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CONNECTIONS BETWEEN MAGNETISM AND SUPERCONDUCTIVITY IN UBe_{13} DOPED WITH THORIUM OR BORON

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ABSTRACT

Magnetism and superconductivity appear to be intimately connected in the heavy electron (HE) superconductors. For example, it has been conjectured but not proven that the exchange of antiferromagnetic spin fluctuations are responsible for pairing in HE superconductors. In this paper we review recent results in $U_{1-x}Th_xBe_{13}$, where specific heat, lower critical field and zero-field μSR measurements reveal another second-order phase transition (below the superconducting transition) to a state which possesses small-moment magnetic correlations for $0.019 \leq x \leq 0.043$. We present a new phase diagram for $(U,Th)Be_{13}$ which indicates that the superconducting and magnetic order parameters are closely coupled. A discussion of the nature of the lower phase is presented, including the consideration of a possible magnetic (time-reversal-violating) superconducting state.

When UBe_{13} is doped with B ($UBe_{12.97}B_{0.03}$) the Kondo temperature is decreased and the specific heat jump at the superconducting transition temperature is significantly enhanced. However, μSR measurements reveal no magnetic signature in $UBe_{12.97}B_{0.03}$, unlike the case for Th doping. The correlation between changes in the Kondo temperature and changes in the superconducting properties induced by B doping provide evidence for the importance of magnetic excitations in the superconducting pairing interaction in UBe_{13}.

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I. INTRODUCTION

The existence[1] of small-moment, f-electron magnetism in the undoped heavy-electron (HE) superconductors $CeCu_2Si_2$, UPt_3 and URu_2Si_2 suggests intriguing connections between magnetism and superconductivity in HE materials. One manifestation of this connection is explicit in UPt_3, where recent neutron scattering experiments[2] have shown that the antiferromagnetic and superconducting order parameters are coupled. However, the general suggestion that antiferromagnetic spin fluctuations are principally responsible for pairing in HE superconductors has not yet been proven. In this paper we review the effects on the superconductivity of doping UBe_{13} with small quantities of the impurity atoms Th and B. We discuss a new phase diagram[3] for $(U,Th)Be_{13}$, which indicates that magnetism and superconductivity are closely coupled in this system as well. Possible interpretations of the magnetic phase found in the superconducting state of $(U,Th)Be_{13}$ are reviewed, including the possibility that a magnetic (time-reversal-violating) superconducting state exists. We also show that when UBe_{13} is doped with B ($UBe_{12.97}B_{0.03}$), the Kondo temperature is decreased and the specific heat jump ΔC at the superconducting transition is significantly enhanced, indicating a possible connection between magnetic excitations and the superconducting pairing interaction in UBe_{13}.[4]

II. $(U,Th)Be_{13}$

Substitution of Th for U in $U_{1-x}Th_xBe_{13}$ produces[5] a non-monotonic depression of the superconducting transition temperature T_{c1}, accompanied by a second phase transition at $T_{c2} < T_{c1}$ for $0.019 \le x \le 0.043$. Theoretical interpretations of this second phase have included a coexisting antiferromagnetic spin-density-wave state,[6] a transition to a second superconducting state possessing orbital[7] or spin[8] magnetic moments, or small local moments on the uranium[9] or thorium[10] sites. Here we test these hypotheses against zero-field muon-spin-resonance (μSR) and lower-critical-field (H_{c1}) data across a broad range of Th concentrations: x = 0.0000, 0.0066, 0.0100, 0.0193, 0.0245, 0.0355, and 0.0600.

The μSR experiments were carried out at the Paul Scherrer Institute using the surface muon beam at the low temperature facility. The experimental setup, data analysis, and sample preparation are discussed elsewhere.[11] The measured zero-field muon spin relaxation functions were well described[11] by fits to the Kubo-Toyabe relaxation function

$$G_{KT}(t) = 1/3 + 2/3\ (1 - \sigma_{KT}^2 t^2)\ \exp\ (-\sigma_{KT}^2 t^2/2), \qquad (1)$$

appropriate for inhomogeneous broadening. Here σ_{KT} is proportional to the root-mean-square field distribution $(\Delta H)_{rms}$ at the muon site, and $\sigma_{KT} = \gamma_\mu (\Delta H)_{rms}$, where γ_μ is the muon gyromagnetic ratio ($8.51 \times 10^4\ s^{-1}Oe^{-1}$). The exact stopping site of the muon is not known. The H_{c1} measurements were performed at the Kamerlingh Onnes Laboratory using a flux-gate magnetometer and a ^{3}He cryostat. Long, thin cylindrical samples requiring negligible demagnetization corrections were cut from the same batches as used for the μSR experiments. The H_{c1} values were consistently obtained[3] both as the first derivation (2%) from linearity of the initial shielding curve following zero-field cooling and by using a different procedure based on the Bean critical-state model. The critical temperatures determined in various ways are given in Table I.

The temperature dependence of σ_{KT} for x = 0.035 is shown in Fig. 1, together with ac susceptibility showing the onset of superconductivity below T_{c1} and specific heat showing a second phase transition below T_{c2}. The μSR measurements show a constant relaxation rate above T_{c2} (due to nuclear dipolar broadening from ^{9}Be) and the onset of an additional magnetic field of electronic origin below T_{c2},. The temperature dependence of the electronic contribution to the μSR linewidth is given by

$$s(t) = \sigma_e(T)/\sigma_e(0), \qquad (2)$$

where $t = T/T_{c2}$ and $\sigma_e^2(T) = \sigma_{KT}^2(T) - \sigma_{KT}^2(T_{c2})$. Eqn. (2) expresses the assumption that the nuclear and electronic (σ_e) contributions to σ_{KT} are uncorrelated. The additional field is about 1.8 Oe, corresponding to an electronic moment of order $(10^{-3}\text{-}10^{-2})\mu_B$/U atom, under the assumption of dipolar coupling to the muon. The value s(t) is plotted in Fig. 2, showing that the transition is clearly second order, ie., typical of a continuous order parameter. The solid curve in Fig. 2 is consistent[11] with a mean-field theory of magnetic order and also is numerically consistent with the pairing amplitude (or order parameter) in the BCS theory of superconductivity.

