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THE LOW-TEMPERATURE PHASE DIAGRAM OF $U_{1-x}Th_xBe_{13}$

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Measurements of specific heat $c_p(T)$ and thermal conductivity $\lambda(T)$ give a detailed picture of the $T_c(x)$ phase diagram of superconducting $U_{1-x}Th_xBe_{13}$ below 1 K. A distinct minimum for T_c is noted for $x \approx 0.018$. Increasing x further results in increasing values for T_{c1} and the occurrence of a second phase transition in the superconducting state at $T_{c2} < T_{c1}$. Two phase transitions are observed between $0.02 < x < 0.045$. T_{c2} is roughly constant with varying x. For $x > 0.05$, only one single transition occurs and T_c decreases with further increasing x.
Anomalous behaviour is also observed for $UBe_{13-y}B_y$ for y values between 0 and 0.05 in the sense that the c_p anomalies at the superconducting transition are distinctly larger than the corresponding discontinuity observed in pure UBe_{13}. The influence of B-doping on the phase diagram of $U_{1-x}Th_xBe_{13}$ has also been studied.

UBe_{13} is a heavy-electron superconductor whose properties strongly suggest that unconventional ideas are necessary to understand its behaviour[1]. One of the most intriguing features is the variation of the critical temperature T_c upon replacing U by Th on the few atomic-% level. While measurements of the electrical resistivity $\rho(T)$ and magnetic susceptibility $\chi(T)$ [2] indicate a non-monotonic variation of T_c of $U_{1-x}Th_xBe_{13}$ as a function of x, specific-heat $c_p(T)$ experiments[3] confirm this behaviour and reveal the appearance of a second phase transition in the superconducting state within a limited range of x. Here we report some progress that has been made in investigations of this $[T_c,x]$ phase diagram.

In fig. 1 we show the phase diagram as it results from various measurements of the low-temperature specific heat $c_p(T)$ of $U_{1-x}Th_xBe_{13}$ compounds for $0 < x < 0.06$. The initial depression of T_c at low x values, which is characteristic also for most of the other impurities that we have studied, terminates in a well defined minimum at $x = 0.018$ and for further increasing x, $T_{c1}(x)$, which denotes the transition to the superconducting state, displays a broad maximum and a subsequent continuation of decreasing $T_{c1}(x)$ for $x \geq 0.035$. The appearance of a second phase transition at $T_{c2} < T_{c1}$ is, with increasing x, first observed for $x = 0.0205$. Although $\partial T_{c1}/\partial x$ is positive for $x = 0.0190$, no second phase transition is observed above 0.2 K for material with this chemical composition. For $x = 0.043$, the two transitions cannot be resolved but the magnitude of the c_p anomaly indicates two transitions. The transition at T_{c2} does not affect the resistive or the low-field magnetic behaviour of the respective compounds indicating that superconductivity, as a macroscopic property, is not affected.

Clearly such a phase diagram is very unusual for a superconductor and the question arises how the different regions in the $[T_c,x]$ plane should be inter-

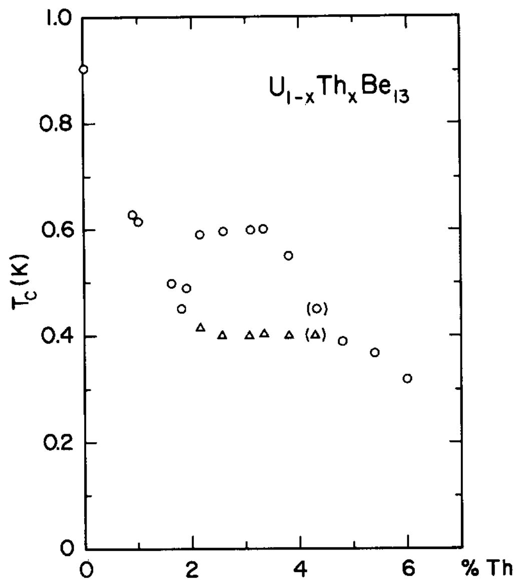

FIGURE 1
Phase diagram for $U_{1-x}Th_xBe_{13}$ as obtained from specific-heat measurements in zero magnetic field. The circles denote the transition to the superconducting state, the triangles show the second transition as described in the text.

preted. Since for pure UBe_{13} there is good reason to assume that an unconventional order parameter is different from that of a conventional BCS superconductor[1], theoretical work has dealt with possibi-

