Uniaxial-Stress Anisotropy of the Double Superconducting Transition in UPt_3

D. S. Jin, S. A. Carter, B. Ellman,[(a)] and T. F. Rosenbaum

The James Franck Institute and Department of Physics, The University of Chicago, Chicago, Illinois 60637

D. G. Hinks

Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439

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We study the specific heat of single-crystal UPt_3 for uniaxial stress $0 \le S \le 4$ kbar. For $S \parallel \hat{c}$, the lower and upper superconducting transitions move towards each other in temperature and merge by $S=2$ kbar and $T=485$ mK. In the normal state, the Sommerfeld constant γ increases 12% by 2 kbar and then plateaus. By contrast, for $S \parallel \hat{a}$, γ decreases steadily and the onset T_c's of the two superconducting transitions remain essentially unchanged, although the specific-heat peaks broaden markedly. Residual strain in the basal plane may account for some crystals of UPt_3 showing only one broad superconducting transition.

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The investigation of anisotropic responses in the heavy-fermion superconductors has revealed essential aspects of the superconducting state. Axial anisotropy in the temperature dependences of the ultrasonic attenuation [1] and the magnetic penetration depth [2] has established the nodal structure of the superconducting gap in UPt_3. Group-theory considerations [3] then limit the available unconventional superconductor representations, but cannot establish uniquely the nature of the pairing. Pronounced anisotropy in the upper critical field [4] provides further clues, and recent detailed analyses [5,6] of the $H_{c2}(T)$ directional dependence allow further constraints on the parity and the order of the non-*s*-wave pairing mechanism.

The discovery of two zero-field superconducting transitions [7] in UPt_3 has opened up a new chapter in heavy-fermion physics. In contrast to the marked axial anisotropy of the normal-state–superconductor transition, the double-transition H-T phase diagrams [8,9] are topologically isotropic with respect to field direction. This striking isotropy in an anisotropic superconductor has spurred additional representation scenarios [10] for the superconducting state in UPt_3 and has raised as well concerns about sample homogeneity.

In addition to magnetic field, uniaxial stress can serve as a symmetry-breaking field. We report here measurements of the specific heat of single crystals of UPt_3 stressed parallel to and perpendicular to the hexagonal basal plane. Our primary result is the first evidence of an anisotropic phase diagram for the superconducting double transition. Moreover, our findings indicate that residual stresses in the basal plane may explain the current mystery of why some UPt_3 crystals have only one superconducting peak in the specific heat.

Single crystals of UPt_3 were grown by the vertical-float-zone refining method, annealed at 950 °C for 12 h, and then slowly cooled. Characterization via ac magnetic susceptibility gives a single $T_c=545$ mK with a 10%–90% transition width of 7 mK. These samples are prepared similarly to those used in the ultrasonic velocity determination of the double-transition H-T phase diagram [9], samples whose heat capacity has not been measured previously. Uniaxial stress was applied to crystals of typical dimensions $1.5\times1.0\times1.0$ mm^3 with 1.5 mm^2 parallel faces cut perpendicular to either the $\hat{a}$ or the $\hat{c}$ axis. The stress cell was a NbTi hollow tapped cylinder, designed to fit into the top-loading chamber of a dilution refrigerator and illustrated in the inset to Fig. 3. Uniaxial stress up to 4 kbar was applied with a torque wrench and mediated via a NbTi spacer which prevented sample rotation during tightening.

The stress was calibrated by using the cell as a Brinell hardness indenter. A small Al block and $D=\frac{3}{32}$ in. diam WC ball replaced the UPt_3 crystal, various torques were applied, and the indentation diameters d in the Al were measured. The load L is then given by the relationship [11] $H_B=2L/\pi D^2\{1-[1-(d/D)^2]^{1/2}\}$, where the Brinell hardness $H_B=86\pm2$ was determined independently by indenting Al with known loads, L is in kg, and d and D are in mm. Absolute values of uniaxial stress should be accurate within 5%.

The heat capacity was determined by measuring the exponential decay of the temperature after application of a known heat pulse. The heater and Speer carbon chip thermometer were mounted on the outside edges of a thin copper foil whose center was compressed between the NbTi spacer and the sample. The heat leak was a narrow graphite block which doubled as the cell mount. The addendum from the stress cell was negligible at all temperatures of interest given the disparity between the superconducting transition temperatures of NbTi and UPt_3 and the large Debye temperature of NbTi.

