dependence on phonon relaxation with frequency, and the strength of the spin part of the many-body interaction. For the experimentalist we feel this work suggests the need for further dHvA g-value measurements where possible, and the extension of CESR to other frequencies.[8] By taking due consideration of the importance of g anisotropy and the relative size of $B/(1+B)$ compared to σ_g/g, it may now be possible to detect CESR in some of the many metals which have been examined, but which have yielded negative results.

Finally, the determination of the many-body spin parameters may eventually allow the observation of the finite-$\vec{k}$ collective modes (spin waves) as seen in the alkalis.[12]

We wish to express our appreciation to Professor P. Nozières for sending us the Master's thesis of his student Ms. Collette de Botton wherein we first had the suggestion of many-body exchange with g anisotropy brought to our attention. It is a pleasure to thank Professor D. Fredkin, Dr. R. Freedman, Dr. Y. Yafet, and Professor W. Kohn for many fruitful discussions.

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[1]See P. Monod and S. Schultz, Phys. Rev. 173, 645 (1968), for a description of our 9.2-GHz apparatus. For further details and for a description of the 35-GHz apparatus see M. R. Shanabarger, thesis, University of California, San Diego (unpublished); and G. L. Dunifer, thesis, University of California, San Diego (unpublished).

[2]At high temperatures the observed linewidth is corrected for finite sample thickness to yield $1/\gamma T_2{}^*$ as described by Dunifer, Ref. 1. At low temperatures the line shape should be a Lorentzian, which we found to be the case for Cu, while in Al the wings of the observed lines decayed somewhat faster than a Lorentzian. The deviations do not significantly affect the analysis of the data.

[3]S. Schultz *et al.*, Phys. Lett. 23, 192 (1966).

[4]Initial observations of a frequency dependence in CESR of Cu and Ag are discussed by Shanabarger, Ref. 1.

[5]R. J. Elliot, Phys. Rev. 96, 266 (1954).

[6]Y. Yafet, Solid State Phys. 14, 1 (1963).

[7]The possibility that motional narrowing has been observed for CESR in Be is reported in T. Kennedy and G. Seidel, Bull. Amer. Phys. Soc. 16, 1428 (1971).

[8]Theoretical, J. Dobson and D. Fredkin (to be published); experimental, D. Pinkel and S. Schultz (to be published).

[9]B. N. Aleksandrov and I. G. D'Yakov, Zh. Eksp. Teor. Fiz. 43, 852 (1962) [Sov. Phys. JETP 16, 603 (1963)].

[10]This interpretation differs from that suggested by R. Dupree and B. W. Holland, Phys. Status Solidi 24, 275 (1967).

[11]D. L. Randles, to be published.

[12]S. Schultz and G. Dunifer, Phys. Rev. Lett. 18, 283 (1967).

Theory of Many-Body Effects on Conduction-Electron Spin Resonance in a g-Anisotropic Metal*

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The variation of g over the Fermi surface leads to a breakdown of Larmor's theorem when scattering is too slow to motionally narrow conduction-electron spin resonance. We show that electron correlations lead to a collective mode whose position is not that of motionally narrowed conduction-electron spin resonance, and whose width would be zero in the absence of scattering. The shift and width of the collective mode are discussed in general and calculated in one experimentally interesting regime.

We have calculated the position and width of conduction-electron spin resonance (CESR) in a model metal in which there is a spread of "g values," due to some kind of spin-orbit coupling, and the conduction electrons are interacting. When the mean free time (τ) is short, we obtain the expected motional narrowing of the resonance, and the center of the Lorentzian line is given by the average g value ($\langle g\rangle \mu_B H_0$); the criterion for motional narrowing is, as usual, $\sigma_g \mu_B H_0 \tau \ll 1$ (σ_g is the rms width of the distribution of g values, μ_B the Bohr magneton, H_0 the dc magnetic field). When τ is sufficiently long, we again obtain a narrow Lorentzian line whose center is shifted slightly from $\langle g\rangle \mu_B H_0$; to observe this shift, we must have $B\langle g\rangle \mu_B H_0 \tau/(1+B) > 1$ (B is a dimensionless interaction parameter defined below; it can be thought of as similar to "B_0" in the Landau theory of a Fermi liquid). Observation of CESR in both of these regimes is reported in the prededing paper. The long-τ regime exhibits the following features:

(1) The linewidth $(1/T_2{}^*)$ would approach zero as $\tau \to \infty$ if there were no spin-lattice relaxation.

