Muon spin relaxation and isotropic pairing in superconducting PrOs$_4$Sb$_{12}$

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(Dated: March 22, 2022)

Transverse-field muon spin rotation measurements in the vortex lattice of the heavy-fermion (HF) superconductor PrOs$_4$Sb$_{12}$ yield a temperature dependence of the magnetic penetration depth $\lambda$ indicative of an isotropic or nearly isotropic energy gap. This is not seen to date in any other HF superconductor, and is a signature of isotropic pairing symmetry, possibly related to a novel nonmagnetic “quadrupolar Kondo” HF mechanism in PrOs$_4$Sb$_{12}$. The $T = 0$ relaxation rate $\sigma_{\mu}(0) = 0.91(1) \, \mu s^{-1}$ yields an estimated magnetic penetration depth $\lambda(0) = 3440(20) \, \AA$, which is considerably shorter than in other HF superconductors.

PACS numbers: 71.27.+a, 74.70.Tx, 74.25.Nf, 75.30.Mb, 76.75.+i
Keywords: superconductivity, heavy fermions, penetration depth, muon spin rotation, PrOs$_4$Sb$_{12}$

In most heavy-fermion (HF) metals and superconductors the $f$ ion (Ce,Yb,U) has a magnetic ground state. HF behavior has, however, been reported in a small number of praseodymium-based alloys and compounds [1, 2, 3], in which the crystalline electric field (CEF) ground state of the non-Kramers Pr$^{3+}$ ion could be nonmagnetic and degenerate. In this case a charge-scattering analog of Kondo spin scattering can give rise to the so-called “quadrupolar Kondo effect” [4], an example of the two-channel Kondo effect that has been invoked to explain non-Fermi-liquid behavior in HF systems [5].

Superconductivity has recently been discovered in the cubic HF compound PrOs$_4$Sb$_{12}$ [6, 7]. From thermodynamic measurements it is found that a large carrier effective mass $m^* \approx 50 m_e$, characterizes both the normal and superconducting states, and the transition temperature $T_c = 1.85$ K is relatively high for a HF superconductor. Although a conventional spin-based HF mechanism has not been completely ruled out, thermodynamic properties of PrOs$_4$Sb$_{12}$ suggest a Pr$^{3+}$ nonmagnetic doublet $\Gamma_3$ CEF ground state, so that the quadrupolar Kondo effect is a candidate mechanism for the HF behavior. The symmetry of the superconducting pairing in such a system is a fundamental question.

Transverse-field muon spin rotation (TF-$\mu$SR) has proved an effective probe of the internal magnetic field distribution $n(B)$ in the vortex state of conventional and unconventional type-II superconductors [8, 9, 10]. In the $\mu$SR technique [11, 12] spin-polarized positive muons ($\mu^+$) are implanted into the sample, where the muons precess in their local fields. In general the muon spin relaxation rate is related to the rms width $\langle (\Delta B)^2 \rangle^{1/2}$ of $n(B)$. In turn $\langle (\Delta B)^2 \rangle^{1/2}$ is inversely proportional to the square of the magnetic penetration depth $\lambda$, which is related to the density $n_s$ of superconducting carriers and $m^*$ by the London equation

$$1/\lambda^2 = 4\pi n_s e^2/m^* c^2.$$  \hspace{1cm} (1)

The temperature dependence of $\lambda$ at low temperatures is therefore sensitive to the lowest-lying superconducting excitations, the thermal population of which reduces $n_s$ with increasing temperature. In the presence of an isotropic or nearly isotropic energy gap $\lambda(T) - \lambda(0)$ varies exponentially with temperature, whereas nodes in the gap function characteristic of non-s-wave pairing lead to power-law dependences [13, 14]. In HF superconductors $\lambda$ is typically very long ($\gtrsim 10^4 \, \AA$), in which case the muon relaxation rate can be dominated by other sources of magnetism in the sample. It is then difficult to extract $\lambda(T)$ from TF-$\mu$SR experiments, but the technique has been successfully applied in a number of systems [12].

