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Dilatometry study of the superconducting phases of UPt_3

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Magnetostriction and thermal expansion measurements on single-crystalline UPt_3 ($\Delta L \| c$) have been performed for magnetic fields in the basal plane. From these experiments a detailed superconducting phase diagram (field versus temperature) has been constructed. Via the Ehrenfest relations an accurate prediction of the pressure dependence of the phase diagram is obtained.

In recent years the heavy-fermion superconductor UPt_3 ($T_c = 0.5$ K) has attracted considerable attention because of its unconventional superconducting properties. Early measurements showed a power-law behaviour of several thermodynamic quantities at low temperatures as proof of unconventional superconductivity [1]. This argument was weakened, however, by the fact that scattering processes at impurities may play a significant role at low temperatures. Recently, solid evidence for unconventional superconductivity was found by the discovery of additional anomalies in the superconducting state. Measurements of the specific heat [2], sound velocity [3] and thermal expansion [4] clearly established the existence of a second superconducting transition approximately 60 mK below the first transition. Measurements in magnetic field revealed a complex superconducting phase diagram in the B–T plane [3–5], with three superconducting phases meeting in a tetracritical point.

Within the framework of the Ginzburg–Landau approach the complex superconducting phase diagram is generally described by a multicomponent order parameter that interacts with a symmetry-breaking field ([6] and references therein). In most models the small antiferromagnetically ordered moment ($0.02\mu_B$/U-atom) along the hexagonal b-axis of the crystal, detected by neutron diffraction below $T_N = 5$ K [7], is considered to act as the symmetry-breaking field. Although the recently reported Ginzburg–Landau models can explain the superconducting phase diagram to a large extent, they are inadequate at several points [6]. Model calculations have been carried out for the thermal expansion and magnetostriction [8] by taking into account the direct strain–order parameter coupling. The main prediction of these calculations is that the thermal expansion and magnetostriction show steps of equal sign at all the superconducting phase transitions.

As little is known about the lattice deformations caused by the superconducting transitions, dilatation measurements were carried out in the temperature interval $0.05\,\mathrm{K} < T < 0.8\,\mathrm{K}$ and in magnetic fields up to 9 T.

Magnetostriction and thermal expansion measurements were performed on a single-crystalline sample of UPt_3 (dimensions $a \times b \times c \simeq 3 \times 1 \times 2\ \mathrm{mm}^3$), using a sensitive three-terminal parallel-plate capacitance dilatometer [9], for an elongation (contraction) along the c-axis of the crystal and magnetic fields in the basal plane (both a- and b-axis). The linear magnetostriction $\lambda_c = (L(B) - L(0))/L(0)$ was recorded at temperature intervals of 20 mK or less, while slowly sweeping the field (20 mT/min). The coefficient of linear magnetostriction $\tau_c = L^{-1}\,\mathrm{d}L/\mathrm{d}B$ is obtained by differentiating λ_c with respect to B. The coefficient of linear thermal expansion $\alpha_c = L^{-1}\,\mathrm{d}L/\mathrm{d}T$ was measured at field intervals of 0.025 T or more, using a temperature modulation technique [4].

In fig. 1 some typical magnetostriction curves ($B \| b$) are shown. At a magnetic field of 0.1 T a small anomaly is detected, which has a weak temperature dependence and is also observed in the normal phase.

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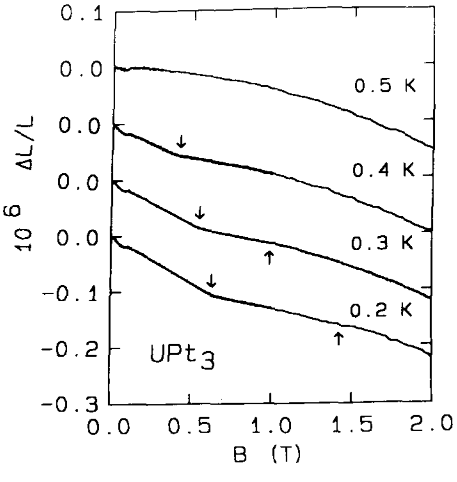

Fig. 1. The magnetostriction of UPt_3 ($\Delta L \| c$) for applied magnetic fields along the b-axis at some characteristic temperatures. The downward arrows indicate the first superconducting transition and the upward arrows the second superconducting transition for increasing fields. At a field of 0.1 T an additional anomaly is observed. The curves are shifted along the vertical axis for clarity.

At higher temperatures the anomaly weakens and vanishes near 4.2 K. The hysteresis at this low-field transition indicates that it is of first order. The superconducting transitions are indicated by small changes of slope in the magnetostriction (step in τ_c) and its derivative (change of slope in τ_c).

The thermal expansion ($\Delta L \| c$) in a magnetic field ($B \| b$) is shown in fig. 2. In zero field two steps of opposite sign are observed at the two superconducting transition temperatures $T_c^- = 0.438(2)$ K and $T_c^+ = 0.493(2)$ K. In applied field the splitting of T_c decreases and eventually only one small anomaly is observed in high fields.

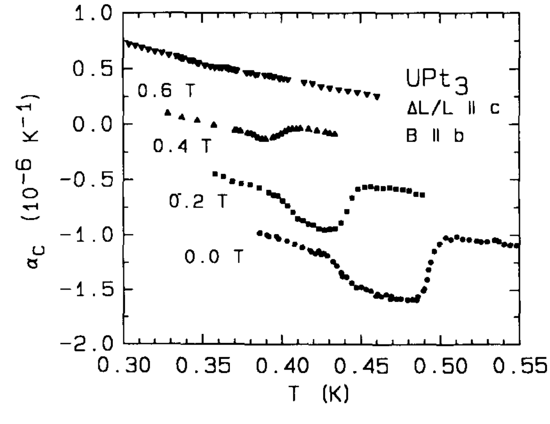

Fig. 2. Thermal expansion of UPt_3 ($\Delta L \| c$) for several magnetic fields along the b-axis (●: 0.0 T, ■: 0.2 T, ▲: 0.4 T and ▼: 0.6 T). The step sizes decrease for increasing fields and above the tetracritical point only one small step is present at the transition. The field curves are shifted along the vertical axis for clarity.

