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Pressure dependence of the superconducting phases in UPt_3

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We report measurements of the specific heat of the heavy-fermion compound UPt_3 under hydrostatic pressure p up to 4.5 kbar. The two superconductive transitions at temperatures T_c^+ and T_c^- observed previously under zero pressure decrease at a rate of $dT_c^+/dp = -24$ mK/kbar and $dT_c^-/dp = -5$ mK/kbar. The transitions merge at $p^* = 3.7$ kbar and $T^* = 419$ mK. These results suggest that the weak antiferromagnetic order, which is suppressed on the same pressure scale, acts as a symmetry-breaking field in splitting the superconductive transition, thus yielding convincing evidence for a non-BCS order parameter for this system.

Superconductivity in the presence of strong electronic correlations has been a fascinating subject of solid-state physics for more than a decade.[1] One of the issues of importance is whether or not materials exhibiting this kind of unusual superconductivity—notably heavy-fermion compounds and high-T_c oxides—can be described within the standard BCS theory of superconductivity. Recently, interest has focused on the heavy-fermion compound UPt_3 that exhibits many features pointing to a non-BCS-like pairing mechanism. From a variety of experimental evidence, a multiplicity of superconducting phases has been inferred (for a recent review see Ref. 2). Particularly compelling are the two distinct discontinuities observed in the specific heat[3–5] and thermal expansion[5] in the zero applied magnetic field. The two transitions converge with the application of a magnetic field of 0.5 T for $\mathbf{B}\perp c$ (Ref. 4) and 0.8 T for $\mathbf{B}\parallel c$.[5] These data have been interpreted[6–10] in terms of a Ginzburg-Landau theory with a two-component-vector order parameter which is coupled to a symmetry-breaking field (SBF). In these models, the SBF lifts the twofold degeneracy of the superconductive transition in the same way that a magnetic field lifts the twofold degeneracy of superfluid ^{3}He phase A. This mechanism requires a superconducting *vector* order parameter, as opposed to a BCS order parameter where an SBF may lead to a modulation of the superconducting state as observed in certain Chevrel phases[11] but not to a multiplicity of superconducting phases.

An obvious possibility for an SBF in UPt_3 is the weak antiferromagnetic order developing in UPt_3 below $T_N = 5$ K with the very weak moments ($\approx 0.02\mu_B$/U atom) aligned in the basal plane of hexagonal UPt_3.[12] Very recently, it has been shown that this magnetic order is suppressed upon application of a moderate external pressure p, with the intensity of the antiferromagnetic $(0,\frac{1}{2},1)$ Bragg-peak measured at 2 K dropping by more than a factor of 2 for $p = 1.8$ kbar.[13] If the weak antiferromagnetism of UPt_3 is indeed responsible for splitting the superconductive transition, as has been suggested on the basis of the pressure dependence of the upper critical field[13,14] and also on the basis of neutron diffraction in applied magnetic fields,[15] one would expect that the two superconductive transitions may eventually collapse into a single transition under pressure.

In this paper, we report on high-resolution heat-capacity measurements of UPt_3 under pressure which allow a determination of the (p,T) superconductive phase diagram. The two superconductive transitions are clearly resolved for low pressures p and merge together at $p = 3.7$ kbar. In the light of the above discussion, this yields strong experimental support of the idea that the two transitions are due to a coupling between superconducting and antiferromagnetic order parameters. Thus, our results constitute yet another piece of evidence for an unconventional superconducting state in UPt_3.

The sample used in the present study is a small polycrystalline cylinder (diameter 2.6 mm, length 5 mm) which was cut from a larger specimen previously investigated.[3] The sample was pressurized in a Cu-Be cell with a methanol-ethanol mixture as a pressure-transmitting medium. A superconducting Sn platelet served as a pressure gauge. The heat capacity of the whole cell was measured with the standard heat-pulse technique for temperatures T between 0.2 and 1.5 K. The cell contribution to the heat capacity was determined in separate runs for several pressures (with a Cu dummy replacing the sample) and was subtracted to yield the heat capacity of the sample. The latter amounted to between 7% and 10% of the total heat capacity. More experimental details can be found elsewhere.[16]

Figure 1 shows the specific heat C in the vicinity of the superconductive transitions for four different pressures plotted as C/T vs T. For low pressures, the two transitions previously observed for $p = 0$ are clearly resolved. Our zero-pressure data are in good agreement with the earlier data of Hasselbach, Taillefer, and Flouquet.[4] With increasing p the upper transition at T_c^+ shifts to lower temperatures much faster than the lower one at T_c^-. At 2.4 kbar, the two transitions can just barely be resolved, until finally at 4.5 kbar they clearly have merged together. It is worth pointing out that the transitions do not become broader with increasing p. If anything, they appear even to sharpen up a little with a width (between 10% and 90%) of only 7 mK for the transition in 4.5 kbar. This indicates that the pressure in the cell is hydrostatic. As an aside we note that the pressure dependence of the normal-state linear specific-heat coefficient γ, i.e., a linear decrease at a rate of -12 mJ mol^{-1}K^{-2}/kbar, agrees very well with the earlier data of Brodale *et al.*[17]

