Critical Point in the Superconducting Phase Diagram of UPt_3

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We report on detailed measurements of the specific heat of UPt_3, performed on a high-quality single crystal in a magnetic field perpendicular to the c axis, at temperatures down to 100 mK. Two distinct phase transitions at zero field are seen to converge at a critical point, near $H=5$ kOe, which coincides with the sharp break in the H_{c2} curve. Beyond that point, there is evidence for only one phase. Combining these thermodynamic results with ultrasonic attenuation and H_{c2} data, an H-T diagram is constructed which consists of a "polycritical" point and several superconducting phases.

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Much of the wide interest shown in the heavy-electron compound UPt_3 since it was discovered[1] to exhibit superconducting properties below $T=0.5$ K has been fueled by the possibility that the superconducting instability in this metal is mostly the result of direct electron-electron interactions. Our current knowledge of the normal state in UPt_3 is perhaps the strongest support for this possibility. At low temperatures, the correlated electrons behave much like a normal Fermi liquid: The resistivity shows a quadratic dependence[2,3] and the specific heat a linear dependence[1,4] on temperature (up to $T=3T_c$), and the NMR relaxation time shows a Korringa law.[5] There exists a well-defined Fermi surface,[6] but the cyclotron masses are extraordinarily high, enhanced on average by a factor of 20 over and above the corresponding band masses. Such an enormous mass enhancement cannot be explained by a *simple* coupling to phonons, and the electron-electron interactions must play a predominant role. The residual interactions between the normalized quasiparticles, responsible for the formation of a superconducting state, may well also share this predominance. To test this idea, much direct information about the superconducting state has been accumulated, so far mainly in the form of temperature dependences of thermodynamic and transport properties. Although this information certainly points to an unusual behavior, it does not yet allow for a firm conclusion that the superconductivity in UPt_3 is truly unconventional. The ambiguity arises partly because of the range of temperatures to which most measurements so far have been restricted; the relevant low-T regime $(0<T<T_c/5)$ has never been probed reliably. In this Letter, we report on experimental results which depend neither on an extrapolation to this low-T regime, nor on any assumption about a particular model for the superconducting state, to support the view that UPt_3 is indeed host to an unconventional type of superconductivity: We present *thermodynamic* evidence for a *multicomponent* superconducting phase diagram. Previous experimental indications of several phases[3,7-11] are consistent with the overall H-T diagram derived from the present study.

The heat capacity of our UPt_3 single crystal was measured for temperatures in the range 100–700 mK and for fields perpendicular to the c axis of the hexagonal crystal structure up to 7.5 kOe. A pulse technique was used with a heat leak adjusted so that the heating time was much shorter than the relaxation time for all temperatures and fields. The temperature of the sample was measured using a phosphorus-doped silicon resistor calibrated for each field value against a RuO_2 thermometer located in a field-compensated region. The relative uncertainty on the ratio C/T is approximately 3% for $100 \leq T \leq 700$ mK. The monocrystalline sample of UPt_3 was cut by spark erosion out of a single grain from a 10.4-g polycrystalline ingot used in previous zero-field measurements.[7] The mass of the single crystal is 270 mg. Its orientation was determined by Laue scattering of neutrons, which also confirmed its monocrystalline character. The ingot was prepared following a procedure known to yield electronic mean free paths well in excess

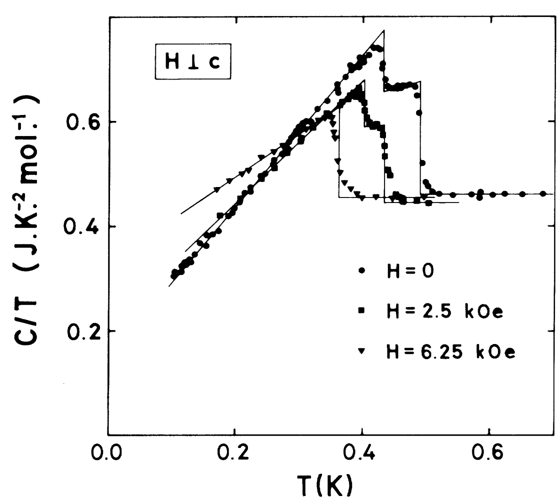

FIG. 1. Specific heat of UPt_3 near the transition for three representative values of the magnetic field, applied perpendicular to the c axis, plotted as C/T vs T. Solid lines are used to define the parameters of Fig. 4.

