Multiband Superconductivity in the Heavy Fermion Compound $PrOs_4Sb_{12}$

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The thermal conductivity of the heavy fermion superconductor $PrOs_4Sb_{12}$ was measured down to $T_c/40$ throughout the vortex state. At lowest temperatures and for magnetic fields $H \approx 0.07H_{c2}$, already 40% of the normal state thermal conductivity is restored. This behavior (similar to that observed in MgB_2) is a clear signature of multiband superconductivity in this compound.

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The low temperature properties of $PrOs_4Sb_{12}$, the first Pr-based heavy fermion (HF) superconductor [1] (filled skutterudite structure with space group Im$\bar{3}$, $T_c \simeq$ 1.85 K), have many unusual features both in the normal and in the superconducting states [2]: the nonmagnetic singlet ground state of the Pr^{3+} ion suggests that the conduction electron mass renormalization comes from inelastic scattering by crystal field transitions, whereas the superconducting transition temperature would be enhanced by the quadrupolar degrees of freedom of the rare earth f electrons [3,4]. Several experiments also point to *unconventional* superconductivity in this compound: a double superconducting transition in the specific heat [5–7] as well as thermal conductivity measurements in a rotated magnetic field [8] could result from different symmetry states of the order parameter; London penetration depth studies [9] or flux-line lattice distortions [10] indicate nodes of the gap. Tunneling spectroscopy reveals a gap of the order of the BCS value, but with a distribution of values as observed in borocarbides or in $NbSe_2$ [11].

In this Letter, we report a study of heat transport at very low temperature and under magnetic field in $PrOs_4Sb_{12}$, intended to probe the low energy excitations, i.e., the gap structure and nodes. Instead, another phenomenon was uncovered: our results provide compelling evidence for multiband superconductivity (MBSC) in this compound. Coming after similar findings in MgB_2 [12], in $NbSe_2$ [13], or in the borocarbides Y and $LuNi_2B_2C$ [14], they show that very diverse mechanisms may lead to MBSC (or strongly anisotropic gaps) so that it could be much more common than presently thought.

Our rectangular-shaped ($\sim 0.4 \times 0.4 \times 2$ mm^3) $PrOs_4Sb_{12}$ single crystal (same as in [8]) was grown by the Sb-flux method [8] and has $T_c \simeq 1.85$ K. The thermal conductivity (κ) parallel to the magnetic field was measured in a dilution refrigerator by a standard two-thermometers–one-heater steady-state method down to 50 mK and up to 2.5 T [$\mu_0 H_{c2}(T \to 0) \simeq 2.2$ T]. The carbon thermometers were thermalized on the sample by gold wires, spot welded on the surface of the $PrOs_4Sb_{12}$ sample. The same contacts and gold wires were used to measure the electric resistivity of the sample by a standard four-point lock-in technique.

Figure 1 shows the temperature dependence of κ/T at different magnetic fields. Defining $L = \kappa\rho/T$, the inset demonstrates the excellent agreement (within 3%) with the Wiedemann-Franz law at the lowest temperatures in the normal state (data in 2.5 T). The minimum of L/L_0 at temperatures about 1 K reveals the growth of inelastic collisions on warming. For $T \geq 3$ K, L/L_0 increases above 1, maybe due to a phonon contribution to the heat transport (about 20% at 6 K).

At the superconducting transition, κ/T (zero field data) exhibits no anomaly, as predicted by ordinary BCS theory.

