Conduction-Electron-Spin Resonance in Sodium and Potassium—Theory of Anomalies near the Critical Angle*

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The combined effects of surface relaxation and quasiparticle interactions on (i) the angular dependence of the g value and linewidth and (ii) the temperature dependence of the linewidth are studied using a formula given previously by the author. The theory predicts features qualitatively similar to the anomalous features observed by Schultz and Dunifer near the critical angle. The explanation is suggestive, but not complete, and further work is necessary.

Spin waves have been observed in sodium and potassium by Schultz and Dunifer.[1] The interpretation of these experiments in terms of the Landau-Silin[2] theory of Fermi liquids by Platzman and Wolff[3] and by Wilson and Fredkin[4] is convincing, but one would like to understand better the nature of the anomalies which occur near the critical angle Δ_c at which the spin-wave peaks coalesce.

Walker[5] has included the effect of surface relaxation, as well as Fermi-liquid interactions, in a theory of transmission-electron-spin resonance (TESR) through a thin slab (see Fig. 1). The present paper discusses a formula given in Ref. 5, and shows that the formula predicts anomalies similar to those observed.

By way of introduction, we give an intuitive discussion of the effects of surface relaxation. Consider a spatially nonuniform disturbance in the resonant component of the magnetization density M_- and let this disturbance be characterized by the wavelength λ. Suppose that the spatial nonuniformity is smoothed out by particles diffusing a distance $\frac{1}{2}\lambda$ from regions of high M_- density to regions of low M_- density. According to elementary diffusion theory, this takes a time $t\sim(\frac{1}{2}\lambda)^2/D$; the time t is the effective relaxation time of the nonuniform wave of spin density. A more accurate description of this process is given by the dispersion relation for the spin-dependent oscillations in an infinite medium, which can be written

$$\omega=\omega_0-i(1/\tau_s+Dk^2),$$

where ω_0 is the bulk electron-spin-resonance frequency and $k=2\pi/\lambda$. The first term in the brackets is the bulk spin-relaxation rate of the infinite-wavelength mode (e.g., due to spin-orbit coupling with impurities) and the second term is the relaxation rate associated with the process just described.

Now consider the normal modes of oscillation of the magnetization density in a metallic slab of thickness L. If there is no relaxation of the spins at the surface the number of spin-up electrons leaving the metallic surface equals the number of spin-up electrons striking it (and similarly for spin down). In this case there is no net current j of spin flowing out of the surface, and hence we must have $j=-D(dM/dz)=0$ at the surface. A spatially uniform precession of spins satisfies this boundary condition and is thus a normal mode; in fact this is just the mode excited in a typical conduction-electron-spin-resonance (CESR) experiment performed on a thin slab at low temperatures [so that $(D\tau_s)^{1/2}\gg L$]. This mode is shown in Fig. 2(a).

Now assume that there is spin relaxation at the surface. A spin-up electron striking the surface and making a transition to a spin-down state causes a loss of two Bohr magnetons at the surface; one because a spin-up is lost, and the other because a spin-down (which makes a negative contribution) is created. This loss of spin at the surface is accounted for in the macroscopic theory by making the spin current $j=-D(dM/dz)$ different from zero at the surface. Hence, the normal mode of oscillation of the magnetization density must have a somewhat bowed shape [see Fig. 2(b)] in order to provide a gradient of M, and hence a spin current, at the surface. Hence, surface relaxation causes the normal mode to be spatially nonuniform, and spatial nonuniformity gives rise to an additional relaxation rate Dk^2.

To calculate the wave number k associated with the inhomogeneous mode of Fig. 2(b), assume a magnetization density of the form

$$M_-(z)=A\cos kz+B\sin kz$$

and apply the boundary conditions describing surface relaxation (e.g., see Ref. 5):

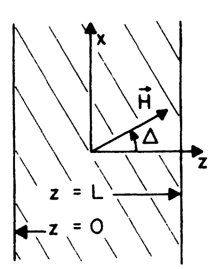

FIG. 1. Experimental geometry; the external field makes an angle Δ with the normal to the slab.

