Magnetic transition and spin fluctuations in the unconventional antiferromagnetic compound Yb$_3$Pt$_4$

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Abstract
Muon spin rotation and relaxation measurements have been carried out on the unconventional antiferromagnet Yb$_3$Pt$_4$. Oscillations are observed below $T_N = 2.22(1)$ K, consistent with the antiferromagnetic (AFM) Néel temperature observed in bulk experiments. In agreement with neutron diffraction experiments the oscillation frequency $\omega_\mu(T)/2\pi$ follows an $S = 1/2$ mean-field temperature dependence, yielding a quasistatic local field of 1.71(2) kOe at $T = 0$. A crude estimate gives an ordered moment of $\sim 0.66 \mu_B$ at $T = 0$, comparable to 0.81 $\mu_B$ from neutron diffraction. As $T \rightarrow T_N$ from above the dynamic relaxation rate $\lambda_d$ exhibits no critical slowing down, consistent with a mean-field transition. In the AFM phase a $T$-linear fit to $\lambda_d(T)$, appropriate to a Fermi liquid, yields highly enhanced values of $\lambda_d/T$ and the Korringa constant $K_2 \mu T/\lambda_d$, with $K_2$ the estimated muon Knight shift. A strong suppression of $\lambda_d$ by applied field is observed in the AFM phase. These properties are consistent with the observed large Sommerfeld–Wilson and Kadowaki–Woods ratios in Yb$_3$Pt$_4$ (although the data do not discriminate between Fermi-liquid and non-Fermi-liquid states), and suggest strong enhancement of $q \approx 0$ spin correlations between large-Fermi-volume band quasiparticles in the AFM phase of Yb$_3$Pt$_4$.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The term quantum criticality refers to the ensemble of phenomena associated with a critical phase transition at zero temperature. For a number of reasons [1] strongly correlated-electron systems in general, and heavy-fermion (HF) metals in particular, are particularly convenient systems in which to study quantum criticality [2–9], since many HF materials possess a quantum critical point (QCP) at $T = 0$. Properties such as unconventional superconductivity, non-Fermi-liquid (NFL) behaviour, weak-moment antiferromagnetism, and quasi-ordered phases such as ‘nematic ordering’ in HF systems [10–13] are all intimately related to quantum critical behaviour.

Recent studies of the binary compound Yb$_3$Pt$_4$ [14–17] indicate that it represents a new type of quantum critical system, possessing an unusual magnetic phase diagram and behaviour near the QCP that differs significantly from that of ‘conventional’ HF systems. Yb$_3$Pt$_4$ crystallizes in the rhombohedral Pu$_3$Pd$_4$-type structure [18–20], and can be represented by parallel chains of alternating ‘Yb$_3$Pt’ octahedra and ‘Pt$_6$Pt’ octahedra [17, 20]. Neutron diffraction reveals...
a \mathbf{k} = 0\) antiferromagnetic (AFM) structure below a Néel temperature \(T_N = 2.4\) K [17], which is also evidenced in transport and magnetic properties measurements [14, 16]. The specific heat gives evidence that the AFM transition is mean-field at low applied magnetic fields, evolving into a lambda-like second-order transition at a critical end point (CEP) \(T_{CEP} = 1.2\) K, \(H_{CEP} = 15.2\) kOe. Finally \(T_N \rightarrow 0\) at \(H_{QCP} \approx 16.2\) kOe, indicating a magnetic field tuned AFM QCP. The abnormal features related to the QCP in Yb3Pt4 are that (a) there is no mass divergence at the QCP, and (b) the Fermi-liquid state is not destroyed across the QCP. This indicates that the magnetic order might be driven by the exchange enhancement of the Fermi liquid itself in this system and could therefore represent a new route to quantum criticality in heavy-fermion metals.

This paper reports a muon spin rotation and relaxation (\(\mu\)SR) study of the static and dynamic magnetism of Yb3Pt4 single crystals. In the time-differential \(\mu\)SR technique [21, 22] spin-polarized positive muons (\(\mu^+\)) are stopped at the sample and precess in their local fields; the time dependence of the ensemble \(\mu^+\) spin polarization, \(P(t)\), is obtained from the asymmetry \(A(t)\) of the decay positron momenta. In Yb3Pt4 the onset of damped oscillations indicates an AFM transition at \(T_N \approx 2.21(1)\) K, slightly lower than but consistent with the value obtained from bulk data. The oscillation frequency yields a crude estimate for the ordered magnetic moment of \(\sim 0.66\) \(\mu_B/\text{Yb ion}\), consistent with the value of 0.81 \(\mu_B/\text{Yb ion}\) obtained from neutron diffraction [17]. The oscillation frequency follows an \(S = 1/2\) mean-field temperature dependence, also in agreement with neutron diffraction experiments.