We plot in Fig. 1 the two superconducting transitions in UPt_3 as a function of magnetic field H parallel and perpendicular to the c axis, respectively. In either orientation, both transitions move to lower temperature with increasing field and eventually merge. The magnetic-field scale is different as is the rate at which the entropy

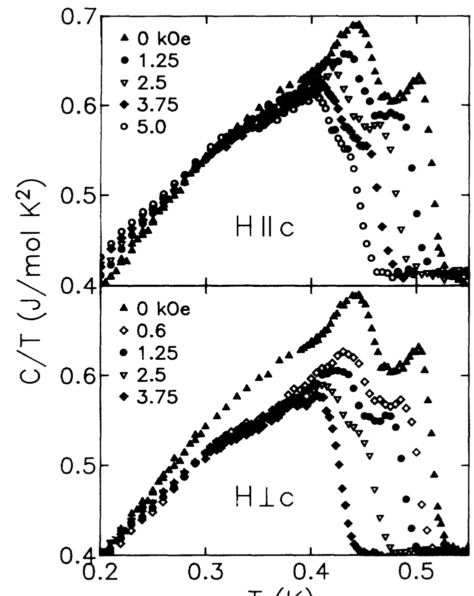

FIG. 1. Specific-heat determination of the double superconducting transition in single-crystal UPt_3 with magnetic field parallel (top) and perpendicular (bottom) to the c axis. Both transitions move down in T with increasing H, eventually merging.

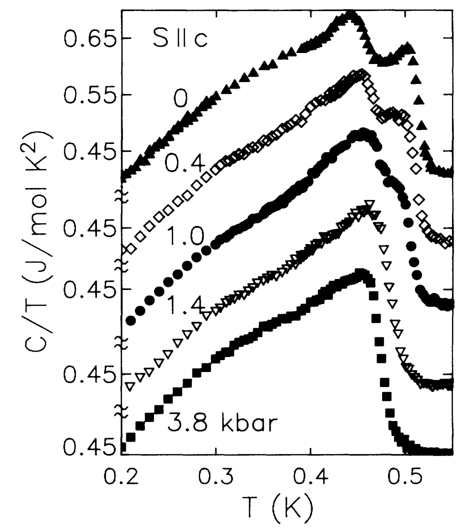

FIG. 2. Double superconducting transition as a function of uniaxial stress parallel to $\hat{c}$. Here, the upper transition moves down in T with increasing S, but the lower transition moves up. The merged single transition then slowly moves down in T with further increase in S.

$[S = \int (C/T)dT]$ shifts out of the peaks, but the phase diagram is topologically isotropic with respect to field direction.

We show in Fig. 2 the response of the double superconducting transition to a series of uniaxial stresses applied perpendicular to the basal plane, $S \| \hat{c}$. Here, the upper transition moves to lower T with increasing S, as it did with H, but the lower transition moves to higher T with increasing S. The two transitions can no longer be resolved at 1.5 kbar, but actually may merge at slightly higher S in light of the narrower transition width at 2, 3, or 4 kbar. The cascade of curves is offset in this plot because of the way in which the Sommerfeld constant γ changes with S (discussed below).

We compare in Fig. 3 the effect of applying stress in the basal plane, $S \| \hat{a}$. Both transitions move to marginally lower T with increasing S, but the major impact of the stress is to broaden the transitions. By 4 kbar the two transitions are almost smeared beyond recognition, although the upper transition onset temperature has remained essentially unchanged [12]. We do not believe that the broadening observed here is due to gradients in the stress field for two reasons: (i) Results on a number of different samples of different geometries with $S \| \hat{a}$ all map onto Fig. 3; and (ii) data obtained using the identical technique for $S \| \hat{c}$ (Fig. 2) do not show any broadening. A zero-stress recheck for samples separately stressed along $\hat{a}$ and $\hat{c}$ above 2 kbar showed no evidence for irreversible effects or the presence of residual strains.

The S-T phase diagrams for uniaxial stress perpendicular and parallel to the basal plane, respectively, are plotted in Fig. 4. We take the midpoint of the jump in C/T vs T to define T_c. The error bars reflect the width of the jump and for $S \| \hat{a}$ all the data are within the error bars because of the smearing of the transition.

