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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. We observe the appearance of a spontaneous internal magnetic field at $T = 475$ mK, which is about 80 mK below T_c. This transition likely corresponds to the lower superconducting transition observed in specific-heat and ultrasound measurements. Our results provide microscopic evidence that the low-temperature/low-field phase in the superconducting phase diagram of UPt_3 may break time-reversal symmetry.

It is widely accepted that UPt_3 is an unconventional superconductor. The splitting of the specific heat transition [1] and the observation of transitions within the superconducting state [2] have been taken as two of the most convincing illustrations of the anomalous behaviour of UPt_3.

Many of the properties of the observed superconducting phase diagram can be understood in terms of a vector superconducting order parameter with complex components. [3] In these models, the various superconducting phases are identified with different forms of the order parameter. One of these states, the low-temperature/low-field phase has been proposed to correspond to the so-called (1, i) state, which violates time reversal symmetry. To date, no direct evidence for such an identification exists. Here, we present results of muon spin relaxation experiments in which we observe the appearance of a spontaneous magnetic field within the superconducting state. Our results provide microscopic evidence that time reversal symmetry is broken in the low-temperature/low-field phase of UPt_3.

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In a μSR experiment, nearly 100% spin-polarized muons are implanted (one at a time) into a specimen. After implantation, the muon spins evolve in the local magnetic environment. The positron from muon decay ($\tau_\mu = 2.2$ μs) is emitted preferentially along the instantaneous muon pin direction. A histogram of many (10^7) such positrons therefore reflects the muon spin polarization function, which in turn reflects the magnetic environment experienced by the muon ensemble.

Several experimental geometries may be employed. In a zero-field μSR experiment, positron detectors are positioned 180° apart, normal to the initial muon polarization direction. In the absence of magnetic order, the polarization is relaxed by the nuclear dipole moments, and is well described by the so-called Kubo–Toyabe function

$$\mathcal{P}_\mu(t) = \tfrac{1}{3} + \tfrac{2}{3}(1 - \Delta^2 t^2)\exp[-\tfrac{1}{2}\Delta^2 t^2] \qquad (1)$$

where (Δ/γ_μ) is the width of the local field distribution and γ_μ is the muon gyromagnetic ratio. The presence of additional weak (relative to nuclear dipolar fields) sources of magnetic field will result in an increase in Δ.

In a transverse field experiment, the muon polarization precesses in the applied field and takes the form

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$$\mathcal{P}_\mu(t) = G_{xx}(t)\cos(\omega t) \quad (2)$$

where $G_{xx}(t)$ is a relaxation function which reflects the inhomogeneity of the magnetic field distribution and is typically approximated by a gaussian function $\exp(-\sigma^2 t^2/2)$.

In the mixed state of a type II superconductor the magnetic field penetrates a sample in the form of vortices which generally from a lattice to minimize their mutual repulsion. The flux lattice provides a source of field inhomogeneity which then manifests itself as an increase in the relaxation of the transverse field muon polarization signal. Several authors [4] have shown that the relaxation rate $\sigma \propto 1/\lambda^2$, where λ is the magnetic penetration depth. By measuring the temperature dependence of $\sigma(T)$, one may deduce $\lambda(T)$, assuming there is no additional source of relaxation.

The experiments described here were performed on two different specimens. The first sample, a polycrystal, was prepared at Grenoble, in an identical manner to the specimen used in previous specific heat experiments which exhibited a clear double superconducting transition [1]. The second sample was a single crystal grown at McMaster University. It was cut so that the basal plane was normal to the external magnetic field.

Figure 1(a) shows the transverse field ($H_{ext} = 180$ G) relaxation rate $\sigma(T)$ for the polycrystalline sample of

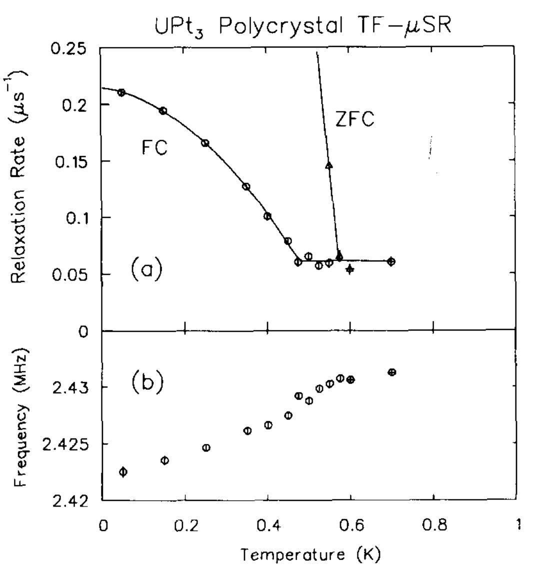

Fig. 1. Transverse-field measurements of polycrystalline UPt_3 following field cooling (circles) and zero-field cooling (triangles). The applied field was 180 G. (a) Transverse field muon relaxation rate σ. The difference between the two measurements is due to flux pinning in the superconducting state. (b) Muon precession frequency showing a diamagnetic shift below T_c.

UPt_3 following both field cooling (σ_{FC}) and zero field (σ_{ZFC}) cooling. Above 550 mK, the two cooling procedures give identical results, whereas for lower temperatures, the ZFC relaxation rate greatly exceeds that for field cooling. The enhancement is due to flux pinning which acts to oppose the formation of a uniform flux lattice in the superconducting state and allows us to estimate $T_c \approx 550$ mK. The muon precession frequency, shown in fig. 1(b) for field cooling exhibits a diamagnetic shift with an onset temperature of 550 mK, giving the same estimate for T_c.

