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MUON SPIN RELAXATION AND KNIGHT SHIFT IN THE HEAVY-FERMION SUPERCONDUCTOR UPt_3

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Positive muon spin relaxation experiments have been conducted on the heavy-fermion superconductor UPt_3 in both the normal and superconducting states for zero, transverse, and longitudinally applied magnetic fields. Below 6 K in zero applied field, the μ^+ relaxation rate is approximately twice that expected from ^{195}Pt nuclear dipolar relaxation alone. Transverse- and longitudinal-field measurements show that the observed relaxation rate depends on magnetic field and is quasistatic in origin. It is suggested that the onset of very weak ($\sim 10^{-3}$ μ_B/U atom) magnetic ordering below approximately 6 K is responsible for the observed increase in the relaxation rate. μ^+ Knight shift measurements in the normal state of UPt_3 show a temperature dependent shift K_μ which tracks the bulk susceptibility χ. From the K_μ vs. χ plot, a μ^+ hyperfine field of approximately 100 Oe/μ_B is extracted.

1. INTRODUCTION

Following the discovery of heavy-fermion superconductivity (HFS) in $CeCu_2Si_2$ /1/ and UBe_{13} /2/, a third HFS system, UPt_3, was reported /3/. In addition to the usual properties /4/ associated with these materials, UPt_3 exhibited spin fluctuation behavior similar to that previously reported for $TiBe_2$ /5/ and UAl_2 /6/. Thus it was suggested that UPt_3 might represent the first known system to exhibit coexistent bulk superconductivity and spin fluctuations. Moreover, it was proposed that the electron pairing responsible for superconductivity in UPt_3 was not the usual BCS type but rather an unusual form, possibly odd-parity pairing. Further evidence supporting this suggestion has been recently inferred from ultrasound measurements /7/ and critical field studies /8/.

Despite extensive theoretical and experimental efforts to understand the low-temperature properties of HFS compounds in general, and UPt_3 in particular, much remains to be learned. For example, no NMR signals have been detected in UPt_3, due presumably to a very large broadening mechanism. Consequently, we have undertaken positive muon (μ^+) spin relaxation (μSR) studies in UPt_3 to investigate the behavior of local magnetic fields.

2. EXPERIMENTAL

Zero-field μSR data were taken in the interval 0.165-20 K, with some additional points taken in either longitudinal or transverse fields, using a conventional μSR spectrometer. For the zero-field measurements the magnetic field at the sample was nulled to ±10 mOe. High transverse field (5 kOe) data were taken in the interval 2.3-300 K, from which the μ^+ relaxation rate and Knight shift were determined. The stability of the applied field was monitored by an NMR probe. Measured μ^+ Knight shifts K_μ were corrected for a copper reference shift of 60 ppm /9/. Temperatures below 2.3 K were attained with a ^3He-^4He dilution refrigerator /10/, whereas a continuous-flow cryostat was used above 2.3 K. An arc-melted polycrystalline ingot (19 mm diameter, 3 mm thick) with light interstitial impurity (C,N,O) concentrations of 100-150 ppm at. was used in this study. From ac susceptibility data the superconducting transition was found to occur at 0.41 ±0.01 K.

3. NORMAL STATE RELAXATION (T > 0.5 K)

Figure 1 shows the μ^+ Gaussian relaxation rate for UPt_3 in zero and various applied magnetic fields. The calculated Van Vleck linewidth σ_{VV} /11/ for random, static ^{195}Pt ($I = 1/2$) nuclear dipoles is indicated by the cross-hatched area in Fig. 1(b). The value of σ_{VV} ranges from 0.031 μs^{-1} to 0.042 μs^{-1}, depending upon which interstitial site is assumed for the μ^+ occupation. For purely dipolar fields, the zero-field linewidth is known /12/ to be larger than σ_{VV} by a factor $\sqrt{5}$, thus the range of values shown in Fig. 1(a) is from 0.070 μs^{-1} to 0.094 μs^{-1}.

