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Experimental Perspectives on Heavy-Electron Metals

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We motivate the description of heavy-electron metals in terms of concepts from the Kondo problem. These concepts are used to discuss magnetism and superconductivity in heavy-electron systems. Particular attention is given to what we view as the principal outstanding questions in this field and directions in which the field is developing. This will include consideration of the differences between Ce and U heavy-electron compounds as well as the occurrence of very small ordered magnetic moments.

1. INTRODUCTION

The class of materials called heavy-electron metals is defined by the very large electronic specific-heat coefficient, γ, of order 1 J/mole K^2. It is now clear that there is a continuum in the physics which connects the heavy-electron metals with metals with γ's one to two orders of magnitude smaller, and that no sharp boundary demarks where heavy-electron behaviour can be said to begin. It may seem surprizing that a meaningful distinction can be made between materials based on γ, but the distinction appears to be a physically valid one and is based, ultimately, on the fact that all the very large γ's are of the same magnetic origin.

The heavy-electron materials are all f-element based (primarily on Ce and U), in particular on those elements whose f-configuration is regarded as unstable, although there appears to be no good reason why such materials might not also be found based on first row transition-metal elements. At temperatures of order room temperature and higher, the heavy-electron metals behave magnetically like local-moment systems. On cooling to helium temperature, the local-moment susceptibility develops into a Pauli-like susceptibility, characterized by its magnetic-field independence. At the same time, the remaining high-temperature spin entropy is smoothly transferred to the conduction-electron system: for effective spin J and temperature T_0 characterizing the change over, we estimate $\gamma \simeq k \ln (2J + 1)/T_0$ per spin, in accord with measured values.

A useful way to think about this change of behaviour is in Kondo terms: the local moments become compensated via a Kondo-type interaction with the conduction electrons, a consequence of which is that a narrow resonance develops at the Fermi level. The negative $d\rho/dT$ often seen in the high-temperature resistance behaviour and the large positive Hall coefficient observed, are consistent with this Kondo impurity picture, as is, of course, the unusual magnetic susceptibility and electronic specific-heat characteristics mentioned above. The magnetic parentage of the large γ's is evident in the plot of γ versus χ (Fig. 1), where we see that all the materials lie on the high-χ side of the Sommerfeld free-electron relation between γ and χ, indicated by the solid line. In our qualitative picture, it is the local moment fluctuations which make the condition electrons heavy.

2. NORMAL STATE

What is different in the atomically-ordered lattices of f-ions from the "impurity" situation is that the electronic state below some characteristic temperature T_0 becomes "coherent". The loss of incoherent scattering, characteristic of the high-temperature state, is clearly seen in the rapid drop of the electrical resistivity and in the loss of the large positive contribution to the Hall constant [1]. The temperature characterizing coherence is of the same order of magnitude as the Kondo temperature connected with the ion lattice. Theory has actually not progressed very far in inter-relating the Kondo and coherence temperature scales. A step in this direction has been the work of Jones and Varma [2] on the two-ion Kondo problem. What they find is an important interaction between the single-ion Kondo scale and the RKKY coupling and present a scheme for extending their considerations to the full lattice. Batlogg et al. [3] have presented evidence for a temperature-dependent Kondo state in UBe_{13} developing in the vicinity of the onset of coherence, signaling a breakdown in the single-ion Kondo approach.

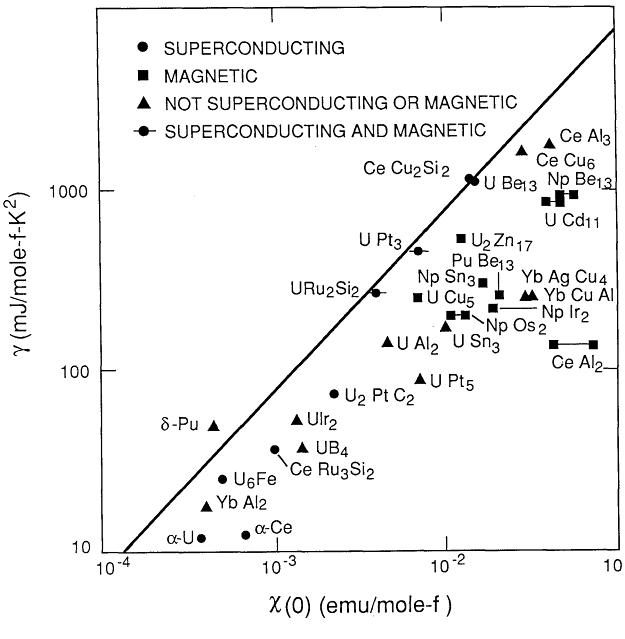

Fig. 1. Ln γ versus $\ell n\ \chi$ for f-element materials.

