Upper Critical Magnetic Field of the Heavy-Fermion Superconductor UBe$_{13}$

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The temperature dependence of the upper critical magnetic field $H_{c2}(T)$ of the heavy-fermion superconductor UBe$_{13}$ was determined resistively. The magnitude of the initial slope of $H_{c2}(T)$, ~ 420 kOe/K, is the largest value ever reported for a bulk superconductor. The curve of H_{c2} vs T has an extremely unusual shape with a linear region that persists to very low temperatures. The anomalous shape of $H_{c2}(T)$ cannot be accounted for by current theories of either conventional or p-wave superconductivity.

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Recently, Ott *et al.*[1] observed bulk superconductivity in the compound UBe$_{13}$ with properties similar to those of CeCu$_2$Si$_2$[2] and UPt$_3$.[3,4] All three materials appear to be examples of a small class of "heavy-fermion" superconductors that are characterized by low values of the superconducting transition temperature $T_c \lesssim 1$ K and large conduction electron effective masses $m^* \sim$ several hundred times the free-electron mass m_e, inferred from the normal-state electronic specific-heat coefficient λ. The remarkable properties of these heavy-fermion superconductors has led to the speculation that they might exhibit p-wave superconductivity.[5,6] The low-temperature T dependence of the specific heat of UBe$_{13}$[7] and $H_{c2}(T)$[4,6] and ultrasonic attenuation[8] measurements on UPt$_3$ appear to be consistent with, but do not constitute definitive proofs of, this possibility.

In order to obtain more information about the nature of superconductivity in heavy-fermion systems, we have measured the electrical resistivity ρ of UBe$_{13}$ for 50 mK $\leqslant T \leqslant 1$ K in applied magnetic fields H up to 175 kOe. The resistivity determined curve of H_{c2} vs T of UBe$_{13}$ has a very anomalous shape and an enormous initial slope $(-dH_{c2}/dT)_{T_c} \sim 420$ kOe/K, the largest value ever observed for a bulk superconducting material. From an analysis of the H_{c2} vs T curve near T_c in terms of a conventional theory of type-II superconductivity, we find that the magnitude of the initial slope $(-dH_{c2}/dT)_{T_c}$ is consistent with $m^* \sim 300 m_e$ and the negative curvature of $H_{c2}(T)$ near T_c can be explained by paramagnetic limiting. However, we are unable to account for the linearity of $H_{c2}(T)$ between 50 mK and 0.7 K in terms of this same theory. Further theoretical work is necessary to see if calculations of $H_{c2}(T)$ for p-wave superconductors can explain these data. We have also observed strong negative magnetoresistance in the normal state which suggests that a Kondo lattice description may be appropriate for this system.

The bar-shaped single-crystal specimen of UBe$_{13}$ used in this investigation was prepared in a manner previously described.[1] Measurements of the electrical resistance at 16 Hz were performed in a ^{3}He-^{4}He dilution refrigerator at the University of California at San Diego for $T \gtrsim 0.07$ K by varying T in fixed H, applied with a superconducting solenoid, up to 60 kOe. The temperature was determined from a 100-Ω Speer carbon resistance thermometer. On the basis of magnetoresistance data for other Speer carbon resistors,[9] the error in temperature for $0 < H \leqslant 60$ kOe and 0.3 K $\lesssim T \leqslant 1.0$ K was estimated to be $\lesssim 12$ mK. Additional measurements for $0 \leqslant H \leqslant 5$ kOe were made in a ^{3}He refrigerator using the vapor pressure of ^{3}He as a thermometer which is known to be relatively insensitive to magnetic fields in this range.[10] Electrical-resistance measurements at 40 Hz with the sample in the mixing chamber of a dilution refrigerator were made at the Francis Bitter National Magnet Laboratory, Massachusetts Institute of Technology, at fixed T by sweeping H, produced by a Bitter solenoid, up to 175 kOe.[11] The temperature was deduced from two carbon resistance thermometers within the mixing chamber and a correction for magnetoresistance was made as described elsewhere.[11]

Selected ρ vs T data, between 80 mK and 1 K in various magnetic fields from 0 to 60 kOe, are shown in Fig. 1. The superconducting transition curves shift to lower temperature and broaden somewhat with increasing magnetic field. Also evident in the ρ vs T data of Fig. 1 is the large negative normal-state magnetoresistance. This is illustrated in the inset of Fig. 1 where isotherms of ρ vs H between 0 and 60 kOe at several temperatures between 0.85 and 1 K are presented. Neither $CeCu_2Si_2$[12] nor UPt_3[4] displays such a large negative magnetoresistance for comparable values of H.

