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Journal
Japanese Journal of Applied Physics, 26(S3-2)

ISSN
0021-4922

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Publication Date
1987

DOI
10.7567/jjaps.26s3.1219

Proc. 18th Int. Conf. on Low Temperature Physics, Kyoto, 1987
Japanese Journal of Applied Physics, Vol. 26 (1987) Supplement 26-3

Specific Heat of $(U_{0.97}Th_{0.03})Be_{13}$ under Pressure*

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The specific heat, C, of $(U_{0.97}Th_{0.03})Be_{13}$ has been measured for $0.1 \leq T \leq 1K$ and $1.6 \leq P \leq 7.7$ kbar, and for $0.1 \leq T \leq 20K$ with P=0. For T>8K both the pure and Th substituted samples have essentially the same C. The peaks in C/T at 0.33 and 0.54K for P=0 are suppressed and shifted to lower T by pressure. Anomalies in C/T can be correlated to corresponding rapid changes in magnetic susceptibility, χ. Rapid suppression of the peaks and shift of T_c to lower values is in marked contrast to the behavior found for pure UBe_{13} whose single peak amplitude decreases approximately linearly with P to about 60% at 9.3 kbar. The broad "shoulder" in C/T near 2K that is found for UBe_{13}, but not for any other heavy-fermion compound, HFC, is commpletely suppressed in the Th substituted sample.

Substitution of non-magnetic Th on U sites in UBe_{13}, $(U_{1-x}Th_x)Be_{13}$, produces unexpected and complex behavior in the superconducting region below 1K. In addition to the anomalous non-monotonic decrease of the superconducting transition temperature, T_c, with increasing Th content, there is the appearance of a second peak in C for $0.0175<x<0.04$ which is not due to a second phase or inhomogeneities [1]. For this range of x, T_c is nearly constant at 0.6K. Substitution of other impurities for U and Be produces a monotonic decrease of T_c, with no special depression of T_c associated with a magnetic moment on the impurity [2-4]. The unique effect of Th substitution on UBe_{13} over a limited range of x has been interpreted both as an antiferromagnetic transition [5] and as a transition between two anisotropic superconducting states [6]. Several attempts to confirm the presence of magnetic ordering in the $(U_{1-x}Th_x)Be_{13}$ system have failed [7,8], while the effect on T_c of magnetic Gd substituted for U (x=0.03) supports the suggestion of two different superconducting phases [2].

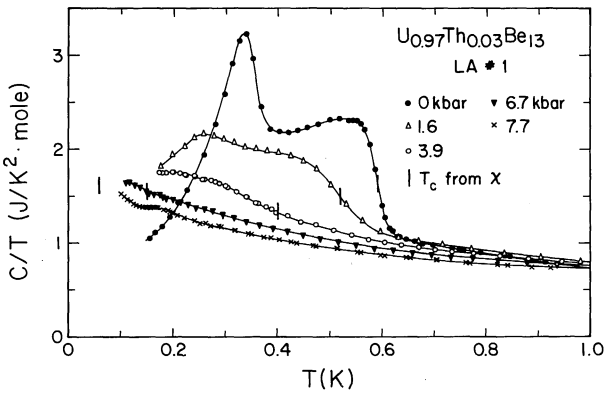

Fig. 1. C/T vs T for $(U_{0.97}Th_{0.03})Be_{13}$.

Measurements of the properties of materials as a function of pressure, P, provides an additional dimension in which to make comparisons with model calculations or theory. They also provide a straightforward basis for establishing correlations between superconductivity and magnetism without the complications of interpretation associated with measurements on a series of structurally and chemically different compounds. Measurements of the P-dependence of properties is a particularly fruitful approach for an HFC because the extreme pressure sensitivity of the 4f and 5f-electrons involved in the phenomena produces large effects at readily attainable pressures.

Recently χ of the $(U_{1-x}Th_x)Be_{13}$ system has been measured in the range $0 \leq P \leq 12$ kbar below 1K for $0 \leq x \leq 0.06$ [9]. Two distinct regions of superconductivity are present for P>9 kbar, which are separated by a range of x where superconductivity does not occur. Except for x=0.06, T_c, determined from changes in χ, decreases monotonically as P increases.

The $(U_{0.9697}Th_{0.0303})Be_{13}$ sample for the specific heat measurements weighed 1.673g and consisted of five right circular cylinders (approximately 6.4mm dia. x 2.4mm long) sparkcut from the center of an arc-melted, unannealed, polycrystalline "button" prepared as described previously [10]. They were placed in a pressure cell [11] and surrounded by AgCl to act as a pressure transmitting medium. A thin Sn plate on top of the sample stack and a Pb plate on the bottom served as superconducting manometers. The pressure gradient across the stack was ~ 15%. For all T and P in the range of the measurements, the heat capacity of the sample was >50% of the total.

