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The Physics and Chemistry of Heavy Fermions

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Abstract

The heavy Fermions are a subset of the f-element intermetallics straddling the magnetic/non-magnetic boundary. Their low temperature properties are characterized by an electronic energy scale of order 1 - 10 K. Among the low temperature ground states observed in heavy Fermion compounds are exotic superconductors and magnets, as well as unusual semiconductors. We review here the current experimental and theoretical understanding of these systems.

Heavy Fermion materials are a subset of the intermetallic compounds containing f-electron elements. These compounds are distinguished at low (Helium) temperatures by an anomalously large electronic specific heat coefficient (γ), with correspondingly large Pauli paramagnetic susceptibility (χ). Here large means two to four orders of magnitude larger than Cu, whose electronic specific heat $\gamma \approx 1$ mJ/mole-Cu K². That the heavy Fermion's large γ derives from the f character is experimentally confirmed by the γ of the isostructural rare-earth or actinide analogous compound which has no f electron (e.g. La, Lu, Y, or Th). It is general practice to normalize the γ to one mole of the f element. The free-electron ratio $\gamma/\chi = \pi^2 k_B^2 / 3 \mu_B^2$ is a limiting value approached by a few heavy Fermion materials, but generally χ is larger than the free-electron estimate from γ : not all the χ shows up in the γ at low temperature (Figure 1).

The integral $\Delta S = \int^T \gamma dT$, representing the entropy being developed in the electronic system of the heavy Fermion compounds as T increases from 0 K, is large and of order $R \ln 2$ per mole of rare earth for T in the range 1 K - 10 K, depending on the particular system. The temperature by which this entropy is developed is called the coherence temperature, T^* , and is seen in many other physical properties. It seems reasonable to regard this entropy as being carried by a set of localized f spins which emerge above temperatures of this order, judging also by the presence at higher temperatures of a Curie-Weiss law in the magnetic susceptibility with Curie constant corresponding to that of the Hund's rule ground state of the fⁿ configuration of the f element. What the physics and

chemistry of these heavy Fermion compounds is concerned with then is the low temperature disappearance (or compensation) of the local f moment character which becomes evident at high temperatures, namely for temperatures greater than a few times T^* .

The f elements among whose intermetallic compounds we can find heavy Fermions are, with a few understandable exceptions, at the start of the 4f and 5f series (the rare earths and actinides), as well as at the end of the 4f series, namely Ce, U, and Yb. It is therefore natural to associate heavy Fermion behavior with an instability of the f configuration, and this could be expected to be sensitive to details of the f-elements' chemical environment. While something similar might be expected to occur in the d-elements, the feature special to the f shell is that it is an inner shell and generally not involved in chemical bonding: the radius of the 4f shell is small compared to interatomic distances. For the first 4f element, Ce, it appears that the electron added to the number of its neighbor La has two close energy configurations, one being an outer spd configuration, the other an inner f-like configuration. We might expect these nearly degenerate energy states to hybridize with each other in the intermetallic compound. The ligands surrounding the f element could influence this hybridization substantially. In metallically conducting compounds, this hybridization will show up as a coupling between the atomic f-like level and the conduction electrons. For the weakly bound f electron (as in Ce), this coupling is known to be antiferromagnetic: conduction-electron spins couple oppositely to the localized spins. A further detail is that the orbital

angular momentum of the f wave function is not quenched, and the effective coupling is both spin and momentum dependent.

The theoretical problem of a localized spin interacting antiferromagnetically with a sea of conduction electrons is the celebrated Kondo problem, whose solution is one of the outstanding achievements of many-body physics. It describes how the local spin is screened out, or compensated, continuously as the temperature, T , falls below a characteristic Kondo temperature $T_K = De^{(-1/\rho J)}$. Here D is the band width characterizing the conduction electrons, J the local spin-conduction electron coupling and ρ the electronic density of states at the Fermi level. The low temperature state is a many-body singlet of the conduction electrons with the local spin, not a bound local spin-conduction electron pair. Something like this compensation appears to be happening below T^* in the heavy Fermions. In the chemically ordered lattice of f ions, rather than the single f ion impurity case appropriate to the Kondo problem, one wonders to what extent the physics carries over. The highly correlated nature of the problem is immediately evident: in the heavy Fermion problem the ratio of conduction to f electrons is of order one, far from the Kondo case. The Kondo lattice is in fact a limiting case of the Anderson lattice Hamiltonian, which will in general provide a closer, albeit related, description of the physics.

