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Thermodynamics and magnetism in $U_{1-x}Th_xBe_{13-y}B_y$

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We report specific heat and μSR measurements on Th ($x = 0.019$) and/or B ($y = 0.03$) substituted UBe_{13}. The specific heat data show that either Th or B substitution reduces the Kondo temperature T_K and increases the entropy at the superconducting transition by almost 20%, indicating an enhanced density of states. However, whereas μSR shows clear evidence for magnetic correlations for Th substitutions ($0.019 < x < 0.043$), no magnetism is observed for B substitutions. The enhanced specific heat jump in the B-substituted material is associated with a change in the superconducting properties as T_K is reduced.

I. INTRODUCTION

The ground state properties of heavy electron (HE) compounds are often very sensitive to doping with small quantities of impurity atoms. Perhaps the most unusual case is that of $U_{1-x}Th_xBe_{13}$, where Th substitution produces[1] both a nonmonotonic depression of the superconducting transition temperature T_c and a second phase transition below T_c for $0.019 \leqslant x \leqslant 0.043$. Recent muon spin rotation (μSR) experiments[2] have demonstrated that this lower phase possesses small magnetic moments, of order 10^{-3}–10^{-2} μ_B/U-atom. Thus the magnetism and superconductivity are closely coupled in this system. Recently it was reported[3] that substitution of B for Be in $UBe_{12.97}B_{0.03}$ drastically increases the specific heat jump at T_c, and it was surmized that magnetic correlations were also produced in $UBe_{12.97}B_{0.03}$, as in $(U,Th)Be_{13}$. In this paper we report μSR and specific heat measurements in $U_{1-x}Th_xBe_{13-y}B_y$ for $x = 0.019$ and $y = 0.03$, to further investigate these phenomena.

The μSR experiments were carried out in zero applied field between temperatures 0.05 and 1.7 K at the low-temperature facility (LTF) of the Paul Scherrer Institute. The samples for μSR were arc-melted polycrystalline ingots about 1 mm thick with 2 cm^2 cross-sectional area.[2] The specific heat data were collected between $T = 0.3$ K and 20 K using a small-mass calorimeter.[4]

II. SPECIFIC HEAT DATA

Figure 1 shows the temperature dependence of the specific heat for pure UBe_{13}, $UBe_{12.97}B_{0.03}$(UBeB), and $U_{0.981}Th_{0.019}Be_{12.97}B_{0.03}$ (UThBeB) between 0.3–20 K. The UBe_{13} data show the characteristic features of a rise in C/T below about 6 K associated with the Kondo resonance, followed by a superconducting specific heat anomaly with an onset temperature $T_c \approx 0.91$ K. (The midpoint of the rise in C/T occurs at about 0.82 K.) The UBeB data, although qualitatively similar, are quantitatively different in several important ways. First, the gentle rise below 6 K in C/T reflecting the Kondo anomaly is pushed to lower temperatures in UBeB. Second, C/T at T_c is somewhat larger in UBeB than in UBe_{13}; and finally, the jump in specific heat ΔC at T_c is much larger in UBeB than in UBe_{13}. The T_c from either the onset or midpoint of the jump in C/T is unchanged and x-ray analysis does not show a distinguishable difference between UBe_{13} and UBeB. The resistivity maximum at about 2.5 K in UBe_{13} is shifted to a slightly lower temperature in UBeB, however. When Th is added to UBeB T_c is reduced to about 0.6 K and two specific heat transitions are clearly visible below 0.6 K. The latter is similar to specific heat data reported previously for unborated $U_{1-x}Th_xBe_{13}$ ($0.019 \leqslant x \leqslant 0.043$).

III. μSR DATA

The μSR time differential spectra were analyzed[2] using the standard zero-field Kubo–Toyabe relaxation function, which gives very good fits to the data. The μSR relaxation rate $\sigma(\mu s^{-1})$ in pure UBe_{13} is due to inhomogeneous broadening from the ^{9}Be nuclear-dipole-field distribution at the μ^+ site. No change in relaxation rate σ for $T < 8$ K has been observed[2] in UBe_{13}. Figure 2 shows σ as a function of temperature for UBeB, UThBeB, and UThBe. As in UBe_{13}, UBeB shows no change in σ between 0.05 and 1.7 K. When Th is added to the system, however, the μSR rate rises dramatically below about 0.4 K, as reported previously[2] for $(U,Th)Be_{13}$. This rise in σ is due to the onset of magnetic correlations associated with the lower-temperature phase in $(U,Th)Be_{13}$, as mentioned in the introduction.

