SPIN-WAVE EXCITATION IN NONFERROMAGNETIC METALS

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Using the Fermi-liquid theory, we have calculated the wave-number-dependent rf spin susceptibility of an interacting electron gas. For large $\omega\tau$ the kernel exhibits a branch of singularities, spin waves, which show up as sidebands on the electron spin-resonance line in thin slabs of metals. The character of these spin waves and their influence on electron spin resonance is discussed in detail.

It is well known[1,2] that, in an interacting Fermi system immersed in a static magnetic field, there exists a branch of excitations (starting at the free-electron spin-resonance frequency at long wavelength) analogous to spin waves in ferromagnetic materials. These oscillations are collective periodic variations of the macroscopic magnetization vector.[3] We believe that such excitations are responsible for the spin-resonance sidebands whose observation in Na and K is described in the following Letter.[4] These experiments enable one to determine, in an essentially spectroscopic way, two of the Landau Fermi-liquid parameters for each of these materials.

Using the Fermi-liquid theory, we have calculated the wave-number-dependent rf spin susceptibility of an interacting electron gas. Orbital and spin-flip collisions have been included in a phenomenological but plausible way. In the long-wavelength limit ($k \to 0$) we have obtained results for an arbitrary exchange integral. To order k^2 the susceptibility depends on only the first two exchange parameters (S and P wave). If the orbital collision time τ is short, the susceptibility kernel is purely diffusive. For large $\omega\tau$ the kernel exhibits a branch of singularities which is the spin-wave spectrum. The character of this branch depends strongly on the propagation direction with respect to the magnetic field. For "attractive" exchange, the curvature is negative for propagation along the field and positive for propagation perpendicular to it.

Using the long-wavelength approximation for the susceptibility it is possible to approximately solve the electron spin-resonance (ESR) problem for a finite slab of metal.[5,6] The transmitted magnetic field shows a resonance at what may be called the noninteracting ESR. In addition, when $\omega\tau$ is large and there is "sufficient" exchange among the electrons, a series of sidebands appears. The sidebands correspond to the excitation of nonuniform modes of magnetization in the slab, with wave vectors $k = n\pi/L$. The splitting of the spin-wave sidebands from the central ESR line enables one to measure the first two Landau exchange parameters for the interacting electron gas.[7] The measurements of spin-wave sidebands described in the accompanying Letter are in excellent agreement with the theoretical model to be presented here.

For slowly varying external disturbances the transport properties of a Fermi liquid are completely described by the quasiparticle density matrix $\rho(p,x)$,[8]

$$\rho_{\alpha\alpha'}(p,x) \equiv n\delta_{\alpha\alpha'} + \vec{m}\cdot\vec{\sigma}_{\alpha\alpha'}. \tag{1}$$

In equilibrium at zero temperature this quasiparticle distribution function $n_0(p,x)$ is a Fermi distribution and the quasiparticle energy $E^0(p) = p^2/2m^*$.[9] In nonequilibrium situations the quasiparticle density matrix satisfies a transport equation similar to the Boltzmann equation. We are interested in the solution of the transport equation in the linear approximation.

The transport equation (for the magnetization vector $\vec{m}$) separates into three scalar uncoupled equations for the quantities $m^{\pm} = m_x \pm i m_y$ and m_z (z = magnetic field direction). Since the quantity m^+ is the only one which shows a resonance at the electron precession frequency, it is sufficient to consider it. Defining the quantity g as

$$m^+ = -\frac{\partial n_0}{\partial E^0(p)} g$$

the transport equation for g becomes[2]

$$\frac{\partial g}{\partial t} + \left[\vec{V}\cdot\nabla + \frac{e}{c}(\vec{V}\times\vec{H}_0)\cdot\frac{\partial}{\partial\vec{p}} + i\Omega_0\right][g + \delta\epsilon_2]$$
$$= -\frac{1}{\tau}\left[g - \int\frac{g\,d\Omega}{4\pi}\right] + \tfrac{1}{2}\gamma_0(\vec{V}\cdot\vec{\nabla} + i\Omega_0)h_+, \tag{2}$$

