM_y$, $\mathfrak{H} = H_x - iH_y$, $K^2 = \kappa^2 \epsilon^2 \delta^2 / 2$, $\epsilon^2 = 2DT_1 / \delta^2$, $\alpha = T_1(\omega - \omega_0)$, and $\beta = \chi_0 \omega_0 T_1$, Eqs. (3) can be simplified to

$$\begin{aligned} [\alpha - i(1 - K^2)] \mathfrak{M} + \beta \mathfrak{H} &= 0, \\ [4\pi \epsilon^2] \mathfrak{M} + [\epsilon^2 + iK^2] \mathfrak{H} &= 0. \end{aligned} \quad (4)$$

For a solution we must have

$$\begin{vmatrix} K^2 - 1 - i\alpha & -i\beta \\ -i4\pi \epsilon^2 & K^2 - i\epsilon^2 \end{vmatrix} = 0, \quad (5)$$

or $K^4 - c_1 K^2 + c_2 = 0$, where

$$\begin{aligned} c_1 &= 1 + i(\alpha + \epsilon^2), \\ c_2 &= \epsilon^2 [4\pi \beta + i(1 + \alpha)]. \end{aligned}$$

Next it is noted that since the equations are all linear:

$$\mathfrak{M}_n = v_n \mathfrak{H}_n, \quad (6)$$

where

$$v_n = \frac{-i[K^2 - i\epsilon^2]}{4\pi \epsilon^2}, \quad (7)$$

¹ H. C. Torrey, Phys. Rev. **104**, 563 (1956).

² Freeman J. Dyson, Phys. Rev. **98**, 349 (1955).

³ W. S. Ament and G. T. Rado, Phys. Rev. **97**, 1558 (1955).

and

$$\mathcal{E} = E_x - iE_y = u_n \mathcal{H}_n, \quad (8)$$

where

$$u_n = iKc\sqrt{2}/4\pi\sigma\epsilon\delta.$$

The boundary conditions are tangential \mathcal{H} continuous and $\mathbf{k} \cdot \nabla \mathcal{N} = 0$, where \mathbf{k} is a unit vector in the z direction. This latter condition can be derived by integrating Eq. (1) over a flat disk with one face just outside the sample and the other face just inside the sample.⁴ In the limit as the disk volume goes to zero all terms except the diffusion term go to zero. The integrated diffusion term becomes $\mathbf{k} \cdot \nabla \mathcal{N}$, which must, therefore, also be zero. In component form the boundary conditions are

$$\begin{aligned} \mathcal{H}_1 + \mathcal{H}_2 &= \mathcal{H}_0, \\ K_1 v_1 \mathcal{H}_1 + K_2 v_2 \mathcal{H}_2 &= 0, \end{aligned} \quad (9)$$

where \mathcal{H}_0 is the magnetic field just outside the metal surface. The surface impedance,

$$Z = (4\pi/c) [\mathbf{k} \cdot \mathbf{E} \times \mathbf{H} / |H|^2], \quad (10)$$

becomes in the complex notation

$$Z = (4\pi/c) \text{Im}(\mathcal{E}/\mathcal{H}_0). \quad (11)$$

Define $X = \mathcal{E}/\mathcal{H}_0$, or

$$\mathcal{H}_0 X = (ic\sqrt{2}/4\pi\sigma\epsilon\delta) [K_1 \mathcal{H}_1 + K_2 \mathcal{H}_2]. \quad (12)$$

Let $X' = (4\pi\sigma\epsilon\delta/ic\sqrt{2})X$, giving finally the result

$$\mathcal{H}_0 X' = K_1 \mathcal{H}_1 + K_2 \mathcal{H}_2. \quad (13)$$

Equations (9) and (13) define the secular equation

$$\begin{vmatrix} 1 & 1 & 1 \\ X' & K_1 & K_2 \\ 0 & K_1 v_1 & K_2 v_2 \end{vmatrix} = 0,$$

with the solution

$$X' = \frac{ic_2^{\frac{1}{2}} [c_1 + 2c_2^{\frac{1}{2}}]^{\frac{1}{2}}}{\epsilon^2 + i[c_1 + c_2^{\frac{1}{2}}]}. \quad (14)$$

