# Anisotropy of Point-Contact Spectra in the Heavy-Fermion Superconductor $UPt_3$

G. Goll, H. v. Löhneysen, and I. K. Yanson[a]
*Physikalisches Institut, Universität Karlsruhe, W-7500 Karlsruhe, Germany*

L. Taillefer
*Department of Physics, McGill University, Montreal, Canada H3A2T8*

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The heavy-fermion superconductor $UPt_3$ has been studied in the superconducting state down to 36 mK by point-contact spectroscopy. Distinct minima in the differential resistance $dV/dI$ versus voltage $V$ related to the energy gap are observed for current flow parallel to the crystallographic $c$ axis, while for current flow within the basal plane such features were almost always absent. This difference, together with a comparison of the Blonder-Tinkham-Klapwijk theory, gives clear evidence for a strong gap anisotropy. This is further supported by the unusual temperature and magnetic-field dependence of the spectra.

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In classical superconductors Cooper pairs with total spin zero and total angular momentum zero (BCS pairing) are formed via the electron-phonon interaction. In recent years, several new classes of superconducting materials such as high-$T_c$ oxide superconductors, organic superconductors, and, notably heavy-fermion superconductors (HFS) have been discovered with the notion of possible "unconventional" superconductivity in one of the following ways: (1) A superconductive order parameter (OP) is denoted as unconventional if below the transition temperature $T_c$ additional symmetries are broken besides the gauge symmetry. In such a case, the OP will in general show strong gap anisotropy, e.g., points or lines of nodes on the Fermi surface where the OP vanishes. (2) The Cooper pair attraction is unconventional, i.e., mediated by nonphononic interactions. There is some connection between these two points as revealed by the case of superfluid $^3$He, the only system where unconventional pairing has been clearly identified [1]. However, there is increasing evidence that some HFS, in particular, $UPt_3$, may exhibit unconventional superconductivity. In $UPt_3$, the strongest evidence is the multiplicity of superconducting phases observed [2–5]. Also, the temperature ($T$) dependence of various physical properties below $T_c$, such as specific heat [5], is indirect evidence for a gap function with zeros on the Fermi surface. More powerful are studies which rely on directional probes, such as ultrasonic attenuation [6], London penetration depth [7], and thermal conductivity [8]. Although these probes have been used to investigate the gap structure of $UPt_3$ [6–8], they have not yet led to an unambiguous identification of the gap function. More direct information on this is highly needed.

Point-contact (PC) spectra, i.e., differential resistance $dV/dI$ versus voltage $V$ of a PC between superconductor ($S$) and normal metal ($N$), through the well-known mechanism of Andreev reflection [9,10] at a $N/S$ interface, should be amenable to unravel the gap structure of anisotropic superconductors [11]. Such experiments have been performed on a number of HFS. Measurements on $UPt_3$ [12] and $URu_2Si_2$ [13,14] have been able to show that a gap does exist. However, detailed information is lacking. For instance, recent experiments on $URu_2Si_2$ [13] could not distinguish between isotropic ("$s$ wave") and anisotropic (e.g., "$d$ wave") gaps. Furthermore, $T$ was not low enough in previous experiments so that thermal smearing of the gap-related features precluded a definite statement.

The present work reports PC spectra of $UPt_3$ which yield the following new results. (1) A gap-related structure in the spectra is generally observed for current ($I$) flow parallel to the crystallographic $c$ direction and only very weak structures—if at all—are seen for $I$ flow within the basal plane. This strong anisotropy is the first direct evidence that the gap vanishes in the basal plane as predicted by various theoretical scenarios of unconventional superconductivity in $UPt_3$ [15]. (2) Our $T$ is low enough to obtain a clear signature of the gap (where present). The PC spectra cannot be modeled with the Blonder-Tinkham-Klapwijk (BTK) theory with an isotropic energy gap [9]. (3) The gap-related features are only observed in the low-temperature, low-field superconducting phase of $UPt_3$. This hints at the different OP's of the superconducting phases.

