RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 32, NUMBER 3 1 AUGUST 1985

Proposed determination of many-body effects in heavy-fermion systems by conduction-electron-spin resonance

Kevin S. Bedell and David E. Meltzer
Department of Physics, State University of New York at Stony Brook, Stony Brook, New York 11794
(Received 25 March 1985)

We propose that it might be possible to determine the contribution of many-body interactions to the large effective mass in "heavy-fermion" materials, e.g., UPt_3, by the method of conduction-electron-spin resonance (CESR). A microwave transmission observation of CESR may show a resonance pattern which, based on already measured parameters, would clearly distinguish among suggested models.

Recent discussions of the so-called "heavy-fermion" superconductors have centered on the possibility of triplet pairing in the superconducting state.[1] However, the normal-state behavior raises very interesting questions in itself. In particular, it is not at all clear what is the origin of the enormous effective masses ($m^* \approx 10^2$–10^3) which characterize these materials.

One possibility is that the large effective masses arise essentially from band-structure effects, which is to say that the interplay of crystal geometry and electron hybridization accounts for the very high density of electronic energy states.[2] It is also possible that the high density of states is due in large part to strong many-body effects, and that unusually powerful correlations among the conduction electrons lead to the observed m^*'s.[3,4] Of course, some combination of the two effects may be at work.

The hypothesis of significant many-body effects arises from a number of observations, including a large "residual" (temperature-independent) component of the magnetic susceptibility, and a $T^3 \ln T$ component of the specific heat (observed in at least one of the materials, UPt_3), both characteristic of Fermi-liquid behavior. In addition, recent theoretical work has indicated that the "almost localized" limit of both the induced-interaction model[4] and Gutzwiller[3] approaches to Fermi-liquid theory lead to consistent explanations of the observed effects.

Within the Fermi-liquid picture, the strong many-body effects are signified by relatively large (absolute) values for the Fermi-liquid parameters, F_l^s and F_l^a, which characterize the effective quasiparticle interaction and are equal to zero in the absence of many-body effects (i.e., in a degenerate free Fermi gas). In ordinary metals, these parameters are expected to be quite small, with typical values $F_0^a \approx -0.2$, $F_0^s \approx -0.5$, and $F_1^s \approx 0.1$.[5] However, the hypothesis of large many-body effects in the heavy-fermion materials leads to explicit predictions of substantially enhanced values for the Landau parameters. For instance, if the $T^3 \ln T$ term in the specific heat for UPt_3 is ascribed to Fermi-liquid effects, a value of $F_0^a = -0.78$ is found.[6] (This is based on the work of Pethick and Carneiro,[7] who pointed out errors in previous calculations of this term in ^{3}He.) Similarly the Gutzwiller model leads to $F_0^a = -0.75$, $F_1^a = -0.75$,[3] while the induced interaction model predicts yet more strongly enhanced values ($F_0^a \approx -0.9$, $F_1^a \approx 30$) although the relative size of the band structure and other effects introduces a significant uncertainty.[4] In general, one has the relation $m^* = m_\lambda(1+F_1^s/3)$,[8] where the Landau parameter F_1^s characterizes the many-body effects, and in which the so-called "dynamical" mass m_λ incorporates the various other effects such as lattice structure, phonons, etc. If the many-body effects are indeed small, one expects virtually the entire m^* to be due to m_λ, and that the Landau parameters will be little changed from those seen in ordinary metals.

It is thus of very considerable interest to make an experimental determination of the Landau parameters in the heavy-fermion materials in some direct fashion, without making recourse to strongly model-dependent assumptions. At least one such method is potentially available—a microwave transmission observation of conduction-electron-spin resonance—and it is the purpose of this paper to show how such an experiment might greatly clarify the issues discussed above.

It was first shown by Silin[9] that it would be possible for wavelike collective excitations of the transverse magnetization to propagate in an interacting Fermi liquid immersed in a static magnetic field. These were dubbed "spin waves" in analogy to the excitations in ferromagnetic systems. Later, Platzman and Wolff[10] included the effects of collisions and calculated, in the long-wavelength limit, an expression for the rf spin susceptibility in an interacting electron gas. (In this limit, only the Landau parameters F_0^a and F_1^a appear.) They used this to solve for the transmitted magnetic field in an electron-spin resonance experiment in a finite slab of metal. Dyson[11] had previously derived the shape of the resonance line in the noninteracting regime for transmission through a metallic slab.

