Expansivity of the superconducting phases of UPt_3

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The superconducting phase diagram of heavy-fermion UPt_3 has been determined with the use of a sensitive dilatometric technique. Discontinuities observed in the coefficients of the linear thermal expansion (α) and magnetostriction (τ) at the phase lines yield the uniaxial pressure dependence of the various phases via the Ehrenfest relation. The results yield important constraints on the analysis within a Ginzburg-Landau approach.

The heavy-fermion superconductor UPt_3 ($T_c=0.5$ K) attracts much attention because of its unconventional superconducting properties. Recently, solid evidence for an unconventional (nonsinglet) superconducting (SC) order parameter has come to the fore by the discovery of additional anomalies in the thermodynamic properties of the superconducting phase. Measurements of the specific heat,[1] sound velocity,[2] and thermal expansion[3] clearly identified a second phase transition that takes place ~60 mK *below* T_c. Moreover, measurements in an external magnetic field revealed a *complex superconducting phase diagram in the B-T plane*,[2–4] with three distinctly different superconducting phases that meet at a tetracritical point. A phenomenological explanation for the splitting of the superconducting transition has been offered within a Ginzburg-Landau formalism including a term that describes the coupling of a superconducting *multicomponent* order parameter with a symmetry breaking field (SBF).[5] The SBF is thought to originate from the extremely small antiferromagnetic (AF) moment [0.02 μ_B/(U atom)] detected by neutron diffraction below $T_N=5$ K.[6] Recent neutron-scattering[7] and specific-heat[8] experiments under hydrostatic pressure have revealed that the splitting $\Delta T_c=T_c^+-T_c^-$ is proportional to the antiferromagnetic moment squared $M^2(T_c^+)$, and that both seem to vanish at a critical pressure of 4.0±0.7 kbar, which suggests a direct coupling of the AF and SC order parameter. However, the available Ginzburg-Landau models[5] are at several points inadequate,[9] in particular as to the topology of the phase diagram. Therefore other explanations should be looked into. In this respect we mention the scenario of two nearly degenerate representations that split the superconducting phase transition in the presence of a spontaneous lattice deformation or stacking faults.[10]

So far, little is known about the lattice deformations that occur at the different phase lines. As the electronic Grüneisen parameter of UPt_3 is unusually large (~70),[11] strong deformations are expected to arise from the coupling of the strains to the SC order parameter.[12] Indeed large effects have been observed in the longitudinal elastic constants.[2,12] In this paper we report on a dilatometry study of the superconducting phases of UPt_3. The lattice deformations at all the phase lines have been identified by either thermal expansion or magnetostriction. The resulting phase diagram allows for an accurate analysis of the phase lines near the tetracritical point. The uniaxial pressure dependence of the various phases is derived via Ehrenfest relations. Moreover, we report on a novel feature at a field of 0.1 T.

Thermal expansion (TE) and magnetostriction (MS) experiments were performed on a single-crystalline sample (dimensions $a\times b\times c\simeq 3\times 1\times 2$ mm^3) for a field directed in the basal plane (along a or b) and a dilatation (contraction) along the hexagonal c axis, using a sensitive parallel-plate capacitance dilatometer, machined of oxygen-free high-conductivity copper, with a detection limit of ~0.01 Å. The linear MS, $\lambda_c=[L(B)-L(0)]/L(0)$, was recorded while slowly sweeping the field at a typical rate of 20 mT/min. The coefficient of linear MS $\tau_c=L^{-1}dL/dB$ was then calculated by differentiating λ_c with respect to B. The coefficient of linear *TE*, $\alpha_c=L^{-1}dL/dT$, was measured using a temperature modulation technique ($f=0.01$ Hz, $\Delta T=5$ mK), with two RuO_2 resistors, which served as heater and thermometer, glued on the sample.

