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SPIN-RESONANCE TRANSMISSION IN PARAMAGNETIC METALS*

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A simple description of spin-resonance transmission in a paramagnetic metal is presented and applied to the recent observation of the effect in gadolinium. The resonance condition is shown to be $\gamma B_z = \omega_0$. As a consequence, resonance transmission occurs at a lower value of applied field than does resonance absorption. In conduction-electron spin-resonance transmission the same condition is shown to hold, and it is suggested that a measurement of the relative separation of the transmission and absorption resonances permits a determination of the conduction-electron spin susceptibility χ_s.

The observation of paramagnetic resonance transmission in gadolinium metal above the Curie point has recently been reported.[1] As in a conduction-electron spin-resonance (CESR) transmission experiment,[2-4] a transverse magnetization is resonantly excited at one surface of a metal foil and coherently detected at the opposite surface. In CESR transmission the specimen thickness is much greater than the skin depth, and transport of magnetization occurs via diffusion of the conduction electrons which, having left the skin layer at the excitation surface, retain phase memory and precess coherently for a transverse relaxation time T_2. Thus the condition for a successful observation of CESR transmission is that the specimen thickness should not be appreciably greater than the distance a conduction electron can diffuse in time T_2.

In Gd, however, the paramagnetic resonance absorption is due to S-state ions,[5] not conduction electrons, and diffusion of the spins is negligible. Moreover, T_2 is short. If CESR is characterized by weak paramagnetism and rapid diffusion, Gd represents the opposite limit: strong paramagnetism and no diffusion. An estimate, based on dc resistivity measurements,[6] gives a skin depth of 6 μ at the frequency used (9.2 GHz).[1] The 75-μ specimen was, therefore, much larger than the classical skin depth, although not as much as in a CESR experiment where the classical skin depth is about 1 μ and a specimen thickness is 30 μ or greater.[2-4] In a specimen many skin depths thick, however, a small increase in the skin depth results in a large increase in transmitted power; this proves to be the case in Gd.

A striking feature of the results in Gd is that the resonant transmission occurs at a lower value of external magnetic field than does the resonant absorption and that the relative displacement is greatest when the external field is perpendicular to the specimen.

The explanation of both types of transmission is that, associated with any resonant absorption, there is an anomalous dispersion, resulting in a resonant modulation of the transmission coefficient. Spin-resonance transmission is then, in principle, a general phenomenon, not restricted to CESR. This Letter contains a simple but detailed explanation of the results in Gd. It also points out that the relative shift of transmission and absorption should occur in CESR as well, where the interpretation is particularly simple.

A complete description of the propagation of electromagnetic fields in a medium requires, in addition to the Maxwell equations, constitutive relations which describe the response of the medium to the fields. In a good conductor the displacement current may be neglected and, with the additional assumption of Ohm's law, the Maxwell equations reduce to the eddy-current equation for the fields transverse to the direction of propagation, which is taken to be perpendicular to the surface of the specimen:

$$\nabla^2\vec{H} = (4\pi\sigma/c)(\partial\vec{H}/\partial t + 4\pi\partial\vec{M}/\partial t). \tag{1}$$

The constitutive relation must describe the resonant dependence of $\vec{M}$ on $\vec{H}$. If it is assumed that the only significant contribution to $\vec{M}$ is the paramagnetism of the ions, the appropriate relation is given by the Bloch equation for the transverse components,

$$\partial\vec{M}/\partial t = \gamma\vec{M}\times\vec{H} - \vec{M}/T_2. \tag{2}$$

(Here "transverse" is with respect to the external magnetic field.) In the following discussion the direction of the external field H is perpendicular to the surface of the specimen. For this geometry the z component of the magnetic field inside the metal is $H_z = H/(1+4\pi\chi)$, since $H_z = H - 4\pi M_z$ and $M_z = \chi H_z$. The x and y components of $\vec{M}$ and $\vec{H}$ will vary as $\exp(i\omega_0 t - kz)$, where ω_0 is the fixed frequency of the experiment. With this substitution, and the use of the complex fields $M^- = M_x - iM_y$ and $H^- = H_x - iH_y$, the Maxwell and Bloch equations are reduced to two homogeneous equations in M^- and H^-. The solution of the 2×2 secular determinant yields one root,

$$k^2 = [2i(1+4\pi\chi)/\delta^2][\gamma(H-H_0)T_2 + i] \times \{\gamma[H - H_0(1+4\pi\chi)]T_2 + i(1+4\pi\chi)\}^{-1}. \tag{3}$$

The expression has been written in a form appropriate to an experiment done at constant frequency in a varying external field H, and H_0 is defined by $\gamma H_0 = \omega_0$.

The numerator of this expression is resonant at $H = H_0$, corresponding approximately to maximum transmission. The denominator is resonant at $H = H_0(1+4\pi\chi)$ corresponding approximately to maximum k, and therefore to maximum absorption. The latter is not unexpected since a standard calculation of resonant absorption taking demagnetizing effects[7] into account indicates that it occurs when $H = H_0(1+4\pi\chi)$ for this geometry. The external field for maximum transmission is nearly independent of χ. Since B_z is continuous and equal to H, this implies that transmission occurs when $\gamma B_z = \omega_0$. The calculation when the external field is parallel to the surface gives similar results. In this geometry maximum absorption occurs when $H = H_0/(1+4\pi\chi)^{1/2}$, again in agreement with the simple demagnetization calculation. Maximum transmission occurs when $H = H_0/(1+4\pi\chi)$, again indicating that the resonance condition is $\gamma B_z = \omega_0$. The transmission signal is not, however, symmetric about this value because of the increased damping on the high-field side.

