

# The superconducting phases of $\text{UPt}_3$ under pressure

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The upper critical field  $H_{c2}(T)$  of the heavy fermion superconductor  $\text{UPt}_3$  for a magnetic field applied in the basal plane has been studied down to 150 mK and under hydrostatic pressure up to 10 kbar. The kink already observed in  $H_{c2}(T)$  at ambient pressure disappears by applying a relatively low pressure. This suggests that the high-temperature low-field superconducting phase reported recently is rapidly suppressed by pressure. A possible interpretation of these results is discussed in connection with recent theories.

## I. INTRODUCTION

The existence of the multicomponent superconducting phase diagram proposed recently for  $\text{UPt}_3$  (Ref. 1) was supported by evidence from ultrasonic attenuation<sup>2</sup> and specific-heat<sup>1,3</sup> measurements, which revealed anomalies in these physical properties within the superconducting state. Moreover, the proposed phase diagram offered a natural explanation for the observed kink at  $H^* = 4$  kOe in the upper critical field<sup>4,5</sup> for a field directed in the basal plane. For such a configuration, the above-mentioned experimental results suggest that the normal phase of  $\text{UPt}_3$  condenses into two distinct superconducting phases depending on the field strength, i.e., whether  $H < H^*$  or  $H > H^*$ .<sup>6</sup> One possible consequence of this distinction would be a different response of the two hypothetical superconducting phases to the variation of an external parameter. Given the large sensitivity of the transition temperature  $T_c$  to pressure in heavy fermion systems in general, and in  $\text{UPt}_3$  in particular,<sup>7</sup> hydrostatic pressure appeared to be a suitable probe to reveal the multiplicity of the superconducting phases.

We present here measurements of the upper critical field  $H_{c2}$  of  $\text{UPt}_3$  under hydrostatic pressure which show that the critical temperature behaves in a strikingly different manner for fields below and above  $H^*$ . Moreover they indicate that the kink in  $H_{c2}(T)$  disappears when the applied pressure exceeds 1.4 kbar, a relatively small value, suggesting that the high-temperature low-field phase is rapidly suppressed by pressure.

## II. EXPERIMENT

Two series of resistivity measurements were performed on two monocrystalline whiskers to determine  $H_{c2}(T)$  in the basal plane for various pressures. The results on both samples (labeled 7 and 8) were found to be similar, and we shall describe here primarily those obtained on whisker No 8. This whisker was made by spontaneous growth from a rapidly quenched melt kept in a water-cooled copper crucible inside a UHV furnace. Its dimensions are approximately  $15 \mu\text{m} \times 150 \mu\text{m} \times 1 \text{mm}$ . The resistivity was measured using an ac bridge. The current and potential electrodes consisted of  $25\text{-}\mu\text{m}$  gold wires and four indium contacts to the sample. The pressure was obtained by using a hydraulic pressure at room temperature in a Cu-Be clamped piston and cylinder device. The superconducting critical temperature of a Sn bar

was used for determining the pressure. The pressure cell was cooled by solid contact to the cold finger of a dilution refrigerator designed and built at the Centre de Recherches sur les Très Basses Températures in Grenoble. The magnetic field was generated by a superconducting solenoid. A Matsushita carbon resistance installed in the compensated zone of the magnet was used for thermometry. Given the low thermal conductivity of Cu-Be special attention was given to the thermalization of the sample via the copper wires going to the four electrodes. For all measurements, a current density of  $0.3 \text{ A cm}^{-2}$  was used. The electrical resistivity at zero field was found to show an initial drop at a higher temperature just before the full superconducting transition. This effect, observed at all pressures, is reminiscent of what was reported in Ref. 5. The application of a magnetic field of 500 G destroyed this initial drop and sharp transitions ( $\Delta T = 5\text{--}7 \text{ mK}$ ) under magnetic field were observed. Although this zero-field effect may be intrinsic to  $\text{UPt}_3$ , one can not exclude the possibility of an inadequate thermalization due to the superconductivity of the indium contacts below 300 G. However this did not modify the results, since  $T_c$  was defined as the temperature by which half of the total fall in resistance was fulfilled, which was always within the sharp drop. Given the relatively small pressures applied, the possible built-in strains of the sample could have a nonnegligible influence on the onset of superconductivity.

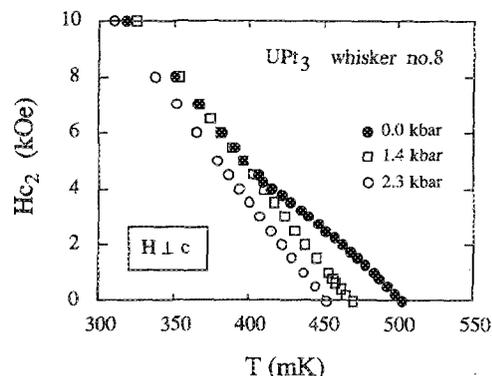


FIG. 1. Temperature dependence of  $H_{c2}(T)$  for three different values of the applied pressure and for a field in the basal plane. Note the complete disappearance of the kink at  $H = H^* = 4$  kOe for  $P > 1.4$  kbar.