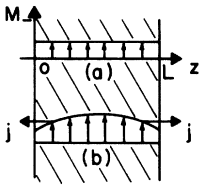

FIG. 2. Normal modes of oscillation of M_- with no modes for the following cases: (a) the absence of surface relaxation, and (b) the presence of surface relaxation. In the presence of surface relaxation, the mode must have a bowed shaped so that a current $j = -D(dM_-/dz)$ resulting from a gradient in M_- can flow out of the surface.

$$D\frac{dM_-}{dz}\bigg|_{z=0} = \frac{\epsilon' v}{2} M_-\bigg|_{z=0}; \quad D\frac{dM_-}{dz}\bigg|_{z=L} = -\frac{\epsilon' v}{2} M_-\bigg|_{z=L},$$

where $\epsilon' = \epsilon(1+B_0)/(1-\epsilon)$, ϵ being the probability of a spin flip at the surface, and B_0 a Landau-Fermi-liquid-theory parameter; v is the Fermi velocity. Thus, one finds that the wavelength of the normal modes are determined by the equation

$$k \tan kL = (\epsilon' v/D) + (\epsilon' v/2D)^2 k^{-1} \tan kL.$$

For the normal mode with no nodes, assuming $|kL| \ll 1$, one finds

$$k^2 = (\epsilon' v/DL) + (\epsilon' v/2D)^2.$$

Substituting this into the dispersion relation given above, one finds

$$\omega = \omega_0 - i\left(\frac{1}{\tau_s} + \frac{\epsilon' v}{L} + \frac{\epsilon'^2 v^2}{4D}\right),$$

which is the equivalent to the result of Eq. (6) below. As will be seen below, at a certain critical angle Δ_c (see Fig. 1 for a definition of Δ), the diffusion constant becomes very small, giving a large contribution to the observed width. Furthermore, since the diffusion constant is in fact a complex number [see Eqs. (3) and (4)] surface relaxation can also result in a change in the observed resonance frequency.

A more complete mathematical discussion of surface relaxation has been given by Walker.[5] Equation (5.13) of that paper states that the amplitude of the transmitted microwave field is given by

$$H_t \propto \frac{1}{D}\left(\frac{1}{k\tan(\frac{1}{2}kL) - C} + \frac{1}{k\cot(\frac{1}{2}kL) + C}\right), \quad (1)$$

where

$$k^2 = (i/D)(\omega - \omega_0 + i/\tau_s), \quad C = \epsilon' v/2D. \quad (2)$$

The diffusion constant D is given by

$$D = D_\parallel \cos^2\Delta + D_\perp \sin^2\Delta, \quad (3)$$

where

$$D_\parallel = \frac{\frac{1}{3}\overline{v}^2 \tau_0}{1 - i\omega_B \tau_0},$$

$$D_\perp = \frac{\frac{1}{3}\overline{v}^2 \tau_0 (1 - i\omega_B\tau_0)}{(1 - i\omega_B\tau_0)^2 + (\overline{\omega}_c \tau_0)^2} \quad (4)$$

and

$$\omega_B/\omega_0 = (B_0 - B_1)/(1+B_0),$$
$$(\overline{v}/v)^2 = (1+B_0)(1+B_1). \quad (5)$$

B_0 and B_1 are Fermi-liquid-theory parameters,[3] $\overline{\omega}_c = (1+B_1)\omega_c$, and τ_0 is the orbital collision time of the electrons.

Bringing the two terms in Eq. (1) to a common denominator, and making the approximation $|\frac{1}{2}kL| \ll 1$, one finds[6]

$$H_T \propto \left[\omega - \omega_0 + i\left(\frac{1}{\tau_s} + \frac{\epsilon' v}{L} + \frac{\epsilon'^2 v^2}{4D}\right)\right]^{-1}, \quad (6)$$

which is in agreement with the results arrived at intuitively above. We estimate the maximum value of $|\frac{1}{2}kL|$, which is of interest in calculations performed below, to be $|\frac{1}{2}kL| \sim 0.2$, and therefore Eq. (6) will be used for further discussion.

The term proportional to ϵ'^2 in Eq. (6) was neglected in the discussion following Eq. (5.13) of Ref. 5, and our interest in its consequences was stimulated by the work of Janossy and Monod.[7]

Separating the term $(\epsilon'^2 v^2/4D)$ into its real and imaginary parts, one finds that the resonance occurs at a frequency

$$\omega_R = \omega_0 - \tfrac{3}{4}\epsilon'^2(\overline{v}/v)^2 \hat{\omega}_B [X/(X^2+Y^2)] \quad (7)$$

and has a width given by

$$\frac{1}{\tau_R} = \frac{1}{\tau_s} + \frac{\epsilon' v}{L} + \frac{3}{4}\epsilon'^2\left(\frac{\overline{v}}{v}\right)^2 \frac{Y}{X^2+Y^2}. \quad (8)$$