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lities considering such states and it has been found [4] that certain combinations of different representations of odd parity states may reproduce the phase diagram shown in fig. 1 Of special interest is the $c_p(T)$ behaviour well below T_{c1} or T_{c2} for x smaller or larger than about 0.02. From our experiments in zero magnetic field, we have no evidence for distinctly different behaviour in the two concentration regimes. For most investigated compositions, we found that rather large c_p/T ratios are adopted for T approaching 0 K, in distinct contrast with pure UBe_{13}. For some samples this ratio is approximately equal to the c_p/T values that are measured in the normal state preceding the c_p anomalies at T_c or T_{c1}. This, of course, poses some difficulties with entropy considerations connected with a second order phase transition involving the heavy quasiparticles forming the superconducting state. So far we can only speculate that either another phase transition at even lower temperatures follows or that a gapless state with a much enhanced density of states at E_F is formed. In view of the magnitude of the c_p anomalies, the latter solution is, however, difficult to justify. Measurements to lower temperatures may clarify the situation.

Recent measurements using μSR [5] revealed the onset of magnetic correlations below T_{c2} in the range where $x \approx 0.03$. Estimates of the moments involved in producing the measured internal fields lead to values of less than 10^{-2} μ_B/U.

In previous work[6] it was found that replacing small amounts of Be by B in UBe_{13} leads to a decrease of T_c but also to an anomalous enhancement and broadening of the c_p anomaly connected with the superconducting transition within a limited range of Be concentration. This was interpreted as an occurrence of two unresolved phase transitions, possibly of similar kind as those observed in $U_{1-x}Th_xBe_{13}$. The largest c_p anomalies were observed for $UBe_{13-y}B_y$ when $y \approx 0.03$. We have now also investigated the influence of similar B dopings in $U_{1-x}Th_xBe_{13}$ compounds. In fig. 2 we demonstrate this influence for the case where x = 0.019 and y = 0.03. It is obvious that for this Th concentration, a tiny amount of B on Be sites induces two phase transitions. In the B-doped sample, the transition at T_{c1} is almost identical to the single transition in the undoped specimen. The second transition at T_{c2} is at about 0.35 K and, as mentioned above, the c_p/T ratio well below T_{c2} is not reduced. The Boron doping gives no resolved second transition for x = 0.015 but the c_p anomaly is again drastically enhanced as mentioned above for pure UBe_{13}. T_c itself is not shifted. Quite harmful is the Boron doping for material with a Th concentration of about 4.3%, where a reduced T_c and a drastically shrunken c_p anomaly results.

Thermal-conductivity measurements were made on pure UBe_{13} and on some samples with different Th concentrations. The most obvious effect of Th-doping is a lower thermal conductivity above T_c but enhanced λ values well below T_c instead. In contrast to pure UBe_{13}, a small contribution varying linearly with T is deduced in the limit where T approaches 0 K. No dramatic effects are observed at the phase transitions.

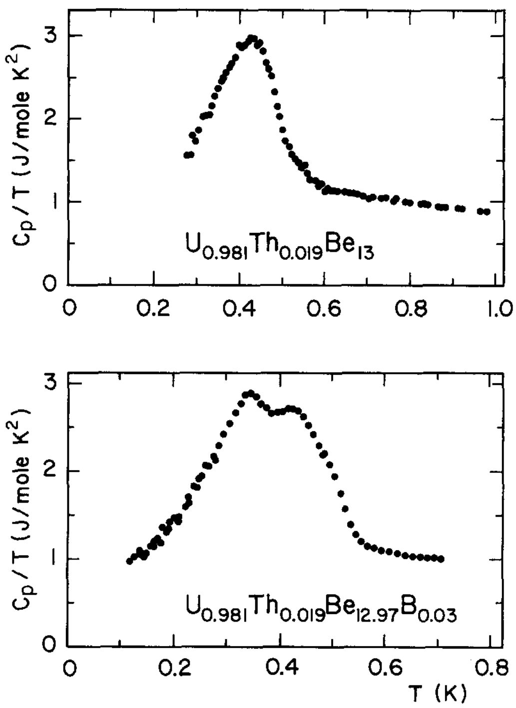

FIGURE 2
Example for the variation of the c_p anomaly of superconducting $U_{1-x}Th_xBe_{13}$ upon replacing a small amount of Be with B. Note the different temperature scales.

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