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## **MUON SPIN RELAXATION STUDIES OF HEAVY FERMION SUPERCONDUCTORS**

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Invited talk presented at the International Workshop on Low Energy Science (LEMS'93), 4-8 April, 1993, Santa Fe, NM, and to be published as Internal Report.

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This talk will focus recent developments in our understanding of heavy fermion (HF) superconductors and the role that positive muon spin relaxation ( $\mu$ SR) studies have played in helping to elucidate their properties. As illustrations two systems will be discussed: (1)  $UPd_2Al_3$ , one of the most recently discovered HF superconductors, which also displays coexisting magnetic order and (2)  $UBe_{13}$  doped with small quantities of Th substituted for U, which displays an interplay between its superconducting and magnetic ground states, leading to multiple superconducting states.

## Muon Spin Relaxation Studies of Heavy Fermion Superconductors

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### Abstract

This talk will focus recent developments in our understanding of heavy fermion (HF) superconductors and the role that positive muon spin relaxation ( $\mu$ SR) studies have played in helping to elucidate their properties. As illustrations two systems will be discussed: (1)  $\text{UPd}_2\text{Al}_3$ , one of the most recently discovered HF superconductors, which also displays coexisting magnetic order and (2)  $\text{UBe}_{13}$  doped with small quantities of Th substituted for U, which displays an interplay between its superconducting and magnetic ground states, leading to multiple superconducting states.

### **I. Introduction**

Heavy fermion behavior arises in regions of the periodic table where the atoms possess f-electrons which are neither highly localized nor highly itinerant [1]. Typically such behavior occurs in cerium-or uranium-based compounds possessing 4-f and 5-f electrons, respectively. At high temperatures the f-electrons act like local moments with close to the full f-shell moment ( $\sim 3\mu_B$ , for example), exhibiting Curie-like susceptibilities. At low temperatures the f-moments are reduced to a fraction of their high-temperature values through hybridization with the ligand conduction electrons. The f-electrons thus take on an itinerant character and the conduction electrons become "heavy". This is manifest through a narrow resonance at or near the Fermi level, producing a high density of states at low temperatures. In addition, below a characteristic temperature the resistivity drops, indicating the onset of coherent electron scattering, which also characterizes the heavy fermion state. The exact mechanism by which these phenomena occur in a lattice of f-atoms is not well understood, and constitutes one of the major challenges of many-body physics in solids today.

The heavy electron ground state has been shown to exhibit paramagnetism, magnetic order and/or superconductivity. The ordered moments are typically small, of order  $10^{-3} - 10^{-1} \mu_B/\text{f-atom}$ . The large entropy collapse associated with the superconducting state points to the f-electrons as the superconducting pairs. Frequently, small-moment magnet order (also from the f-electrons) coexists with the superconductivity, each presumably occupying different parts of the Fermi surface

Heavy electron superconductors have drawn considerable attention because their properties are distinctly different from conventional Type II BCC superconductors like niobium, for example. In conventional

superconductors the superconducting gap exists over the entire Fermi surface, and the electrons pair in a zero angular momentum, spin-singlet state which is produced by the electron-phonon interaction. A non-vanishing gap gives rise to exponential temperature dependences for all measurements involving the thermal excitation of quasiparticles across the gap: specific heat, magnetic-field penetration depth ( $\lambda$ ) and nuclear spin lattice relaxation rate ( $1/T_1$ ), as examples. However, measurements of these quantities in heavy fermion superconductors all exhibit power-law temperature dependences, usually considered as evidence for nodes in the energy gap. Table I gives the expected power-laws for two different kinds of nodal structures on a spherical Fermi surface, lines and points.

**TABLE I**

	<b>Polar</b>	<b>Axial</b>
$C_p$	$T^2$	$T^3$
$1/T_1$	$T^3$	$T^5$
$1/\lambda^2_{\parallel}$	$T^3$	$T^2$
$1/\lambda^2_{\perp}$	$T$	$T^4$
$\chi_{\text{spin}}$	$T^2$	$T^3$

Table I. Temperature dependence of specific heat ( $C_p$ ), spin lattice relaxation rate ( $1/T_1$ ), magnetic field penetration depth (for vector potential parallel ( $\lambda_{\parallel}$ ) and perpendicular ( $\lambda_{\perp}$ ) to the order-parameter symmetry axis) and spin susceptibility ( $\chi_{\text{spin}}$ ). The polar and axial gap structures correspond to odd-parity states with an equatorial line of nodes and two point nodes, respectively. An isotropic gap produces more nearly exponential temperature dependencies

