Superconducting $PrOs_4Sb_{12}$: A Thermal Conductivity Study

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The superconducting state of the heavy fermion $PrOs_4Sb_{12}$ is studied by heat transport measurements on a highly homogeneous single crystal exhibiting only one transition peak in the specific heat. The field *and* temperature dependence of the thermal conductivity confirm multiband superconductivity and point to fully open gaps on the whole Fermi surface.

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Several unusual features have been reported on the superconducting (sc) state of $PrOs_4Sb_{12}$, the first Pr-based heavy fermion superconductor (SC) [1]: A double sc transition in the specific heat (C_p) has been observed on different samples [2–5], like in the well-known case of UPt_3, but its intrinsic nature has not been clearly established yet [4,6]. Other experiments also point to a sc gap with nodes: thermal conductivity (κ) measurements in a rotated magnetic field [7], London penetration depth studies [8], and flux-line lattice distortion [9]. These results contrast with scanning tunneling microscope (STM) spectroscopy [10], Sb nuclear quadrupole resonance (NQR) [11], or muon spin relaxation (μSR) [12], which measured a fully opened gap. Our first low temperature κ study under magnetic field (sample A) provided strong evidence for multiband superconductivity (MBSC) in $PrOs_4Sb_{12}$, but sample quality did not allow analysis of the gap topology from the low temperature regime of κ [13].

In this Letter, we report a new study of thermal transport at very low temperatures on a highly homogeneous $PrOs_4Sb_{12}$ single crystal. Supplementary C_p measurements of this sample show only one single, sharp sc jump at T_c, supporting an extrinsic origin for the double transition reported so far. Improved sample quality also has profound impact on κ, mainly on the temperature dependence $\kappa(T)$ in zero field. It provides compelling evidence for a rather "conventional" MBSC scenario with fully opened gaps on the whole Fermi surface.

Our thin, platelet-shaped $PrOs_4Sb_{12}$ single crystal [sample B2, $\sim 760 \times 340 \times 45\ \mu m^3$, $T_c \simeq 1.75$ K, residual resistivity (ρ) ratio $\rho(300\ \mathrm{K})/\rho(T_c) \sim 30$ instead of ~ 15 in sample A] has been extracted (gently "ground down" against the disk of a diamond saw) from a conglomerate of several small cubes of $PrOs_4Sb_{12}$ (sample B1, $\sim 1 \times 0.75 \times 0.6\ mm^3$), grown by the Sb-flux method [7]. $C_p/T(T)$ in zero field has been measured on a physical property measurement system by Quantum Design, first the entire conglomerate (B1) and then sample B2 alone (see Fig. 1). $\kappa(T, H)$ of sample B2 parallel to the magnetic field has been measured in a dilution refrigerator by a standard two-thermometers–one-heater steady-state method down to 30 mK and up to 3 T [$\mu_0 H_{c2}(T \to 0) \simeq 2.2$ T]. The carbon thermometers were thermalized on the sample by gold wires, held by silver paint on gold stripes evaporated on the surface of the sample after ion gun etching. The gold stripes are essential for the stability and the quality of the electrical contacts (resistance $R_c^e \approx 10\ \mathrm{m}\Omega$ at 4 K). The same contacts and gold wires were used to measure the electric resistivity of the sample by a standard four-point lock-in technique. The reliability of the experimental setup was checked against the Wiedemann-Franz law, giving quantitatively similar results as reported in Ref. [13].

The excellent homogeneity of our sample B2 is documented by the fact that the bulk sc transition appears at exactly the same temperature on C_p/T, κ/T, and ρ (see Fig. 1). Another criterion regarding crystal purity is the residual value of κ/T in the $T \to 0$ limit. For platelet B2, it is smaller than $1.6\ \mu\mathrm{W\,K^2\,cm^{-1}}$, which corresponds to 0.07% of $\kappa/T(T \to 0, \mu_0 H = 2.5\ \mathrm{T} > H_{c2})$ and is significantly lower than in sample A. These signatures of high crystal quality allow us to use thermal transport at very low

