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Spin fluctuations and anisotropic nuclear relaxation in single-crystal UPt_3

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The anisotropy and temperature dependence of the ^{195}Pt spin-lattice relaxation rate T_1^{-1} have been measured in a single crystal of the heavy-fermion compound UPt_3. Between 1.5 and 4.2 K T_1^{-1} is proportional to temperature. The observed anisotropy $T_{1\parallel}^{-1}/T_{1\perp}^{-1}=1.7\pm0.1$, together with the nearly isotropic ^{195}Pt hyperfine field, imply that the spin fluctuations are predominantly in the basal plane. The Korringa product is close to the value expected for a noninteracting Fermi gas, which indicates that the relaxation is not enhanced by antiferromagnetic (AFM) fluctuations. The effective fluctuation rates derived from these NMR data (9 ± 1 meV) and from inelastic neutron scattering by AFM fluctuations are similar, indicating that a rate of this order characterizes a range of fluctuation wave vectors.

The importance of spin fluctuations in the behavior of heavy-fermion materials has been understood for some time. Spin fluctuations in the heavy-fermion superconductor UPt_3 have been extensively studied,[1] most directly by neutron scattering[2] where a rich variety of behavior has been found. Inelastic neutron scattering for relatively high-energy transfers $\hbar\omega\gtrsim1$ meV revealed short-range spin correlations of both ferromagnetic (FM) and antiferromagnetic (AFM) character,[3] the latter exhibiting a characteristic fluctuation rate ~10 meV in energy units and setting in at ~20 K. Below this temperature FM correlations are found between spins at uranium sites in the hexagonal basal plane, with AFM correlations between planes. These fluctuations were found to account for the normal-state bulk static susceptibility to within $\sim20\%$.

Following initial indications of an AFM transition below a Néel temperature $T_N\sim5$ K in muon-spin rotation studies,[4] further low-energy ($\hbar\omega\lesssim0.6$ meV) and Bragg neutron-scattering experiments[5] in UPt_3 showed AFM fluctuations at a rate ~1 meV and confirmed the AFM state. In this state the magnetic unit cell is doubled in the basal plane by very weak [ordered moment $(0.02\pm0.01)\mu_B$/U atom] AFM order. Recent neutron-scattering measurements in the superconducting state of UPt_3 revealed intricate correlations between the AFM and superconducting order parameters,[6] which give strong support to an unconventional mechanism for superconductivity in UPt_3.

In heavy-fermion compounds nuclear magnetic resonance (NMR) measurements also probe properties of the heavy electrons.[7] Nuclear spin-lattice relaxation, in particular, is dominated by thermal excitations of the same $4f$-derived heavy-fermion spins which can be probed by neutron scattering. In the normal state of heavy-fermion materials (as in ordinary metals) the nuclear spin-lattice relaxation rate T_1^{-1} is proportional to temperature in the Fermi-liquid region,[7] i.e., below a characteristic temperature T_0. Above T_0, $T_1^{-1}(T)$ varies less rapidly as the correlated heavy-fermion state is destroyed.

This article describes measurements of the ^{195}Pt spin-lattice relaxation rate in a single-crystal sample of UPt_3. Low-temperature ^{195}Pt NMR in powder samples of UPt_3 has been reported previously.[8-10] Above the superconducting transition at ~0.5 K very large and anisotropic ^{195}Pt Knight shifts were found,[8-10] corresponding to the anisotropy of the bulk susceptibility.[1] The relaxation was observed to be proportional to temperature,[8] although there was some indication[9] of a downturn in $T_1^{-1}(T)$ at ~10 K. An analogous anisotropy in the spin-fluctuation amplitude would therefore be expected to affect T_1^{-1}. Very recently Kohori *et al.*[10] have reported ^{195}Pt relaxation measurements in field-aligned powder samples of $U(Pt_{1-x}Pd_x)_3$, $x=0$, 0.01, and 0.05. The present data complement ^{195}Pt Knight-shift measurements carried out in the same single crystal by Koster, Williams, and Bucher[11] using a cw NMR technique. Where comparable the cw and pulsed NMR spectra are in reasonable agreement, as are the single-crystal NMR data and previously published results.[8-10]

Nuclear spin-lattice relaxation is caused by thermal fluctuations of the heavy-fermion spins, coupled to the nuclei via a transferred hyperfine interaction. These fluctuations are related by the fluctuation-dissipation theorem to the dissipative component $\chi_s''(\mathbf{q},\omega)$ of the corresponding dynamic spin susceptibility. Furthermore, in heavy-fermion systems the strong f-shell spin-orbit coupling renders the spin and total susceptibilities proportional to each other: $\chi_s''(\mathbf{q},\omega)\propto\chi''(\mathbf{q},\omega)$.

