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# Magnetism and superconductivity of $UPt_3$ by muon spin techniques

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## Abstract

We summarize our recent μSR results on the magnetic and superconducting properties of the heavy-fermion superconductor $UPt_3$. Whereas zero-field measurements show no signature of the superconducting and magnetic phase transitions, the experiments in high transverse field evidence the presence of two inequivalent magnetic regions for temperatures ranging from 0.2 to 115 K with an anomaly near the magnetic transition. From low transverse field experiments in the low-temperature superconducting phase (B phase), we deduce the temperature dependence of the superfluid densities. We briefly discuss all these results in relation to those obtained for other compounds of this family. © 2000 Elsevier Science B.V. All rights reserved.

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The hexagonal heavy-fermion superconductor $UPt_3$ is attracting much interest because it has been established as an unconventional superconductor as seen by the existence of three distinct superconducting phases in the magnetic field-temperature plane [1,2]. In zero-field the two superconducting phase transitions occur at ~ 0.475 K (B phase) and ~ 0.520 K (A phase). It is usually thought that this complex phase diagram arises from the lifting of the degeneracy of a multi-component superconducting order parameter.

The most popular candidate for such a symmetry-breaking field is the short-range antiferromagnetic order characterized by a Néel temperature of $T_N \simeq 6$ K and an extremely small ordered

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magnetic moment (0.02 (1) $\mu_B$/U-atom). The magnetic order has only been observed by neutron and magnetic X-ray diffractions. Nuclear magnetic resonance (NMR) [3] and zero-field muon spin [4] measurements as well as macroscopic studies have failed to prove the existence of static antiferromagnetic order.

We summarize the results obtained by μSR techniques on high-quality crystals. They differ substantially from the formerly published data measured on first generation samples [2,5]. Because of the limited available space we will not mention the experimental details which can be found in the original reports [4,6,7]. The investigation was started by the zero-field and low transverse-field measurements performed at the pulsed muon source ISIS (UK) and was more recently completed by high transverse-field experiments carried out at the pseudo-continuous muon source of the Paul Scherrer Institute (PSI, Switzerland). The bulk of the work was done on crystals grown in Grenoble. Part of the PSI measurements were perfomed on a crystal prepared in Amsterdam.

Similar to NMR but opposite to the neutron and X-ray diffraction techniques, the zero-field measurements did not exhibit any signature of the magnetic phase transition. The combined results of all these techniques, are consistent with a characteristic fluctuation frequency of the uranium magnetic moments in the range between 1 MHz and 1 GHz [8]. No signature of the breaking of the time reversal symmetry in the A and B phases has been detected either in zero-field experiments. This conclusion has been supported by later magnetization [9] and μSR measurements [10].

Small-angle neutron scattering (SANS) experiments have nicely revealed a well-ordered flux line lattice in the B phase with a structure deviating from the triangular one [11] for a field applied perpendicular to the $c$-axis. These results were not consistent with earlier μSR studies on single crystals, the analysis methodology of which was disputed. By low transverse field measurements [6], we have obtained a consistency with the SANS results in both the magnitude and anisotropy of the low-temperature London penetration depth $\lambda$ ($\lambda_a$(0.05 K) = 604 (13) nm and $\lambda_c$(0.05 K) = 426 (15) nm). Furthermore we got the full temperature dependence of $\lambda_a$ and $\lambda_c$ (Fig. 1). We then derived the characteristic angle $\alpha_a$ of the vortex lattice which is approximately temperature independent. The analysis of the temperature dependence of $\lambda_a^{-2}$ ($\lambda_c^{-2}$) which is proportional to the superfluid density in the $a$ ($c$) direction, is consistent with the existence of a line of nodes at the equator and point nodes at the poles of the Fermi surface.

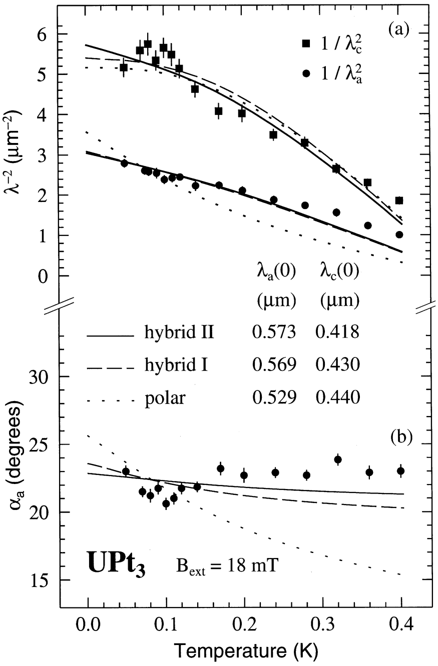

Fig. 1. Temperature dependence of parameters deduced from the analysis of the μSR data. (a) $\lambda_c^{-2}$ (squares) and $\lambda_a^{-2}$ (circles). (b) opening angle $\alpha_a$ ( $= \alpha_{a^*}$) of the vortex lattice for the field applied in the basal plane. The data are presented for $T \leqslant 0.4$ K. Lines give the predictions of models described in Ref. [6].

