Muon Spin Relaxation in UPt_3

G. M. Luke, A. Keren, L. P. Le, W. D. Wu, and Y. J. Uemura
Department of Physics, Columbia University, 538 West 120th Street, New York, New York 10027

D. A. Bonn
Department of Physics, University of British Columbia, Vancouver, British Columbia, Canada V6T 2A3

L. Taillefer
Department of Physics, McGill University, Montreal, Canada H3A 2T8

J. D. Garrett
Department of Physics and Institute for Materials Research, McMaster University, Hamilton, Ontario, Canada L8S 4M1
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We report muon spin rotation-relaxation measurements of the heavy fermion superconductor UPt_3. The broadening of the transverse field muon precession signal sets in approximately 60 mK below T_c, a temperature which corresponds to the lower superconducting transition. In zero applied magnetic field, we observe an increase in the internal magnetic field within the superconducting state which can be explained if the "lower superconducting phase" in the H-T phase diagram of UPt_3 is characterized by broken time-reversal symmetry.

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The heavy fermion compound UPt_3 is likely the most widely studied material in its class. The heavy fermion state in this compound is characterized by a large linear specific heat coefficient $\gamma = 450$ mJ/mole K^2. At $T = 5$ K there is a transition to a weak antiferromagnetic state, as detected by both muon spin relaxation (μSR) [1] and neutron scattering [2] while at still lower temperature, UPt_3 becomes superconducting, with $T_c \approx 0.5$ K. It is now widely accepted that UPt_3 is an unconventional superconductor [3,4]. The splitting of the specific heat transition [5] and the observation of transitions within the superconducting state from ultrasound velocity [6] are taken as two of the most convincing illustrations of UPt_3's anomalous behavior. On the basis of these measurements, at least three distinct superconducting phases have been identified.

Although the precise nature of the superconducting state is not yet agreed upon, there is some emerging agreement of some of its general features [3,4]. Most of the different scenarios for explaining the existence of the various phases rest on the idea that the antiferromagnetic state (existing below $T_N = 5$ K) provides a symmetry-breaking field which couples to the superconducting order parameter, splitting the transition to an otherwise degenerate ground state. Different superconducting phases are then identified with different forms of the superconducting order parameter. This idea is supported by the recent observation that the destruction of magnetic order through the application of hydrostatic pressure [7] also causes a collapse of the two superconducting transitions into one [8].

Previously, Broholm *et al.* [9] measured the transverse field muon spin relaxation rate $\sigma(T)$ in external field $B_{\rm ext} \sim 180$ G. They interpreted their results in terms of the penetration depth, finding $\lambda(0) \sim 7000$ Å, and suggested that the anisotropic temperature dependence was evidence for a superconducting gap containing point axial nodes and a line node in the basal plane. Our subsequent measurements [10] raised questions about this interpretation, based on the field dependence of σ. Bulk measurements have provided estimates [11,12] for the penetration depth which vary over a wide range: $\lambda \approx 19\,000$ Å [11], 21 000 Å [12], and 9000 Å [13]. For the longer values of λ, we would expect virtually no detectable enhancement of the transverse field relaxation rate σ, suggesting that the observed increase in σ could be the result of some mechanism other than the superconducting screening of the external field.

It has been proposed [14] that one of the superconducting phases, the low-temperature–low-field phase, corresponds to a state which violates time-reversal symmetry, analogous to the A phase of superfluid ^{3}He. To date, no direct experimental evidence for such an identification exists. Here, we present results [15] of μSR experiments in which we observe the appearance of a spontaneous magnetic field within the superconducting state. This result supports the picture that time-reversal symmetry is broken in the low-temperature–low-field phase of superconducting UPt_3.

We have performed μSR experiments on three different specimens of UPt_3. Sample A was a polycrystal, which was prepared at Grenoble in an identical manner to several specimens which exhibit a clear double peak in heat capacity measurements. Samples B and C were cut from a large single crystal grown at McMaster University from iron free electromigrated uranium (from Ames Laboratory) and 99.99% pure platinum (from Johnson-Matthey Aesar). Sample B was oriented such that the magnetic

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field was aligned normal to the $\hat{c}$ axis, whereas the field was parallel to the $\hat{c}$ axis for sample C. Stray magnetic fields at the sample position were reduced to less than 0.05 G.

