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Doppler-Shifted Ultrasonic Spin Resonance in Metals*

Harold N. Spector
Physics Department, Illinois Institute of Technology, Chicago, Illinois 60616

and

J. B. Ketterson
Argonne National Laboratory, Argonne, Illinois 60439
(Received 11 December 1970)

A new method of measuring the conduction-electron g factor by ultrasonic Doppler-shifted spin resonance is presented. A Boltzmann-equation approach is used to obtain the attenuation arising from both the self-consistent-field and the Yafet mechanisms. The magnitude of the spin-dependent part of the attenuation is too small at the usual ultrasonic frequencies to be observed directly but the derivative of the attenuation should be observable.

I. INTRODUCTION

There now exist well-developed experimental techniques such as the de Haas–van Alphen (dHvA) effect, ultrasonic geometric resonance, microwave cyclotron resonance, etc., by which the shape of the Fermi surface and the Fermi velocity may be explored. Another interesting physical quantity associated with conduction electrons is their g factor. If the g factor is isotropic, the conduction-electron spin-resonance (CESR) technique may be used.[1] If the g factor is anisotropic, this technique is less useful, and other experimental techniques should be explored. The harmonic content of the amplitude of the dHvA oscillations contains information on the g factor associated with extremal orbits, but precise

measurements using this approach are difficult.

In this paper we propose that a variant of the well-known Doppler-shifted cyclotron-resonance (DSCR) technique[2-5] may be used to obtain information on the g factor in metals. Consider a sound wave in a metal propagating at an angle θ with respect to an external magnetic field H_0. Provided that H_0 is not too large, certain of the electrons will be in Doppler-shifted cyclotron resonance or Doppler-shifted spin resonance (DSSR) with the wave, i.e.,

$$\begin{Bmatrix}\omega_c\\ \omega_s\end{Bmatrix} = \left(1 \pm \frac{\langle V\rangle}{V_s}\cos\theta\right)\omega ,$$

where $\omega_c = eH_0/m^*c$ is the cyclotron frequency, $\omega_s = eH_0/m_sc$ is the Larmor frequency ($m_s \equiv 2m_0/g$), $\langle V\rangle$ is the average drift velocity along the field, and ω is the sound frequency. Electrons near the limiting point of the Fermi surface where $\vec{V}_F \| \vec{H}_0$ give rise to an absorption edge, the position of the edge being determined by m^*V_F or m_sV_F. (Another effect, the "density-of-states resonance" should also be noted.[4,5]) Thus, by varying the angle θ and from a knowledge of V_F over the surface, we can determine the limiting point value of m^* or m_s. (V_F can in principle be determined from the splitting of the absorption edge caused by the plus and minus sign in the Doppler condition.)

A theory of ultrasonic spin resonance in metals has been developed by Gerasimenko[6] using a Boltzmann-equation approach. However, he made certain approximations in his treatment which make his final results not applicable to the high-frequency region, where $qL \gg 1$. In Sec. II, we generalize his solution to the Boltzmann equation to treat the high-frequency region. We then calculate the ultrasonic absorption coefficient for the particular case where a circularly polarized shear wave is propagating along the magnetic field. In Sec. III we compare the absorption coefficient assuming different mechanisms for the induced magnetic field which couples the spins to the sound wave. We discuss our results and their possible experimental verification in Sec. IV.

II. ULTRASONIC SPIN MAGNETIZATION

We take as our model an electron gas, with each electron having a spin magnetic moment $\mu\vec{\sigma}$, interacting with a dc magnetic field $\vec{H}_0$ and an ac magnetic field $\vec{H}_1$ induced by the sound wave. The Boltzmann equation for this system is[7]

$$\frac{\partial \underline{f}}{\partial t} + \vec{V}\cdot\frac{\partial \underline{f}}{\partial t} + \frac{\partial \underline{f}}{\partial \vec{p}}\cdot\frac{e}{c}\vec{V}\times\vec{H} + \frac{i}{\hbar}[\mu\vec{\sigma}\cdot\vec{H},\ \underline{f}] + \left(\frac{\partial \underline{f}}{\partial t}\right)_{\text{coll}} = 0 , \tag{1}$$