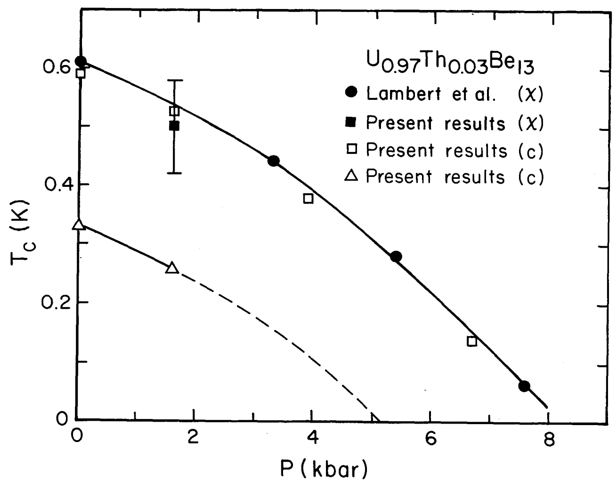

Fig. 2. T_c vs P for $(U_{0.97}Th_{0.03})Be_{13}$ as determined from χ and C measurements.

Figure 1 is a plot of C/T vs T below 1K in the range 0≤P≤7.7 kbar. A vertical bar for a particular P in Fig. 1 indicates T at the midpoint of the rapid change in χ [9], and is interpreted as T_c. At P=0, C/T has a finite intercept at T=0, which in the case of UBe_{13} has been shown [12] to be sample dependent rather than an intrinsic property of this material. For 0.6≤T≤1K, C(P)/C(0) varies by a relatively small amount. Over some of this range C(P) increases with respect to C(0) for P≤3.9 kbar, while at higher P, C(P) decreases over the entire range. The peaks in C/T at 0.33 and 0.54K for P=0 are strongly suppressed, broadened, and shifted to lower T for P>0. At 1.6 kbar the peaks are barely resolved at 0.26 and 0.43K. Only a single broad maximum is observed for P=3.9 kbar with an onset of the anomaly near 0.4K. For P=6.7 kbar only a very small anomaly remains, near 0.15K. At 7.7 kbar an apparently new feature develops -- a small maximum centered near 0.17K. This anomaly may be present at lower pressures but is obscured by the other anomalies, and it could be an impurity effect.

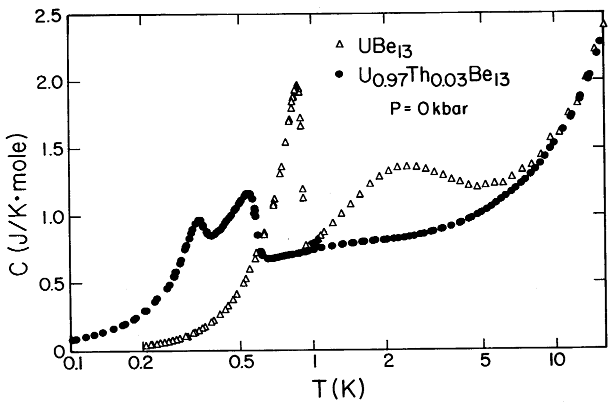

Fig. 3. C vs log T for UBe_{13} and $(U_{0.97}Th_{0.03}Be_{13}$ at P=0.

Figure 2 is a plot of T_c vs P. Values of T_c from Ref. [9] are the midpoints of the changes in χ taken from Fig. 1 and are displayed as filled circles. From the C measurements, T_c is taken as the midpoint of the rise in C/T at the anomaly and is graphed as an open square. During the present measurements, χ was measured at 1.6 kbar. The T_c derived from it is shown as a filled square with the vertical bar indicating the transition width, and is comparable to other data [9]. A satisfactory correlation exists between T_c determined by χ and C. Variation of the temperature of the lower temperature peak with P is more difficult to define. Except for 0 and 1.6 kbar there is no obvious indication of an anomaly and for P≥3.9 kbar it is presumably below the range of T investigated, and/or obscured by broadening and superposition of the two anomalies. If the maximum of the lower peak in C/T is used to mark the second transition, it is represented by open triangles in Fig. 2. (The dashed curve is drawn parallel to the solid curve.) The average dT_c/dP between 0 and 1 kbar is -40mK/kbar for both transitions. dT_c/dP increases to -80mK/kbar at 4 kbar where it remains essentially constant to 8 kbar for the higher T transition. These rates of decrease of T_c with P are in contrast to the constant and lower rate of -24mK/kbar for UBe_{13}, and the 60% decrease in peak amplitude from 0 to 9.3 kbar.

In Fig. 3, C is plotted vs log T for both UBe_{13} and $(U_{0.97}Th_{0.03})Be_{13}$. The broad maximum near 2K for UBe_{13} has been completely suppressed by the Th substitution. Substitution of Th, Lu and Sc for U gave similar results in an earlier investigation [4]. This feature in C has been interpreted as due to development of coherence in a Kondo lattice [13]. Suppression of the anomaly by a non-magnetic impurity is consistent with this idea. Above ~ 8K, C for both the pure and substituted samples are essentially identical as found previously [4].

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*Work at Berkeley supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences Division of the U. S. Department of Energy under Contract Number DE-AC03-76SF00098. Work at Los Alamos was supported by the U. S. Department of Energy.

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