Fig. 3 shows[3] the temperature dependence of σ_{KT} for all of the samples studied. One sees that the μSR linewidth is temperature independent except for those Th concentrations where two specific jumps are seen (x = 0.0193, 0.0245, 0.0355). In each of these cases the μSR linewidth increases below T_{c2}. Furthermore, the extrapolated zero-temperature linewidths $\sigma_e(0)$ increase with Th concentration, as given in Table II.

The $H_{c1}(T)$ data for x = 0.0000, 0.0066, 0.010 show a single quadratic temperature dependence $H_{c1} \propto (1 - \bar{t}^2)$ over the entire temperature range measured (about 0.3 K $\leq T \leq T_{c1}$). Here $\bar{t} = T/T_{c1}$. However, two regions of quadratic temperature dependence are observed for those materials where two specific heat peaks are seen and the μSR linewidth increases below T_{c2}. The $H_{c1}(t)$ vs. T^2 are shown in Fig. 4 for x = 0.0000 and x = 0.0355. This latter behavior in H_{c1} is qualitatively similar to that observed previously[12] for x = 0.033. Values for the slopes $|dH_{c1}/dt^2| = H_{c1}(0)$ are given in Table I, where $H_{c1}^L(0)$ and $H_{c1}^H(0)$ refer to the low and high temperature slopes, respectively. We note that $H_{c1}^L(0)$

increases with x, as does the μSR linewidth σ_e, for x = 0.0193, 0.0245, and 0.0355.

Based upon these data, augmented with specific heat results[13] for other Th concentrations, we have constructed[3] the overall T-x phase diagram for $U_{1-x}Th_xBe_{13}$ as shown in Fig. 5. The uncertainties in x are about 0.005. We draw the following conclusions. (1) There are steep phase boundaries separating magnetic from non-magnetic regions near x = 0.019 and 0.043, between which two specific heat peaks are seen. (2) The fact that within errors the transitions at T_{c2} begin and terminate on the line of superconducting phase transitions at T_{c1} means that the order parameters for the two phases must be strongly coupled.

We now discuss the nature of the phase below T_{c2}. Taking into account the observation of electronic magnetism below T_{c2} and the large specific heat anomaly associated with this transition (comparable to that at T_{c1}), two plausible possibilities for this phase suggest themselves. The first is an antiferromagnetic transition accompanied by a superconducting phase transition, and the second is a transition to a magnetic (time-reversal-violating) superconducting phase. Both of these possibilities require an unconventional or multicomponent superconducting order parameter. A third possibility, that there is only an antiferromagnetic spin-density-wave phase transition but no change in the superconducting phase below T_{c2} seems unlikely for the following reason. The large specific heat jump ΔC at T_{c2} would be very surprising for a spin-density-wave state because the Fermi surface is largely consumed by the superconducting transition at T_{c1}. Thus a large ΔC would require an exceptional enhancement of the density of states near the zeros of the superconducting gap.

The observation of electronic magnetism below T_{c2} can thus be associated with either an antiferromagnetic transition in conjunction with a new superconducting phase or a magnetic superconducting phase. Regarding the former case we note that if the moments were associated with the thorium sites[10] (as "Kondo holes", for example) then the dipolar linewidth $\sigma_e(0)$ should be proportional to x, which is not observed (Table II). Consequently, under the assumption of an antiferromagnetic phase coexisting with superconductivity, the moments are most likely on the uranium sites.

A multi-component, complex superconducting order parameter for (U,Th)Be_{13} could also in principle explain the observed T-x phase diagram. Important experimental facts are that both $\sigma_e(0)$ and $H_{c1}^{L}(0)$ increase with x below T_{c2}, and that the magnetic phase is induced by doping with nonmagnetic Th impurities. We note that $H_{c1} \propto n_s/m^*$, where n_s is the superfluid density and m* is the effective mass. If $n_s(0)$ increases with x, the correlation between $H_{c1}^{L}(0)$ and $\sigma_e(0)$ might be explained by recent theoretical models[14-15] in which orbital currents are induced when electron scattering from nonmagnetic impurities distorts the superconducting order parameter in a complex superconducting phase. The induced currents (and hence the dipolar field $|\underset{\sim}{B}_L| \propto \sigma_e$) would be proportional to $n_s(0)$. If the field sensed by the muon, averaged over the sample volume, were nearly random in direction and magnitude then one would observe a sublinear dependence of $\sigma_e(0)$ on $n_s(0)$, which is seen in the roughly square-root correlation between $\sigma_e(x)$ and $H_{c1}^{L}(x)$ (Table II). Complete randomness would yield $\sigma_e(x) \propto \sqrt{H_{c1}^{L}(x)}$ for a Gaussian distribution.

It is also possible that the increase in $H_{c1}(0)$ with x could be due to a decrease in m*, as expected for an antiferromagnetic transition.[11] However, we note that H_{c1} follows a T^2 law both above and below T_{c2}, which is the T-dependence expected for a change in n_s. If m* changes at T_{c2} it would have to change abruptly at this temperature and not evolve significantly in temperature below T_{c2}. This seems unlikely. However, it is not possible to predict how m* should change with temperature or with x without a detailed microscopic theory.

