## Scaling and Thermal Conductivity in Unconventional Superconductors: The Case of UPt<sub>3</sub>

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We present extensive measurements of the thermal conductivity  $\kappa$  of a high quality single crystal of the heavy fermion superconductor UPt<sub>3</sub> at low fields and very low temperatures (down to 16 mK). Our  $\kappa(B,T)$  data under magnetic fields scale as a function of a single parameter  $x = T/T_c\sqrt{B_{c2}/B}$ yielding to the first observation of the magnetic field-temperature scaling relations recently predicted for a superconductor with a line of zeros in the gap. Both the zero field and the mixed phase measurements of  $\kappa$  give now a consistent picture regarding the gap structure of UPt<sub>3</sub>. [S0031-9007(97)04888-6]

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At present, similarities between heavy fermion and high  $T_c$  copper oxide superconductors are such that most theories aiming at a probe of the unconventional nature of their superconducting phase or at an exploration of its specific properties are elaborated for both types of compounds. Nevertheless, this should not obliterate differences in the current focus of the debate. As regards the high  $T_c$  compounds, a lot of activity is still devoted to the question of extended s-wave versus d-wave scenarios. A consensus towards a *d*-wave scenario is emerging mostly due to microscopic experiments sensitive to the phase of the order parameter (see, e.g., Ref. [1] for a review). In heavy fermion compounds, a direct microscopic evidence for phase changes on the order parameter is not available, but the consensus in favor of the unconventional scenario has emerged with the discovery of multiple superconducting transitions in UPt<sub>3</sub> ( $T_c = 0.5$  K) and extensive work on its complete pressure-field-temperature (P-B-T)phase diagram (see, e.g., Ref. [2] for a review). The central topic concerning UPt<sub>3</sub> is now to identify, among the various order parameters classified according to the irreducible representations of the hexagonal  $D_{6h}$  crystal point group, which one is actually realized.

Recently, a renewed interest for the thermal conductivity ( $\kappa$ ) of UPt<sub>3</sub> has been shown both by theorists [3] and experimentalists [4–6] as it appears that very low temperature measurements of  $\kappa$  might be sensitive not only to the gap anisotropy but also to the order of the gap nodes which distinguish today's most popular  $E_{1g}$  (even parity) and  $E_{2u}$  (odd parity) models. Both models produce a hybrid gap with a line node in the basal plane and point nodes along the *c* axis, suggested by early thermal conductivity and London penetration depth measurements [4,7]. The difference concerning the gap structure for the  $E_{1g}$  and  $E_{2u}$  models lies in the point nodes which are linear for  $E_{1g}$  and quadratic for  $E_{2u}$  [2,3].

The main goal of our paper is to present our new extensive low temperature–low magnetic field measurements of  $\kappa$  in a high quality single crystal of UPt<sub>3</sub>. These mea-

surements give the first demonstration of the scaling relations recently predicted for superconductors having a line of zeros in the gap [8–10]. We argue that the scaling relations we find give a new and up to now completely unexplored appealing probe of the order parameter and that the future theoretical work on the basis of this new probe might lead to a more precise identification of the gap structure of UPt<sub>3</sub>.

At the origin of the scaling hypotheses is the prediction by Volovik [8] that the Doppler shift of the excitation spectrum due to the supercurrent velocity field around a vortex core would lead to a dominant  $\sqrt{B/B_{c2}}$  contribution to the density of states in a superconductor having a gap with a line of nodes. This triggered careful low temperature specific heat experiments under magnetic fields on high  $T_c$  superconductors and a first claim for the experimental observation of such a term in YBaCuO [11]. Later on, the debate runs over the complex analysis of the different low temperature-low field contributions to the specific heat (Schottky anomalies due to impurities, lattice specific heat, parasitic phases, and hyperfine contributions [12]). So at present, both the observation of a  $T^2$  power law at zero field coming from the thermal excitations close to the gap nodes and that of the square root behavior of the T-linear term in the specific heat  $(C/T \propto \sqrt{B/B_{c2}})$  remain a matter of controversy [12].

In UPt<sub>3</sub>, a similar claim has also been made [13]. But strong doubts are shed on this claim because the specific heat of UPt<sub>3</sub> has a huge anomaly below 100 mK which is not of Schottky type [14] and is of unknown origin. This anomaly restricted the analysis of Ref. [13] to the range  $T/T_c > 0.3$ .