where h is the rf magnetic field ($h_x + ih_y$) and

$\gamma_0 \equiv ge\hbar/2mc$. The quantity

$$\delta\epsilon_2 = \frac{2}{(2\pi)^3}\int d^3p'\, \zeta(p,p')g(p')\delta(\mu - E^0(p'))$$

is the change in the quasiparticle energy due to a change in the distribution function. The correlation or interaction function $\zeta(p,p')$ is the basic phenomenological quantity characterizing the "magnetic" properties of the Fermi liquid. Since p and p' are fixed on the Fermi surface, ζ may be expanded as

$$\zeta(p,p') \equiv \sum_n [\zeta_n P_n(\cos\theta)].$$

For convenience we also define the set of dimensionless quantities[2]

$$B_n \equiv m^* p_{\mathrm{F}} \zeta_n / \pi^2(2n+1).$$

The frequency Ω_0 in Eq. (2) is a renormalized spin frequency ($\hbar\Omega_0$ = energy to reverse a single electron's spin) given by

$$\Omega_0 = \omega_s/(1+B_0) \equiv \gamma_0 H_0/(1+B_0),$$

where ω_s is the ESR frequency in the dc field H_0. The term in $1/\tau$ is a phenomenological or orbital relaxation time characteristic of the dc resistance measurements. The collision term is chosen so that the system relaxes to local equilibrium compatible with the local density of magnetization and quasiparticle number.

For disturbances of the form $e^{i(\vec{k}\cdot\vec{x}-\omega t)}$, Eq. (2) may be solved exactly to order k^2. The expansion parameter is essentially $kR \equiv (kv_{\mathrm{F}}/\omega_c^*)$, which is of order 0.1 in the experiments. Neglecting terms of order $(kR)^4$, we find

$$\chi(k,\omega) = -(m^*/m)\chi_{\mathrm{free}}\Omega_0/P(k,\omega), \tag{3}$$

where

$$\frac{1}{(2\pi)^3}\int m^+(p,k,\omega)d^3p \equiv h\chi(k,\omega)$$

and

$$P(k,\omega) = \omega - \omega_s + \tfrac{1}{3}k^2 v_{\mathrm{F}}^{*2}(1+B_0)(1+B_1)(\tilde{\omega}_s - \bar{\Omega}_0) \times \left\{\frac{\sin^2\Delta}{\bar{\omega}_c^{*2} - (\tilde{\omega}_s - \bar{\Omega}_0)^2} - \frac{\cos^2\Delta}{(\tilde{\omega}_s - \bar{\Omega}_0)^2}\right\} \equiv \omega - \omega_s + iD^* k^2. \tag{4}$$

The quantity χ_{free} is the long-wavelength static susceptibility of the noninteracting gas, $\bar{\omega}_c^* = (eH_0/m^*c)(1+B_1)$, $\bar{\Omega}_0 = \Omega_0(1+B_1)$, $v_{\mathrm{F}}^* = p_{\mathrm{F}}/m^*$, $\tilde{\omega} = \omega + i/\tau$, and Δ is the angle the wave vector k makes with the z axis.[10]

As B_0 and $B_1 \to 0$, the coefficient D^* becomes a real number and the susceptibility is a purely diffusive kernel with an anisotropic diffusion coefficient. In this limit our results are equivalent to those of Ref. 6 and, for small $\omega\tau$, to those of Dyson.[5] On the other hand, if $|(B_0 - B_1)\omega\tau/(1+B_0)| \gg 1$, D^* approaches a pure imaginary number and $\chi(k,\omega)$ exhibits a branch of singularities along the curve $P(k,\omega) = 0$ in ω, k space. These are the spin waves. If we allow $\bar{\omega}_c^* \to 0$, i.e., turn off the coupling of the dc field to the orbital motion of the quasiparticles, and let $B_1 \to 0$, D^* becomes isotropic and $\chi(k,\omega)$ reduces to the known long-wavelength susceptibility for an uncharged Fermi liquid like He^3.[2] In general, however, the spin-wave dispersion relation for a charged gas is highly anisotropic. In fact, the bracketed angular expression in Eq. (4) will generally change sign since $(\omega - \bar{\Omega}_0)^2$ is usually less than $\bar{\omega}_c^{*2}$.