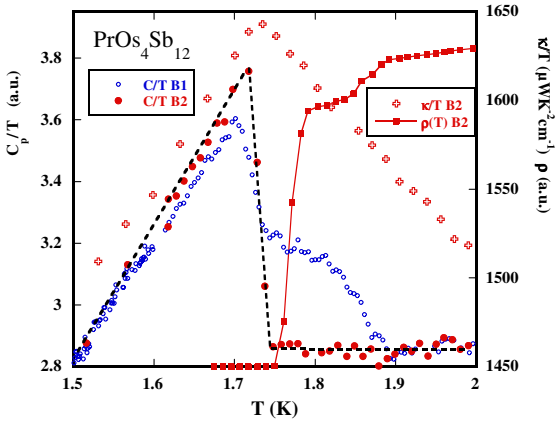

FIG. 1 (color online). Specific heat C_p/T, electric resistivity ρ, and thermal conductivity $\kappa(T)/T$ in zero field around T_c, documenting the collapse of the double transition (observed only in sample B1) and the high homogeneity of sample B2. Nevertheless, a kink at 1.9 K on $\rho(T)$ reveals some "traces" of the upper transition in sample B2, but the bulk sc transition clearly corresponds to the lower T_c from C_p/T of sample B1.

temperatures as a sensitive probe of the low lying energy excitations in $PrOs_4Sb_{12}$.

Figure 1 displays C_p/T, κ/T, and $\rho(T)$ of sample B2 in the vicinity of T_c, as well as the sc transition of sample B1 (C_p/T only). Sample B1 exhibits a double transition on C_p/T, comparable to that reported in Ref. [4]. The fact that both transitions behave similarly under magnetic field and that the upper one always appears inhomogeneous [4] had cast doubt on their intrinsic nature. The remarkable result is that the double transition collapses to a single, sharp jump of C_p/T (at the lowest T_c and of about the same overall height) just by reducing the crystal dimensions (sample B1 $\rightarrow$ B2). Obviously, macroscopic regions with a single and a double transition coexist within the same sample. It suggests that, like in URu_2Si_2 [14], the observed double sc transition in $PrOs_4Sb_{12}$ is related to sample inhomogeneity. A hint for extracting samples with a single transition comes from the preparation stage: In order to remove all small cavities appearing during the sawing process of B1, we had to reduce the thickness of B2 down to only 50 μm. Similarly, such tiny dimensions were reported for another sample exhibiting a single C_p/T jump [6]. Further systematic (structural) investigations are required to determine the nature of defects which might be at the origin of the sharp double transition in $PrOs_4Sb_{12}$.

In Fig. 2, we compare the temperature dependence of κ/T in zero field of samples A (former) and B2 (present). For B2, κ/T has dropped by about 2 orders of magnitude from T_c down to 30 mK, with a clean T^3 behavior of κ below 100 mK. In sample A, the low temperature behavior is probably dominated by impurities, defects, and/or other inhomogeneities, resulting in a sort of crossover regime $\kappa/T \sim T$ [13]. By contrast, at higher temperatures, κ/T is qualitatively the same for both samples, except that there is now a sharp peak in κ/T precisely at the sc transition (as seen by C_p/T and ρ). In sample A, these features were much broader and the local maximum of κ/T appeared below the resistive T_c or the onset of the C_p/T jump.

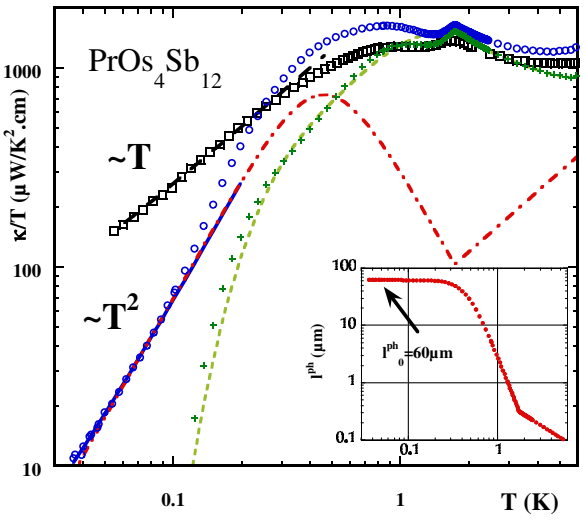

FIG. 2 (color online). $\kappa/T(T)$ in zero field: comparison of former (A, open squares) and present (B2, open circles) samples. For sample B2, we separate the phonon κ^{ph} (dashed-dotted line) and electronic κ^{el} (crosses) contributions. The dashed line is a fit of κ^{el} within the MBSC scenario, with exponential behavior (fully open gaps); see text. The inset shows the estimated temperature dependence of the phonon mean free path.