The study of the temperature dependence of the muon Knight shift through the superconducting phase transitions is expected to yield information on the nature of the spin pairing (singlet or triplet) of the Cooper pairs. We carefully measured the relative frequency shifts in high transverse field μSR experiments for different magnitudes and orientations of this field. We found very small anomalies at the superconducting phase transitions. The Knight shift is obtained from these data after correction for the Lorentz and demagnetization fields. Because of the uncertainty on the demagnetization field

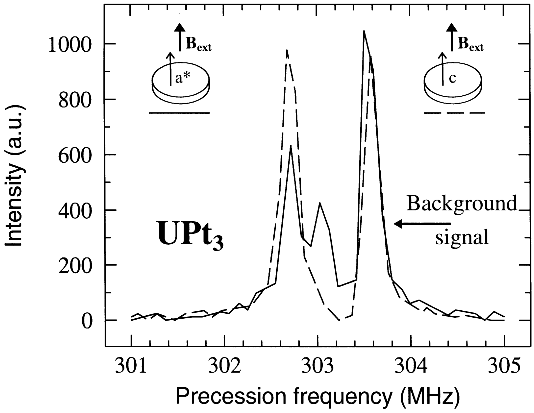

Fig. 2. Two Fourier transforms of spectra recorded at 2.6 K with $B_{\text{ext}} = 2.3$ T. $\boldsymbol{B}_{\text{ext}}$ is either parallel to the $a^*$- or $c$-axis. The line at ~ 303.6 MHz originates from the backing plate.

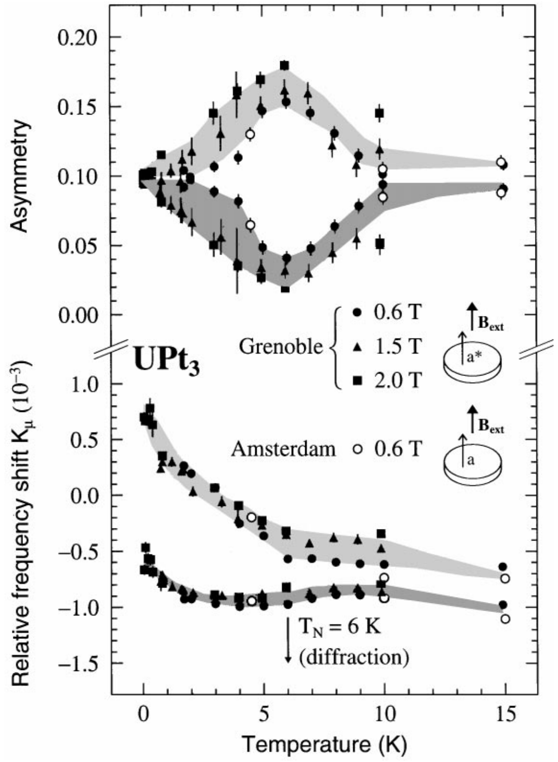

Fig. 3. Temperature dependence of the asymmetries and Knight shifts $K_\mu$ with $\boldsymbol{B}_{\text{ext}}$ perpendicular to the $c$-axis. The data are for $0.5\ \text{K} \leqslant T < 15$ K, three field intensities and two samples denoted Grenoble and Amsterdam. $K_\mu$ is corrected for Lorentz and demagnetization fields. The actual value of $K_\mu$ is subject to an uncertainty due to the demagnetization correction. Nevertheless, the shape of $K_\mu(T)$ is independent of this correction.

coefficient, no reliable information could be extracted from these data. Nevertheless, these data allowed us to get interesting information at higher temperature. We display in Fig. 2 the Fourier transforms of spectra recorded for two directions of the transverse field $\boldsymbol{B}_{\text{ext}}$ applied parallel to the $a^\star$- or $c$-axis. Two signals arising from the sample are observed for $\boldsymbol{B}_{\text{ext}} \parallel a^\star$ whereas only one is seen for $\boldsymbol{B}_{\text{ext}} \parallel c$. This situation persists at least in the temperature range 0.2–115 K. The temperature dependence of the asymmetries and frequency shift of both components is displayed in Fig. 3. The most striking feature is that the two muon precession frequencies have essentially approximately equal asymmetries in the whole temperature range (the region $T > 15$ K is not shown in Fig. 3) except near $T_{\text{N}}$ ($T_{\text{N}} \pm 4$ K) where one asymmetry increases at the expense of the other one. The analysis of the data indicates that the presence of the two components is not due to the existence of two muon sites. Therefore, we conclude that $\text{UPt}_3$ as seen by a local probe such as the muon, is characterized by two magnetically inequivalent regions or domains at least up to 115 K. In Fig. 4 we plot the Knight shift as a function of the bulk susceptibility, the temperature being an implicit parameter. This is the so-called Clogston–Jacarino plot. Whereas the data present no anomaly at $T_{\text{N}}$ for $\boldsymbol{B}_{\text{ext}} \parallel c$, a clear break is observed for one of the components when $\boldsymbol{B}_{\text{ext}} \perp c$. Therefore contrary to the zero-field case a μSR signature of the magnetic transition is obtained in relatively strong magnetic field. Two origins for the Knight shift anomaly found below $T_{\text{N}}$ may be invoked. For the first one the external field slightly rotates the ordered uranium moment. Then the symmetry of the muon site which results in a cancellation of the field for the antiferromagnetic order is broken. A non-vanishing field is then present at the muon in the ordered phase. This interpretation is not consistent with the measured isotropy in the basal plane of the muon response in $\text{UPt}_3$. The second possible origin focuses on the itinerant character of the magnetism of $\text{UPt}_3$. In this picture the additional shift is a measure of the enhancement of the magnetic susceptibility of the conduction