At T = 0 K, we expect the Bloch theorem to apply to the atomically-ordered compounds. Dramatic confirmation for the existence of coherent Bloch states for heavy electrons has been provided by the observation of de Haas-van Alphen oscillations in $CeCu_6$ [4] and UPt_3 [5]. The interesting fact is that no light masses have been observed: all electrons on the Fermi surface have become heavy. The largest mass observed to date is 90 m_e in UPt_3. It is interesting that in the case of UPt_3, for which detailed band-structure calculations exist, the extremal cross-sectional areas agree quite well with the calculations. This is in line with our qualitative remarks: Kondo-type interactions can only influence states within kT_0 of the Fermi level, affecting areas little.

Inelastic neutron-scattering experiments are providing their own slant on the question of coherence. The most complete experiments addressing this are on $CeCu_6$ [6] and UPt_3 [7]. Strong antiferromagnetic correlations are found to be a property of the heavy-electron ground state, and these correlations are washed out above the coherence temperature (Fig. 2). The energy scale characterizing these fluctuations appears to vary from compound to compound in the same way that the coherence temperature does. It is also interesting to note that there has been little indication of significant phonon softening in reported neutron-scattering experiments on heavy-electron metals.

3. SUPERCONDUCTIVITY

Much interest attends the various phase transitions observed in heavy-electron metals although a number occur just as coherence begins setting in. First among these are the superconducting phase transitions, now firmly established for $CeCu_2Si_2$, UBe_{13}, and UPt_3. URu_2Si_2, with a somewhat smaller γ, should also be considered with this group. T_c in all cases is near 1 K, and it is also observed that (γ,χ), for these metals, lies near the Sommerfeld line. The important questions raised by the occurrence of superconductivity in materials with strong magnetic characteristics were both whether a new mechanism is involved and whether a new superconducting pairing is being seen.

It turns out that these questions are not easy to answer experimentally and that an accumulated body of data rather than a single definitive result is determining the current thinking on these matters. While the case has been made by Steglich and co-workers [8] for an isotropic singlet superconducting state in $CeCu_2Si_2$, there is growing evidence that in UPt_3 and UBe_{13} the superconducting state is anisotropic. The evidence comes from numerous measurements of non-BCS temperature dependences below T_c including ultrasonic attenuation [9], specific heat [10], thermal conductivity [11], and London penetration depth (Fig. 3). The point is that the anisotropic states have zeroes of the gap on parts of the Fermi surfaces, either points or lines. These zeroes yield power laws in T rather than exponential dependence, as in the case where the gap is non-zero everywhere. These power laws are also to be distinguished from the more conventional gaplessness in magnetically doped conventional superconductors, which gives a linear in T contribution to the specific heat. This linear term has been claimed to be obseved in UPt_3, although recent work refutes this [13]. There also exists an unusual anisotropy in the ultrasonic attenuation in UPt_3 [14], as well as a peak below T_c in the ultrasonic attenuation in UBe_{13} [9].