(2) The shift in the center of the resonance is a combined effect of the spread of g values and the electron-electron interactions. If there is no g-value spread, interactions have no effect whatever on CESR. If there are no interactions among the electrons, we should have a non-Lorentzian CESR which reflected the distribution of g values.

(3) If we neglect the variation of the spin-lattice relaxation time with temperature, we should expect the linewidth to pass through a maximum with increasing temperature, provided the sample is pure enough for it to be in the long-τ regime at the lowest temperature.

The narrow resonance in the long-τ regime, which we call the collective CESR, furnishes some information on the Landau parameter B in metals with complex Fermi surfaces in which spin waves[1] are difficult to observe. Moreover, as we explain at the end of this Letter, the frequency shift from the motionally narrowed CESR to the collective CESR can be substantial when $B/(1+B) \ll \sigma_g/g$; this large shift may be relevant to the failure to observe CESR in some metals.

Following Landau,[2] we view the conduction electrons as forming a gas of quasiparticles with effective Hamiltonian

$$E(k) = \mathcal{E}(k) + \tfrac{1}{2} g(k)\, \mu_{\mathrm{B}} \vec{\mathrm{H}} \cdot \vec{\sigma} + \delta E(k, r).$$

For simplicity, we use a scalar $g(k)$. Electron interactions are included by means of $\delta E(k, r)$, which, to avoid serious complication when $\mathcal{E}$ is anisotropic, we express as

$$\begin{aligned}\delta E_{\alpha\beta}(k, r) &= a \delta_{\alpha\beta} \operatorname{tr} \int \frac{d^3k'}{(2\pi)^3}\, \delta f(k', r) + b \sigma_{\alpha\beta} \cdot \operatorname{tr} \int \frac{d^3k'}{(2\pi)^3}\, \sigma f(k', r) \\ &= (a-b) \delta_{\alpha\beta} \operatorname{tr} \int \frac{d^3k'}{(2\pi)^3}\, \delta f(k', r) + 2b \int \frac{d^3k'}{(2\pi)^3}\, \delta f_{\alpha\beta}(k', r),\end{aligned}$$

with a and b constants; $\delta f = f - \eta(\mathcal{E}_{\mathrm{F}} - \mathcal{E})$, with $\mathcal{E}_{\mathrm{F}}$ the Fermi energy and f the quasiparticle distribution function. Our object is to calculate the rf (zero wave number) susceptibility $\chi_-(\omega)$ for the resonant rotating wave.[3] We do this by solving the Landau kinetic equation with collision term

$$\left(\frac{\partial \delta f_{\alpha\beta}}{\partial t}\right)_c = -\frac{1}{\tau} \delta\tilde{f}_{\alpha\beta} + \frac{1}{\nu} \int \frac{d^3k'}{(2\pi)^3}\, \delta(\mathcal{E}(k) - \mathcal{E}(k')) \left[\frac{2}{\tau_0} \delta\tilde{f}_{\alpha\beta}(k') + \frac{1}{\tau_s} \delta_{\alpha\beta} \operatorname{tr} \delta\tilde{f}(k')\right].$$

We have defined

$$\delta\tilde{f} = \delta f + \delta(\mathcal{E} - \mathcal{E}_{\mathrm{F}})(\tfrac{1}{2} g \mu_{\mathrm{B}} \vec{\mathrm{H}} \cdot \vec{\sigma} + \delta E),$$

which is the deviation of f from local equilibrium. The spin-flip scattering time is τ_s, the non–spin-flip scattering time is τ_0, $1/\tau = 1/\tau_0 + 1/\tau_s$, and ν is the density of states per unit volume for $\mathcal{E} = \mathcal{E}_{\mathrm{F}}$ (we have anticipated the fact that $\delta\tilde{f} \propto \delta(\mathcal{E} - \mathcal{E}_{\mathrm{F}})$).[4]

