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Large magnetic field effect on the heavy fermion superconductor

$U_{1-x}Th_xBe_{13}$ just above T_c

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Magnetic fields as small as 4 kOe substantially increase the resistivity of $U_{1-x}Th_xBe_{13}$ for $x = 0.0175$ and 0.0378 between 1.4 K and T_c , the superconducting transition temperature of this heavy fermion material. At the intermediate value of $x = 0.026$, T_c reaches a local maximum, and no normal-state field dependence is observed up to 4 kOe. At $x = 0.0603$, away from the region of nonmonotonic dependence of T_c on x , again no field dependence is seen. The observations are discussed with regard to specific-heat measurements and in terms of the interaction of superconductivity with narrowband features.

I. INTRODUCTION

The intermetallic compound UBe_{13} is a member of the small but growing class of so-called heavy fermion materials. Such systems at low temperatures are characterized by large, temperature-independent susceptibilities and large linear electronic specific-heat coefficients γ . These enhanced γ values correspond to an effective electronic mass of the order of 100 electron masses. Such enhanced properties suggest a description of the low-temperature state of these systems as an electronic Landau-Fermi liquid, the classic example of which is normal 3He .

Three of these heavy fermion systems undergo a superconducting transition below 1 K, notably $CeCu_2Si_2$,¹ UBe_{13} ,² and UPt_3 .³ What is remarkable, focusing on UBe_{13} in particular, is that the magnitude of the specific-heat jump at the superconducting transition temperature is enormous, and of the same order of magnitude as the low-temperature γ value of 1.1 J/mol K^2 . This indicates that it is indeed an electronically enhanced γ and that the electrons responsible for the unusual normal-state properties are the same electrons which bring about superconductivity.

There have been suggestions that the nature of the Cooper pairs in these heavy fermion superconductors is not of the conventional, singlet BCS type. There have been proposals that UBe_{13} ⁴ and UPt_3 ^{5,6} are anisotropic ($L \neq 0$) superconductors by analogy with the $L = 1$ triplet polar state of superfluid 3He at very low temperature. In contrast, $CeCu_2Si_2$ has been modeled as a Kondo lattice system with a conventional BCS ground state.^{7,8}

Nonmagnetic impurities have been shown to substantially modify both the normal and superconducting properties of UBe_{13} .^{9,10} Substitution of thorium, which has no $5f$ electrons, for uranium in compounds of the form $U_{1-x}Th_xBe_{13}$ has been the most heavily studied case. The T_c is strongly depressed by Th in the $x = 0-0.06$ concentration limit, and between about $x = 0.017$ and 0.026 shows an extremely unusual increase of T_c , as determined by resistivity or (less dramatically) by ac susceptibility.⁹ In addition, as x is increased, the shoulder in the resistivity at about 25 K becomes a broad peak and moves to higher temperature. This feature looks as if it is caused by crystal field scattering,

but no crystal field levels have been observed to date.⁹ The narrow low-temperature peak, at 2.5 K for pure UBe_{13} , is similar to a feature in the resistivity of $CeCu_2Si_2$,¹¹ which has been ascribed to the development of the highly correlated Fermi liquid state. This peak moves to lower temperatures rapidly on Th addition, disappearing below T_c for $x \geq 0.026$. The net effect of the motion of these two features is that at a particular low temperature, the resistivity over a range of x values decreases with increasing x .

More unusual behavior yet has been observed recently in the specific-heat C_p data of single-phase $U_{1-x}Th_xBe_{13}$ below T_c .¹² Between about $x = 0.0216$ and 0.0378 a second transition is observed below T_c . It is argued that this can only be a transition between superconducting configurations with different $L \neq 0$ pairing states, although there is some concern about how to interpret entropy balance for these samples below T_c . Very recent NMR results¹³ also reject the possibility that the second transition is structural or magnetic in nature. The spin-lattice relaxation rate $1/T_1$ is roughly proportional to T^3 for $x = 0$ and $x = 0.033$. This is the expected temperature dependence for an energy gap that vanishes on one or more lines on the Fermi surface, which is the case for an $L = 1$ triplet polar state superconductor.

Because small Th additions produce large effects in UBe_{13} , we thought that modest magnetic fields might also strongly affect the properties of $U_{1-x}Th_xBe_{13}$, and indeed they do.