How can the conduction electrons compensate the f moment in this case? We are reminded that chemical bonds do this in the pair bond. A "Kondoized" variant of the electron-pair bond might provide

a way to approach the physics and chemistry of the heavy Fermion problem, but this approach has not been taken. There is a small energy scale in the problem, T^* or T_K , and this must emerge naturally, and its rather modest variations in diverse environments suggest that it is best thought of as an atomic property.

We return now to the properties of the heavy Fermion materials [1,2]. For $T > T^*$, we have a dirty metal with a set of independent local paramagnetic spins strongly scattering conduction electrons. This scattering gives rise in many cases to a negative coefficient of electrical resistivity, reminiscent of the Kondo impurity effect (Figure 2). The magnetic susceptibility obeys a Curie-Weiss law with negative Curie-Weiss temperature. Below T^* the electrical resistivity drops rapidly, often into a low temperature T^2 behavior, characteristic of a Fermi liquid (Figure 3). It is at these low temperatures that the large γ develops, seen as a strong upturn in a C/T versus T^2 specific heat plot, usually but not always becoming constant at sufficiently low T , provided some kind of phase transition does not occur. It is generally believed that a low- T Fermi liquid state is achieved by heavy Fermion materials, just as in the Kondo impurity case. It makes sense to think about a Fermi surface, in that for a number of these materials deHaas-van Alphen oscillations have been observed. Large electron masses have been found over significant portions of the Fermi surfaces. While some materials appear to have a large and fairly uniform mass enhancement over their entire Fermi surface, other materials appear to have enhanced electron masses over only part of their Fermi surfaces [3]. There is

evidence in some cases that the heavy Fermion state significantly affects the topology of the Fermi surface. A close correspondence between the measured γ and the masses seen in deHaas-van Alphen is generally not found, presumably due to the great difficulty in observing those orbits with the largest masses.

There is an obvious competition between the low-T compensated state and a magnetically ordered state of the uncompensated moments. This has been the subject of various theoretical and experimental studies [4,5]. The competition is generally couched in terms of competing scales T^* and T_{RKKY} , the latter the energy scale characterizing moment-moment interactions in a metal. Theory has gone farthest with the two impurity Kondo model, finding two stable Fermi liquid fixed points and (with some controversy) an unstable fixed point between the two. Varma [6] has argued that the unstable fixed point corresponds to a marginal Fermi liquid, a topic we address further below.

It is an interesting fact that heavy Fermion materials which order magnetically at a low temperature, T_N , fall into two classes: those with small ($< 0.1 \mu_B$) and those with large moments ($> 0.5 \mu_B$). A few interesting Ce compounds cross this distinction, having modulated moments and sometimes having both compensated- and large- moment sites. It appears to be the rule that the large-moment order is found in those systems for which $T_N \geq T^*$, the small moments for $T_N \ll T^*$. It makes sense then to separate off the larger-moment

systems as of more conventional type and regard the low-moment cases as examples of Fermi-surface instabilities.

The typical Fermi-surface instabilities are spin-density waves, charge-density waves and superconductivity. There are now known about a half dozen heavy Fermion superconductors (see Table), and it is these materials which first attracted the great interest to the field. This was because the heavy mass ground state appeared to have its origin in a magnetic interaction, and as such could well be expected to lead to an exotic type of superconductivity, both as to pairing state and attractive interaction. The first-known heavy Fermion superconductors were CeCu_2Si_2 [7], UPt_3 [8] and UPt_3 [9]. All have transition temperatures in the 0.5 K to 1 K range. These superconductors were investigated for signs of higher angular momentum pairing, mainly by looking for evidence of nodes of the superconducting gap on the Fermi surface. This was sought as power law rather than BCS-type exponential temperature dependencies in various physical properties below T_c , such as penetration depth, ultrasonic attenuation and specific heat (Figure 4). While good evidence for these was found, they proved not to be quite as definitive as hoped: even Nb-based superconductors show various deviations from strict BCS behavior.

More compelling evidence for exotic superconductivity was found with the discovery of multiple superconducting phases at T_{c1} and T_{c2} in UPt_3 [16,17] (Figure 5) and Th-alloyed UPt_3 (Figure 6). A curious aspect of UPt_3 is that neutron studies [20] find a small-

moment ordered magnetic state below 5 K, with ordered moment $\approx 0.01\mu_B$. Not all samples show this order however. In addition, a transmission electron microscopy study [21] finds evidence that a structural modulation exists in this compound, and that the coherence lengths characterizing the superconductivity, the magnetic order, and the structural modulation are all comparable, approximately 250 Å. So there is some chance that the magnetic order is a defect phenomenon and that, a perhaps smaller chance, the various superconducting transitions are intimately connected with the structural modulation. It is true, for example, that some samples show two transitions in resistivity, indicating inhomogeneous material.