In Fig. 3, we compare the normalized $\kappa(H)/T$ data at 50 mK of samples A and B2: In this temperature region, the quasiparticle mean free path is governed by *elastic* impurity scattering [13]. The rapid increase of κ at low fields is perfectly reproducible and even more pronounced in sample B2 because of the significant drop of κ/T for $T \ll T_c$ in zero field (see Fig. 2): A magnetic field of only 5% of $H_{c2}(0)$ is enough to restore about 40% of the normal state heat transport. This robust feature, similar to observations in MgB_2 [15], confirms MBSC in $PrOs_4Sb_{12}$ [13]. The plateau at $0.4\kappa_n$ can be interpreted as the "normal state" contribution of the small gap band at $T \rightarrow 0$, observed once the vortex cores of that band overlap. Moreover, despite improved sample quality, there is clearly no sign of a phase transition in the mixed state as suggested in Ref. [7]: $\kappa(H, T \rightarrow 0)$ has no anomaly at the proposed H^* line [$H^*(T \rightarrow 0) \approx 0.8$ T], whereas the $B \rightarrow C$ transition in UPt_3 was clearly seen on $\kappa(H)$ [16].

Let us proceed with a more detailed analysis of the temperature behavior of κ/T. Just above T_c, the Wiedemann-Franz law is recovered [13], which indicates a phonon thermal conductivity (κ^{ph}) negligible compared to the electronic heat transport (κ^{el}) in the neighborhood of T_c. The change of slope observed at T_c [$d(\kappa_S/\kappa_N)/d(T/T_c)$] is of order 1.4. In conventional SC, it is generally ascribed to the combined effects of the opening of the gap and the energy dependence of the electron-phonon scattering rate on κ^{el} [17,18]. In the

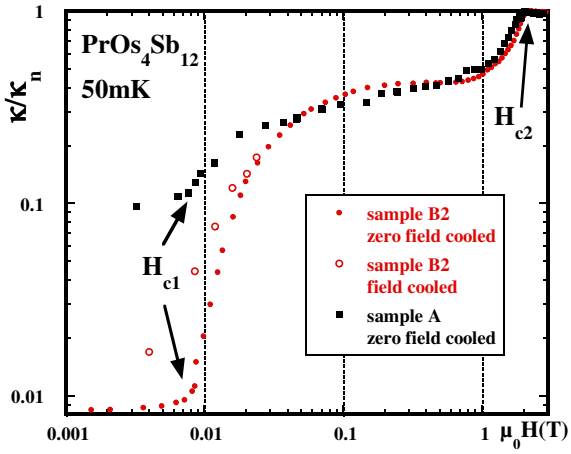

FIG. 3 (color online). $\kappa/T(H)$, normalized to κ_n (its value in the normal state) at 50 mK for samples A and B. The arrows indicate the lower (H_{c1}) and upper (H_{c2}) critical fields. The data in the "field cooled" mode reveal residual flux pinning below 50 mT and a sensitivity of κ to fields as low as 5 mT.

BCS weak-coupling limit, its maximum value is of order 1.4, when lattice scattering is the limiting mechanism for κ^{el} (see measurements on very pure In [19–21]). For $PrOs_4Sb_{12}$, electronic inelastic scattering may replace the effect of electron-lattice scattering. Nevertheless, taking into account the relative weight of elastic to inelastic scattering, as well as MBSC (negligible effect of gap opening in the small gap band), the value of $d(\kappa_S/\kappa_N)/d(T/T_c) \approx 1.4$ appears very large. It is likely a signature of strong-coupling effects, as observed (and calculated), for example, in lead [$d(\kappa_S/\kappa_N)/d(T/T_c) \approx 7$ [17,22]]. Indeed, Sb NQR [11] or C_p analysis [5] have already stressed strong-coupling effects in $PrOs_4Sb_{12}$.

As regards now the origin of the local maximum of κ/T slightly below 1 K, it had been ascribed to an enhanced inelastic mean free path due to the condensation of electronic scattering centers for $T < T_c$. But the question of phonon or electronic origin (or both) was left open [13]. It is now possible to give a quantitative estimate of the phonon contribution: Indeed, a maximum of κ/T below T_c followed by a T^3 behavior of κ at low temperature is well-documented from sc Pb and Nb [22,23], from the rare earth nickel borocarbides RNi_2B_2C (R = Lu, Y) [24–26], and many other materials.