III. UBe_{13} doped with boron.

A second striking example for the effects of impurity doping on the superconducting properties of UBe_{13} is in the substitution of B for Be. Initial studies[16] of $UBe_{12.97}B_{0.03}$ showed a depression of T_c to about 0.77 K (initial onset), accompanied by an enhanced though broader (in temperature) specific heat jump ΔC compared to pure UBe_{13}. In this paper we review more recent experiments[4,17] on different samples of $UBe_{13-y}B_y$. Figure 6 shows the temperature dependence of the specific heat (plotted as C/T vs. log T) for pure UBe_{13} together with $UBe_{12.97}B_{0.03}$ (UBeB) and $U_{0.981}Th_{0.019}Be_{12.97}B_{0.03}$ (UThBeB). Several differences between pure UBe_{13} and the doped samples are immediately apparent from Fig. 6. First, both UBeB and UThBeB show an enhanced linear coefficient of specific heat γ at the onset of superconductivity, compared to UBe_{13} (see Table III). Second, the Kondo temperature T_K, is reflected in the rise of C/T (the shoulder below 6 K in UBe_{13}, for example), reduced by doping. Third, Th doping produces both two specific heat peaks for the concentration shown and a reduction in T_c, while B doping does neither. Finally, Th and B doping each produce a larger ΔC.

A possible explanation for the enhanced ΔC in UBeB is that a second (magnetic) transition is induced by B doping, as in $(U,Th)Be_{13}$. This hypothesis was tested with μSR. As seen in Fig. 7, only Th doping induces an enhanced μSR linewidth and hence a magnetic signature below T_{c2}. This fact, plus the narrowness in the specific heat anomaly for UBeB indicates that only a <u>single</u> transition with an enhanced γ and ΔC exists in UBeB. Recently, Beyermann <u>et.al</u>[17] have shown that the ΔC enhancement appears to be largest for B concentrations near $UBe_{12.97}B_{0.03}$, and that the addition of B increases the high temperature effective moment in the magnetic susceptibility, consistent with a reduction of T_K.

We now compare UBe_{13} and UBeB, both cases where no electronic magnetism is observed by μSR. The discussion focuses on examining the connection between a change in T_K and a change in the superconducting properties with B doping.

Table III gives a summary of the relevant thermodynamic parameters for UBe_{13} and UBeB. While T_c is essentially unchanged for our samples, γ at T_c is enhanced by about 10% in UBeB, whereas the entropy $S(T_c)$ released up to T_c is about 15% larger in UBeB. Furthermore, because $S(T_c) = \gamma T_c$ for a temperature independent γ, entropy is not quite conserved (for a constant γ) in either material (see Table III). For simplicity, we have defined a value $\bar{\gamma}(T_c/2)$ necessary to conserve entropy (ie., $S(T_c) = \bar{\gamma}T_c$), assuming that γ increases linearly below T_c. The fact that γ is not temperature independent, indicates that the heavy electron state is still forming when the materials becomes superconducting, ie., that T_K is comparable to T_c. Thus one can compare the relative specific heat jumps in the two materials, where $\Delta C = \beta\bar{\gamma}T_c$ and β is related to the strength of the pairing interaction. We find $\beta \approx 1.5$ in UBe_{13} and ≈ 2.5 in UBeB, compared to 1.43 for the weak-coupling BCS case.[4] For strong coupling the value of β is given approximately by[18]

$$\beta = 1.43\ [1+53(T_c/\omega_0)^2\ln(\omega_0/3T_c)], \tag{3}$$

where ω_0 is the characteristic boson frequency for the pairing interaction. One obtains $\omega_0 \approx 4$ meV for UBe_{13} and $\omega_0 \approx 0.7$ meV for UBeB. For comparison ω_0 is about 25 meV, 15 meV and 4 meV in Al, V and Pb, respectively.[18] Thus, B doping at this concentration may significantly reduce ω_0.

Another useful comparison arises between β and the quantity $2\Delta_0/k_BT_c$, for which there appears to be a universal relation[18] for crystalline superconductors. Here Δ_0 is the zero-temperature value of the superconducting gap energy. Using this relation one finds $2\Delta_0/k_BT_c \approx 3.6$ and 4.4 for UBe_{13} and UBeB, respectively, compared to the BCS value of 3.53. Thus $UBe_{12.97}B_{0.03}$ appears to be a strong-coupling superconductor, whereas UBe_{13} is not. The fact that T_c is not significantly reduced as ω_0 is reduced may be accidental, but may also be explained qualitatively by observing that[18] $T_c \propto \omega_0 \exp(-1/N(o)V)$, and that a reduction in ω_0 may be offset by an increase in either the pairing potential V or the electronic density of states N(o). Further experimental and theoretical work are clearly required to clarify this issue.

In conclusion we note that a reduction in T_K is accompanied by a reduction in ω_0 and an enhancement of the specific heat jump in UBeB compared to UBe_{13}. This is qualitatively consistent if the superconducting pairing interaction in UBe_{13} is driven largely by spin fluctuations. Such a case has been hypothesized for HE systems, though no direct evidence (comparable to the isotope effect in BCS superconductors) has yet been produced. In this regard, we note that measurements of the specific heat of UBe_{13} under pressure[19] increase T_K while producing a reduced specific heat jump, yielding further evidence for this hypothesis.

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Figure Captions

Fig. 1. Temperature dependence of (a) zero-field μSR linewidth σ_{KT}, (b) specific heat and (c) ac susceptibility in $U_{0.965}Th_{0.035}Be_{13}$.

Fig. 2. Dependence on reduced temperature t of normalized zero-field , ;R linewidth s(t) from Eqn. 2.

Fig. 3. Temperature dependence of zero-field μSR linewidth σ_{KT} in $U_{1-x}Th_xBe_{13}$.

Fig. 4. Lower critical field $H_{c1}(T)$ plotted vs. T^2 in $U_{1-x}Th_xBe_{13}$ for x = 0.0000 (top) and x = 0.0355 (bottom). The lines are guides to the eye.