Magnetostriction curves have been taken at temperature intervals of 20 mK or smaller. A few typical curves are shown in Fig. 1. At a relatively low field of 0.1 T (that we denote B_d) a small anomaly is visible. This anomaly is also observed in the normal phase and, therefore, cannot be attributed to superconductivity. We will comment on this later. At higher fields a second anomaly appears, i.e., a kink in λ_c (step in τ_c). Below $T_{cr}=0.387$

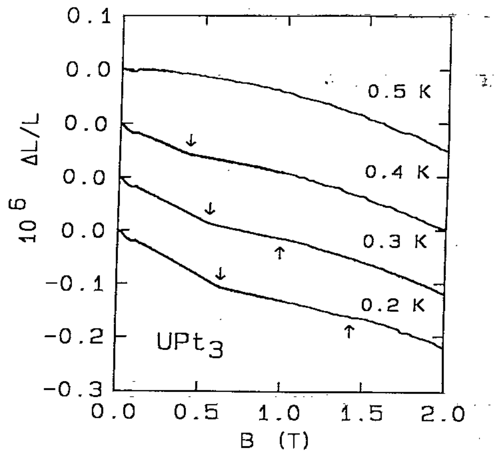

FIG. 1. Magnetostriction along the c axis for $\mathbf{B}\|b$. ↓ indicates the NA boundary (at 400 mK) or the BC boundary (at 200 and 300 mK). ↑ indicates the NC boundary. The curves are shifted along the vertical axis for the sake of clarity.

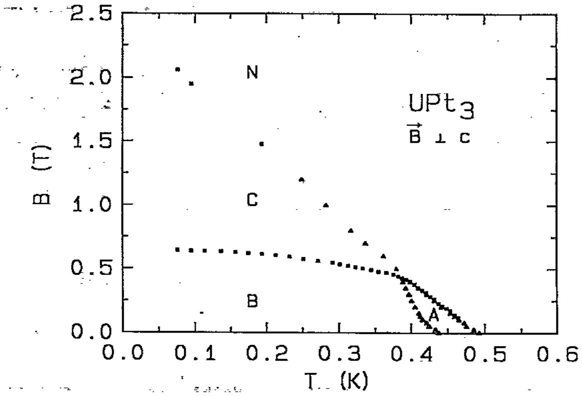

FIG. 3. Superconducting phase diagram of UPt_3 for $\mathbf{B}\perp c$, determined from the magnetostriction (■) and thermal expansion (▲) data.

K this kink separates the low-B low-T phase (labeled B) from the high-B low-T phase (C), whereas above T_{cr} it delimits the low-B high-T phase (A) from the normal phase (N). The normal to superconducting phase transition appears as a step in τ_c at the NA boundary, while it appears only as a change of slope at the NC boundary. This underlines the different nature of the A and C phases.

The thermal expansion curves in constant applied fields have been taken at field intervals of $\Delta B \geq 0.025$ T. A few typical curves are shown in Fig. 2. The zero-field curve shows two sharp jumps *of opposite sign*, at $T_c^+=0.493(2)$ K and $T_c^-=0.438(2)$ K. The jumps at T_c^+ and T_c^- identify the NA and AB transitions, respectively. Under a magnetic field the transition temperatures are both suppressed, initially at an almost equal rate $dT_c^+/dB \simeq dT_c^-/dB \simeq -0.24$ K/T, while for $B>0.1$ T the lower transition is suppressed more slowly, $dT_c^-/dB=-0.07$ K/T, so that the transitions meet at a critical point. For $\mathbf{B}\|\mathbf{a}$ and $\mathbf{B}\|\mathbf{b}$ identical results have been obtained revealing the absence of a marked basal-plane anisotropy. For $B>B_{cr}$ the $\alpha_c(T)$ curves exhibit only a single small discontinuity, which signals the NC boundary.

Having determined all the transition points from the $\alpha(T)$ curves in fixed field and the $\lambda(B)$ curves at fixed temperature, we collect them in the B-T phase diagram in Fig. 3. The phase diagram is in good agreement with previously reported diagrams,[2–4] however, as the accuracy is higher, a clear change of slope becomes visible in the AB phase line at 0.1 T. An enlargement of the region near the critical point (Fig. 4) clearly reveals a *tetracritical* point located at $T_{cr}=0.387(3)$ K and $B_{cr}=0.443(5)$ T. The thermodynamic stability of the phase diagram has been investigated by Yip, Li, and Kumar,[13] by relating the slopes of the phase lines to the discontinuities in the specific heat at the phase transitions. The absence of latent heat at the NA, AB, and NC phase lines indicates that these transitions are of second order. This is supported by the lack of hysteresis in $\alpha_c(T)$ and $\lambda_c(B)$ at the phase transitions. The order of the BC phase line can be

