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Fermi-liquid renormalization in the superconducting state of UBe_{13}

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ABSTRACT

The specific heat in the mixed phase of superconducting UBe_{13} is observed to vary as the square of the applied magnetic field. This is shown to imply a Fermi-liquid-type suppression of the exchange enhancement as the superconducting gap opens, similar to that seen in ^3He. Consistent with this effect is the observed non-monotonic temperature dependence of the Maki parameter κ_2 which is related to the Pauli susceptibility.

Heavy-fermion (HF) intermetallic compounds continue to provide a rich source of new phenomena for the study of strong interactions in electronic materials. The enormous low-temperature metallic entropy in these systems arises from the magnetic degrees of freedom of the 4f or 5f valence electrons of the Ce and U-based compounds respectively (Varma 1985). Because magnetism is the source of this most obvious feature of the low-temperature behaviour, it is interesting that superconductivity is observed in several compounds; usually intrinsic magnetism and superconductivity do not occur together. This apparent contradiction has been addressed theoretically by invoking magnetic fluctuations for mediating superconducting pairing (Schmitt-Rink *et al.* 1986) in contrast with usual superconductors where pairing is mediated by lattice vibrations (phonons). A natural outcome of this theory of superconductivity is unconventional (d-wave) symmetry of the pairing state (Miyake *et al.* 1986, Heffner and Norman 1996). Similar ideas have been proposed for high-T_c cuprate superconductors, and the possibility of d-wave pairing in these materials is a subject of intense debate.

Despite the recognized importance of magnetic fluctuations for superconducting pairing in HF systems, there is little direct evidence for their behaviour in the superconducting state. Here we present a new approach to this problem using specific heat against field measurements, $C(H)$. We show that in UBe_{13}, among the heaviest of HF superconductors (Ott *et al.* 1984), $C(H) \propto H^2$ in the mixed state. This is, to our knowledge, unique behaviour among superconductors, but further the *temperature dependence* of the effect implies a Fermi-liquid renormalization of the exchange enhancement similar to that observed in the B phase of ^3He. We also observe an anomalous non-monotonic temperature dependence of the Maki parameter κ_2 consistent with strong Fermi-liquid renormalization.

The data presented here are on a high-quality single crystal (SX) of UBe_{13}, grown from an Al flux. The recognition of this particular crystal's quality came in the course of scanning several different samples using the width and height of the specific heat jump at the superconducting–normal transition as the primary measure of quality. As shown in figure 1, the SX sample displays a much larger normalized jump at T_c, $\Delta C/\gamma T_c = 2.6$, than found in an arc-melted polycrystalline (PX) sample where $\Delta C/\gamma T_c = 1.9$, a value typical of previously studied samples, both SX and PX (De Visser *et al.* 1992). Similarly, as shown in the inset of figure 1, the width of the jump in $C(H_{c2})$ is about a factor of 50% smaller in the SX than in the PX crystal, another indication of better quality. The thermal expansion coefficient (Kleiman *et al.* 1990) and nonlinear susceptibility (Ramirez *et al.* 1994) have also been measured in this crystal. The present measurements were made using a semiadiabatic technique, with a field scan method that has been described previously (Ramirez *et al.* 1995).

In figure 2 are shown $C(H)$ data for the SX specimen throughout a field range encompassing the upper critical field H_{c2}. (All data have been corrected for the Be nuclear specific heat contribution of the form valid for $k_B T \gg g\mu_N H$: $C(H,T) = 13 \times \frac{5}{4} R(g\mu_N H/k_B T)^2$, where μ_N is the nuclear magneton and R the gas constant.) Of most importance is the monotonic *superlinear* H-dependence of $C(H)$ for $H < H_{c2}$. Data taken for the PX sample (not shown), by contrast,