More recently [9,10] it has been shown that the same effects responsible for the  $\sqrt{B/B_{c2}}$  behavior of the density of states should yield more general scaling relations of the various thermodynamic and transport properties with respect to the scaling variable  $x = T/T_c\sqrt{B_{c2}/B}$ . In particular, the  $\sqrt{B/B_{c2}}$  behavior for the specific heat is predicted for  $T/T_c \ll 1$ ,  $B/B_{c2} \ll 1$ , and  $x \ll 1$ ,

conditions which were not reached in the measurements of Ref. [13] in UPt<sub>3</sub>. For high  $T_c$  compounds, the only published report for such a scaling behavior concerns thermal Hall data on YBaCuO for only five temperatures between 20 and 30 K, a quite limited temperature range [10].

By contrast, our low temperature measurements in UPt<sub>3</sub> allow one for the first time to span the whole H, T plane at low temperatures and fields. Figure 1 shows some of the measured  $\kappa(T, B)$  curves between 16 mK (0.03  $T_c$ ) and 0.1 K below 0.6 T for fields and heat currents parallel to the basal plane or perpendicular to it. We insist that these are raw data: Owing to the temperature range and the low residual resistivity, the phonons give a negligible contribution to  $\kappa$  (contrary to most other superconductors, like YBaCuO). Below 0.1 K, inelastic electron-electron collisions, which are still very important close to  $T_c$ in UPt<sub>3</sub> can be safely ignored: The zero field thermal conductivity is controlled by purely electronic excitations scattered by the crystal defects and impurities [4-6]. No sign of the specific heat anomaly is observed in  $\kappa(T)$ , suggesting that it is due to rather localized excitations [6]. Also, by contrast to high  $T_c$  cuprates,  $B_{c2}(T)$  is accessible down to  $T \rightarrow 0$  K [15] and displayed in Fig. 2 as measured by the resistive transition. The insets show a zoom between 10 and 100 mK showing no detectable sign of the anomaly reported in the specific heat. The low temperature-low field part of the diagram, which will be

15 15 (mW/K<sup>2</sup>cm) 10 c/T (mW/K<sup>2</sup>cm) 5 10 Ž <sup>0.4</sup> B (T) 0.56 T 0.3 T 0.2 T 5 0.11 T 0.08 T UPt<sub>3</sub> 0.04 T B // j // b 0 T 0 (mW/K<sup>2</sup>cm 20  $\kappa/T (mW/K^2 cm)$ 20 10 Ž 15 0.8 B (T) 0.6 T 0.4 T 10 0.3 T 0.2 T 5 0.1 T UPt\_ 0.05 T ~~~~ B // j // c 0 T 0 0 0.02 0.04 0.06 0.08 0.1 T (K)

FIG. 1. The figure shows the thermal conductivity divided by temperature  $\kappa/T$  as a function of T for different magnetic fields below 100 mK for  $B \parallel j \parallel b$  (upper part) and  $B \parallel j \parallel c$  (lower part). The insets show the magnetic field behavior of  $\kappa/T$ extrapolated at  $T \rightarrow 0$  K.

the region of interest in this paper has been shadowed in Fig. 2.

The samples and the experimental apparatus are described in Ref. [6]. The samples were cut on adjacent sites of the same mother crystal along the b and c axes (basal plane and perpendicular to it) and measured down to 15 mK with the field applied always parallel to the heat current  $(B \parallel i \parallel b \text{ and } B \parallel i \parallel c)$ . Their high quality is demonstrated by the specific heat [5] and the residual resistivities ( $\rho_{b,0} = 0.54 \ \mu\Omega$  cm and  $\rho_{c,0} = 0.17 \ \mu\Omega$  cm) which are among the lowest ever reported. The method was a standard steady heat transport measurement, and the magnetoresistance of the thermometers was corrected systematically at every magnetic field by comparison to a thermometer located in the zero field region of the magnet. The temperature-field sweep is almost continuous, and more than 10 different magnetic field values were applied in the shadowed region of Fig. 2 for each field direction (not all of them are shown in Figs. 1, 3, and 4 for clarity).