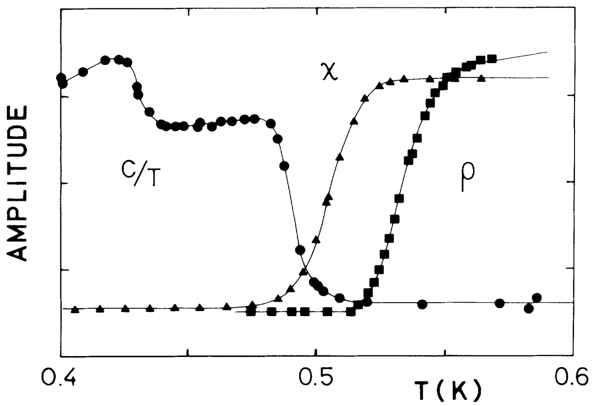

FIG. 2. Characterization of the superconducting transition by three measurements ($H=0$): (1) specific heat (C/T), (2) ac susceptibility (χ) on the single crystal used in this study, and (3) resistivity (ρ) on a *polycrystal* cut from the same ingot, next to the single crystal. The values of the transition temperature T_c (at 50%) and of the width of transition ΔT_c (90%–10%) are, respectively, (1) 490 mK ($\equiv T_2$), 15 mK; (2) 503 mK, 30 mK; and (3) 532 mK, 25 mK.

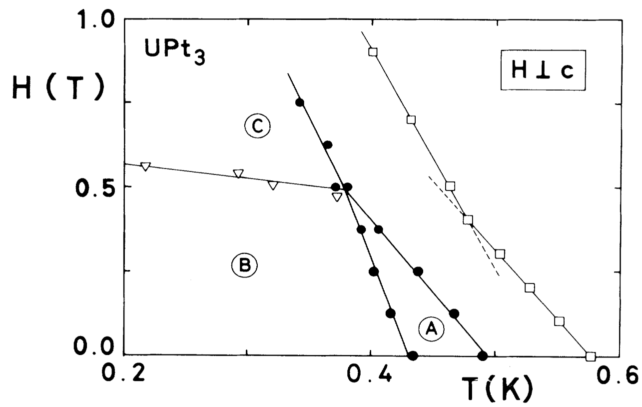

FIG. 3. Proposed superconducting phase diagram of UPt_3 for $\mathbf{H}\perp\mathbf{c}$. The existence of two phases at low fields ($0 \leq H < 5$ kOe), labeled A and B, is established by the present specific-heat measurements (circles). The transition from phase A to some other phase at $H \cong 5$ kOe is supported by the H_{c2} data (squares; from Ref. 3). Evidence that this other phase, labeled C, might be distinct from phase B comes from the peak in the ultrasonic attenuation vs field (triangles; from Ref. 9).

of 1000 Å (see Ref. 6).

Our main experimental results are presented in Figs. 1–4. In Fig. 1, the temperature dependence of the specific heat is shown as C/T vs T for three representative values of the magnetic field H.

$H=0$.—Since the zero-field behavior has been reported and discussed in a recent paper (see Ref. 7), we shall concentrate on the additional information one may derive from the lower temperatures achieved here, and from further sample characterization. The data of Fig. 1 clearly confirm the presence of two discontinuities, called 1 and 2 (with 1 referring to the lower temperature). These discontinuities are separated by 56 mK, occurring, respectively, at 434 and 490 mK; they are both *sharp*, with widths of 10 and 15 mK (see Fig. 2), and they are both *large*, with $\Delta C_i/T_i = 310$ and 207 $\mathrm{mJ\,K^{-2}mol^{-1}}$, for $i=1$ and 2, respectively. The *magnitude* of the discontinuities implies that they both involve the heavy quasiparticles. The *sharpness* eliminates the possibility of a *single* superconducting transition split by inhomogeneity of the sample, i.e., occurring at two different critical temperatures in two different parts of the crystal. Further confirmation of the intrinsic nature of the double peak is (i) its equally clear observation in the full ingot (40 times larger) out of which our single crystal was cut (i.e., sample in Ref. 7) and (ii) the high quality of the sample. This quality is ascertained by detailed study of the resistivity, the ac susceptibility, and the specific heat as a function of temperature (measured with the same thermometer) near the transition. The results are given in Fig. 2. A few features should be stressed: (i) All three curves show narrow transitions (quite a bit narrower than for previously investigated samples); (ii) the resistive T_c is high, a reliable sign of quality;[3] (iii) the susceptibility remains fully diamagnetic through both discontinuities in the specific heat; and (iv) the three onset temperatures do not coincide, with T_2 being the lowest. A possible explanation for this last point is the presence of dislocations in a bulk sample with their associated strain fields producing local regions of higher T_c

