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INVITED PAPER

SUPERCONDUCTIVITY AND HEAVY FERMIONS

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Experimental studies are discussed which shed light on the nature of the heavy fermion normal state, the occurrence of heavy fermion superconductivity and anomalous properties of the superconducting state i) Magnetization and magnetoresistance of the Kondo lattice UBe_{13} demonstrate the breakdown of a description in terms of a single energy scale T_K Here T_K becomes temperature dependent below ≈ 10 K, and the canonical relationship between $R(H)$ and $M(H)$ does not hold ii) Superconductivity in UPt_3 occurs at the borderline to magnetism spin-density wave-like instabilities are induced by a few percent of Th or Au, whereas mass enhancement and susceptibility are rapidly reduced by Ir substitution This suggests that spin fluctuations, rather than phonons, are important in mediating superconducting pairing of heavy fermions in UPt_3 iii) The possibilities of noncubic strain induced by anisotropic superconductivity in UBe_{13} is investigated by ultrasound experiments The suggestion of tetragonal strain in (U Th)Be_{13} is found to be not applicable, and observed frequency and amplitude dependent absorption anomalies at $T \ll T_c$ might reflect domain wall motion

1. Introduction

The subject of heavy fermions and their superconductivity continues to offer challenging problems for both experiment and theory In this paper we chose three topics from our current work to address central themes in this field heavy fermions as the low temperature quasiparticles in a Kondo lattice, relationship between superconductivity and magnetism to elucidate the role played by spin fluctuations in mediating superconductivity and the anisotropic superconducting state itself

2. Kondo-lattice UBe_{13}: similarities and differences to the single site physics

Heavy fermions are the quasiparticles in a Kondo-lattice Whereas the single site Kondo problem is well understood in its details by now, attempts to solve the lattice problem have not been particularly successful so far [1] One of the questions is whether low and high temperature properties can be described by a single scaling parameter (T_K), and to what extent intersite interactions modify the behavior at the lowest temperatures (Vanishing of resistance is an obvious consequence) Here we report two experimental observations pertinent to this question

One of the hallmarks of the Kondo effect is the logarithmic resistance increase upon cooling, which is also seen in the Kondo lattices, with the notable exception of UPt_3 This resistance can be suppressed by magnetic fields of strength $g\mu H \approx kT_K$, as we have found in magnetoresistance measurements of UBe_{13} in fields up to 23 T and from 0 5 to 25 K In the range of overlap our results are in agreement with other studies, which also found a pronounced decrease of R in magnetic fields [2–5] Much quantitative insight is gained by realizing that the functional form of $R(H)$ is well described by $R(H)$ calculated for the $J = 1/2$ Coqblin–Schrieffer model [6] In fig 1 the

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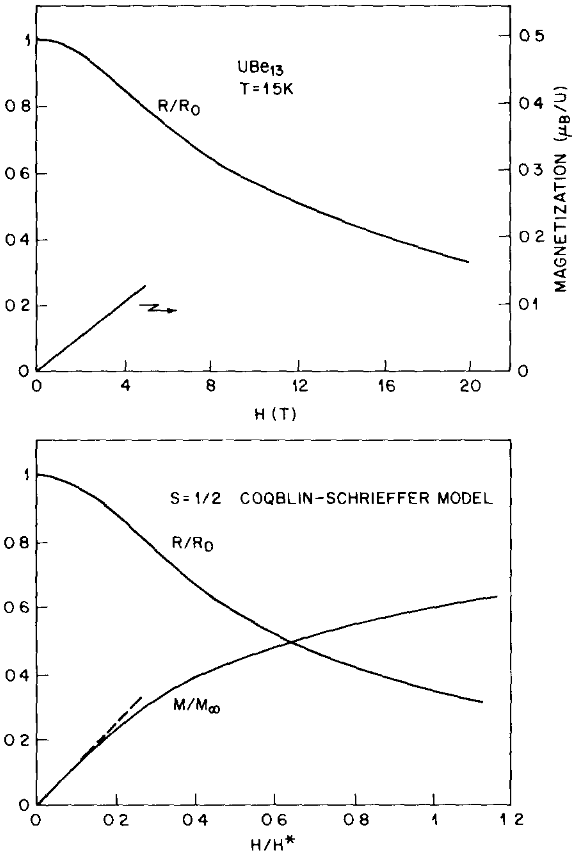