Calculation of $\chi_-(\omega)$ follows a standard pattern. We define $k(t)$ to be the electron trajectory in k space; it depends on an independent variable $k = k(0)$. We further let $g(t) = g(k(t))$,

$$\Phi(t, k) = \left[i \frac{\langle g \rangle \mu_{\mathrm{B}} H_0}{1+B} - i\omega + \frac{1}{\tau}\right] t + i \mu_{\mathrm{B}} H_0 \int_0^t dt'\, \Delta g(-t'),$$

where $\Delta g = g - \langle g \rangle$ and for any function F defined on the Fermi surface,

$$\langle F \rangle = \frac{2}{\nu} \int \frac{d^3k}{(2\pi)^3}\, \delta(\mathcal{E}(k) - \mathcal{E}_{\mathrm{F}}) F(k);$$

ν is the electron density of states at the Fermi surface. We find that

$$\chi_-(\omega) = \chi_0 + \frac{i\omega\nu\mu_{\mathrm{B}}{}^2}{4} \times \left\{ \frac{\langle g \rangle}{1+B} G_{10} + G_{11} + \left(\frac{\langle g \rangle}{1+B} G_{00} + G_{01}\right) \left[\frac{\langle g \rangle}{1+B} + \left(\frac{1}{\tau_0} - i\omega \frac{B}{1+B}\right) G_{10}\right] \left[1 - \left(\frac{1}{\tau_0} - i\omega \frac{B}{1+B}\right) G_{00}\right]^{-1} \right\},$$

where

$$G_{nm} = \langle \Delta g^n \int_0^\infty dt\, e^{-\Phi(t)} [\Delta g(-t)]^m \rangle.$$

The functions $G_{nm}(\omega)$, and therefore the rf susceptibility $\chi_-(\omega)$, have a series of branch cuts corresponding to power absorption by simple quasiparticle spin-flip transitions. Such absorption occurs whenever, for some cyclotron orbit characterized by k_z,

$$\omega = [\langle g \rangle/(1+B) + \langle \Delta g \rangle_{k_z}]\, \mu_B H_0 + n\omega_c(k_z),$$

where $\langle \Delta g \rangle_{k_z}$ is the average of $\Delta g(t)$ over the orbit, $2\pi/\omega_c(k_z)$ is the period of the orbit, and n is an integer; we put $1/\tau = 0$ for the purpose of studying elementary excitations. The shape of the single-particle absorption spectrum reflects the detailed distribution of g values and cyclotron frequencies. However, that interesting structure, intrinsic to the material, is blurred by $\Delta\omega \sim 1/\tau$.

In addition to the branch cuts just mentioned, $\chi_-(\omega)$ has poles when

$$1 - [1/\tau_0 - i\omega B/(1+B)] G_{00}(\omega) = 0, \tag{1}$$

corresponding to additional absorption with a Lorentzian line shape. At this point we confine our attention to the one pole whose imaginary part (width of the associated resonance) is dominated by $1/\tau_s$ rather than by the much larger $1/\tau_0$; we shall refer to this as collective CESR. We use as coordinates on the Fermi surface k_z and $\bar{t}$, the time required for an electron to move, under cyclotron motion, from the k_x-k_z plane to the point in question. We note that $\Delta g(k_z, \bar{t})$ is a periodic function of $\bar{t}$, and write

$$\Delta g(k_z, \bar{t}) = \sum_{m=-\infty}^{\infty} \Delta g_m(k_z) \exp[im\omega_c(k_z)\bar{t}].$$