The dynamic \(\mu^+\) spin relaxation rate \(\lambda_d(T)\) increases upon approaching \(T_N\) from above but shows no sign of a critical divergence, consistent with a mean-field transition. Below \(T_N\) \(\lambda_d(T)\) drops rapidly from \(T_N\) to \(\sim 1.8\) K, then decreases less rapidly with decreasing temperature, passing through a shallow minimum at \(T_{min} \approx 0.1\) K. Below \(\sim 1.5\) K \(\lambda_d(T)\) can be fitted with a sublinear power law (above \(T_{min}\)), suggestive of NFL behaviour, or alternatively a two-component form that assumes Korringa relaxation of \(\mu^+\) spins and Yb\(^{3+}\) moments by spin scattering of Fermi-liquid quasiparticles. Highly enhanced values are obtained for the coefficient \(\lambda_d/T\) and the Korringa product \(K^2\mu/T\lambda_d\), where the \(\mu^+\) Knight shift \(K_\mu\) is estimated from the magnetic susceptibility. Strong field-induced suppression of \(\lambda_d\) is observed below \(T_N\) but not in the high-temperature paramagnetic phase. These results are suggestive of strong enhancement of \(q \approx 0\) spin correlations in Yb3Pt4, and confirm the unusual nature of the phase transition in this compound.

**2. Experiments**

\(\mu\)SR experiments were carried out on Yb3Pt4 single crystals at the M15 beam line at TRIUMF, Vancouver, Canada. A number of needle-like single crystals (the \(c\)-axis is the long direction) were aligned on a rectangular silver plate of area \(\sim 1\) cm\(^2\) and fastened with GE varnish with the crystalline \(c\) axis perpendicular to the beam direction. The experiments were carried out in the top-loading \(^3\)He–\(^4\)He dilution refrigerator at TRIUMF using the standard time-differential \(\mu\)SR technique [21, 22]. Asymmetry data \(A(t)\) were taken in longitudinal fields up to 10 kOe at 25 mK, 1.2 K and 2.3 K, and over the temperature range 25 mK–4 K in weak longitudinal fields of 15 and 20 Oe. The latter served to ‘decouple’ \(^{195}\)Pt nuclear dipolar fields [23] that otherwise would have made an unwanted contribution to \(A(t)\). These fields also affected the Yb\(^{3+}\) moment fluctuations, as discussed in section 3.2.

Magnetic susceptibility measurements were carried out on a single crystal from the \(\mu\)SR sample in a Quantum Design MPMS magnetometer for applied fields of 100 Oe, 10 kOe, and 20 kOe over the temperature range 2–300 K. The crystal was aligned with the field perpendicular to the \(c\) axis. The data (not shown) agree within errors with previously published results [15, 16].

**3. Results and discussion**

Representative early time asymmetry plots \(A(t)\) are shown in figure 1. At low temperatures the data exhibit damped oscillation due to the precession of \(\mu^+\) spins in the spontaneous local field \(B_{loc}\) due to the ordered Yb\(^{3+}\) moments. The late-time \(A(t)\) data (not shown) are from \(\mu^+\) spin components parallel to \(B_{loc}\) [23], and exhibit dynamic (spin–lattice) relaxation due to thermal fluctuations of the \(\mu^+\) local field.

![Figure 1](image-url)
To model this situation the following two-component function was fitted to the asymmetry data for $T < T_N$:

$$A(t) = A_s \exp(-\lambda_s t) \cos(\omega_{\mu} t) + A_d \exp(-\lambda_d t^2) + A_{\text{bgf}}.$$  

(1)

The first term describes the early time oscillation in a static field with an exponential damping envelope, and the second term is the ‘stretched exponential’ often used to describe an inhomogeneous distribution of dynamic $\mu^+$ relaxation rates [24]. Here $\omega_{\mu}$ is the angular frequency of the $\mu^+$ spin precession, $\lambda_s$ is the exponential damping rate, $\lambda_d$ is the dynamic relaxation rate, and $K < 1$ is the ‘stretching power’. The third term represents a constant background signal arising from muons that stop in the silver sample holder. The total initial asymmetry $A(0) = A_s + A_d + A_{\text{bgf}}$ is assumed to be independent of temperature and applied field. The fitting results based on (1) are shown in figure 1 as solid curves. Equation (1) is also expected to model $A(t)$ data for $T > T_N$ with $A_1 = 0$.