The following definitions have been introduced:

$$X = \cos^2\Delta - (\hat{\omega}_B/\hat{\omega}'_B)\sin^2\Delta,$$
$$Y = (\omega_B\tau_0)^{-1}[\cos^2\Delta + (\hat{\omega}_B\omega_B/\hat{\omega}'_B\omega''_B)\sin^2\Delta] \quad (9)$$

and

$$\hat{\omega}_B = \omega_B[1 + (\omega_B\tau_0)^{-2}],$$
$$\omega'_B = \omega_B^{-1}(\overline{\omega}_c^2 + \tau_0^{-2} - \omega_B^2), \quad (10a)$$

$$\hat{\omega}'_B = \frac{\omega'_B[1 + 4(\omega_B\tau_0)^{-2}]}{1 - (2/\omega_B\omega'_B\tau_0^2)},$$
$$\omega''_B = \frac{\omega_B\omega'_B[1 - (2/\omega_B\omega'_B\tau_0^2)]}{2\omega_B + \omega'_B}. \quad (10b)$$

In the limit $\omega_B\tau_0 \gg 1$, the equations for X and Y reduce to

$$X = \cos^2\Delta - [B^2/(1-B^2)]\sin^2\Delta \quad (11)$$

and

$$Y = (\omega_B\tau_0)^{-1}\{\cos^2\Delta + [B^2(1+B^2)/(1-B^2)^2]\sin^2\Delta\}, \quad (12)$$

where $B = (\omega_B/\overline{\omega}_c)$. The critical angle Δ_c is by def-

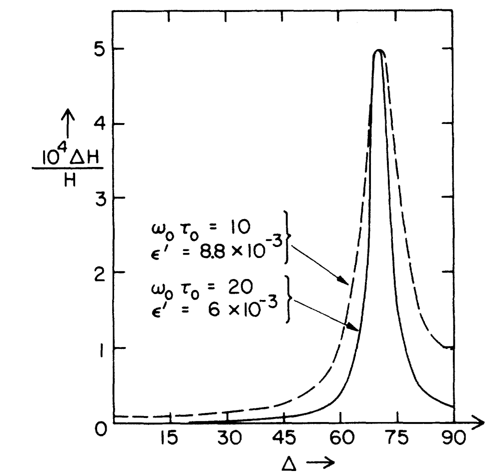

FIG. 3. Ratio $\Delta H/H$ where ΔH is the contribution of terms quadratic in ϵ' to the linewidth, and H is the field for resonance. Δ is plotted in degrees.

inition the angle at which $X(\Delta)=0$. From experimental observation,[1] $\Delta_c = 70°$ and Eq. (11) gives $B=\cos\Delta_c = \frac{1}{3}$.

Consider the contribution of the term quadratic in ϵ' to the width of the resonance. Figure 3 shows the ratio of the width of the resonance line, ΔH (in gauss), to the field for resonance, H, for two typical[1] values of $\omega_0\tau_0$. The parameter ϵ' is chosen so that $(\Delta H/H) = 5\times 10^{-4}$ at the critical angle Δ_c. The curve in Fig. 3 for the value $\omega_0\tau_0 = 10$ is similar to the experimental data[1] on linewidths as a function of angle.

Having chosen the magnitude of ϵ' to obtain the correct magnitude of the linewidth, there are no further adjustable parameters. The shift of the resonance field due to surface relaxation can now be calculated from Eq. (7) and is plotted as a function of Δ in Fig. 4. The curve for $\omega_0\tau_0 = 10$ agrees with experiment[1] in that it has the correct shape, and predicts the correct sign and magnitude of the shift in the field for resonance.

The observed temperature dependence of the linewidth is very unusual.[1] At the critical angle, the line narrows initially as the temperature is raised, whereas at $\Delta = 0°$ and $\Delta = 90°$ there is no initial narrowing. As the temperature is raised further to above 20 °K, the lines at all angles begin to broaden and have approximately equal widths. These features are accounted for at least qualitatively in our expression (8). Note that for $\Delta=\Delta_c$, $X=0$ and therefore $(\Delta H/H)$ is proportional to Y^{-1} which in turn is proportional to $\omega_0\tau_0$ [see Eq. (12)]. As the temperature is raised, τ_0 decreases and therefore so does $\Delta H/H$, which explains the initial narrowing of the line at $\Delta = \Delta_c$. For $\Delta = 0°$ or $\Delta = 90°$, $|X| > |Y|$ and therefore $(\Delta H/H)$ is proportional to Y which in turn is proportional to $(\omega_0\tau_0)^{-1}$; hence in these cases the line is expected to broaden as the temperature is raised; if, as may be the case at $\Delta = 0°$, the main contribution to the linewidth is the bulk relaxation rate τ_s^{-1}, the line is also expected to broaden as the temperature is raised.