In this experiment, a thin metallic slab separates two tuned microwave cavities. Microwave energy is coupled into one cavity, and is absorbed by the conduction electrons in the slab in the form of an rf magnetization. The nonequilibrium motion of the electrons transports part of the energy to the other side of the slab, where it is reemitted into the second cavity, and detected. In practice, the sample is studied at constant applied frequency ω, and the external field H_0 is swept in the region near $\omega_s = g\mu_B H_0/\hbar$, the electron resonant frequency.

The transverse spin susceptibility is of the form $\chi^+ \sim \chi_0\omega_s/(\omega-\omega_s+iD^*k^2)$, where D^* is a complex diffusion constant and k a wave number. The result of Platzman and Wolff is as follows:

$$D^* = -i\frac{v_F^2}{3}(1+F_0^a)(1+F_1^a/3)\left[\tilde{\omega}_s - \omega_s\frac{(1+F_1^a/3)}{(1+F_0^a)}\right]$$
$$\times\left[\frac{\sin^2\Delta}{\omega_c^2(1+F_1^a/3)^2 - \{\tilde{\omega}_s - \omega_s[(1+F_1^a/3)/(1+F_0^a)]\}^2} - \frac{\cos^2\Delta}{\{\tilde{\omega}_s - \omega_s[(1+F_1^a/3)/(1+F_0^a)]\}^2}\right] ,$$

which leads to

$$H_t \sim \frac{i}{T_2\Gamma^2}\frac{\csc(2W)}{W} ,$$

where

$$4W^2 = -(\alpha+i)/T_2\Gamma^2 ,$$
$$T_2\Gamma^2 = \frac{T_2 v_F^2}{3L^2}(1+F_0^a)(1+F_1^a/3)\left[\tilde{\omega}_s - \omega_s\frac{(1+F_1^a/3)}{(1+F_0^a)}\right]$$
$$\times\left[\frac{\sin^2\Delta}{\omega_c^2(1+F_1^a/3)^2 - \{\tilde{\omega}_s - \omega_s[(1+F_1^a/3)/(1+F_0^a)]\}^2} - \frac{\cos^2\Delta}{\{\tilde{\omega}_s - \omega_s[(1+F_1^a/3)/(1+F_0^a)]\}^2}\right] , \quad (1)$$

where T_2 is the spin-spin relaxation time, $\alpha = (\omega - \omega_s)T_2$, L is the thickness of the slab, $\omega_c = eH_0/m^*c$, $\omega_s = g\mu_B H_0/\hbar$ (where H_0 is the static external field), Δ is the angle between the wave vector **k** and the z axis, $\tilde{\omega}_s = \omega_s + i(1+F_1^a/3)/\tau$, where τ is the relaxation time for the momentum. H_t is the transmitted rf magnetic field; a complex constant which is approximately magnetic field and angle independent is omitted, which is why the "$\sim$" is used. [Note that in Platzman and Wolff's original expression for $\tilde{\omega}_s$, the $(1+F_1^a/3)$ is missing; see Ref. 15.]

In the case where there are no interactions ($F_0^a = F_1^a = 0$), or when the product $\omega\tau$ is very small, this result agrees with Dyson's and only a diffusional broadening (with a distinct shape) of the electron-spin resonance line is obtained. However, in the interacting case, and with sufficiently large $\omega\tau$, a series of sidebands of the central resonance appears which corresponds to the spin-wave excitations, with wave numbers $k = n\pi/L$.

These spin waves were actually observed for the first time in alkali metals by Schultz and Dunifer,[12] in good agreement with the predictions of Platzman and Wolff. Additional experiments with improved sensitivity have been made.[13,14]

The particularly interesting aspects of these transmission CESR experiments are, first, that they provide an unambiguous signature of the many-body interactions in the electron Fermi liquid, and second, that they focus very specifically on the conduction electrons themselves. The only way a signal can propagate from one side of the metal slab to the other—assuming experimental care in minimizing leakage—is for the electrons to carry it in the form of their transverse magnetization as they move through the metal. Thus, it is the interactions in the electron system itself which are being probed, and complications introduced by local moments are not a factor. This provides a very nice separation of the effects due to collective behavior in the Fermi liquid, and those due to effects restricted to the local lattice sites.

We proceed immediately to our main point: It may be possible to make transmission CESR observations on some of the heavy-fermion materials—in particular, on UPt_3—which could clearly distinguish whether or not many-body effects are the predominant cause of the large effective masses.