Before further analysing the relaxation data, we discuss the choice of (1) as a fitting function. In ZF- and LF-$\mu$SR, fluctuations of $B_{\text{loc}}$ are often treated using the ‘dynamic Kubo–Toyabe’ relaxation function [23], in which $B_{\text{loc}}$ fluctuates as a whole with a single correlation time. Such a procedure is not appropriate in an ordered magnet, where a quasistatic contribution to $B_{\text{loc}}$ due to the ordered electronic moments is present. Then $B_{\text{loc}}$ is the sum of quasistatic and fluctuating components [13].

3.1. $\mu^+$ spin precession

The temperature dependence of $\omega_{\mu}$ is shown in figure 2(a). As for the neutron diffraction results [17], $\omega_{\mu}(T)$ is well described by an $S = 1/2$ mean-field model (curve in figure 2(a)). A fit to this function yields $T_N = 2.22(1)$ K, which is slightly lower than the value obtained from susceptibility and specific-heat measurements [15]. The zero-temperature frequency $\omega_{\mu}(0)/2\pi = 23.23(3)$ MHz corresponds to a quasistatic local field $B_{\text{loc}}(0) = \omega_{\mu}(0)/\gamma_B = 1.71(2)$ kOe.

A very rough estimate of the static Yb$^{3+}$ moment can be obtained by taking the local field $B_{\text{loc}}$ at the $\mu^+$ stopping site to be the contribution of the uniformly polarised Yb$^{3+}$ magnetization $M$ to the total magnetic field: $B_{\text{loc}} \approx B_{\text{loc}}^\text{stat} = 4\pi M$. Obviously this procedure takes no account of any transferred hyperfine contribution to $B_{\text{loc}}$, the specific $\mu^+$ site, or the Yb$^{3+}$ moment configuration in the AFM phase; it most likely overestimates $B_{\text{loc}}$. A more accurate estimate would use a lattice-sum calculation of the dipolar field at the $\mu^+$ stopping site from the Yb$^{3+}$ magnetic structure determined by neutron diffraction; unfortunately this cannot be done at present because the stopping site is not known.

For Yb$_3$Pt$_4$ the unit cell volume $v_c = 811.65$ Å$^3$, and a unit cell contains 18 Yb ions [15]. Thus $M = 18 \mu_B/v_c = 206$ emu/\mu$_B$, so that $B_{\text{loc}}^\text{stat} = 2.58$ kG/\mu$_B$. Comparison of the measured field with this value yields an estimated moment of $\sim 0.66$ \mu$_B$/Yb ion, in better than expected agreement with the neutron diffraction result of 0.81(5) \mu$_B$/Yb ion [17].

The late-time fraction $\eta_d$, defined by

$$\eta_d = \frac{A_d}{A_s + A_d},$$

is given in the inset of figure 2(a). The step-like change indicates the disappearance of the quasistatic component of $B_{\text{loc}}$ above $T_N$, consistent with the evidence for a sharp mean-field-like transition obtained from $\omega_{\mu}(T)$. The sharpness of the step indicates that the spread in $T_N$ is small.

The exponential damping rate $\lambda_s$ is shown in figure 2(b). We shall see below that the dynamic relaxation rate is much smaller than $\lambda_s$; the latter is therefore dominated by a static inhomogeneous distribution of $\mu^+$ precession frequencies [21]. The decrease of $\lambda_s$ at low temperatures is not understood. The inset to figure 2(b) gives the ratio $\lambda_s/\omega_{\mu}$. Except for a spike at $T_N$, most likely due to a narrow spread of transition temperatures, this ratio is fairly small (20–30%); the spontaneous field in the AFM phase is relatively homogeneous.

3.2. $\mu^+$ spin relaxation

The temperature dependence of the dynamic $\mu^+$ spin relaxation rate $\lambda_d$ in longitudinal fields $H$ of 15 and 20 Oe is plotted in figure 3. Even for such small fields there is a visible field dependence, discussed in more detail below. Below $T_N$ $\lambda_d$ drops rapidly with decreasing temperature (upper left inset of
relaxation data (figure 3). See text for details.