The problem of transmission through a slab of metal can be approximately solved. The technique used in solving the slab problem is similar to that of Ref. 6 and our results reduce to those when the exchange parameters are set equal to zero. The macroscopic magnetization vector is assumed to obey a second order, diffusionlike equation with D^* [Eq. (4)] as the diffusion coefficient. The magnetization vector may then be expanded in normal modes with the boundary condition $\nabla M(z)|_{z=0,\,z=L} = 0$. The rf exciting field is treated as a delta function. In this approximation the transmitted magnetic field H_t is

$$H_t \sim (i/T_2\Gamma^2)W^{-1}\csc 2W, \tag{5}$$

where

$$4W^2 = -[(1-\omega_s/\omega)\omega T_2 + i]/T_2\Gamma^2 \tag{6}$$

and

$$T_2\Gamma^2 = \frac{v_{\mathrm{F}}^{*2}}{3L^2\omega_s^{\,2}}(\omega T_2)\left\{\frac{\sin^2\Delta}{\bar{\omega}_c^{*2} - (\tilde{\omega}_s - \bar{\Omega}_0)^2} - \frac{\cos^2\Delta}{(\tilde{\omega}_s - \bar{\Omega}_0)^2}\right\} \times (1+B_0)(1+B_1)(\tilde{\omega}_s - \bar{\Omega}_0). \tag{7}$$

A phenomenological spin relaxation time T_2 has been included in Eqs. (5)-(7) and the proportionality sign in Eq. (5) indicates that a complex number, which is approximately magnetic-field and angle independent, has been omitted. In addition to the spin-resonance line near $W=0$, the transmitted field shows a series of sidebands for $W=n\pi$ ($n=\pm1, \pm2, \cdots$). In the infinite-$\omega\tau$ limit these peaks are the same peaks found by setting $k=n\pi/L$ in Eq. (4) and finding the zero of Eq. (4).

Figure 1 shows a series of transmission curves, including the central ESR line and the first spin-wave sideband, calculated from Eq. (5) with parameters typical of the alkali metals. Both theoretically[11] and experimentally,[7] B_0 is expected to be about -0.2, with B_1 smaller (we used $B_1=0.01$). Other parameters had the values $v_F^*=10^8$ cm/sec, $L=0.32$ mm, and $\omega=10^{11}$ cps. In this figure the sideband splitting is typically 3 to 5 G. For $|B_0-B_1|\simeq 0.2$, the condition $|(B_0-B_1)\omega\tau/(1+B_0)|>1$ is satisfied for $\omega\tau>10$. The sideband has been symmetrized, and is labeled with the angle Δ. In Fig. 2 we plot the same curve as in Fig. 1 for $\Delta=90°$ and $\omega\tau=20$, 60, and 100. As $\omega\tau$ is increased the symmetrized sideband does not move relative to the ESR line, though it narrows.

We briefly summarize some of the important general features of these curves. (1) For $|B_1|<|B_0|$ and $B_0<0$ ("attractive" exchange), the sidebands are split to the low-field, high-frequency side of the ESR for propagation across the field. (2) As the field is swung around,