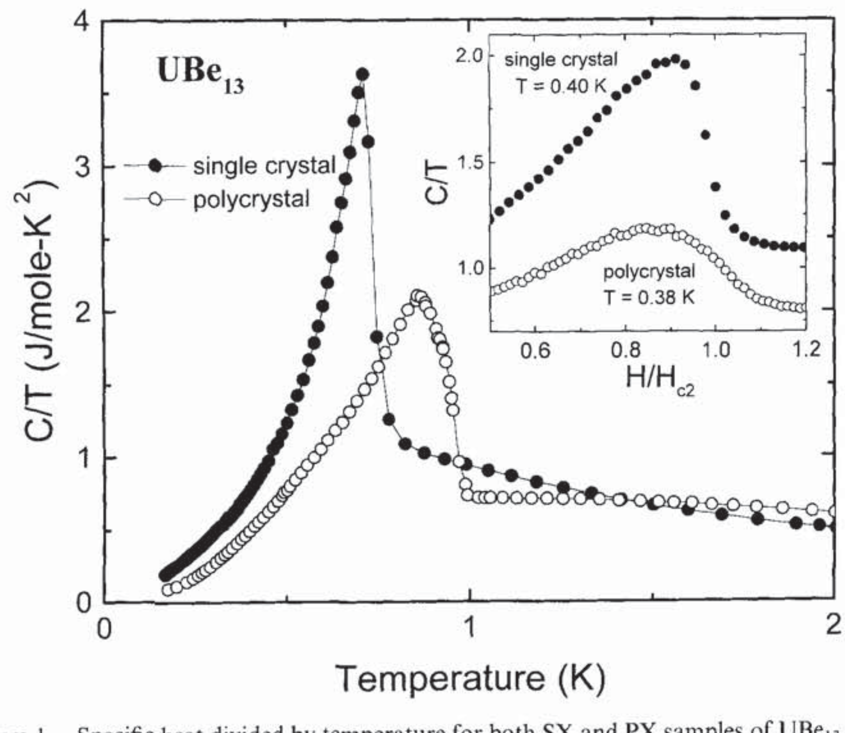

Figure 1. Specific heat divided by temperature for both SX and PX samples of UBe_{13} in zero field. The inset shows data taken against field at fixed temperatures for both samples.

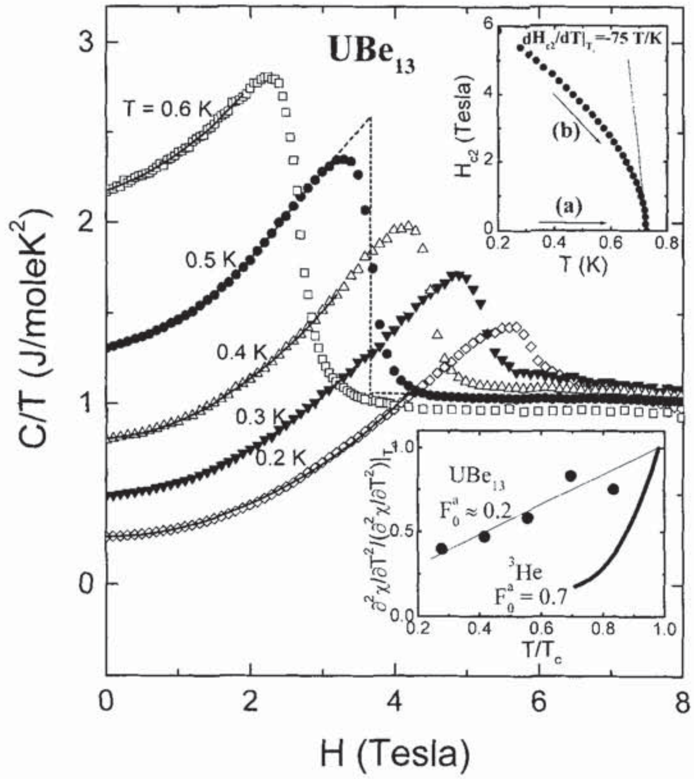

Figure 2. Field dependence of the specific heat divided by temperature, C/T, at several temperatures. The solid lines are fits, incorporating constant, linear and quadratic H-dependent terms over the range $0 \leqslant H \leqslant 0.7H_{c2}$, as discussed in the text. The equal area construction used to determine $\Delta C(H)$ is shown for $T = 0.5$ K as a broken line. The upper inset shows the upper critical field temperature dependence and the initial slope determined over the range $0.99T_c < T < T_c$. The lower inset shows the normalized second derivative of the spin susceptibility in both the superconducting state of UBe_{13} (full circles and solid line to guide the eye) and in the B phase of ^3He (bold curve).

display a rounded peak superimposed on a monotonically rising $C(H)$ within the mixed state, similar to that observed earlier on a different sample (Ellman *et al.* 1991). Since this rounded peak is seen in PX samples with broad transitions and is absent in our SX sample with sharp transitions, we believe it is not intrinsic to pure UBe_{13} and that the observed SX behaviour is that of a homogeneous superconductor.

