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Heavy-fermion superconductivity in  $PrOs_4Sb_{12}$  studied by  $\mu SR$ 

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After the discovery of superconductivity in  $PrOs_4Sb_{12}$  [1], which is to date the only known Pr-based heavy-fermion superconductor, much of experimental evidence have been reported suggesting that the superconductivity is unconventional. In this talk, after a brief review of the present understanding of the superconducting properties of  $PrOs_4Sb_{12}$ , the results of our muon spin relaxation ( $\mu$ SR) study are discussed, especially focusing on the discovery of broken time-reversal symmetry (TRS) in the superconducting state [2]. TRS-broken superconducting states are quite rare; an unambiguous observation of such fields has only been made in  $Sr_2RuO_4$ [3].

Zero-field (ZF)  $\mu$ SR is the most powerful method to investigate microscopic internal magnetic fields in materials with high sensitivity, as small as 10  $\mu$ T. We applied this technique to study the superconductivity of PrOs<sub>4</sub>Sb<sub>12</sub> with a superconducting (SC) transition temperature (hereafter referred to as  $T_{c1}$ ) of 1.82 K. The measurements have been done at the  $\pi$ A-port in KEK-MSL using small single crystals with sizes of . 1 mm grown by the Sb-flux method, Clear de Haas-van Alphen (dHvA) oscillations observed in one of the crystals [4] are indicative of their high quality.

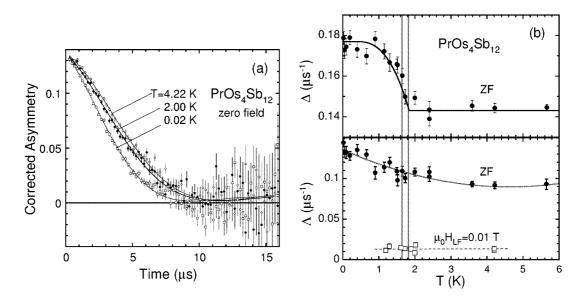


Figure 1: (a) ZF- $\mu$ SR spectra in PrOs<sub>4</sub>Sb<sub>12</sub>. The curves are fits to the model (see text). (b) Temperature dependences of  $\Delta$  and  $\Lambda$ . The vertical lines indicate  $T_{c1} = 1.82$  K and  $T_{c2} = 1.64$  K, at which a sharp jump and a kink structure, respectively, are observed in the specific heat. (from ref.[2])

The ZF- $\mu$ SR spectra are shown in Fig. 1(a). It is obvious that the relaxation becomes stronger with decreasing temperature. As shown by the solid curves in Fig. 1(a), excellent

fits to the data are obtained using  $P_{\mu} = \exp(-\Lambda t)G_z^{\mathsf{KT}}(\Delta, t)$ , where  $G_z^{\mathsf{KT}}$  represents the Kubo-Toyabe function. The best-fit values of  $\Delta$  and  $\Lambda$  are shown in Fig. 1(b). It is remarkable that  $\Delta$  shows a significant increase in the superconducting state (with an onset temperature of around  $T_{c1}$ ), indicating the appearance of a spontaneous internal field associated with the superconductivity. Note that the observed increase in  $\Delta$  is smaller than the experimental uncertainty in a previous measurement [5]. Provided that the increase is of electronic origin, its width  $\Delta_{\rm e}(T \to 0)/\gamma_{\mu} = 1.2$  G is obtained. The temperature dependence of  $\Delta_{\rm e}$  below  $T_{c1}$ agrees with that of the BCS order parameter  $\Delta_{\rm BCS}(T)$ , as evident in Fig. 1(b), supporting our interpretation. The background contribution of  $\Delta = 0.14 \mu s^{-1}$  is attributable to the nuclear dipole fields caused by Sb nuclei located around stopped muons (the  $\Delta$  value is appropriate compared to the results on  $\Pr Fe_4 P_{12}$ ).

The distinct anomaly in  $\Delta$  is in marked contrast with that of  $\Lambda$ , which increases gradually with decreasing temperature across  $T_{c1}$  and  $T_{c2}$  (=1.65 K, where another anomaly appears in the specific heat) without showing any distinct anomalies. This observation suggests the existence of magnetically fluctuating internal fields in the background of the superconductivity. The magnetic fluctuations might be related to the gap-opening feature below 3 K in the relaxation rate of Sb-NQR [6] and a recently observed anomaly in inelastic neutron scattering studies [7].

The present observation of the spontaneous internal fields provides unambiguous evidence that TRS is broken in the SC state, i.e., the spin and/or orbital parts of Cooper pairs carry ordered magnetic moments (it is not conclusive in the temperature range between  $T_{c1}$  and  $T_{c2}$ ). These fields have two types of possible sources depending on the spin and/or orbital parts of Cooper: (1) (for both non-zero spin and orbital moments) spontaneous undumped supercurrents induced in the vicinity of impurities, surfaces, and/or domain walls between the degenerate SC phases, where the order parameter has spatial inhomogeneities (as in the interpretation for Sr<sub>2</sub>RuO<sub>4</sub> [3], and (2) (for non-zero spin moments) a finite hyperfine field induced at the  $\mu^+$ sites. Although we cannot distinguish between the two at this stage, the present finding will help us to narrow down the number of possibilities for the symmetry of the SC order parameter.

Recently we have performed similar experiments on a reference material LaOs<sub>4</sub>Sb<sub>12</sub> and found no evidence both for an increase in  $\Delta$  and for the presence of temperature dependent fluctuating fields. These results are in line with the present understanding of LaOs<sub>4</sub>Sb<sub>12</sub> to be a conventional superconductor. In the temperature vs the La concentration (x) phase diagram for Pr<sub>x</sub>La<sub>1-x</sub>Os<sub>4</sub>Sb<sub>12</sub>, both superconducting states are connected to each other without any suppression of  $T_c$  [8]. Therefore,  $\mu$ SR experiments on the Pr<sub>x</sub>La<sub>1-x</sub>Os<sub>4</sub>Sb<sub>12</sub> system should be very useful to investigate the anomalous superconductivity in PrOs<sub>4</sub>Sb<sub>12</sub>.

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