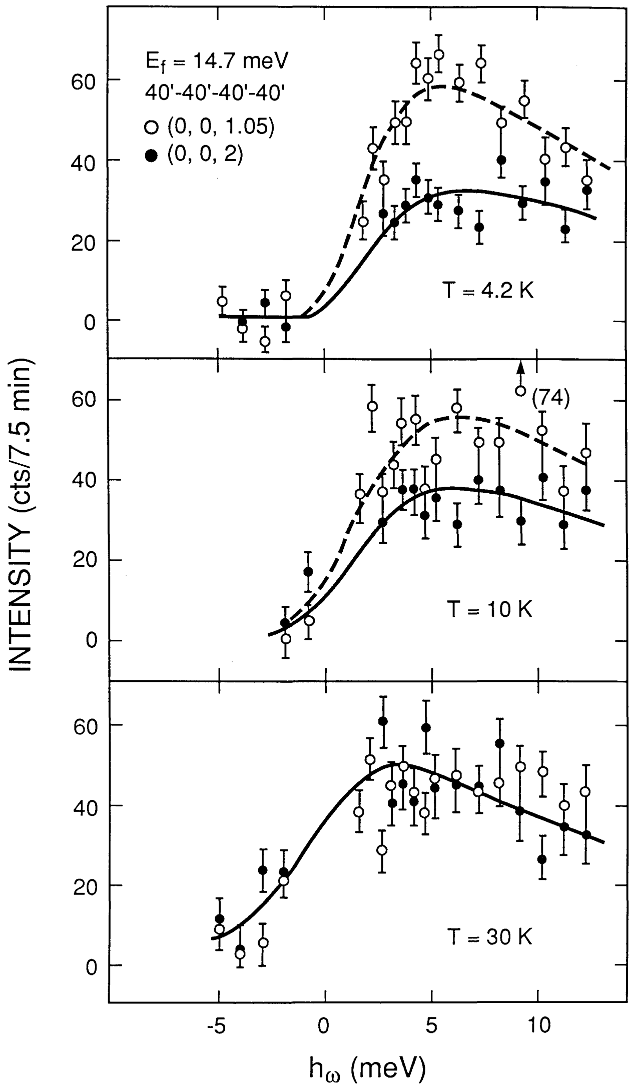

Fig. 2. Inelastic neutron scattering spectra of UPt_3 for Q = (0,0,2) and (0,0,1.05) at three different temperatures. Dashed lines are fits to the magnetic scattering. The antiferromagnetic correlations, contributing only in the (0,0,1.05)-scan, are gone by T = 30 K (from ref. 7).

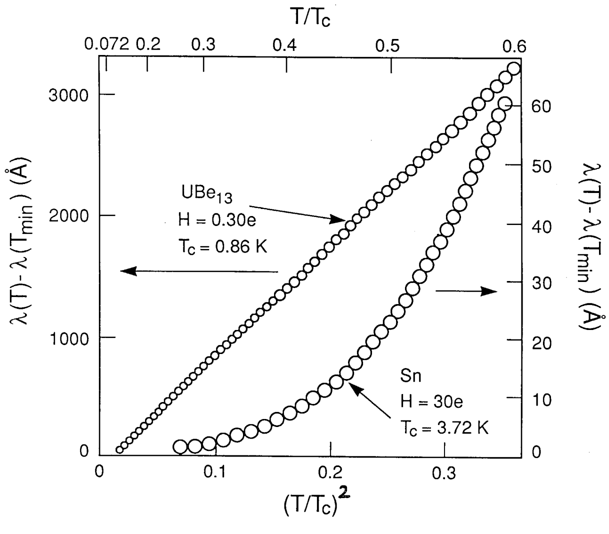

Fig. 3. Comparison between the London penetration depths of UBe_{13} and Sn (from ref. 12).

One of the curiosities of heavy-electron superconductivity is the behaviour of UBe_{13} with Th impurities. The curve of T_c versus Th concentration is not monotonic, and an extensive set of measurements on the pressure dependence of T_c as a function of Th doping gives support to the idea that there are at least two different superconducting states involved as x varies [15] (Fig. 4). It has already been known for some time that there is a second phase transition of unknown origin below T_c for x > 1.7% Th [16]. μSR experiments [17] have been interpreted as showing that the lower phase transition is to a state with an ordered moment of ≈ 0.001 μ_B/U. (We note that superconducting pairings with some net ordered moment are possible). Specific-heat measurements have determined in addition that the upper and lower transitions for x > 1.7% track each other in applied magnetic fields [18].