First we note that the relaxation time τ in the above expression is that associated with the resistivity, as discussed by Wilson and Fredkin.[15] Then we may make an estimate of this τ from the expression $\tau = m^*/ne^2\rho$, where ρ is the resistivity. This m^* is in fact the full m^* associated with specific-heat measurements, regardless of the relative proportions of band-structure and many-body effects which compose it.[16] Using the value proposed by Chen *et al.*[17] of $k_F = 1.08\times10^8$ cm^{-1}, which was derived by assuming a spherical Fermi surface, and the value for the resistivity near 1 K of 1 $\mu\Omega$ cm,[18] we obtain $\tau \approx 2\times10^{-11}$ sec. (The resistivity of UPt_3 is unusually low relative to the other "heavy-fermion" materials, which makes it particularly attractive for this experiment.) This value for k_F also leads to an effective mass m^*/m of 187, and a Fermi velocity $v_F = 6.7\times10^5$ cm/sec.

The Platzman and Wolff result shows that, when $\omega\tau$ is sufficiently small, D^* is purely real, and the Dyson result is recovered as stated above. However, when the condition

$$\omega_s\tau|(F_0^a - F_1^a/3)/(1+F_0^a)(1+F_1^a/3)| >> 1$$

is satisfied (referred to by Platzman and Wolff as "sufficient exchange"), D^* will become purely imaginary and there will be a branch of singularities in the susceptibility which corresponds to the spin waves. In the expression for H_t, these occur at wave number values of $k = n\pi/L$.

If many-body effects are relatively small, one expects the Landau parameters to be similar to those in ordinary metals, and so we will use the values measured for sodium, $F_0^a = -0.21$, $F_1^a = -0.03$.[14] (The experiments indicate that the values for $l > 1$ are very small.) The values given by the induced interaction model (if one assumes $m^*/m_\lambda \sim 20$) are $F_0^a = -0.9$, $F_1^a = 30$. To find a value for the parameter F_1^a which will correspond to the "experimental" assignment of $F_0^a = -0.78$, we assume (as in Refs. 3 and 4) that we are in the "almost localized" regime. In this case, the Landau scattering amplitudes

$$\{A_l^{s,a} = F_l^{s,a}[1 + F_l^{s,a}/(2l+1)]^{-1}\}$$

quickly converge to the values $A_0^s \to 1$, $A_1^s \to 3$, and the forward scattering sum rule $\sum_l (A_l^a + A_l^s) = 0$ gives a value $F_1^a = -0.4$. (Here, we assume that the Landau parameters for $l > 1$ are small, which is not expected to be as good an approximation as in the case of ordinary metals.)

In the case of an applied field of 3000 G ($\omega_s = 5.3\times10^{10}$ sec^{-1}), the "sufficient exchange" parameter will have the following values: "ordinary" metal 0.27; "experimental" values 3.6; induced interaction model 10.5. These widely separated values result in predictions for the observed resonance pattern which are very different in appearance from each other, and which—if a resonance can be observed at all—should allow for a clear experimental determination to be made of the actual contribution of many-body interactions.

In Fig. 1 we display plots of the transmitted magnetic field H_t determined from Eq. (1) above, for the three different sets of values for the Landau parameters. The other parameters used in the plots are $L = 0.001$ cm and $T_2 = 10^{-7}$ sec.