where $\vec{H} = \vec{H}_0 + \vec{H}_1$, $\vec{\sigma}$ is the Pauli spin matrix and $[,]$ denotes the commutator. The distribution function $\underline{f}$ is an operator in spin space which can be represented by a 2×2 matrix and we represent the Pauli spin matrices by the standard notation:

$$\sigma_x = \begin{pmatrix}0 & 1\\ 1 & 0\end{pmatrix},\quad \sigma_y = \begin{pmatrix}0 & i\\ -i & 0\end{pmatrix},\quad \sigma_z = \begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix}.$$

The change in the distribution function because of collisions is due both to collisions in which the spin is flipped and collisions which do not flip the spin. Using a relaxation time ansatz we can write

$$\left(\frac{\partial \underline{f}}{\partial t}\right)_{\text{coll}} = \left(\frac{\underline{f} - \underline{f}_0}{\tau_s}\right) + \left(\frac{\underline{f} - \bar{\underline{f}}}{\tau}\right), \tag{2}$$

where τ_s is the spin-lattice relaxation time and τ is the ordinary electron relaxation time. In this expression $\underline{f}_0$ is the equilibrium distribution function to which the electron spins relax and $\bar{\underline{f}}$ is the average of $\underline{f}$ over the Fermi surface since collisions which conserve the electron spin do not lead to a change in the electron density:

$$\underline{f}_0 = \begin{pmatrix} f_0(\epsilon - \mu H_0) & 0\\ 0 & f_0(\epsilon + \mu H_0)\end{pmatrix},$$

$$f_0(\epsilon) = [e^{(\epsilon-\epsilon_F)/k_BT} + 1]^{-1} , \tag{3}$$

$$\bar{\underline{f}} = \frac{1}{4\pi}\int_{FS} d\Omega\, \underline{f} .$$

The magnetization induced by the magnetic field is

$$\vec{M} = (\mu/\hbar^3)\,\mathrm{Tr}\int d^3p\,\vec{\sigma}\underline{f} . \tag{4}$$

Since any 2×2 matrix can be expanded in terms of the identity matrix $\underline{I}$ and the Pauli spin matrices $\vec{\sigma}$, we can decompose $\underline{f}$ and $\underline{f}_0$:

$$\underline{f} = f\underline{I} + \vec{f}\cdot\vec{\sigma} , \qquad \underline{f}_0 = \gamma_+\underline{I} + \gamma_-\vec{n}\cdot\vec{\sigma} , \tag{5}$$

where $\gamma_\pm = \frac{1}{2}[f_0(\epsilon - \mu H_0) \pm f_0(\epsilon + \mu H_0)]$ and $\vec{n}$ is a unit vector in the direction of the dc magnetic field, $\vec{H}_0 = H_0\vec{n}$. Then

$$\vec{M} = (2\mu/\hbar^3)\int d^3p\vec{f} , \tag{6}$$

and using the commutation relations for the Pauli spin matrices $[\sigma_i, \sigma_j] = 2i\epsilon_{ijk}\sigma_k$, the Boltzmann equation for $\vec{f}$ takes the form

$$\frac{\partial \vec{f}}{\partial t} + \left(\vec{V}\cdot\frac{\partial}{\partial \vec{r}}\right)\vec{f} + \frac{e}{c}(\vec{V}\times\vec{H})\cdot\frac{\partial}{\partial \vec{p}}\vec{f} - \frac{2\mu}{\hbar}(\vec{H}_0 + \vec{H}_1)\times\vec{f}$$
$$= -\frac{(\vec{f} - \gamma_-\vec{n})}{\tau_s} - \frac{(\vec{f} - \bar{\vec{f}})}{\tau} . \tag{7}$$

Linearizing the Boltzmann equation to first order in quantities proportional to the ac magnetic field