Inset: λ rate H power. In turn contribute to moment fluctuations, also via a Korringa mechanism, which

Equation (4) can, however, be interpreted as a model for $\mu^+$ relaxation by two mechanisms, both of which involve quasiparticle excitations in a Fermi liquid. These excitations couple directly to $\mu^+$ spins (the Korringa mechanism [25], first term in (4)), and to localized Yb$^{3+}$ moment fluctuations, also via a Korringa mechanism, which in turn contribute to $\mu^+$ spin relaxation (second term in (4)). For a Fermi liquid the relaxation is linear in $T$ in both cases. The Korringa relaxation rate of Yb$^{3+}$ moments decreases with decreasing temperature, so that Yb$^{3+}$ fluctuation noise power is transferred to low frequencies. This results in an inverse- $T$ contribution to $\lambda_d$, which samples the noise power at the $\mu^+$ Zeeman frequency.

We do not consider the low-temperature anomaly further, concentrating instead on the temperature region 0.2–1.8 K. There is evidence for a Fermi liquid in the AFM phase of Yb$_3$Pt$_4$ [16], so that we consider further consequences of the Fermi-liquid ansatz for the muon spin relaxation in this region. The Korringa product [25]

relates the $\mu^+$ Knight shift $K_{\mu}$ and the Korringa relaxation rate $AT$ (cf equation (4)) in a Fermi liquid. For a non-interacting Fermi gas

The (rough) order-of-magnitude agreement suggests that itinerant electrons in Yb$_3$Pt$_4$ are indeed in a Fermi-liquid state in the AFM phase at intermediate temperatures. Better agreement cannot be expected for a number of reasons, including the stretched-exponential (inhomogeneous) nature of the relaxation function, the uncertainty in the estimation of $B_{\text{loc}}$, effective spin $\neq 1/2$, and the correlated nature of the conduction-electron band.

We particularly note the effect of enhancement of the $\gamma$ parameter $\gamma_{\mu}$, which is also a function of $B_{\text{loc}}$, and the Korringa relaxation $\lambda_{\mu}$. The large Sommerfeld–Wilson and Kadowaki–Woods ratios in the AFM phase of Yb$_3$Pt$_4$ [16] also confirm this with the band quasiparticles involved in resistive scattering.

The low specific-heat coefficient $\gamma \approx 0.05$ J mol$^{-1}$ K$^{-2}$ in Yb$_3$Pt$_4$ at low fields [16] suggests that the Fermi-liquid band electrons are not particularly heavy. For a Fermi liquid a crude upper bound on $\lambda_d$ is given by the general expression

$$\lambda_d \lesssim T (\gamma_{\mu} B_{\text{loc}})^2 \frac{\hbar}{k_B T^2}.$$ (8)
where $T^*$ is an effective ‘Fermi’ temperature [27]. This yields a very low value of $T^* \lesssim 1.3$ K, suggesting strong enhancement of spin fluctuations sampled by $\lambda_d$. We conclude that in Yb$_3$Pt$_4$ both the uniform response and local magnetic fluctuations are anomalously strong at low frequencies in a way that is not reflected in the specific heat; this is of course already indicated by the anomalously large Wilson ratio.

The upper left inset of figure 3 gives $\lambda_d(T)$ between 25 mK and 4 K. It can be seen that in the paramagnetic phase between $T_N$ and 4 K $\lambda_d$ increases with decreasing temperature, with a cusp at $T_N$. The increase is slow, and shows no sign of the divergence at $T_N$ often observed in other systems [24, 28–31]. Such a divergence is due to critical slowing down of spin fluctuations, which moves the fluctuation noise power down to the low frequencies sampled by $\mu$SR. Thus the absence of a divergence is consistent with a continuous mean-field-like transition with a very narrow critical width.

The lower right inset of figure 3 shows the stretching power $K$ associated with $\lambda_d$. Below $T_N$ $K \approx 0.4$, indicating considerable inhomogeneity in the relaxation rate. There is only a weak temperature dependence below $\sim 1$ K, suggesting the stability of the fitting function in the AFM phase. Above $T_N$ $K \rightarrow 1$, i.e. the relaxation is exponential as expected in a homogeneous paramagnetic state.

Figure 4(a) shows the field dependence of $\lambda_d$ at 25 mK, 1.2 K, and 2.3 K. It can be seen that at 25 mK $\lambda_d$ is strongly suppressed by the applied field, dropping by an order of magnitude in fields of a few tens of Oe. Above 1 kOe $\lambda_d$ is negligible. The measured rates in the range 0.1–1 kOe are not reliable, being so slow that small systematic errors are important. Fits to the power law $\lambda_d \propto H^{-\gamma}$ yield $\gamma = 0.13, 1.2$, and 2.6 for 2.3 K, 1.2 K, and 25 mK, respectively. The suppression becomes quite small above $T_N$. Field-induced rate suppression is often a sign of ‘decoupling’ of the muon spin from a distribution of static internal fields [23]. This cannot be the case in the present situation, however: in the AFM phase the internal field is strong ($\sim 1.6$ kOe), and would not be decoupled by fields less than ten times this value.

