Angular Dependence of the Upper Critical Field of the Heavy Fermion Superconductor UPt_3

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The angular dependence of the upper critical field H_{c2} of UPt_3 in magnetic fields up to 0.85 T has been measured. In spark cut single crystals a sixfold modulation of the resistivity is observed at the superconducting transition in the basal plane. This modulation vanishes as the temperature approaches T^* [temperature of the discontinuity in slope of the $H_{c2}(T)$] and reappears for $T < T^*$ but with opposite sign. The results provide important new evidence on the nature of the change in the superconducting order parameter at T^* and its possible coupling with the antiferromagnetism.

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The unconventional nature of superconductivity (SC) in the heavy fermion superconductor UPt_3 is now well established [1], although the details of its mechanism and even the choice among the possible symmetries for the superconductivity remain unresolved. Transitions between three distinct superconducting phases (denoted A, B, and C) are seen as a function of applied magnetic field (H) and temperature (T). The three phases meet in a tetracritical point at (T^*, H^*) [2,3]. One phase, denoted by C, exists only under applied magnetic fields above H^*, while the A and the B phases persist to zero field. This is illustrated by specific heat measurements [2] where one sees two distinct second-order transitions in zero field corresponding to entering the A phase from the normal state and then at lower temperature to the change from the A phase to the B phase. The occurrence of two successive superconducting states suggests a multicomponent order parameter with a degeneracy broken by a weak symmetry breaking field (SBF) [4]. The most promising candidate for this SBF is generally considered to be an antiferromagnetic order ($T_N \approx 5$ K) comprised of small moments $\mathbf{m}\,[|\mathbf{m}| \approx (0.02 \pm 0.01)\mu_B]$ lying along the a^* directions in the hexagonal plane [5] with domain sizes of 150 Å. Evidence that the SC and antiferromagnetism (AF) are indeed coupled comes from neutron diffraction [6], upper critical field [7,8], and specific heat [9] measurements under hydrostatic pressure. Yet another candidate for the SBF could be a macroscopic incommensurate structural modulation [10] which has been observed by recent transmission electron microscope studies [11].

Several attempts based on phenomenological theories have been made to describe the temperature dependence of the in-plane upper critical field (H_{c2}). In the absence of a SBF Burlachkov argued that $H_{c2}(\Phi, T)$ would be isotropic in the basal plane of a hexagonal crystal, regardless of the nature of the superconducting state [12]. The inclusion of coupling to a SBF gives a possible explanation for the discontinuity in slope of the in-plane $H_{c2}(T)$ for both a two-dimensional representation (2D-REP) [13] and a one-dimensional representation (1D-REP) [14] of the superconducting order parameter. The 1D-REP model can reproduce the observed discontinuity in slope of H_{c2} for all in-plane field direction [7,8,15,16], whereas for the 2D-REP as it stands the discontinuity in slope of H_{c2} would be expected to vanish for $\mathbf{H} \parallel \mathbf{m}$. In both cases a small anisotropy of H_{c2} in the basal plane is expected if the SBF is locked to the crystal lattice.

In this Letter we present a new detailed study of the angular dependence of the upper critical field $H_{c2}(\Theta, \Phi, T)$ on whiskers and Czochralski grown single crystals of UPt_3 that supports the choice of a 1D-REP order parameter and gives further evidence relating to the change in the nature of the order parameter between the different superconducting phases.

The whiskers were grown by rapidly cooling a high purity melt in ultrahigh vacuum and have typical dimensions of $5 \times 0.2 \times 0.3$ mm^3, with their length parallel to the hexagonal c axis. The whisker denoted $W1$ has $T_c = 508$ mK, transition width $\Delta T_c = 18.4$ mK, and $T^* = 430$ mK. Two other samples, $S1$ ($T_c = 527.9$ mK, $\Delta T_c = 6$ mK, and $T^* = 437$ mK) and $S2$ ($T_c = 526.9$ mK, $\Delta T_c = 10$ mK, and $T^* = 421.2$ mK), were carefully spark-cut (dimension $6 \times 0.4 \times 0.6$ mm^3) from two large single crystals of UPt_3 grown separately by the Czochralski method under ultrahigh vacuum from zone-refined depleted uranium. The as-grown crystals were annealed for several days at (1200–1300) °C (see Ref. [17]), and the cut and etched bars were subsequently annealed for an additional week at 950 °C, improving the sharpness of the resistive transition, which coincides with the onset of the specific heat jump. A comparison of the resistive transitions of samples $W1$ and $S1$ is shown in the inset of Fig. 1.

