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Upper critical fields of the heavy-fermion superconductor UBe_{13}

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We report high-precision measurements of the upper critical magnetic field of UBe_{13} as determined from the behavior of magnetic forces and torques acting on our samples. Our results suggest the presence of two sharp features or "kinks" in the upper critical field as a function of temperature in both single-crystal and polycrystalline samples. We discuss these results in light of recent theoretical and experimental work on the coexistence of heavy-fermion superconductivity with magnetic and/or additional superconducting states.

The growing body of experimental evidence supporting the existence of multiple superconducting states in the heavy-fermion compound UPt_3 has fueled interest in whether similar states also exist in UBe_{13} , URu_2Si_2 , and CeCu_2Si_2 . Ultrasonic measurements within the superconducting state of UPt_3 ,¹ and mechanical measurements of the flux lattice,² are consistent with the appearance of multiple superconducting states in the H - T plane, a situation suggested by earlier low-temperature thermodynamic measurements.³ There are two peaks in the specific heat of superconducting UPt_3 ,⁴ which come together as the magnetic field increases.⁵ These two peaks coincide at a point that is experimentally indistinguishable from the position of a sharp break or "kink" in the slope of the upper critical magnetic field, $H_{c2}(T)$.⁵ Theoretical work has shown that the degeneracy of multiple, coexisting, superconducting states can be lifted by a coupling between the superconducting order parameter and a symmetry-breaking field,⁶ such as the antiferromagnetism exhibited by UPt_3 .⁷

A similar situation may exist in the heavy-fermion superconductor $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ in the range $0.02 \leq x \leq 0.04$ where a second peak appears in the specific heat below the superconducting transition in zero magnetic field.⁸ This second peak was observed by Ott *et al.*,⁹ who suggested that the transition was either to a magnetic or a second superconducting state. Although definitive evidence for the onset of magnetic correlations at the second transition has been reported,¹⁰ it has been shown that spontaneous magnetism can exist in both singlet and triplet superconductors¹¹ [the latter being similar to the A phase of superfluid ^3He (Ref. 12)]. In a sample with $x=0.0331$, these peaks approach each other as the magnetic field increases.⁹ High-precision measurements of $H_{c2}(T)$ on another specimen of the same sample have resolved a kink near the position where these two peaks could intersect.¹³

Rauchschwalbe *et al.*¹⁴ suggested that the second transition of thoriated UBe_{13} could exist in the pure material. They propose that two superconducting states exist in

UBe_{13} and cross with increasing thorium concentration near $x=0.02$. Theoretical work has shown that a system which allows pairing in two even-parity states of angular momentum (e.g., s - and d -wave states) can have thermodynamic properties, such as $H_{c2}(T)$, which resemble those observed in $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$, and that these two states can cross with increasing impurity concentration.¹⁵

Pure UBe_{13} , with cubic symmetry and a superconducting transition temperature T_c near 0.9 K, is the "heaviest" of the heavy-fermion superconductors. The upper critical magnetic field of UBe_{13} has been investigated by several groups.^{16,13} The temperature dependence of H_{c2} is unusual and has been difficult to describe theoretically. The initial slope of $H_{c2}(T)$ is enormous; indeed, a vertical slope cannot be ruled out by existing data.¹⁶ At $T=0$, H_{c2} is surprisingly large, reflecting a small superconducting coherence length. The unusual behavior of $H_{c2}(T)$ in polycrystal samples has been qualitatively described by including a field dependence of the condensation energy as estimated from magnetoresistivity experiments.¹⁷ There is, in addition, a qualitative difference between the low-temperature behavior of single-crystal and polycrystal samples.⁸ At low temperatures $H_{c2}(T)$ of single crystals is linear, while in polycrystals $H_{c2}(T)$ exhibits a positive curvature which sets in near 0.4 K.¹⁶ Ott has suggested that this peculiar upturn may be due to an anisotropy of H_{c2} .⁸ In this paper we present high-precision measurements of $H_{c2}(T)$ determined from magnetic forces and torques acting on both single and polycrystalline samples. Our results suggest the presence of two kinks in $H_{c2}(T)$ of both samples. The confirmed existence of such kinks would constitute evidence for additional phase transitions in UBe_{13} .