Fig 1 Comparison between physical properties of UBe_{13} and calculations for the $J = 1/2$ Coqblin-Schrieffer model [6] While the field dependence of the magnetoresistance is well described by the single site results, the magnetization is not

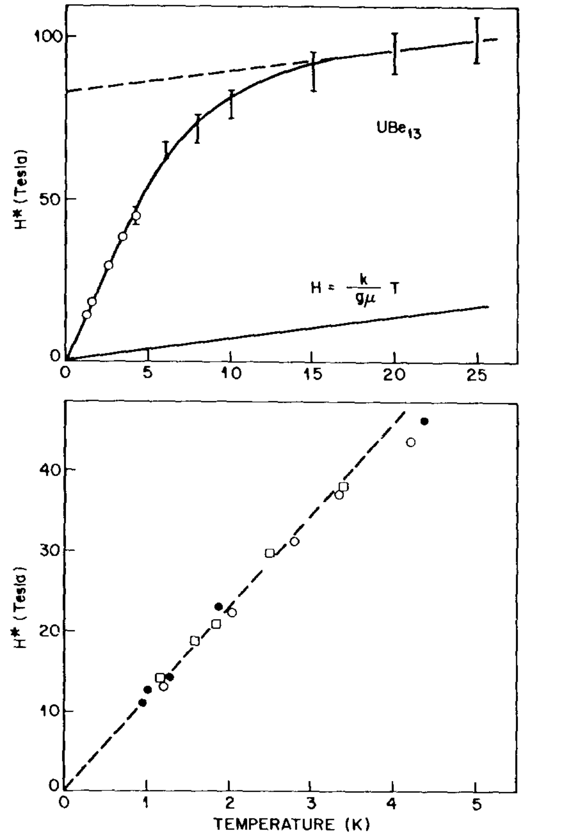

Fig 2 Temperature dependence of the characteristic field H^* from magnetoresistance For a Kondo impurity H^* is essentially the Kondo energy scale (T_K) and as such a constant In UBe_{13}, however H^* is temperature dependent below ≈ 10 K

theoretical curve, scaled to the field H^*, is compared to the experimental curve at $T = 1.5$ K At least in this respect the Kondo lattice UBe_{13} resembles the single Kondo site New behavior is found, however, when the characteristic field H^* is evaluated for each temperature by adjusting H^* such as to get optimum agreement between experiment and calculation Recall, H^* for the single impurity is simply given by $H^* \sim (T + T_K)$, reflecting the fact that the physics of the Kondo impurity is dominated by only one energy scale, T_K In UBe_{13} this is indeed observed for $T \gtrsim 10$ K, as shown in fig 2, where H^* is plotted as function of T At these high temperatures $H^* = H_K + (k/g\mu)T$ with $H_0 \approx 80$ T and $g = 2$, $\mu = \mu_B$ (More information on the precise values for g and μ appropriate for the effective doublet could, in principle, be extracted) $H_0 = 80$ T corresponds to a low T Kondo temperature $T_0 \sim 50$ K, defined by $\chi(T = 0) = S(S+1)/3T_0$ [6] Below 10 K, however, H^* changes rapidly and extrapolates linearly to 0 as $T \rightarrow 0$ Note that at all temperatures $T \ll H^*$ and H_K *itself becomes temperature dependent*, or, to put it differently, the "Kondo scale" ceases to be a constant The crossover temperature for H_K coincides with the gradual onset of mass enhancement as measured in specific heat

A second point of difference to the simple site behavior is found in the field dependent magnetization For a single site there is a strict relationship between $R(H)$ and $M(H)$, given by the field-induced variation of the $2J+1$ sublevel population When $J \leq 1$ this leads to a sublinear magnetization curve Take, for example, the case of $J=1/2$ at $H=0\ 4\ H^*$ the resistance has dropped by $\approx 33\%$, and the magnetization deviates from linearity by $\approx 17\%$ [6] We have measured the magnetization of UBe_{13} with high accuracy in a SQUID magnetometer at $T = 1\ 5$ K in fields up to 5 T and found no deviation from linearity within $\pm 0\ 2\%$ This is to be contrasted to the expectation from the resistance drop which would give a reduction from linearity by $\approx 6\%$ (see fig 1) Thus, in the Kondo lattice UBe_{13} the relationship between $R(H)$ and $M(H)$ is not the same, universal one, as for a single Kondo site

These results demonstrate in two important ways, the qualitatively new physics in a lattice of Kondo centers