Then for

$$\left| \frac{\langle g \rangle \mu_B H_0}{1+B} - \omega - \frac{i}{\tau} \right|^{-1} |\Delta g_0|\, \mu_B H_0 \ll 1, \tag{2}$$

$$|\Delta g_m|\, \mu_B H_0/m\omega_c \ll 1, \quad m \neq 0, \tag{3}$$

we can expand $G_{00}(\omega)$ in powers of Δg_m, substitute in (1) and obtain the complex pole

$$\omega \equiv g_{\text{obs}}\, \mu_B H_0 - i/T_2^*,$$

$$\frac{1}{T_2^*} = \frac{1+B}{\tau_s} + \frac{(1+B)\langle \Delta g_0^2 \rangle (\mu_B H_0)^2 \tau^*}{1+X^2} + 2(1+B)\tau^* \sum_{m=1}^{\infty} \left\langle \frac{|\Delta g_m|^2 (\mu_B H_0)^2 [1 + X^2 + (m\omega_c \tau^*)^2]}{[1 - X^2 + (m\omega_c\tau^*)^2]^2 + 4X^2} \right\rangle, \tag{4}$$

$$g_{\text{obs}} = \langle g \rangle \left[1 + \frac{B\langle \Delta g_0^2 \rangle (\mu_B H_0)^2 \tau^{*2}}{1+X^2} + 2B\tau^{*2} \sum_{m=1}^{\infty} \left\langle \frac{|\Delta g_m|^2 (\mu_B H_0)^2 [1 + X^2 - (m\omega_c\tau^*)^2]}{[1 - X^2 + (m\omega_c\tau^*)^2]^2 + 4X^2} \right\rangle \right], \tag{5}$$

where $1/\tau^* = 1/\tau_0 - B/\tau_s$ and $X = [B/(1+B)]\langle g \rangle \mu_B H_0 \tau^*$. Note that when $\omega_c\tau^* \ll 1$, we have, simply,

$$\frac{1}{T_2^*} = \frac{1+B}{\tau_s} + \frac{(1+B)\langle \Delta g^2 \rangle (\mu_B H_0)^2 \tau^*}{1+X^2}, \quad g_{\text{obs}} = \langle g \rangle \left[1 + \frac{B\langle \Delta g^2 \rangle (\mu_B H_0)^2 \tau^{*2}}{1+X^2} \right],$$

and when $\omega_c\tau^* \gg 1$, the terms involving Δg_m are negligible compared to those involving Δg_0. The regime $\omega_c\tau^* \gg 1$ is more interesting in practice, and in the preceding paper the notation $\langle \Delta g_0^2 \rangle = \sigma_g^2$ is adopted.

We conclude that, for large scattering, $|X| \ll 1$, we have ordinary motional narrowing [the factors $(1+B)$ can be absorbed into τ_s and τ^*, which already contain many-body effects]. Many-body effects do not manifest themselves, and the observed spectrum is shown in Fig. 1. When $|X| \gg 1$, on the other hand, many-body effects qualitatively change the spectrum from that arising from the free-electron picture. For noninteracting electrons, motional narrowing ceases when $|\Delta g|\, \mu_B H_0 \tau \gtrsim 1$, and we should expect a spectrum reflecting the g-value distribution [Fig. 1(a)]. In fact, for $|X| \gg 1$ the spectrum consists of the free-electron continuum shifted in frequency by $-[B/(1+B)]\langle g \rangle \mu_B H_0$, and the collective CESR [see Fig. 1(b)]. The narrow line must be understood as a collective oscillation; its width vanishes in the no-scattering ($\tau \to \infty$) limit.

We shall demonstrate elsewhere that the collective CESR always exists for $B \neq 0$. When $|\Delta g|$

