Anisotropic dc Magnetization of Superconducting UPt_3 and Antiferromagnetic Ordering Below 20 mK

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We have measured the dc magnetization of three high-quality single crystals of UPt_3 with a SQUID magnetometer down to ≈5 mK in magnetic fields below 2 mT. With $\vec{B}$ in the $\hat{c}$ direction we find a double superconducting transition, while for $\vec{B} \parallel \hat{a}$ we observe only one. At lower temperatures the temperature dependence of the magnetization follows power laws indicating unconventional behavior for the penetration depth. Below 20 mK a steep diamagnetic drop occurs, coinciding with the specific heat anomaly which we found earlier at 18 mK, pointing to static antiferromagnetic order. [S0031-9007(99)08720-7]

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Among the heavy fermion compounds which are known for their unusual superconducting (SC) and magnetic properties UPt_3 is the most likely candidate for unconventional superconductivity in the sense that the symmetry of the order parameter is lower than that of the Fermi surface. Despite the fact that the exact symmetry has not yet been identified, an unconventional order parameter is strongly indicated by the existence of multiple superconducting phases including measurements of specific heat [1], also under applied magnetic field [2], and pressure [3], as well as ultrasonic attenuation [4], tunneling [5], and anisotropy of the lower and upper critical fields [4,6]. These results show that there are three SC phases (usually denoted A, B, and C) in the magnetic field-temperature (B-T) plane. The existence of these phases leads to a double transition at T_c^+ and T_c^-, separated by about 60 mK, and to kinks in $B_{c1}(T)$ and $B_{c2}(T)$. In addition, evidence for unconventional behavior of UPt_3 follows from specific heat [1,2,7], thermal conductivity [8], and superconducting penetration depth [9–12], that exhibit power laws at low temperatures instead of the classical exponential behavior.

On the theoretical side, models have been proposed for a two-component order parameter coupled to a symmetry breaking field for which antiferromagnetism (AF), observed below 5 K in neutron scattering experiments, is considered the most likely candidate [13,14]. Alternative scenarios have been suggested having accidentally near-degenerate order parameters [15,16] or one-dimensional representations combined with weak spin-orbit coupling [17]. However, there is as yet no consensus as to which class of models is correct.

Of key importance to this problem is the magnetic nature of the various SC phases of UPt_3. So we have performed new static magnetization measurements down to very low temperatures in low magnetic fields with a SQUID magnetometer. In earlier work extensive studies of the penetration depth of various samples have been performed, notably an investigation of four samples using ac inductance methods [11], finding a significant frequency dependence in some cases. These results suggested the need for static measurements, particularly at very low temperatures, performed on high quality single crystals. Owing to special annealing procedures [18], our samples have very high T_c's (551 mK) and residual resistivity ratios rrr $\gtrsim 900$; see Table I. We have cooled these samples to temperatures close to 5 mK, which has allowed us to detect, for the first time, a magnetization signal associated with AF ordering, a key component of theoretical models that require a symmetry breaking field. We identify this transition with the low temperature specific heat anomaly at 18 mK that we have reported previously [7].

The static magnetic susceptibility was measured in a SQUID pickup system (described in Ref. [19]), in

TABLE I. Properties of the samples used in this work; rrr_c is the residual resistivity ratio, measured along the c axis.

Sample	Mass (mg)	μmole	T_c^+ (mK)	T_c^- (mK)	Annealing temperature (°C)	rrr_c
#1	485.5	589.7	540	476	Not annealed	200
#2	19.214	23.338	551	427	800, 6 days	892
#3	14.020	17.029	549	−	970, 6 days	957

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magnetic fields between 0.05 and 2 mT. The thermal coupling of the samples to a nuclear demagnetization stage was provided by a 2-mm-diam Ag wire. To reach the lowest temperatures the samples were squeezed in a Ag clamp attached to the Ag wire, all of high purity (99.999%). The temperature was measured with carbon resistors down to 10 mK. After demagnetization to $\approx$1.6 mK (sufficient to cool the samples to their limit) the temperature of the nuclear stage, T_{nst}, was calculated from the warm-up curve using its known heat capacity. Below $\approx$20 mK the sample temperature was determined from the heat leak, the thermal conductance of the link, measured independently, and T_{nst}. A conservative estimate taking a typical value of 6 μW/mole for the radioactive heat leak of UPt_3 grown from depleted uranium [19], resulted in a final temperature for the samples of $\lesssim$5 mK.