Power-law temperature dependencies do not provide definitive evidence for an unconventional gap structure, however [2] (A conventional gap-less superconductor can also exhibit power-law behavior, for example). More decisive conclusions can only be drawn from tests of the symmetry-breaking nature of the order parameter  $\Lambda(\mathbf{k})$ . In conventional superconductors  $\Lambda(\mathbf{k})$  obeys the symmetry of the Hamiltonian, which includes rotational, reflection (parity) and time-reversal symmetry. An unconventional superconductor has a lower symmetry in at least one of these respects. Experimental tests

of this property include a transition from one superconducting state to another, observation of magnetism associated with the superconducting order parameter, or anisotropy in the temperature dependence of the penetration depth or the critical fields.

This paper concerns  $\mu$ SR studies in two heavy fermion superconductors,  $\text{UPd}_2\text{Al}_3$  and  $(\text{U,Th})\text{Be}_{13}$ , and will address some of the evidence for unconventional superconductivity in these systems. The  $\mu$ SR experiments were carried out at the Paul Scherrer Institute in Villigen, Switzerland, and involve a collaboration between Los Alamos, ETH-PSI, Leiden University, U.C. Riverside, Darmstadt and Tohoku Universities.

## II. $\text{UPd}_2\text{Al}_3$

This material has a superconducting transition temperature  $T_c = 1.5 - 2.0\text{K}$  (depending on sample quality) and an antiferromagnetic transition at  $T_N = 14\text{K}$ , with an ordered moment  $\sim 0.85\mu_B$  [3]. The magnetic structure consists of ferromagnetic sheets in the basal plane coupled antiferromagnetically along the c-axis. The NMR  $1/T_1$  has a  $T^3$  temperature dependence [4] below  $T_c$ .

Zero-field and transverse-field  $\mu$ SR data on both single crystalline [5] and polycrystalline [6]  $\text{UPd}_2\text{Al}_3$  are consistent with the positive muon occupying the  $(0,0,1/2)$  site in the unit cell. The temperature dependence of the transverse-field  $\mu$ SR rate for polycrystalline  $\text{UPd}_2\text{Al}_3$  is shown in Fig 1 between about 0.1 and 300K.

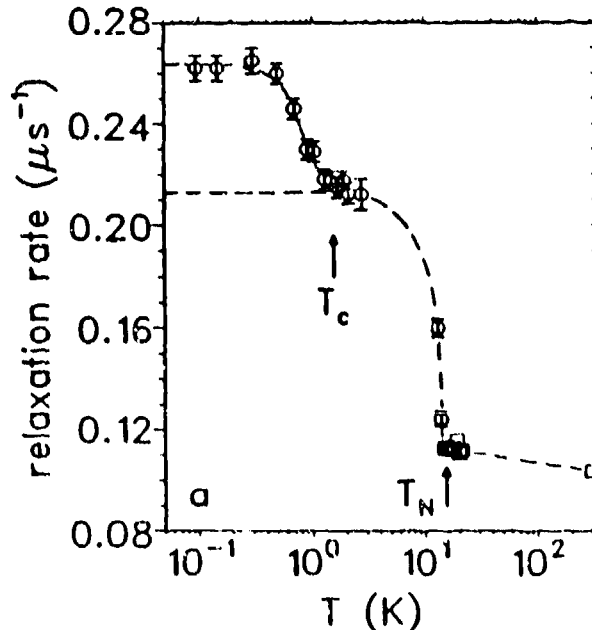


Fig. 1. The  $\mu$ SR relaxation rate for  $\text{UPd}_2\text{Al}_3$  in transverse applied field of 340 G for  $T < 10\text{K}$  and 117 G for  $T > 10\text{K}$ .  $\circ$ - exponential rate,  $\square$  - Gaussian rate (corrected by  $1/\sqrt{2}$  to allow comparison). From Ref. 6.

There is a rapid rise below  $T_N$  due to the AFM order and a further rise below  $T_c$  due to the flux lattice formed in the superconducting state. This indicates a coexistence of superconducting and magnetic f-electron states. The data below  $T_c$  was further analyzed as a product of a Gaussian times an exponential function. The exponential relaxation rate accounts for the AFM ordering and was held constant for  $T < T_c$ . The Gaussian rate roughly describes the inhomogeneous broadening from the flux lattice and for an ordered triangular lattice is proportional to  $1/\lambda^2$ . The temperature dependence of  $1/\lambda^2$  is compared to several models of superconductivity in Fig 2. The  $\lambda(0) = 6250 \pm 1250$  Å.