3. UPt_3: proximity of superconductivity and magnetism

Among the heavy fermion compounds UPt_3 is special because its resistivity decreases monotonically below room temperature and because the specific heat at low temperatures indicates strong spin fluctuations [7,8] In loose terms, UPt_3 has a high "coherence" temperature, meaning that its physical properties are dominated by quasiparticle band-structure effects up to much higher temperatures than in the other heavy fermion compounds We have investigated this aspect of UPt_3 by systematically substituting both on the U and on the Pt sublattice Substitution changes the Fermi surface either by variation of the c/a ratio of this hexagonal compound and/or by shifting the Fermi level We found that superconductivity in UPt_3 is replaced by a spin-density wave-like instability when Au or Th substitution decreases c/a, and that spin fluctuations are effectively suppressed when Ir substitution increases c/a

An example of the magnetic instability is discussed here (For more detail on $(U,Th)Pt_3$ see ref [9]) When Au or Th is substituted in UPt_3, the magnetic susceptibility at $T \to 0$ increases and develops for more than $\approx 3\%$ Au or Th an anomaly at 6 5 K with a drop in $\chi(T)$, typical for

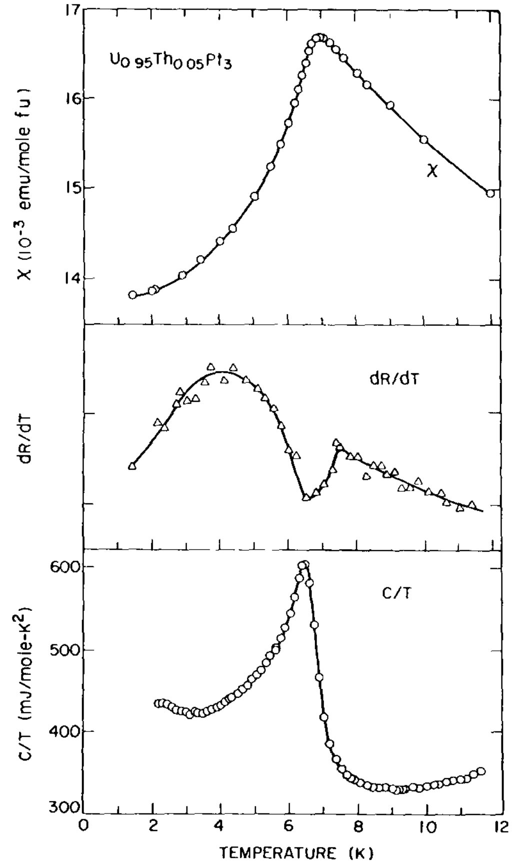

Fig 3 A comparison of the anomalies at $T_0 = 6\ 5$ K in $U_{0\ 95}Th_{0\ 05}Pt_3$, which indicate a Fermi surface instability of the same type as in chromium (spin-density wave)

an antiferromagnetic-like ordering (fig 3) Specific heat measurements reveal the corresponding anomaly in $C(T)$ Analysis of the excess specific heat $\Delta C(T)$ and of the resistance change at $T=6\ 5$ K rules out local moment magnetism and suggests a spin-density-wave-like Fermi surface instability the specific heat $\Delta C(T)$ is of an activated nature, obeying well an Arrhenius law with an activation energy of $26k_B$, resulting in a strong coupling value of 4 for Δ/T_0 The second evidence for the development of an energy gap on part of the Fermi surface is seen in the upturn of $R(T)$ below T_0, just as in chromium The fraction of the Fermi surface affected by this instability is small ($\approx 10\%$), as estimated either from the

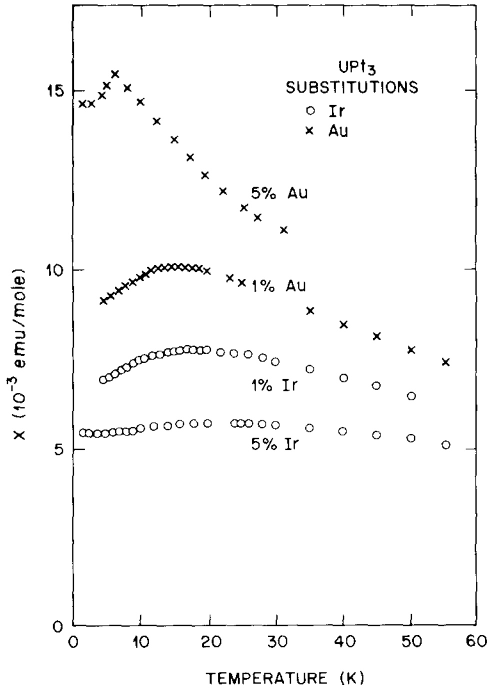

Fig 4 Susceptibility of Au and Ir substituted UPt_3. Ir suppresses $\chi(T)$ and also the electronic specific heat, whereas Au enhances $\chi(T)$ and induces the same Fermi surface instability as Th