We have investigated three samples in this work. Sample #1 is a single crystal grown at the University of Konstanz by electron beam melting using especially depleted uranium. Its shape is a 2-mm-diam cylinder with its axis along the $\hat{c}$ direction and a slanted top face. Samples #2 and #3 were two single crystals grown at Northwestern University using float-zone refining and annealing in ultrahigh vacuum [18]. Their dimensions are ($0.5 \times 0.5 \times 4$ mm^3) cut out of a larger piece by spark erosion with the crystallographic $\hat{c}$ axis (#2) and $\hat{a}$ axis (#3) oriented along its length. They were subsequently etched, removing 10% by weight, and annealed at 800 °C and 970 °C, respectively. Both have a very high and sharp T_c, as well as very high rrr values given by $\rho_{300\text{-K}}/\rho_{0\text{-K}}$ where $\rho_{0\text{-K}}$ is defined as the $T \rightarrow 0$ extrapolation of the quadratic T dependence of the resistivity measured below 1 K. For samples #2 and #3, respectively, $\rho_{0\text{-K}}$ was 0.148 and 0.477 $\mu\Omega$ cm, and the resistive transition widths (90%–10%) were 5.7 and 2.3 mK. Sample #3 was found to have nearly 2 orders of magnitude smaller hysteresis in high-field magnetic torque measurements [20] as compared to sample #1, and even to sample #2, attesting to its excellent quality.

The field cooled (Meissner) signals showed increasing diamagnetic values upon successive temperature sweeps and only after a few sweeps the signal was quasireversible with only a slight long-term drift from flux creep processes [21]. We subtracted this creep from the raw data as well as a T-dependent background from the Ag cooling finger which resulted in a maximal systematic error of 15% at the lowest temperatures. The Meissner signal was approximately 2% of the shielding signal (zero field cooled) owing to strong flux pinning in UPt_3, leaving a large background of trapped flux inside the sample, in spite of $B < B_{c1}$. The T dependence of $\lambda(T) - \lambda(0)$ was deduced from the quasireversible Meissner signals.

The data for the Meissner signals of the three samples are shown in Fig. 1 after subtraction of the drift and of the Ag background. For the samples $\vec{B} \parallel \hat{c}$, a double transition

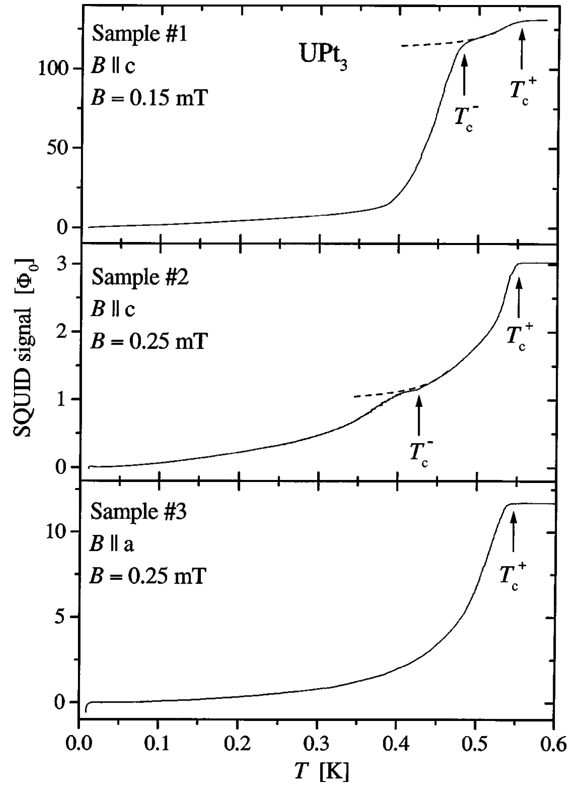

FIG. 1. T dependence of the penetration depth of three UPt_3 single crystals. The solid curves are the flux changes in the SQUID magnetometer measured in units of the flux quantum Φ_0. Their T dependence is proportional to $\lambda(T) - \lambda(0)$. Different absolute values are due to different sample sizes. The field is oriented parallel to the long geometrical axes of the samples. The transitions T_c^+ and T_c^- are indicated as the onset of the χ decrease. For the dashed lines, see text.

at T_c^+ and T_c^- is clearly observed. This is also true for the unannealed sample #1 which did not show a double peak in the specific heat at T_c [7]. In the case $\vec{B} \parallel \hat{a}$ we observed only a single transition at T_c^+. In Fig. 1 the data are shown for sample #3 but the absence of a second drop in this orientation was confirmed with sample #1. Below 30 mK a magnetization anomaly consisting of a small increase starting near 30 mK and an additional drop below 15–20 mK is observed in both samples #2, and #3 (Fig. 2). We identify it with the onset of AF order (see below). After numerical subtraction of this anomaly, power law fits for $\lambda(T) - \lambda(0)$ to the intermediate parts of different curves (between 30 and 120 mK) gave exponents between 0.97(1) and 1.00(1) for $\vec{B} \parallel \hat{a}$. The additional systematic error from the background subtraction leads to an exponent of 1.0 (+0.3/−0.1) in this case. For the $\vec{B} \parallel \hat{c}$ orientation the resulting exponent is 2.0 ($\pm$0.1).