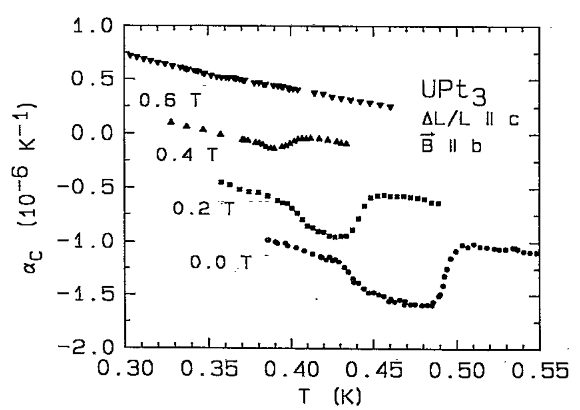

FIG. 2. The coefficient of thermal expansion along the c axis in fields ($\mathbf{B}\|b$) as indicated. The curves in field are shifted along the vertical axis for the sake of clarity.

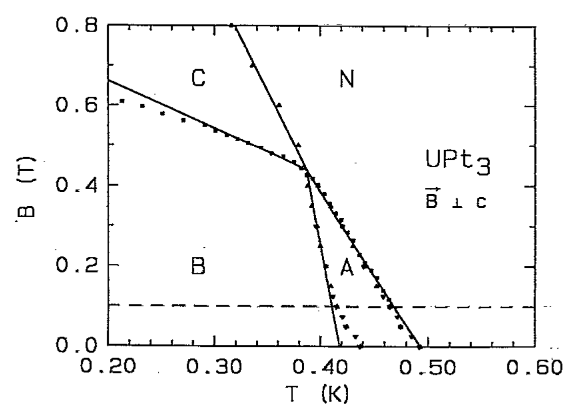

FIG. 4. Enlargement of the superconducting phase diagram of UPt_3 near the tetracritical point. The phase lines are obtained from the thermal expansion data for $\mathbf{B}\|b$ (▲) and $\mathbf{B}\|a$ (▼) and from the magnetostriction data for $\mathbf{B}\|b$ (■). The full lines represent the slopes near (T_{cr}, B_{cr}). The dashed line locates the temperature-independent anomaly at $B_d=0.1$ T.

inferred from the thermodynamic constraints.[13] For our phase diagram the relevant slopes near the tetracritical point (Fig. 4) are given by $dT_{NA}/dB=-0.241$ K/T, $dT_{AB}/dB=-0.070$ K/T, $dT_{BC}/dB=-0.840$ K/T, and $dT_{NC}/dB=-0.188$ K/T, while the specific-heat data published in Ref. 4 yield jump ratios $\Delta C_{NA}/\Delta C_{NC}=0.65(8)$ and $\Delta C_{AB}/\Delta C_{NC}=0.35(8)$. The stability condition for a tetracritical point of four second-order phase lines with the measured slopes is satisfied for $\Delta C_{NA}/\Delta C_{NC}=0.58(4)$ and $\Delta C_{AB}/\Delta C_{NC}=0.43(4)$. With these parameters we reach the conclusion that the BC transition is also of second order. This conclusion is supported by the absence of hysteresis in the magnetostriction at the BC transition.

Having established that all the phase lines are of second order we next determine the pressure dependence of the phase diagram via the Ehrenfest relations. As, so far, only the linear coefficients α_c and τ_c, i.e., along the c axis, have been measured, only the effect of a uniaxial stress along c (p_c) can be evaluated. The relevant Ehrenfest relations are

$$\left[\frac{\partial T}{\partial p_c}\right]_{B,p'}=\frac{V_m\Delta\alpha_c}{\Delta(C_p/T)}, \qquad (1)$$

$$\left[\frac{\partial B}{\partial T}\right]_{p}=-\frac{\Delta\alpha_c}{\Delta\tau_c}, \qquad (2)$$