The superlinear $C(H)$ in UBe_{13} is, to our knowledge, unique among superconductors. For large-κ materials such as UBe_{13} ($\kappa \approx 100$) the behaviour of $C(H)$ should be dominated over most of the field range ($H_{c1} \ll H \ll H_{c2}$) by a term linear in applied field (Fetter and Hohenberg 1969),

$$C(H,T) - C(0,T) = \frac{\kappa_1(0)}{\kappa_3(0)}\,\gamma T\,\frac{B}{H_{c2}(0)}\,[1 + \mathrm{O}(t^2)], \tag{1}$$

where $\kappa_1(0)/\kappa_3(0) \approx 1$, B is the internal field, and $t = T/T_c$. Since H_{c1} is small (less than 0.1 T), $B = H$ to good accuracy over the field range of interest, and equation (1) yields $C(H) \propto H$, in contrast with our observations. Corrections to this expression near H_{c1}, owing to vortex interactions, yield a downward-curving $C(H)$ (Ramirez 1996).

It is, perhaps, not surprising that $C(H)$ does not conform to expectations for conventional (s-wave) superconductors given the large body of evidence for gaplessness in UBe_{13}. Recently, Volovik (1993) explored the consequence of line nodes on the low energy density of states of normal excitations in the presence of vortex lines. Instead of equation (1) as for conventional superconductors, one finds the form $C(H)/T \propto H^{1/2}$ for $H_{c1} \ll H \ll H_{c2}$, which is clearly not the dominant behaviour in UBe_{13}.

To understand the significance of the observed quadratic behaviour of $C(T,H)/T$, we note that the free energy quite generally contains a term $-\frac{1}{2}\chi(T)H^2$. Therefore, simple thermodynamics dictates a strongly temperature-dependent susceptibility since

$$\frac{1}{T}\frac{\partial^2 C}{\partial H^2} = \frac{\partial^2 \chi(T)}{\partial T^2}. \tag{2}$$

Of course, for common metals with weak exchange enhancement, the spin susceptibility is given by the normal state (Pauli) term χ_N which is orders of magnitude smaller than the shielding susceptibility. Since the latter dominates the temperature dependence of the long-wavelength magnetic response near T_c, it is usually impossible to observe effects due to a temperature-dependent χ_N. From equation (2) we see that the observed quadratic $C(H)/T$ implies that the analytic part of $\chi(T)$ contains terms T^n, where $n \geqslant 2$. As already mentioned, vortex contributions to $C(H)$ are either linear or sublinear in H. Thus, the source of the quadratic behaviour must arise from the normal fluid in the region outside the core of the vortices. (The field varies with a screening length λ, while the vortices are separated by a mean distance $L \approx (H_{c2}/H)\xi$ so that for $H/H_{c2} \gg (\xi/\lambda)^2$ the field is nearly uniform. For UBe_{13}, $(\xi/\lambda)^2 \approx 10^{-2}$ and $H_{c2}(T=0) \approx 6\,\mathrm{T}$. So, for $H \geqslant 0.1\,\mathrm{T}$, the field can also be considered uniform.)

For systems with strong exchange enhancement, however, one must consider the effect of the superconducting gap $\Delta(T)$ on χ_s. As $\Delta(T)$ increases with decreasing T below T_c, the fraction of material in normal regions will decrease, leading to a commensurate reduction in the spin susceptibility χ_s. In conventional s-wave superconductors, this temperature dependence is $\chi_s = \chi_N \exp(-2\Delta_0/T)$, where Δ_0 is the superconducting gap at $T = 0$. This form, as shown below, does not describe our data. For unconventional superconductors with nodes on the Fermi surface there will be a weaker temperature dependence. Line nodes yield $\chi_s \propto T$ which, by equation (2), cannot contribute to $C(H)T$. Point nodes yield $\chi_s \propto T^2$. To see whether the susceptibility obtained from equation (2) can be explained by point nodes, we fit the field dependence of $C(H)/T$ at different temperatures to the expression

$$\frac{C(H,T)}{T} = \frac{C(0,T)}{T} + aH + bH^2, \tag{3}$$

where the linear term is included to capture the effect of equation (1). (All fits were made over the region $0 < H < 0.7H_{c2}$). In figure 2 is shown the result of this fit at several temperatures; clearly the fit is very good. However, if the H^2 term in $C(H)$ were due to point nodes alone, it would yield a temperature-independent value for b. While the present data do not allow us to exclude a contribution from point nodes, it is clear that the dominant source of nonlinearity in $C(H)$ has another origin which we now discuss.