In the case of UBe_{13} , Th-doping at the few percent level results in two superconducting phases. UBe_{13} is interesting in that its superconductivity develops on cooling before a coherent, normal electronic ground state is completely developed. In this case, moreover, the superconductivity appears very robust, with T_c not delicately dependent on sample quality as in UPt_3 . This is particularly apparent in the pronounced anomalies in specific heat at T_c . Evidence that a small magnetic moment exists below T_{c2} in the Th-doped samples showing two transitions has been found in μSR measurements [22]. Some types of superconducting pairing can carry a small moment. General opinion favors at present some kind of magnetically-mediated pairing in a relative d-wave state.

There are a number of other heavy Fermion superconductors. CeCu_2Ge_2 is magnetically ordered at ambient pressure and low T , but pressure drives it non-magnetic. Above 75 kbar it becomes a superconductor [15]. URu_2Si_2 shows both magnetic and superconducting order [10-12]. Antiferromagnetic order sets in at $T_N=17$ K with $\mu_{\text{ord}}=0.01 \mu_B$, thought here to derive from induced-moment ordering out of a crystal-field singlet ground state but with considerable differences of opinion. Additionally, this material shows very sample dependent T_C 's. The low-temperature specific heat $\gamma = 90$ mJ/mole-U K^2 is modest, and an interpretation has been made that the superconductivity and magnetism compete for Fermi surface, somewhat at odds with the singlet crystal field ground state argument. The isostructural compounds UPd_2Al_3 [13] and UNi_2Al_3 [14] also show magnetic order and superconductivity, the latter appearing local-moment like, the former itinerant. These two compounds seem analogous to the ternary rare-earth magnetic superconductors studied heavily in the 1970's where the physics was interpreted in terms of two weakly coupled electronic subsystems.

Neutron studies of heavy Fermion magnets are not as extensive as one would like. Cooling below T^* results in $\chi(q)$ developing finite- q correlations from the higher temperature single-ion response. In the case of U_2Zn_{17} , with $T_N=9.8$ K, one finds an ordered moment of $0.6 \mu_B/\text{U}$, placing it in the large moment class [23]. It is however interesting that the ordering is driven by the temperature dependence of the U-U coupling J . Generally, a Stoner-like condition $\chi J=1$ must be satisfied for some q at T_N . For U_2Zn_{17} it is not the

temperature dependence of χ that drives the ordering; it is the temperature dependence of J . The criterion is just barely satisfied, and it turns out that Cu substitution for Zn at the percent level completely suppresses T_N [24].

An unusual case of heavy Fermion antiferromagnetism is that found in UCu_5 . The $T_N=15$ K, and the ordered moment is large, about $0.6\mu_B$. Below T_N , as is typical, γ drops to 1/3 its value at T_N . Optical measurements show, however, that this order is best interpreted as a Fermi surface instability [25]. More surprising, a second phase transition occurs at 1.5 K in sufficiently high quality material [26]. This second transition only weakly perturbs the underlying magnetic order, and its true nature remains unknown. But the electrical resistivity of the material rises by a factor of 8 below 1.5 K. While the possibility of this being some kind of charge-density-wave instability seems remote, it is worth keeping in mind that both elemental U and Pu apparently support charge-density waves at low temperature.

There is another small subgroup of the heavy Fermion materials that are small gap semiconductors, with gaps in the range of a few K to 1000 K [27]. For these it appears that T^* and the gap temperature are roughly the same. There is a reasonable argument that this class of materials represents a particularly simple limit of the lattice problem in which there is a single half-filled conduction band crossing an occupied f orbital: the hybridization of these two opens a gap at the Fermi level. There also appear to be some near

misses to this behavior, as well as a number of surprising cases of semimetals with heavy Fermion properties. An interesting feature of this subgroup of materials is seen in the temperature evolution of their frequency-dependent optical conductivities [28]: a redistribution of spectral weight over an energy of order $10 T^*$ occurs on going from low T to about $1/2 T^*$. This is a clear indication of the strong coupling nature of the physics.

The question of marginal and/or non-Fermi liquid ground states has also received considerable theoretical and experimental attention [29,30]. The experimental evidence involves reduced entropy development with temperature and linear-in- T resistivities. Recent experiments on CeCu_6 [31] support the idea that what is really involved here is a $T=0$ phase transition. This is consistent with the somewhat troubling aspect of these studies, that the marginal Fermi-liquid properties only show up in heavily alloyed compounds, not stoichiometric, atomically-ordered ones.