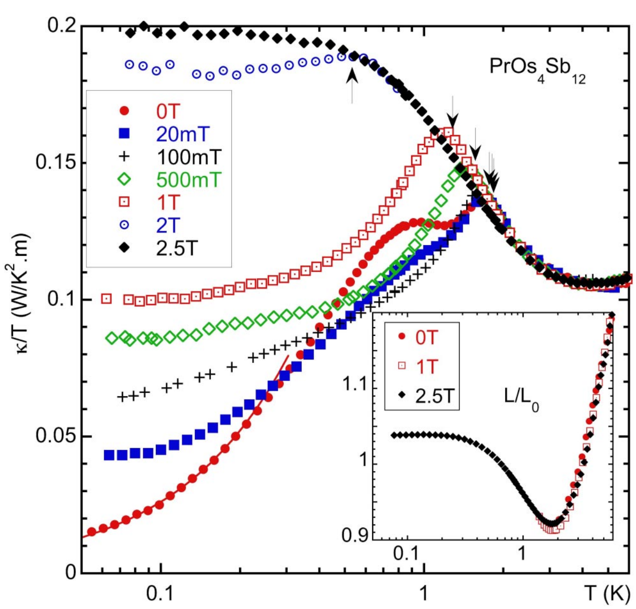

FIG. 1 (color online). $\kappa(T)/T$ at different fields. The anomaly at $T \approx 1$ K is rapidly suppressed under magnetic fields. Solid line: pure $\kappa/T = aT$ law valid in zero field below 0.2 K. Arrows: $T_c(H)$. Inset: test of the Wiedemann-Franz law.

The decrease of $\kappa(T, H = 0)$ seems to take place only slightly below T_c, when the number of excited quasiparticles is reduced by the gap opening. In our case, κ/T exhibits a significant enhancement at about $T_c/2 \approx 1$ K. This feature is suppressed by a field of only 20 mT (see Fig. 1), whereas the specific heat remains unchanged under such small magnetic fields (results not shown here). So this anomaly should be controlled by the scattering mechanism. One possibility is an enhanced phonon contribution below T_c [15], suppressed by the mixed state. According to the measured phonon contribution to the specific heat and a sound velocity of order 2000 m/s [16], a saturation of the phonon mean free path at ≈ 20 μm is required to reproduce the temperature and amplitude of the maximum of the anomaly. This is 10 times smaller than the crystal's smallest length, and might come from extended defects in our crystal, like macroscopic voids, flux inclusions, etc. But other explanations like a boosted quasiparticle inelastic scattering lifetime (as in the high-T_c cuprates [17–19]) are also possible. In fact, strong enhancement of the microwave conductivity is observed below T_c, indicating a rapid collapse in quasiparticle scattering [20].

In the case of a phonon origin of the anomaly, we expect below 0.3 K a contribution to κ of order $0.2T^3$ W/K m, which should not change the observed temperature dependence of our thermal conductivity data below 0.2 K: $\kappa \approx 0.26T^2$ W/K m (full line on Fig. 1). In the case of an electronic contribution, it should have disappeared when inelastic scattering is suppressed, which is certainly the case below 0.3 K (see data at 2.5 T). The T^2 dependence of κ down to $T_c/40$ indicates low energy quasiparticle excitations. However, it does not fit with the simple theoretical predictions for anisotropic gap with nodes: $\kappa \sim T^3$ for line nodes or second order point nodes and $\kappa \sim T^5$ for linear point nodes. It may result from a crossover regime, toward a finite residual value of κ/T expected, for example, in any imperfect sample displaying unconventional superconductivity. We also note that experimentally the T^2 dependence of κ is observed in several unconventional superconductors believed to host line nodes, such as $CeRIn_5(R = Co, Ir)$ [21], Sr_2RuO_4 [22,23], and $CePt_3Si$ [24].

So let us concentrate on the field dependence $\kappa(H)$ at very low temperatures. As discussed above, the quasiparticle mean free path below 0.3 K is governed by elastic impurity scattering. We assume also that the phonon scattering in the same temperature range is governed by static defects and is therefore field independent (at least, it cannot be lowered by the field). Under small magnetic fields (20 or 100 mT) at very low temperatures (50 or 100 mK), we observe a pronounced increase in the thermal conductivity with increasing field (Figs. 1 and 2). At intermediate fields, we observe a crossover to a plateau (for $H/H_{c2} \approx 0.4$), which might be related to the symmetry change observed on thermal conductivity experiments under rotating field [8].

The H dependence of $\kappa(H)$ in $PrOs_4Sb_{12}$ in low fields is in dramatic contrast to that in conventional superconductors. For conventional superconductors in the clean limit, small magnetic fields hardly affect the very low temperature thermal conductivity. By contrast, in unconventional superconductors with nodal structure in the gap function, the Doppler shift experienced by the quasiparticles in the mixed state induces a field dependence of $\kappa(H)$. The initial decrease of $\kappa(H)$ at high temperature (where the condition $\sqrt{H/H_{c2}} < T/T_c$ is satisfied) can be explained by the Doppler shift [25]. But the observed H dependence for $PrOs_4Sb_{12}$ at low fields is intriguing, since it increases with H steeper than expected with the Doppler shift [26]. So, though the Doppler shift can explain the low field behavior of $\kappa(H)$ qualitatively, it is obvious that it cannot explain the whole H dependence: for $T \ll T_c$, the extremely strong field dependence of $\kappa(H)$ in $PrOs_4Sb_{12}$ bears resemblance to that of MgB_2 [27] (see Fig. 3). Indeed, half of the normal state thermal conductivity is restored already at $H \approx 0.05H_{c2}$ for MgB_2, and about 40% of κ at $H \approx 0.07H_{c2}$ in the case of $PrOs_4Sb_{12}$.

