Low-Field Magnetic Investigations of the Superconducting State in $PrOs_4Sb_{12}$

Tomasz Cichorek [1], Ana Celia Mota, Frank Steglich, Niel A. Frederick [2], Wilhelm M. Yuhasz [2], and M. Brian Maple [2]

The discovery of superconductivity in $CeCu_2Si_2$ pointed to an experimental realization of Cooper pairing different from the indirect electron-electron interaction via phonons [1]. It is now a rather well established fact that unconventional superconductivity (sc) in this and other heavy-fermion metals is mediated by magnetic dipole fluctuations. An interesting exception is the filled skutterudite compound $PrOs_4Sb_{12}$ [2], for which a pair formation via electric quadrupole fluctuations seems to be realized. Such a possibility is suggested by the nonmagnetic ground state of the Pr^{3+} ion combined with an antiferroquadrupolar ordering which emerges between 4.5 T and 16 T and below 1 K, as inferred from neutron diffraction measurements [3]. Furthermore, a large number of experiments point to the unconventional superconductivity in $PrOs_4Sb_{12}$. For example, Sb-NQR data shows the absence of a coherence peak in the temperature dependence of nuclear-spin-lattice-relaxation rate [4] and zero-field μSR data provide evidence for breaking of time-reversal symmetry [5]. Finally, structure in the jumps of both the specific heat [6] and thermal expansion [7] associated with the sc transition suggests that there may be two distinct superconducting phases with superconducting critical temperatures T_{c1} = 1.85 K and T_{c2} = 1.74 K. However, the intrinsic nature of this double phase transition remains open, since a lack of a substantial difference in dT_c/dH has been inferred from the H-T phase diagram [8,9].

Quality of the $PrOs_4Sb_{12}$ single crystal studied (a rectangular parallelepiped of 5×0.2×0.3 mm^3) was checked by measurements of its temperature dependence of both the specific heat $c_p(T)$ and the ac magnetic susceptibility $\chi_{ac}(T)$ in the vicinity of the superconducting transition [10]. As presented in Fig. 1, the $c_p(T)$ data for our sample shows a sharp "double jump" structure. Also shown in Fig. 1 is the in-phase component of the ac susceptibility which indicates that almost the whole superconducting transition occurs already at $T_{c1} \approx$ 1.81 K and only the last 10 % "foot" is extended to T_{c2} = 1.72 K. All the low-field magnetic measurements, namely ac susceptibility, dc magnetization, critical currents (proportional to the remanent magnetization), and vortex creep rates have been taken with a custom built SQUID arrangement.

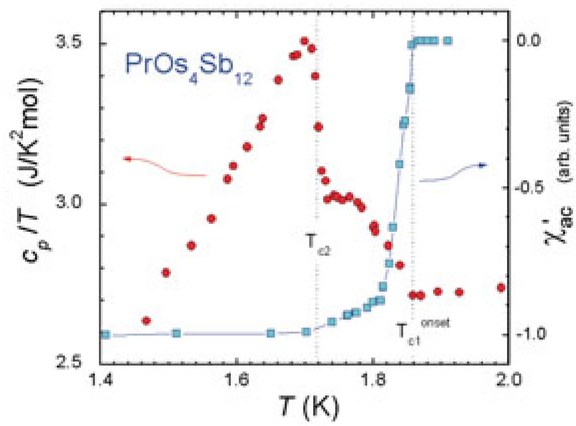

Fig. 1: Specific heat, as c_p/T vs T, (red circles, left scale) and ac magnetic susceptibility (blue squares, right scale) for the $PrOs_4Sb_{12}$ single crystal studied.

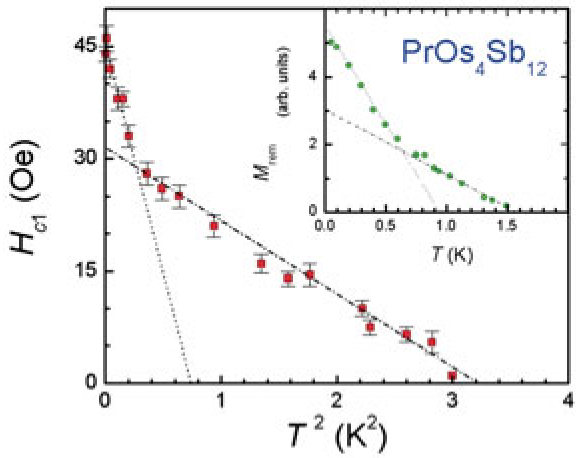

Fig. 2: Lower critical field for $PrOs_4Sb_{12}$ as a function of temperature. Inset: Remanent magnetization M_{rem} vs T for the same single crystal of $PrOs_4Sb_{12}$.

The lower critical field H_{c1} was determined from magnetization (shielding) isotherms [10]. Each magnetization curve was taken after zero-field cooling of the sample to the desired temperature. H_{c1} was defined as the first deviation from the shielding slope in the $M(H)$ curve. The resultant H_{c1} data are plotted in Fig. 2 on a squared-T scale. We observe a pronounced enhancement of H_{c1} below $T \approx$ 0.6 K. Remarkable that similar behavior of $H_{c1}(T)$ have been observed by various groups for thoriated

UBe_{13} and UPt_3 below their second sc transitions. We have fitted the $H_{c1}(T^2)$ data with two straight lines, whose extrapolation to $T = 0$ yields values of $H_{c1}(0)$ of 31 and 44 Oe for the high ($T \lesssim 0.6$ K) and low ($T \gtrsim 0.6$ K) temperature regime, respectively. Since our crystal was in the shape of a rectangular parallelepiped and the magnetic field was aligned parallel to its largest dimension, we have not introduced demagnetization correction to the given values of the lower critical field. We note that our experimental findings are consistent with the positive curvature of $H_{c1}(T)$ deduced from previous magnetization measurements on different $PrOs_4Sb_{12}$ single crystals [11], although these measurements did not have enough resolution to reveal the sharp kink at around 0.6 K.