$\vec{H}_1$, we have $\vec{f} = \gamma_- \vec{n} + \vec{f}_1$, where $\vec{f}_1 \propto \exp i(\vec{q}\cdot\vec{r} - \omega t)$ for a sound wave of wave vector $\vec{q}$ and frequency ω. This yields the following equation for $\vec{f}_1$:

$$\left[-i(\omega - \vec{q}\cdot\vec{V}) + \frac{1}{T}\right]\vec{f}_1 - \frac{2\mu}{\hbar}(\vec{H}_0\times\vec{f}_1 + \vec{H}_1\times\vec{n}\gamma_-) + \frac{e}{c}(\vec{V}\times\vec{H}_0)\cdot\frac{\partial}{\partial\vec{p}}\vec{f}_1 + \frac{(\gamma_-\vec{n} - \vec{f})}{\tau} = 0, \quad \frac{1}{T} = \frac{1}{\tau} + \frac{1}{\tau_s} . \tag{8}$$

For the magnetic field along the z direction and using notation $f_\pm = f_{1x} \pm i f_{1y}$ for circular polarization, the above equation reduces to the form

$$\left[-i(\omega \pm \omega_s - \vec{q}\cdot\vec{V}) + \frac{1}{T}\right]f_\pm + \frac{e}{c}(\vec{V}\times\vec{H}_0)\cdot\frac{\partial}{\partial\vec{p}}f_\pm = \mp\frac{2i\mu\gamma_- H_\pm}{\hbar} + \frac{\bar{f}_\pm}{\tau} , \tag{9}$$

where $\omega_s = 2\mu H_0/\hbar$ and $H_\pm = H_{1x} \pm iH_{1y}$. The solution to the above equation is

$$f_\pm = \int_{-\infty}^{s} ds' \left(\mp\frac{2i\mu\gamma_- H_\pm}{\hbar} + \frac{\bar{f}_\pm}{\tau}\right) \times\left\{\exp\left[-\int_{s'}^{s} ds''\left(-i[\omega \pm \omega_s - \vec{q}\cdot\vec{V}(s'')] + \frac{1}{T}\right)\right]\right\} , \tag{10}$$

where[6]

$$V_x(s) = V\sin\theta\cos(\omega_c s + \varphi), \quad V_y(s) = V\sin\theta\sin(\omega_c s + \varphi) ,$$
$$V_z(s) = V\cos\theta , \qquad \omega_c = eH_0/m^*c .$$

For a general orientation between the direction of propagation of the wave $\vec{q}$ and the dc magnetic field $\vec{H}_0$, the calculation follows the same lines as that of Cohen *et al.*[8] for the ordinary ultrasonic absorption

$$f_\pm = \left(\mp\frac{2i\mu\gamma_- H_\pm}{\hbar} + \frac{\bar{f}_\pm}{\tau}\right)I(\theta, \varphi) , \tag{11}$$

where

$$I(\theta, \varphi) = \int_{-\infty}^{s} ds' \exp\left(-\int_{s'}^{s} ds''\left[-i(\omega \pm \omega_s - q_z V\cos\theta) + \frac{1}{T} + iq_x V\sin\theta\cos(\omega_c s'' + \varphi)\right]\right)$$
$$= \sum_{n=-\infty}^{+\infty} \frac{J_n(X\sin\theta)\exp\{i[n(\omega_c s + \varphi) - X\sin\theta\sin(\omega_c s + \varphi)]\}}{1/T - i(\omega \pm \omega_s - n\omega_c - q_z V\cos\theta)} \tag{12}$$

and $X = q_x V/\omega_c$. Then we find that

$$\bar{f}_\pm = \mp\frac{2i\mu\gamma_- H_\pm}{\hbar}\int_{FS}\frac{d\Omega}{4\pi}I(\theta, \varphi)\Big/\left(1 - \frac{1}{\tau}\int_{FS}\frac{d\Omega}{4\pi}I(\theta, \varphi)\right) , \tag{13}$$

$$f_\pm = \mp\frac{2i\mu\gamma_-\tau H_\pm}{\hbar}\left[I(\theta, \varphi)\Big/\left(\tau - \int_{FS}\frac{d\Omega}{4\pi}I(\theta, \varphi)\right)\right] . \tag{14}$$