Fig. 5 Phase diagram for $U_{1-x}Th_xBe_{13}$. Open symbols are from this work. Squares, T_{c1} from χ_{ac}; circles, T_{c1} from magnetization M(H); inverted triangles, T_{c2} from kink in $H_{c1}(T^2)$. The solid upright triangles are T_{c1} and T_{c2} from specific heat in Ref. 13. The symbol (Δ) at x = 0.043 indicates a merging of T_{c1} and T_{c2}, as described in Ref. 13. T_{c1} = 0.39 K for x = 0.0600 was determined resistively.

Fig. 6. Temperature dependence of specific heat per Kelvin C/T.

Fig. 7. Temperature dependence of zero-field μSR linewidth (σ_{KT} in text.)

Table I

Th(%)	T_{c1}(K) χ_{ac}	T_{c1}(K) M(H)	T_{c2}(K) M(H)	$H_{c1}^{L}(o)$ (mT)	$H_{c1}^{H}(o)$ (mT)
0.00	0.86	0.86			4.32
0.66	0.67	0.67			3.27
1.01	0.65	0.65			2.64
1.93	0.48	0.48	0.44	3.79	2.28
2.45	0.58	0.59	0.41	4.91	2.89
3.55	0.55	0.55	0.39	5.59	3.53

Collected parameters and transition temperatures of $U_{1-x}Th_xBe_{13}$.

The notation is explained in the text.

Table II

x(%)	x/1.93	$\sigma_e(x)/\sigma_e(1.93)$	$[H_{c1}^L(x)/H_{c1}^L(1.93)]^{1/2}$
1.93	1.00	1.00	1.00
2.45	1.27	1.11 ± 0.06	1.14 ± 0.07
3.55	1.84	1.31 ± 0.07	1.21 ± 0.07

The x dependence of σ_e and $[H_{c1}^L]^{1/2}$ at T = 0 in $U_{1-x}Th_xBe_{13}$.

Table III

	UBe_{13}	$UBe_{12.97}B_{0.03}$
T_c(K)	.91	.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
$S(T_c)$ (J/mol·K)	1.06	1.23
$\bar{\gamma}(T_c/2)$ (J/mol·K^2)	1.17	1.35
$\Delta C/(\bar{\gamma}\cdot T_c)$	1.5	2.5

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

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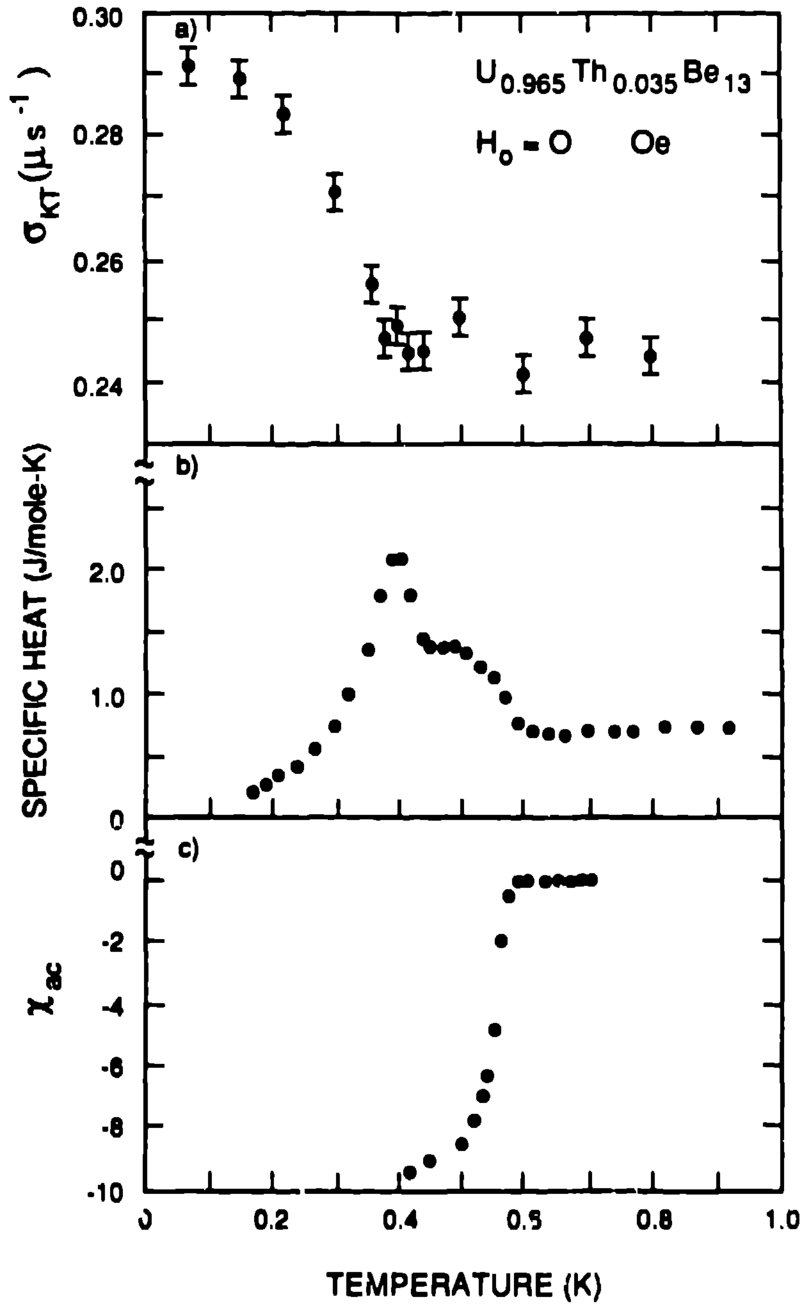

a)
$U_{0.965}Th_{0.035}Be_{13}$
H_o = O Oe
$\sigma_{KT}(\mu s^{-1})$
0.30
0.28
0.26
0.24
b)
SPECIFIC HEAT (J/mole-K)
2.0
1.0
0
c)
χ_{ac}
0
-2
-4
-6
-8
-10
0
0.2
0.4
0.5
0.8
1.0
TEMPERATURE (K)

