Muon spin relaxation in the heavy fermion system UPt_3

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We report muon spin rotation/relaxation (μSR) measurements of the heavy fermion superconductor UPt_3 in external fields $H_{ext}\|\hat{c}$. We find that the muon Knight shift is unchanged in the superconducting state, consistent with odd-parity pairing (such as p wave). The transverse field relaxation is observed to be strongly field dependent, decreasing with increasing field. Below T_c the increase is barely detectable in an applied field of 4 kG$\|\hat{c}$. On the basis of the high field measurements, we estimate the low temperature penetration depth to be $\lambda(T\to 0)\gtrsim 11\,000$ Å.

There is a growing body of experimental evidence that shows that the heavy fermion system UPt_3 (Ref. 1) is a non-s-wave superconductor. Neutron scattering[2] and heat capacity measurements detect strong spin fluctuations in the superconducting state, thought to favor anisotropic pairing (such as p or d wave). In addition, ultrasound velocity[3] and heat capacity[4] measurements have detected possible phase boundaries within the superconducting state. The existence of several superconducting phases has spurred the development of theories to identify the various states. In this vein, several authors[5] have suggested that UPt_3 possesses a multicomponent superconducting order parameter transforming according to a nonidentity representation of the hexagonal D_{6h} group. There is still great uncertainty about the properties of UPt_3: even the parity of the superconducting pair state has not been unambiguously determined.

Many properties of the superconducting state can reflect the underlying symmetry of the pairing. Among these are the spin susceptibility χ and the magnetic field penetration depth λ. In an even parity (such as s wave) superconductor, the electrons are paired in states with opposite spin. The combined susceptibility of that pair is 0; the measured spin susceptibility reflects that of the normal state electrons, approaching zero as the temperature approaches zero [following the Yosida function $Y(T)$]. In odd parity superconductors, different susceptibilities are possible, depending on the pair wavefunction, and can be markedly different from the even parity case. For example, the triplet ABM and BW states of ^{3}He have susceptibilities $\chi_{ABM}/\chi_n = 1$, $\chi_{BW}/\chi_n = \frac{2}{3} + \frac{1}{3}Y(T)$, respectively, where χ_n is the normal state susceptibility.

The magnetic field penetration depth λ describes the screening effects of the superconducting electrons. Its temperature dependence can also provide information about the pairing symmetry.[6] If there are nodes in the superconducting gap, characteristic of higher l pairing, thermal pair breaking will give rise to a power law temperature dependence in λ. By comparison, s-wave superconductors have no nodes in the gap, and as a result, the penetration depth shows little temperature dependence for $T < T_c/3$.

Muon spin relaxation measurements are useful for determining both $\lambda(T)$ and $\chi(T)$ simultaneously.[7] In a time differential μSR experiment, 100% spin polarized positive muons are injected individually into a specimen, where the muon spins precess in the local magnetic field. The μ^+ decays (lifetime $\tau_\mu = 2.2\,\mu s^{-1}$), emitting a positron, preferentially along the instantaneous muon spin direction. A histogram of positrons detected versus the time interval after implantation will exhibit the lifetime exponential decay superimposed on the muon spin polarization function. The "asymmetry," which is the ratio of the difference and the sum of spectra from two opposing counters, is directly proportional to the muon polarization. Typically, in transverse field, the polarization function is given by

$$\mathscr{P}(t)=A_0\,[\cos(\omega t+\phi)]\,\exp(-\sigma^2 t^2/2), \qquad (1)$$

where the frequency is given by the local field $\omega = (\gamma_\mu/2\pi)B_{loc}$ and the relaxation rate σ reflects the inhomogeneity in the local field.