The angular dependence of the critical field was determined by both four point ac resistivity and susceptibility measurements. The samples were mounted on the cold finger of a miniaturized dilution refrigerator (ϕ 38 mm) and at the center of a three axis magnet consisting of

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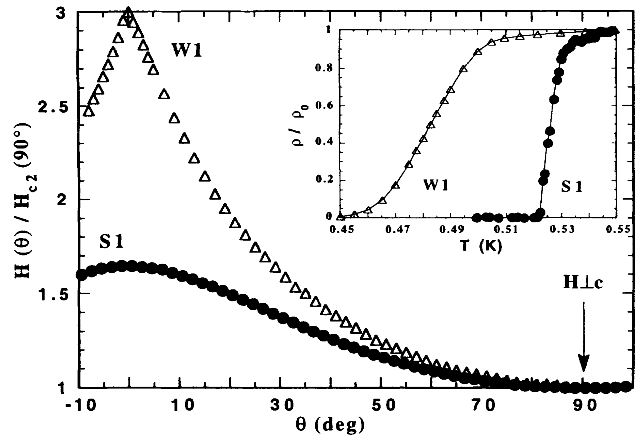

FIG. 1. Angular dependence $H_{c3}(\Theta)/H_{c2}$ of the whisker $W1\,(\Delta)$ compared to $H_{c2}(\Theta)/H_{c2}$ of the sample $S1$ ($\bullet$) at $T/T_c = 0.932$ for H oriented between the **c** axis and the basal plane. The inset shows the resistive transitions of sample $W1\,(\Delta\!: H = 0.05$ T) and sample $S1\,(\bullet\!: H = 0$ T).

two pairs of Helmholtz coils and one solenoid. The applied magnetic field (maximum 0.85 T) can be oriented spatially with an angular accuracy of 0.1°, while keeping its magnitude constant to better than 1%. As no mechanical movement is needed, a great stability in the sample temperature and a good reproducibility in the results are achieved. We used a low current density of $J \approx 0.4$ A/cm^2 to preserve the sharpness of the resistive transition under magnetic field. The resistive superconducting transition is shifted without change of form to higher or lower temperatures as the angle of the applied field is varied in the basal plane or between the basal plane and the **c** axis. Therefore, by keeping the actual temperature $T = T_c$ fixed, a rotation of the magnetic field results in a variation of the measured resistance if H_{c2} changes with direction. Alternatively taking the midpoint of the resistive transition as a definition of T_c, the strength of the magnetic field was adjusted to maintain a constant value of the resistance for each direction of the magnetic field. In both cases the results can be interpreted as measuring the angular dependence of H_{c2} for each given temperature.

For the whisker $W1$, the angular behavior of the critical field $H_{c2}(\Phi)$, which is presented in Fig. 2, shows sharp peaks with a periodicity of 60° for rotation of the applied field in the hexagonal plane, as has been observed in previous resistivity measurements [8,16]. In fact, the sharp nature of the variations of the critical field at $\Phi \sim 30°$ and 90° suggests that the critical field probed is, in fact, $H_{c3}(\Phi)$ due to surface superconductivity [18], which becomes strongly enhanced when the field is oriented in the hexagonal plane and parallel to two of the six surfaces of the whisker (the surface normals show a sixfold symmetry). The extension of the simple model describing the angular dependence of the H_{c3} [19] to the