1 - 1

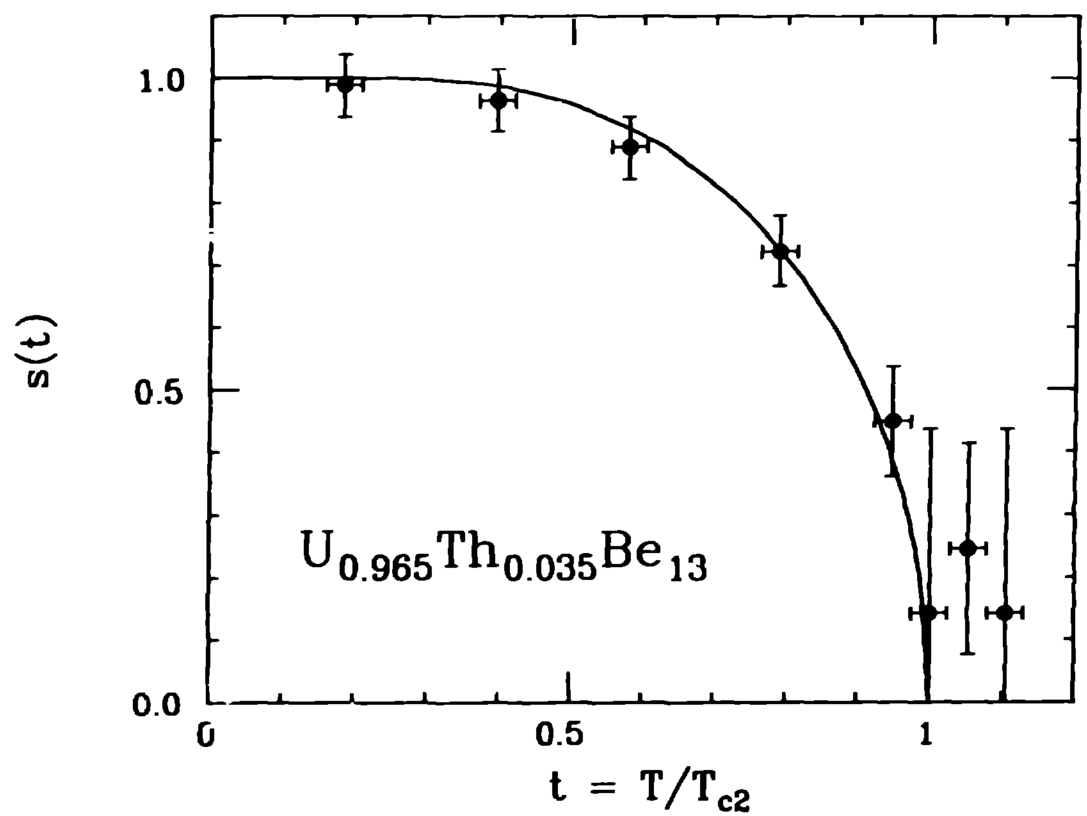


Fig. 2

IS-12/90-702

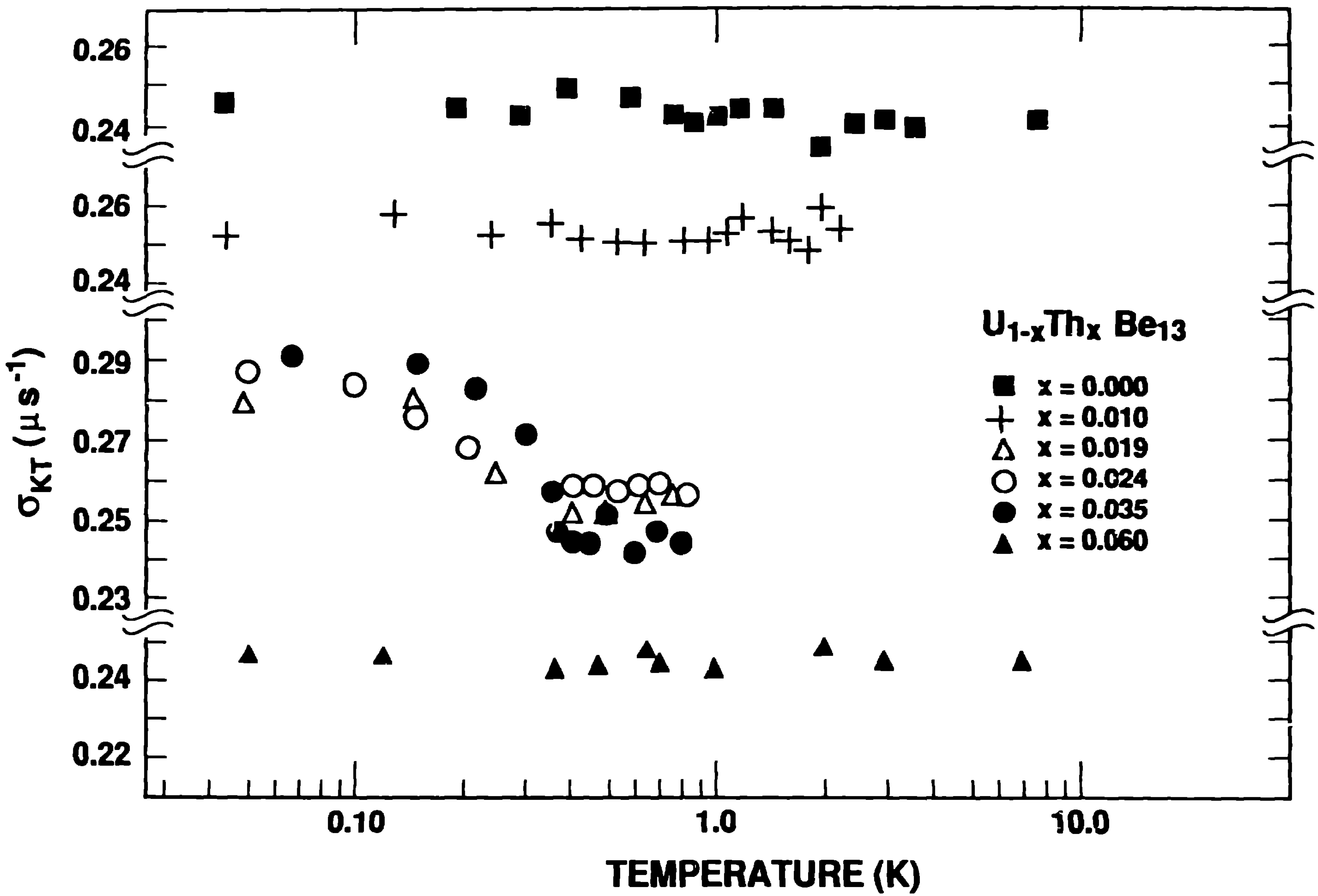


Fig. 2

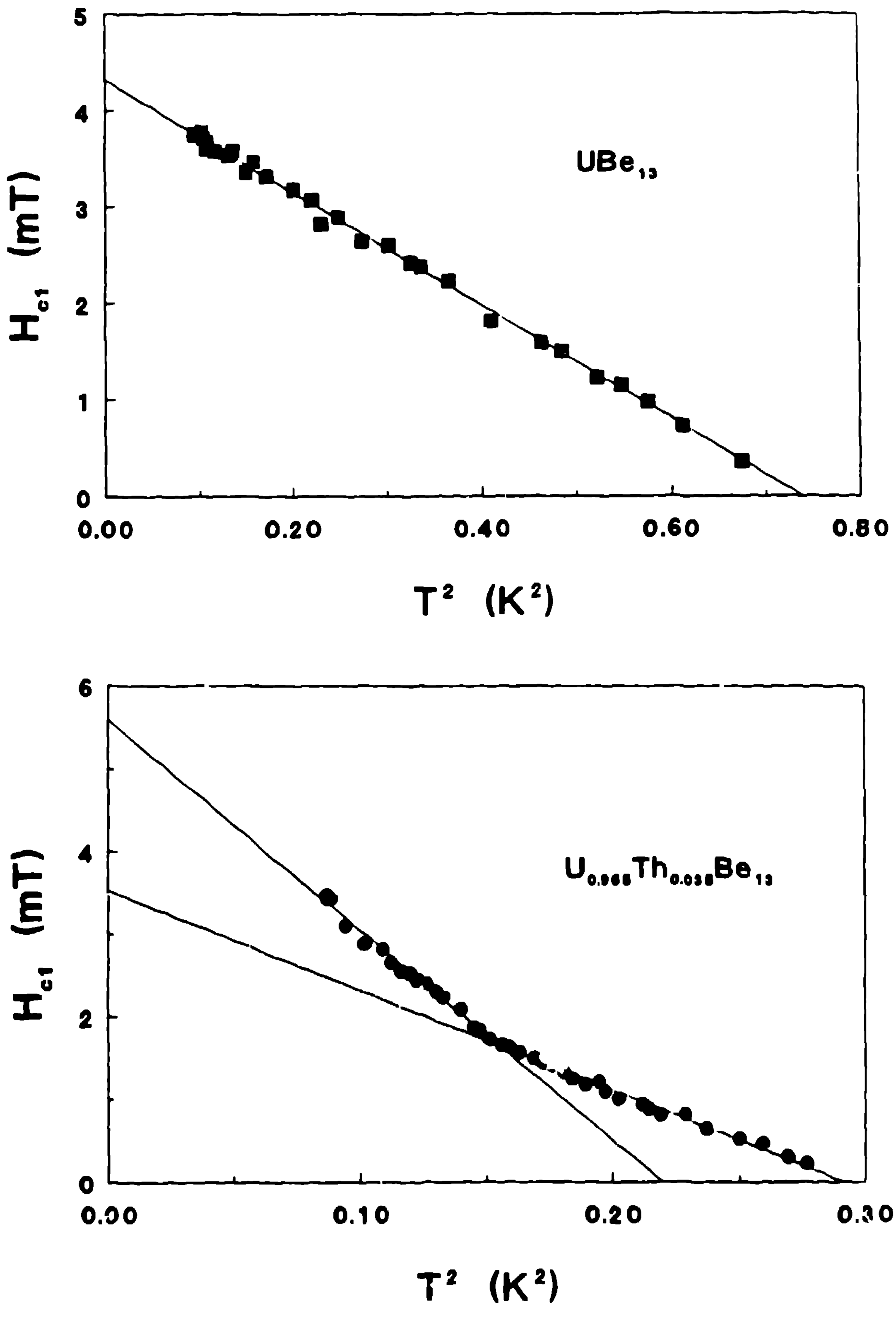