We now address the general problem. There is a continuum of mass enhancement linking simple metals with heavy Fermions. No apparent limit to conduction-electron mass enhancement seems to exist. One can say that the mass goes inversely with T^* and that the lower T^* is, the more likely some order (e.g. magnetic) can occur. This is roughly because T^* is expected to be exponential in J , T_{RKKY} algebraic. Increasing T^* takes one through the valence-fluctuation regime to (eventually) the fully-hybridized metals. The various phase transitions found in d-band materials are found in heavy

Fermions - namely superconductivity, magnetism, and, maybe, charge-density waves, and in some cases nothing. So our question is: is there anything more to the heavy Fermions than a situation characterized by an unusually large effective mass?

We come back to the chemical view. The small energy scale in the problem, T^* , appears to be an atomic, on-site property which can be fiddled with somewhat via surrounding ligands. There seems to be something like an atomic state which develops into the Kondo resonance in these solids, but ultimately the many-body physics is played out in the first coordination shell of the f atom. Placing the f atoms on geometrically frustrated lattice suppresses the tendency to magnetic order of the f moments. The electronic instabilities that then occur in such lattices involve quite small energy scales, against the quiescent background of low temperature. The experimental conditions are ideal.

What do we really know? Not so much. The reliance on the Kondo model for guidance in the lattice problem is, in our view, really little beyond recognizing some sum rules. We know how much entropy is involved and the scale over which it develops. We do not have a clue experimentally about f-f interactions. The attempts to derive them from specific heat measurements cannot really give us the interaction terms, since the coupling and T^* scales are not distinguished, and only the largest is apparent. We are in an experimental regime where the physics is clean in the sense that not much else is happening. It is ideal for understanding most aspects of

the strongly-correlated-electron problem. Many of the properties of the lattice resemble those of isolated impurities, so what is new in the concentrated problem?

This question is similar to the chemist's question in the more general context of solid state chemistry: we still see the bonds in the solid, so what is different? Obviously the various phase transitions are, but some of these are presaged at the molecular level. The heavy Fermion problem is a peculiar mix of an atomic problem and a lattice one. The properties of the coherent state, such as the temperature-dependent development of antiferromagnetic correlations are not atomic-like. But we know, especially from epr-probes, that the delicate physics is at the local level. The various possible ways that sites can be coupled is large, and the problem is how to recognize them.

Related to these considerations is the question: can coherence be established via a first-order phase transition? There are two cases where this can be claimed: the γ - α volume collapse in elemental Ce [32], and the volume dilation in YbInCu₄ [33,34]. Both are isostructural, first-order phase transitions involving large electrical resistivity drops and local moment loss. One immediately thinks of the gas-liquid phase transition in these cases. Generalizing, can we regard the continuous development of coherence on cooling through T* as a gas-liquid system beyond its critical point in parameter space? This seems qualitatively reasonable, but also needs detailed examination.

The correlated electron liquid is distinguished by having a very close-by-in-energy magnetic excited state. This is even apparent in the magnetic properties of defects. From the high-temperature side, it seems that there are many kinds of order in which to freeze the large entropy of the systems. The competition between the chemical freeze-out, namely the compensation of moments, versus various magnetic ways to reduce degeneracy is what leads to the richness of the observed phenomena. And added to this are the Fermi-surface instabilities, which are not entropy-driven.

Acknowledgments

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Figure Captions

Figure 1. $T \rightarrow 0$ limiting values of γ and χ for selected uranium compounds. The line represents the free-electron relation.

Figure 2. Temperature dependent properties of UBe_{13} . The superconducting upper critical field is shown in the upper left.

Figure 3. Temperature dependent properties of UPt_3 .

Figure 4. Power laws seen in various physical properties below T_c in UBe_{13} .

Figure 5. Superconducting phase diagram of UPt_3 as determined by sound velocity measurements. After Adenwalla et al. [18].

Figure 6. T_c -composition phase diagram of $\text{Th}_x\text{U}_{1-x}\text{Be}_{13}$. After Ott et al. [19].

Table. Heavy Fermion Superconductors

Compound	T_c (K)	Reference
CeCu ₂ Si ₂	0.65	[7]
UBe ₁₃	0.9	[8]
UPt ₃	0.50	[9]
URu ₂ Si ₂	1.5	[10-12]
UPd ₂ Al ₃	2.0	[13]
UNi ₂ Al ₃	1.0	[14]
CeCu ₂ Ge ₂	0.64*	[15]