In zero field  $\kappa$  follows roughly a  $T^3$  power law for  $j \parallel b$  and  $j \parallel c$  due to the hybrid gap. The magnetic field behavior of  $\kappa/T$  extrapolated at  $T \rightarrow 0$  K is shown in the insets of Fig. 1. Let us point out that this linear field behavior of  $\kappa/T(0, B)$  is not a trivial result for a clean superconductor like UPt<sub>3</sub> where the electronic mean free path  $(l \ge 5000 \text{ Å})$  greatly exceeds the superconducting coherence length ( $\xi_0 \sim 150$  Å). In clean superconductors, it has been demonstrated that the contribution to the thermal conductivity of the electrons in the vortex cores

3.5

2.5

1.5

0.5

1

0

B (J) 2

3

// b



В

0.04 T (K)

2.3

field applied along the basal plane and perpendicular to it measured by the resistive transition [points,  $B_{c2}(T)$ ]. The lines are taken from Ref. [2] and show the different phase transitions found in this compound. The inset shows  $B_{c2}(T)$  between 10 and 100 mK.

is negligible because the mean free path of the core excitations is reduced from the normal state mean free path lto about  $\xi_0 \ll l$ . In *s*-wave superconductors, the excitations outside the vortex cores are fully gapped and  $\kappa(B)$ at low fields and low temperatures is nearly field independent contrasting the linear field increase of the specific heat [16]. Therefore the continuous increase of  $\kappa$  with the magnetic field found here is qualitatively completely different from the one found in clean *s*-wave superconductors. The question now is whether or not we can relate the magnetic field behavior with the proposed models of a hybrid gap, suggested by the low temperature zero field  $\kappa(T)$  measurements.

The scaling relations recently proposed [9,10] should provide a first answer to the question. Indeed, they connect the zero field temperature dependence of thermodynamic and transport properties to their field behavior, under the assumption that everything is governed by thermal excitations of wave vectors in the neighborhood of the nodes of the gap, and located spatially outside the vortex cores. This last point is particularly relevant for the thermal conductivity of clean superconductors. The scaling relations are functions of a single reduced parameter x = $T/T_c\sqrt{B_{c2}/B}$  and are valid for  $T/T_c$  and  $\sqrt{B/B_{c2}} \ll 1$ . But Kopnin and Volovik [9] have shown that they should hold both for  $x \ll 1$  and  $x \gg 1$ , although the scaling function should show a change of regime for  $x \sim 1$ . This function has been explicitly calculated for the specific heat of a superconductor having a line of nodes under the form:  $C(B,T)/T^2 \sim f(x)$  [9,10] and the asymptotic behavior of  $f [f(x) \to \text{const for } x \to 0 \text{ and } f(x) \to 1/x \text{ for } x \to \infty]$ give indeed  $C(0,T) \propto T^2$  and  $C(B,0) \propto \sqrt{B/B_{c2}}$ . As regards thermal conductivity, theoretical predictions have been presented only for 2D superconductors [10], but, following the spirit of Refs. [9,10], we expect a scaling relation close to  $\kappa(B,T)/T^3 = g(x)$  assuming the zero field behavior  $\kappa(0,T) \propto T^3$ .

The knowledge of g(x) is, of course, not necessary to check if the experimental data follow a scaling relation: It is already a challenge to find if the curves can scale as a function of the single parameter  $x = T/T_c\sqrt{B_{c2}/B}$ . Owing to the fact that the zero field curve does not follow a strict  $T^3$  behavior [6], we have searched scaling relations of the form  $\kappa(B,T)/T^n = g(x)$  with *n* an adjustable parameter which should be close to n = 3.

The best scaling relation is found for  $x = T/T_c\sqrt{B_{c2}/B}$ and  $\kappa/T^{2.7}$  for  $B \parallel j \parallel b$  and  $\kappa/T^{3.1}$  for  $B \parallel j \parallel c$ , that is, for power laws in T quite close to the zero temperature power law  $T^3$ . Figure 3 shows the data for  $T/T_c < 0.5$  and  $\sqrt{B/B_{c2}} < 0.5$  plotted in an appropriate way to show the scaling relations. We have plotted the data at low temperatures  $T/T_c < 0.2$  and low fields  $\sqrt{B/B_{c2}} < 0.4$  as filled symbols, at higher temperatures  $T/T_c > 0.2$  as open symbols and at higher fields  $\sqrt{B/B_{c2}} > 0.4$  as crosses. The scaling works in the low magnetic field range ( $\sqrt{B/B_{c2}} < 0.4$ ), however, only