The $UPt_3$ sample was a piece of a single crystal grown by the Czochralski method. It was annealed for 2 d at 1473 K under UHV. The specific heat of this sample showed a superconductive double transition with $T_c^+ = 521$ mK and $T_c^- = 460$ mK. It was then cut into a rectangular shape by spark erosion with edges of 0.5 mm along the crystallographic $a$ direction of hexagonal $UPt_3$, 2.3 mm along $c$, and 3.7 mm along the $b$ direction perpendicular to $a$ and $c$. The surface was cleaned by etching with 5 vol% HCl in $HNO_3$. PC spectra were obtained by pressing a Pt counterelectrode against one of the sample's surfaces, presumably resulting in a preferred $I$ flow in the direction perpendicular to the surface. Throughout this paper the $I$ direction is to be understood in this sense. Additionally, a piece of a polycrystal where the superconductive double transition was previously observed (sample 1 of [3,5]), was also investigated. This piece contained a large grain (surface area $1.5\times0.5$ mm$^2$) as detected with an optical microscope. The surface normal of this grain as determined from Laue pictures had an angle of 20° to 30° with the $c$ axis. The orientation with respect to $a$ and $b$ could not be determined. We measured $T_c^-=439$ mK and $T_c^+=506$ mK with the specific heat [16] in good agreement with earlier data [5].

The sample and Pt counterelectrode were mounted in a device that was immersed directly in the mixing chamber of a $^3$He/$^4$He dilution refrigerator in order to avoid heating of the PC region. A PC between Pt and UPt$_3$ could be both established and changed at low $T$ from the outside by means of mechanical feedthroughs. This proved to be essential not only because of thermal contraction of the device, but also because experience showed that a "gentle touch" resulted in the most reliable contacts. Typical zero-bias resistances $R_0$ were of the order of 1 $\Omega$ from which a PC diameter of $d=(16\rho l/3\pi R_0)^{1/2}\sim 600$ Å is estimated for the ballistic limit [17]. Here $\rho$ is the resistivity and $l$ the electron mean free path of UPt$_3$. The product $\rho l$ for UPt$_3$ is $2\times10^{-15}$ $\Omega\,\mathrm{m}^2$ [7]; hence the residual resistivity of 1–2 $\mu\Omega\,$cm corresponds to 2000–1000 Å. (The estimate $d=\rho/R=130$ Å appropriate for the thermal limit also yields $d<1$.) The PC spectra were obtained by standard lock-in technique in a constant-current mode.

Figure 1 shows representative PC spectra for $I$ along different directions measured at our lowest $T$ (between 36 and 53 mK). About twenty spectra were taken for each direction. The first point to note is that the spectra are almost symmetric with respect to voltage bias and, in addition, a given spectrum was reproducible with up and down voltage sweeps to almost within the scatter of the data points. The salient feature to be read directly off Fig. 1 is the minimum of $dV/dI$ vs $V$ for small $V$ and the leveling off near 100 $\mu$V for $I\parallel c$. In some cases, a double-minimum structure was found (curves 5 and 6). The upper branch of curve 5 was taken above $T_c^+$ at 520 mK. The feature has now completely vanished. This proves that these features are due to the superconducting energy gap in UPt$_3$. Most remarkably, for $I\parallel c$ this feature was found nearly always while for $I\parallel a$ and $I\parallel b$ the absence of such a feature was the rule. Only very rarely a shallow minimum near zero bias was observed for these $I$ directions (curves 2 and 4 in Fig. 1). Of course, we do not know the microscopic structure of our PC and hence cannot make definite statements about $I$ flow and $I$ spread. By statistics, however, we may state that the features observed most often for a given $I$ direction (adjusted macroscopically) are representative for PC's with microscopic $I$ flow in that direction. Hence our experiment provides compelling evidence that the gap-related structures are much stronger for $I\parallel c$ than for $I\perp c$, i.e., within the basal plane. To our knowledge, this is the first time that a strong gap anisotropy has ever been directly observed in a HFS. In view of the most often observed absence of any gap-related feature for $I\perp c$, our data even give some direct evidence for a line of gap nodes in that plane, as has been indirectly inferred from the $T$ dependence of thermodynamic and transport data [5–8]. In principle, quite different features in PC spectra are conceivable if the ratio of the superconducting coherence length $\xi$ to the PC diameter varies. However, for UPt$_3$ $\xi_c=125$ Å and $\xi_{ab}=113$ Å [18] do not differ too much; hence this possibility is unlikely. Also, $l>d$ for all directions; hence the directional dependence of $l$ should have no pronounced effect on the spectra.