On cooling, the phonon mean free path l^{ph} increases from a law $l^{ph} \sim T^{-1}$ (when it is limited by electron-phonon interactions above T_c) up to a typical crystal dimension at the lowest temperature. But an intermediate regime, described empirically by $l^{ph} \sim T^{-1}(T_c/T)^n$, starts below T_c due to the reduction of scattering by electrons (see inset in Fig. 2). Quantitatively, an estimation for κ^{ph}/T is plotted in Fig. 2 from:

$$\frac{1}{\kappa^{ph}_{sc}/T} = \frac{1}{\kappa^{ph}_{normal}/T}\left(\frac{T}{T_c}\right)^{n=3} + \frac{1}{b l_0^{ph} T^2}. \qquad (1)$$

We used $\kappa^{ph}_{normal} = aT^2$, with $a = 60\ \mu W/K^3\,cm$ (from deviation of the Lorenz number above T_c [13]); bl_0^{ph} is fixed by κ/T at the lowest temperatures ($T < 100$ mK), yielding $l_0^{ph} \sim 60\ \mu m$ $b = 10.9 \times 10^3\ W\,K^{-4}\,m^{-2}$). l_0^{ph} is of order the smallest crystal dimension. The adjustable parameter is mainly the power law (n) for the boosted temperature dependence of l^{ph}: It proved impossible (by adjusting n) to account for the local maximum only by the phonon contribution, so that a situation similar to that of Pb [22] or the high-T_c oxides [27–29] is recovered. In any case, it is seen that κ^{ph} should follow a T^3 behavior up to 0.3 K, giving, in the temperature range 0.1–0.3 K, a robust estimate of κ^{el}/T.

Nevertheless, we tried to understand the normalized electronic contribution $\kappa^{el}/\kappa^{el}_{2.5\ T}$ up to $T \leq 0.6$ K (the region with dominant elastic impurity scattering). The most striking feature is the exponential behavior of κ^{el}, which points to nodeless gaps (see Fig. 2). Moreover, it is not possible to fit $\kappa^{el}(T)$ with a BCS gap corresponding to $T_c = 1.729$ K: κ^{el}/T starts to rise at much lower temperatures than expected, requiring a smaller gap value. This cannot be compensated by strong-coupling effects (which only make it worse, increasing the ratio Δ/T_c) nor by another estimation of the phonon contribution [$\kappa^{ph}(T)$ cannot be larger than $bl_0^{ph}T^3$, constrained by the measurements below 0.1 K]. The data can be quantitatively reproduced within a MBSC scenario, when we include a small $\Delta_s(T)$ and a large $\Delta_l(T)$ gap function with the same T_c, and two associated conduction channels: $\kappa^{el}/T = n_s\kappa^{el}_{\Delta_s}/T + (1 - n_s)\kappa^{el}_{\Delta_l}/T$. The best fit is then obtained for a zero temperature gap ratio of about $\Delta_l/\Delta_s(T \to 0) \sim 3$, with $\Delta_s(T \to 0) \sim 1$ K, and a "weight" for the small gap band $n_s \sim 0.35$. This value is close to the 40% deduced from the "plateau" of $\kappa(H, T \to 0)$ (Fig. 3). The characteristic field scale H^S_{c2} for the vortex core overlap of the small band gap can now be estimated from $H_{c2}/H^S_{c2} \sim (\Delta_l v_{F,s}/\Delta_s v_{F,l})^2$, where $v_{F,i}$ is the average Fermi velocity of band i. In Ref. [4], we assumed that the small gap band is also a light carrier band, with $v_{F,s}/v_{F,l} \sim 5$. We then get $H^S_{c2} \sim 10$ mT, which is of the order of H_{c1} and seems reasonable owing to the data of $\kappa(H)$. The main outcome of this analysis is the existence of a small but *finite* gap Δ_s in $PrOs_4Sb_{12}$, quantitatively consistent with the MBSC scenario deduced from $\kappa(H)$.