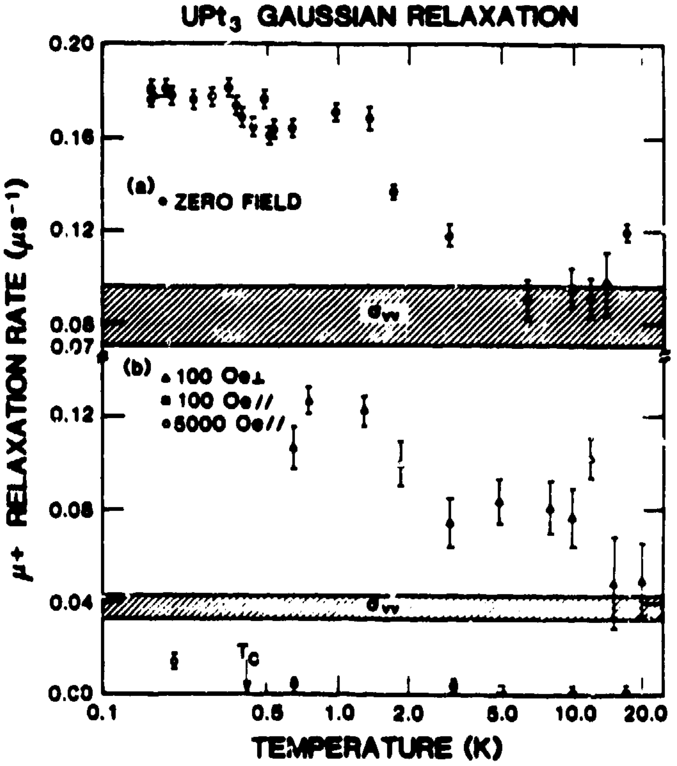

Fig. 1. μ^+ Gaussian relaxation rates in polycrystalline UPt_3. The cross-hatched areas correspond to the calculated Van Vleck lindwidths, σ_{VV}, with the limits being μ^+ site dependent.

For 6 K < T < 15 K, the zero-field relaxation rate, σ_{ZF}, [Fig. 1(a)] is consistent with σ_{VV}; however, for T < 6 K, σ_{ZF} increases by approximately a factor of two, attaining a value of 0.165 μs^{-1} near 1.4 K. In a low (100 Oe) transverse applied field, the μ^+ relaxation rate σ_{LF} is consistent with σ_{VV} for 15 K < T < 20 K [Fig. 1(b)]. As the temperature decreases, there is a 50% increase in σ_{LF} between 15 and 10 K; an additional 50% increase is observed between 3 and 1 K, where σ_{LF} reaches an approximate value of 0.12 μs^{-1}. Below ≈2 K, $\sigma_{ZF}/\sigma_{LF} = 1.45 \pm 0.15$.

High-field (5 kOe) transverse μSR relaxation rates σ_{HF} are shown in Fig. 2 for the interval 2.3 K < T < 300 K. Over comparable temperature intervals, σ_{HF} is 2-4 times as large as σ_{LF}, indicating field-dependent relaxation. There exists a plateau in σ_{HF} near 30 K, followed by a marked increase as T is reduced from 15 K to 2.3 K.