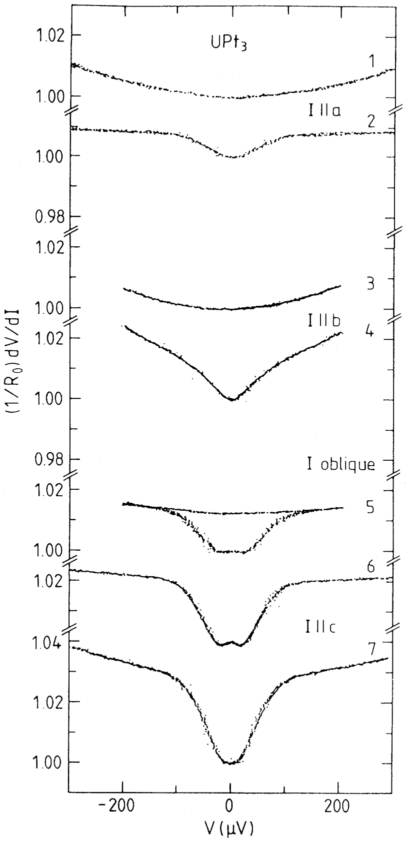

FIG. 1. Differential resistance $dV/dI$ vs voltage $V$ for different PC's of the UPt$_3$ single crystal (curves 1–4, 6, and 7) and the grain (curve 5) normalized to the zero-bias resistance $R_0$. The temperatures $T$ (mK) and zero-bias resistances $R_0$ ($\Omega$) are 37, 0.877 (curve 1); 37, 1.284 (2); 50, 0.447 (3); 36, 0.631 (4); 53, 1.077 (5); 42, 1.617 (6); 37, 1.377 (7). The upper branch of curve 5 (measured at $T=520$ mK) was shifted to coincide with the lower branch at high voltages.

Even for the "strong-gap" direction along $c$ the gap-related feature is in fact weak compared to the simple expectation of Andreev reflection (AR). At a $N/S$ interface, an electron in $N$ is backscattered as a hole with probability $A$ and induces a Cooper pair in $S$ to take up the momentum. This leads to a zero-bias resistance which for $A=1$ is reduced by a value of $r=0.5$ with respect to the resistance at $V \gg \Delta e$ where $\Delta$ is the superconducting energy gap. AR can occur also in gapless superconductors (with, of course, a reduced $A$) as long as there is a nonzero probability of Cooper pairs. Hence it probes, strictly speaking, the superconductive OP and not the gap. Our spectra show a reduction of $r=3\%$ at most. For an isotropic superconductor, impurities are not expected to influence $r$ to a large extent [19]. However, a strongly anisotropic scattering rate may lead to a deviation of $r$ from the theoretical value $r=0.5$. A second, and in our view more likely, possibility of $r$ reduction is that an anisotropic gap with nodes reduces the probability of AR. Also, it is not known how the weak antiferromagnetism of UPt$_3$ acts on the superconductivity at the surface, and thus on $r$.