Using the above distribution function in Eq. (6) we find that $M_\pm = \chi_\pm H_\pm$, where

$$\chi_\pm = \mp\frac{2i\mu^2}{\hbar}\int\left[d\epsilon g(\epsilon)\gamma_-\tau\int\frac{d\Omega}{4\pi}I(\theta, \varphi)\Big/\left(\tau - \int_{FS}\frac{d\Omega}{4\pi}I(\theta, \varphi)\right)\right] \tag{15}$$

and $g(\epsilon)$ is the density of states for the electrons. When $\mu H_0 \ll \epsilon_F$, where ϵ_F is the Fermi energy, we get $\gamma_- = \mu H_0(\partial f_0/\partial\epsilon)$ and

$$\chi_\pm = \mp\frac{3i\mu^2 n_0\omega_s}{2\epsilon_F}\left[\tau\int_{FS}\frac{d\Omega}{4\pi}I(\theta, \varphi)\Big/\left(\tau - \int_{FS}\frac{d\Omega}{4\pi}I(\theta, \varphi)\right)\right] , \tag{16}$$

where n_0 is the electron density.

A. Propagation Parallel to Magnetic Field

When the sound wave is propagating parallel to the magnetic field $q_x = 0$, $q_z = q$ and we have

$$I(\theta, \varphi) = \frac{T}{1 - i(\omega \pm \omega_s - qV\cos\theta)T} , \tag{17}$$

$$\int_{FS}\frac{d\Omega}{4\pi}I(\theta, \varphi) = M + iH , \tag{18}$$

where[9]

$$M = \frac{1}{2qV_F}\left[\arctan qL\left(\frac{\omega \pm \omega_s}{qV_F} + 1\right) - \arctan qL\left(\frac{\omega \pm \omega_s}{qV_F} - 1\right)\right] , \tag{19}$$

$$H = \frac{1}{4qV_F}\ln\left\{\left[1 + (qL)^2\left(\frac{\omega \pm \omega_s}{qV_F} + 1\right)^2\right]\Big/\left[1 + (qL)^2\left(\frac{\omega \pm \omega_s}{qV_F} - 1\right)^2\right]\right\} .$$

In Eqs. (19), V_F is the Fermi velocity and $L = V_F T$. We then are left with the following expression for $\chi_\pm$:

$$\chi_{\pm}=\frac{\mp 3in_0\mu^2\omega_s\tau(M+iH)}{2\epsilon_F(\tau-M-iH)} . \tag{20}$$

B. Propagation Perpendicular to Magnetic Field

When the sound wave is propagating perpendicular to the magnetic field $q_x=q$, $q_z=0$, and we have

$$\int_{FS}\frac{d\Omega}{4\pi}I(\theta,\varphi)=\sum_{n=-\infty}^{+\infty}\frac{g_n(X)}{1/T-i(\omega\pm\omega_s-n\omega_c)} , \tag{21}$$

where[8]

$$g_n(X)=\int_0^{\pi/2}d\theta\sin\theta J_n^2(X\sin\theta) . \tag{22}$$

We then have the following result for $\chi_{\pm}$:

$$\chi_{\pm}=\left(\frac{\mp 3in_0\mu^2\omega_s\tau}{2\epsilon_F}\sum_{n=-\infty}^{+\infty}\frac{g_n(X)}{1/T-i(\omega\pm\omega_s-n\omega_c)}\right)\Big/\left(\tau-\sum_{n=-\infty}^{+\infty}\frac{g_n(X)}{1/T-i(\omega\pm\omega_s-n\omega_c)}\right) . \tag{23}$$

III. ABSORPTION COEFFICIENT

The power absorbed per unit volume in a system of changing magnetization is

$$Q=\mathrm{Re}\vec{H}\cdot\frac{d\vec{M}}{dt}=\omega\,\mathrm{Im}\vec{H}\cdot\vec{M}=\omega|H_1|^2\mathrm{Im}\chi . \tag{24}$$

The absorption coefficient is the power absorbed per unit volume divided by the incident acoustic flux

$$\alpha=\frac{Q}{\frac{1}{2}\rho V_s\omega^2|\xi|^2}=\frac{2}{\rho\omega V_s}\left|\frac{H_1}{\xi}\right|^2\mathrm{Im}\chi , \tag{25}$$

where ξ is the amplitude of the acoustic wave. The absorption due to the interaction between the ultrasound and the electron spins depends both upon the imaginary part of the ac magnetic susceptibility and the magnitude of the magnetic field induced by the sound wave.