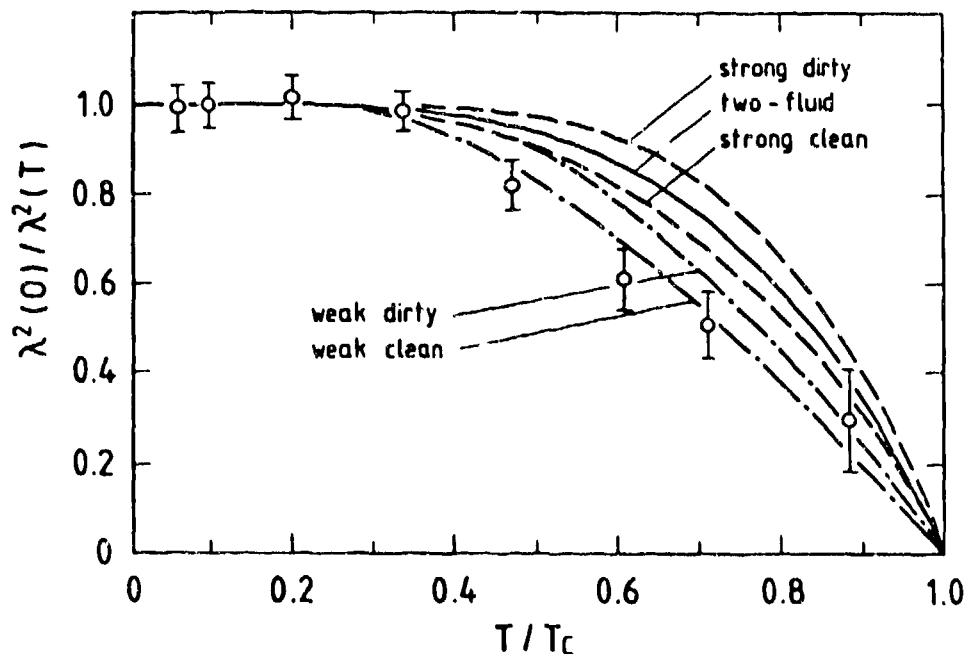


Fig. 2. The temperature dependence of the inverse-square magnetic field penetration depth in  $UPd_2Al_3$  compared to various models for the superconducting state. From Ref 6.

The data are consistent with a clean, weak-coupling BCS superconductor, i.e., no nodes in the gap. This in turn is consistent with the upper critical field data, which show no anisotropy and Pauli limiting at low temperatures, characteristic of spin-singlet pairing. On the other hand the  $1/T_1$  data do not show the exponential temperature dependence expected for a node-less gap. Furthermore, the  $\mu$ SR-determined  $1/\lambda^2$  data are equally well fit to a power-law temperature dependence ( $1/\lambda^2 \propto 1-T^\nu$ ,  $\nu = 2.3 \pm 0.2$ ), consistent with a nodal structure in the gap. (At this writing, no single-crystal  $\mu$ SR data below  $T_c$  are available, and so the nodal structure from  $\lambda(T)$  cannot be specified.) It is possible that

all of the data could be consistent with a d-wave ( $l = 2$ ), spin-singlet pairing state, however.

### III. (U,Th)Be<sub>13</sub>

This material has been studied for many years [7], but it is only recently that its peculiar superconducting phase diagram may be qualitatively understood. Pure UBe<sub>13</sub> is superconducting at  $T_C \cong 0.9$ K, but no AFM order has yet been established at any temperature above 10 mK. Upon doping with Th the  $T_C$  for U<sub>1-x</sub>Th<sub>x</sub>Be<sub>13</sub> becomes non-monotonic, first decreasing as  $x$  increases, and then sharply increasing at  $x \cong 0.019$ . The  $T_C$  passes through a maximum near  $x \cong 0.030$ , after which  $T_C$  monotonically decreases again. In the region  $0.019 < x < 0.043$ , a second superconducting transition at  $T_{C2}$  is observed in specific heat, accompanied by the onset of very small moment ( $10^{-3} - 10^{-2} \mu_B/U$ ) magnetism, which has been well characterized by  $\mu$ SR and other techniques [8]. The phase diagram is shown in Fig. 3.