To explain the temperature dependence of χ_s in low fields, we appeal to a theory developed for liquid ^{3}He which explains the effect on the susceptibility due to interactions in a Fermi-liquid with a temperature-dependent density of states. For this case, Larkin and Migdal (1963) and Leggett (1965) have calculated the change in Fermi-liquid effects, finding for the magnetic susceptibility

$$\frac{\chi_s(T/T_c)}{\chi_s(T_c)} = \frac{(1 - F_0^a)g(T/T_c)}{1 - F_0^a g(T/T_c)}. \quad (4)$$

Here F_0^a is the exchange enhancement parameter and $g(T/T_c) = -N(0)^{-1}\Sigma_p(\mathrm{d}f/\mathrm{d}E_p)$ is the effective density of states at the chemical potential relative to its value $N(0)$ in the normal state. Because the exchange-enhanced susceptibility depends on the density of normal quasiparticles in a mean-field sense, as the superconducting gap opens and $g(T/T_c)$ decreases, the magnitude of the enhancement itself decreases. The net result is a much stronger temperature dependence of χ_s below T_c than in the absence of exchange enhancement. This effect has been known for some time to exist in ^{3}He where $F_0^a = 0.7$. In figure 2 (lower inset) we compare $(\partial^2\chi_s/\partial T^2)/(\partial^2\chi_s/\partial T^2)|_{T_c}$ data for UBe_{13} and for ^{3}He, the latter representing a fit to nuclear magnetic resonance measurements in the B phase (Ahonen *et al.* 1975). In order to extract an F_0^a value for UBe_{13} from the present data, we must assume a specific form for $g(T/T_c)$. Using $g(T/T_c)$ for either point (proportional to T^2) or line (proportional to T) nodes yields a value of $F_a^0 = 0.2(^{+0.10}_{-0.05})$. A Bardeen–Cooper–Schrieffer type of $g(T/T_c)$ cannot fit the data.

Although the temperature dependence of χ_s can be explained within a Fermi-liquid model, it is not accurate enough to allow a firm determination of the bare density of states, and hence the node symmetry. However, more information can be obtained from the magnitude of χ_s. If one integrates the fitting parameter $b = \partial^2\chi_s/\partial T^2$, assuming that $\chi_s(0) = 0$, then one obtains for the nonlinear contribution $\chi_{s,nl}(T_c) = 3.9\ (\pm 0.5) \times 10^{-3}$ emu mol^{-1}, which is a factor or four lower than the directly measured value in the normal state, 0.014 emu mol^{-1}. The most likely source of this discrepancy is a term in χ_s that is linear in T, that is due to line nodes, corresponding to d states, which arise naturally from exchange of spin fluctuations (Miyake *et al.* 1986, Schmitt-Rink *et al.* 1986).

The above discussion focused on the low-field development of χ_s, that is along route (a) in figure 2 (upper inset). We now turn to the behaviour of χ_s at H_{c2}, that is along route (b). This is most easily discussed in terms of the Maki parameter κ_2 defined by the Clausius–Clayperon relation

$$\frac{\Delta C}{T} = \left(\frac{\mathrm{d}H_{c2}}{\mathrm{d}T}\right)^2 \frac{1}{4\pi[(2\kappa_2^2 - 1)\beta_A]}, \quad (5)$$

where $\beta_A = 1.16$ (triangular flux-line lattice). To determine κ_2, ΔC was measured using an equal-area construction as shown in figure 2 and H_{c2} from the midpoint of the transition. The data are shown in figure 3 together with existing theoretical results (at present available only for s-wave systems), for both clean ($l \gg \xi_0$) (Eilenberger 1965) and dirty ($l \ll \xi_0$) (Maki 1966) systems and in the presence of Pauli limiting (Maki 1966), here characterized by $\beta^2 = 3\tau_{so}\Delta_0\alpha^2$, where τ_{so} is the spin–orbit relaxation time, and $\alpha = 2^{1/2}H_{c2}(0)/H_P$ is the usual Pauli parameter,