This Letter reports $\mu$SR experiments in the superconducting state of PrOs$_4$Sb$_{12}$. The penetration depth derived from the vortex-lattice field distribution width exhibits the temperature dependence characteristic of isotropic $s$-wave pairing. Isotropic $p$-wave pairing [13], which is indistinguishable from $s$-wave pairing by thermodynamic or electrodynamic measurements, is also possible. To our knowledge this is the only example to date of an isotropic gap in a HF superconductor. It suggests the possibility of (a) marked differences in superconducting properties between HF materials with magnetic and nonmagnetic $f$-ion ground states, and (b) a relation between the pairing symmetry and the mechanism (spin or quadrupole Kondo effect) for the HF normal state from which the superconductivity evolves.
µSR measurements were carried out in the dilution refrigerator at the M15 beam line, TRIUMF, Vancouver, Canada, on an unaligned powder sample of PrO$_4$Sb$_{12}$ prepared by growth from an antimony flux as described previously [5, 6]. Gallium arsenide, which rapidly depolarizes muons in transverse field, was placed around the sample to minimize any background signal due to muons that stop in the sample holder. TF-µSR and zero-field µSR (ZF-µSR) data were obtained for temperatures between 0.05 and 2.1 K.

In a number of HF superconductors local fields due to spontaneous static magnetism below $T_c$ are inferred from ZF-µSR data [3, 4]. Such magnetism can originate from a spin-wave instability or local-moment ordering, as observed in CeCu$_{2.2}$Si$_2$ [4, 5] where magnetic ordering and superconductivity compete for the ground state. Alternatively, static magnetism below $T_c$ may be due to a superconducting order parameter that breaks time-reversal symmetry, as observed in, e.g., Th-doped UBe$_{13}$ [6]. Muon local fields due to static magnetism could masquerade as vortex lattice inhomogeneity, and falsify the interpretation of the TF-µSR results given above.

We have therefore carried out ZF-µSR experiments to determine whether such static magnetism exists in PrO$_4$Sb$_{12}$. The zero-field data are well fit by a “damped Kubo-Toyabe” function, i.e., the product of a damping exponential and the Kubo-Toyabe function [14] expected from nuclear dipolar fields. Figure 3 gives the temperature dependence of the corresponding relaxation rates $W$ and $\Delta_{KT}$. The exponential damping rate $W(T)$ is already appreciable in the normal state, increases only slightly more than experimental uncertainty below $T_c$, and is (negatively) correlated with $\Delta_{KT}(T)$ in the fitting process. Thus there is no statistically significant evidence for static magnetism below $T_c$; the present data place an upper limit of $\sim 50$ $\mu$T on any static field below $T_c$. We argue below that the relaxation rate changes of Fig. 1 are too small to affect the considerably increased TF-µSR rates in the superconducting state.

Representative TF-µSR muon-spin precession signals at an applied field of 20 mT are shown in Fig. 2 in the normal and superconducting states. A small ($\sim 10\%$)

![TF-µSR spin precession signals in PrO$_4$Sb$_{12}$](image)

**FIG. 2:** TF-µSR spin precession signals in PrO$_4$Sb$_{12}$ ($T_c = 1.82$ K), applied field 20 mT. (a) Normal state ($T = 2.1$ K). (b) Superconducting state ($T = 0.1$ K). The weak nonrelaxing signal in (b) is due to muons that do not stop in the sample.

FIG. 1: Damped Kubo-Toyabe ZF-µSR relaxation in PrO$_4$Sb$_{12}$. Circles: Kubo-Toyabe relaxation rate $\Delta_{KT}(T)$. Triangles: exponential damping rate $W(T)$.
field distribution in an ideal vortex lattice [1] was not observed. The Gaussian relaxation rate $\sigma_s$ is then given by $\gamma_\mu [(\Delta B)^2]^{1/2}$, where $\gamma_\mu \approx 8.516 \times 10^8 \text{ s}^{-1} \text{T}^{-1}$ is the $\mu^+$ gyromagnetic ratio.

The temperature dependence of $\sigma_s$ is shown in Fig. 3 for an applied field of 20 mT. Salient features of these data are that $\sigma_s(T)$ varies only slowly at low temperatures, and that the $T = 0$ value $\sigma_s(0) = 0.91 \mu$s$^{-1}$. This is large for a HF superconductor [8, 9] and is much larger than changes in the ZF-$\mu$SR rates (Fig. 1), so that $\sigma_s$ is dominated by the vortex-state field distribution.

We obtain $\lambda$ from $\sigma_s$ and the expression

$$\frac{\sigma_s(T)}{\sigma_s(0)} \propto \langle B^2 \rangle^2 = 0.00371 \Phi_0^2 \lambda^{-4}$$

appropriate to an isotropic extreme type-II superconductor [12], where $\Phi_0$ is the flux quantum. The data are shown in the inset to Fig. 3. The BCS low-temperature expression $\lambda(T) = \lambda(0)[1 - (T/T_c)^2]^{1/2} \exp(-\Delta(T)/\kappa)$ was fit to data for $T \leq 0.8$ K. The fit value $\lambda(0) = 3440(20) \lambda$, $\Delta(T)/\kappa = 2.1(2)$, from fit to data for $T \leq 0.8$ K.