For propagation of the ultrasound parallel to the magnetic field we have

$$\mathrm{Im}\chi=\frac{3n_0\mu^2\omega_s}{2\epsilon_F}\left(\frac{M-(M^2+H^2)/\tau}{(1-M/\tau)^2+(H/\tau)^2}\right) . \tag{26}$$

In the limit $qL\ll 1$ and $qV_F\gg|\omega\pm\omega_s|$ we get the result

$$\mathrm{Im}\chi=\frac{3n_0\mu^2\omega_s}{2\epsilon_F}\left(\frac{1/\tau_s+\frac{1}{3}(qL)^2/\tau}{[1/\tau_s+\frac{1}{3}(qL)^2/\tau]+(\omega-\omega_s)^2}\right) , \tag{27}$$

which is the result obtained by Gerasimenko.[6] In this limit, the condition for observing ultrasonic spin resonance is that $1/\tau_s<\omega<(3/\tau)|V_S/V_F|^2$ which favors low frequencies and high temperatures. Also under these conditions, the spin-resonance frequency will yield only an average value for the g factor since it is being averaged over all the electrons on the Fermi surface.

The other limit of interest when $\vec{q}\parallel\vec{H}_0$ is $qL\gg 1$. In that case

$$\mathrm{Im}\chi=\begin{cases}\dfrac{3\pi n_0\mu^2\omega_s}{4\epsilon_F qV_F}, & qV_F>|\omega-\omega_s|\\ 0, & qV_F<|\omega-\omega_s|\end{cases} \tag{28}$$

and we obtain a Doppler-shifted spin resonance. The condition for observing Doppler-shifted spin resonance is $qL\gg 1$ which favors low temperatures and in addition, the resonance frequency will yield the factor g/V_F at limiting points on the Fermi surface. If one knows the Fermi velocity at the limiting points from other experiments, one can determine the g factor at the limiting points of the Fermi surface from the resonance frequency.

For propagation of the ultrasound perpendicular to the magnetic field we will only discuss the same case as was considered by Mikoshiba,[9] $qL\gg 1$ and $\omega_c T\gg 1$. Under these conditions since $\omega\sim\omega_s\sim\omega_c$ and $X=(\omega/\omega_s)(V_F/V_s)\gg 1$ we can use the asymptotic forms for $g_n(X)$, i.e., $g_n(X)\approx 1/(2X)$ for $n<X$ and we find that[8]

$$\chi=\frac{3in_0\mu^2\omega_s\tau}{4qV_F\epsilon_F}\,\frac{\coth\pi\{[1-i(\omega-\omega_s)T]/\omega_c T\}}{\tau-(\pi/2qV_F)\coth\pi\{[1-(\omega-\omega_s)T]/\omega_c T\}} . \tag{29}$$

Although this expression yields the same conditions for observation of the spin resonance as Mikoshiba obtained, the expression for the absorption coefficient differs considerably from Mikoshiba's result. This is because the Boltzmann equation must be used to obtain valid results when $X>1$.[10]

The magnetic field which appears in Eq. (24) can arise from either of two mechanisms. For transverse waves propagating in a metal, a self-consistent magnetic field is induced because of the presence of transverse currents accompanying the ultrasound. This self-consistent magnetic field can be obtained using Maxwell's equations and the expression for the current density induced by the ultra-

TABLE I. Parameters used for bcc sodium.