μSR experiments [17] also gave some evidence for a small moment present in pure UPt_3. This is distinct from the magnetic ordering found at 5 K in Th- and Pd-doped UPt_3 [19], without superconductivity at lower temperatures. Now neutron experiments [20] have found a magnetic ordering with T_N = 5 K, and an ordered moment of ≈ 0.02 μ_N/U. This is believed to be a feature of the pure and not the defect lattice. A very interesting point is that below 5 K the order parameter grows to a value which becomes constant abruptly at T_c = 0.5 K. It is important to also remark that superconductivity co-exists with small moment magnetism in URu_2Si_2 [21] and perhaps $U_{1-x}Th_xB_{13}$, as mentioned above. The presence of the small-moment ordering and the strong antiferromagnetic correlations in the superconducting state makes a magnetic mechanism for T_c in these cases plausible.

4. ANTIFERROMAGNETISM

Besides the superconducting phase transition, a number of heavy-electron metals are known to undergo antiferromagnetic phase transitions. The compound U_2Zn_{17} is one such, with T_N = 9.7 K. An extensive neutron investigation has been conducted on this material [22]. Magnetic-fluctuations, highly localized in reciprocal space, are found. A Kondo-like response function fits the data well, and it is an unusual result of this analysis that the phase transition is driven by the temperature dependence of the RKKY interaction and not that of χ. An instability criterion of the form $J\chi = 1$ becomes satisfied because J increases with decreasing temperature, χ remaining nearly constant near T_N. This temperature dependence of the RKKY interaction is the subject of the work of Jones and Varma. A possible explanation for the extreme sensitivity of T_N in U_2Zn_{17} to Cu additions [23] may then lie in the interference of these additions with the low temperature development of the RKKY interaction.

An interesting feature of the heavy-electron uranium compounds showing low-temperature magnetic transitions is that the spin configurations have been found to be commensurate. Spin configurations in many Ce systems, we note, are often incommensurate. Another feature found for the heavy-electron uranium systems is that the ordered moments have either of two values: ≈ 0.7 μ_B/U or ≈ 0.02 μ_B/U [24]. Admittedly, the data set for this observation is small, but it is remarkable, for example that Th-doped UPt_3 orders at 5 K with 0.7 μ_B/U while the pure UPt_3 has the same T_N with 0.02 μ_B/U.

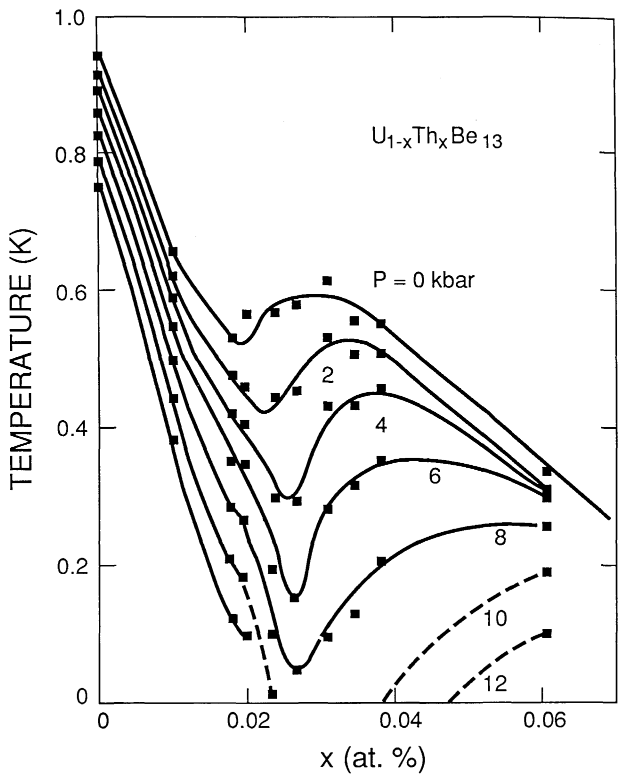

Fig. 4. Phase diagram of T_c versus x for $U_{1-x}Th_xBe_{13}$ at various pressures (from ref. 15).