The local field can be different from the applied field due to a muon-conduction electron hyperfine interaction. The measured fractional shift in the muon precession frequency from this interaction is the sum of the muon Knight shift (K_μ), Lorentz and demagnetizing shifts, and a diamagnetic shift in the superconducting state; and is given by

$$\frac{B_{loc}-B_{ext}}{B_{ext}}=K_\mu+4\pi\,(\tfrac{1}{3}-n)(\chi-\chi_d)+4\pi\chi_d(1-n). \qquad (2)$$

All of the present experiments were performed at the TRIUMF M15 surface muon channel. A mosaic of pieces 2 mm$\times$0.3 mm thick cut from a single crystal using spark erosion was mounted on pure silver. Below 4 K, the mosaic was then mounted with the sample $\hat{c}$-axis oriented along the applied field direction on the mixing chamber extension of an Oxford Model 400 dilution refrigerator. Measurements above $T = 4$ K were performed using a conventional

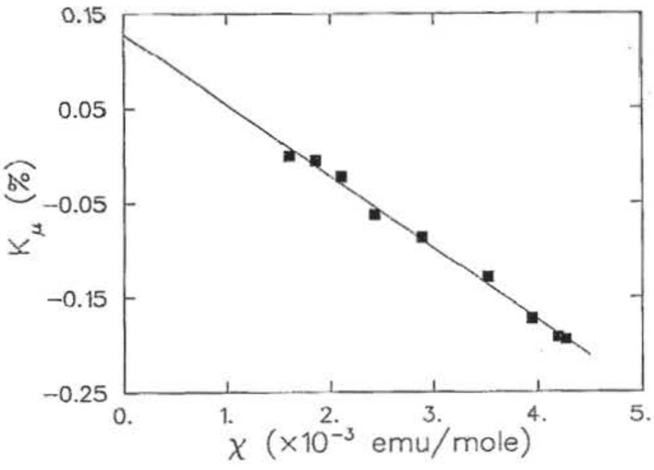

FIG. 1. Muon Knight shift (including Lorentz and demagnetizing corrections), plotted as a function of dc susceptibility, for $H \| \hat{c}$. Temperature is an implicit variable.

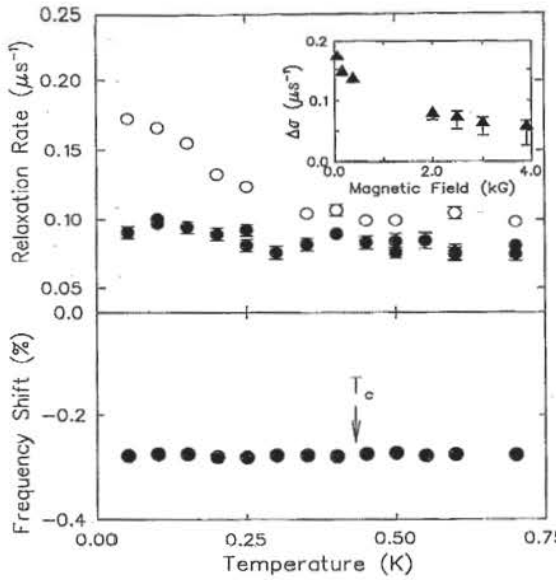

FIG. 2. (a) Transverse field muon spin relaxation rate vs temperature for $B_{ext} = 150$ G (open circles) and 4 kG (filled circles). (inset) Field dependence of increase in σ below T_c. (b) Fractional muon frequency shift (measured in $B_{ext} = 4$ kG), which is unchanged below T_c.

He flow cryostat. Field stability was controlled using an NMR-based feedback loop.

In UPt_3, K_μ is large due to the large Pauli-like susceptibility of the heavy electrons. Fourier transforms of data taken in a large external field (~ 4 kG) show two signals, one from UPt_3, the second from muons in the silver sample holder. Silver has a very small, temperature-independent Knight shift, providing a useful reference measurement of B_{ext}.

Comparing measurements of the muon Knight shift with those of the dc susceptibility (both with the field applied along the $\hat{c}$ axis), we see a similar temperature dependence; plotting K_μ vs χ in Fig. 1, we obtain a linear relationship. Since the susceptibility in the basal plane displays a different temperature dependence,[1] we see that the muon Knight shift reflects the susceptibility for fields along the $\hat{c}$ axis. The susceptibility χ is largely due to the spin susceptibility χ_S; extrapolating to $\chi_S = 0$, we find $K_\mu(\chi_S = 0) = +0.13\%$. The slope of the K_μ vs χ curve gives us a hyperfine field of about -4.2 kG/μ_B. This is substantially larger (and of opposite sign) than reported in previous measurements of polycrystalline UPt_3,[8] which is not surprising in view of the strong anisotropy of χ in UPt_3.