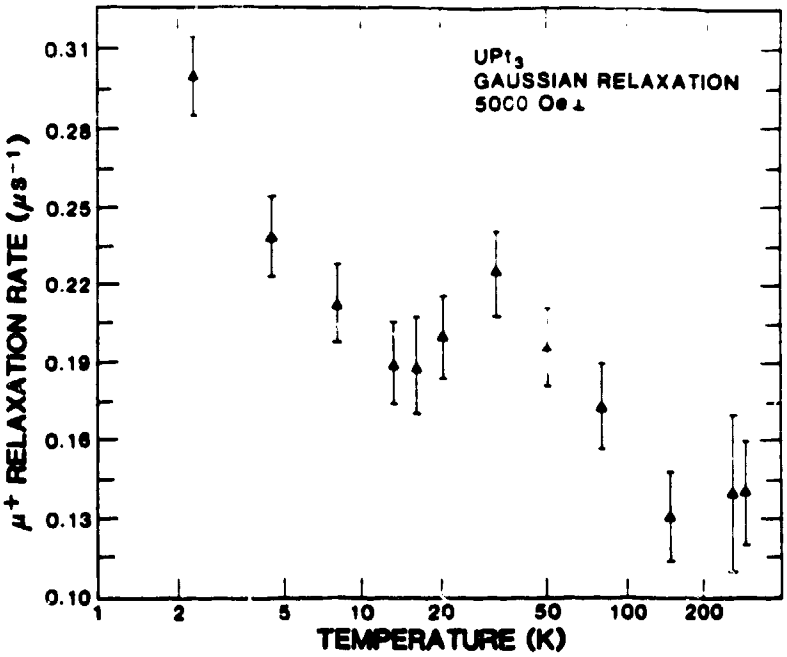

Fig. 2. μ^+ Gaussian relaxation rates in polycrystalline UPt$_3$ taken in a 5 kOe transverse field as a function of temperature.

All of the μ^+ relaxation rates in longitudinally applied fields are essentially zero below about 17 K [see Fig. 1(b)]. These results are consistent with a quasistatic relaxation mechanism in transverse and zero fields. This means that the temperature dependence σ_{ZF} is due neither to μ^+ hopping nor to motional narrowing of the μ^+ local field by relaxation of ^{195}Pt nuclei.

The principal experimental result presented here is that one observes a larger-than-expected, field-dependent, quasistatic μ^+ relaxation rate for $T < 15$ K in UPt$_3$. To explain this finding one must postulate the existence of an increased μ^+ local field distribution, of which three hypothetical mechanisms are discussed below. First, precipitations of a spurious magnetic second phase, e.g., UPt /13/, may have contaminated our sample and might give rise to a macroscopic magnetic field inhomogeneity. Second, the increased μ^+ linewidth may be caused by a strong indirect coupling between μ^+ and ^{195}Pt moments, mediated by band electrons. Third, the band (heavy) electrons themselves might possess a weak form of magnetic order, which could result in a μ^+ local field distribution via the μ^+ hyperfine interaction. We discuss these possibilities in turn.

3.1. Magnetic Second Phase

To check for the possible existence of magnetic phases, magnetization measurements were conducted on a portion of the sample used for μSR. No remanent moment was observed, and we can place an upper limit of $\approx 2 \times 10^{-3}$ emu/gm on the remanent magnetization. If UPt (T_c = 27 K) is assumed to be the magnetic second phase, its mole fraction must therefore be less than 0.064%.

The Walstedt-Walker (WW) mechanism /14/ for inhomogeneous line broadening by a static distribution of paramagnetic impurities is also applicable to broadening by dipolar fields of inclusions of a magnetic second phase, as long as the inclusions are well dispersed and small. The Lorentzian linewidth λ in the WW theory is given by

$$\lambda = 5.065\ \gamma_\mu m_{eff} \quad , \qquad (1)$$

where γ_μ is the μ^+ gyromagnetic ratio ($8.51 \times 10^4\ G^{-1}s^{-1}$), and m_{eff} is the magnetization due to the second phase. If a UPt concentration of 0.064% is assumed, then a WW calculation yields an upper limit of 0.016 μs^{-1} for the μ^+ linewidth. A Lorentzian fitted to the data below 2 K (not shown in Fig. 1) yields 0.057 μs^{-1}, which is 3.5 times larger than the calculated width. We conclude, therefore, that the observed upper limit on the remanent magnetization implies a small contribution to the observed μ^+ linewidth from magnetic inclusions.