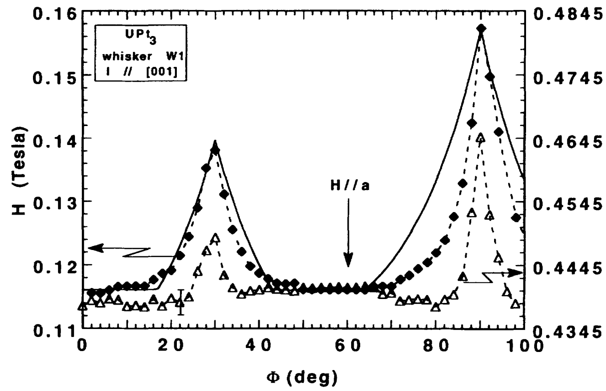

FIG. 2. $H_{c3}(\Phi)$ measured on the whisker $W1$ at 470 mK ($\blacklozenge$) and 415 mK (Δ) for $\mathbf{H} \perp \mathbf{c}$. The solid line is a fit of the H_{c3} model to the 470 mK data (see text). The dashed lines are guides to the eye.

case of six plane surfaces as presented by the whisker gives a theoretical variation,

$$H_{c3}(\Phi)/H_{c2} = \max|(|\sin(\Phi - \Phi_n)| + \varepsilon_n^*|\cos(\Phi - \Phi_n)|)^{-1}|_n, \quad (1)$$

where $\Phi_n = (2n + 1)\pi/6$, $\varepsilon_n^* = H_{c2}/H_{c3}(0°)|_n$, and $n = 1, 2, 3$.

The solid line in Fig. 2 shows a fit to the data ($T = 470$ mK), taking $\varepsilon_1^* = 0.831$ and $\varepsilon_2^* = \varepsilon_3^* = 0.739$ (Ref. [20]. Experimentally the relative amplitude $H_{c3}(\Phi)/H_{c2}(60°)$ at $\Phi = 30°$ and 90° decreases with decreasing temperature and increasing field strength [$H_{c3}(90°)/H_{c2}(60°) = 1.35, 1.15$, and 1.05 for $T = 470$, 450, and 415 mK, respectively], whereas theoretically $H_{c3}(T)/H_{c2}(T)$ should increase with decreasing temperature [21]. This unusual behavior can, however, be understood by considering either a crossing from orbital to paramagnetic limitation or the action of an internal exchange field. The interpretation that the critical field seen is H_{c3} is strengthened by the observation of a similar angular dependence of the upper critical field for directions between the basal plane and the **c** axis (Fig. 1). A very pronounced maximum is observed for the whisker near $\mathbf{H} \parallel \mathbf{c}$, which cannot be accounted for in terms of a simple effective mass model ($\varepsilon^2 = m_\parallel/m_\perp$):

$$H_{c2}(\Theta)/H_{c2}(\pi/2) = (\sin^2\Theta + \varepsilon^2\cos^2\Theta)^{-1/2}. \quad (2)$$

These surface effects observed in whiskers are not apparent in measurements on the Czochralski grown single crystals ($S1, S2$). The phase diagram (see Fig. 3) determined by resistivity and confirmed by susceptibility measurements shows a discontinuity in slope of $H_{c2}(T)$ at $T^* \approx T_c - 0.09$ K for $\mathbf{H} \perp \mathbf{c}$ and nearly no anomaly for $\mathbf{H} \parallel \mathbf{c}$. The angular dependence $H_{c2}(\Theta)$ for $S1$ shown in Fig. 1 is well fitted ($\varepsilon^2 \approx 0.372$) by the anisotropic mass model [Eq. (2)] for $H_{c2\parallel c}/H_{c2\perp c} \sim 1.6$, consistent with other published values [3].