Upon cooling through $T_c \approx 0.45$ K, we see that there is *no* discernable change in K_μ. The measured shift, shown in Fig. 2(b), remains about -0.3%, of which about -0.12% comes from Lorentz and demagnetizing shifts (which like the Knight shift are proportional to χ). Diamagnetic contributions to the shift $[4\pi\chi_d(1 - n)]$ due to superconductivity are negligible since n is near to 1 in our geometry and χ_d is small in high fields. We therefore conclude that the spin susceptibility is unchanged below T_c. This is in agreement with ^{195}Pt-NMR (Ref. 9) and induced moment form factor measurements.[10]

In addition to measuring the frequency of the precession signal, we have simultaneously determined the relaxation rate $\sigma(T)$ for several fields $(H \| \hat{c})$ up to 3.9 kG. The inhomogeneity in the local fields from the vortex lattice[11] is manifested in an increase of the relaxation rate σ. In the London limit,[12] $(H < H_{c2}/4)$, the relaxation rate $\sigma \propto \lambda^{-2} \propto n_s/m^*$, where λ is the magnetic field penetration depth, n_s is the superconducting carrier density, and m^* is the carrier effective mass. In 3.9 kG, there is at most a very small increase in σ below T_c [filled circles, shown in Fig. 2(a)]. After subtracting the temperature-independent background, we find $\sigma(0) < 0.06\ \mu s^{-1}$, corresponding to $\lambda(0) \geqslant 11\,000$ Å.

Lower field measurements [$H \sim 200$ G, open circles, see Fig. 2(a)] show enhanced relaxation below T_c, in agreement with previous work.[13] Plotting the field dependence of the increase in the relaxation rate below T_c [inset of Fig. 2(a)], we see that there is a reasonably smooth decrease in σ with increasing applied field. We expect that σ should be field independent over a large range of fields between H_{c1} and H_{c2} (Refs. 11,12) in order to extract λ. If the measured inhomogeneity is field dependent it generally implies that the measured relaxation does not accurately reflect the penetration depth. In this case, the value of 11 000 Å can only serve as a *lower* bound for the penetration depth, which may in fact be much longer.

There are several possible sources of increased broadening on low fields that could account for the enhanced relaxation. One of these is flux pinning, acting to prevent formation of a uniform flux lattice. Zero field cooled measurements in 3.9 kG show greatly enhanced relaxation, characteristic of strong flux pinning. Other possible sources of low field broadening below T_c include proximity to H_{c1} and shape-dependent inhomogeneities in the demagnetizing factor.[14]

Ultrasound measurements[3] have detected an anomaly in UPt_3 around $H = 12$ kG (for $H \| \hat{c}$). It has been suggested that this anomaly indicates a phase boundary between different superconducting states. There is a possibility that our field-dependent relaxation may be related to this anomaly. However, we are prevented from accessing the feasibility of such an effect by a lack of theoretical understanding of the superconducting states of UPt_3. Nevertheless, we note that all of these μSR measurements lie in the London limit, where we do not expect significant field dependence in the relaxation rate.

In conclusion, we find that the muon Knight shift is unchanged in the superconducting state of UPt_3, supporting the idea of odd-parity pairing. Although it is possible for spin orbit scattering to reduce changes in the Knight shift,[15] we would argue the the long mean free path [$l \sim 1000$ Å in UPt_3 (Ref. 16)] suggests that scattering is not important here.

We estimate that the low temperature penetration depth is in excess of 11 000 Å, roughly consistent with the estimate from a flux confinement measurement [$\lambda(0) \sim 19\,000$ Å].[17] Since the penetration depth $\lambda \propto m^*/n_s$ and the effective mass is large [e.g., cyclotron effective mass $m_c = 25 \rightarrow 90 m_e$ (Ref. 16)], we expect λ to be rather large. The change below T_c in relaxation rate is so small in high fields that is is not possible to discuss its temperature dependence in terms of different possible gap node structures.