Then T_1^{-1} is proportional to a weighted sum over wave vector $\mathbf{q}$ of $k_BT\chi''(\mathbf{q},\omega)/\hbar\omega$, evaluated at the nuclear frequency $\omega=\omega_n$. The weighting factor is the so-called

hyperfine form factor $|A(\mathbf{q})|^2$, which is the squared magnitude of the Fourier transform of the hyperfine coupling $A(\mathbf{r})$ between nuclei and heavy-electron spins. The relaxation rates $T_{1\parallel}^{-1}$ and $T_{1\perp}^{-1}$ for applied field $\mathbf{H}_0$ parallel and perpendicular to the crystalline c axis, respectively, are[12]

$$T_{1\parallel}^{-1} = 2(T/\omega_n)\sum_{\mathbf{q}} |A_\perp(\mathbf{q})|^2 \chi_\perp''(\mathbf{q},\omega_n) \quad (1)$$

and

$$T_{1\perp}^{-1} = (T/\omega_n)\sum_{\mathbf{q}} [|A_\perp(\mathbf{q})|^2 \chi_\perp''(\mathbf{q},\omega_n) + |A_\parallel(\mathbf{q})|^2 \chi_\parallel''(\mathbf{q},\omega_n)] , \quad (2)$$

where constants have been absorbed into the form factors. These relations express the fact that only components of the fluctuating hyperfine field perpendicular to $\mathbf{H}_0$ are effective in relaxation. The factor of 2 in Eq. (1) accounts for the two basal-plane components.

The single-crystal sample was a disk approximately 5 mm in diameter and 1 mm thick. It was cut from an ingot used in previous neutron-scattering[5,6] and specific-heat[13] experiments, and was also used in a previous muon-spin rotation study of the superconducting state.[14] Its low residual resistivity[13] is indicative of good sample quality.

Nuclear relaxation measurements in single-crystal metallic specimens are difficult for several reasons. The signal arises only from the skin depth of the sample and is therefore weak; the rf pulses which excite the NMR cause appreciable eddy-current heating of the sample; and magnetoacoustic ringing of the sample, due to excitation of rf acoustic modes in the static magnetic field, generates a large spurious signal. In our experiments this signal lasted a time comparable to T_1, which was always less than 700 μsec. It was dealt with by subtracting the signal obtained for applied fields on either side of the ^{195}Pt resonance from the on-resonance signal. The magnetoacoustic ringing, which does not vary strongly with field, was largely canceled out by this procedure.

A sequence of two rf pulses, each of tipping angle θ and separated by a variable delay time, was used to measure the recovery of the longitudinal nuclear magnetization, which is proportional to the difference between the free-induction signals after the two pulses. As usual in pulsed NMR $\theta = \gamma_n H_1 t_w$, where γ_n is the nuclear gyromagnetic ratio, H_1 is the magnitude of the pulsed rf magnetic field, and t_w is the rf pulse width. The total eddy-current heat Q_p per pulse then varies as $H_1^2 t_w \propto \theta^2/t_w$. A signal induced by the rf pulse in a small link placed near the NMR coil provided an uncalibrated measure of H_1, so that the effect of Q_p on the relaxation-rate measurement could be determined.

Pulse heating was found to be prohibitive unless the sample was immersed in liquid helium, which accounts for the limited temperature range (1.5–4.2 K) of the experiments reported here. Even with immersion, pulse heating produced a significant effect on the measured relaxation, but when the above sequence was used the observed relaxation rate could be reliably extrapolated to $Q_p = 0$ as shown in Fig. 1. To ensure that the initial pulse perturbed the resonance uniformly t_w was kept much shorter than the free-induction decay time. This constraint together with the need for small pulse heating limited θ to values much less than $\pi/2$, which rendered the already weak

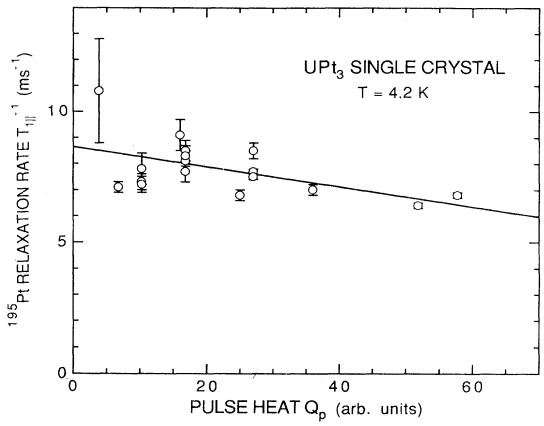

FIG. 1. Dependence of observed ^{195}Pt spin-lattice relaxation rate $T_{1\parallel}^{-1}$ (applied field $\mathbf{H}_0$ oriented parallel to the crystal c axis) on eddy-current pulse heat Q_p in single-crystal UPt_3. The sample was immersed in liquid helium ($T = 4.2$ K). The straight line is the best fit to the data.