change in $\chi(T)$, the resistance anomaly or the entropy under the $\Delta C(T)$ curve ($\approx 8\%$ R ln 2). Neutron scattering experiments [10] have confirmed the magnetic nature of this Fermi surface instability, and the ordered moment was found to be $\approx 0.6 \mu_B/U$. When Ir is substituting for Pt in UPt_3, spin fluctuations are rapidly suppressed. The susceptibility and the T^3 ln T part of the specific heat decrease, as well as the linear coefficient γ (fig 4)

These results can be summarized by plotting, e g , the low temperature susceptibility χ, as function of substituent concentration (fig 5). The data fall on one line when the atomic percent concentration is used as a scale to measure the small changes in the c/a ratio. It is apparently the variation of c/a, and not so much the electron/atom ratio, that determines the physical properties. The unconventional nature of superconductivity in UPt_3 is suggested by fig 5 as c/a decreases only slightly (e g , 0.8517 for UPt_3, 0.8511 for 10% Th in UPt_3), details of the

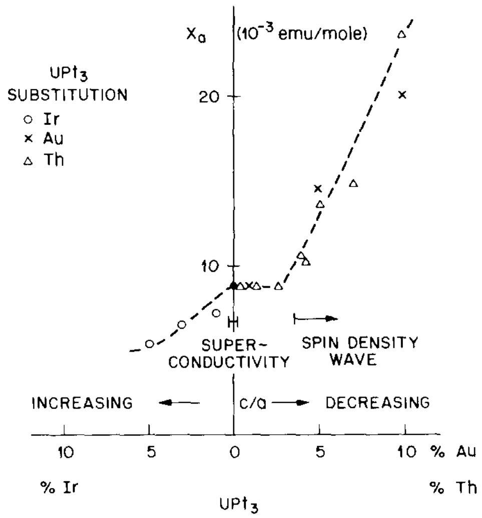

Fig 5 An example of the systematic variation of physical properties when Th, Au or Ir are added to UPt_3. This demonstrates that superconductivity in UPt_3 occurs as a "precursor" to a spin-density wave and thus is most likely mediated by spin fluctuations

quasiparticle band structure are modified. The resulting changes in the electronic behavior are quite drastic: moderately heavy on the Ir side ($\gamma \approx 260$ mJ/mol K^2 for 5% Ir), spin-fluctuations and superconductivity for pure UPt_3, and finally a spin-density wave for more than $\approx 3\%$ Th or Au. A similar systematic variation is observed for the temperature at which the susceptibility maximum occurs (23 K for 5% Ir, 17 K for UPt_3 and 10 K for $\approx 3\%$ Th). We also note that substitution of Pd for Pt results in the same transition as we found for Au and Th substitution [11,12], and more recent resistivity measurements seem to confirm our original interpretation in terms of Fermi surface instabilities [13]

This sequence of electronic ground states is quite unusual and highlights the peculiarity of heavy fermion superconductivity in UPt_3. As the band parameters (Fermi surface nesting conditions, enhancement factors, ...) vary in such a way as to allow the development of a Fermi surface instability in the form of a spin-density wave, superconductivity occurs just when spin fluctuations are strong, but not yet strong enough to drive a magnetic instability

This remarkable proximity of superconductivity and magnetism supports the view that superconductivity in UPt_3 is not mediated by phonons, but rather by spin-fluctuations The present results, in addition to the ones obtained from neutron scattering studies [14] imply that the spin fluctuations are dominantly antiferromagnetic, not ferromagnetic as commonly inferred from the $T^3 \ln T$ term in the specific heat The q-dependence of the spin fluctuations is important in determining the effective potential for superconducting pairing and the symmetry of the pair wavefunction Recent calculations indicate that antiferromagnetic fluctuations enhance anisotropic even-parity pairing and suppress odd-parity pairing [15]