Another feature readily apparent from Fig. 1 is that the ratio of the two transition heights at T_c^+ and T_c^- remains roughly constant, although the overall transition height does decrease a little. The total discontinuity ΔC_{tot}^- at T_c^- (normalized to the normal-state specific heat) decreases from $\Delta C_{\text{tot}}^-/\gamma T_c^- = 0.71$ ($p=0$) to 0.60 ($p=2.4$ kbar). The specific-heat discontinuities and the transition temperatures are obtained in the usual way, i.e., by replacing the observed transition of finite width by an ideal sharp transition keeping the entropy constant.

The (p,T) phase diagram derived from the data of Fig. 1, and from additional data not shown, is displayed in Fig. 2. We extract $dT_c^+/dp = -(24 \pm 5)$ mK/kbar and $dT_c^-/dp = -(5 \pm 1)$ mK/kbar. The two transitions merge at approximately $p^* = 3.7$ kbar and $T^* = 419$ mK assuming a linear pressure dependence for both. The data for dT_c^+/dp are very close to the T_c data obtained from resistivity measurements under pressure on whiskers[18] ($dT_c^+/dp = -24$ mK/kbar for $p < 1.5$ kbar) and from ac-susceptibility measurements under uniaxial stress[19] up to 0.8 kbar where $dT_c/d\varepsilon = -26$ mK/kbar for strain ε along the c axis and no change in T_c was found for ε along a or b (see Ref. 14). Hence our results unambiguously show that the onset temperature T_c measured by these authors[18,19] is indeed to be associated with the upper transition at T_c^+. However, a discrepancy should be noted. The data on whiskers[18] suggest $p^* \sim 2$ kbar, which is smaller than p^* determined directly from our (p,T) phase diagram by a factor of 2. In view of the completely independent sets of experiments, an error in the pressure determination appears unlikely. Rather, the role of different samples (bulk and whisker) should receive close scrutiny, as well as the possible presence of uniaxial strains.

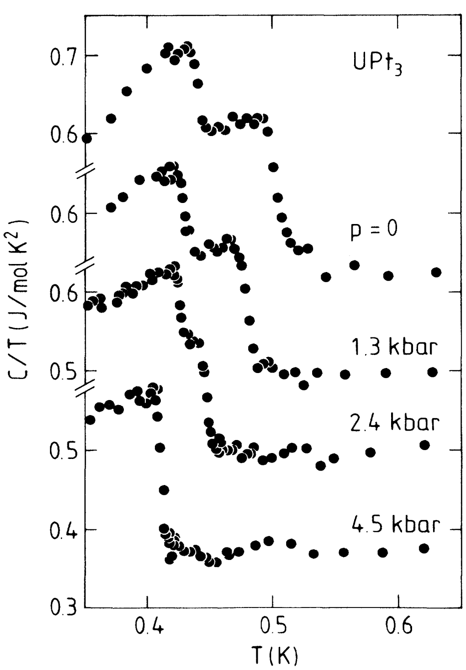

FIG. 1. Specific heat C divided by temperature T of UPt_3 vs T in the vicinity of the superconductive transition for different pressures p.

We can further compare the pressure dependences of T_c^+ and T_c^- with the prediction of the Ehrenfest relation $(dT_c/dp)_{p=0} = V_m T_c \Delta\alpha_v/\Delta C$ where V_m is the molar volume, $\Delta\alpha_v$ is the discontinuity of the volume thermal-expansion coefficient at T_c, and ΔC the corresponding jump in the specific heat. The pressure dependence of T_c^+ derived[5] from the discontinuity $\Delta\alpha_v^+$ at the upper transition is $dT_c^+/dp = -(20 \pm 3)$ mk/kbar, in good agreement with our results. From our data for dT_c^-/dp and ΔC^-, we expect a discontinuity at the lower transition $\Delta\alpha_v^- \approx -1.5 \times 10^{-7}$ K^{-1}. This *negative* jump is seven times smaller than the positive one observed at T_c^+ (Ref. 5) and results from a near compensation of $\Delta\alpha_\parallel$ and $2\Delta\alpha_\perp$, the anomalies for expansion parallel and perpendicular to the c axis. In conclusion, the present data in conjunction with earlier work, yield a coherent thermodynamic description of the pressure dependence of the two superconducting phases in UPt_3.