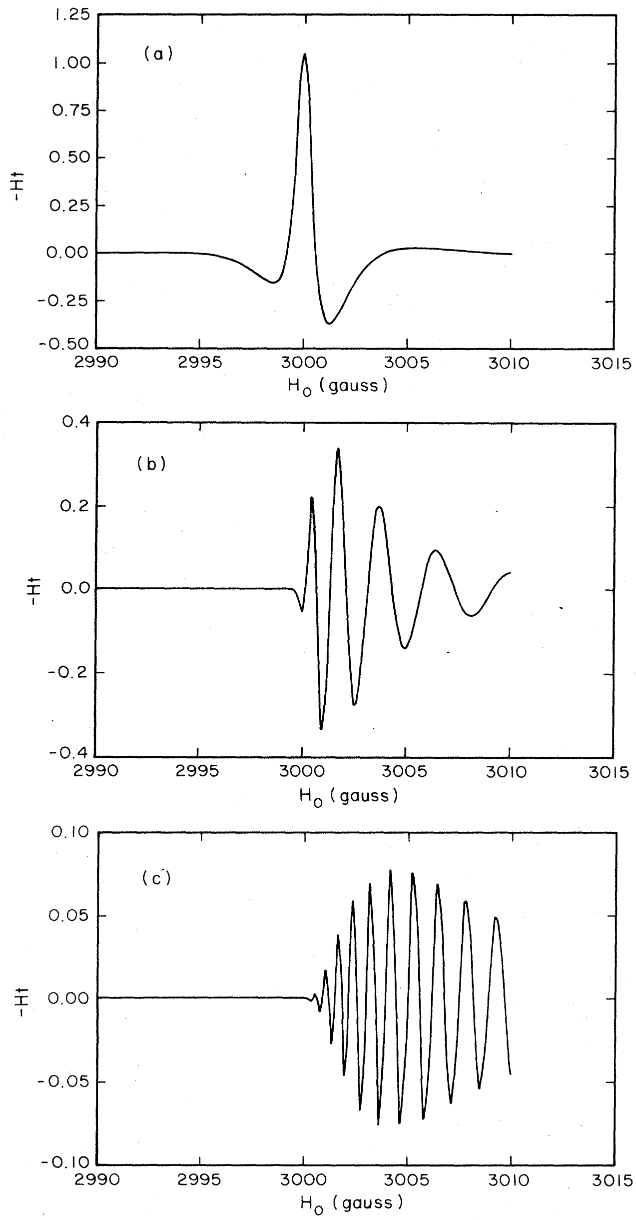

FIG. 1. Plots of the transmitted magnetic field H_t assuming a value of $T_2 = 10^{-7}$ sec. Other parameters used are $m^* = 187$, $v_F = 6.7\times10^5$ cm/sec, $\omega = 5.3\times10^{10}$ sec^{-1}, $L = 0.001$, $\tau = 2\times10^{-11}$ sec. (The results are essentially independent of Δ.) (a) $F_0^a = -0.21$, $F_1^a = -0.03$; (b) $F_0^a = -0.78$, $F_1^a = -0.4$; (c) $F_0^a = -0.9$, $F_1^a = 30$.

Figure 1(a) shows the results for the "ordinary metal" parameters, and has the appearance of a somewhat distorted "Dysonian" line shape, not very different from CESR results where spin waves are not a factor. In Fig. 1(b) spin-wave "satellites" appear quite prominantly on the high-field side of the CESR resonance frequency. Figure 1(c), using the induced-interaction parameter values, shows an even more distinct spin-wave pattern, with considerably narrowed widths for the wave peaks.

It is important to point out that, even if the value of τ differs substantially from the one we have assumed, the resonance patterns which would result are not only quite distinguishable from each other, but *also* from those in Fig. 1, *even though* the "sufficient exchange" parameters may match. That is, it turns out that the figure for ordinary metal parameters, with $\tau \sim 10^{-10}$ sec, is not at all similar to that for the "experimental" parameters with $\tau \sim 10^{-11}$ sec, although the sufficient exchange parameters would then be about the same. The parameter value does not in itself sufficiently specify the pattern shape to allow the patterns to be confused.

These resonance patterns are essentially independent of Δ, the angle between the static field and the propagation direction of the spin waves. This sharply differs from ordinary metals, and is due to the very large m^* which allows one to set $\omega_c \approx 0$ in the $\sin^2\Delta$ term in Eq. (1). This is remarkable in itself, and may help considerably in picking out the CESR signal from the experimental background, which generally varies with angle.

The value for the thickness L, although quite small, was chosen because larger values produce a very great reduction in transmitted field. The very small Fermi velocity of the "heavy" electrons imposes this condition, since large distances cannot be traversed before the electrons, due to spin-flip collisions, lose their spin "memory" of the magnetization signal. (Observations have been made with other materials of approximately this thickness.[19])

The parameter T_2 introduces the most uncertainty. It is not possible to specify its value in advance of the experiment, even to within an order of magnitude. The theory of CESR relaxation times is quite complicated, and not particularly reliable (see, for example, Ref. 20). Although a T_2 of $\sim 10^{-7}$ sec is typical of the alkali metals at temperatures ~ 1 K, it is not at all obvious, and is perhaps unlikely, that this will correspond to that of UPt_3. It is not possible, in advance, to untangle the various competing processes which affect T_2. For instance, one might expect greater spin-orbit coupling to result in increased scattering from impurities, thus reducing T_2. However, as Wilson and Fredkin point out, a greater degree of "correlation" among electrons may make it substantially harder for an impurity to flip a given spin, since it must in some sense drag the correlated electrons along with it. Thus, one expects an enhancement of the T_2 by a factor of the order of $(1/1+F_0^a)$, which could be substantial.