FIG. 3. The upper part (lower part) of the figure shows at low temperatures and low fields  $(T < 0.5T_c \text{ and } \sqrt{B/B_{c2}} < 0.5)$  plotted as  $\kappa/T^{2.7}$  for  $B \parallel j \parallel b \kappa/T^{3.1}$  for  $B \parallel j \parallel c$ ) as a function of  $x = (T/T_c)\sqrt{B_{c2}/B}$ . For  $T > 0.2T_c$  we use open symbols, whereas for  $T < 0.2T_c$  we use filled symbols. For high fields  $(\sqrt{B/B_{c2}} > 0.4)$  we use crosses.

at temperatures even lower than  $0.2(T/T_c)$ . A detailed analysis shows that the departure from scaling laws on increasing temperature for a given field value if found at the temperature  $T^*$  where the  $\kappa(B,T)$  curves cross the zero field curve  $\kappa(0, T^*)$  (see Fig. 1), in such a way that  $\kappa(B,T) > \kappa(0,T)$  for  $T < T^*$  and  $\kappa(B,T) < \kappa(0,T)$  for  $T > T^*$ . This indicates probably that above  $T^*$  some electron scattering process which was not included in the theories of Refs. [9,10] contributes appreciably to the thermal conductivity  $\kappa$ . Note that in UPt<sub>3</sub> it is very difficult to reach the regime x > 1, i.e., magnetic fields as low as  $\sqrt{B/B_{c2}} \ll T/T_c \ll 1$ , because, in this case, we quickly approach  $B_{c1}(\approx 0.01T)$ ; nevertheless, the data show clearly a curvature towards an x-independent behavior at large  $x \left[ \kappa(B,T) / T^3 = g(x) \rightarrow \text{const} \right]$ , as expected to recover the zero field behavior.

When plotting the same data as a function of other parameters, e.g., a more "classical"  $x = (T/T_c) (B_{c2}/B)$ , as shown in Fig. 4 and still  $\kappa(B,T)/T^n$  with  $n \sim 3$  in the ordinate to recover the zero field behavior for  $x \to \infty$ , the measured curves for different magnetic fields are always well separated. This shows that the scaling law is indeed sensitive to the  $\sqrt{B}$  dependence of the parameter x, expected for unconventional superconductors. Even if we relax the constraint of recovering the zero field behavior for  $x \to \infty$  (i.e., treating *n* as an adjustable parameter) in order to concentrate on the region  $x \ll 1$ , we could not find a valid scaling relation. Note that the trivial relation  $\kappa(B,T)/T^2 \approx 1/x$  is followed for temperatures such that  $\kappa(B,T)/T \sim \kappa(B,0)/T \sim \text{const}$ , but no scaling is observed, as this is only a sophisticated formulation of the linear field dependence of  $\kappa(B, 0)$ .



FIG. 4. This figure shows the same data (same symbols) as Fig. 3 plotted against  $x = (T/T_c) (B_{c2}/B)$ . We have included data (crosses) taken at 1 T.

By contrast, the scaling shown in Fig. 3 is valid up to values of x ( $x \sim 1$ ) which include a temperature range where  $\kappa(B,T)$  has already a large departure from the  $\kappa(B,0)/T \sim \text{const}$  found at the lowest temperatures. Therefore, the scaling relations shown in Fig. 3 tell more than the linear increase of  $\kappa(B,0)$ : They also trace the evolution of the temperature dependence of  $\kappa(B,T)$ , governed by the gap nodes, under magnetic field.

Our result demonstrates that the magnetic field thermal conductivity in clean superconductors is related to the gap structure, as much as the thermal conductivity at zero field. The consistent picture regarding the superconducting gap measured by zero field and magnetic field thermal conductivity in UPt<sub>3</sub> is a confirmation of the validity of the thermal conductivity as a probe of the gap structure of unconventional superconductors.

This also gives a new way for measuring the gap structure that we hope will stimulate new theoretical calculations which could take full advantage of scaling and might help to discriminate between different possible order parameters. The goal might be to provide a microscopic connection between the temperature and field dependences of  $\kappa$ . As regards the superconducting gap of UPt<sub>3</sub>, we note that we find similar scaling behaviors for the heat current in the basal plane and along the *c* axis: This seems quite natural for a hybrid gap as theories [3] already predict only weak differences on the temperature

dependence of  $\kappa(0, T)$  for both directions, but has not yet been theoretically established.

In conclusion, our thermal conductivity measurements have provided a first thorough test of the predicted scaling behavior of the low field–low temperature properties of superconductors having a line of zeros in the gap.

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