[1] A. De Visser, A. Menovsky, and J. J. M. Franse, Physica **147B**, 81 (1987).
[2] G. Aeppli, A. Goldman, G. Shirane, E. Bucher, and M.-Ch. Lux-Steiner, Phys. Rev. Lett. **58**, 808 (1987).
[3] G. Bruls, D. Weber, B. Wolf, P. Thalmeier, B. Lüthi, A. de Visser, and A. Menovsky, Phys. Rev. Lett. **65**, 2294 (1990); S. Adenwalla, S. Lin, Q. Ran, Z. Zhao, J. Ketterson, J. Sauls, L. Taillefer, D. Hinks, M. Levy, and B. Sarma, Phys. Rev. Lett. **65**, 2298 (1990).
[4] R. A. Fisher *et al.*, Phys. Rev. Lett. **62**, 1411 (1989); K. Hasselbach, L. Taillefer, and J. Flouquet, Phys. Rev. Lett. **63**, 93 (1989).
[5] See for example, T. A. Tokuyasu, D. W. Hess, and J. A. Sauls, Phys. Rev. B **41**, 8891 (1990).
[6] D. Einzel, P. J. Hirschfeld, F. Gross, B. S. Chandrasekhar, K. Andres, H. R. Ott, J. Beuers, Z. Fisk, and J. L. Smith, Phys. Rev. Lett. **56**, 2513 (1986); F. Gross, B. Chandrasekhar, D. Einzel, K. Andres, P. J. Hirschfeld, H. R. Ott, J. Beuers, Z. Fisk, and J. L. Smith, Z. Phys. B **64**, 175 (1986).
[7] For a review of aspects of the μSR technique, see A. Schenck, *Muon Spin Rotation Spectroscopy: Principles and Applications in Solid State Physics* (Hilger, Bristol, 1985).
[8] R. H. Heffner, D. W. Cooke, A. L. Giorgi, R. L. Hutson, M. E. Schillaci, H. D. Rempp, J. L. Smith, J. O. Willis, D. E. MacLaughlin, C. Boekema, R. L. Lichti, J. Oostens, and A. B. Denison, Phys. Rev. B **39**, 11 345 (1989).
[9] Y. Kohori, T. Kohara, H. Shibai, Y. Oda, T. Kaneko, Y. Kitaoka, and K. Asayama, J. Phys. Soc. Jpn. **56**, 2263 (1987).
[10] C. Stassis, J. Arthur, C. F. Majkrzak, J. D. Axe, B. Batlogg, J. Remeika, Z. Fisk, J. L. Smith, and A. S. Edelstein, Phys. Rev. B **34**, 4382 (1986).
[11] P. Pincus, A. C. Gossard, J. Vaccarino, and J. H. Wernick, Phys. Lett. **13**, 21 (1964).
[12] E. H. Brandt, Phys. Rev. B **37**, 2389 (1988).
[13] C. Broholm, G. Aeppli, R. N. Kleiman, D. R. Harshman, D. J. Bishop, E. Bucher, D. Ll. Williams, E. J. Ansaldo, and R. Heffner, Phys. Rev. Lett. **65**, 2062 (1990).
[14] Shivaram, Gannon, Jr., and Hinks [Phys. Rev. Lett. **63**, 1723 (1989)] and de Visser and co-workers (Ref. 1) measured $H_{c1} \sim 150$–200 G in UPt_3 using rf field penetration and dc-magnetization techniques, respectively. Typically $B_{ext} > 5H_{c1}$ is necessary to ensure a uniform flux lattice. If their techniques give correct values for H_{c1}, then 200 G is clearly inadequate for determining $\lambda(T)$. However, thermodynamic arguments give values for H_{c1} of several Gauss. In an applied field of only 20 G we do not observe any flux expulsion. This is consistent with either strong flux pinning or a small H_{c1}.
[15] R. A. Ferrell, Phys. Rev. Lett. **3**, 262 (1959); P. W. Anderson, Phys. Rev. Lett. **3**, 325 (1959).
[16] L. Taillefer and G. G. Lonzarich, Phys. Rev. Lett. **60**, 1570 (1988).
[17] F. Gross, K. Andres, and B. S. Chandrasekhar, Physica C **162–164**, 419 (1989).