Magnetic Field Dependent Specific Heat Across the Middle and Upper Critical Field Lines In UPt_3

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The specific heat C of the heavy fermion vector superconductor UPt_3 has been measured with high resolution as a function of magnetic field across the middle and upper (H_{c2}) critical field boundaries for temperatures below the tetracritical point. For $T \ll T_c$ and $H_{c1} \ll H \ll H_{c2}$, $C(H)$ grows at $H^{1/2}$, in agreement with a recent prediction for a superconductor with lines of zeros in the gap function. At higher fields, a distinct jump ΔC is found across both boundaries and in both principal crystallographic directions. The inferred susceptibility jumps imply highly anisotropic Ginzburg-Landau parameters κ_2. At H_{c2}, $d\kappa_2/dT$ displays a sign reversal between the two directions, unique among superconductors. This suggests a strong influence of the gap symmetry on thermodynamic quantities.

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The nature of superconducting order parameter in the heavy fermion compound UPt_3 remains one of the most intriguing problems in condensed matter physics. Among all superconductors, UPt_3 is unanimously viewed as possessing a vector order parameter, in contrast to the usual BCS s-wave state. Evidence for a vector order parameter in UPt_3 comes from the occurrence of the following: (i) characteristic power-law temperature dependences of thermodynamic properties [1]; (ii) the highly anisotropic gap [2,3]; and (iii) the existence of distinct phase boundaries within the mixed state, as sketched in the insets to Figs. 1 and 2 [4–7]. Although it is possible in principle for s-state systems to display features (i) and (ii), the multiple-phase character (iii) of the mixed state is unique to a vector order parameter system. Theories abound to describe the nature of the order parameter between these phases—however, no consensus has been reached concerning the microscopic description of the pairing and the symmetry relationships between the different phases.

The various models advanced to explain the multiple phases in UPt_3 take the form of Ginzburg-Landau (GL) free energies dictated in a phenomenological way by symmetry considerations [8–11]. These theories are, therefore, amenable to the usual thermodynamics tests. In the present Letter we provide new thermodynamic data which serve to constrain such models. We present high-resolution field-dependent specific heat $C(H)$ data and show that the jump in $C(H)$ (ΔC) across both BC and CN boundaries is highly anisotropic. Across CN, the GL parameter $\kappa_2(T)$ exhibits marked anisotropy between the two crystallographic directions, parallel and perpendicular to the hexagonal crystal axis c. More surprising is that $d\kappa_2/dT$ displays a sign reversal between these two directions. For $H \perp c$, $d\kappa_2/dT < 0$, in accord with Abrikosov-Gor'kov theory and also as is found for traditional s-wave superconductors. For $H \parallel c$, however, $d\kappa_2/dT > 0$, which, to our knowledge, is unique behavior among superconductors. Across BC, a much smaller $\Delta C(H)$ is observed which yields a more isotropic effective $d\kappa_2/dT$.

The measurements were made on a Czochralski-grown single crystal possessing two cleanly resolved zero-field specific heat peaks and an extrapolated resistivity ratio of $\rho(300)/\rho(0) \approx 190$. Measurements were performed using a standard semi-adiabatic heat-pulse technique. To achieve fine resolution in magnetic field, data were taken in a small temperature range ($\approx$10–20 mK) around the temperature of interest in fixed fields. The C versus T data were then fit to a straight line from which C at a particular T within the small temperature range was calculated. In order to minimize torquing (estimated to