The high- and low-field limits of Eq. (3) are different:

$$k^2(0) = 2i/\delta^2,$$
$$k^2(\infty) = 2i(1+4\pi\chi)/\delta^2. \tag{4}$$

The low-field limit gives the classical skin effect in a medium of unit permeability, while the high-field limit gives the classical skin effect in a medium of permeability $1+4\pi\chi$. At low external fields, the variation of the high-frequency fields is nonadiabatic and the spins cannot follow; far above resonance the spins precess rapidly and can follow the high-frequency fields, which now vary adiabatically.

The transmitted fields will then vary as e^{-kd}, where d is the sample thickness. Since coherent detection is used, the detected signal will be proportional to

$$\exp(-k_R d)\cos(\theta - k_I d), \tag{5}$$

where k_R and k_I are the real and imaginary parts of k, and θ is the phase of the coherent reference. In Fig. 1, the results of machine calculations are shown for a choice of parameters corresponding to the reported measurements. Absorption and transmission signals are shown for both orientations of the external field. The absorbed power is proportional to the real part of the surface impedance, which is in turn proportional to k_R. The line shapes, positions, and relative intensities are all in good agreement with experiment.[1]

The only modification required to extend this calculation to CESR is the addition of a diffusion term to the Bloch equation, which accounts for the transport of magnetization by the mo-

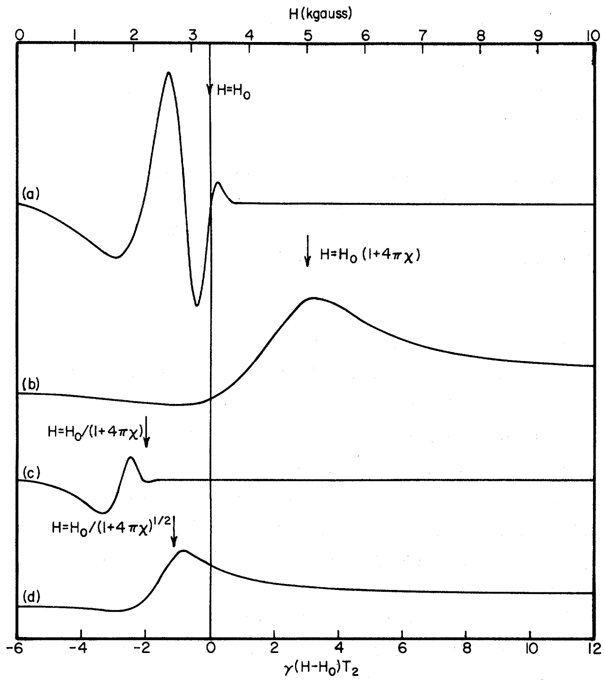

FIG. 1. Spin-resonance transmission and absorption signals versus $\gamma(H-H_0)T_2$ for the following parameters: $\gamma H_0T_2=6$, $4\pi\chi=0.5$, $d/\delta=12$, $\theta=60°$. (a) Transmitted signal with H perpendicular to the surface. (b) Absorption signal with H perpendicular to the surface. (c) Transmitted signal with H parallel to the surface. The scale is the same as (a). (d) Absorption signal with H parallel to the surface. The scale is the same as (b).

tion of the conduction electrons.[3,8] Greater care, however, must be used in writing the the torque term, which should properly be written as $\vec{M}_S\times\vec{B}$, where $\vec{M}_S$ is the spin contribution to the magnetization. If $\vec{M}=\vec{M}_S$ then $\vec{M}_S\times\vec{B}=\vec{M}_S\times\vec{H}$ and no distinction need be made. In CESR the distinction should be made, since orbital diamagnetism and the diamagnetism of the ion cores are a significant, although nonresonant, contribution to the magnetization. The appropriate Bloch equation is then

$$\partial\vec{M}_S/\partial t=\gamma\vec{M}_S\times\vec{B}-\vec{M}_S/T_2+D\nabla^2\vec{M}_S. \tag{6}$$

Once again the calculation described is for H perpendicular to the specimen surface. The modified Bloch equation, together with the Maxwell equations, results in a secular determinant yielding two roots,[3]

$$k_1^2=2i/\delta^2+i4\pi\chi_S H_0T_2/\delta_{\text{eff}}^2$$

and

$$k_2^2=2[1+i\gamma(H_0-H)T_2]/\delta_{\text{eff}}^2, \tag{7}$$

where $\delta_{\text{eff}}^2=2DT_2$. The second root describes CESR transmission; the resonant field is $H=H_0$, or equivalently, $\gamma B_z=\omega_0$. In CESR the spins, once they have diffused out of the skin layer, no longer interact with the exciting rf fields, and they precess in the static field B. The transmitted signal is now symmetric about the resonance condition because diffusion "decouples" the transmission and absorption resonances. To calculate the field for resonance absorption one must include the interaction with the exciting rf field; the result is $H=H_0(1+4\pi\chi_S)$, where χ_S is the spin susceptibility. The latter result is significant in an accurate determination of the g value and has been verified experimentally.[9]

The difference in the resonant fields for absorption and transmission depends only on χ_S, and suggests that a careful measurement of the relative shift may be a useful technique for measuring spin susceptibilities. Measurements are now being carried out on lithium, where the results may be compared with accurate measurements obtained by other methods.[10] An obvious extension would be to those metals in which the standard method, for various reasons, is not applicable.

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