*P=101 kbar

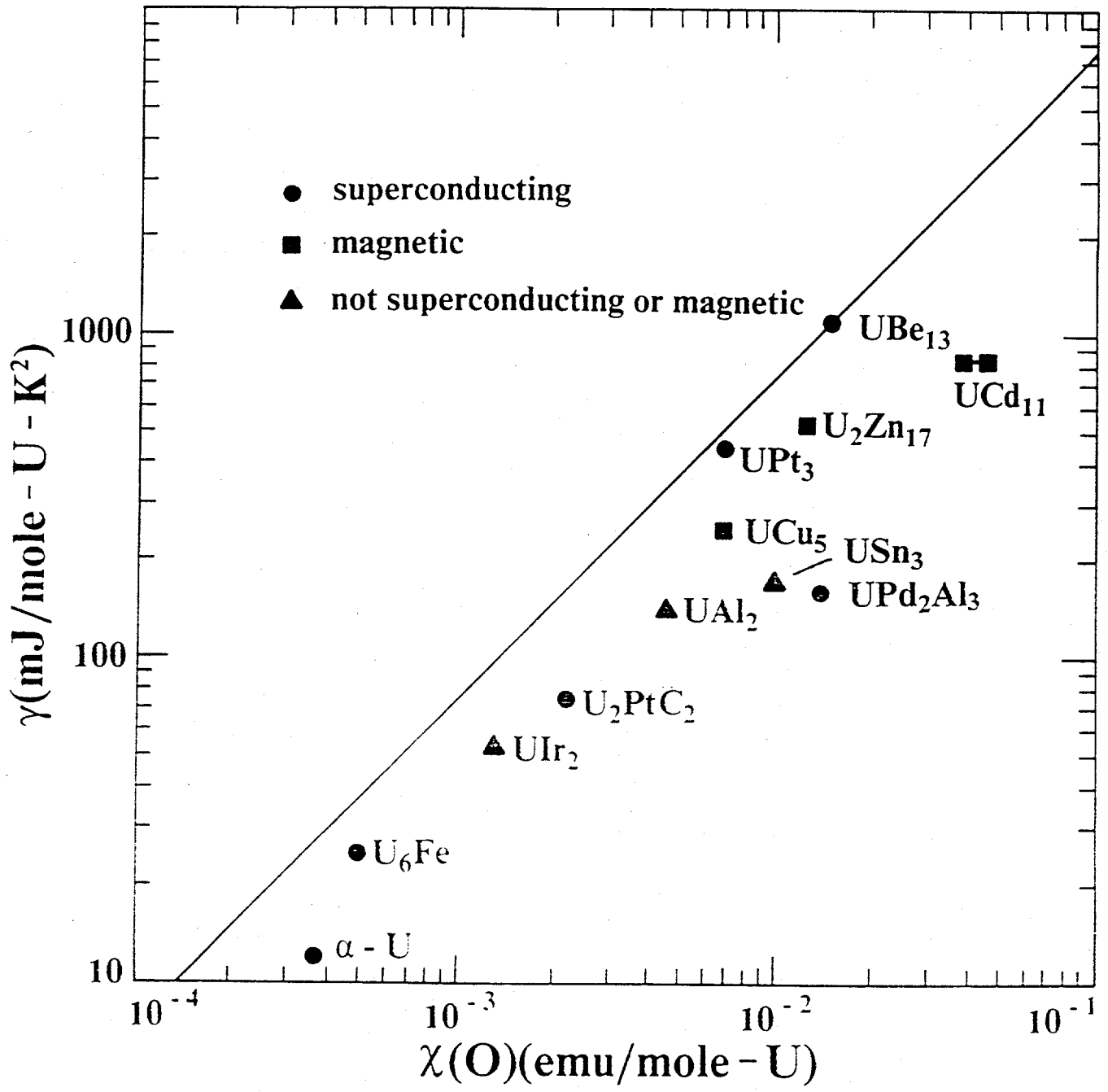


Fig. 1 Fisk et al.

