Title
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Journal
Physica B+C, 130(1-3)

ISSN
0378-4363

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Publication Date
1985-05-01

DOI
10.1016/0378-4363(85)90210-4

Physica 130B (1985) 159–162
North-Holland, Amsterdam

SUPERCONDUCTIVITY OF URANIUM COMPOUNDS

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Invited paper

The superconducting compounds of uranium exhibit a large range of lattice spacings and of electronic specific heat γ values. Recent work has revealed superconductivity to be more common in such systems than formerly believed.

Until approximately two years ago, the occurrence of superconductivity in uranium and its compounds was believed to be remarkably restricted. Stabilized γ-uranium and the α-form under pressure were known to be superconducting, and only the compounds U_6X where $X =$ Mn, Fe, Co, and Ni, and the compounds UCo and $U_2Pt_2C_2$ were known superconductors. With the discovery of superconductivity in UBe_{13} in 1983 began a new experimental search for uranium superconductors, and some results of this search are listed in the table. As more ternaries are examined, one can now expect that many more superconductors containing uranium will be discovered in the future. By far the most interesting superconductors among those listed are UBe_{13} [1] and UPt_3 [2], and we will discuss first these two materials before concluding with an overview.

UBe_{13} is a simple cubic compound with 8 formula units per unit cell, the U–U distance equals 5.13 Å. This is much too large for direct overlap of 5f wave functions from adjacent U atoms. The macroscopic physical properties of UBe_{13}, when taken together, are quite anomalous (fig. 1). We see, first of all, a negative $d\rho/dT$ between 2.5 K and 300 K with a well-defined peak at 2.5 K, there being a drop in resistivity below this temperature, which is intercepted by the superconducting transition temperature at $T_c = 0.9$ K. The resistivity at T_c is large and in excess of 100 $\mu\Omega \cdot$ cm. We secondly notice that the high-temperature magnetic susceptibility is large and obeys a Curie–Weiss law with $\mu_{eff} = 3.1\ \mu_B$. For $T \to 0$ K, χ approaches a large constant value. The low-temperature specific heat is perhaps the most unusual of all the properties: the electronic specific heat coefficient γ at low temperatures is increasing on cooling below 10 K and is approaching the enormous value of 1.1 J/mol $U \cdot K^2$ as T approaches T_c. The huge specific heat anomaly at T_c indicates that the superconducting gap opens in the high density of states band, and that it is these "heavy fermions" that are superconducting, as also occurs in the case of $CeCu_2Si_2$ [3]. We note additionally, that the slope of the upper critical field at T_c, $dH_{c2}/dT = 440$ kOe/K, is the highest known for any superconductor [4].

A detailed analysis of the specific heat in the superconducting state finds a power rather than a exponential law in T [5] and this in addition to the strong coupling aspects of the transition, suggests that one is dealing here with an unconventional type of superconductivity with zeroes in the gap at points or lines on the Fermi surface, as expected for various forms of p-wave superconductivity.

Further support for this idea comes from data on $Th_xU_{1-x}Be_{13}$ alloys (fig. 2). Not only is the T_c depression with x not monotonic [6], but one finds, in the flat region of T_c versus x, two bulk specific-heat anomalies. The upper transition is without doubt a superconductivity one. The lower one has been shown by NMR measurements [7] to be very unlikely either a structural or magnetic transition. It seems at present that this

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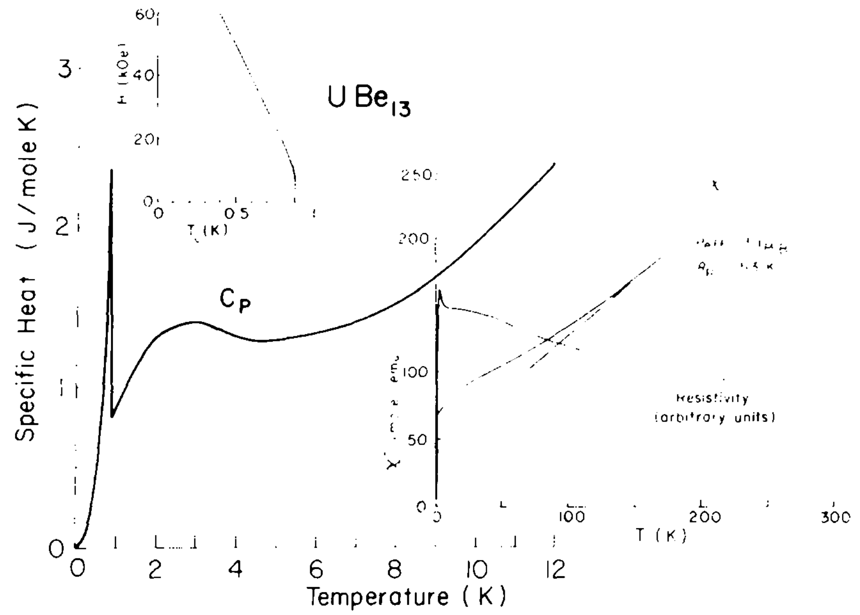

Fig. 1. Temperature dependence of some properties of UBe_{13}.