Early studies of the penetration depth in cuprate superconductors using unaligned powders also observed slight temperature dependence of $\lambda$ at low temperatures, and concluded that the pairing was $s$-wave [10]. Later measurements in high-quality aligned crystals [13] revealed the linear temperature dependence characteristic of $d$-wave pairing. The early results appear to have been due to a combination of circumstances: strong anisotropy in $\lambda$, oxygen inhomogeneity, and sensitivity to hole doping due to the low density $n_s$ of superconducting carriers. None of these factors would seem to affect the current measurements. The crystal structure of PrOs$_4$Sb$_{12}$ is cubic, so that the penetration depth is isotropic. With $m^* \approx 50 m_e$ we find $n_s \approx 10^{22}$ carriers/cm$^3$ from Eq. (1), so that PrOs$_4$Sb$_{12}$ is a good metal and the carrier concentration is insensitive to chemical inhomogeneity. Thus it seems unlikely that the observed temperature dependence of $\lambda(T)$ is due to extrinsic effects.

The resistive mean free path $\ell$ can be calculated from the residual resistivity $\rho(0) \approx 5 \mu\Omega \text{cm}$ [6, 8] using $n_s$ as an estimate of the normal-state carrier concentration, and can be compared with the superconducting coherence length $\xi_0$ [6]. We find $\ell/\xi_0 \approx 3$, so that PrOs$_4$Sb$_{12}$ is a rather clean superconductor [and Eq. (1), derived in the clean limit, is valid]; this is of importance in the analysis of thermodynamic properties.

The field dependence of $\sigma_s$ at $T = 0.1$ K in PrOs$_4$Sb$_{12}$ is shown in Fig. 4. In these experiments the field was set with the temperature above $T_c$, and the sample was cooled into the superconducting state at constant field; otherwise trapped flux broadens the field distribution and causes a spurious increase of the $\mu^+$ relaxation rate. The field dependence of $\sigma_s$ is non-linear, decreasing at low fields and then saturating above $\sim 100$ mT. The origin of this non-linearity is not understood, but might be due to disorder in the vortex lattice [11, 12] or to a change of vortex-lattice symmetry, e.g., from a triangular to a square lattice. Measurements at fields higher than $\sim 200$ mT will be difficult because of the growing importance of the normal-state paramagnetic relaxation.
rate.

Figure 5 is an Uemura plot [10, 20] of superconducting transition temperature $T_c$ vs zero-temperature muon relaxation rate $\sigma_s(0)$ in a number of HF superconductors: UBe$_{13}$ [21], CeCu$_2$Si$_2$ (derived from the penetration depth estimate of Ref. [1]), UPd$_2$Al$_3$ [22], CeCoIn$_5$ and CeIrIn$_5$ [23], UPt$_3$ [24], and PrOs$_4$Sb$_{12}$ (this Letter). It can be seen that $\sigma_s(0)$ is larger in PrOs$_4$Sb$_{12}$ than in other HF superconductors. The values of $\sigma_s(0)$ shown in Fig. 5 are in rough inverse proportion to the HF effective masses of these systems, as expected from Eq. (1) and $\sigma_s(0) \propto 1/\lambda^2(0)$, so that the position of PrOs$_4$Sb$_{12}$ in this plot is not surprising.

The isotropy of the superconducting energy gap indicated by the temperature dependence of $\sigma_s$ (Fig. 4) strongly suggests that superconductivity in PrOs$_4$Sb$_{12}$ is in some sense “conventional.” In this regard PrOs$_4$Sb$_{12}$ differs markedly from other HF superconductors studied to date, a difference that may possibly be related to a nonmagnetic or quadrupolar Kondo state in this compound. Better understanding of the quadrupolar Kondo lattice will be needed to elucidate the questions of whether PrOs$_4$Sb$_{12}$ is indeed such a system and, if so, whether this property is related to the conventional superconducting behavior.

We are grateful to D. Arsenau, B. Hitti, and S. Kreitzman for help with these experiments, and to A. Amato and D. Cox for useful discussions. This work was supported in part by the U.S. NSF, Grant nos. DMR-0102293 (UC Riverside), DMR-9820631 (CSU Los Angeles) and DMR-0072125 (UC San Diego), the Canadian NSERC (Simon Fraser), the U.S. DOE, Grant no. DE-FG03-86ER-45230 (UC San Diego), and the NEDO International Joint Research Program (UC San Diego), and was performed in part under the auspices of the U.S. DOE (Los Alamos).

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