In the inset of Fig. 2, we present the temperature dependence of the remanent magnetization $M_{\rm rem}$ obtained by cycling the zero-filed-cooled crystal up to the field corresponding to the critical state (i.e., full penetration of vortices into the sample), removing the magnetic field, and finally recording the number of expelled vortices with a digital quantum flux counter as the crystal is heated to $T \gg T_{c1}$. In this case, $M_{\rm rem}$ is proportional to the critical current I_c. Coincident with the enhancement of $M_{\rm rem}$ at $T \approx 0.6$ K, we observe a dramatic increase of I_c below the same temperature. By comparison with thoriated UBe_{13} and UPt_3, one is tempted to identify the pronounced enhancements of both H_{c1} and I_c deep in the sc state of $PrOs_4Sb_{12}$ with a transition into another superconducting phase at $T_c^* \approx 0.6$ K. However, there is no evidence of a jump in the specific heat around 0.6 K, as in thoriated UBe_{13} and UPt_3. One of possible reason could be that the transition at T_c^* is of first order, like between the A and B phases of superfluid ^{3}He, or of a higher order than second. A small feature in $c_p(T)$ could be also obscured by the nuclear Schottky contribution which increases rapidly with decreasing temperature below 0.6 K. On the other hand, several experiments related to the sc gap symmetry also point to an unexplained "0.6 K" anomaly in $PrOs_4Sb_{12}$: the abrupt leveling off of the inverse nuclear spin lattice relaxation time sets in at the same temperature [3]. Additionally, the measurements of the penetration depth in a magnetic field of 200 Oe exhibit a feature that clearly hints at an increase in the superfluid density for $T \lesssim 0.6$ K [12]. Finally, whereas tunneling spectroscopy reveals a well-defined gap of the order of the BCS value, a small feature around 0.6 K was reported [13]. Therefore, in view of the enhancement of H_{c1} and I_c, it seems plausible that the nature of the gap function in $PrOs_4Sb_{12}$ changes deep in its sc state.

Previous study on another sample consisting of many single-crystalline pieces showed a strong irreversibility of isothermal dc-magnetization curves [14]. This was already indicative of large pinning in $PrOs_4Sb_{12}$, which prevents the vortices from entering freely into the sample and inhibits free vortex motion. Most importantly, however, our very recent relaxation measurements of the remanent magnetization for the same $PrOs_4Sb_{12}$ single crystal discussed before [15] clearly point at the very week flux creep (cf. Fig. 3). Furthermore, a relaxation rate of $PrOs_4Sb_{12}$ turns out to be significantly lower that in other heavy-fermion superconductors [16]. Only exceptions are unconventional superconductors with broken time reversal symmetry like, e.g., $U_{1-x}Th_xBe_{13}$ with $x = 0.027$ below its second sc transition at $T_{c2}/T_c \approx 0.6$ [17]. For those systems, an extremely week (practically zero) flux creep was reported. Therefore, we tentatively relate the very strong pinning in $PrOs_4Sb_{12}$ to the spontaneous appearance of static internal magnetic fields below the sc transition temperature — the hallmark of a breaking of time-reversal symmetry, as inferred from zero-field μSR experiments [5].

In summary, our low-field magnetic investigations showed an enhancement of H_{c1} and I_c deep in

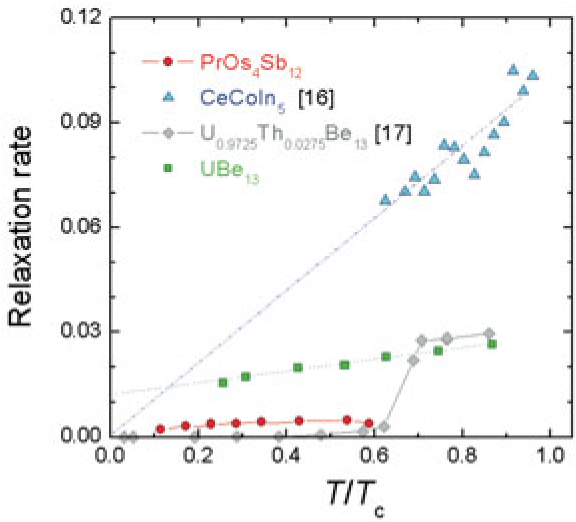

Fig. 3: Normalized logarithmic relaxation rate as a function of reduced temperature for our $PrOs_4Sb_{12}$ single crystal. For comparison, relaxation rates for other heavy-fermion superconductors are also shown. Lines are a guide to the eyes only.

the superconducting state of the filled skutterudite heavy fermion $PrOs_4Sb_{12}$. Since similar enhancements in $H_{c1}(T)$ have been observed for thoriated UBe_{13} and UPt_3, the archetypes of multiphase superconductors, we speculate that low-field anomalies discussed reflect a transition into another sc phase at $T_c^* \approx 0.6$ K. An examination of the literature revealed unexplained features in other physical quantities whose further investigation would be very valuable to clarify an origin of the "0.6 K" anomaly. Finally, a low relaxation rate of the remanent magnetization gives further evidence for unconventional superconductivity in $PrOs_4Sb_{12}$.

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[1] Present address: Institute of Low Temperature and Structure Research, Polish Academy of Science, Wroclaw, Poland
[2] University of California at San Diego, La Jolla, USA