In previous work Gross *et al.* [9] used a dc magnetization method, similar to that reported here, but at much higher temperatures. Broholm *et al.* [10] analyzed their μSR measurements finding a linear behavior of λ for $\vec{B} \parallel \hat{c}$ and a quadratic dependence for $\vec{B} \parallel \hat{a}$, in contrast

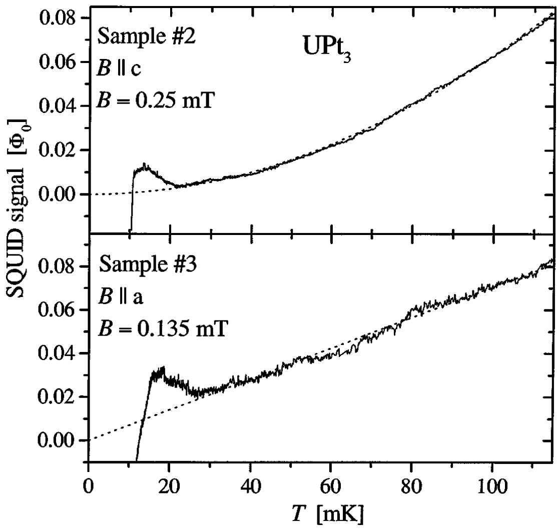

FIG. 2. Low temperature part of the UPt_3 penetration depth (same scales as in Fig. 1). Dotted lines: Quadratic fit for $\vec{B} \parallel$ c and linear fit for $\vec{B} \parallel$ a. The diamagnetic drop below 18 mK points to AF order. It is preceded by a "paramagnetic" precursor.

to our work. However, based on absolute measurements of the penetration depth [9], it was claimed [12,22] that the intrinsic T dependence was not observed in the muon spin rotation (μSR) experiment. Similarly, NMR experiments [23] did not have the resolution to see the effects of $\lambda(T)$ on the lineshape. Signore *et al.* [11] used ac inductance methods with a survey of four samples to determine the temperature dependence of λ for $\vec{B} \parallel \hat{c}$, finding at low frequency a clearly observable double transition and a quadratic power law for two unannealed crystals. For an annealed sample it was linear. The power law exponents became larger at high frequency, up to 16 MHz. Our static magnetization results are consistent with their observation of a double transition for $\vec{B} \parallel \hat{c}$ and, in addition, we have shown that the double transition is not observed for $\vec{B} \parallel \hat{a}$. However, our observation of a quadratic power law behavior for $\vec{B} \parallel \hat{c}$, with the annealed sample #2, is inconsistent with results from their annealed crystal. Differences between the experiments include sample purity, annealing, surface conditions, and the effects of trapped flux. We argue that the systematic errors and impurity scattering always favor power laws higher than linear and that if a linear dependence is observed, it should be closer to the intrinsic temperature dependence. Indeed, our sample #2 showed a much higher hysteresis in torque measurements compared with sample #3, and its T^2 dependence might not be intrinsic. In this respect, combining the results of Signore *et al.* [11] for $\vec{B} \parallel \hat{c}$ and our present finding for $\vec{B} \parallel \hat{a}$, there is good evidence for linear temperature dependence for the penetration depth in UPt_3 in both directions.

We interpret these penetration depth data in terms of a simple model of screening currents combined with unconventional superconductivity with point and line nodes of a two-dimensional order parameter. The penetration of magnetic flux into the superconductor is given by the London formula $\lambda^{-2} = \frac{4\pi e^2}{c^2}\frac{n_s}{m^*}$. In an anisotropic superconductor n_s/m^* is a tensor quantity. The movement of the quasiparticles in k space is confined to the intersection lines of Landau cylinders (very closely spaced in low fields) with the Fermi surface. Whenever supercurrents have to pass a node of the order parameter, there is a vanishing number n_s of Cooper pairs available to carry the current, and the screening of flux is weaker, i.e., the penetration depth is larger. The effect of point nodes is small because only very few k-space trajectories are influenced. From Fig. 1 it is seen that after the initial drop below T_c^+ (different for samples #1 and #2 due to smaller flux pinning in the latter) the SQUID signal would follow the temperature variation of n_s; see dashed lines. For $\vec{B} \parallel \hat{c}$, but not for $\vec{B} \parallel \hat{a}$, below T_c^- a second mechanism sets in leading to an increased Meissner effect (shorter λ) which means a *qualitative* change of the order parameter between the A and B phase. This can be explained by an enhancement of n_s due to development of a second component of the OP and the disappearance of meridional lines, present in the A phase, but not in the B phase. It is hard to explain this observation in 1D models, e.g., by an additional isotropic OP component as we will show in the following discussion (where we use the notation of a spherical Fermi surface).