II. EXPERIMENTAL PROCEDURE

High-purity U, Th, and Be were melted together in a conventional argon atmosphere arc furnace. The buttons were turned and remelted at least seven times. Compositions were corrected for weight loss using the measured ratio of constituents in the arc furnace residue.¹² Samples were subjected to x-ray diffraction and metallographic analysis. All specimens were measured in the as-cast condition. Semicircular slices were spark cut from the buttons. A standard four-probe ac resistivity technique in a dilution refrigerator and superconducting magnet were employed in the measurements.

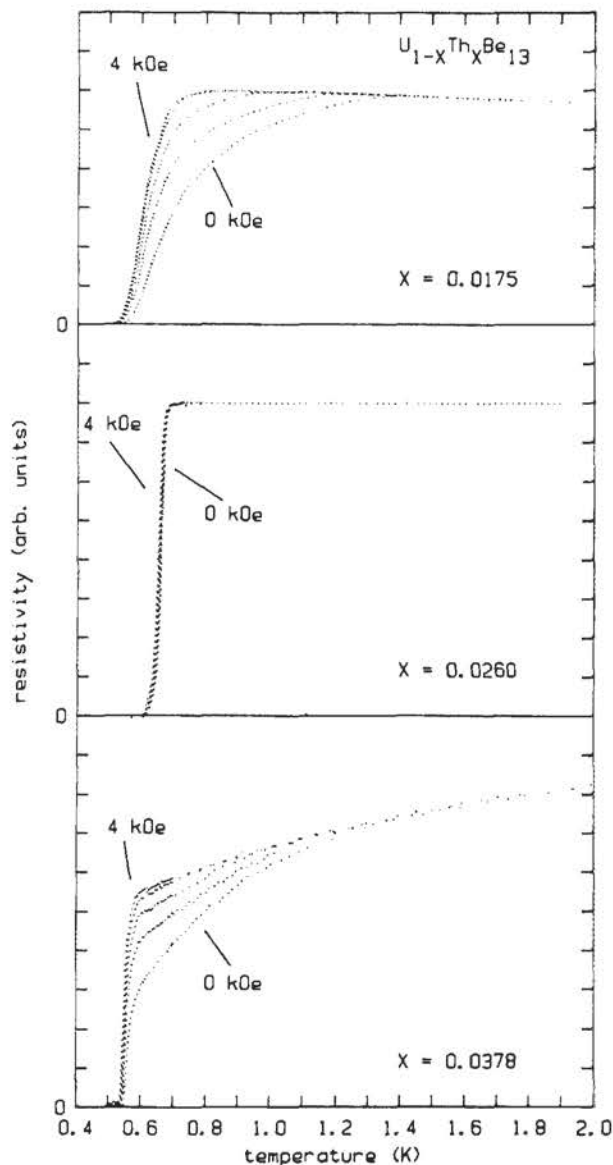


FIG. 1. Resistivity of $U_{1-x}Th_xBe_{13}$ vs temperature with magnetic field as a parameter. Magnetic fields are, from right to left: 0, 1, 2, 3, and 4 kOe. Thorium compositions shown are $x = 0.0175$, $x = 0.026$, and $x = 0.0378$.

III. RESULTS

The main results are summarized in Fig. 1, which shows the resistivities of $U_{1-x}Th_xBe_{13}$ as a function of T for several values of applied magnetic field H . For $x = 0.0175$, the low-temperature peak in the resistivity moves to lower temperatures in an applied field until it appears to move below T_c for $H \sim 4$ kOe. Putting it another way, the low-temperature decrease in the resistivity is inhibited by a magnetic field. For $x = 0.026$, the sample with x nearest to the relative maximum in T_c , the magnetic field has essentially no effect on the resistivity. Near the end of the anomalous region in x at $x = 0.0378$, the $H = 0$ resistivity shows no low-temperature peak; the slowly falling resistivity as T decreases is the low-temperature side of the broad, high-temperature (~ 30 K) feature. There does, however, appear to be a rather more rapid decrease in the resistivity below 1 K than for $x = 0.0175$. The application of a field

inhibits this decrease, as it does in the $x = 0.0175$ case. Finally, for $x = 0.0603$ (not shown) there is a decrease in the resistivity below the 30-K peak all the way to T_c . This is in a region where only one transition is observed in C_p data, and here only a very slight increase ($\sim 1\%$) of the normal-state resistance is observed in a 4-kOe field just above T_c .