$$\left[\frac{\partial p_c}{\partial B}\right]_{T,p'}=\frac{\Delta\tau_c}{\Delta_{33}}, \qquad (3)$$

where $\Delta(C_p/T)$ and Δs_{33} are the discontinuities at the phase lines in the molar specific heat divided by T (at constant pressure) and the longitudinal compliance along the c axis s_{33}, respectively, $V_m(=42.4\ \mathrm{cm}^3/\mathrm{mol})$ is the molar volume, and p' refers to uniaxial pressures in the basal plane. From the field variation of the values of $\Delta\alpha_c$ (Fig. 2) and $\Delta(C/T)$ (Ref. 4), the uniaxial pressure dependence of the phase lines NA, AB, and NC has been calculated using Eq. (1). The resulting temperature-field-pressure phase diagram is shown in Fig. 5. Because of the opposite signs of the values for $\Delta\alpha_c$ at the NA and AB transitions, the A phase will be suppressed under uniaxial pressure. The critical uniaxial pressure for suppression of the A phase (p_{cr}) is relatively small, e.g., $p_{cr}=2.5$ kbar and $T=0.460$ K in zero field. For $p>p_{cr}$ only one phase line remains, as was shown by recent pressure experiments.[8,14] We identify this phase line as the NC boundary. For increasing fields we calculate [Eq. (1)] that p_{cr} decreases and drops to zero at the tetracritical point in the B-T plane. The C phase appears to be the most stable phase under uniaxial pressure. From the small negative value of $\Delta\alpha_c(\simeq-0.02\times10^{-6}\ \mathrm{K}^{-1})$ at the NC transition we infer that the C phase is suppressed at a low rate. The thermodynamic consistency of the measurements was cross-checked by using Eq. (2). For instance, the slope (dB/dT) of the NA phase line calculated from the discontinuities $\Delta\alpha_c$ and $\Delta\tau_c$ was found to be in excellent agreement with the experimental one (Figs. 3 and 4). The uniaxial pressure dependence of the BC phase line cannot be evaluated from Eq. (1), as no clear anomalies in $\Delta(C/T)$ and $\Delta\alpha_c$ have been observed at the BC boundary. The absence of a sizable value for $\Delta\alpha_c$ follows directly from Eq. (2), as $\Delta\tau_c$ is relatively large and $(\partial B/\partial T)_p$ is small. However, the uniaxial pressure dependence of the BC phase line can be inferred from Eq. (3), where Δs_{33} can be evaluated from the discontinuities in the longitudinal elastic constants, Δc_{11} and Δc_{33}.[2] With the estimate $\Delta s_{33}\simeq26\times10^{-6}\ \mathrm{Mbar}^{-1}$ (Ref. 2) and the experimental value $\Delta\tau_c=-0.12\times10^{-6}\ \mathrm{T}^{-1}$ (Fig. 1), we deduce $dB/dp_c\simeq-0.2$ T/kbar (at 200 mK). Consequently, the B phase is also suppressed at moderate uniaxial pressures (2–3 kbar). This is consistent with the thermodynamic constraints on the phase diagram which impose that also in the p_c-T plane a tetracritical point exists, i.e., a fourth phase line (the BC phase line) should emerge from the point where the phase lines NA, AB, and NC meet (see Fig. 5). Note that for our thermodynamically derived phase diagram we assumed a linear uniaxial pressure dependence of the different phase lines. This approximation is justified by the good agreement with the zero-field uniaxial pressure diagram.[14]

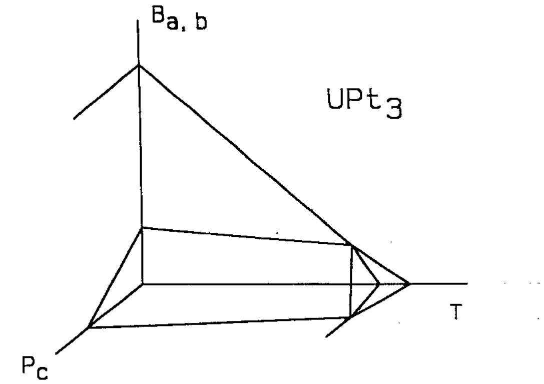

FIG. 5. Schematic uniaxial pressure dependence (along the c axis) of the superconducting phases of UPt_3 for a field in the basal plane. In the B_{ab}-T plane a tetracritical point is located at $B_{cr}=0.443$ T and $T_{cr}=0.387$ K. In the p_c-T plane a tetracritical point is located at $p_{cr}=2.5$ kbar and $T_{cr}=0.460$ K. Both the A and B phases disappear at moderate uniaxial pressures ($\sim$2.5 kbar in zero field).