These data provide a plausible explanation for why some UPt_3 crystals have one broad superconducting specific-heat peak while others have two, and why annealing to remove strains is an important step in preparing samples with two narrow transitions. From Fig. 3 it is clear that strains left in the basal plane during sample preparation can convert two superconducting peaks into one. It is also clear that this process can occur without significant modification of either the crystal's T_c onset or

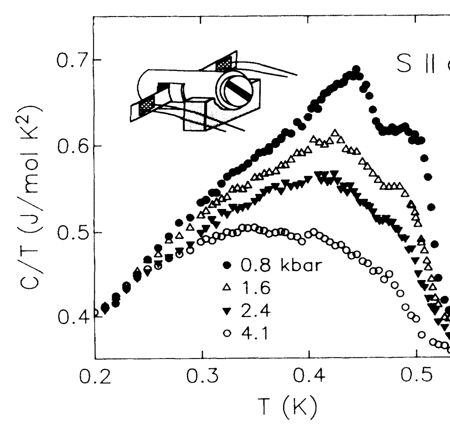

FIG. 3. Double superconducting transition as a function of uniaxial stress in the basal plane, parallel to the a axis. The major effect of stress is to smear the transitions; the onset T_c's barely move.

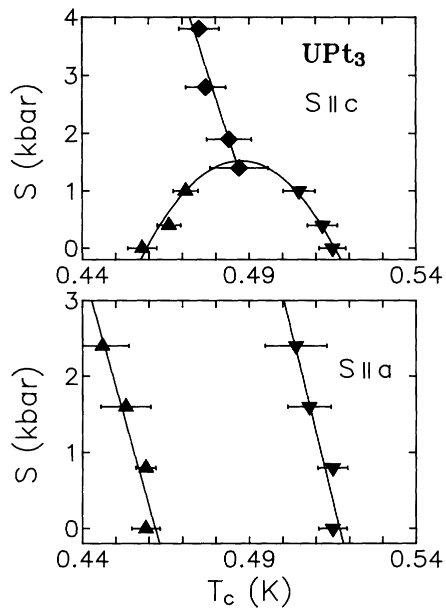

FIG. 4. Axially anisotropic stress-temperature phase diagrams for the superconducting double transition.

the value of the extrapolated residual linear specific heat $\gamma_0 = C/T|_{T=0}$, both commonly taken as measures of sample quality [7,13]. The fact that the low-temperature ($T < 300$ mK) specific heat is independent of S means that the $T \to 0$ power-law behavior and, hence, the placement and number of nodes in the gap, is not changed through the application of uniaxial stress.

The double transition in UPt_3 has been explained in terms of a degeneracy lifted by a coupling of the superconducting order parameter to the coexisting [14] antiferromagnetism, which breaks the hexagonal symmetry and can be described in terms of various one- [10] and two-dimensional [15–19] representations of the hexagonal crystal (D_{6h}). In the context of the Ginzburg-Landau theory developed for the two-dimensional representation, the transition temperatures are given by $T_{c1} = T_{c0} + \tau$ and $T_{c2} = T_{c0} - (\beta_1/\beta_2)\tau$. Here, T_{c0} is the transition temperature in the absence of antiferromagnetism, β_1 and β_2 are the Ginzburg-Landau coefficients of the quartic terms, and τ is proportional to both the strength of the antiferromagnetic order parameter and its coupling to the superconducting order parameter. The ratio β_1/β_2 can be determined from the specific-heat jumps at the two transitions, where

$$\frac{\Delta C_1}{\Delta C_2} = \frac{T_{c2}}{T_{c1}} \frac{\beta_1}{\beta_1 + \beta_2}.$$

We find $\beta_1/\beta_2 = 0.31 \pm 0.05$ for $S = 0$, giving $\tau = 13$ mK and $T_{c0} = 502$ mK. By comparison, other values reported from specific-heat measurements include $\beta_1/\beta_2 = 0.5$, $\tau = 19$ mK [20] and $\beta_1/\beta_2 = 0.15$, $\tau = 8$ mK [16], while BCS weak-coupling theory predicts $\beta_1/\beta_2 = 0.5$.

Given the phase diagram of Fig. 4, we now can extrapolate back from high stress where the transitions have merged to find the experimental value $T_{c0}(S=0) = 495 \pm 10$ mK. This value agrees well with the prediction from the two-dimensional representation of the superconductivity, but disagrees with the one-dimensional, odd-parity representation [10] prediction where $T_{c1} = T_{c0} + 2\tau$ and $T_{c2} = T_{c0} - (3\beta_2 - \beta_1)\tau/2\beta_2$. With $\beta_1/\beta_2 = 0.31$, we find $\tau = 31$ mK and $T_{c0} = 454$ mK, below both transition temperatures.