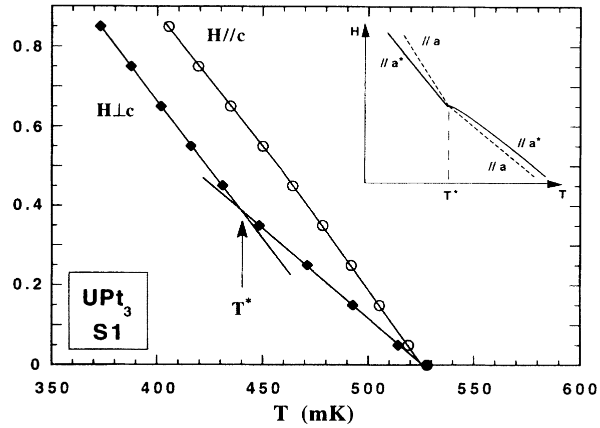

FIG. 3. Phase diagram (sample $S1$) determined for $\mathbf{H} \parallel \mathbf{c}$ (∘) and $\mathbf{H} \perp \mathbf{c}$ (◆). The lines are guides to the eye. The inset shows schematically the anisotropy of the phase diagram in the hexagonal plane where the phase lines ($\mathbf{H} \parallel \mathbf{a}$ and $\mathbf{H} \parallel \mathbf{a}^*$) cross at T^*.

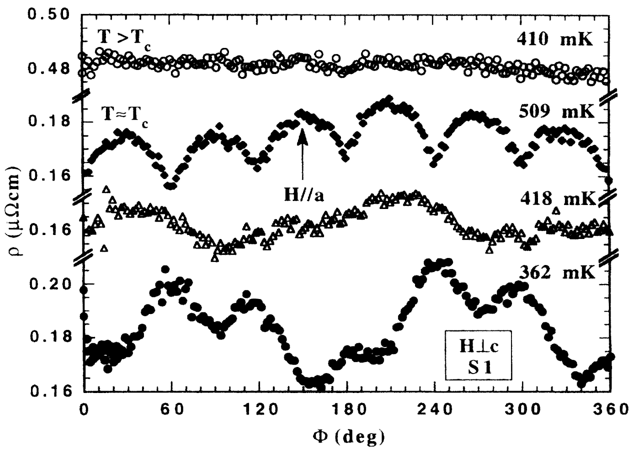

FIG. 4. Angular variation of the resistivity ($\mathbf{H} \perp \mathbf{c}$) at different temperatures and fields (sample $S1$). The resistivity is isotropic in the normal state (∘: $T = 410$ mK, $H = 0.75$ T). The resistivity curves measured in the middle of the superconducting transition at 509 mK (◆), 418 mK (Δ), and 362 mK (•) show a sixfold modulation, which changes its phase by passing the discontinuity at T^* (= 437 mK) in the phase diagram.

We next discuss our detailed study of the anisotropy in the basal plane. The angular dependence of $\rho(\Phi)$ measured on samples $S1$ and $S2$ is similar but more pronounced for sample $S1$ which has a sharper drop in the resistivity at T_c. In normal state ($T > T_c$ and $|\mathbf{H}| = 0.75$ T), the resistivity showed no anisotropy in the hexagonal plane to the limit of our experimental resolution. For $T \sim T_c$ in a small applied magnetic field ($|\mathbf{H}| = 0.05$ T), the resistivity as a function of angle presents a small but well pronounced sixfold modulation (Fig. 4). The maximum amplitude of the modulation in sample $S1$ is about 3% of the initial resistivity value at the midpoint of the transition. The angular dependence of the resistivity around the minimum (Φ close to 60°, angular resolution of 0.5°) is smooth, and it can be described by a Taylor series expansion in powers of the angle. The sixfold modulation in $\rho(\Phi)$ disappears progressively as the temperature decreases towards $T \approx T^*$ ($T^* = 437$ mK, $S1$). At lower temperatures $T < T^*$ this modulation reappears but with opposite sign (see Fig. 4). A Fourier transform of the discrete set of N data points $R(\Phi_k)$, written $R(n) = \sum_{k=0}^{N-1} R(\Phi_k) \exp(-i2\pi n \Phi_k/N)$, confirms the presence of a sixth-order component [$R(6)$], which decreases continuously in amplitude from T_c to T^*, vanishes near T^*, and increases below T^* with opposite sign. A superposed fourth-order component [$R(4)$] is related to the sample geometry, and its amplitude increases continuously with decreasing temperature. A second-order component [$R(2)$] is attributed to the misalignment of the contacts and to a residual angle between the basal plane and rotation plane of the field.