NMR signal even weaker and led to long signal-averaging times.

Figure 2 gives the temperature dependences of the ^{195}Pt relaxation rates $T_{1\parallel}^{-1}$ and $T_{1\perp}^{-1}$. Linear fits to both sets of data yield zero intercepts within error, and give values of the T-linear coefficients $T_{1\parallel}^{-1}/T = 1.81 \pm 0.07$ ms^{-1}K^{-1} and $T_{1\perp}^{-1}/T = 1.05 \pm 0.08$ ms^{-1}K^{-1}. We therefore obtain $T_{1\parallel}^{-1}/T_{1\perp}^{-1} = 1.72 \pm 0.15$.

The transferred hyperfine coupling to the heavy-electron susceptibility has been obtained in the usual way from the slope of a plot of K vs χ with temperature an implicit parameter[15] from the data of Kohori *et al.*[9,10] and Koster, Williams, and Bucher.[11] Relatively little anisotropy (~15%) is found. This indicates that the fluctuations themselves are anisotropic and, in fact, lie principally in

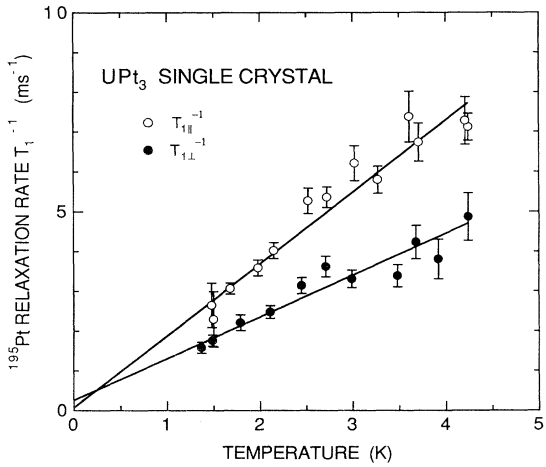

FIG. 2. Temperature dependence of ^{195}Pt spin-lattice relaxation rates $T_{1\parallel}^{-1}$ and $T_{1\perp}^{-1}$ in single-crystal UPt_3, measured with applied field $\mathbf{H}_0$ oriented parallel and perpendicular, respectively, to the crystal c axis. The straight lines were fitted to the data with both slopes and intercepts varied for best fit; the intercepts vanish to within error.

the basal plane. For example, if the hyperfine interaction were perfectly isotropic and the fluctuating spins (and therefore the fluctuating hyperfine field) lay completely in the basal plane, one would obtain $T_{1\parallel}^{-1}/T_{1\perp}^{-1}=2$ from Eqs. (1) and (2).

We next consider the Korringa product $\mathcal{S}=K^2T_1T$. For a noninteracting Fermi gas

$$\mathcal{S}=\mathcal{S}_0\equiv(\hbar/4\pi k_B)(\gamma_e^2/\gamma_n^2)\,,$$

where γ_e is the gyromagnetic ratio of the electron.[15] Thus $\mathcal{S}_0$ is independent of material parameters. The ratio $\mathcal{S}/\mathcal{S}_0$ is a measure of the effect of interactions on both the static susceptibility and the fluctuations which give rise to T_1^{-1}, but is also affected by the nature of the transferred hyperfine coupling [cf. Eqs. (1) and (2)].

In the present situation both the static and dynamic susceptibilities are markedly anisotropic, and each component of the generalized dynamic susceptibility must be independently characterized. Equation (1) therefore motivates the definition of a transverse Korringa product $\mathcal{S}_\perp=K_\perp^2T_{1\parallel}T$. Using the Knight-shift data of Koster, Williams, and Bucher,[11] we find $\mathcal{S}_\perp=(1.18\pm0.10)\mathcal{S}_0$. Inspection of Eqs. (1) and (2) leads to the corresponding definition of a longitudinal Korringa product

$$\mathcal{S}_\parallel=K_\parallel^2/(2T_{1\perp}^{-1}/T-T_{1\parallel}^{-1}/T)\,.$$

Again using the data of Koster, Williams, and Bucher, we find $\mathcal{S}_\parallel=(1.2\pm0.4)\mathcal{S}_0$. Thus there is no marked enhancement or reduction of the Korringa product for either orientation.