The resistively determined $H_{c2}(T)$ data for UBe_{13} are shown in Fig. 2 where the 50% points of the transitions have been plotted. The horizontal or vertical bars represent the widths defined from the 10% and 90% points of the transitions. Measurements at current densities of 0.34 and 0.10 A/cm^2 with the applied field either parallel or perpendicular to the direction of the current gave essentially identical results. Displayed in the inset of Fig. 2 are more detailed low-field H_{c2} vs T data where the temperature was inferred from the vapor pressure of ^{3}He. The values of $H_{c2}(T)$ at each field were obtained by averaging the data from six separate sets of measurements, and the horizontal lines represent the uncertainties that were estimated from the scatter in the data. Within experimental error, the $H_{c2}(T)$ curve is linear between 0 and 4 kOe with a slope $(-dH_{c2}/dT)_{T_c} = 420 \pm 20$ kOe/K. To our knowledge, this initial slope is the highest value ever observed for any three-dimensional bulk superconducting material. The previously reported value of 257 kOe/K for UBe_{13}[1] neglected the magnetoresistance of the Allen-Bradley carbon resistance thermometer which would reduce the initial slope from its actual value.[9] The largest values previously reported for $(-dH_{c2}/dT)_{T_c}$ are 230 and 63 kOe/K, respectively, for the other two heavy-fermion superconductors $CeCu_2Si_2$[12] ($T_c \simeq 0.6$ K) and UPt_3[4] ($T_c = 0.54$ K), and ~ 70 kOe/K for the Chevrel phase compound $LaMo_6Se_8$[13] ($T_c \simeq 11$ K). Between 0.850 K and ~ 0.7 K, $H_{c2}(T)$ exhibits negative curvature, whereas below ~ 0.7 K, $H_{c2}(T)$ becomes linear with a slope $(-dH_{c2}/dT) = 91$ kOe/K with no indication of saturation down to our low-temperature limit of 50 mK at which $H_{c2} = 90$ kOe.

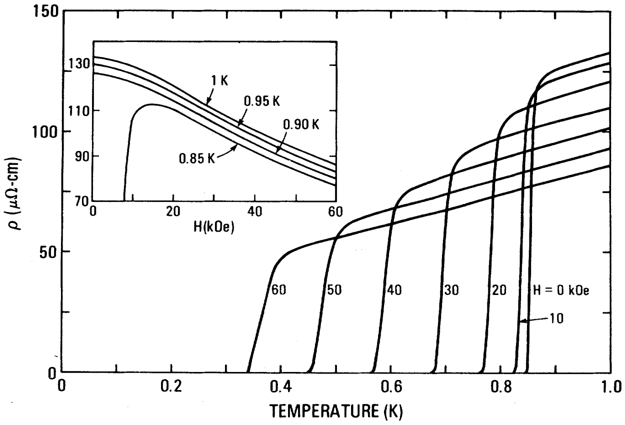

FIG. 1. Selected electrical resistivity ρ vs temperature data for a single-crystal specimen of UBe_{13} in various applied magnetic fields between 0 and 60 kOe. Shown in the inset are ρ vs H isotherms between 0 and 60 kOe at 0.85, 0.90, 0.95, and 1.00 K. The lines are smooth curves that have been drawn through the data points.