The AB model [15] involves two accidentally near-degenerate 1D order parameters (A and B representation) of the same parity. The A_{1g} OP transforms as unity, A_{1u} as $\hat{z}k_z$. Neither have meridional line nodes. The A_{2g} representation includes line nodes along six meridional great circles [basis function (BF), $k_xk_y(k_x^2 - 3k_y^2)(k_y^2 - 3k_x^2)$], and A_{2u} additionally at the equator [BF(A_{2g}) times $\hat{z}k_z$]. The appearance or disappearance of such a nodal structure would affect both $\lambda \parallel$ and $\lambda \perp$ to $\hat{c}$, because in both cases a high number of Landau cylinders intersect with the line nodes. The B representations have the BFs $k_y(3k_x^2 - k_y^2)(B_1)$ and $k_x(3k_y^2 - k_x^2)(B_2)$, multiplied by k_z for even parity or by $\hat{z}$ for odd parity. This means nodes on three meridians (and for even parity also on the equator) so that the same argument as for A_2 applies and anisotropy is not expected. The same holds for the nearly degenerate OP model using the A_1 and E_1 representations [16]. Here the OP has line nodes around two circles parallel to the equatorial plane ($k_x^2 + k_y^2 - 2k_z^2 = 0$) which would disappear at the lower transition. In this case screening currents would be influenced only when the field is in the basal plane, contrary to what we observe. In the scenario of weak spin-orbit coupling [17] the orbital part of the OP belongs to a single 1D odd-parity representation in the whole SC region, and only the spin part changes at T_c^- which would not influence λ. The weak SO coupling model also does not account for the observed anisotropy.

The E_{1g} and E_{2u} models incorporate two-dimensional representations with basis functions $k_z\binom{k_x}{k_y}$ and $\hat{z}k_z\binom{k_x^2-k_y^2}{2k_xk_y}$, respectively. In the whole SC region both models exhibit point nodes at the poles and line nodes at the equator of the Fermi surface. The line nodes reduce n_s for shielding currents in the situation $\vec{B} \parallel \hat{a}$, but do not change at T_c^-. In the A phase additional line nodes appear on one or two meridians, respectively, which additionally reduce n_s for the shielding of $\vec{B} \parallel \hat{c}$. Therefore, an effect on $\lambda_{a,b}$ is expected in the E models, whereas $\lambda_{b,c}$ is not influenced. We conclude that both E models can explain our observation of an anisotropic double transition in the penetration depth.

We now look at the low temperature part of $\lambda(T)$. The two E models differ in the k dependence of the gap around the point nodes which leads to different power laws. The E_{1g} model predicts a linear T dependence for $\lambda_{a,b}$ ($\vec{B} \parallel \hat{c}$) and a quadratic for λ_c. With $\vec{B} \parallel \hat{a}$ we measure a combination of λ_b and λ_c and at low temperatures the linear term should dominate. For the E_{2u} model a linear behavior is expected in both field directions [24,25]. We infer that either E model is consistent with our experiment.

There have been attempts to explain larger than linear exponents. One of them has been resonant impurity scattering [24], which should, however, produce the same T^2 dependence in both field directions. Another possibility includes nonlocal effects [26], but they should occur only at very low temperatures.

At the lowest temperatures the "diamagnetic" drop coincides with anomalies in specific heat [7] and thermal expansion [27]. On the other hand, $B_{c2}(T)$ shows no effect in this temperature region [19]. This suggests that the anomaly is not due to the SC state, but to a change of magnetization in the normal state. Fomin and Flouquet [28] predict long-range antiferromagnetic ordering at a very low temperature with only short-range fluctuations setting in at 5 K, a view that has been recently supported by neutron scattering experiments [29] where it was found that the $[\frac{1}{2}, 0, 1]$ magnetic reflection, present below 5 K, shows a distinct narrowing below 20 mK. This was interpreted as due to AF correlations of a typical length of 40 nm below 5 K and long-range magnetic order below 20 mK. With our new observations of static magnetic ordering there is growing evidence in support of this view. This might be significant for theoretical models that invoke magnetic order as a symmetry breaking field.

In summary, we have reported on dc magnetization studies on high-quality UPt_3 single crystals. The data are interpreted within a model of critical currents in an anisotropic superconductor with unconventional 2D order parameter symmetry. Unfortunately, the issue of the E_{1g} versus the E_{2u} model is still unresolved and a clarification is hindered by an anomaly which we have discovered near 18 mK. We believe that this anomaly is the magnetization signature for onset of static long-range antiferromagnetic order.

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