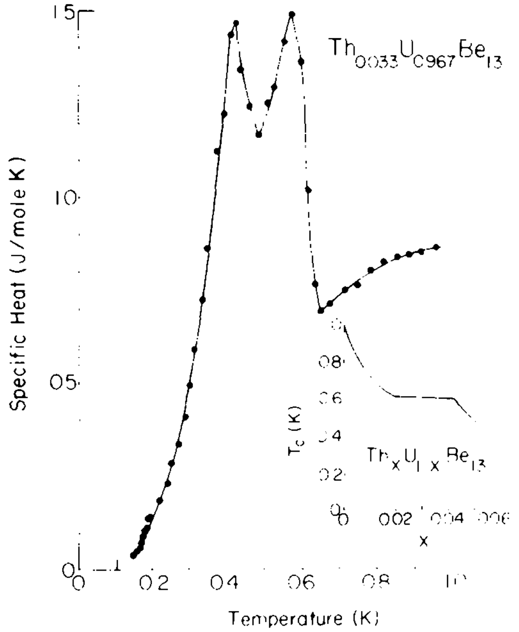

Fig. 2. Specific heat of $U_{0.9667}Th_{0.0333}Be_{13}$ polycrystal inset schematically shows T_c versus x for $U_{1-x}Th_xBe_{13}$ polycrystal.

second, lower temperature transition could then well be another superconducting one, and this is easy to envision only within the context of $l \neq 0$ superconductivity.

The properties of UPt_3 (fig. 3) are in a number of ways different. In this hexagonal structure we have the U–U distance equal to 4.1 Å. The electrical resistivity is large (~100 μΩ · cm) at room temperature, but decreases on cooling as expected in typical metals. Again we have a Curie-Weiss law at high temperature, with $\mu_{\text{eff}} = 2.6\ \mu_B$, but with considerable anisotropy of χ, the basal plane susceptibility at low temperatures being roughly twice that of the c-direction. In addition, there is a peak in the susceptibility at 14 K, which has no specific heat anomaly. The really unusual behavior, in view of the superconductivity at 0.5 K of this compound, is its specific heat, which shows a T^3 ln term, but rise at low temperatures (to a value of 450 mJ/mol U · K²), believed to be the signature of ferromagnetic spin fluctuations [2].

There are two experiments on the superconductivity of UPt_3, which are suggestive of $l \neq 0$ pairing. The first, somewhat weak evidence is

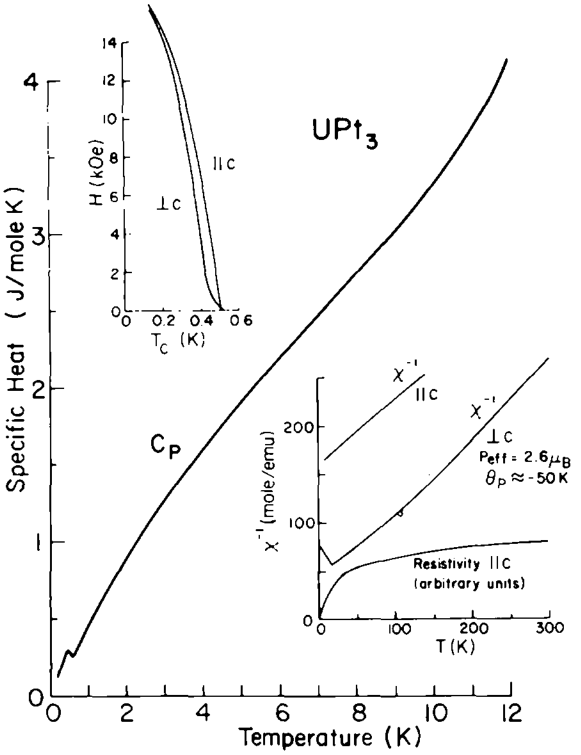

Fig. 3. Temperature dependence of some properties of UPt$_3$ [17].

from the upper critical field anisotropy [8], which has been used by Varma as pointing to a polar p-wave superconducting state. The second stronger evidence comes from ultrasonic absorption below T_c [9]. In this case, a good T^2 law has been found, consistent with a line of zeroes in the superconducting gap, as predicted for the polar p-wave state.

An examination of the table shows the remarkable variation of γ of almost two orders of magnitude across the series of uranium superconductors. We see that there are superconductors represented in all ranges of U–U spacing and γ, and it is a curious fact that magnetically ordering uranium compounds cover nearly the same range of these parameters. It is strange, however, than no extremely large γ U compounds have so far been found, which show neither superconductivity nor magnetic order as seen in CeAl$_3$ and CeCu$_6$. No measurements at present point to other than conventional superconductivity for the other uranium superconductors.

Table I

Material	T$_c$ {K}	γ {mJ/mol U · K^2}	d(U–U) {Å}	Ref.
γU	2.1	—	2.96	11
α-U	0	25	3.12	12
UCo	1.5	—	3.21	13
U$_6$Mn	2.4	—	3.20	11
U$_6$Fe	3.8	26	3.20	14
U$_6$Co	2.6	21	3.20	15
U$_6$Ni	0.4	—	3.20	11
U$_2$PtC$_2$	1.47	75	3.52	16
UBe$_{13}$	0.9	1000	5.13	1
UPt$_3$	0.5	450	4.1	2
UAl$_2$Si$_2$	1.34	27.9	4.15	10
URu$_3$	0.145	12.4	3.98	10
UAl$_2$Ge$_2$	1.60	—	4.22	10
UGa$_2$Ge$_2$	0.87	—	4.22	10

There is an additionally strange behavior seen in the compound UAl$_2$Si$_2$. This forms in the same Cu$_3$Au structure adopted by UAl$_3$ and USi$_3$, and in fact has a γ which is half way between the respective γ's of the end members. Yet neither UAl$_3$ nor USi$_3$ is superconducting [10]. One wonders if other cases of this unexpected superconductivity might be found. Finally, we note that many of the superconducting compouds of uranium have γ's, which are close to what one expects for density of states given by assuming that the measured low temperature susceptibility is all Pauli-type in origin.

Work performed under the auspices of the U.S. Department of Energy.

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