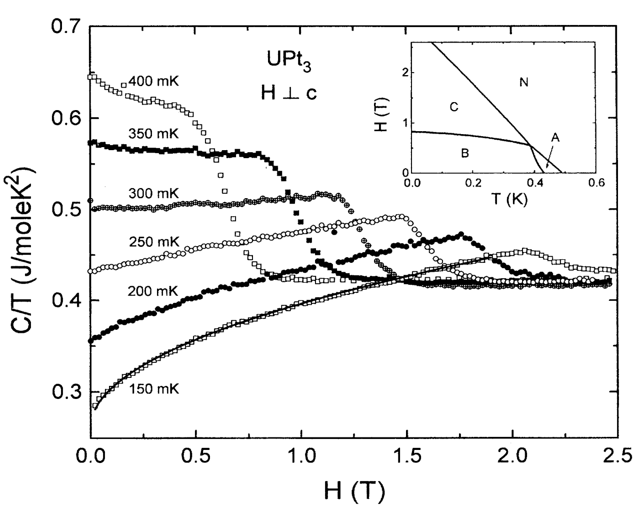

FIG. 1. Specific heat divided by temperature C/T, as a function of applied field $H \perp C$ for different temperatures. The rounded discontinuities correspond to $\Delta C/T$ at H_{c2}. The inset shows a schematic drawing of the phase diagram for $H \perp C$ based on the magnetostriction data of Ref. [7]. Here A, B, and C are different superconducting states and N denotes the normal phase. The solid line is the result of a least-squares fit of the C/T data to $\{a + b\sqrt{H}\}$ over the field 0–1.9 T, and yields $a = 0.262$ J/mole K^2, $b = 0.134$ J/mole K^2 T$^{1/2}$.

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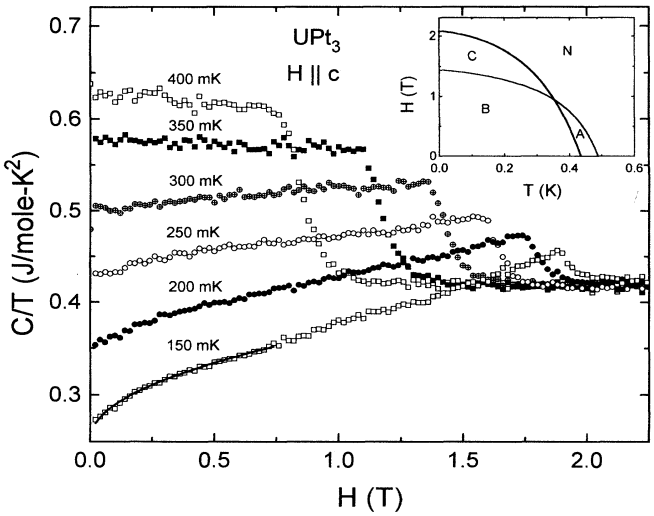

FIG. 2. Specific heat divided by temperature C/T, as a function of applied field $H \parallel C$ for different temperatures. The rounded discontinuities correspond to $\Delta C/T$ at H_{c2}. The inset shows a schematic drawing of the phase diagram for $H \parallel C$ based on the magnetostriction data of Ref. [7]. The solid line is the result of a least-squares fit of the C/T data to $\{a + b\sqrt{H}\}$ over the field 0–0.75 T, and yields $a = 0.254$ J/mole K^2, $b = 0.114$ J/mole K^2 T$^{1/2}$.

be small at the critical fields) of the sample in response to the applied field, the sapphire calorimeter supporting the sample was rigidly held by a number of cotton threads anchored to the top-loading sample holder.

In Figs. 1 and 2 we show $C(H)$ data for H perpendicular and parallel to the c axis, respectively. At low fields, ~0.5 T, $C(H)$ exhibits downward curvature. This feature is clearly distinct from H_{c1}—the intrinsic H_{c1} is 2 orders of magnitude smaller [3], and surface-pinning features usually occur at fields 1 order of magnitude smaller [12,13]. Volovik [14] has recently shown that the density of states in a superconductor with line nodes in the gap displays a characteristic square root dependence on H for $T \ll T_c$. With a view towards this interpretation, we have fit the data for $T = 150$ mK and $H < H_{c2}$ to a function $C/T = a + b\sqrt{H}$, and show the results in Figs. 1 and 2. The clear nonlinearity of $C(H)$ and the good agreement with this fitting form strongly suggest the presence of line nodes, in agreement with μSR studies [3].