Various measurements on the sc state of $PrOs_4Sb_{12}$ have been interpreted as pointing to either gap nodes [7–9] or fully open gaps [10–12]. Focusing on the latest, we can compare the extracted gap values. The NQR [11] as well as the μSR [12] studies propose large ratios of $2\Delta/k_BT_c$, respectively, ~5.2 and ~4.2, supporting strong-coupling effects but not the presence of a small gap. Nevertheless, the NQR data show a large residual relaxation rate ($1/T_1$) below 0.5 K, which may point, as for our sample A, to crystal inhomogeneities which prevent observation of the smallest gap. Moreover, like specific heat, the nuclear relaxation rate should be rather sensitive to bands with a large density of states. So if our interpretation [13] of the small gap band as being also a "light mass" band is correct, it may have indeed little contribution to $1/T_1$. The muon relaxation rate (σ_s) measured with μSR is controlled by the field distribution, which may not put more weight on the heavy than on the light bands. But the measurements were performed in a residual field of 20 mT, already larger than H^S_{c2}, so that again $\sigma_s(T)$ is probably governed by the high energy excitations. However, the "unusual" nonlinear field dependence of $\sigma_s(T = 0.1\ K)$ compares well with $\kappa(H)$: The MBSC scenario, with the small gap band associated to light carriers, can even "explain" the increase of σ_s at low fields as $\sigma_s \propto 1/m^*$. Eventually, the STM measurements [10] proposed a gap distribution, which may extend from 120 to 325 μV ($2\Delta/k_BT_c \sim 1.5$–4.1), not so far from our analysis of $\kappa(T)$ ($2\Delta_S/k_BT_c \sim 1.15$, $2\Delta_L/k_BT_c \sim 3.5$).

A further robust and reproducible [13] experimental observation supports a *fully open* gap in $PrOs_4Sb_{12}$. Indeed, we can also measure on our setup the electrical

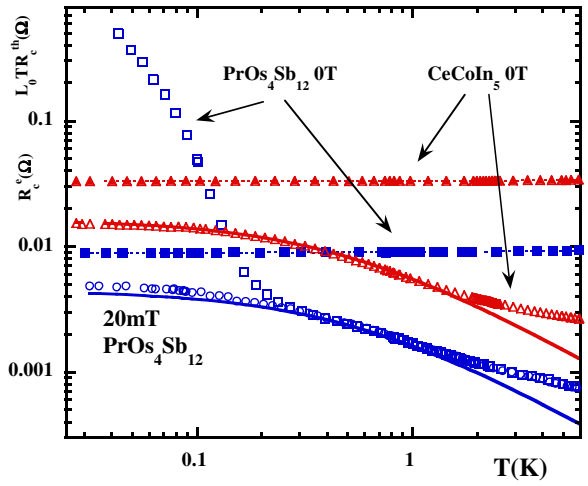

FIG. 4 (color online). Electric $R_c^e(T)$ (solid symbols) and thermal $R_c^{\rm th}(T).L_0T$ (open symbols) resistance of the Au-$PrOs_4Sb_{12}$ or Au-$CeCoIn_5$ contacts. Solid lines: Fits of $R_c^{\rm th}(T)$ according to Eq. (2). $R_c^e(T)$ are temperature and field independent, but $R_c^{\rm th}(T)$ shows a highly singular behavior in zero field below 200 mK on $PrOs_4Sb_{12}$. This further supports a fully open gap in this system (see text).

contact resistance $[R_c^e(T)]$ between the sample and the gold wire of the thermometer thermalization, as well as its *thermal* contact resistance $R_c^{\rm th}$. $R_c^{\rm th}$ is defined as $P/\Delta T$, with P the Joule power dissipated in $R_c^e(T)$ and ΔT the thermal gradient produced by P. For large contact areas, we expect that at low temperatures

$$R_c^{\rm th} \approx 1/2 \frac{1}{\frac{L_0 T}{R_c^e} + aT^2} \qquad (2)$$

(aT^2 being the phonon contribution to the thermal conductivity of the contact). Figure 4 shows that this is well observed below 1 K for $PrOs_4Sb_{12}$ in a field of 20 mT, or $CeCoIn_5$ in zero field (new measurements on the same setup). But for $PrOs_4Sb_{12}$ in zero field, an unexpected divergence is observed below 200 mK, with no correspondence on $R_c^e(T)$ (field and temperature independent). On the same footing, $CeCoIn_5$ in zero field, like $PrOs_4Sb_{12}$ in 20 mT, could be cooled below 10 mK, whereas $PrOs_4Sb_{12}$ in zero field remained stuck above 25 mK. A reason could be that electronic heat transport is suppressed at the normal-sc interface when $k_BT \ll \Delta_s$ (whereas electric current can always go through thanks to Andreev processes [30]): Of course, this barrier is suppressed in very low field in $PrOs_4Sb_{12}$ (multiband effects) or in zero field in $CeCoIn_5$, which has line nodes of the gap [31].