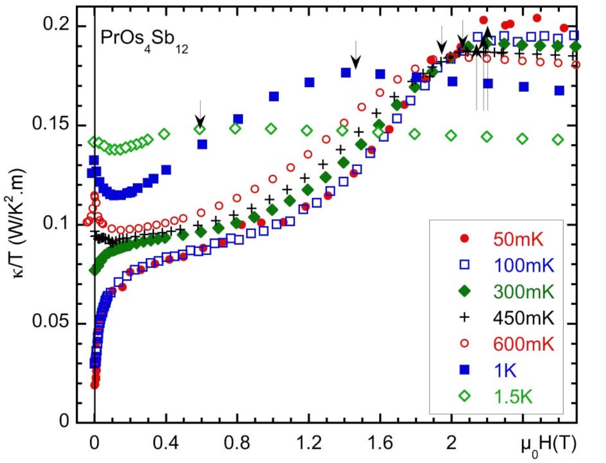

FIG. 2 (color online). $\kappa(H)$: Field dependence of κ: at about $T_c/2$, it may arise from the strong decrease of the "1 K anomaly" (see Fig. 1), whereas at low temperatures, the increase signs MBSC (see Fig. 3). Arrows: $H_{c2}(T)$.

As mentioned in the beginning, MgB_2 is now recognized as the archetype of a two-band superconductor with full gaps, and it is well established experimentally [12,27,28] that the smallest gap on the minor band is highly field sensitive. Theoretically, for κ, as well as for the specific heat, the field dependence of the smallest gap is controlled by a "virtual" H_{c2} (named H_{c2}^S), corresponding to the overlap of the vortex cores of the band with the smallest gap (Δ_S), having a coherence length of order $\frac{\hbar v_F}{\Delta_S}$ [29,30]: above H_{c2}^S, the contribution to κ of the small band with full gap is close to that in the normal state, *only* when it is in the dirty limit (a condition easily satisfied owing to the large coherence length of that band). This remains true even if small interband coupling prevents a real suppression of Δ_S at H_{c2}^S [29]. In the case of $PrOs_4Sb_{12}$, the large ratio of H_{c2}/H_{c2}^S may originate both from the difference in the gap and from the difference in the Fermi velocity between the bands.

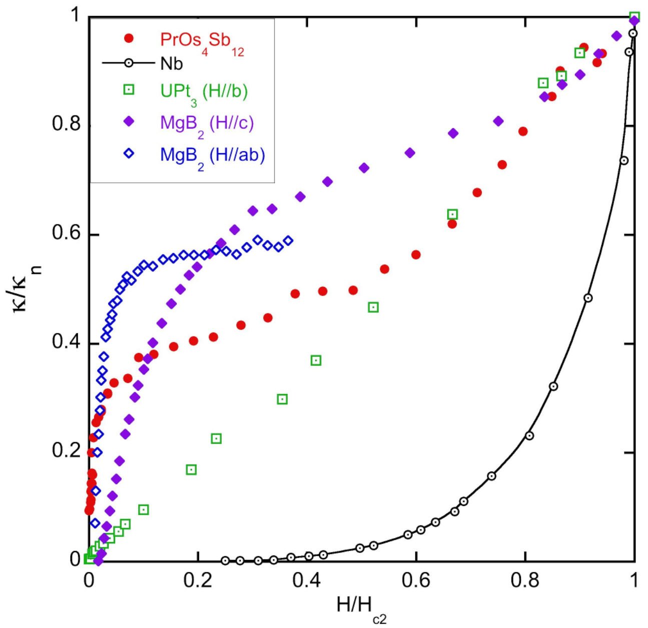

FIG. 3 (color online). Low temperature behavior of $\kappa(H)$ in a conventional one band superconductor (Nb [37]), UPt_3 [32], MgB_2 [27], and our own data on $PrOs_4Sb_{12}$, adapted from [27]. The comparison between MgB_2 and $PrOs_4Sb_{12}$ is striking, and supports two-band superconductivity in this system.