Examining σ_{FC} in fig. 1(a), we see that there is an increase within the superconducting state, onsetting at about 475 mK, significantly below T_c. The fact that the increase in σ_{FC} does not begin until well below T_c indicates that some mechanism other than the formation of a flux lattice is likely responsible for the relaxation. Several bulk measurements [5,6] have estimated the penetration depth $\lambda \approx 19\,000$ Å. For such a value, we would expect an increase (which would be added in quadrature to the relaxation rate above T_c) of only 0.02 μs^{-1} in the transverse field relaxation rate σ. Such a small change would not be within our experimental resolution, providing further evidence that the increase in σ_{FC} is from some source other than the penetration depth.

Figure 2 shows the zero field relaxation rate Δ measured for the UPt_3 polycrystal. We see that there is an increase in Δ below roughly 500 mK. The increase in Δ is significantly greater than that resulting from the 5 K antiferromagnetic transition and is unambiguous evidence for the presence of an additional internal magnetic field of ~0.25 G at the muon site. However, the field inhomogeneity is substantially smaller than seen in the transverse field measurements.

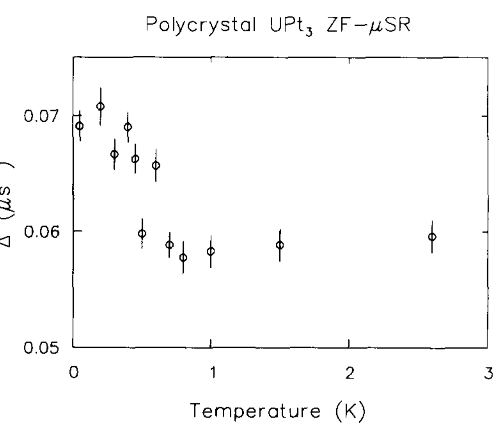

Fig. 2. Zero-field relaxation rate Δ in UPt_3, indicating the appearance of a spontaneous magnetic field within the superconducting state.

The results of similar measurements of the single-crystal specimen are shown in fig. 3. Comparing the FC/ZFC transverse field ($H_{ext} = 213$ G) relaxation rate σ_{FC}/σ_{ZFC}, indicates $T_c \sim 550$ mK for this sample. Again, we see that the increase in σ_{FC} begins about 80 mK below T_c. An increase in the zero-field relaxation rate Δ is also apparent, somewhat below 500 mK, in agreement with the results for the polycrystal.

In both samples, the change in the internal magnetic field occurs at approximately $0.85T_c$. We note that this corresponds to the temperature where the lower superconducting transition has been observed in specific heat [1] and ultrasound velocity [2] measurements. It seems likely that the magnetic change we observe is a manifestation of the transition found in bulk measurements. Similar results have been found in μSR studies of $U_{1-x}Th_xBe_{13}$ for $0.02 < x < 0.04$. [7] In those experiments, a spontaneous magnetic field appeared at the same temperature as the lower superconducting transition seen in heat capacity. While in the randomly substituted $U_{1-x}Th_xBe_{13}$ system there is always some uncertainty about the uniformity of the stoichiometry, we have demonstrated that the same effect can occur in a pure compound.

The source of the additional internal magnetic field is unclear. Blount [8] has suggested that a roughly 5% decrease of the $(1, \frac{1}{2}, 0)$ magnetic Bragg peak intensity observed in the superconducting state may be due to a reorientation of the small antiferromagnetic moments. Such a reorientation could give an increase in the field seen by the muon, since the local field is the vector sum of the fields from neighbouring spins. However, in this case, we would expect roughly similar effects in both zero- and transverse-field measurements. Another possibility is that the lower transition is to a state with broken time reversal symmetry. The additional internal field associated with the breaking of time reversal symmetry would increase the field experienced by the muons, giving an increase in the relaxation rate. In the model of Choi and Muzikar [9], a charged impurity (such as the positive muon) will be screened by a supercurrent, which produces a magnetic field at the site of the impurity. They obtained a rough estimate of 0.006 G for the internal field, which is somewhat smaller than our observed value.

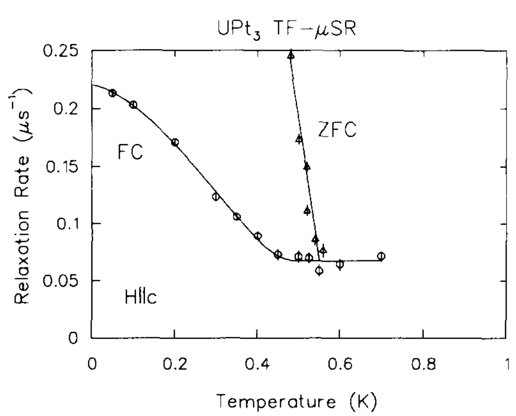

Fig. 3. Transverse field ($H_{ext} = 213$ G) relaxation in single-crystal UPt_3 following field cooling (circles) and zero-field cooling (triangles).

Previously, we had reported TF-μSR measurements [10], finding a large field dependence in the change in the relaxation rate. We found that the increase in σ_{FC} in the superconducting state decreased with increasing field. Our new results show an increase in the spontaneous internal magnetic field in the superconducting state and that the increase in the transverse field relaxation rate doesn't appear until $T = 475$ mK, significantly below T_c. In both previous μSR measurements, [11,10] σ_{FC} increases only below about 450 mK. The low temperature relaxation in these previous measurements may also reflect the effects of the same spontaneous internal magnetic field.

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