Our theory also makes predictions about the angular dependences of the linewidth and g value as functions of temperature. For example, since the width of the curve of $(\Delta H/H)$ vs Δ is proportional to $(\omega_B\tau_0)^{-1}$, this width should increase as the temperature is raised. A rough check of this is the experimental result that at high temperatures the linewidth is independent of angle.[1]

The contribution of the term linear in ϵ' to the linewidth can also be calculated. Using the value $\epsilon' = 8.8\times 10^{-3}$ deduced above in fitting the angularly dependent part of the linewidth, and also using $L = 10^{-2}$ cm,[1] we find

$$\frac{\Delta H}{H} = \frac{2\epsilon v}{\omega_0 L} = 3\times 10^{-3}\,. \tag{13}$$

This is about two orders of magnitude larger than the experimentally observed angularly independent contribution to the linewidth. Hence, there is very definitely something wrong with our model.

Schultz has informed me[8] that the experiments were done under conditions such that strong surface relaxation of the conduction electrons is much more likely to take place where the sodium film comes into contact with the metal of which the microwave cavity is made than at the surfaces $z=\pm\frac{1}{2}L$ of the film indicated in Fig. 1. In such a model, the appropriate value of L to be inserted into the formula $(\Delta H/H) = (2\epsilon' v/\omega_0 L)$ would be the order of the size of the window in the microwave

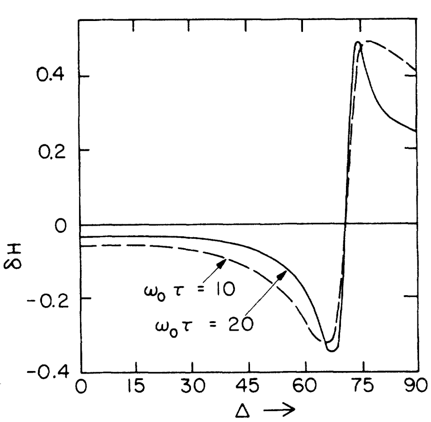

FIG. 4. δH is the shift in gauss in the field for resonance due to surface relaxation. A negative value of δH means that surface relaxation lowers the field for resonance. Δ is plotted in degrees.

cavity and the estimate (13) would be reduced by about two orders of magnitude. Thus it seems likely that the failure of our model is due at least in part to the fact that it does not correspond sufficiently closely to the actual experimental geometry.[9] The most obvious modification of Eq. (6) which is necessary for the new geometry is the redefinition of L to be the size of the window in the microwave cavity.

We shall use the name "edge relaxation" to describe the process in which electrons relax by coming into contact with the brass cavity walls at the edges of the sodium film. It appears that edge relaxation can account for the anomalous behavior observed near the critical angle by Schultz and Dunifer.

In concluding we note that our investigation of surface relaxation has led us to an expression for the linewidth proportional to the real part of D^{-1} and an expression for the shift in the resonance field proportional to the imaginary part of D^{-1}. The excellent agreement of these expressions with the experimental data means that we have at least established an empirical correlation between the real and imaginary parts of D^{-1} and the linewidth and g-value, respectively.

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[1]S. Schultz and G. Dunifer, Phys. Rev. Lett. **18**, 283 (1967); G. Dunifer, Ph.D. thesis (University of California, San Diego, 1968) (unpublished); detailed experimental data on the angular dependence of the g value and linewidth appear in the thesis.

[2]V. P. Silin, Zh. Eksp. Teor. Fiz. **35**, 1243 (1958) [Sov. Phys.-JETP **8**, 879 (1959)].

[3]P. M. Platzman and P. A. Wolff, Phys. Rev. Lett. **18**, 280 (1967).

[4]Andrew Wilson and D. R. Fredkin, Phys. Rev. B **2**, 4656 (1970).

[5]M. B. Walker, Phys. Rev. B **3**, 30 (1971).

[6]Dr. S. Schultz has informed me that P. Nozières and Mlle. Colette de Botton have independently studied the effects of surface relaxation and that their work is contained in the Masters thesis of Mlle. Colette de Botton (now Mme. Lewiner); (Colette de Botton: Memoire presenté á l'Université de Paris, January, 1968) (unpublished). In particular, they pointed out the existence of the contribution to the linewidth proportional to ϵ^2/D.

[7]A. Janossy and P. Monod (unpublished).

[8]S. Schultz (private communication); experimental work in progress by S. Schultz and D. Pinkel and is expected to clarify further the role of surface relaxation.

[9]J. F. Dobson has recently informed us that he had independently completed a theoretical investigation of the finite-slab problem some time ago, and that a summary of his results appear in Phys. Lett. A **44**, 171 (1973).