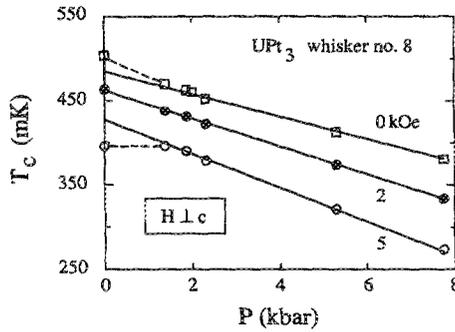


FIG. 2. Pressure dependence of the transition temperature ( $T_c$ ) for three values of magnetic field in the basal plane.  $T_c$  is defined as the middle of the drop in resistivity between the normal state and the superconducting regime. Note the deviations from linearity at pressures below  $P_c = 1.4$  kbar, upwards (by 20 mK) at  $H = 0$  and downwards (by 27 mK) at  $H = 5$  kOe. The disappearance of these opposite deviations at  $P = P_c$  suggests a symmetric collapse of the two transitions in zero field (see text).

### III. RESULTS

The principal results are presented in three Figs. 1–3. Figure 1 shows the  $H_{c2}(T)$  for several different pressures. The distinct kink at ambient pressure is seen to disappear under the lowest applied pressure. At ambient pressure and for  $H < H^*$  we observe a slight nonlinearity in  $H_{c2}(T)$  which has not been reported in other samples. The ratio of the slopes above and below  $H^*$  is comparable to what has already been reported. The low  $T_c$  (504 mK) at ambient pressure compared to a relatively small residual resistivity (0.45  $\mu\Omega$  cm) suggests that the correlation between these two quantities is not as simple as previously thought.<sup>5</sup> The disappearance of the kink was also seen in sample 7 for which, however, the lowest investigated pressure was 3.9 kbars.

To illustrate the difference in the behavior of  $T_c$  below and above  $H^*$  we present  $T_c(P)$  curves for selected fields in Fig. 2. The figure shows that for  $P < 2$  kbar, while  $T_c$  falls drastically in the presence of a small field ( $< H^*$ ) it hardly changes at all for fields just above  $H^*$  and it increases for  $H > 5$  kOe. For pressures greater than 2 kbar we find a linear decrease of  $T_c$  for all fields up to 10 kOe with a slope  $-(dT_c/dP)$  which gets more pronounced with increasing field. This can roughly be expressed as

$$-dT_c/dP(H) = -dT_c/dP(H=0) + \beta H.$$

The corresponding values of  $dT_c/dP(H=0)$  and  $\beta$  for the two samples are given in Table I.

TABLE I. Comparison of the kink in  $H_{c2}(T)$  at ambient pressure for the two samples studied. The effect of the pressure on zero-field transition temperature (for pressures exceeding 2 kbars) is compared with the results reported by Willis *et al.*<sup>7</sup>  $\beta$  is a parameter indicating the influence of the magnetic field on the slope of  $dT_c/dP$  (see text).

	$T_c$ (mK)	$\rho_0$ ( $\mu\Omega$ cm)	$H^*$ (kOe)	$-H'_{c2}(T)(P=1 \text{ bar})$		$-dT_c/dP(H=0)$ (mK/kbar)	$\beta$ (mK/kOe bar)
				$H > H^*$	$H < H^*$		
				(kOe/K)			
Sample 7	537	0.5	3.5	63	43	11.3	1.0
Sample 8	504	0.45	4	63	(45)	13.2	1.2
Ref. 5	490	0.5	...	...	...	12.6	...

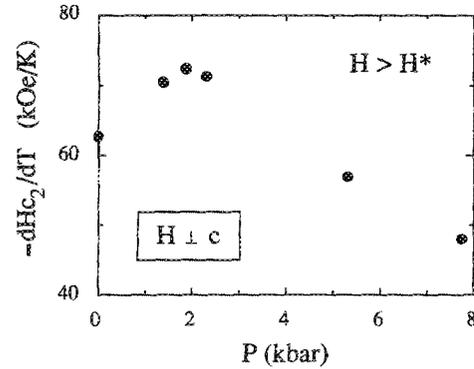


FIG. 3. Variation of the slope of  $H_{c2}(T)$  with pressure for the high-field ( $H > H^*$ ) phase.  $H^*$  is assumed to be zero for  $P > 1.4$  kbar.

Figure 3 shows the variation under pressure of the slope of  $H_{c2}(T)$  for fields exceeding  $H^*$  (in fact, for any field  $H$  as soon as  $P > 1.4$  kbar). A linear behavior of  $H_{c2}(T)$  was observed at least up to 12 kOe and  $dH_{c2}/dT$  was thus easily and clearly defined. While  $-(dH_{c2}/dT)$  falls rapidly when  $P > 2$  kbar it rises somewhat surprisingly for low pressures. This initial rise is associated with a noticeable increase in  $T_c$  at high fields. A reasonable extrapolation of our data to lower temperatures implies an initial rise and a maximum in  $H_{c2}(0)$  under pressure. However a detailed investigation of the low-temperature, high-field phase boundaries is needed to confirm this.