4. The superconducting state

By now little doubt exists that superconductivity involving heavy fermions is not of conventional type Specifically, all experimental probes sensitive to the quasiparticle density of states agree in that their temperature dependence is nonexponential [1] The discussions are only whether the gap vanishes along lines or on points on the Fermi surface, and it has been suggested that all experimental results (on all compounds) are consistent with lines of zero gap [16] Instead of entering this discussion, we focus here on a different aspect

When a few percent of Th are substituted for U in UBe_{13} the superconducting transition temperature T_{C1} is reduced nonmonotonically [17] and a second phase transition at T_{C2} occurs below T_{C1} This lower transition was first seen in specific heat measurements [18] and its nature is not clear yet Ultrasound studies found a large absorption peak associated with T_{C2} [19] From the character of the attenuation and the variation in sound velocity, which are very reminiscent of an antiferromagnetic transition, the possibility of a weak magnetic transition was suggested Estimates for the ordered moment gave $\approx 10^{-2} \mu_B/U$

An alternative explanation for the ultrasound absorption peak was proposed [20] assuming a transition between two different superconducting states Here the idea is that an anisotropic superconducting state in a cubic crystal drives a tetragonal or rhombohedral lattice distortion Domain wall motion would dissipate ultrasound energy and, depending on the superconducting state, the symmetry of the distortion and the propagation and polarization direction of the sound wave, prediction can be made about the presence or absence of attenuation peaks In the original ultrasound experiments longitudinal sound was propagated along a (100) direction The model in ref [20] predicted for the specific case of tetragonal strain ultrasound absorption for (100) but not for (111) propagation To test this, we performed studies with longitudinal sound propagating along (111) or (100) in two crystals of $U_{1-x}Th_xBe_{13}$ with $x = 0\,03$ A typical temperature dependence at 120 MHz is shown in fig 6 The attenuation peak is present not only for (100) but also for the (111) direction, and the peak values are of comparable size Thus, we conclude

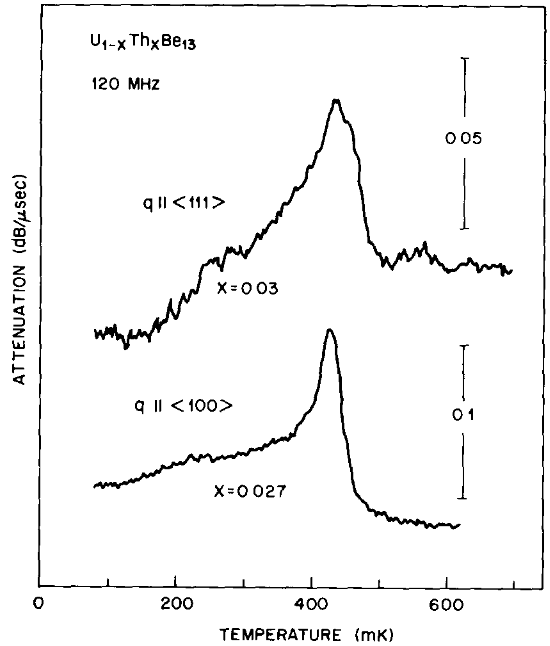

Fig 6 Ultrasound absorption associated with the lower transition in superconducting (U, Th)Be_{13} Longitudinally polarized sound propagates either in 100 or 111 directions The two peaks are of comparable magnitude If they were caused by motion of tetragonally strained domains due to anisotropic superconductivity coupling to the lattice, the peak for 111 propagation would be absent

that coupling of ultrasound to a tetragonally distorted superconducting phase cannot explain the observed attenuation peak

While this particular prediction based on anisotropic superconductivity has not been verified in ultrasound experiments, we have, however, observed different anomalies which indeed might reflect anisotropic superconductivity For low frequencies (≤50 MHz) a weak absorption and change in sound velocity occurs well below the two transition temperatures (100–150 mK) The anomalies are frequency and amplitude dependent and become more pronounced at low frequencies and small amplitudes So far, three samples with various concentrations of Th (0 9, 1 7 and 3%) are found to exhibit this coupling to ultrasound and experiments on undoped UBe_{13} are currently in progress At this point only speculations on the origin of these anomalies can be offered Domain wall motion with pinning on impurities, similar in spirit to the above discussed model, is one possibility Alternatively, it needs to be explored if texture of an anisotropic state would cause ultrasound anomalies of the observed character

5. Summary

The three fields of current research which we discussed here reveal insight into the normal state of heavy electrons, the close proximity of superconductivity and magnetism and the anisotropic nature of the superconducting state

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