Our results convincingly support the notion of a symmetry-breaking field that lifts the degeneracy of the superconductive transition by breaking the hexagonal symmetry of UPt_3. An external pressure of 3.7 kbar appears to restore the degeneracy that exists in the absence of an SBF. Hess, Tokuyasu, and Sauls[7] and Machida, Ozaki, and Ohmi[8] have given a detailed phenomenological description based on symmetry arguments of the coupling of the superconducting order parameter, assumed to belong to the E_{1g} representation, to an SBF in UPt_3 arising from spatially homogeneous antiferromagnetic order.[8] Very recently, Joynt *et al.*[10] considered a model of a randomly oriented antiferromagnetic order parameter, in which the symmetry is broken only locally; this takes account of the fact that the antiferromagnetic domains in UPt_3 are

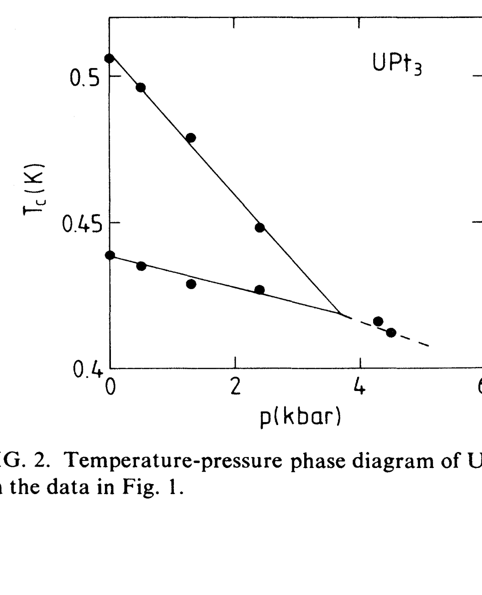

FIG. 2. Temperature-pressure phase diagram of UPt_3 derived from the data in Fig. 1.

small,[15] of a size comparable to the superconducting coherence length. In all these models, the presence of the ordered phase at T_c^+ affects the formation of the low-T phase if the ratio of the two coefficients β_1 and β_2 of the quartic terms in the Ginzburg-Landau free energy for a two-dimensional superconducting order parameter differs from unity. These coefficients are readily determined from the specific-heat discontinuities ΔC^+ and $\Delta C_{\rm tot}^-$, $\Delta C_{\rm tot}^-/\Delta C^+ = (1+\beta_2/\beta_1)(T_c^-/T_c^+)$. From our data, $\beta_2/\beta_1 = 0.49$ for $p=0$, close to the weak-coupling limit 0.5 and in good agreement with other measurements.[4,5] Furthermore, it increases only slightly with pressure, reaching $\beta_2/\beta_1 = 0.64$ at $p=2.4$ kbar. The strength of the SBF is given by the parameter $\tau(p)$ obtained from $T_c^+(p) = T_c^0(p) + \tau(p)$ and $T_c^-(p) = T_c^0(p) - (\beta_1/\beta_2)\tau(p)$.[7,8] Our analysis based on Fig. 2 yields $\tau(p) = (22-6\ p)$ mK where p is measured in kbar, and $dT_c^0/dp = -18$ mK/kbar which is somewhat larger than observed[18,20] at high pressures $p > p^*$. The $p=0$ value $\tau(0)$ is in reasonable agreement with $\tau = 18$ mK estimated independently from the kink in the measured upper critical field.[7]

As to the microscopic origin of the SBF, we note that the critical pressure p^* determined in the present work is close to the pressure of ~ 3 kbar where the integrated intensity of the $(0,\frac{1}{2},1)$ antiferromagnetic Bragg-peak measured at 2 K extrapolates to zero.[13] This coincidence is far from circumstantial since these pressures are low compared to the pressures needed to cause appreciable changes either in the normal-state properties [application of 3 kbar decreases γ by 7% (Ref. 17) and the T^2 coefficient of the electrical resistivity by 9% (Ref. 20)] or, for that matter, in T_c itself. The argument can be made quantitative by noting that the magnetic Bragg intensity I depends linearly on T for $p=0$ (Ref. 12) and linearly on p for fixed temperature $T=2$ K (Ref. 13). If dI/dT is assumed to be independent of p at low p, a linear dependence of T_N on p follows, and $T_N \sim 0.5$ K at 4 kbar. This extrapolated pressure agrees very well with p^*, and the case of a coupling between the superconducting and antiferromagnetic order parameters is thereby considerably strengthened. Whether this coupling operates on a macroscopic scale—thus breaking the overall hexagonal symmetry of a crystal[6-8]—or more locally with a random orientation, thus preserving the overall symmetry,[10] remains to be determined. The rather isotropic behavior of both the upper and lower critical fields of UPt_3 would seem to favor the latter.[2,5]

In conclusion, we have established firm experimental evidence that the two superconductive transitions in UPt_3 result from a splitting of a twofold-degenerate transition arising from a symmetry-breaking field whose origin is the weak antiferromagnetic order. Upon application of external pressure both the splitting of the superconductive transition and the antiferromagnetic order disappear at roughly the same critical pressure. However, the detailed mechanism of the splitting and the precise representation to which the order parameter belongs are not yet firmly established. Of course, the origin of superconductivity itself in UPt_3 remains to be understood.

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