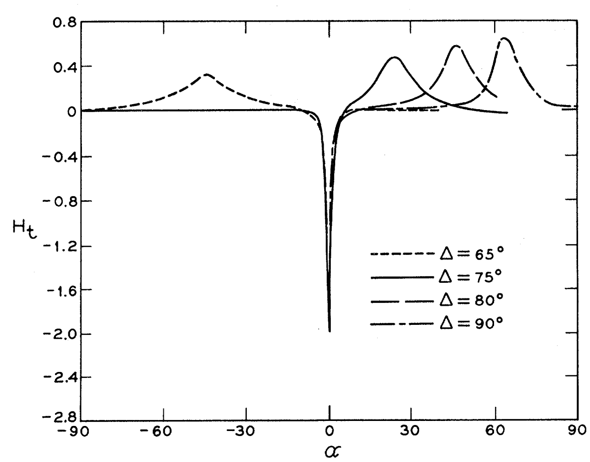

FIG. 1. Plot of the transmitted magnetic field as a function of $\alpha \equiv (1-\omega_s/\omega)\omega T_2$ in the neighborhood of the first spin-wave sideband. We used $B_0=-0.2$ and $B_1=0.01$. Other parameters in the figure are $v_F^{*2}/3L^2\omega^2=3\times10^{-4}$, $\omega\tau=60.0$, and $\omega T_2=6\times10^4$. The angle Δ is the angle the dc magnetic field makes with the normal to the surface of the specimen.

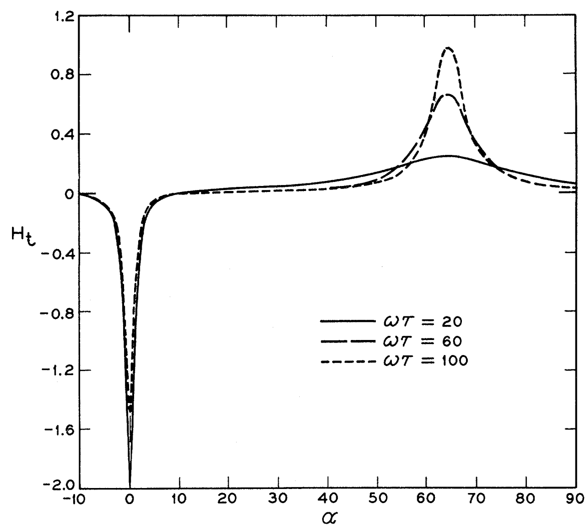

FIG. 2. Plot of the transmitted magnetic field as in Fig. 1. The angle Δ was fixed at 90° and $\omega\tau$ was varied from 20 to 100.

all sidebands coalesce at the ESR for some angle and then reappear on the high-field side of ESR. (3) The angular distribution of the splitting of each sideband goes as $1-A\cos^2\Delta$ where A is a function of both B_0 and B_1. (4) The magnitude of the splitting is geometry dependent and for small splitting goes like $1/L^2$. The actual comparison of these results with the experimental data in Na and K appears in the following Letter.[4]

The calculations described in this Letter were motivated by Walsh, Rupp, and Schmidt's observation[12] of a small splitting (~0.5 G) of the ESR line in K. Our subsequent work has shown that this splitting is not due to spin waves and it remains a mystery. We wish to thank Professor S. Schultz and Dr. W. M. Walsh for encouragement and many fruitful discussions of the ESR problem in metals. Discussions with Professor D. R. Fredkin have also been very helpful.

[1]V. P. Silin, Zh. Eksperim. i Teor. Fiz. 33, 1227 (1957) [translation: Soviet Phys.—JETP 6, 945 (1958)].

[2]V. P. Silin, Zh. Eksperim. i Teor. Fiz. 35, 1243 (1958) [translation: Soviet Phys.—JETP 8, 870 (1959)]; G. Lill, Phys. Letters 8, 171 (1964); D. R. Fredkin, to be published.

[3]The spin-wave modes are excitations above the ground state and do not in any way correspond to spin-density waves [see A. W. Overhauser, Phys. Rev. 128, 1437 (1962)] which are modifications of the ground state.

[4]Sheldon Schultz and Gerald Dunifer, Phys. Rev. Letters 18, 283 (1967).

[5]F. J. Dyson, Phys. Rev. 98, 349 (1955).