At slightly higher fields, a region of roughly linear $C(H)$ behavior is found corresponding to creation of normal regions in the vortex cores. For $H > H_{c2}$, C/T shows no H dependence and a weak rise as T decreases, consistent with previous studies [15]. Finally at the upper critical field H_{c2}, a sharp anomaly is seen. We note that previous measurements of this type observed a much broader feature, most likely the result of lower sample quality, as indicated by the absence of double peaks in zero field [16]. To determine $\Delta C/T$ at H_{c2}, an extrapolation of straight line fits from above and below H_{c2} to the middle of the transition was used. The temperature dependence and anisotropy of $\Delta C/T$ will be discussed below. With the quality of data shown in Figs. 1 and 2, it is difficult to ascertain any sharp feature which might be associated with the transition between B and C phases as shown in the inset. This observation is consistent with a previous estimate of a few mJ/mole K^2 for the magnitude of the expected jump in C based on the slopes of the phase boundaries and the observed jumps across NC, NA, and AB [7,17]. In order to directly observe this jump and track its T dependence, we performed a series of higher resolution measurements, the results of which are shown in Figs. 3 and 4.

The data in Figs. 3 and 4 show the results of higher-resolution scans. As a result of surface pinning of flux lines, there can be significant low-field hysteresis in the magnitude of $C(H)$, depending on the thermal history of the sample—for this reason, every field scan was done both for increasing as well as decreasing field. In addition to the hysteresis, however, distinct and reproducible jumps are observed at fields consistent with those where sharp features are seen in both sound velocity [5,6] and magnetostriction measurements [7]. These features define the BC boundary (note that there exists ~10% variability in the location of the phase boundaries among different samples). The existence of $\Delta C/T$ jumps suggests a second-order phase transition. In addition, as alluded to above, the agreement of the size of the jumps (as the tetracritical point is approached) with predicted values

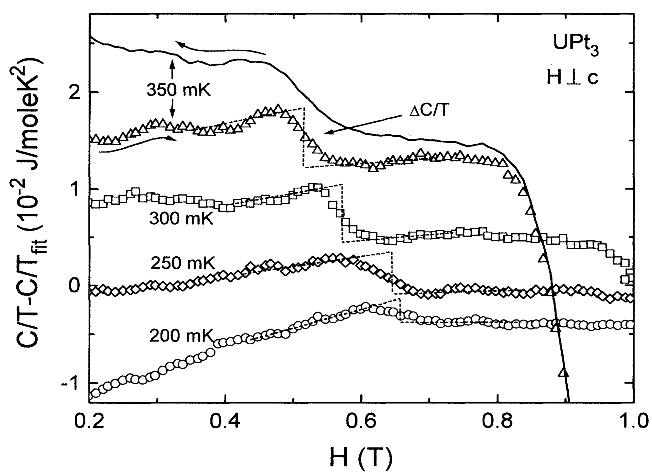

FIG. 3. The specific heat divided by temperature, as a function of H, for $H \perp c$ at various temperatures, with a linear least-squares fit to the data in the C region of the phase diagram subtracted. The different data sets have been offset vertically for clarity. The rounded discontinuity depicted as $\Delta C/T$ marks the BC phase boundary. Examples of equal area constructions used to determine $\Delta C/T$ are shown. For 350 mK, data are shown both for increasing as well as decreasing field showing an example of hysteresis, presumably related to surface-boundary pinning of flux lines.

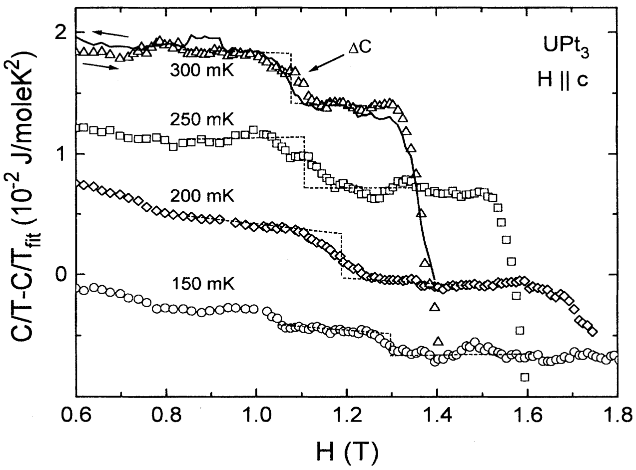

FIG. 4. The specific heat divided by temperature, as a function of H, for $H \parallel c$ at various temperatures, with a linear least-squares fit to the data in the C region of the phase diagram subtracted, as in Fig. 3. For 300 mK, data are shown for both increasing as well as decreasing field.