However, this "dirty limit" scenario seems incompatible with the unconventional superconductivity revealed by several experiments [8–10]. On the other hand, in case of unconventional superconductivity, MBSC might as well give rise to a rapid increase of $\kappa(H)$ at low temperature even in the clean limit. Indeed, for an unconventional superconductor, one expects an increase of the contribution of the small gap band on a field scale of order H_{c2}^S provided the condition $\sqrt{H/H_{c2}^S} \gg T/T_c$ is satisfied [25]. In $PrOs_4Sb_{12}$, this will be the case at $T = 0.05$ or 0.1 K whatever the field above H_{c1} (≈ 2 mT) [31].

An additional experimental observation gives support to the MBSC scenario. Indeed, we faced unexpected difficulties in getting reliable measurements in $PrOs_4Sb_{12}$ compared to previous works on other systems (see, e.g., [32]): it was not until very thin (17 μm) Kevlar fibers were used for the suspension of the thermometers that reliable values of κ (satisfying the Wiedemann-Franz law above H_{c2}) were obtained, and it proved very hard to cool down the thermometers below 30 mK. Curiously, if a very small field (≈ 10 mT) is applied, the thermometers cool down below 15 mK. It is nowadays recognized in the community of low temperature thermal measurements that thermal contacts are a central issue for the reliability of such measurements [33]. Before invoking a possible intrinsic mechanism for bad thermal contacts (electron phonon coupling, etc.), we fully characterized these contacts, measuring both their electrical (R_c^e) and the thermal resistance (R_c^{th}).

The results are shown in Fig. 4: despite a constant (Ohmic) R_c^e below T_c, R_c^{th} diverges strongly at low temperatures, explaining the difficulties encountered on cooling the thermometers below 30 mK. It is also seen that this divergence is strongly suppressed in a field of 100 mT, much smaller than the upper critical field H_{c2}, in agreement with the observation of the field sensitivity of the base temperature of the thermometers. R_c^e has the usual Maxwell contribution (coming from the concentration of current and field lines in the contact area): $R_M^e = \rho/2d$, with $d = 17$ μm the gold wires diameter, and ρ the resistivity of $PrOs_4Sb_{12}$ [34] (we can neglect the resistivity of the gold wires), which controls the temperature dependence of R_c^e above T_c and its jump at T_c. It also has an additional constant contribution ($R_{cc} \approx 35$ mΩ), coming from scattering at the Au-$PrOs_4Sb_{12}$ interface. We define R_c^{th} as $\Delta T/P$, $P = R_c^e i^2$ the heat power generated by direct joule heating (with current i), and ΔT the thermal gradient across the contact. Following the analysis of R_c^e, ΔT should have two contributions so that

$$R_c^{\text{th}} = \frac{R_{cc}}{2R_c^e} \frac{1}{L_0 T/R_{cc} + \alpha T^2} + \frac{1}{4d\kappa}\left(1 + \frac{R_{cc}}{R_c^e}\right). \quad (1)$$

The first term is coming from ΔT across the interface. We assume a linear increase of the heat power up to $R_{cc} i^2$ within R_{cc}, a thermal conduction following the Wiedemann-Franz law for the electronic contribution (L_0: Lorentz number), and a αT^2 law for the phonon contribution: $\alpha \approx 0.18 \times 10^{-6}$ W/K^3 is the only free parameter of expression (1), and has the same value for all fields. The second term is the Maxwell contribution from