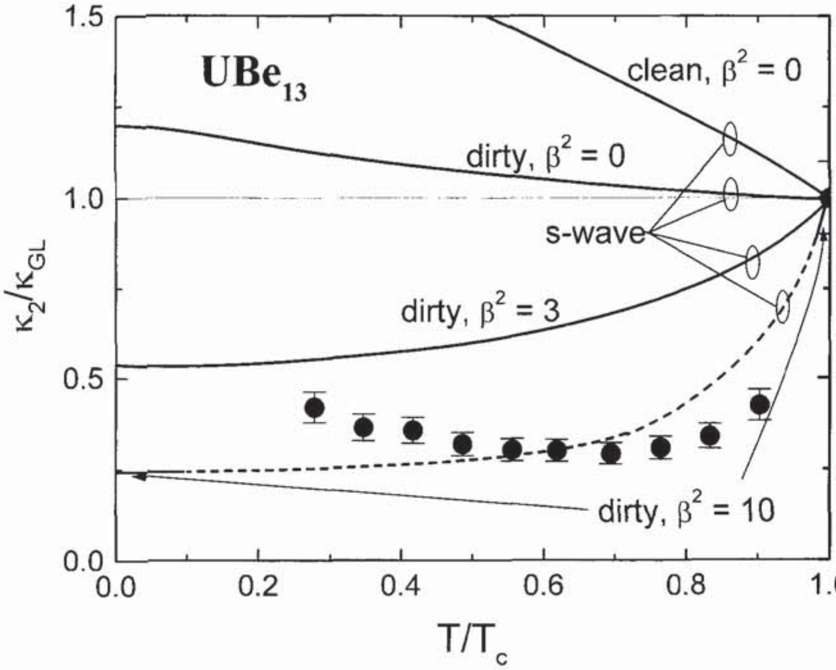

Figure 3. The ratio of the Maki parameter κ_2 to the Ginzburg–Landau parameter κ against temperature. The full curves are calculations for s-wave superconductors in various limits of transport lifetime and Pauli limiting (Eilenberger 1965; Maki 1966), as described in the text. The broken curve is a line to guide the eye, interpolating between the limiting behaviour, $T/T_c \ll 1$, $T/T_c \approx 1$ (Maki 1966), for $\beta^2 = 10$.

given $H_P = (N_0\Delta_0/\chi_{sn})^{1/2}$ where N_0 is the density of states and χ_{sn} is the normal-state spin susceptibility. Clearly, the κ_2 data near T_c are consistent with strong Pauli limiting. However, below $0.7T_c$, κ_2 *increases* as T is lowered further. This anomalous behaviour is qualitatively consistent with the Fermi-liquid effect described above where χ_{sn}, now identified with χ_s in equation (5), decreases rapidly below T_c, hence rapidly decreasing the Pauli parameter α. To analyse the data quantitatively, however, a full theory of κ_2 for low-symmetry superconductors is needed.

We note another important manifestation of the Fermi-liquid effect in UBe_{13}. In particular, one long-standing puzzle is the shape of the $H_{c2}(T)$ curve (the data in figure 1, upper inset, are typical SX data). At T_c, the slope is extremely large, $dH_{c2}/dT = -75T/K$ but, when T is decreased to only $0.9T_c$, dH_{c2}/dT has decreased by an order of magnitude. Since $H_c^2/8\pi = F_N - F_S$, where F_N and F_S are the free energies of the normal and superconducting phases respectively, a rapidly increasing H_c, and hence H_{c2}, implies a rapidly decreasing F_S because the normal-state properties are much less field sensitive. It is likely that F_S decreases with increasing field in the same way as with increasing temperature; this would stabilize a higher than usual value of H_{c2} for temperatures close to T_c. Another puzzle surrounding UBe_{13} is the large size of the jump in C/T at $H = 0$. As mentioned earlier, $\Delta C/\gamma T_c = 2.6$, which is a factor of three larger than in other HF compounds. Although the Fermi-liquid idea above deals only with long-wavelength excitations, the loss of enhancement in uniform susceptibility due to decreasing quasiparticle excitations in the superconducting state is expected to carry over to a loss of enhancement at all q and ω. Since the specific heat measures the excitations at all q for $\omega(q) < O(T)$, it is likely that the jump in $C(T, H = 0)$ at T_c has a significant contribution from the suppression of the (antiferromagnetic) spin fluctuations responsible for the large exchange enhancement in the normal state.

In summary, we have provided the first evidence for a Fermi-liquid effect in the superconducting state of a HF system, indeed of any superconductor. It would be of interest to search for related effects in the other HF systems, especially the two other heavy superconductors, $CeCu_2Si_2$ and UPt_3.

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