Aspects of the magnetic ordering fit nicely into the qualitative model discussed earlier. Below the coherence temperature, the heavy-electron lattice is a kind of van Vleck singlet lattice, only here the singlets are not crystal-field singlets but Kondo singlets. Magnetic order can be thought about as occurring in such a lattice by a bootstrap process. Imagine a small field at one f-site. The Kondo ion there, in the small field, becomes slightly uncompensated, yielding a small net moment at that site. This small moment, via the RKKY interaction , generates a field at neighboring f-sites in the lattice. If the generated field at the neighboring site is as large as the initial small field at our original sites, a self-sustaining internal field can be generated. The internal field generated, in partially quenching the Kondo compensation, will reduce the effective mass of the conduction electrons. The loss of γ below T_N is then a loss in mass, not a loss of the Fermi surface. These losses in γ are, in fact, in accord with the comparison of high-temperature effective moments and the ordered, except in the case of URu_2Si_2. Small-moment ordering of a strange kind is also indicated in μSR experiments below 1 K in $CeAl_3$ [25].

It has not been so easy to identify heavy-electron cerium magnetic compounds because the γ's in the magnetically ordered states are much smaller than in U cases. The reason for this appears to be that the Kondo temperature of Ce in its compounds is, fairly generally, an order of magnitude smaller than for U in its compounds. If the internal fields generated upon ordering are roughly the same for both compounds, we expect a much stronger quenching of the Kondo compensation, since this will vary with the ratio H_{int}/T_0.

Another aspect of the Ce system is that because T_0 is typically of order 1 K for them, RKKY interactions often cause magnetic ordering before the heavy-electron state can develop. In some cases, such as $CeCu_5$, it appears possible to develop, with impurity substitution, a heavy-electron state from what appears to be a simple magnetic state (fig. 5).

The most interesting substitutions which alter the properties of heavy-electron systems are ordered ones, since the possibility for coherence survives. Unfortunately, such substitutions can not often be achieved. An interesting case is that of adding Au to UPt_5 to produce $UAuPt_4$, where the Au is believed to fill a special position in the lattice. Here the γ changes from 90 mJ/mole K^2 to 725 mJ/mole K^2 for $UAuPt_4$ [26].

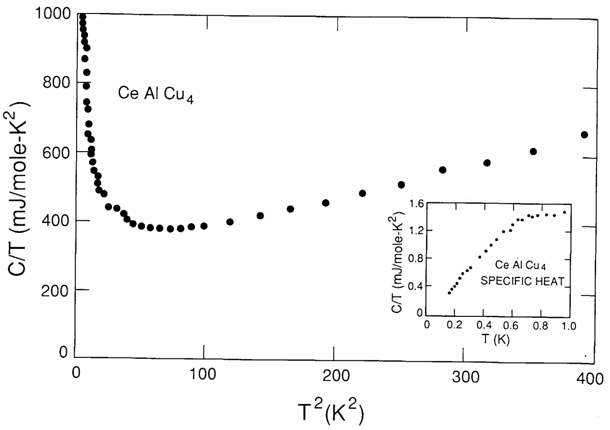

Fig. 5. Low temperature C/T versus T^2 for $CeAlCu_4$. Inset shows specific-heat data below 1 K, the slope giving a low-temperature limiting γ = 2.20 J/mole K^2 (from ref. 27).

5. CONCLUDING REMARKS

It is believed that a theory for heavy-electron metals can be developed from studies of the Anderson lattice Hamiltonian. What one hopes to achieve is a phase diagram encompassing the rich variety of phenomena observed experimentally. Do the small-moment orderings, for example, only occur near the Sommerfeld line in the (γ,χ) plot? And how are we to think about phase transitions occurring, as in UCu_5, in a heavy-electron state which develops in the presence of higher temperature magnetic order [28]?

It is important to understand how the very small energy scales observed come into being, Yb compounds can provide a useful approach to such a question because here pressure can be used as a clean probe to push the f^{13} configuration towards integral occupancy. It would be interesting if superconductors were to be found in the process.

The heavy-electron problem appears as a piece of the more complicated transition-metal problem. Spin fluctuations appear to dominate the physics of the heavy-electron metals, not charge fluctuations. In the transition metals the magnetic electrons enter into the bonding, and are consequently importantly involved in the free energy of the materials. There must exist transition-metal compounds where the spin-fluctuations dominate, and if external conditions could be changed so as to mix-in charge fluctuations, an important new attack on the larger problem could be made from the heavy-electron arena.

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