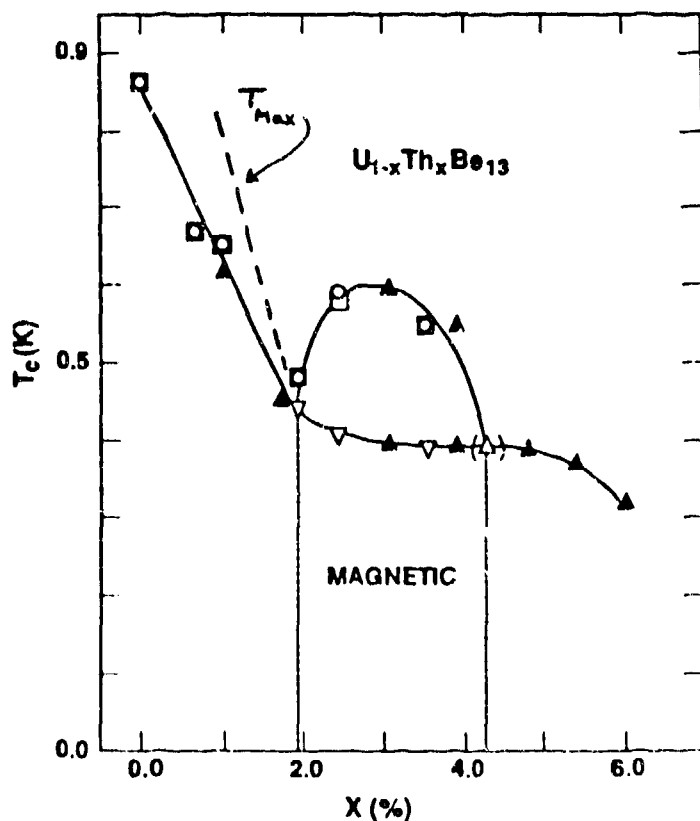


Fig. 3. Phase diagram for U<sub>1-x</sub>Th<sub>x</sub>Be<sub>13</sub>. The various symbols represent different types of measurements, explained in Ref. 8. The dotted line shows the approximate Th-dependence of the resistivity maximum  $T_{max}$ .

An important question has been whether the magnetic correlations are intrinsic to the superconducting state (thus violating time reversal invariance), or whether the correlations are due to weak AFM order coexisting with a changed superconducting state. Recent normal-state measurements [9] of the magnetoresistance and specific heat, combined with previous measurements of the resistivity under pressure [10] have helped to resolve this controversy and qualitatively explain the non-monotonic temperature dependence of  $T_C$  in  $(U,Th)Be_{13}$ .

The resistivity and specific heat of  $UBe_{13}$  both show a maximum near  $T_{max} = 2.2K$  which moves down with Th doping until it passes below  $T_C$  at  $x \approx 0.019$ , exactly where  $T_C$  begins to increase again and where magnetic correlations are seen in  $\mu SR$ . Application of pressure [10] to  $U_{1-x}Th_xBe_{13}$  increases both  $T_{max}$  and the resistivity  $\rho$ , such that  $\rho/\rho_{max}$  scales with  $T/T_{max}$  for many different pressures, indicating a new energy scale given approximately by  $T_{max}$ . This idea is qualitatively consistent with theoretical models of the normal state behavior of heavy fermion systems which show that a new energy scale emerges from the intersite exchange coupling between Kondo ions [11]. Recently a systematic study [9] of resistivity and specific heat as a function of Th concentration and magnetic field has strongly suggested that the feature at  $T_{max}$  is produced when the conduction electrons are scattered by the correlated motion of the U spins, so-called spin fluctuations. This spin-fluctuation temperature is reduced by impurities such as Th, as is the superconducting temperature  $T_C$ . (See Fig. 3) When  $T_{max} < T_C$  the magnetic fluctuations are "frozen-out," so to speak, yielding weak magnetic order. It is possible that the superconducting and magnetic order parameters are coupled, causing an increase in  $T_C$  at  $x \approx 0.019$ . Another possible explanation for the non monotonic behavior of  $T_C$  is that the pairbreaking effects of the spin fluctuations are reduced when  $T_{max} < T_C$ , thus driving up  $T_C$ . Therefore the complicated superconducting phase diagram of  $(U,Th)Be_{13}$  appears to arise from the interaction of the superconducting f-electrons with the incipient magnetic correlations also present in the f-electron system.

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