3.2. Indirect Interaction

We now consider consequences of a strong electron-mediated interaction between μ^+ and ^{195}Pt moments /15/. The order of magnitude of the μ^+ linewidth σ_{ind} due to this mechanism is

$$\sigma_{ind} \approx A_\mu(UPt_3)\, A_{Pt}(UPt_3)\, \rho(UPt_3) \quad , \tag{2}$$

where $A_\mu(UPt_3)$ and $A_{Pt}(UPt_3)$ are the hyperfine (hf) coupling constants between band electrons and μ^+ and ^{195}Pt spins, respectively, and $\rho(UPt_3)$ is the density of band states at the Fermi energy. A value $\sigma_{ind} \approx 10^5 s^{-1}$ is required to explain the observed low-temperature μ^+ linewidth in UPt_3 [Fig. 1(a)]. This is coincidentally the approximate value of the indirect (RKKY) interaction $J_{Pt} \approx [A_{Pt}(Pt)]^2\, \rho(Pt)$ between ^{195}Pt spins in platinum metal /16/, where $A_{Pt}(Pt)$ is the hf coupling constant and $\rho(Pt)$ is the density of states.

Available data can be used to estimate the value of σ_{ind}. As we have noted above,

$$\frac{\sigma_{ind}}{J_{Pt}} \approx \frac{A_\mu(UPt_3)}{A_{Pt}(Pt)} \times \frac{A_{Pt}(UPt_3)}{A_{Pt}(Pt)} \times \frac{\rho(UPt_3)}{\rho(Pt)} \quad . \tag{3}$$

Now the ratio $A_\mu(UPt_3)/A_{Pt}(Pt) \approx 8 \times 10^{-5}$ can be estimated from our μ^+ Knight shift measurements and NMR Knight shift data from Pt metal /17/. Similarly, the ratio $\rho(UPt_3)/\rho(Pt) = 70$ is obtained from measured specific-heat coefficients C/T in the two systems /18/. This yields

$$\sigma_{ind} \approx 5.6 \times 10^{-3} \frac{A_{Pt}(UPt_3)}{A_{Pt}(Pt)} J_{Pt} \quad . \tag{4}$$

In order that $\sigma_{ind}/J_{Pt} \approx 1$ the Pt hf coupling constant would have to be more than 175 times larger in UPt_3 than in Pt metal. An experimental value for $A_{Pt}(UPt_3)$ is unfortunately not available, as ^{195}Pt NMR has not yet been observed in UPt_3.

Nevertheless some evidence points toward a reduction, often considerable, of the hf coupling upon passing from a pure nonmagnetic metal to the same metal incorporated in an f-atom compound. For example, in $CePt_6$ the transferred hf coupling is some 30 times weaker than the d-band core polarization interaction in pure Pt /19/. Thus there is perhaps a discrepancy of as much as four orders of magnitude between the estimated value of σ_{ind} and the experimental μ^+ linewidth. This discrepancy seems difficult to make up with inaccuracies of the above estimates, such as unknown details of the indirect interaction, for example. We conclude that the contribution of the observed μ^+ linewidth due to indirect interactions with the surrounding Pt nuclei is in all probability negligible.

3.3. Heavy-Electron Magnetism

Finally, we examine consequences of an assumed quasistatic magnetic order in the band electron system. We first note that, in general, μ^+ hyperfine fields are of the order of 10^3 Oe/μ_B, via either indirect Fermi contact or direct dipolar interactions. Thus the observed increase in linewidth σ/γ_μ (~1 Oe in field units) occurring below 6 K implies an order-of-magnitude moment per U atom of only 10^{-3} μ_B. It is important to note that recent specific heat and susceptibility measurements /20/ on UPt_3 alloyed with Pd, exhibit low-temperature magnetic anomalies. For example, in U(Pt[illegible]Pd[illegible])$_3$ there is a peak at 3.6 K in the specific heat data which the authors suggest may be due to an antiferromagnetic type of ordering. Similar studies /21/ on Pd- and Th-doped UPt_3 report an anomaly at 6 K in the magnetic susceptibility and specific heat data which, based upon field dependence, appears to be due to antiferromagnetism. Other studies /22/ on Th-doped UPt_3 reveal a Fermi-surface instability occurring at 6.5 K. We believe that our μSR results are consistent with these findings, the difference being that μSR is more sensitive to weak magnetic ordering thereby allowing us to observe such an effect in "pure" UPt_3 in zero field. The strong field dependence of the μ^+ relaxation rate further supports the notion of some kind of quasistatic magnetic state which sets in below about 10 K.