[6]M. Lampe and P. M. Platzman, Phys. Rev. 150, 340 (1966).

[7]In principle, an absolute measurement of the susceptibility determines the value of the isotropic exchange parameter. In practice, this measurement [R. Schumacher, T. Carver, and C. P. Slichter, Phys. Rev. 95, 1089 (1954)] is extremely difficult to perform and gives results which necessarily have poor accuracy.

[8]P. Nozières, Theory of Interacting Fermi Systems (W. A. Benjamin, Inc., New York, 1964).

[9]The mass m^* is the effective quasiparticle mass which includes band-structure effects, phonons, and electron-electron effects. It can be shown {see M. Ya. Azbel', Zh. Eksperim. i Teor. Fiz. 34, 766 (1958) [translation: Soviet Phys.—JETP 7, 527 (1958)]} that, to a high degree of accuracy, the mass measured in Azbel'-Kaner cyclotron resonance {M. Ya. Azbel' and E. A. Kaner, Zh. Eksperim. i Teor. Fiz. 32, 896 (1956) [translation: Soviet Phys.—JETP 5, 730 (1957)]} is, in fact, the quasiparticle mass.

[10]The long-wavelength susceptibility, Eq. (4), depends only on B_0 and B_1. The terms in k^4 in $\chi(k, \omega)$ will involve B_2 as well. Experiments involving small wavelengths should give information about the higher moments of the Fermi-liquid function.

[11]T. M. Rice, Ann. Phys. (N.Y.) 30, 100 (1965).

[12]W. M. Walsh, Jr., L. W. Rupp, Jr., and P. H. Schmidt, Phys. Rev. 142, 414 (1966).

OBSERVATION OF SPIN WAVES IN SODIUM AND POTASSIUM*

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We report the first observation of spin waves in sodium and potassium at low temperatures. Utilizing the theory of Platzman and Wolff, we are able to deduce the first two Legendre coefficients of the Landau correlation function for a Fermi liquid.

In the course of making precise g-value and linewidth measurements of the conduction-electron spin resonance (CESR) in sodium foils at 1.4°K, utilizing the selective transmission technique, we found significant departures from the predictions of previous theories.[1] Further investigation showed that there was extra structure in the vicinity of the CESR. It is the purpose of this Letter to show that the extra structure that we have observed is proof of the existence of spin waves in the metal which are excited by the applied rf field. We have verified the detailed theory of Platzman and Wolff[2] (henceforth called P-W), and furthermore, we are able to extract from the data the relevant Landau Fermi-liquid theory parameters B_0 and B_1. We wish to point out that these spin-wave data appear as a primary effect in themselves and vanish in the limit of zero electron correlation. We believe that this direct observability, plus the fact that we are doing a resonance experiment, will allow the measurement of the relevant parameters with a significant precision.[3] Since we have seen the spin-wave signals in both sodium and potassium, we feel the technique is generally applicable to all the alkali metals and in principle to all metals.[4]

The basic experimental arrangement is similar to that which we have used to discover the CESR in several new metals.[5] The major change has been the use of solid dielectric cavities, which help to insure the parallelism of the surfaces of the ductile alkali samples while also protecting them from any deteriorating atmosphere. In brief, the technique consists of placing the sample between a pair of X-band cavities tuned to the same frequency in the presence of a uniform dc field H_0. Microwave power is coupled into one cavity (transmitter), and a sensitive superheterodyne receiver is connected to the second cavity (receiver). If the leakage between the cavities has been sufficiently reduced (by experimental care), any power that appears in the receiver cavity has been transmitted through the sample.[6] In the vicinity of a magnetic resonance, electrons absorb power within a skin depth on the transmitter side of the sample and carry this information via their nonequilibrium transverse magnetization to the far side of the sample where they radiate power.[7] As is shown in P-W, at selected values of the applied dc field near the main CESR (for a fixed frequency), other modes of the spin system are excited depending on