Our UBe_{13} samples are approximately bar-shaped with dimensions $\sim 2 \times 1 \times 1 \text{ mm}^3$ and $\sim 4 \times 1 \times 1 \text{ mm}^3$ for the polycrystal and single crystal, respectively. The polycrystal was prepared by arc-melting stoichiometric quantities of the elements on a water-cooled copper hearth in an argon atmosphere. The single crystal was grown from an

Al melt and was oriented with the [100] direction parallel to the magnetic field. The Meissner effect, as determined by ac susceptibility measurements, is featureless to about 0.8 K (0.6 K) with an onset temperature of about 0.88 K (0.94 K) and a transition width of about 0.02 K (0.04 K) for the single-crystal (polycrystal) sample.¹⁸

Magnetic forces and torques are sensed with an *in situ* capacitive magnetometer.¹⁹ The sample is suspended by two fine copper wires over a silvered glass plate, forming a capacitor in which the sample itself acts as one of the capacitor plates. The assembly is positioned about 2 cm below magnetic center of a superconducting or resistive magnet, in the mixing chamber of a top-loading dilution refrigerator. Magnetic forces and torques result in a displacement of the sample. The displacement is sensed by a standard three-terminal capacitance bridge. Modeling the sample as an oblate spheroid and assuming a small, isotropic magnetic susceptibility χ , one can show that, to leading order in χ , the magnetic force is proportional to χH^2 and the magnetic torque to $\chi^2 H^2$.²⁴ If the change in capacitance ΔC is small, both force and torque contribute linearly to $\Delta C(T, H)$.¹⁹ Our thermometer is a carbon resistor mounted near magnetic center, calibrated against the susceptibility of cerium magnesium nitrate (CMN), and corrected for magnetoresistance.²⁰ We define H_{c2} to be the point at which magnetic hysteresis, resulting from flux-pinning effects, disappears in a transition to the normal state as described previously.¹³ This definition is equivalent to that of extrapolating the flux-pinning force to zero.²¹ Typical data are shown in the inset of Fig. 1, where the amplitude of the magnetic hysteresis ΔM is plotted against magnetic field across the transition. The data on each side of the transition are fitted to straight lines which intersect at H_{c2} as shown. We attribute the finite amplitude of ΔM in the normal state to eddy currents induced by the changing magnetic field. Taking the difference between ΔM and the linear fits mentioned above results in a peak centered near H_{c2} . We take the transition width to be the full width at half maximum of this peak. In the vicinity of the kinks in the critical field, the transition widths increase with decreasing temperature from 0.03 to 0.04 T (0.17 to 0.24 T) for the single-crystal (polycrystal) sample.

Our upper critical field measurements are shown in Fig. 1. Polycrystal and single-crystal data are denoted by solid circles (upper curve) and open circles (lower curve), respectively. The initial slopes of $H_{c2}(T)$ are consistent with earlier evaluations.¹⁶ The lower H_{c2} values of the single crystal are consistent with the lower T_c of this sample, which may result from the known presence of Al inclusions from the growth process. Kinks in $H_{c2}(T)$ appear to exist at the positions shown by arrows. Note that, although the data sets seem to scale at temperatures above about 0.5 K, below this temperature region they diverge: Ott's suggestion of anisotropy⁸ thus remains operative.

Hereafter, we shall use the terms *A* feature and *M* feature to refer to the colder and warmer of the two kinks in each $H_{c2}(T)$ curve, respectively. The proposed kinks occur at coordinates (T_A, H_A) and (T_M, H_M) on the $H_{c2}(T)$ curve; these coordinates are given in Table I. The

TABLE I. Coordinates of the *A* feature and *M* feature (see text) observed in the upper critical fields of single and polycrystalline UBe_{13} .

	T_A (K)	H_A (T)	T_M (K)	H_M (T)
Single crystal	0.38	6.4	0.68	3.4
Polycrystal	0.35	7.7	0.52	5.6

kinks appear as abrupt increases in slope with the exception of the *M* feature of the single crystal, which appears cusplike (making it difficult to observe in Fig. 1). To accentuate these kinks we define ΔH_{c2} as the difference between all the H_{c2} data in the vicinity of a particular kink and an extrapolation of the data just above the transition. This extrapolation is linear except in the case of the single-crystal *M* feature where a quadratic term is added to the fit of the data above the transition. Plots of ΔH_{c2} versus T are shown in Figs. 2 and 3 for the *A* feature and *M* feature, respectively.