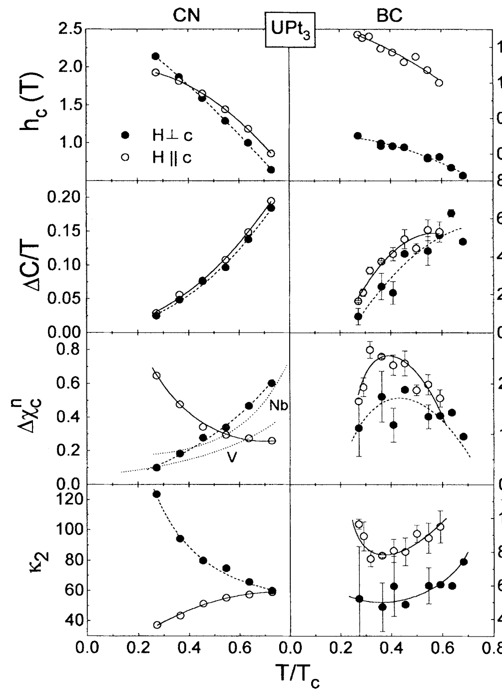

FIG. 5. The left side shows the critical field in Tesla, the specific heat discontinuity $\Delta C/T$ in J/mole K^2, the normalized susceptibility discontinuity $\Delta\chi_c^n$, and the Ginzburg-Landau parameter κ_2 for the upper critical field (CN) boundary in UPt_3. The right side shows the same quantities for the middle (BC) phase boundary, where $\Delta C/T$ is now expressed in mJ/mole K^2. Error bars are statistical uncertainties in $\Delta C/T$, based on the results of several field sweeps. All lines through the data are only guides to the eye. Also shown as dotted lines are $\Delta\chi_c^n$ at H_{c2} for the type-II superconductors Nb and V. The marked anisotropy in $d\kappa_2/dT$ for the CN phase boundary is discussed in the text.

demonstrates thermodynamic consistency among all four phase boundaries.

To analyze the temperature dependence of ΔC at the BC and CN phase boundaries, recall from thermodynamics that for any magnetic phase transition governed by a phase boundary $h(T)$,

$$\Delta C = T(dh/dT)^2\Delta\chi/4\pi\,, \quad (1)$$

where $\Delta\chi$ is the jump in magnetic susceptibility across the boundary. At H_{c2} in a type-II superconductor, $\Delta\chi$ is given by the Abrikosov solution $[(2\kappa_2^2 - 1)\beta_A]^{-1}$, where κ_2 is a GL parameter, and β_A is a constant of order unity, determined by the symmetry of the flux lattice [18]. To compare the temperature dependences of $\Delta\chi$ at H_{c2} among different materials, we defined the quantity $\Delta\chi_c^n \equiv (\Delta c/t)/(dh/dt)^2$, where c, h, and t are normalized to $\Delta C(T_c)$, $H_{c2}(T = 0)$, and T_c(=0.55 K for UPt_3), respectively. For the BC transition, the normalization is done with respect to ΔC(0.35 K), h(0.15 K), and $T_c = 0.55$ K.