g factor (g)	2.0015
Effective mass (m^*)	$1.27m_0$ g
Density (ρ)	1.01 g/cm^3
[100] sound velocity (V_s)	2.392×10^5 cm/sec
Fermi velocity (V_F)	8.40×10^7 cm/sec
Fermi energy (E_F)	4.09×10^{-12} erg
Lattice spacing (a_0)	4.225 Å

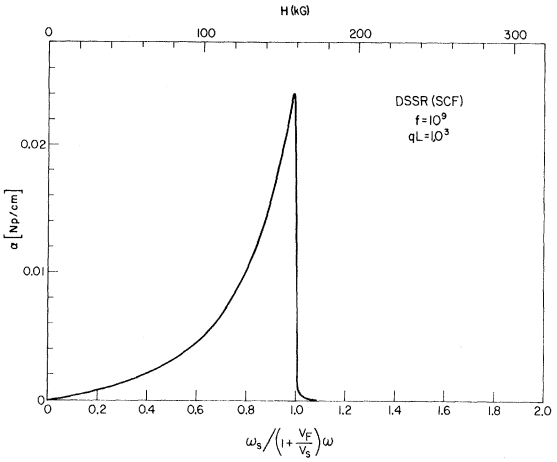

FIG. 1. Magnetic field dependence of the DSSR attenuation for the case of sodium and for $f=10^9$ Hz and $qL=10^3$.

sound[8]:

$$\vec{J}=n_0 e\frac{d\vec{\xi}}{dt}+\vec{J}_e\ , \tag{30}$$

where $\vec{J}_e=\overleftrightarrow{\sigma}\cdot\vec{E}$ is the induced electron current density, $\overleftrightarrow{\sigma}$ is the ac-conductivity tensor for the electrons, and $\vec{E}$ is the self-consistent electric field accompanying the ultrasound in a metal. The self-consistent magnetic field is then found to be

$$H_1=\frac{4\pi n_0 e(V_s/c)\xi}{1-(4\pi/i\omega)\sigma(V_s/c)^2}\ . \tag{31}$$

The second mechanism which can give rise to an induced magnetic field accompanying the ultrasound in a metal is that put forth by Yafet.[11] The ultrasound interacts with the electron spins through the ordinary electron-lattice interaction via spin-orbit coupling by the ultrasound. As a result of this mechanism, the matrix element for spin reversal is given by $Cq^2\Delta ga\xi$, where C is the deformation potential, $\Delta g=g-2.0023$ is the g shift, and a is the lattice spacing. Equating the matrix element of the term giving the interaction of the electron spin with the effective magnetic field induced by the ultrasound to the matrix element given by Yafet we find that the effective magnetic field is

$$H_1=q^2(C\,\Delta ga/\mu)\xi\ . \tag{32}$$

The induced magnetic fields in Eqs. (31) and (32) may be used in Eq. (24) to determine the absorption coefficient versus dc magnetic field for values of qL with propagation of the ultrasound parallel to the dc field.

IV. NUMERICAL RESULTS AND DISCUSSIONS

We must now inquire as to whether the predicted magnitude of the attenuation arising from DSSR is observable in the presence of the much stronger background attenuation caused by the DSCR. Since our formalism has been developed in the free-electron picture we will carry out our calculations for the case of sodium for which this approximation is applicable. The parameters used in the calculations have been collected in Table I. Calculations were carried out for frequencies of 10^8, 10^9, and 10^{10} Hz and for values of qL equal to 10, 10^2, and 10^3 (we have taken $\tau_s=\infty$). Figure 1 shows the spin portion of the attenuation for $f=10^9$ Hz and $qL=10^3$ for the self-consistent-field (SCF) mechanism. The Yafet mechanism is much smaller in this frequency range due to the small g shift in sodium.