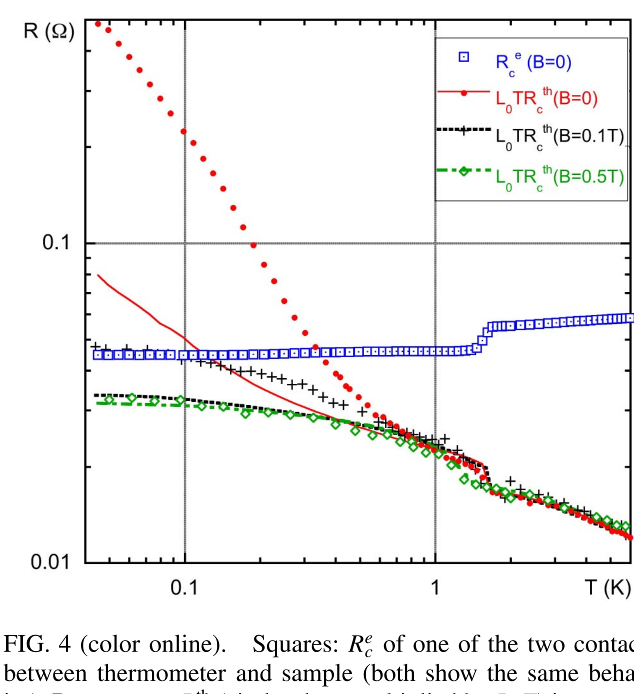

FIG. 4 (color online). Squares: R_c^e of one of the two contacts between thermometer and sample (both show the same behavior). By contrast, R_c^{th} (circles, here multiplied by $L_0 T$) is strongly diverging at low temperature in zero field, and highly field dependent. Lines are calculated from expression (1) for each field. On this graph, the thermal leak due to the Kevlar suspension remains always above 10 Ω.

the thermal conductivity of the sample ($1/2\kappa d$ [34]), with a uniform heat power ($R_{cc}i^2$) plus a nonuniform heat power generated by R_M^e (nonzero only above T_c) [34].

Expression (1) gives a fair account of R_c^{th} in 500 mT over the whole temperature range, and above 0.8 K in 100 mT and in zero field, including the observed jump of R_c^{th} at T_c (due to the change in the distribution of heat power when R_M^e is suppressed). The additional divergence below 0.8 K might well come from the Sharvin surface resistance, which gives additional thermal impedance due to the gap opening even in a metallic contact (Andreev scattering does not contribute to heat transport [35]). This divergence of R_c^{th}, whereas R_c^e remains Ohmic and stable, puts drastic constraints on the thermal insulation of the thermometers from the refrigerator, and can be faced in any other experiments with a constriction at a normal-superconducting interface. On the other hand, the suppression of this divergence in low field in $PrOs_4Sb_{12}$ is easily explained by the MBSC scenario, because above $H_{c2}^S \ll H_{c2}$, thermal excitations from the normal metal will be transferred in the small gap band without an additional barrier. This effect seen on the interface thermal conductivity is even stronger than that observed on the bulk thermal conductivity.

So both $\kappa(H)$ and $R_c^{\text{th}}(H)$, which probe the excitation spectrum, give support to multiband superconductivity in $PrOs_4Sb_{12}$. A possibility for the origin of multiband superconductivity in this system is the spread in density of states among the various bands of that compound [7]: comparison of de Haas–van Alphen [36] and specific heat measurements [1] reveals that some of them contain quasiparticles with large effective masses ($m^* \sim 50m_e$) and the other only light quasiparticles ($m^* \sim 4m_e$) [36], a situation similar to that of most Ce heavy fermion compounds. Theoretical work combining band calculations (for the determination of v_F) and a realistic fit of $\kappa(H)$ (to extract H_{c2}^S) is needed to evaluate precisely the smallest gap. If instead we take the inflection point at low field of $\kappa(H)$ at 50 mK as a "typical value," we get $H_{c2}^S \approx 15$ mT. With the ratio of the Fermi velocities of both bands extracted from $H_{c2}(T)$ [7], we find a gap ratio of order 2: this (rough) estimate contrasts with the very large field effect. It is a direct consequence of the hypothesis that the two bands of the model have very different renormalized Fermi velocities, which may be taken as indicative of weak interband scattering.

So our thermal transport measurements under magnetic field provide clear evidence for MBSC in the HF compound $PrOs_4Sb_{12}$. Strong electronic correlations are thought to be at the origin of the different coupling between the various electronic bands, as opposed to the difference in the dimensionality of the various sheets of the Fermi surface in MgB_2. The low field behavior of κ is consistent with unconventional superconductivity. Further work on purer samples with improved thermal contact would be very valuable to reveal the low temperature behavior of $\kappa(T)$ in this MBSC superconductor in zero field, a study that has not been possible in MgB_2 or $NbSe_2$ owing to a dominant phonon contribution.

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