We now discuss these kinks individually assuming that they do indeed exist. Kinks in $H_{c2}(T)$ can result from phase transitions in either the superconducting state itself (as in UPT_3 discussed above) or the normal state [as in the antiferromagnetic transition of the magnetic superconductor SmRh_4B_4 (Ref. 22)]. To elucidate potential ori-

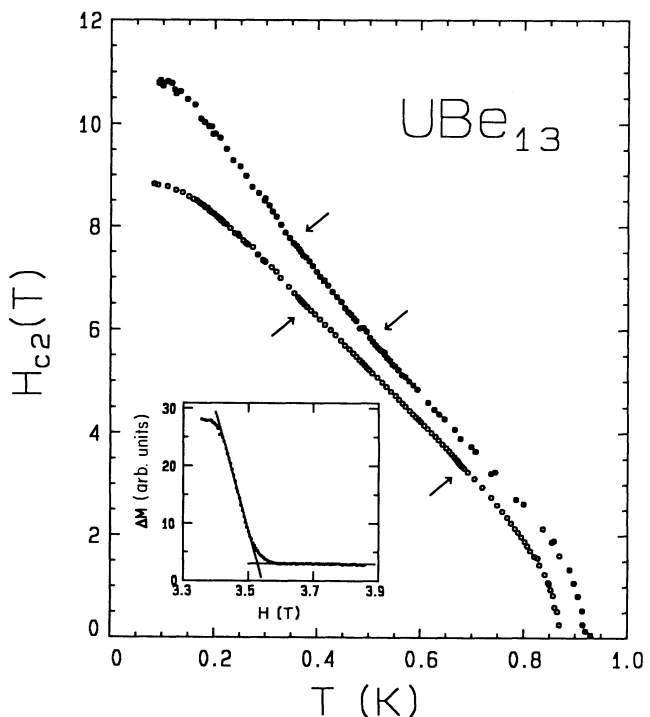


FIG. 1. Upper critical magnetic field of UBe_{13} plotted against temperature. Single-crystal data are denoted by open circles (lower curve) with solid circles for the polycrystal data (upper curve). Positions of “kinks” in the critical field are shown with arrows (see text). The inset shows typical data depicting the disappearance of magnetic hysteresis used to determine the critical field (see text).

gins for these kinks, we studied the behavior of the magnetic force and torque acting on the samples in both normal and superconducting states.

We first discuss the *A* feature. ΔH_{c2} data for the single and polycrystalline samples are shown in Figs. 2(a) and 2(b), respectively. Also shown in Fig. 2(c), for comparison, is the kink previously observed in a thoriated polycrystalline sample with $x = 0.0331$.¹³ We have not found any structure in $\Delta C(T, H)$, in either superconducting or normal state, associated with this kink. (However, the magnetization in the superconducting state is dominated by flux pinning, which is known to “smear out” the magnetic signature of phase transitions such as the lower critical field.²³) The temperature coordinate of the *A* feature is similar in both pure UBe_{13} samples. Also note the qualitative similarity between these kinks and that previously observed in the 3% thoriated sample. These observations suggest that the colder kink could be the result of a phase transition within the superconducting state itself, perhaps a state similar to that observed in $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$.

We now discuss the *M* feature. ΔH_{c2} data for the single and polycrystalline samples are shown in Figs. 3(a) and 3(b), respectively. Here the temperature and field

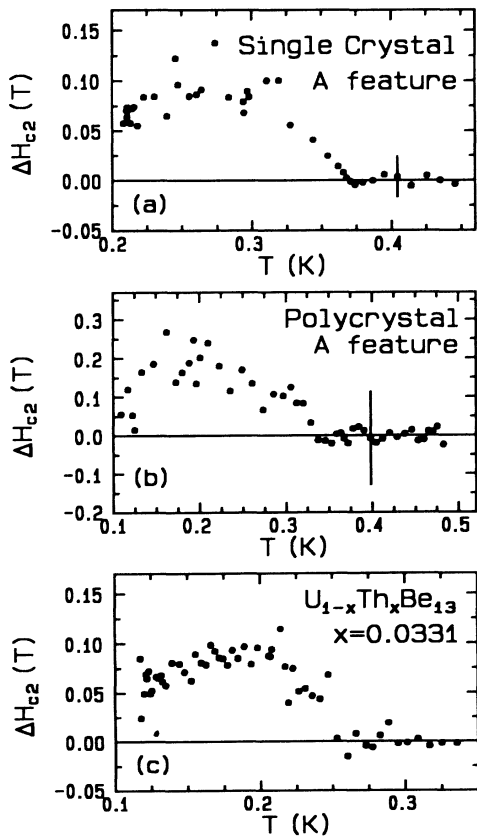


FIG. 2. $\Delta H_{c2}(T)$ plotted against temperature in the vicinity of the *A* feature in the upper critical field (see text) of (a) the single-crystal sample, and (b) the polycrystalline sample. A kink observed earlier in a polycrystalline sample of $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ ($x = 0.0331$) is shown in (c) for comparison. Vertical bars depict the width of the transition (see text).