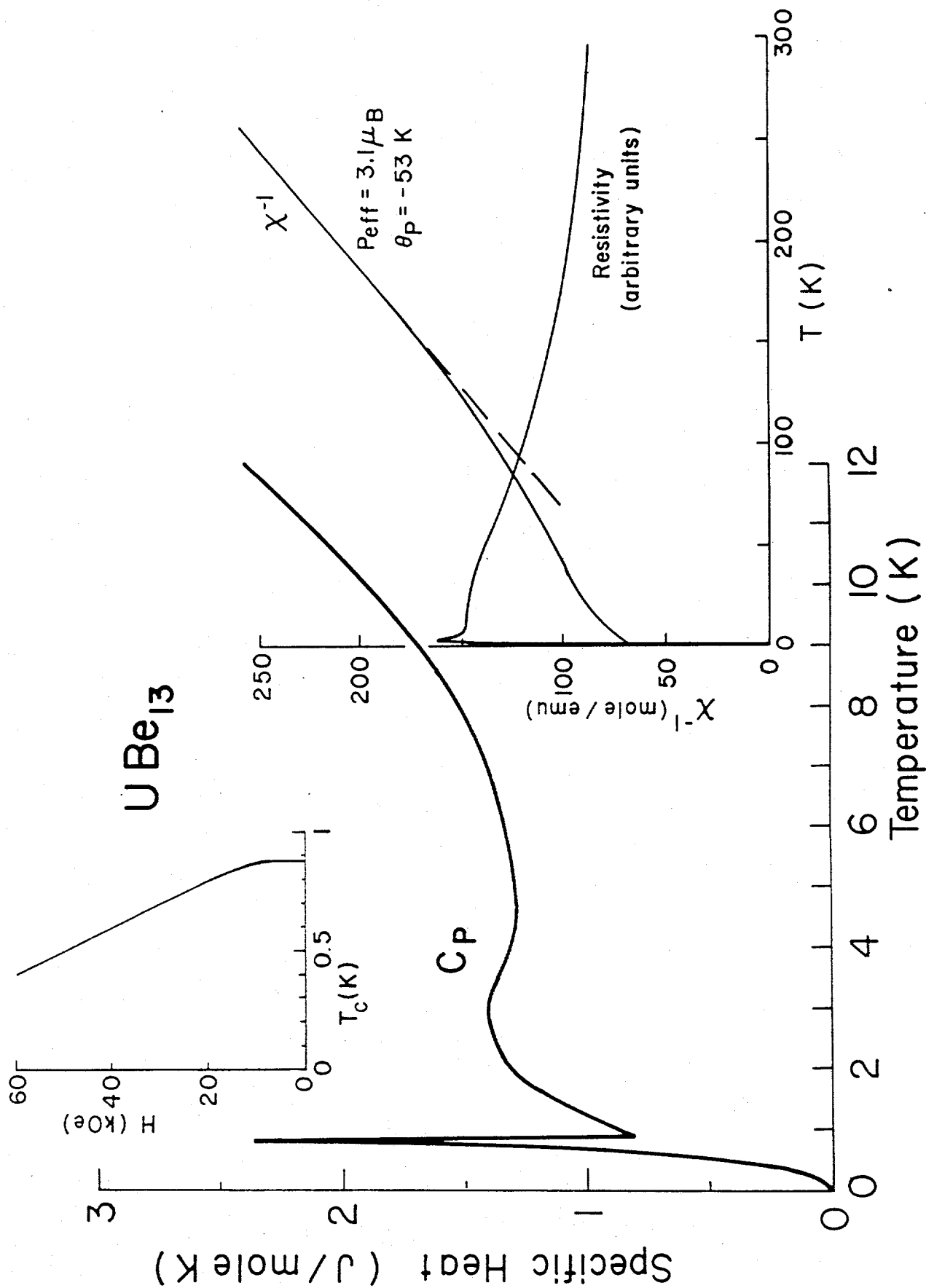


Fig. 2 Fisk et al.