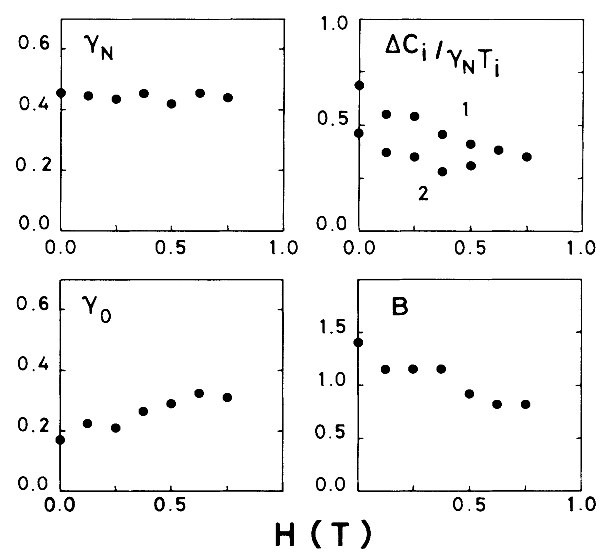

FIG. 4. Field dependence of four parameters which characterize the specific heat of UPt_3 in the vicinity of the superconducting transitions. T_1 and T_2 are the lower and upper transition temperatures, respectively; γ_N is the normal-state linear coefficient of C just above T_2; ΔC_i is the size of the discontinuity in C for both transitions ($i=1,2$); γ_0 and B are the fitting parameters of Eq. (1). Units are in K, J, and mol.

due to the strong dependence of T_c on stress. Although these regions would amount to a negligible volume, and hence have little effect on the specific heat, they would show up in the more sensitive dynamic measurements.

No sign of a latent heat could be deduced from the detailed shape of either jump in C/T, even though the steps in temperature and the size of the heat pulses were kept down to 2–3 mK. This suggests that the lower transition, at T_1, is also of the second kind.

The temperature dependence of the specific heat below T_1 follows rather closely a behavior of the form

$$C/T = \gamma_0 + BT\,, \tag{1}$$

even down to 100 mK. The most straightforward extrapolation as $T \rightarrow 0$ of both the normal-state specific heat (as $\gamma_N T$) and the observed superconducting specific heat [as in Eq. (1)] leads to an entropy of the superconducting state at $T = T_2$, which exceeds by 6% the entropy the system of electrons would have if it remained in the normal state down to $T = 0$. Hence one (or both) of these extrapolations is wrong. Since the correlated electrons appear to have firmly settled into a Fermi liquid prior to the onset of superconductivity, it is fairly natural to expect them to remain well-defined fermions as $T \rightarrow 0$. If this were the case, and $C = \gamma_N T$ were indeed a correct description of the normal-state behavior down to $T = 0$, then the real $T \rightarrow 0$ limit of C/T would not be given by γ_0 but would have to be much lower. Since the true dependence is far from established in the relevant range (i.e., $0 < T < T_c/5$), one can only conclude that Eq. (1) is not likely to hold much below 100 mK and that it may be misleading to regard the T^2 dependence of the specific heat in UPt_3 as diagnostic of the low-temperature superconducting state (i.e., of the gap structure). As a consequence, γ_0 is just a parameter which, along with others given in Fig. 4, can be used to describe the specific heat of UPt_3 in the vicinity of the superconducting transitions.