In Fig. 5 we show for both BC and CN boundaries, the temperature dependence of the critical field, $\Delta C/T$, and $\Delta\chi_c^n$. We see first for CN that, whereas $\Delta C/T$ displays a similar temperature dependence in both crystallographic directions, $\Delta\chi_c^n$ is highly anisotropic. For $H\perp c$, $\Delta\chi_c^n$ appears to follow a temperature dependence similar to that found in other clean type-H superconductors, Nb [19] and V [20]. One expects that as $T \to 0$, $\Delta\chi_c^n$ will vanish logarithmically, consistent with the clean limit behavior of κ_2 [21]:

$$\kappa_2 \approx 1.22\kappa[\ln(T_{c0}/T)]^{1/2}. \quad (2)$$

For $H \parallel c$, however, $\Delta\chi_c^n$ has the inverse temperature dependence—it *increases* with decreasing T. This results from the combination of a smaller critical field slope [22] and a similar-size $\Delta C/T$ for $H \parallel c$ compared to $H\perp c$.

In Fig. 5, these data are expressed as a temperature-dependent $\kappa_{2,(\perp,\parallel)}$. Here both the difference in absolute values of $\kappa_{2,\perp}$ and $\kappa_{2,\parallel}$, as well as their opposing temperature dependences, vanish, as expected, as $T \to T_c$. The average value of κ_2 between the two directions near T_c is in good agreement with $\kappa \approx (\lambda_L/\xi_0)_{T=0} \approx 7000/120 = 58$, where λ_L is the London penetration length and ξ_0 the coherence length, both expressed in Å [3]. We note that there is no indication of a sign reversal in $d\kappa/dT$ between the two directions using $\kappa(T) \equiv \lambda(T)/\xi(T)$, using $\lambda_L(T)$ from Ref. [3], $\xi(T)$ extracted from $H_{c2}(T)$ measurements [22]. This suggests that κ and κ_2 depend on the gap structure in different ways.

Anisotropic values for κ itself are not uncommon—for instance, in the layered material $NbSe_2$, $\kappa_\parallel = 30$, while $\kappa_\perp = 9$ [23] (the anisotropy of κ_2 has not been measured for $NbSe_2$). *The unusual aspect of the present data stems from the sign reversal in $d\kappa_2/dT$ between the two different directions.* Important in this regard is that in $NbSe_2$, the orientation dependence of H_{c2} [23] shows no crossing behavior between different temperatures, as is found in UPt_3 [22]. Other anisotropic superconductors also show the effects of energy scale anisotropy. For instance, in the organic superconductor $(TMTSF)_2ClO_4$, the magnitudes of H_{c2} vary widely among the three principal crystal directions, but no sign of crossing or anomalous slope changes is observed [24]. In the high-T_c cuprate materials, H_{c2} is beyond experimental accessibility for this type of study.

The behavior at the middle transition (BC) also displays anisotropy, though less pronounced than across CN. In Fig. 5 is also plotted an effective κ_2 for this transition. Physically, this κ_2 probably corresponds to a change in the degree of type-II character related to a presumed rotation of the superconducting order parameter. Interestingly, although κ_2 itself is anisotropic, $d\kappa_2/dT$ is nearly isotropic across BC.

To understand the anomalous behavior in $d\kappa_2/dT$ it will be necessary to go beyond traditional s-wave descriptions of electron pairing. From Eq. (2), for instance, it is clear that the discrepancy between $d\kappa_2/dT$ and $d\kappa/dT$ cannot be understood in simple microscopic terms, say, as a quasiparticle mass anisotropy. Towards this, it has been noted that the unusual crossing of H_{c2} in UPt_3 can be explained by invoking paramagnetic limiting for $H \parallel c$ within the context of vector superconductivity for odd-parity pairing [25]. This calculation essentially determines anisotropy in energy scales for the two directions. It would be of interest to examine whether the anisotropy observed in the *scale-invariant* thermodynamic parameter κ_2 can be described by the same ground states as proposed to explain H_{c2} alone. Similarly, the behavior of the effective κ_2 across the BC boundary should place constraints on the possible low field B phases.

In conclusion, we have measured the specific heat jumps across the middle and upper phase boundaries and in both principal directions for the vector superconductor UPt_3. The observed anisotropy in the temperature derivative of the inferred Ginzburg-Landau parameter κ_2 is unique among superconductors, apparently reflecting an unusual temperature-dependent gap structure.

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