This result is somewhat surprising, given the evidence for the importance of antiferromagnetic spin fluctuations in UPt_3. For example, in the heavy-fermion superconductor UBe_{13} the isotropic Korringa product is reduced by the AFM enhancement of T_1^{-1}: $\mathcal{S}\simeq0.3\mathcal{S}_0$ (Ref. 16). The effect of such fluctuations on relaxation of nearby nuclei is suppressed, however, when the form factor $|A(\mathbf{q})|^2$ vanishes for $\mathbf{q}$ equal to the wave vector $\mathbf{Q}_{AF}$ of the AFM spin fluctuations. In a real-space description this vanishing is due to the fact that for such a form factor spins on the two AFM sublattices are equidistant from nuclear sites, leading to cancellation of hyperfine fields from correlated fluctuations of oppositely oriented spins on the two sublattices. For example, the lack of enhancement of ^{17}O relaxation at Y and planar O sites in the normal state of the high-temperature superconductor $YBa_2Cu_3O_7$ has been attributed[17] to such a vanishing of $|A(\mathbf{Q}_{AF})|^2$.

In UPt_3 the crystallographic situation is not this simple, but Pt sites are coordinated at nearly the same distance by two U sites in the same plane and two U sites in adjacent planes (one above and one below the plane of the Pt site).[1] It seems likely that this near-neighbor configuration, together with a nearly isotropic transferred hyperfine interaction, suppresses enhancement of the ^{195}Pt relaxation rate by the "high-energy" (10 meV) AFM correlations *between* planes.

There is no such approximate cancellation [i.e., $|A(\mathbf{Q}_{AF})|^2\neq0$ for the appropriate value of $\mathbf{Q}_{AF}$] of the "low-energy" (1 meV) fluctuations associated with the AFM transition at 5 K.[3] If these fluctuations were important in ^{195}Pt relaxation the Korringa product would again be reduced, contrary to experiment. We conclude that for different reasons the ^{195}Pt relaxation is affected by neither the high-energy nor the low-energy AFM fluctuations. The latter result is not obvious *a priori*: although the high-energy AFM fluctuations dominate the fluctuation spectrum,[2] weaker motional narrowing of these lower-frequency AFM fluctuations might have allowed them to play a role in nuclear relaxation.

In the absence of strong correlations the Korringa product $\mathcal{S}_i$ for each orientation i, together with the bulk susceptibility χ_i for that orientation, yield an estimate[18]

$$\Gamma_i^{(\mathrm{eff})}=2k_B\gamma_n^2\mathcal{S}_i/\chi_i \qquad (3)$$

for the effective fluctuation rate $\Gamma_i^{(\mathrm{eff})}$ of fluctuations in direction i. If the fluctuation spectrum is Lorentzian this estimate is equal to the half-width, and in general $\hbar\Gamma_i^{(\mathrm{eff})}$ gives the energy scale for the fluctuations. In the present case Eq. (3), together with $\mathcal{S}_\perp$ and the low-temperature bulk susceptibility $\chi_\perp$ (Ref. 1), yields $\hbar\Gamma_\perp^{(\mathrm{eff})}=9\pm1$ meV. This is within a factor of 2 of the value obtained from neutron scattering for the high-energy AFM fluctuations.[2] Since the latter fluctuations appear to be ineffective in ^{195}Pt relaxation, we conclude that *the same energy scale appears to characterize the fluctuation spectrum over a wide range of values of* $\mathbf{q}$. For the longitudinal fluctuations the rate is 3 to 4 times higher, but is still 100–1000 times slower than conduction-electron fluctuation rates characteristic of ordinary metals.

The ^{195}Pt relaxation rate is observed to be rapidly depressed by the onset of superconductivity.[9,19] It therefore appears that the *entire* fluctuation spectrum, and not only that part associated with the AFM transition, is suppressed by the opening of the superconducting energy gap below T_c.

In conclusion, we have determined the anisotropy in the ^{195}Pt relaxation rate in UPt_3 between 1.5 and 4.2 K. The observed anisotropy indicates that the fluctuations lie principally in the basal plane. The absence of AFM reduction of the transverse Korringa product is inconsistent with relaxation by AFM fluctuations, which in the case of high-energy AFM fluctuations is attributed to a small form factor for the ^{195}Pt hyperfine coupling. The fluctuations effective in nuclear relaxation have a characteristic fluctuation rate ~10 meV, like the dominant AFM fluctuations observed in neutron scattering, but are not antiferromagnetically correlated. Such a rate therefore characterizes the dissipative susceptibility over a wide range of wave vectors. It is remarkable that this entire complex spectrum is affected by the onset of superconductivity, as indicated by recent neutron-scattering results[6] and ^{195}Pt relaxation in the superconducting state.[9,19]

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