If we assume a conventional type of superconductivity for UBe_{13}, we can analyze the H_{c2} vs T data accordingly.[14] Because of the high value of the normal-state ρ for UBe_{13}, it is appropriate to use the dirty-limit approximation[15] for $(-dH_{c2}/dT)_{T_c}$ which, in units of oersteds per kelvin, is given by $(-dH_{c2}/dT)_{T_c}$

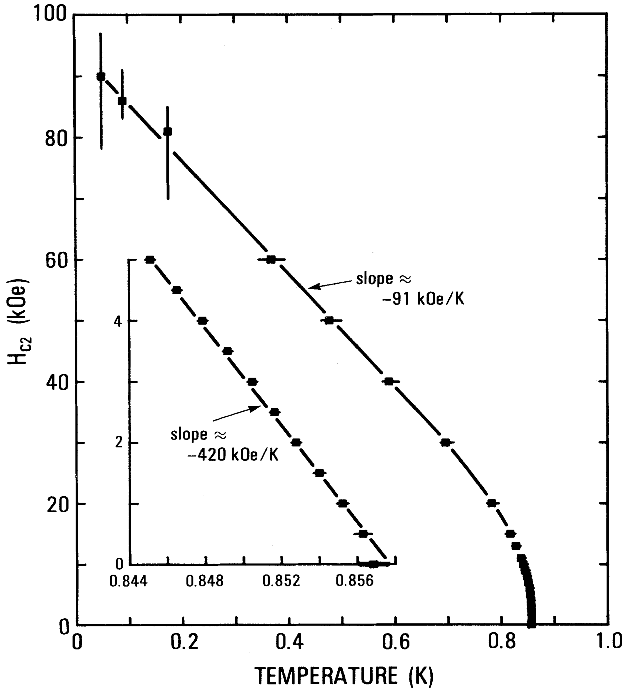

FIG. 2. Upper critical magnetic field H_{c2} vs temperature T for a single-crystal specimen of UBe_{13}. Shown in the inset are H_{c2} vs T data in the vicinity of the zero-field T_c of 0.857 K. The lines are a guide to the eye. Horizontal and vertical bars indicate experimental uncertainties as discussed in the text.

$=(4.41\times10^4)\rho\gamma$, where ρ is measured right above T_c in Ω cm and γ is in erg/cm^3 K^2. Using the value $\rho=125\ \mu\Omega$ cm from this work and the previously reported value $\gamma=1.1$ J/mole$\cdot$K$^2=2.36\times10^5$ ergs/cm^3 $\cdot$K^2 measured for a different sample,[1] we find $(-dH_{c2}/dT)_{T_c}=749$ kOe/K, in order of magnitude agreement with the value of 420 kOe/K determined from the H_{c2} vs T data.

The slope of the H_{c2} vs T curve at T_c can also be used to estimate the zero-temperature orbital critical field $H_{c2}^*(0)$ via the weak-coupling formula[15]

$$H_{c2}^*(0)=0.693[(-dH_{c2}/dT)_{T_c}]T_c$$

which gives $H_{c2}^*(0)\sim250$ kOe. This value of $H_{c2}^*(0)$ can then be used to estimate the coherence length at $T=0$, ξ_0, by means of the dirty-limit expression[16]

$$H_{c2}^*(0)=\Phi_0/4.54\xi_0 l_{\text{tr}}\qquad(l_{\text{tr}}<<\xi_0),\tag{1}$$

where $\Phi_0=ch/2e=2.07\times10^{-7}$ Oe cm^2 is the flux quantum, and l_{tr} is the transport mean-free path. The transport mean-free path in cm is given by the expression[17]

$$l_{\text{tr}}=(1.27\times10^4)/\rho(Z/\Omega)^{2/3},\tag{2}$$

where ρ is in Ω cm, Z is the number of conduction electrons per unit cell, and Ω is the unit cell volume in cm^3. As a rough approximation, we assume that there are three $5f$ "heavy electrons" contributed by each U atom, yielding $Z=24$ since there are eight UBe$_{13}$ formula units per unit cell. From the lattice parameter of the UBe$_{13}$ specimen used in this investigation, $a=10.254$ Å, we find $\Omega=1.08\times10^{-21}$ cm^3. We then obtain $l_{\text{tr}}=12.9$ Å from Eq. (2), and $\xi_0=142$ Å from Eq. (1). If this calculation of ξ_0 using the conventional theory of type-II superconductivity in the dirty limit is valid, then UBe$_{13}$ would not be expected to support triplet superconductivity due to the strong destructive effect of nonmagnetic scattering.