IV. ANALYSIS AND CONCLUSIONS

Although the substitution of a few percent B for Be or Th for U in UBe_{13} produces a significant increase in ΔC at T_c compared to pure UBe_{13}, only Th produces an onset of magnetic correlations, at least for the boron concentration studied here. This may be due to the fact that, unlike B, Th is substituted at the f-electron site. We confine the remainder of this discussion to a comparison of UBe_{13} and UBeB,

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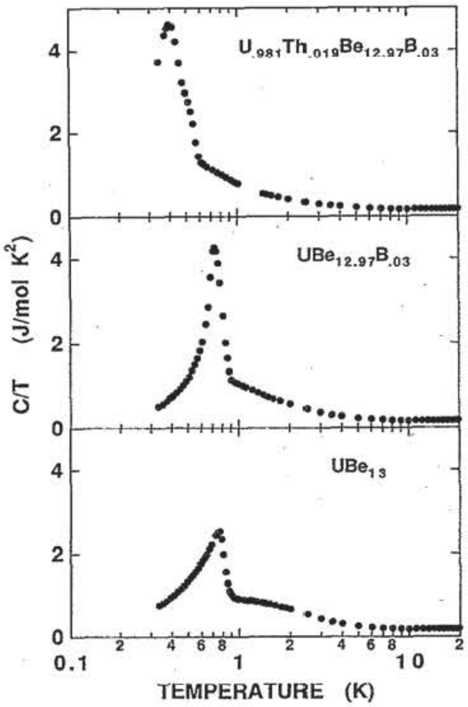

FIG. 1. Temperature dependence of specific heat per Kelvin C/T.

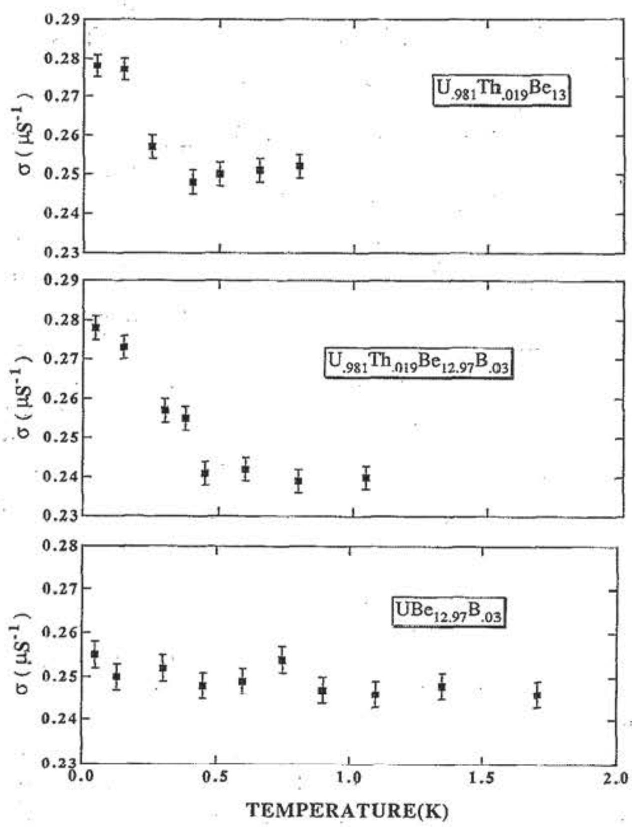

FIG. 2. μSR Kubo–Toyabe relaxation rate σ as a function of temperature.

where no magnetism is present, and the interpretation of the specific heat data is therefore less complicated.

The entropy S removed by the superconducting transition is given by[5]

$$S(T_c)=\int_0^{T_c}\frac{C}{T}dT. \quad (1)$$

One must have $S(T_c) = \gamma T_c$ to conserve entropy, where at low temperatures in a free electron picture the Sommerfeld constant γ is independent of temperature. Table I gives $S(T_c)$, $\gamma(T_c)$, and T_c, showing that in our UBe_{13} sample entropy is not quite conserved with the assumption of a temperature-independent γ. The entropy conservation is somewhat worse in UBeB. This indicates that the heavy-electron state is still forming when the superconducting transition occurs [i.e., γ(T) is increasing as T decreases].