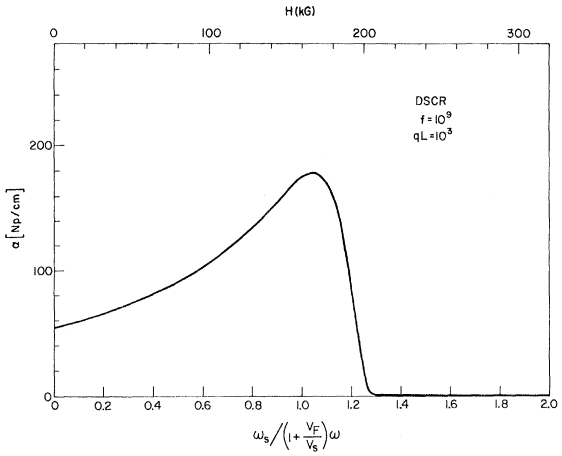

FIG. 2. Magnetic field dependence of the DSCR attenuation for the case of sodium and for $f=10^9$ Hz and $qL=10^3$.

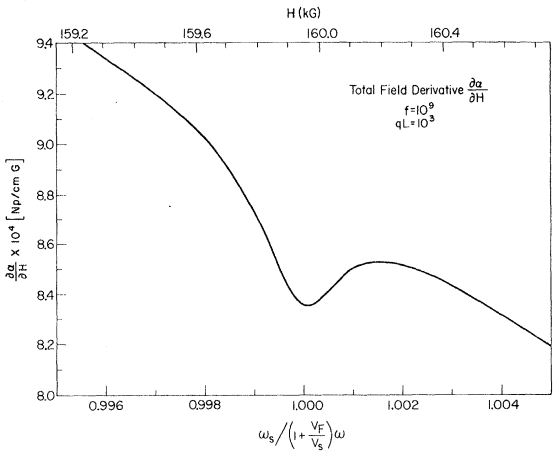

FIG. 3. Field derivative of the total attenuation in sodium as a function of magnetic field with $f=10^9$ Hz and $qL=10^3$.

For other materials (Bi, heavy transition metals, etc.) where larger g shifts are encountered, the Yafet mechanism may contribute significantly to the attenuation. The attenuation α is plotted as a function of both H and $\omega_s/(1+V_F/V_s)\omega$, the latter quantity being unity at the limiting point. Figure 1 clearly shows that, providing it is observable, the position of the DSSR edge can be used to accurately determine the limiting point g factor. Figure 2 shows the attenuation arising from DSCR which we observe is larger than the corresponding DSSR by a factor of order 10^4. Note that the shape of the DSCR attenuation curve is different from that usually expected[2] due to the breakdown of screening at high frequencies. At a frequency of 10^8 Hz the DSSR is approximately 10^6 times smaller than the DSCR; higher frequencies are clearly desirable. Owing to the lack of availability of steady-state fields larger than ~250 kG, frequencies larger than about 10^9 Hz are not of practical interest. Although the direct observation of DSSR in the attenuation is impractical, the same statement does not hold true for the derivatives with respect to field $\partial\alpha/\partial H$ or frequency $\partial\alpha/\partial f$. Figure 3 shows the total field dependence of $\partial\alpha/\partial H$ (arising from DSSR and DSCR) for a frequency of 10^9 Hz and a qL of 10^3. We note that there is a clearly observable anomaly in the magnitude of the derivative at the position of the DSSR edge. Since the magnitude of the derivative at the spin-resonance edge is proportional to qL, very pure samples are required. The maximum achievable qL would be limited by size-effect considerations which for an 0.1-cm sample of sodium corresponds to $qL \cong 2.6\times10^3$ with $f=10^9$ Hz. Thus the parameters used in calculating Fig. 3 represent conditions which are achievable in the laboratory with present experimental techniques. (Since the skin effect makes a field derivative experiment difficult to perform, a frequency-modulation approach may be more favorable.) Observation of $\partial^2\alpha/\partial H^2$ or $\partial^2\alpha/\partial f^2$ (by detection at the second harmonic of the modulation frequency) would, of course, enhance even more the DSSR near the position of the edge.

The present technique, being a resonance experiment similar to microwave CESR, determines a g factor which presumably does not contain the Stoner (or susceptibility) enhancement factor. This is to be compared with the g factor obtained from spin-wave transmission[12] or the harmonic content of the dHvA effect since these latter experiments contain the Stoner enhancement. Thus a comparison of these two types of experiments would yield information on the magnitude and anisotropy of the enhancement.

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