Not all of the superconducting phase lines were detected by the magnetostriction and the thermal expansion measurements, but by combining both techniques a detailed phase diagram was constructed, as shown in fig. 3. The four superconducting phase lines meet at a tetracritical point with $T_{cr} = 0.387(3)$ K and $B_{cr} = 0.443(5)$ T. No anisotropy of the phase diagram was observed for fields along the a or b-axis in the basal plane. In low fields a clear change in the slope of the phase line of T_c^- is detected near 0.1 T. This change in slope coincides with the low-field anomaly in the magnetostriction.

The absence of hysteresis and latent heat at the superconducting transitions in the thermal expansion and magnetostriction suggests that the superconducting phase transitions are of second order. Using the stability criterion for a tetracritical point of four second-order phase lines of Yip et al. [10], we found no indications of a thermodynamically instable tetracritical point [11]. For second-order phase transitions the uniaxial pressure dependence of the B–T phase diagram can be determined by applying one of the Ehrenfest relations:

$$\left(\frac{T}{p_c}\right)_{Bp_{a,b}} = \frac{V_m \Delta\alpha_c}{\Delta(C_p/T)} \,. \qquad (1)$$

For a given molar volume ($V_m = 4.24 \times 10^{-5}$ m^3 mol^{-1}), the steps in the specific heat (divided by

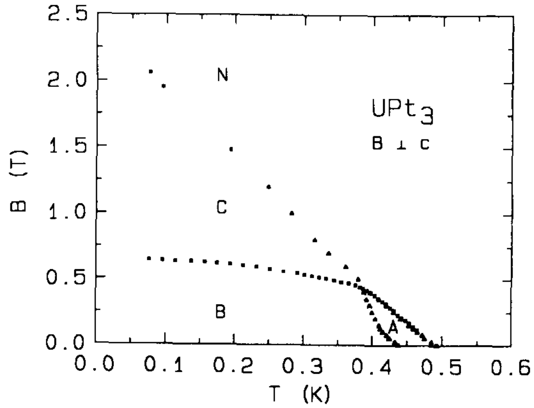

Fig. 3. Superconducting phase diagram of UPt_3 for fields in the basal plane constructed from the measurements of α_c and τ_c. For the low-temperature transition a change in slope is observed in low fields, which coincides with an anomaly in the magnetostriction.

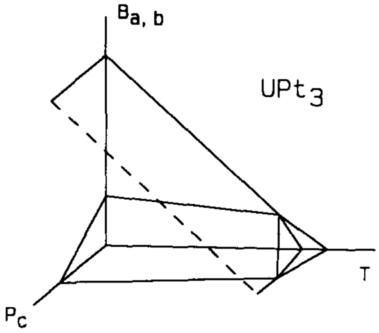

Fig. 4. The calculated uniaxial pressure dependence ($p \| c$) of the $B-T$ phase diagram for magnetic fields in the basal plane.

temperature) $\Delta(C_p/T)$ and the thermal expansion $\Delta\alpha_c$, determine the pressure dependence along the c-axis of the critical temperature. Although not all of these steps are known, they can be calculated using other Ehrenfest relations which relate them to the steps in the magnetostriction $\Delta\tau_c$, the compliance Δc_{33} and the measured slopes of the phase lines in the $B-T$ plane [11].

The calculated uniaxial pressure dependence of the $B-T$ phase diagram is schematically shown in fig. 4, where a linear extrapolation of the initial pressure dependence is used. The tetracritical point is suppressed by uniaxial pressure along the c-axis and vanishes at $T_{cr} = 0.460$ K and $p_{cr} = 2.5$ kbar. A tetracritical point is also observed in the p_c-T plane, in accordance with the stability criteria formulated by Yip et al. [10]. These results are in good agreement with recent specific heat measurements under uniaxial pressure [12].

The observed steps in the thermal expansion in zero field have opposite sign, in sharp contrast with earlier mentioned model calculations [8] of direct strain–order parameter coupling, that predict the steps to have equal sign. Within the model the measured step sizes can only be explained by taking into account a strong and anisotropic strain dependence of the symmetry-breaking field. Therefore, the thermal expansion measurements impose serious constraints on the proposed Ginzburg–Landau models [11].

The low-field anomaly in the magnetostriction and the phase diagram at $B \simeq 0.1$ T can possibly be explained by the effects of domain orientation in field. As the antiferromagnetic order in zero field has a limited correlation length (~250 Å) [7], magnetic domains oriented along the different hexagonal b-axes are formed. At low fields an averaging over the domain orientations leads to nearly parallel phase lines, while at higher fields ($B > 0.1$ T) the domains tend to orient perpendicular to the field in the basal plane, resulting in a change of slope of the phase line of T_c^- [13]. Further measurements are needed to clarify this point.

In summary, dilatometry studies on UPt_3 revealed a superconducting phase diagram ($B \perp c$) of unprecedented accuracy. As the superconducting transitions are of second order the Ehrenfest relations determine the pressure dependence of the $B-T$ phase diagram. The tetracritical point is suppressed under uniaxial pressure along the c-axis and vanishes at a critical pressure of $p_{cr} \simeq 2.5$ kbar.

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