We now turn to a more detailed analysis of the form of the gap feature for $I \parallel c$. The double-minimum structure has been observed here for the first time in a HFS. This is the clearest possible identification of an energy gap in PC spectroscopy. A detailed analysis requires knowledge of the normal-state PC spectrum for low bias. This background was determined by measurements in the normal state (cf. curve 5 in Fig. 1). Of course, one has to be aware of the effect of thermal smearing. However, because the $dV/dI$ curve in the normal state varies smoothly, it is not noticeably affected by thermal smearing. Indeed, the normal-state $dV/dI$ curve at $-50$ mK obtained in a high magnetic field looks identical. Examples of $dV/dI$ curves with background subtracted are shown in Figs. 2(a) and 2(b), respectively, for two different samples (single crystal and grain), and for spectra with $I \parallel c$ and $I$ oblique, respectively, where the double-minimum structure was observed. This structure is, in fact, expected for isotropic superconductors when the probability $A$ for AR is smaller than unity and a probability $T$ for quasiparticle tunneling through a barrier at the $N/S$ interface exists [9,10]. This leads to minima of $dV/dI$ at $V \approx \Delta/e$. Figures 2(a) and 2(b) also show the theoretical BTK curves (calculated for finite $T$) for isotropic superconductors. The fit parameters are $\Delta=29$ $\mu$eV and the barrier strength $Z=0.2$ [Fig. 2(a)] and $\Delta=39$ $\mu$eV and $Z=0.15$ [Fig. 2(b)]. The values of $2\Delta/k_BT_c=1.3$ and 1.8, for single crystal and grain, respectively, fall well short of weak-coupling BCS theory, which might be attributed to an anisotropic gap with nodes. Although the low-bias structure can be modeled quite well with isotropic BTK theory, it is not surprising to find deviations from the BTK fit at higher bias which *cannot* be attributed to thermal smearing. Rather, it is due to the averaging over an anisotropic gap.

Certain PC features, e.g., a double-minimum feature in $dV/dI$ for $A<1$, are also expected for anisotropic superconductors, dependent on the $k$ value of the electrons incident on the $N/S$ interface [11]. The averaging over current directions for different PC's may easily explain why we have not always observed such a feature.

For the often proposed [15] two-dimensional (2D) even-parity representation $E_{1g}$ of the OP in UPt$_3$ $\Delta(\mathbf{k}) \sim \eta_x k_z k_x + \eta_y k_z k_y$, with a line of nodes in the basal plane and point zeros at the poles, one would expect no gap feature in $dV/dI$ for $I$ strictly parallel to $c$. However, because of only point zeros the directional averaging with $I$ contributions off the $c$ direction allows observation of a gap feature. Our results for $I \parallel a$ and $\parallel b$ suggest that averaging over line zeros, i.e., preferred $I$ flow parallel to a plane containing a line of nodes, yields no gap feature.

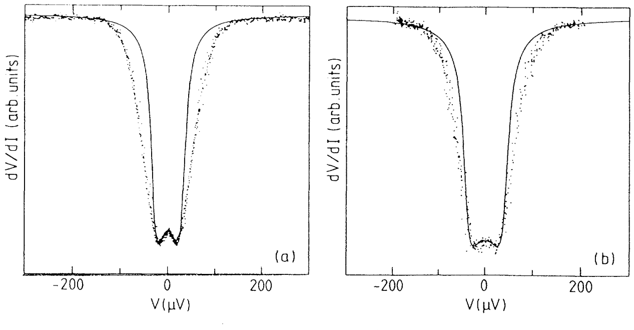

FIG. 2. Differential resistance $dV/dI$ vs voltage $V$ after subtraction of the background for curve 6 [single crystal, (a)] and curve 5 [grain, (b)]. Solid lines indicate a BTK fit for an isotropic superconductor.