The absorption rate is given as

$$P = (c/4\pi) \mathcal{H}_0^2 \text{Im} X.$$

A comparison with the previous solution² is made by noting that β is very small as it is proportional to the Pauli susceptibility. Equation (14) is expanded in powers of β and terms only up to those linear in β are kept. The result then can be shown to be identical with the section in the previous calculation, called "(c) Thick Case."²

⁴ This method was suggested by G. T. Rado, who had used it for deriving the boundary condition resulting from the $\mathbf{M} \times \nabla^2 \mathbf{M}$ exchange term (to be published).

III. EFFECT OF DIFFUSION TERM ON FERROMAGNETIC RESONANCE

The Landau-Lifschitz Equation appropriate for ferromagnetic resonance including exchange and diffusion is

$$\frac{\partial \mathbf{M}}{\partial t} = \gamma \mathbf{M} \times [\mathbf{H} + (2A/M_s^2) \nabla^2 \mathbf{M} - (\lambda/\gamma M_s^2) \mathbf{M} \times \mathbf{H}] + D \nabla^2 \mathbf{M}, \quad (18)$$

where A is the exchange factor, M_s the saturation magnetization, and λ is a relaxation parameter. What needs to be compared is the ratio

$$D/[(2A\gamma)/M_s] = \phi. \quad (19)$$

For a good metal $D \simeq 10^8$, and for some iron-nickel alloys $A \simeq 10^{-6}$ and $M_s \simeq 10^3$. Therefore, ϕ is approximately 10^5 . Ament and Rado,³ using Eq. (18) without the diffusion term, have been able to obtain good agreement with the experimental line shape. The conclusion from this is that D for the magnetic electrons in ferromagnetic metals must be of order 10^{-5} times smaller than in good metal conductors.

IV. SATURATION

Saturation can be treated by numerically integrating Eqs. (1) and (2) with the appropriate boundary conditions. If it is assumed that $\epsilon \gg 1$, which implies the spin-lattice relaxation time is much greater than the time to cross the skin depth, the z component of magnetization should be relatively constant in a layer several skin depths thick inside the metal. Thus, a first order correction to saturation can be made by replacing M_0 in Eq. (3) by $\langle M_z \rangle$, where $\langle M_z \rangle$ is obtained from the equation

$$\partial M_z / \partial t = \gamma [\mathbf{M} \times \mathbf{H}]_z - [M_z - M_0] / T_1 + D \nabla^2 M_z;$$

therefore,

$$\langle M_z \rangle \simeq M_0 + \gamma T_1 \langle [\mathbf{M} \times \mathbf{H}]_z \rangle + D \langle \nabla^2 M_z \rangle,$$

where the averaging is to be performed at the surface. Assuming then that $D \langle \nabla^2 M_z \rangle$ can be neglected, as it has been assumed that $\langle M_z \rangle$ is almost constant near the surface, we have for $\langle M_z \rangle$ the expression

$$\langle M_z \rangle = M_0 + \gamma T_1 \mathcal{H}_0^2 \text{Im} Y, \quad (15)$$

where Y is defined from the determinant

$$\begin{vmatrix} 1 & 1 & 1 \\ Y & v_1 & v_2 \\ 0 & K_1 v_1 & K_2 v_2 \end{vmatrix} = 0,$$

in analogy with the calculation of X . Expanded, this becomes

$$Y = \left(\frac{-i}{4\pi\epsilon^2} \right) \left[\frac{c_2 - i\epsilon^2 c_1 - \epsilon^4}{c_1 + c_2^{\frac{1}{2}} - i\epsilon^2} \right]. \quad (16)$$