If we assume for simplicity that γ increases linearly below T_c, then we arrive at a value $\overline{\gamma}(T_c/2)$ necessary to conserve entropy in the superconducting transition [i.e., $S(T_c)=\overline{\gamma}T_c$]. One obtains $\overline{\gamma}(T_c/2) = 1.17$ and 1.35 J/mol K^2 for UBe_{13} and UBeB, respectively (Table I). Thus adding B to UBe_{13} increases the low-temperature density of states, which is proportional to γ. This could happen if the Fermi energy is changed as electrons are added to the conduction band with B doping, or through a shift in the Kondo temperature T_K, or both. As noted above, however, the shape of the C/T data between about 1 and 6 K indicates that T_K has been lowered in UBeB. We note that the total entropy S(20 K) released up to 20 K is the same within 5% for the two systems (Table I).

The specific heat jump ΔC is given by[5] the difference in the rate of change of entropy with temperature above and below T_c:

$$\Delta C=T_c\left[\left(\frac{\partial S}{\partial T}\right)_s-\left(\frac{\partial S}{\partial T}\right)_n\right]=\alpha\gamma T_c. \quad (2)$$

In BCS theory $\alpha = 1.43$ for the assumption of weak S-wave coupling. Taking $\gamma=\overline{\gamma}(T_c/2)$ in Eq. (2) one has $\alpha = 1.5$ and 2.5 in UBe_{13} and UBeB, respectively. Thus the large ΔC in UBeB is not simply a consequence of an increased density of states (larger $\overline{\gamma}$), but reflects a significant change in the properties of the superconducting state. This is evident from a plot of $S(T)$ (not shown). The slope $(\partial S/\partial T)_s$ just below T_c is seen to be significantly larger in

TABLE I. Specific heat data for $UBe_{13-y}B_y$. Symbols are defined in the text.

	UBe_{13}	$UBe_{12.97}B_{0.03}$
T_c(K)	0.91	0.91
$\gamma(T_c)$ (J/mol K^2)	1.04	1.13
$\gamma(T_c)\cdot T_c$(J/mol K)	0.95	1.03
$\overline{\gamma}(T_c)$ (J/mol K^2)	1.17	1.35
$S(T_c)$ (J/mol K)	1.06	1.23
S(20 K) (J/mol K)	5.91	5.69
$\Delta C/(\overline{\gamma}\cdot T_c)$	1.5	2.5

UBeB than in UBe_{13}, whereas the slope $(\partial S/\partial T)_n$ above T_c is roughly the same in the two materials.

The actual values of α extracted above clearly depend on the linear extrapolation of γ below T_c, where an average $\overline{\gamma}$ was used in Eq. (2). Therefore, it is the change in α between UBe_{13} and UBeB which is most significant, and not its absolute magnitude. The conclusion that α changes significantly with B doping is valid only as long as $\gamma(T)$ varies smoothly below T_c in each material. Only a very anomalous temperature dependence for $\gamma(T)$, such as a rapid drop below T_c followed by a rise as T approaches zero, would invalidate our conclusions, however.

We note again that T_c is unchanged with B doping for this sample. Previously, a depression of T_c to about 0.77 K, accompanied by a smaller and broader (in temperature) ΔC jump, was reported[3] for $UBe_{13-y}B_y$ ($y=0.03$). Nevertheless, an enhanced value of α was also observed in this UBeB sample,[3] though less than in the present sample. The differences from sample to sample are not yet understood. All samples in the present study, however, were prepared at the same time with the same materials and were the same size.

In conclusion, we have shown that both Th and B depress the Kondo temperature in $U_{1-x}Th_xBe_{13-y}B_y$ and lead to an enhanced specific jump ΔC compared to UBe_{13}. Only Th induces detectable magnetic correlations, however. The enhancement of ΔC in UBeB is larger than expected from an increase in the density of states at the Fermi surface, reflecting a change in the superconducting properties. This change could be due to an increase in the strength of the pairing or to a softening of the characteristic mode frequency associated with the pairing interaction. The fact that changes in the superconducting state appear to be associated with changes in the Kondo temperature provide evidence for the importance of magnetic excitations in the superconducting pairing interaction in UBe_{13}.

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