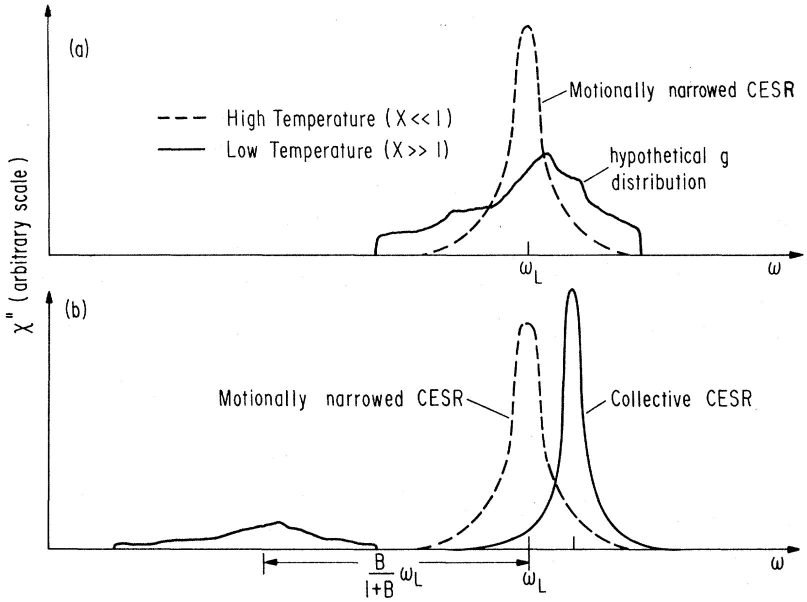

FIG. 1. Schematic absorption spectra for (a) free electrons and (b) interacting electrons, drawn to a common scale. $\omega_L = \langle g\rangle \mu_B H_0$. For free electrons X should be understood as $\omega\tau$. For interacting electrons $X = B\omega\tau/(1+B)$; motional narrowing requires the stronger condition $\omega\tau \ll 1$.

$\times \mu_B H_0 \tau / |X| = (|\Delta g|/g)(1+B)/|B| \ll 1$, almost all of the oscillator strength (area under the absorption line) is in the collective CESR; this is the circumstance for which (4) and (5) are valid. If we imagine reducing $|B|$, we should find that the collective mode approaches the edge of the continuum, which shifts to meet it; at the same time, the oscillator strength of the collective CESR would decrease, and the integrated oscillator strength of the continuum would increase. In the limit $|B| \to 0$, the collective CESR can no longer be excited; this limit cannot be studied with (4) because (4) is only valid if $|B|/(1+B) \gg |\Delta g_0|/g$. That the collective CESR always lies outside the continuum may have an important experimental consequence: Let

$$\min g_0(k_z) = \langle g\rangle - \Delta g_-$$

$$\max g_0(k_z) = \langle g\rangle + \Delta g_+.$$

Then the collective CESR occurs at frequency ω such that

$$\omega/\mu_B H_0 > \langle g\rangle/(1+B) + \Delta g_+ \quad (B>0),$$

$$\omega/\mu_B H_0 < \langle g\rangle/(1+B) - \Delta g_- \quad (B<0).$$

This is shifted from the position of the motionally narrowed CESR, observed at high temperature, by

$$|\omega/\mu_B H_0 - \langle g\rangle| > \Delta g_\pm - B\langle g\rangle/(1+B) \sim \Delta g_\pm$$

when $|B|/(1+B) < \Delta g_\pm/\langle g\rangle$. Therefore, when the scattering is sufficiently weak, the collective CESR may be shifted substantially from the position of high-temperature CESR.

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[1]P. M. Platzman and P. A. Wolff, Phys. Rev. Lett. 18, 280 (1967); S. Schultz and G. Dunifer, Phys. Rev. Lett. 18, 283 (1967).

[2]D. Landau, Zh. Eksp. Teor. Fiz. 30, 1058 (1956) [Sov. Phys. JETP 3, 920 (1957)]; V. P. Silin, Zh. Eksp. Teor. Fiz. 33, 495 (1957) [Sov. Phys. JETP 6, 387 (1958)].

[3]The observed signal, for example, in transmission CESR is not given by $\chi'' = \mathrm{Im}\chi$, but there are standard corrections which are applied to the observed signal to make it comparable to χ''. G. L. Dunifer, thesis, University of California, San Diego (unpublished); S. Schultz, G. Dunifer, and D. Pinkel, to be published.

[4]Strictly, to take electron-phonon scattering into account, we should work with $\delta\tilde F = \int d\mathcal{E}\,\delta\tilde f$. For the present purpose, with $T \ll \Theta_D$, we achieve the same result by writing $\delta\tilde f = \delta\tilde F\,\delta(\mathcal{E} - \mathcal{E}_F)$.