Replacing M_0 by $\langle M_z \rangle$ will, in effect, change β to $\beta \langle M_z \rangle / M_0$, and thus the previous calculation is only

altered by a multiplicative factor. To the term linear in β , Y is given by

$$Y \simeq (-i\beta) \left[\frac{1}{(1+i\alpha) + i^3 \epsilon (1+i\alpha) - i\epsilon^2} \right]. \quad (17)$$

Note that the assumptions for the validity of this approximation correspond to Dyson's section "(c₂) Thick Case, Natural Line."²

In this calculation of electron spin resonance in metals it has been assumed that the electrons are

acted on by H and not B .⁵ In the case of no saturation this is inconsequential, but the saturation expressions would depend on whether H or B is used. It is suggested that such experiments, interpreted using the numerical solution of Eqs. (1) and (2), could throw light on this question.

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⁵ G. H. Wannier, Phys. Rev. **72**, 304 (1947).

Conversion Electron Angular Correlations: General K -Shell Formulation and Threshold Limit*

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A general treatment of polarization-angular correlations involving conversion electrons is presented. Formulas are given for the b , coefficients for all types of polarization of the conversion electron, electric and magnetic multipoles, and arbitrary complexity ν . It is shown that only three coefficients are needed to characterize completely correlations of conversion electrons from the K shell, for a given type of transition. Curves are given of the threshold values for these coefficients for $L=1-5$, electric and magnetic, versus atomic number. The relativistic Coulomb functions (point nucleus, no screening) are presented, along with a number of properties and expansions, in a convenient and consistent notation. Several identities and relations involving the vector addition coefficients are included.

INTRODUCTION AND SUMMARY

ALTHOUGH not many measurements of angular correlations involving internal conversion electrons have been performed,¹ such measurements appear to be potentially useful experimental tools. In particular, the nonconservation of parity in beta decay has given a new impetus to the examination of the beta conversion-electron correlations.

Frauenfelder, Jackson, and Wyld² pointed out that the polarization of the recoil nucleus resulting from a beta decay (in an originally unpolarized nucleus) could be analyzed by angular correlations between the beta decay and a subsequent radiation (gamma or conversion electron) if the longitudinal polarization were measured. This analysis furnishes additional information on the beta interaction.

Rose and Becker in an interesting extension³ have noted that internal conversion electrons in coincidence with beta particles are partially transversely polarized

for odd values of ν . If \mathbf{e} and \mathbf{k} denote the directions of motion of the beta and conversion electrons, respectively, this polarization term is typified by $\boldsymbol{\sigma} \cdot \mathbf{k} \times (\mathbf{e} \times \mathbf{k})$. The longitudinal polarization terms also have odd ν and are of the type $(\boldsymbol{\sigma} \cdot \mathbf{k})(\mathbf{e} \cdot \mathbf{k})$. In addition to these terms, however, there are also transverse polarization terms for even ν , typically $(\boldsymbol{\sigma} \cdot \mathbf{k} \times \mathbf{e})(\mathbf{e} \cdot \mathbf{k})$.

Physically, the observation of the conversion electrons is analogous to analyzing circularly polarized gamma quanta by the photoelectric effect. Since the relative phase of the electron transition matrix elements is important, the use of internal gammas should enhance the effect because outgoing, as opposed to standing, waves occur in the internal conversion process.

A general treatment of the conversion electron- x (i.e., gamma, beta, etc.) correlation is presented in an extension of the method of Biedenharn and Rose.⁴ The importance of this method is that it greatly simplifies the problem, by the use of a parametrization due to Lloyd. It is shown below that for the K shell, with given values of multipolarity and parity, there exist only three quantities to characterize completely any x -conversion electron correlation [see Eq. (23) ff].

In the limit of high energy, the conversion electron

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¹ See, however, S. Frankel *et al.*, Phys. Rev. **93**, 1425 (1954); **100**, 1350 (1955); **102**, 1053 (1956); **105**, 1293 (1957); and **106**, 755 (1957).

² Frauenfelder, Jackson, and Wyld, Phys. Rev. **110**, 451 (1958).

³ M. E. Rose and R. L. Becker, Phys. Rev. Letters **1**, 116 (1958).

⁴ L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953).