Although the strong sixfold surface effects observed in hexagonal whiskers are absent in these rectangular samples $S1$ and $S2$, one can ask whether the remaining sixfold modulation in $S1$ and $S2$ cannot be attributed to some small scale hexagonal structure of the surfaces, which might give a small surface superconductivity contribution to $R(6)$. Electron-microscope studies, however, did not show any microscopic faceting of the surface orientation down to the scale of 1 μm, which rules out surface superconductivity as an explanation of the sixfold modulation in $S1$ and $S2$. Moreover, the fact that $H_{c2}(\Theta)$ is described well by the effective mass model [Eq. (2)] further excludes the possibility that surface properties play a role.

The in-plane anisotropy implies the existence of separate phase lines in the phase diagram ($\mathbf{H} \perp \mathbf{c}$) for the two main directions $\mathbf{H} \parallel \mathbf{a}^*$ and $\mathbf{H} \parallel \mathbf{a}$ (inset Fig. 3, schematic representation). To confirm this directly, we also measured the complete resistive transition as a function of temperature for two orientations ($\mathbf{H} \parallel \mathbf{a}^*$ and $\mathbf{H} \parallel \mathbf{a}$ at $\Phi = 0°$ and 30°, respectively) of the magnetic field. A small difference [$\Delta T_c(H)$] in $T_c(H)$ between these two directions in the A phase ($H < 0.4$ T) is clearly distinguishable. Near T_c the maximum value of the difference is $\Delta T_c \approx 0.5$ mK. Above 0.4 T (C phase), no important anisotropy can be detected to the limit of the experimental resolution. The suppression of ΔT_c coincides with the discontinuity of the slope of $H_{c2}(T)$ in the phase diagram (Fig. 3).

Our results clearly show a weak anisotropy of H_{c2} in the basal plane. We suggest that its origin stems from the antiferromagnetic order and that the macroscopic 60° anisotropy reflects the three possible magnetic domain directions in the hexagonal plane. As $H_{c2}(\Phi, T)$ is not isotropic, the observed anisotropy between the $\mathbf{a}$ and the $\mathbf{a}^*$ directions (Fig. 4, inset Fig. 3) excludes the possibility of continuous domain rotation to follow the applied in-plane field. Furthermore, while both the 2D-REP [13] and the 1D-REP [14] models predict an anisotropy of $H_{c2}(T)$ for the orientations $\mathbf{H} \perp \mathbf{m}$ and $\mathbf{H} \parallel \mathbf{m}$ (with $\mathbf{m}$ aligned to $\mathbf{a}^*$), the 2D-REP model also predicts the absence of the discontinuity of $H_{c2}(T)$ along certain in-plane directions in contradiction to our experimental results where the discontinuity is present for all angles in the basal plane. The reversal of sign in the sixfold modulation of the resistivity takes place as the working point on the $H_{c2}(T)$ line changes from the A phase into the C phase. This change in sign of the modulation is direct evidence for a change in nature of the SC order parameter between the two phases as proposed in the 2D-REP and the 1D-REP models.

In summary, we have measured an anisotropy of $H_{c2}(T)$ and a sixfold modulation of $\rho(\Phi)$ at the superconducting transition in the hexagonal plane of spark-cut single crystals of UPt_3 ($S1, S2$). The observed change in sign of the sixfold modulation passing from the A phase into the C phase gives direct evidence for a change in nature of the unconventional superconducting order parameter at T^*. As the sample geometry is rectangular (samples $S1$ and $S2$), surface superconductivity as seen for the whisker $W1$ cannot account for the observed anisotropy. The determined phase diagram for all the samples presents a topologically identical behavior with a discontinuity in slope for every direction of the applied magnetic field in the basal plane [7,8,15,16], supporting the idea of a coupling between the SBF and the unconventional SC order parameter. The measurements show clearly the existence of a small anisotropy between the $\mathbf{a}$ and the $\mathbf{a}^*$ axes in the basal plane, which in this interpretation indicates that the antiferromagnetic domains do not continuously follow the rotation of the magnetic field.

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