The coherence length can also be obtained from[16]

$$\xi_0=0.18\hbar v_{\text{F}}/k_{\text{B}}T_c.\tag{3}$$

However, it is first necessary to estimate the Fermi velocity $v_{\text{F}}=\hbar k_{\text{F}}/m^*$ where k_{F} is the Fermi wave vector. With the assumption of a spherical Fermi surface, $k_{\text{F}}=(3\pi^2Z/\Omega)^{1/3}=8.69\times10^7$ cm^{-1} and $m^*=\hbar^2k_{\text{F}}^2\gamma/\pi^2(Z/\Omega)k_{\text{B}}^2=296m_e$, which gives $v_{\text{F}}=3.39\times10^5$ cm/s. Equation (3) then yields the value $\xi_0=54$ Å, which is in reasonable agreement with the value of 142 Å inferred from $(-dH_{c2}/dT)_{T_c}$, considering the approximations that we have made. This value of m^* is about 50% larger than the value given in Ref. 1 where a different procedure for estimating Z was used. It is interesting to note that with such a large electron mass enhancement, there is no sizable T^2 contribution to the low-temperature electrical resistivity of UBe$_{13}$. Temperature dependences of ρ of T^n with $n\sim2$ have been reported for other heavy-fermion systems such as CeCu$_2$Si$_2$,[18] UPt$_3$,[3,4] and CeAl$_3$.[19]

The paramagnetic limiting field in the absence of spin-orbit scattering is given by $H_{p0}(0)=18.4T_c$ (kOe) at $T=0$,[20] which for $T_c=0.857$ K gives $H_{p0}(0)=15.8$ kOe. However, the paramagnetic limiting field can be increased by spin-orbit scattering which could explain why $H_{c2}(0)$ exceeds $H_{p0}(0)$ by a considerable amount. The negative curvature of $H_{c2}(T)$ between ~0.7 and 0.850 K is consistent with the T dependence expected if $H_{c2}(T)$ is determined by the paramagnetic limiting field H_p [e.g., $H_p\propto(T_c-T)^{1/2}$ close to T_c in the absence of spin-orbit scattering[21]]. To our knowledge, this feature in $H_{c2}(T)$ has never been observed in any other bulk superconducting material.

The H_{c2} vs T data were next compared to the theory of Werthamer, Helfand, and Hohenberg.[14] After matching the observed initial slope, the linear variation of H_{c2} with T for 30 kOe $\lesssim H\lesssim$ 90 kOe could not be reproduced for any value of the spin-orbit scattering parameter. Scaling $H_{c2}^*(0)$ with the extrapolated normal-state values of $\rho(T,H)$ to account for the T and H dependence of ρ only increased the discrepancy and produced a maximum in the calculated $H_{c2}(T)$ curve. It is possible that the inclusion of the T and H dependence of other quantities, presently assumed constant, would yield the observed behavior. This analysis may be applicable to CeCu$_2$Si$_2$ where a maximum in $H_{c2}(T)$ has been observed.[12]

The unusual H_{c2} vs T curve of UBe$_{13}$, particularly the enormous value of $(-dH_{c2}/dT)_{T_c}$, suggests the possibility of an unconventional type of superconductivity, such as p-wave superconductivity, that is insensitive to an applied magnetic field. The decrease of the slope $(-dH_{c2}/dT)$ for $H\gtrsim20$ kOe might then be due to the degradation of the highly correlated state responsible for the superconductivity when $H\gtrsim H_0$, where H_0 is a characteristic magnetic field. The substantial negative magnetoresistance displayed in the inset of Fig. 1 supports this conjecture. Within the context of a Kondo lattice model for UBe$_{13}$, a value of the order of magnitude of 10 kOe for H_0 would be inferred from $H_0\sim k_{\text{B}}T_0/\mu_{\text{eff}}$ with $T_0\sim2$ K where $\rho(T)$ is maximum and $\mu_{\text{eff}}=3.08\mu_{\text{B}}$ from high-temperature $\chi(T)$ data.[22] Further theoretical work is needed to see whether the $H_{c2}(T)$ data can be described by a p-wave pairing model.[23]

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