4. SUPERCONDUCTING STATE RELAXATION ($T < 0.5$ K)

The zero-field relaxation rate σ_{ZF} increases by approximately 10% just below T_c, as shown in Fig. 1(a). A change of this magnitude corresponds to an increased μ^+ local field distribution of approximately 175 mOe. This cannot be explained by external field inhomogeneities because the field was nulled to ±10 mOe. Similar experiments by us on $Pb_{0.9}In_{0.1}$ (BCS Type II Superconductor) and UBe_{13} (HFS) exhibit no change in μ^+ relaxation rate upon entering the superconducting state. The most likely explanation for the UPt_3 data is that the local field distribution produced by the weak magnetism discussed above is changed slightly upon entering the Meissner state.

5. μ^+ KNIGHT SHIFT

Figure 3 shows both the normal state μ^+ Knight shift K_μ and bulk magnetic susceptibility χ measured as a function of temperature. K_μ is approximately zero for $T \lesssim 20$ K, but decreases to $\approx 0.030\%$ near room temperature. A plot of K_μ vs. χ yields a slope of 0.086 mol emu^{-1}; however, the Lorentz and demagnetization fields give rise to a linear K_μ vs. χ relation with a slope of 0.068 mol emu^{-1}, i.e., 80% of the observed value. The macroscopic corrections therefore account for the majority of the observed slope. This places an upper limit on the hyperfine field of 100 (223) Oe/μ_B, two orders-of-magnitude smaller than obtained in UBe_{13} /23/. Such a small value might be due to the presence of two nearly equal hyperfine couplings of opposite sign.

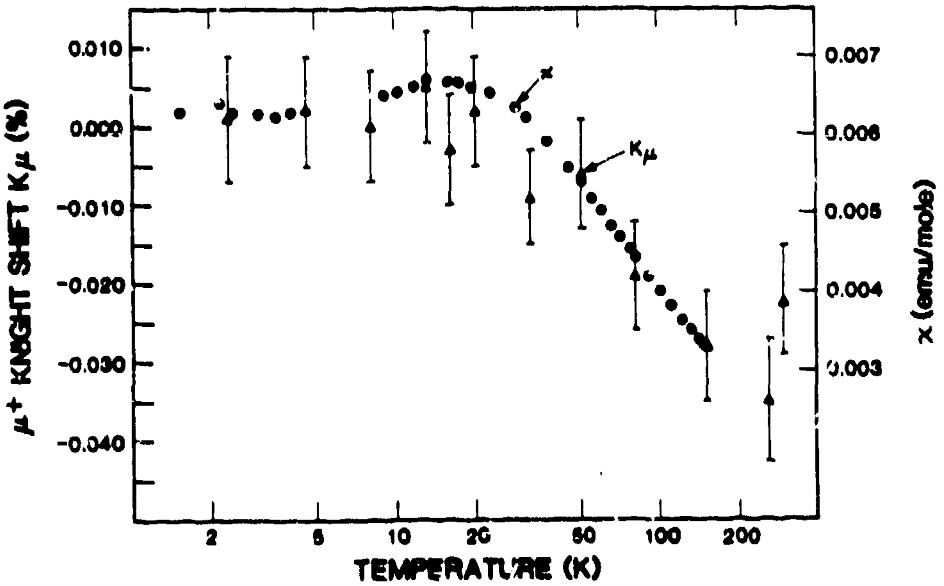

Fig. 3. μ^+ Knight shift and susceptibility of UPt_3.

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