In a μSR experiment, 100% spin polarized muons are implanted one at a time into a sample. After implantation, the spins evolve in the local magnetic environment. The muon decay positron ($\tau_\mu = 2.2\ \mu$s) is emitted preferentially along the direction of the muon spin at the instant of decay. By accumulating histograms of many (10^7) such positrons, one may deduce the muon polarization function. In a transverse field experiment, the muon polarization, which is initially perpendicular to the applied field, precesses in the applied field and takes the form

$$\mathcal{P}_\mu(t) = G_{xx}(t)\cos(\omega t)\ , \qquad (1)$$

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 presence of a flux lattice results in a distribution of internal magnetic fields, where the degree of field inhomogeneity depends on the magnetic penetration depth. By measuring the inhomogeneity, we may deduce the penetration depth [16]. In UPt_3, the penetration depth is relatively long, which results in a rather slow depolarization. In this case, the Gaussian approximation for the relaxation signal is reasonable; significant differences from the more exact Redfield field distribution do not appear until times $t > 12\ \mu$s, which are beyond the range of a typical μSR measurement.

Figure 1 shows the transverse field ($H_{\text{ext}} \approx 180$ G) relaxation rate $\sigma(T)$ for each of the samples, following both field cooling (σ_{FC}) and zero field cooling (σ_{ZFC}). Above about 530 mK, the relaxation rate for the two procedures is identical; whereas for lower temperatures $\sigma_{\text{ZFC}} \gg \sigma_{\text{FC}}$. The enhancement of σ_{ZFC} is due to flux pinning in the superconducting state [17]. We use the temperature where the relaxation rates for the two procedures coincide as a conservative estimate for T_c, obtaining [18] $T_c = 545 \pm 10$, 526 ± 10, and 533 ± 10 mK for samples A, B, and C, respectively.

Examining $\sigma_{\text{FC}}(T)$ in Fig. 1, we see that the relaxation rate is small and temperature independent, from 1 K down to at least 500 mK. The fit curve for each of the three samples is a phenomenological power law of the form $\sigma_{\text{SC}} = \sigma_0[1 - (T/T')^\alpha]^\beta$ for $T < T'$, and $\sigma_{\text{SC}} = 0$ for $T > T'$. The total relaxation rate is $\sigma = \sqrt{\sigma_{\text{SC}}^2 + \sigma_{T>T'}^2}$. The condition $\beta \le 1$ ensures that σ_{SC} has negative curvature near T'. The parameter α is 0.89 for sample A, and 1.0 for samples B and C. The power β we find to be 1.91 for sample A, 2.1 for sample B, and 1.53 for sample C. In agreement with the data of

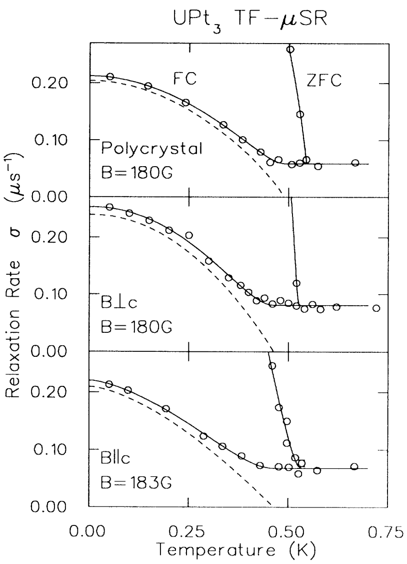

FIG. 1. Transverse field muon spin relaxation rate following field cooling (σ_{FC}) and zero field cooling (σ_{ZFC}). The solid curve is a fit to a generalized power law as described in the text. The dashed line shows the fit result for the enhanced relaxation signal. For each sample, the increase in σ_{FC} occurs at a temperature about 60 mK below the superconducting T_c, as determined by comparison of σ_{FC} and σ_{ZFC}.

Broholm *et al.* [9], we see that the slope of σ_{FC} as $T \to 0$ is greatest for $\mathbf{B} \parallel \hat{c}$, least for $\mathbf{B} \perp \hat{c}$, and intermediate for the polycrystalline sample.