In conclusion, we measured $\kappa(T, H)$ of a highly homogeneous $PrOs_4Sb_{12}$ single crystal exhibiting a *single* jump of C_p/T at T_c. The reproducible field dependence $\kappa(H)$ at $T \ll T_c$ confirms the MBSC scenario, i.e., the existence of 2 (or more) sc gaps sharing the same T_c (like in MgB_2). Further support now comes from the low temperature $\kappa(T)$ and $R_c^{\rm th}(T)$ data which both point to isotropic, *fully opened* gap functions with $\Delta_l/\Delta_s(T \to 0) \sim 3$. Owing to the still mysterious homogeneity problems and to the strong field sensitivity, all analysis requires a close look at the experimental conditions (sample, field, and temperature range). This may have induced the different interpretations on other measurements (concluding gap nodes). As regards the origin of the different coupling strength among the bands, we expect that in $PrOs_4Sb_{12}$, the heavy bands with f character are strongly coupled [4]. Whether this is due to density of states effects [4], or to a pairing mechanism involving f electrons, remains a challenging issue.

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[1] E. D. Bauer *et al.*, Phys. Rev. B **65**, 100506(R) (2002).
[2] M. B. Maple *et al.*, J. Phys. Soc. Jpn. Suppl. **71**, 23 (2002).
[3] R. Vollmer *et al.*, Phys. Rev. Lett. **90**, 057001 (2003).
[4] M. A. Méasson *et al.*, Phys. Rev. B **70**, 064516 (2004).
[5] K. Grube *et al.*, Phys. Rev. B **73**, 104503 (2006).
[6] M. A. Méasson *et al.*, Physica (Amsterdam) **378B–380B**, 56 (2006).
[7] K. Izawa *et al.*, Phys. Rev. Lett. **90**, 117001 (2003).
[8] E. E. M. Chia *et al.*, Phys. Rev. Lett. **91**, 247003 (2003).
[9] A. Huxley *et al.*, Phys. Rev. Lett. **93**, 187005 (2004).
[10] H. Suderow *et al.*, Phys. Rev. B **69**, 060504(R) (2004).
[11] H. Kotegawa *et al.*, Phys. Rev. Lett. **90**, 027001 (2003).
[12] D. E. MacLaughlin *et al.*, Phys. Rev. Lett. **89**, 157001 (2002).
[13] G. Seyfarth *et al.*, Phys. Rev. Lett. **95**, 107004 (2005).
[14] A. P. Ramirez *et al.*, Phys. Rev. B **44**, 5392 (1991).
[15] A. V. Sologubenko *et al.*, Phys. Rev. B **66**, 014504 (2002).
[16] H. Suderow *et al.*, J. Low Temp. Phys. **108**, 11 (1997).
[17] J. Beyer Nielsen *et al.*, Phys. Rev. Lett. **49**, 689 (1982).
[18] V. Ambegaokar *et al.*, Phys. Rev. **139**, A1818 (1965).
[19] R. E. Jones *et al.*, Phys. Rev. **120**, 1167 (1960).
[20] L. Tewordt, Phys. Rev. **128**, 12 (1962).
[21] L. P. Kadanoff *et al.*, Phys. Rev. **124**, 670 (1961).
[22] M. H. Jericho *et al.*, Phys. Rev. B **31**, 3124 (1985).
[23] A. Connolly *et al.*, Proc. R. Soc. A **266**, 429 (1962).
[24] M. Sera *et al.*, Phys. Rev. B **54**, 3062 (1996).
[25] E. Boaknin *et al.*, Physica (Amsterdam) **341C–348C**, 1845 (2000).
[26] B. D. Hennings *et al.*, Phys. Rev. B **66**, 214512 (2002).
[27] J. L. Cohn *et al.*, Phys. Rev. B **45**, R13 144 (1992).
[28] K. Krishana *et al.*, Phys. Rev. Lett. **75**, 3529 (1995).
[29] R. C. Yu *et al.*, Phys. Rev. Lett. **69**, 1431 (1992).
[30] A. F. Andreev, Sov. Phys. JETP **19**, 1228 (1964).
[31] R. Movshovich *et al.*, Phys. Rev. Lett. **86**, 5152 (2001).