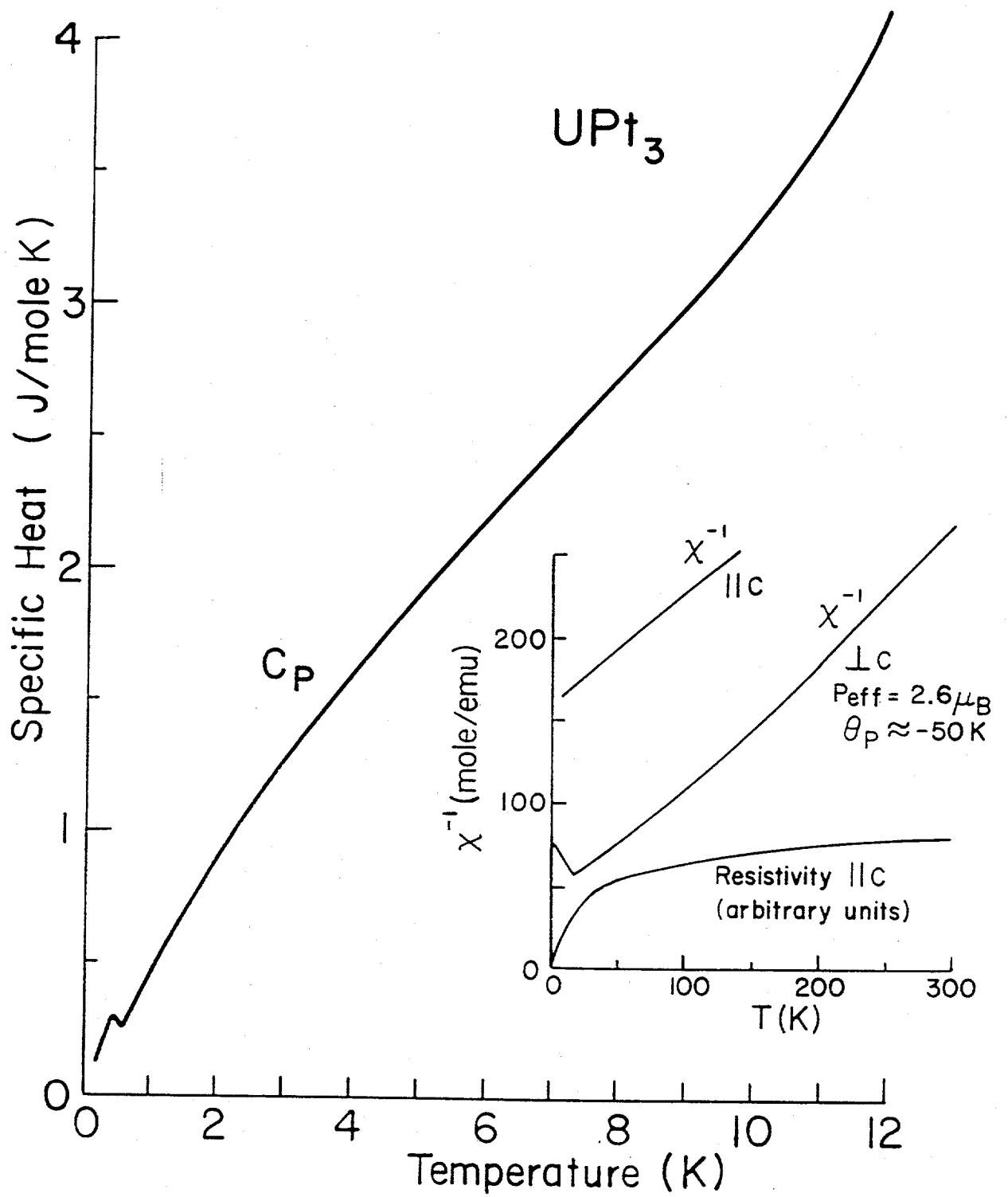
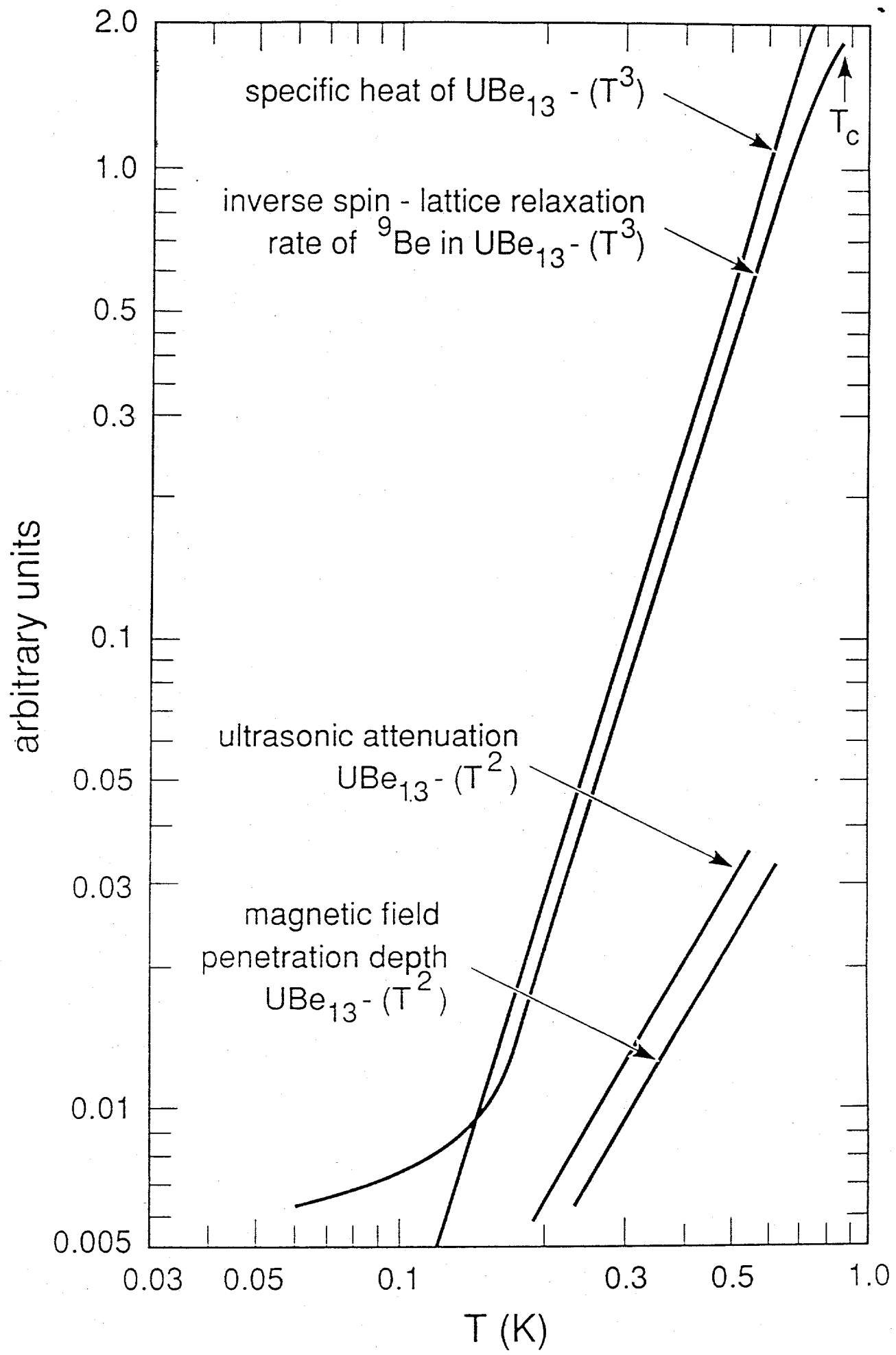