The interaction of the magnetic and superconducting order parameters also can explain the smearing of the transitions which we observe exclusively for $S \parallel \hat{a}$. The antiferromagnetism and the uniaxial stress couple similarly to the superconductivity in such a way as to lower the symmetry to orthorhombic. However, the UPt_3 crystal contains many antiferromagnetic domains with the spins lying in the basal plane [14]. For each such domain, the stress in the basal plane distorts the lattice in a random direction relative to the magnetic vector, thereby adding a random weight to any coupling term between the antiferromagnetic and superconducting order parameters and providing a natural broadening mechanism.

A recent study of the specific heat of polycrystalline UPt_3 under hydrostatic pressure [20] finds that both superconducting transitions move to lower temperature with increasing pressure, merging at $p \approx 3.7$ kbar and $T \approx 420$ mK. It is difficult to imagine a combination of our data for $S \parallel \hat{c}$ and $S \parallel \hat{a}$ to match the hydrostatic pressure results. The differences may lie in the manner by which the c/a ratio can be altered by uniaxial stress, but not by hydrostatic pressure or, perhaps, with the role of grain boundaries under pressure, present only in the polycrystalline sample.

We demonstrate in Fig. 5 that the normal-state response is also anisotropic with respect to the application of uniaxial stress. For $S \parallel \hat{a}$, we find that the Sommerfeld constant γ decreases with increasing stress at a rate of -13 ± 2 (mJ/mol K^2)/kbar, slightly faster than previous hydrostatic pressure results [21]. For stress perpendicular to the basal plane, we see that γ actually increases over its $S=0$ value, indicating that the stress increases the hybridization so as to enhance the effective mass of the f electrons. There is an initial rise ($0 \leq S \leq 2$ kbar) with $d\gamma/dS = 17 \pm 3$ (mJ/mol K^2)/kbar. For $S > 2$ kbar, where the antiferromagnetic order has been suppressed [22] and the double superconducting transition has merged into one transition, γ remains constant within the error bars. We note that data on a UPt_3 whisker stressed along $\hat{c}$ showed an increase in the coefficient of the T^2 term in the resistivity qualitatively consistent with the increase in γ observed here [23].

In summary, we have measured the specific heat of single-crystal UPt_3 for magnetic field H and uniaxial stress S applied both parallel and perpendicular to the

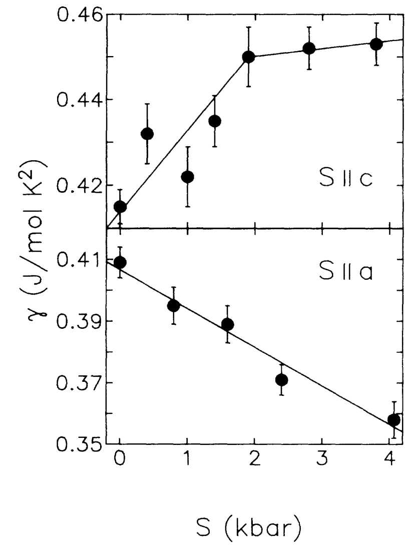

FIG. 5. Sommerfeld constant γ from the normal state also has a distinct directional dependence on uniaxial stress. For S‖ĉ, γ initially increases, then levels off when the two transitions merge. For S in the basal plane, γ decreases at a steady rate with increasing S, as do the transition T_c's. Lines are guides to the eye.

hexagonal basal plane. The superconducting-double-transition phase diagram is topologically isotropic in the H-T plane but axially anisotropic in the S-T plane. The lower and upper superconducting transitions always move to lower T with increasing H, but they move in opposite directions in T with the application of **S**‖ĉ. Data for **S**‖â point to uniaxial strains in the basal plane as an agent which broadens the superconducting transition. Finally, the anisotropic response of the superconductor to uniaxial stress is mirrored by the increase (decrease) of the normal-state Sommerfeld constant γ with **S** perpendicular (parallel) to the basal plane.

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[a]Present address: Center for Particle Astrophysics, The University of California, Berkeley, CA 94704.

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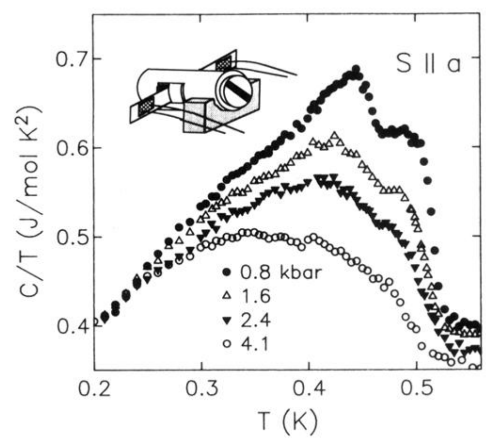

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