If the increased relaxation is the result of field inhomogeneity associated with the flux lattice, we would expect the temperature $T' = T_c$. However, we find values of $T' = 489$, 463, and 464 mK for samples A, B, and C, all well below the measured T_c. The fact that the relaxation rate does not start to increase at T_c indicates that some mechanism other than the straightforward formation of a flux lattice is likely responsible for the relaxation. In each of the samples T' is about 60 mK below T_c, a temperature which corresponds to the lower superconducting transition, suggesting that, in fact, the increased relaxation is associated with the lower transition.

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,

$$\mathcal{P}_\mu(t) = 1/3 + 2/3\,(1 - \Delta^2 t^2)\exp\left[-\frac{1}{2}\Delta^2 t^2\right]\ , \qquad (2)$$

where Δ/γ_μ is the width of the local field distribution and γ_μ is the muon gyromagnetic ratio. If the spontaneous magnetic field is sufficiently large (and uniform) then precession will be observed. Weaker magnetism will

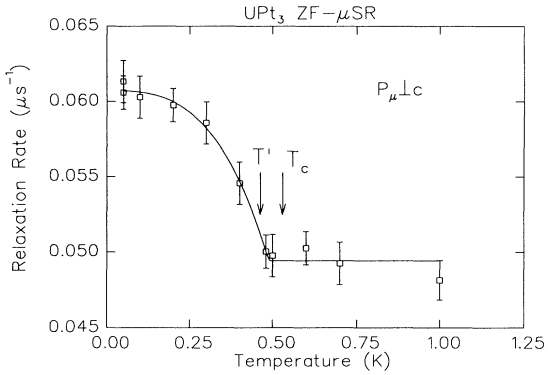

FIG. 2. Zero field muon spin relaxation rate $\Lambda(T)$ measured in sample B, showing the enhancement of relaxation in the superconducting state. The curve is the result of a fit to a generalized power law (see text). The onset temperature for the increase in relaxation is 488 ± 35 mK. The arrows indicate the values of T_c and T' (described in text) determined from transverse field measurements.

give only an increase in the observed relaxation. To characterize our zero field data, we have used the product of a temperature-independent Kubo-Toyabe function and an exponential $\exp[-\Lambda(T)t]$.

Figure 2 shows the zero field relaxation rate $\Lambda(T)$ measured in sample B. We see that there is increased relaxation occurring within the superconducting state. Roughly similar increases were seen in the other two samples. The increase in the relaxation is greater than that which occurs at the 5 K antiferromagnetic transition (but smaller than what we observe in transverse fields) and is unambiguous evidence for the presence of an additional spontaneous internal magnetic field, ~ 0.1 G, at the muon site. Fitting the temperature dependence of the relaxation rate to the same phenomenological power law as before, we find that the onset of the relaxation is $T' = 488 \pm 35$ mK, consistent with the lower superconducting transition.

There are two possible sources of additional magnetic field at the muon site. Blount *et al.* [19] have suggested that a roughly 5% decrease in the (1,1/2,0) magnetic Bragg peak intensity in the superconducting state seen in neutron scattering [2] could be due to a reorientation of the small antiferromagnetic moments. Such a reorientation could, in theory, give an increase in the field seen by the muon, since the local field is the vector sum of the fields from neighboring spins. We have calculated the internal field distribution due to the antiferromagnetic order (using a point dipole approximation for the moments). At sites where the field is less than 0.5 G (a conservative estimate for the field at the muon site below 5 K), the rotation of the moments in the basal plane would have to be on the order of 30° to give the observed change in field; with a 10° rotation the field change is less than about 0.03 G. Neutron scattering measurements at $\mathbf{Q} = (1/2, 0, 0)$ would give the most definitive answer regarding the possible existence of a spin rotation.

The second possibility is that the lower superconducting transition is to a state with broken time-reversal symmetry. The additional internal field associated with this breaking of time-reversal symmetry would increase the field experienced by the muons, giving an increase in the relaxation rate. This effect is similar to that which was envisioned for anyons, which have been searched for in high temperature superconductors [20]. In the model of Choi and Muzikar [21] for a superconducting state with broken time-reversal symmetry, a charged impurity (such as the positive muon) will be screened by a supercurrent, which produces a field at the site of the impurity. They estimated the field to be of the order of 0.006 G. More recently, Ohmi and Machida [22] calculated the internal magnetic field in the low-temperature–low-field phase. They concluded that the expected internal field should be on the order of 0.1 G, in agreement with our results, if the phase was assumed to correspond to a spin-triplet odd-parity phase, belonging to a 1D representation for the superconducting state (as suggested by Knight shift measurements [10] and references therein). In the case of the 2D representation, they found that the local field was reduced by a factor of about 100, much smaller than our experimental value.