Fig. 4

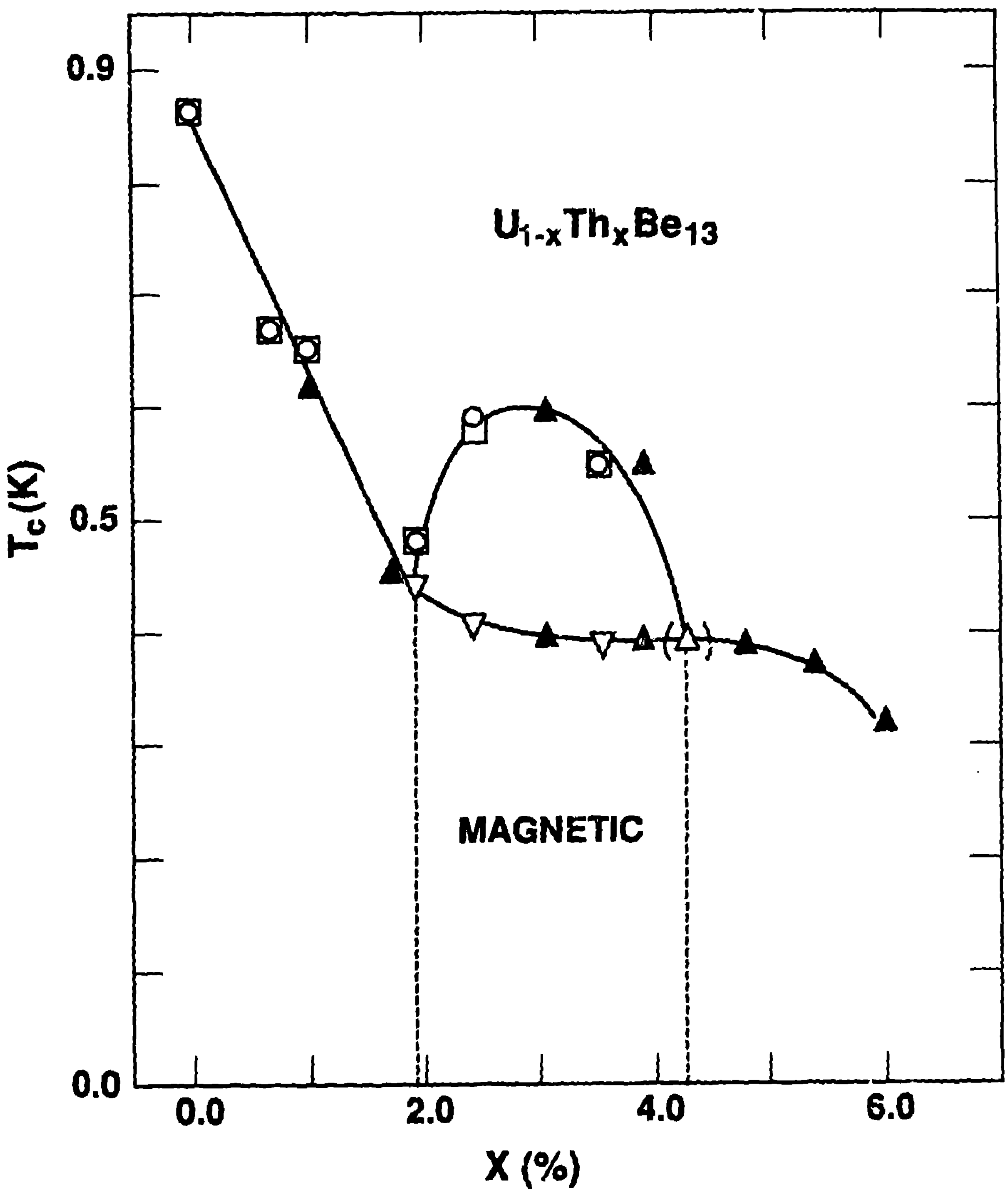
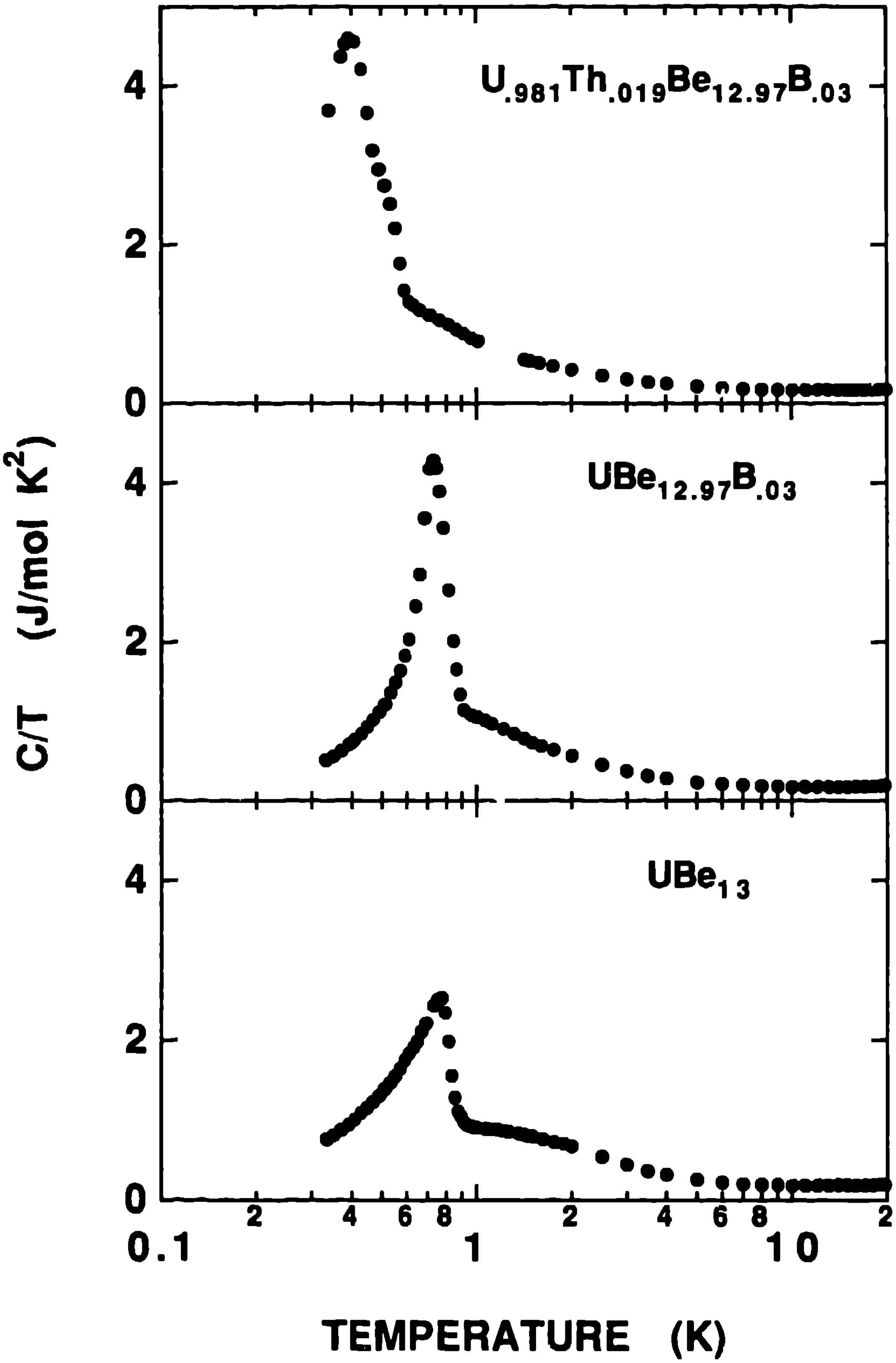
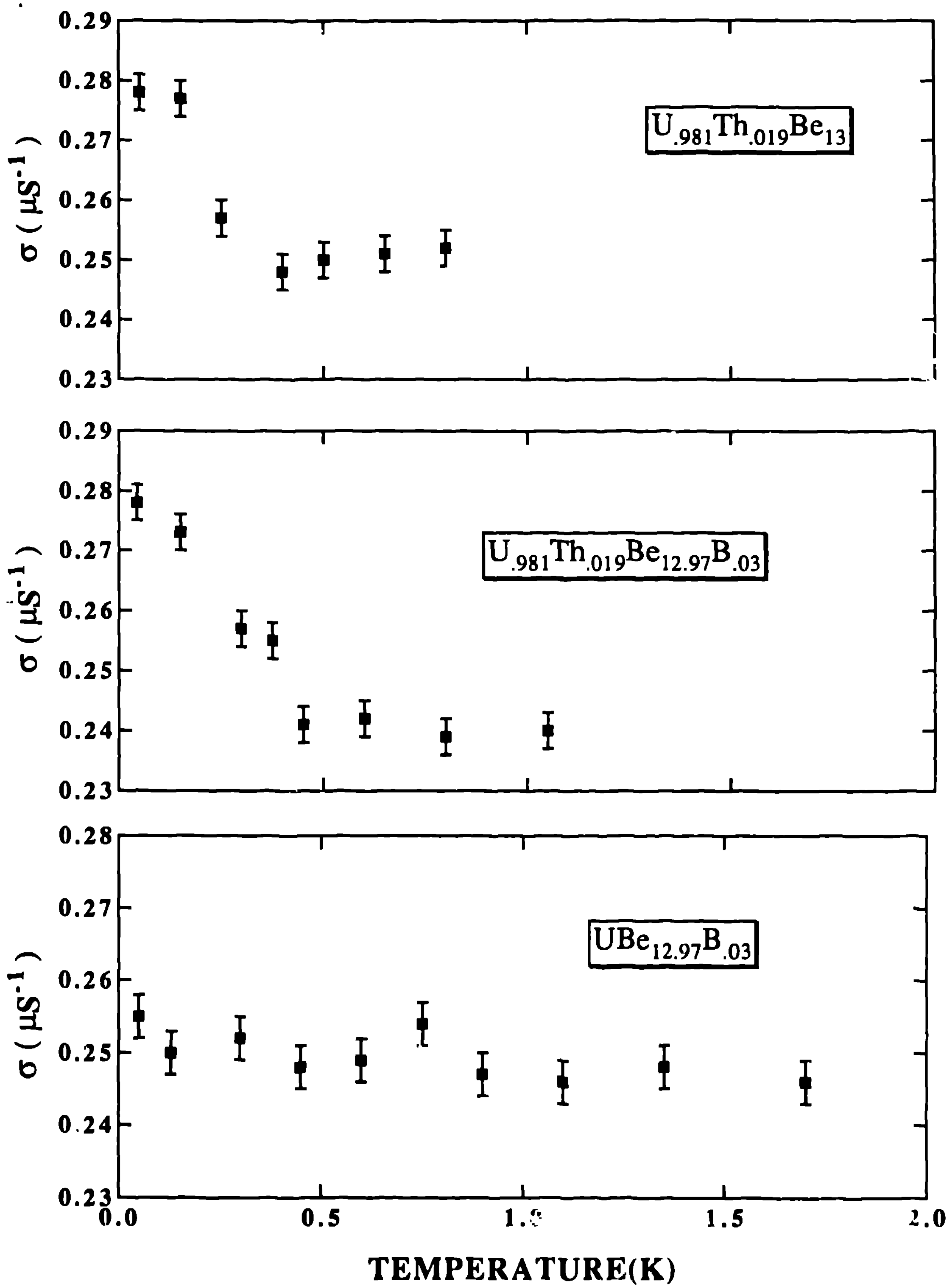


Fig. 5



$U_{.981}Th_{.019}Be_{12.97}B_{.03}$
$UBe_{12.97}B_{.03}$
UBe_{13}
C/T (J/mol K^2)
0
2
4
0.1
1
10
2
4
6
8
TEMPERATURE (K)

FIG. 6



$U_{.981}Th_{.019}Be_{13}$
$U_{.981}Th_{.019}Be_{12.97}B_{.03}$
$UBe_{12.97}B_{.03}$
σ (μs⁻¹)
0.29
0.28
0.27
0.26
0.25
0.24
0.23
0.0
0.5
1.0
1.5
2.0
TEMPERATURE(K)

fig 4