All of the compositions studied have a very large $(-dH_{c2}/dT)_{T_c}$. Magnetic field corrections to the thermometry account for more than one half of the measured T_c shift. Thus the determination of the T_c depression is somewhat inaccurate. Furthermore for $x = 0.0175$, as seen in Fig. 1, it is rather difficult to assign a T_c because the normal-state resistance is dropping so rapidly. Therefore, we can only state that $(-dH_{c2}/dT)_{T_c}$ for $0.0178 < x < 0.0603$ is the same order of magnitude as the value for pure UBe_{13} , which is 420 kOe/K.¹⁴

IV. SAMPLE QUALITY CONSIDERATIONS

We have rejected the possibility that the normal-state magnetic field effects observed here may actually be caused by free Th filaments in the sample. Pure thorium has a T_c of 1.374 K and $H_{c2}(0)$ of 162 Oe. This is approximately the same temperature for which field-dependent effects occur for $x = 0.0175$ and 0.0378. If it is Th superconductivity, then the observed " H_{c2} " is much too large and the transition width is extraordinarily wide, greater than 0.8 K, which would be quite broad for a precipitated element. Furthermore, one would expect the amount of free Th to increase with x , but no excess conductivity is observed for $x = 0.0089$,⁹ 0.026, 0.0603, and 0.0675.⁹ In addition, the shape of the resistivity curves for $x = 0.0378$ and $x = 0.034$ of Ref. 9 are very similar near T_c , suggesting a systematic trend with x rather than random variations with free Th.

Metallography and x-ray diffraction analysis also reveal these samples to be essentially single phase. Very minor ($\sim 1\%$) amounts of UO_2 and Be were observed by x-ray diffraction. The presence of free Be implies that the stoichiometry is on the (U, Th) poor side of the $U_{1-x}Th_xBe_{13}$ compound, and thus free U or Th would be very unlikely. In contrast, flux-grown "single crystals" were of much poorer quality. Fractionation of Th in the melt was clear from lattice parameter determination, and an additional unidentified phase was found. Small amounts of free superconducting Al (the flux) were observed in the resistivity measurements. These transitions were very sharp and were quenched by very small magnetic fields on the order of 100 Oe. Again, all of the measurements reported here are on arc-melted material. The above remarks lead us to believe that the observed resistivity curves are intrinsic to $U_{1-x}Th_xBe_{13}$ and are not caused by second phases, in agreement with the conclusions in Ref. 12.

V. DISCUSSION

The nature of the 2.5-K feature in the resistivity is not known; however, it should be noted that the position of a similar peak in $CeCu_2Si_2$, which is attributed to Kondo lattice scattering, has a strong influence on superconductivity in that heavy fermion system.^{11,15} Superconductivity is de-

stroyed in CeCu_2Si_2 if this peak moves from near 20 K to below 6.5 K, which occurs for some stoichiometries; in UBe_{13} , T_c is also depressed as the 2.5-K feature moves to lower temperature upon Th substitution. Although neither the extreme magnetic field dependence of the low-temperature resistivity nor the two specific heat transitions observed for this small range of x are clearly understood, we suggest that these two phenomena may be related. The apparent lack of sensitivity to magnetic field for $x = 0.026$, the midpoint of the anomalous x region with the local T_c maximum by resistivity and with approximately equal entropies in the two specific-heat anomalies, is also not fully understood. We speculate that these phenomena may be caused by a subtle interplay between superconductivity and the source of the scattering mechanism related to the low-temperature resistivity peak as the relative temperatures or energies of these two interactions change with x . Volovik and Khmel'nitskii very recently suggested¹⁶ that some of these unusual effects may be caused by the formation of a "superconducting glass" state with short range order just below T_c followed by a transition to normal superconductivity with long range order at lower temperatures. A crucial point to bear in mind is that the energy scales and bandwidths relating to these phenomena are extremely small; as a result, modest changes to the system, either by impurity additions or magnetic fields, can cause substantial modification of the physical properties of UBe_{13} .

Specific-heat measurements on $\text{U}_{1-x}\text{Th}_x\text{Be}_{13}$ in an applied magnetic field are currently under way to further elucidate this subtle interplay and to probe the nature of the specific-heat anomalies observed in zero magnetic field.

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