Next we discuss the anomalous behavior of the AB phase line near 0.1 T (Fig. 4). Locating also the anomaly observed in $\lambda_c(B)$ at $B_d=0.1$ T in Fig. 4, we observe that this temperature-independence phase line intersects the AB boundary just where the slope changes. This strongly suggests that both phenomena are coupled. The anomaly at $B_d=0.1$ T in the $\lambda_c(B)$ curves is likely related to the antiferromagnetic order, as it weakens upon increasing the temperature and is no longer detected at 4.2 K. As the anomaly at B_d displays some hysteresis, we suggest that it arises from small lattice distortions caused by the reorientation of magnetic domains perpendicular to the field. Assuming a two-dimensional (2D) E_1 representation for the SC order parameter, the slopes of the phase lines NA and AB have been investigated within the Ginzburg-Landau approach, for various orientations of the SBF with respect to the field.[5] Within this framework the kink in the AB phase line can be explained by a crossover from an averaging random (along the equivalent b axes) domain orientation, yielding parallel phase lines NA and AB, to the configuration SBF$\perp B$ for $B > 0.1$ T, when all the domains are oriented perpendicular to the field.

The zero-pressure phase diagram can be explained to a large extent within the Ginzburg-Landau formalism including a symmetry breaking term. The E_1 representation model[5] appears to be the most relevant model. It is able to describe correctly the topology of the zero-pressure phase diagram for $\mathbf{B}\perp c$. Furthermore, various quantities, such as the specific heat and the upper and lower critical fields can be analyzed in a consistent way, yielding a ratio of the fourth-order stability parameters $\beta_2/\beta_1 \simeq 0.4$ (Ref. 15) and a ratio of the slopes of the upper critical field $R = B'_{c2}(NC)/B'_{c2}(NA) = 1.28$. However, a number of serious problems arises when we include the TE data in the analysis in the E_1 representation model as formulated by Thalmeier *et al.*[12] In particular, a negative ratio $\beta_2/\beta_1 = \Delta_{AB}\alpha_c/\Delta_{NA}\alpha_c = -0.3$ results, while from the $\lambda_c(B)$ data we deduce $\Delta_{NC}\tau_c/\Delta_{NA}\tau_c \simeq 1/R \simeq 0$. In fact this analysis confirms that direct strain–order-parameter coupling is not enough to explain the phase diagram.[12] In order to overcome these problems we have investigated the influence of higher- (fourth-) order terms of the direct coupling, which can be estimated from the change of slope of the thermodynamic quantities at the phase transitions. This effect is, however, far too small. Next we have investigated the influence of the strain dependence of the SBF (denoted by M^2). This effect should be strongly anisotropic in order to make the Ginzburg-Landau analysis consistent. Furthermore, the strain dependence of the SBF should be larger than the strain dependence of T_c (we derive $\partial \ln M/\partial \ln\epsilon_{xx} = 166$ and $\partial \ln M/\partial \ln\epsilon_{zz} = 444$, while $\partial \ln T_c/\partial \ln\epsilon_{xx} = 84$ and $\partial \ln T_c/\partial \ln\epsilon_{zz} = 128$). Another important constraint on the Ginzburg-Landau models follows from the deduced uniaxial pressure dependence of the different phases. For the relevant ratio $\beta_2/\beta_1 \simeq 0.4$, the models predict the B phase to be the most stable phase in absence of a SBF (under pressure). However, our Ehrenfest analysis shows that the A and B phases are rapidly suppressed and that the C phase is the stable phase under uniaxial pressure along the c axis. This strongly suggests that a SBF is not the origin of the splitting of T_c.

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