A strong power law field dependence of the dynamic relaxation is often observed in spin glasses, paramagnetic NFL materials, and geometrically frustrated magnets. It can be evidence for a divergence in the fluctuation noise power spectrum $J(\omega)$ as $\omega \rightarrow 0$; the relaxation rate, proportional to $J(\omega = \omega_p)$, tracks the noise spectrum as the field and hence the $\mu^+$ Zeeman frequency $\omega_p$ is varied [32]. In Yb$_3$Pt$_4$ the origin of the field dependence is not such a divergence: in a magnetically ordered phase the spontaneous local field $B_{loc}$ dominates, and $\omega_p$ is essentially unchanged until the applied field $H \approx B_{loc}$. The observed field dependence is not well understood, but reflects suppression of low-frequency fluctuations by the uniform field. Such suppression would be particularly effective for fluctuations with $\mathbf{q} \approx 0$, consistent with the previously discussed evidence for correlations with small $\mathbf{q}$.

Below $T_N$ the stretching power $K$ is 0.3–0.5 for all fields, i.e. considerably smaller than 1, indicating strong inhomogeneity of the relaxation process. Above $T_N$ $K < 1$ is observed in an applied field, extrapolating to 1 as $H \rightarrow 0$. Here the applied field seems to induce an inhomogeneity similar to (but not as strong as) that in the AFM phase.

4. Conclusions

We have carried out a $\mu$SR study of single crystals of the unconventional antiferromagnet Yb$_3$Pt$_4$. Well-defined $\mu^+$ spin precession is observed in weak-field $\mu$SR data below a Néel temperature $\approx 2.2$ K, in reasonable agreement with previous results for $T_N$ [15, 16]. The corresponding quasistatic field yields a rough estimate of the ordered Yb$^{3+}$ moment of 0.66 $\mu_B$, in agreement with the value 0.81 $\mu_B$ from neutron diffraction experiments [17]. An $S = 1/2$ mean-field temperature dependence of the ordered magnetization is obtained from both $\mu$SR and neutron diffraction.

In low applied field the temperature dependence of the dynamic $\mu^+$ spin relaxation rate $\lambda_d$ is consistent with $T$-linear Fermi-liquid behaviour over much of the AFM phase. Enhanced magnitudes of both $\lambda_d/T$ and the Korringa constant are observed, and $\lambda_d$ is rapidly suppressed by applied field in the AFM phase. These properties strongly suggest enhancement of spin correlations near $\mathbf{q} = 0$. Below $\sim 0.05T_N$ additional $\mu^+$ spin relaxation is observed, possibly due to Yb$^{3+}$ moment fluctuations due to Korringa scattering. Above $T_N$ $\lambda_d$ exhibits a weak increase with decreasing temperature but no sign of a critical divergence, consistent with a continuous mean-field transition.
The $\mu$SR evidence for some form of Fermi-liquid behaviour (large Fermi volume) in the AFM phase of Yb$_3$Pt$_4$, with enhanced spin correlations in the neighbourhood of $q = 0$, seems in conflict with a previous conclusion [17], based on the small specific-heat coefficient, the relatively large ordered Yb$^{3+}$ moment, and well-defined crystalline-field (CEF) excitations, that the Yb$^{3+}$ electrons are localized (small Fermi volume). It should be noted that observation of CEF excitations, with or without magnetic ordering, is fairly common in heavy-fermion metals [33], as are AFM phases with large ordered moments [34]. We know of no other case of coexistence of large ordered moments with a strongly spin-enhanced Fermi liquid, however.

In antiferromagnetic Yb$_3$Pt$_4$ the enhanced magnitude of $\lambda_d$ is not associated with heavy quasiparticles, since the specific-heat coefficient shows no such enhancement. This difference, like the large Sommerfeld–Wilson and Kadowaki–Woods ratios, indicates that most of the entropy is removed at or just below the AFM transition but that significant low-frequency spin-fluctuation degrees of freedom remain at low temperatures to contribute to these ratios and $\lambda_d$. Further work will be required to understand this unusual compound in detail.

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