Fig. 3 Fisk et al.



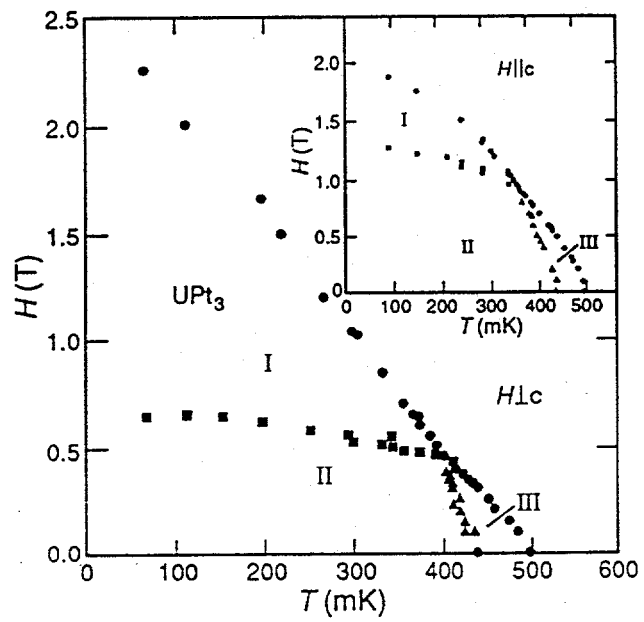


Fig. 5 Fisk et al.

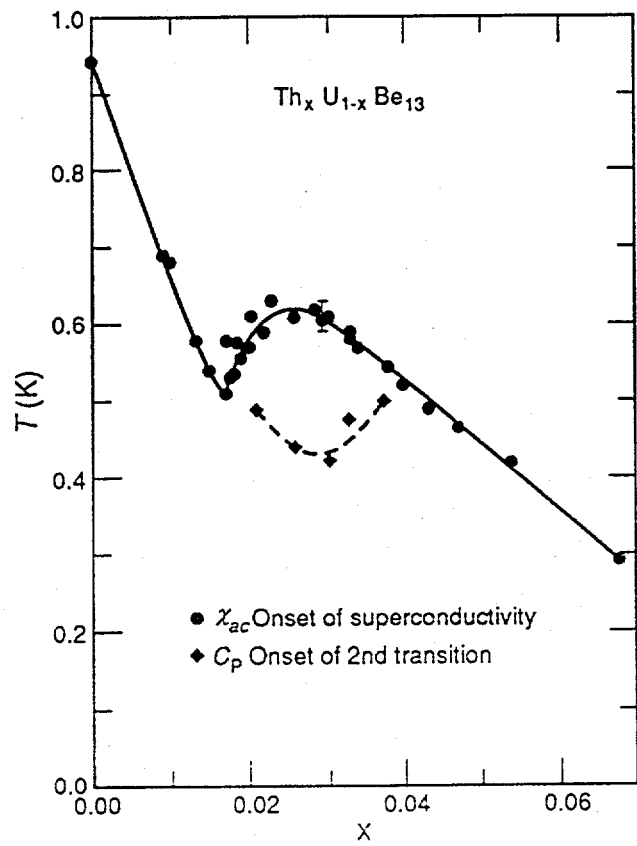


Fig. 6 Fisk et al.