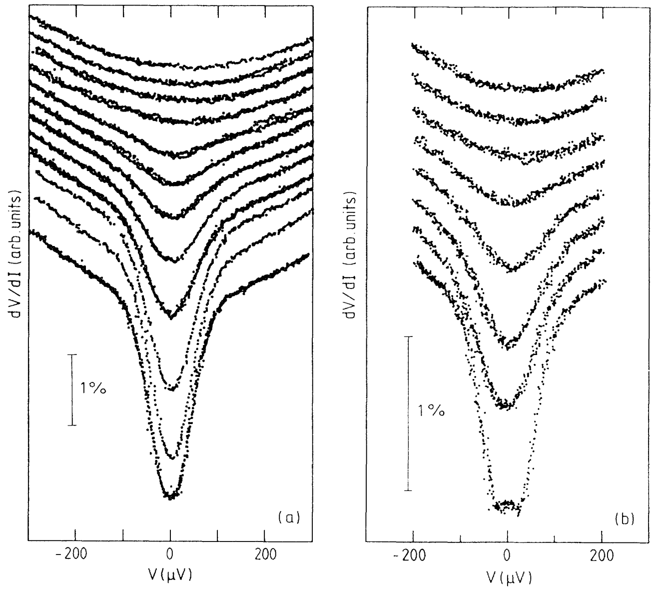

FIG. 3. Temperature dependence of the differential resistance for curves 7 (a) and 5 (b) in Fig. 1. The temperatures $T$ (mK) are (from bottom to top): (a) 34, 102, 198, 299, 352, 394, 421, 442, 471, 499, 538, and 617; (b) 53, 235, 300, 364, 407, 445, 488, and 520. The spectra are shifted vertically for clarity.

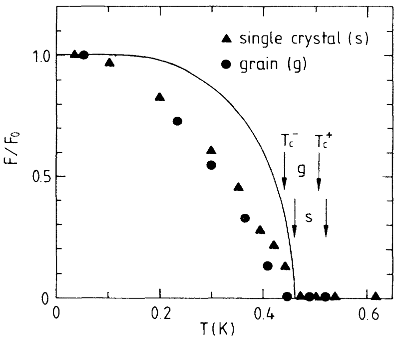

FIG. 4. Temperature dependence of the area $F$ of the gap-related feature normalized to the value $F_0$ obtained at lowest $T$ for the single crystal (▲) and the grain (●). The solid line describes the $T$ dependence of the OP in BCS theory. The arrows indicate the values of $T_c^-$ and $T_c^+$ (see text).

One expects that the averaged gap value is largest for oblique $I$ direction. This is indeed suggested by our experiments, with the grain showing a larger value.

As a final point, we discuss the $T$ dependence of the PC spectra. Figures 3(a) and 3(b) display spectra where in one case a double-minimum structure was observed, while the other showed only a single minimum at $V=0$. The features vanish when the normal state of UPt$_3$ is approached with increasing $T$. Lacking the exact knowledge of the anisotropic gap function and a corresponding BTK expression, we take the area $F$ between the $dI/dV$ vs $V$ curve and the corresponding normal-state background as a measure of the relative magnitude of the (averaged) gap [9]. Figure 4 shows $F$ vs $T$ (obtained via numerical inversion of the measured $dV/dI$ curves) normalized to the value $F_0$ at the lowest $T$. The $F/F_0$ vs $T$ dependence is quite different from the standard BCS behavior of an isotropic superconductor. The faster decrease at low $T$ again is in line with gap zeros. Perhaps even more striking, the gap-related intensity vanishes for both samples well below $T_c^+$ and in fact very close to $T_c^-$. (It would be very coincidental if $T_c^+$ of the PC's in two different samples would be lowered just to look like $T_c^-$.) This suggests that between $T_c^-$ and $T_c^+$ AR is suppressed to a low probability for reasons yet to be explored. Preliminary studies in a magnetic field indicate that no feature in PC spectra is seen in the high-field phase, either. Thus, in terms of the $E_{1g}$ representation for the superconductive OP, only the $\eta=(1,\pm i)$ phase appears to exhibit AR for $I\parallel c$, and not the (0,1) or (1,0) phases with an additional line of nodes in a plane $\parallel c$. This suggests that gap features do not appear for preferred current flow parallel to a plane with lines of nodes, in nice consistency with the directional dependence of the PC spectra discussed above. Our results thus clearly support the notion that the different superconducting phases of UPt$_3$ have distinctly different OP's.

In conclusion, we have found pronounced differences in point-contact spectra in UPt$_3$ measured for different current directions, a direct evidence for an anisotropic superconductive energy gap. Further evidence for an anisotropic gap comes from the form of the gap-related feature and from its temperature dependence. The occurrence of gap-related features for $I\parallel c$ only in the low-field, low-temperature phase of superconducting UPt$_3$ clearly points to different superconductive OP's.

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[a] Permanent address: Institute for Low Temperature Physics and Engineering, Academy of Sciences of the Ukraine, 310164 Kharkov, Ukraine.

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