$H > 0$.—Upon application of a magnetic field transverse to the hexagonal axis of the crystal, the transition temperatures of the two discontinuities are observed to merge rapidly into each other and become indistinguishable at a field value around 5 kOe. Beyond that critical field, only one specific-heat jump is resolved. The evolution in field from a double-peak to a single-peak structure is illustrated in Fig. 1 for field strengths below and above 5 kOe, and displayed in more detail in Fig. 3, where we construct the multicomponent phase diagram of UPt_3, based on the various measurements to date (for $\mathbf{H} \perp \mathbf{c}$). The central feature of this phase diagram is what appears to be a *polycritical point* (or perhaps a critical region), given approximately by $T_c = 370$ mK and $H_c = 5$ kOe. Indeed, at that point, three or perhaps four phase boundaries seem to meet: (1) between the normal phase and the high-T-low-H superconducting phase "A"; (2) between that phase A and the low-T-low-H phase "B"; (3) between the normal phase and the high-H superconducting phase "C"; and (4) between phase B and phase C, if these two are indeed distinct. The first indication of a critical point came from detailed measurements of the temperature dependence of the upper critical field $H_{c2}(T)$ in single crystals.[3,8] For $\mathbf{H} \perp \mathbf{c}$, the $H_{c2}(T)$ curve was observed to be very linear near $H = 0$ (in the best samples), with a slope $H_{c2}' \cong 4$ T/K, and then undergo a sharp break in slope to $H_{c2}' \cong 7$ T/K at a field around 4 kOe.[3] Recently, Schenstrom *et al.*[9] have observed a peak in the attenuation of longitudinal ultrasound as a function of field in superconducting UPt_3 for a direction $\mathbf{H} \perp \mathbf{c}$ (this is the same peak as was previously observed by other groups[10] for different field directions). The position of this peak is nearly independent of temperature, and it suggests the presence of a nearly flat phase boundary at $H \cong 5$ kOe. These data (Ref. 9), reproduced here in Fig. 3, when combined with the existing H_{c2} data (of Ref. 3), with which it ties in nicely, suggest the existence of two superconducting phases at $T = 0$: one at fields below 5 kOe (the A phase) and one at fields above (the C phase). The occurrence of a zero-field transition from the A phase to some other phase (B) was discovered by recent thermodynamic measurements of Fisher *et al.*[7] The importance of the present results is to show that the phase boundary between these last two phases (A and B), as they evolve under the influence of a magnetic field, appears to end at the *same* critical point as the other phase boundary, within experimental accuracy. Of course, on the basis of the thermodynamic measurements alone, a simpler H-T diagram is possible, with only two superconducting phases: a low-T phase (where B is the same as C) and a high-T phase A rapidly suppressed by field, and no longer present above 5 kOe.

The influence of a magnetic field on the overall superconducting behavior is summarized in Fig. 4, in terms of the field dependence of the four parameters γ_N, $\Delta C_i/\gamma_N T_i$ ($i = 1,2$), γ_0, and B, which describe the specific heat of UPt_3 above, at, and below the transition region, respectively. *In the normal state*, the linear coefficient γ_N of the specific heat remains essentially constant for low values of the field (in fact this is the case up to 10 T), i.e., the "normal" Fermi liquid is little affected by a magnetic field. *In the transition region* itself, in addition to the above-mentioned convergence of T_1 and T_2 at $H \cong 5$ kOe, it is seen that in the B phase $\Delta C_1/\gamma_N T_1$ decreases with field by almost a factor of 2, i.e., the specific-heat jump at the $A \rightarrow B$ transition is suppressed more rapidly by field than the critical temperature itself. It should be noted that for both transitions the ratio $\Delta C_i/\gamma_N T_i$ is much smaller than the standard BCS value of 1.43. *In the superconducting state*, and in the B phase, the T^2 fit with the specific heat [given by Eq. (1)], which was adequate at $H = 0$, remains roughly valid in a field ($H \leq 7.5$ kOe), for the range 150 mK $< T$

$<T_1$. The two parameters γ_0 and B are H dependent (Fig. 4): the former increasing, the latter decreasing, by approximately a factor of 2, in reaching 5 kOe.

In conclusion, we have shown that with the present thermodynamic results there is now a body of evidence for the existence of several superconducting phases in UPt_3 with an H-T diagram characterized by what appears to be a critical point, as shown in Fig. 3. A coexistence of several superfluid phases is reminiscent of liquid ^3He, where this situation is well established, and of Th-doped UBe_{13}, for which such an occurrence was also suggested.[12] In all three cases, we would have a multicomponent phase diagram (H-T in UPt_3, P-T in ^3He, and x-T in $U_{1-x}Th_xBe_{13}$), with a low-temperature phase separated from the normal state by an intermediate phase. However, the differences between these fermion systems are numerous. In contrast to UPt_3, Th-doped UBe_{13} is neither pure nor stoichiometric, it may be inhomogeneous, and it does not condense into a Fermi liquid above T_c. As far as calculations are concerned, it has been shown by several authors, arguing on the basis of different superconducting states, that a superconductor of hexagonal crystal symmetry can, in principle, exhibit more than one transition[9,13,14] (at least under the assumption of d-wave pairing). However, whether it is to identify the observed phase diagram to a specific set of superconducting states, or to shed light on the underlying mechanism or type of pairing, a much more detailed knowledge of the newly discovered phases of UPt_3 is needed.

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