In discussing the transverse field (TF) relaxation there are two aspects which must be addressed: first, the enhanced transverse field relaxation occurs only in the lower superconducting phase, and second, the field inhomogeneity observed in the transverse field ($H_{\rm ext} \approx 200$ G) experiment is substantially greater than in the zero field measurement. The TF relaxation rate can be due to the combined effect of the flux lattice and the spontaneous field. In general, it is not possible to separate the different sources of TF relaxation (especially when the effect of each may be field dependent). However, we may identify two possible explanations. Since they are not mutually exclusive, a combination of the two is also possible.

If the relaxation is due to the presence of a vortex lattice, it is only present in the lower superconducting phase. This implies that there must be a change in the superconducting state at T' which results in a reduction of the penetration depth, and therefore increases the TF relaxation rate (since $\sigma \propto \lambda^{-2} \propto n_s/m^*$), either an increase in n_s or a decrease in m^*. In the only other microscopic measurement which has been able to differentiate between the different superconducting states, Goll *et al.* [23] detected an energy gap using point-contact spectroscopy, but only below the lower transition, indicating that the electronic properties of the individual superconducting states differ. A change in the nature of the vortex lattice itself at T' could also give an increase in the relaxation rate. Several authors have shown that in a time-reversal breaking state, the vortex lattice can take forms different from that expected for a conventional superconductor. In this case, one could expect a different proportionality between the field inhomogeneity and the magnetic penetration depth. For example, doubly quantized vortices would give twice the relaxation rate for the same penetration depth. The complex vortex structures found by Tokuyasu *et al.* [24], and Joynt [25] would also have a markedly different field inhomogeneity than for a singly quantized one-component order parameter.

If the transverse field relaxation reflects the spontaneous field, then this implies that the application of an external field causes an enhancement of the spontaneous magnetic field. The field inhomogeneity would then be greater in the transverse field measurements. The field dependence of the spontaneous magnetic field predicted in the various theoretical models [21,22] has not been calculated, which makes it difficult to assess the likelihood of this possibility.

In transverse fields, the difference in the relaxation rate between the superconducting and normal states $[\sqrt{\sigma_{\rm TF}^2(T=0)-\sigma_{\rm TF}^2(T>T_c)}]$ decreases with increasing applied field (for $H_{\rm ext} > 200$ G) [10]. The field dependence in $\sigma_{\rm SC}(T \to 0)$ is consistent with that predicted theoretically [15] for the effect of a cutoff field in the vortex cores and the variation of the sample volume occupied by the vortices and seen in the neutron scattering experiment of Kleimen *et al.* [26]. However, it is also possible that the same field dependence could be present in the other scenarios described above; for example, as the volume fraction occupied by normal cores increases, the average spontaneous field (which would be present only in the regions not occupied by the cores) would decrease. Therefore, the field dependence of the relaxation rate above $H_{\rm ext} > 200$ G does not allow us to distinguish effects of the spontaneous field from the vortex lattice.

The appearance of a spontaneous magnetic field below 0.5 K is consistent with the picture that time-reversal symmetry is broken in at least the low-temperature–low-field superconducting phase of UPt_3. Similar results have been found in μSR studies of $U_{1-x}Th_xBe_{13}$ for $0.02 < x < 0.04$ [1,27]. In this case, the observed magnetism could indicate either the existence of a time-reversal symmetry breaking superconducting state, or a purely magnetic transition, possibly associated with the thorium impurities. In the present work, we have shown that a similar phenomenon can occur in a pure heavy fermion compound without impurities.

Previous μSR measurements [9] of the transverse field relaxation rate were interpreted in terms of the penetration depth and a specific gap structure. The present results indicate a possibility that the transverse field relaxation rate may reflect effects other than the penetration depth. Hopefully, as more microscopic information is discovered, we will obtain a complete understanding of the superconducting states of UPt_3.

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