However, despite this uncertainty, it should still be possible to distinguish between the strongly and weakly interacting regimes. Even if the value of T_2 is changed by orders of magnitude, clearly different resonance patterns still result, which moreover could not possibly be confused with those corresponding to other values of T_2. For instance, in Fig. 2, we present plots for $T_2 = 10^{-8}$ sec for (a) the "ordi-

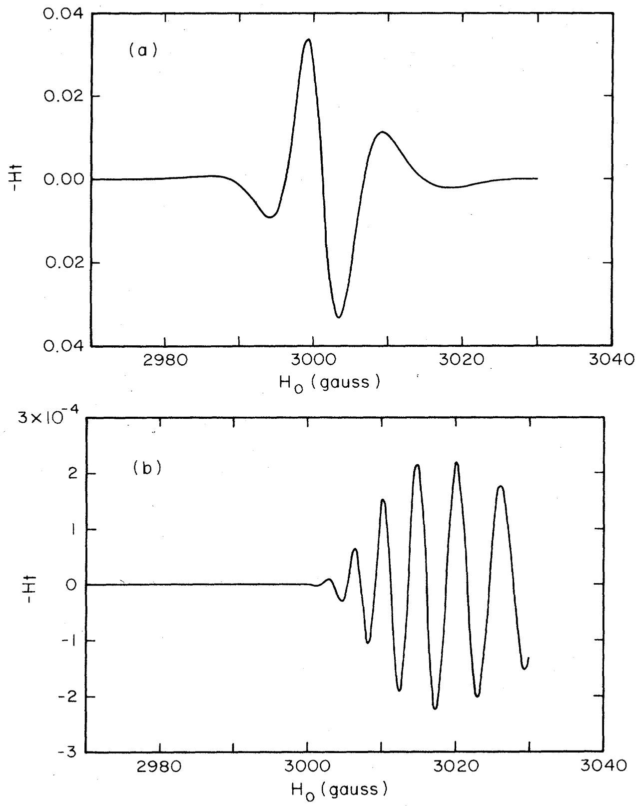

FIG. 2. Plots of the transitted magnetic field H_t assuming a value of $T_2 = 10^{-8}$ sec. (a) $F_0^a = -0.21$, $F_1^a = -0.03$; (b) $F_0^a = -0.78$, $F_1^a = -0.4$.

nary metal" parameters, and (b) for the "experimental" parameters. It remains true, though, that as in any ESR experiment, too small a value of T_2 will remove any possibility of observing a signal.

Recent speculations about the "effective" magnetic moment of the conduction electrons[21] could be tested by observing the deviation of the observed resonance from the CESR resonant frequency. In general, a different μ_{eff} simply shifts the external field at which the spin waves are observed, without very substantial changes in the pattern shapes. It would be particularly useful to have CESR observations regarding this issue, since the transmission experiment would directly test the effective moment characteristic of the electrons in their role as conductors.

Although the Platzman and Wolff expressions are derived for the long-wavelength regime, and the small L we have assumed corresponds to reduced wavelengths, this does not appear to be a significant problem. Dunifer, Pinkel, and Schultz[13] point out that, for $\Delta = 0$, the condition for Eq (1) to be reliable can be determined from Wilson and Fredkin's work to be $k v_F/\omega_s << |F_0^a/(1+F_0^a)|$, which is very well satisfied for the above parameters. As the angle Δ varies from 0, this condition may become significantly more stringent, but the observed spin-wave patterns show deviations from the Platzman and Wolff expressions mainly in the precise locations of the higher-order spin waves. The basic shapes of the patterns are not likely to differ substantially from those shown here, at least for modes corresponding to n's which are not too high.

The preparation of a 10-μm sample appears to be well within the realm of available technology.[22]

It should be pointed out that, while the CESR experiment is directly sensitive only to the many-body interactions reflected in the parameters F_0^a and F_1^a, there are good grounds for inferring from this the order of magnitude of F_1^s, and therefore the approximate value of m^*/m_λ. The analogy with ^{3}He, where the parameters rapidly decrease in magnitude with increasing l, and the forward-scattering sum rule strongly suggest that enhanced (negative) values of $(A_0^a + A_1^a)$ would be accompanied by substantial (positive) values for F_1^s. Indeed, all the models with many-body enhancements (paramagnon, induced interaction, Gutzwiller) reflect this, while experience with ordinary metals, and models without many-body effects[2] likewise suggest the self-consistency of small magnitudes for this set of parameters. Nonetheless, conclusions regarding the effective mass are ultimately indirect and model dependent.

We have benefited from conversations with L. de Long, G. Dunifer, J. Jaffe, S. Schultz, and G. Stewart. This work was supported in part by the U.S. Department of Energy through Contract No. DE-AC02-76ER13001.

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