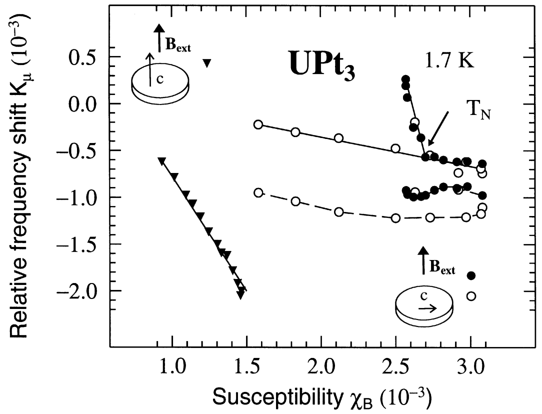

Fig. 4. Clogston–Jaccarino plots obtained for $\boldsymbol{B}_{ext} = 0.6$ T. For $\boldsymbol{B}_{ext} \perp c$ we use the same symbol convention as in Fig. 3. The filled triangles correspond to $\boldsymbol{B}_{ext} \parallel c$. The lines are guides to the eyes.

electrons below $T_N$. The fact that UPt$_3$ is a planar magnet with strong anisotropy between the $c$-axis and the basal plane explains that the anomaly at $T_N$ is only observed for $\boldsymbol{B}_{ext} \perp c$.

Three other uranium heavy-fermion superconductors have been subject to intensive μSR studies. A recent work on UBe$_{13}$ [12] confirms that this compound does not order magnetically and that the uranium magnetic moment is $\lesssim 10^{-3}$ $\mu_B$. In the same work it is shown that $\lambda \gtrsim 1.2$ μm. In UPd$_2$Al$_3$ Feyerherm et al. [13] have provided evidence by Knight shift measurements for the existence of two distinct 5f electronic substates, related either to the superconducting electrons or to the electron responsible for the magnetic ordering. In addition, $\lambda$ is found basically isotropic with $\lambda \sim 0.5$ μm. Finally, measurements on URu$_2$Si$_2$ have been performed but their impact is limited by the quality of the available samples. In agreement with neutron scattering data, the magnetic correlation length is found to be relatively short [14]. Tranverse field data indicate that $\lambda \gtrsim 1$ μm.

In conclusion, the magnetic phase transition of UPt$_3$ which is not observed by macroscopic experimental methods, NMR and zero-field μSR has been detected by the neutron and X-ray scattering techniques and by high transverse field μSR. This gives boundaries on the characteristic fluctuation time of the uranium magnetic moment. In addition two inequivalent magnetic regions have been evidenced up to at least 115 K by this latter method. Concerning the superconducting properties, the μSR techniques have given a rather low upper bound on any spontaneous internal field induced by the Cooper pairs in the low field phases. A relatively large anisotropy of the penetration depth is measured in the full temperature range of the B phase in agreement with the SANS data obtained at low temperature. The analysis of the temperature dependence of the superfluid density is consistent with the existence of a line of nodes at the equator and point nodes at the poles of the Fermi surface. The A phase is characterized by a larger density of nodes than the B phase.

A step forward in the study by μSR of the heavy-fermion superconductors could be the direct observation of the field distribution arising from the flux line lattice. These experiments are difficult because of (i) the involved sample environment needed for such low-temperature experiments and (ii) the relatively large London penetration length characterizing these compounds. In view of this latter constraint the best candidates are UPt$_3$ and UPd$_2$Al$_3$. As far as UPt$_3$ is concerned, another point of interest would be the search for a magnetic signal in zero-field measurements at extremely low temperature, i.e. in the range of 10 mK. This experiment would eventually confirm that the huge anomaly observed by specific heat measurements below 18 mK [15] is of magnetic origin.

Some of us have benefited from the collaboration of R.H. Heffner and J.L. Smith for the UBe$_{13}$ study and of P. Lejay for the preparation of the URu$_2$Si$_2$ crystals.

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## Comments

**Luke:**
*Can the existence of 2 lines with* $B \perp c$ *be due to the presence of external field breaking symmetry (as in the poster of Sonier et al. for* $UBe_{13}$*)?*

**Dalmas de Réotier:**
*In the experiments you are referring to, we have applied the field parallel to the a or a*-axis. Due to a symmetry breaking, a lifting of the degeneracy of the lines can indeed be observed. However, the relative population of the components in this case is not expected to be 1 : 1 as we find.*