For the higher pressures ( $P > 2.3$  kbar), the fall of  $T_c$  and that of  $-(dH_{c2}/dT)$  with pressure may be related through the Ginzburg–Landau formula for clean superconductors,

$$H'_{c2}(T) \propto \gamma^2 T_c. \quad (1)$$

The data yields  $\Delta T_c/(T_c \Delta P) = -25 \text{ Mbar}^{-1}$  and  $\Delta H'_{c2}(T)/H'_{c2}(T \Delta P) = -66 \text{ Mbar}^{-1}$  so that by taking  $\Delta \gamma^2/(\gamma^2 \Delta P) = -43 \text{ Mbar}^{-1}$  from Ref. 8, the correlation expressed by Eq. (1) is found to be essentially correct.

### IV. DISCUSSION

The principal and the most obvious outcome of this experiment is the high-pressure sensitivity of the low-field, high-temperature superconducting phase of  $\text{UPT}_3$  and its suppression (below  $T_c$ ) by a relatively low pressure. The residual phase, however, cannot be identified as the ambient pressure high field phase (B phase in the phase diagram in

Ref. 1) because the  $T_c$  behavior for pressures exceeding 2 kbars falls linearly with pressure for all fields, while in low pressures it increases for fields above  $H^*$ .

Recently Hess, Tokuyasu, and Sauls<sup>9</sup> and Machida, Ozaki, and Ohmi<sup>10</sup> have suggested a possible coupling between a symmetry-breaking field (SBF), probably the antiferromagnetic long-range order observed below  $T_N = 5$  K, and the superconducting order parameter. They use this coupling to explain the splitting of the superconducting transition in  $\text{UPt}_3$ . Their calculations, both similar and along the lines of Joynt's earlier work,<sup>12</sup> show that such a coupling can produce a double transition in zero field with a one-dimensional order parameter appearing at the higher temperature transition and a two-dimensional one with the full symmetry of the crystal below the lower critical temperature. The kink in  $H_{c2}(T)$  for a definite direction in the basal plane is a consequence of a competition between the coupling energy of the vector order parameter with the magnetic field on the one hand and the SBF on the other when the two fields are perpendicular to each other.

A rather simple way to interpret our results in the framework of this theory is to attribute the disappearance of the  $H_{c2}(T)$  kink to a possible suppression of the AFM order under pressure. At zero field, the two superconducting transitions are expressed as<sup>9,10</sup>  $T_{c1} = T_{c0} + \tau$  and  $T_{c2} = T_{c0} - (\beta_2/\beta_1)\tau$ , where  $T_{c0}$  is the critical temperature in the absence of a symmetry breaking field,  $\tau$  is a parameter reflecting the strength of that field (associated to the AFM order), and  $\beta_1$  and  $\beta_2$  are the coefficients of the quartic terms in the Ginzburg-Landau expansion of the superconducting free energy. Now,  $T_{c0}$  is assumed to be a decreasing function of pressure, and that  $\tau$  (which, according to Ref. 10, is proportional to  $M^2/T_N^2$ ,  $M$  being the sublattice magnetization) may be expected to vary with pressure. A fall of  $\tau$  with increasing pressure would make  $T_{c1}$  to go down even faster than  $T_{c0}$ . On the other hand  $T_{c2}$  would decrease less and it can even go up if the pressure variation of  $\tau$  exceeds  $-(dT_{c0}/dP)$ . When at a critical pressure  $\tau$  completely disappears (a destruction of AFM order) the two transitions will be reunified and we will have a unique phase. Our experimental observations seem to agree with this picture.  $T_{c1}$  is found to fall rapidly in a low-pressure regime, and while there is no direct observation of  $T_{c2}$  and its pressure variation, the constancy or the small increase of high-field critical temperatures indicate that it falls much less rapidly than  $T_{c0}$ . To test this idea quantitatively one must investigate the  $T_c$  variation in a low-pressure region. Nevertheless the linear

fall of  $T_c$  at  $P > 2$  kbar for all fields and the disappearance of the kink in  $H_{c2}(T)$  may be an indication of the absence of symmetry breaking field at higher pressures. Recently Hayden and co-workers<sup>13</sup> have established directly by neutron scattering that the AFM order in  $\text{UPt}_3$  is highly sensitive to small pressures, thus providing strong evidence for the role of AFM in shaping the superconducting phase diagram.

The initial rise in the slope of  $H_{c2}(T)$  (for  $H > H^*$ ) is more difficult to understand. One cannot exclude that this rise is related to a possible pressure dependence of different parameters determining this slope for the two phases.<sup>9,10</sup>

In conclusion, the pressure dependence of  $H_{c2}(T)$  in the basal plane measured in two  $\text{UPt}_3$  whiskers reveals a high sensitivity of the high-temperature, low-field phase to an applied pressure and lend support to the view that the two transitions at  $H = 0$  are intimately related and perhaps the result of a lifted degeneracy as proposed by recent theories.

## ACKNOWLEDGMENTS

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