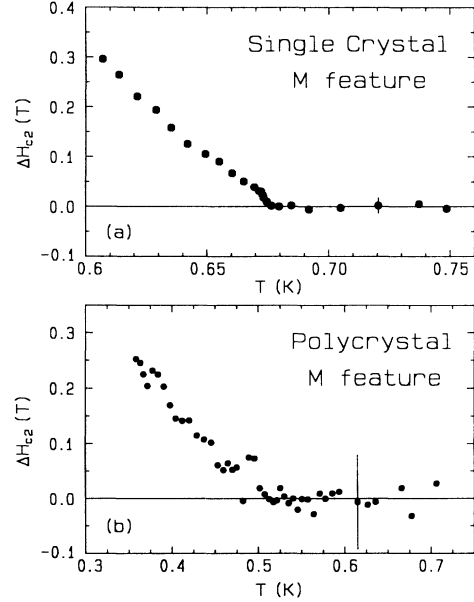


FIG. 3. $\Delta H_{c2}(T)$ plotted against temperature in the vicinity of the *M* feature in the upper critical field (see text) of (a) the single-crystal and (b) the polycrystalline sample. Vertical bars depict the width of the transition (see text).

coordinates of the two *M* features are quite different (see Table I). The cusplike form of this kink in the single crystal is clearly dissimilar to the increase in slope displayed by the polycrystal. (Recall that a quadratic term is added to the linear function used in the determination of the single crystal’s ΔH_{c2} . This procedure enhances the magnitude of ΔH_{c2} over that of the polycrystal.)

Measurements of the magnetic torque in both normal and superconducting states reveal an anomalous behavior. In the superconducting state of the single crystal, the hysteresis due to flux pinning is superimposed on this anomalous behavior, which will be discussed elsewhere²⁴ (although some data on the polycrystal have already appeared²⁵). Briefly, since χ is virtually independent of field to at least 24 T,²⁶ we expect both force and torque to be proportional to H^2 (Ref. 24) and hence to each other (in measurements at constant temperature). At temperatures in the vicinity of the superconducting state, however, an abrupt increase in the magnitude of the torque term relative to the force term is observed in $\Delta C(H)$ measurements. The field at which the onset of this behavior is observed is reproducible and in good agreement with the field coordinate of the *M* feature of both samples, even though the field coordinate of this kink differs by nearly a factor of 2 between these samples. Below about 2 K the field at which the onset of this behavior occurs appears to be independent of temperature. To leading order in χ , the torque term is sensitive to effects resulting from the shape of the sample, while the force term is not.²⁴ These shape effects are characterized by the well-known demagnetization coefficients. Brug and Wolf have shown that these coefficients are generally field and temperature dependent, particularly near a magnetic phase transi-

tion.²⁷ Perhaps this anomalous behavior reflects the presence of a magnetic phase transition, although no sharp feature in $\Delta C(T, H)$ (except for that at H_{c2}) has yet been observed. If this kink is the result of a magnetic phase transition, the different shapes of the M feature, as well as the divergent low-temperature behavior of $H_{c2}(T)$ between the two samples, could be the result of the anisotropy associated with any magnetic ordering. As shown in Fig. 1, the $H_{c2}(T)$ curves indeed diverge near the M feature of the polycrystal; the M feature of the single crystal is cusplike and does not significantly affect the slope of $H_{c2}(T)$ in its vicinity. On the other hand, the break in slope observed at the M feature of the polycrystal could result from the superposition of a cusplike kink with local positive curvature in $H_{c2}(T)$.

Heat-capacity measurements from several groups also suggest the presence of additional phase transitions in UBe_{13} . The unusual behavior of the low-temperature specific heat of UBe_{13} can be described by the existence of a small, broad, second peak below T_c .¹⁴ A robust second peak in the heat capacity has also been observed to develop at about 150 mK and 6 T in polycrystalline UBe_{13} .²⁸ Very recent heat-capacity measurements taken as a function of H at fixed T show "two broad peaks in the superconducting state. The low-field feature occurs at $H \approx 20$ kG, independent of temperature."²⁹ [The discrepancy be-

tween the positions of the heat-capacity features and our features in the H - T plane could either reflect a field dependence to the position of the peak (A feature) or sample-to-sample variations which are currently characteristic of UBe_{13} samples.] Perhaps the external magnetic field helps to stabilize, or even induce, phase transitions responsible for these features.

To conclude, we have presented high-precision measurements of the upper critical magnetic field of UBe_{13} determined from the magnetic forces and torques acting on single and polycrystal samples. Our results suggest the presence of two sharp features or kinks in $H_{c2}(T)$ of each sample.

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