Andreev Reflections on Heavy-Fermion Superconductors

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(Received 7 June 1993; revised manuscript received 13 October 1993)

In a comparative Andreev-reflection study of normal-superconductor point contacts on the c plane of UPt_3, $CeCu_2Si_2$, and URu_2Si_2 crystals at temperatures down to $T=30$ mK and in a magnetic field up to $B=10$ T, differences are found in the current-voltage characteristics. The behavior of the differential resistance $dV/dI(V)$ of UPt_3 is attributed to the presence of nodes in the superconducting order parameter $\Delta(\hat{\mathbf{k}})$ and is compared with theoretical predictions for the symmetry of the pairing state. The data for $CeCu_2Si_2$ and URu_2Si_2 point to the absence of zeros in $\Delta(\hat{\mathbf{k}})$.

PACS numbers: 74.70.Tx, 73.40.Jn

In many experiments performed to clarify the nature of the pairing of quasiparticles in heavy-fermion (HF) superconductors, superconducting properties have been found, which deviate strongly from those described by the weak coupling BCS theory. In particular, the power laws of the temperature dependence of both the specific heat and the kinetic properties of these systems observed below the critical temperature T_c give indirect evidences that the superconducting order parameter $\Delta(\hat{\mathbf{k}})$ should vanish at points or lines on the Fermi surface [1]. Although group theory gives a classification of all possible symmetries of $\Delta(\hat{\mathbf{k}})$ of these systems [2], no experiments up to now have been able to clearly distinguish between the different symmetries. Point-contact (PC) experiments have already been done on the HF superconductors UPt_3, URu_2Si_2, and UBe_{13} [3–6] and showed that it is possible to observe the Andreev-reflection process in normal-superconductor (NS) PC's. The point-contact technique allows a direct study of the superconduction gap Δ. For the special case of UPt_3 the results obtained are not in agreement with the predictions for isotropic superconductors [4].

UPt_3 has a complex B-T phase diagram including a tetracritical point and at least three distinct superconducting phases, in which the observed splitting of T_c is most likely due to the influence of the antiferromagnetism that develops below $T_N=5$ K and lifts the degeneracy of the two pairing states [7]. A pairing function belonging to a degenerate 2D representation has often been proposed [8]. However, it was recently argued that such a 2D scenario cannot easily explain the existence of the tetracritical point for $B\parallel\hat{c}$ and a 1D nonunitary state was instead proposed [9] and supported by experimental μSR results [10]. Among the directional probes, ultrasound attenuation data on UPt_3 [11] are compatible with a Δ which has a line of nodes in the basal plane [1]. Measurements of the anisotropy of the temperature dependence of the magnetic-field penetration depth also suggest that Δ has a line of nodes in the basal plane and point nodes at the poles [12].

In this Letter we present new results on the current-voltage (I-V) characteristics of NS PC's for the three HF superconductors UPt_3, $CeCu_2Si_2$, and URu_2Si_2. The low temperatures reached (down to 30 mK) reduce considerably the thermal broadening of the spectra. A systematic study of the temperature (T) and magnetic-field (B) dependence of the data at zero-bias voltage yields information about the presence of quasiparticles with zero excitation energy in the superconducting state. The experimental results for PC's on UPt_3 indicate the presence of zeros in Δ. Different possible symmetries are tested within the framework of a generalized Blonder-Tinkham-Klapwijk (BTK) model. In contrast, the data for $CeCu_2Si_2$ and URu_2Si_2 show no traces of nodes in Δ.

Electronic transport in NS junctions is described by the BTK formalism [13]. When a voltage V is applied over the junction, electrons injected through a PC with an excess energy $E=eV\geq\Delta$ can cross the NS interface and enter into quasiparticle states in the superconductor. For $E<\Delta$, the traversing electron condenses into the superconducting state by forming a Cooper pair with another electron which leaves behind it a hole with the opposite velocity in the normal electrode. This so-called Andreev-reflection process [13] leads to an excess current I_{exc} and to a lower PC resistance for $eV<\Delta$. The voltage dependence of the PC resistance therefore contains direct information about Δ.

The BTK theory for isotropic superconductors [13] can be extended to the anisotropic case by including the momentum $(\hat{\mathbf{k}})$ dependence of the superconducting energy gap $\Delta(\hat{\mathbf{k}})$ in the expressions for the Andreev-reflection probability A and the normal-reflection probability B [13]. The differential resistance R_{NS} of the NS junction normalized to the normal state value R_{NN} can, at $T=0$,

0031-9007/94/72(14)/2278(4)$06.00

be written as

$$\frac{R_{NS}}{R_{NN}}(V)=\frac{(\partial I_{NS}/\partial V)^{-1}}{(\partial I_{NN}/\partial V)^{-1}}(V)=\frac{(\partial/\partial V)\int d^3k\hbar^{-1}(\partial\varepsilon/\partial k_z)[1-Z^2/(1+Z^2)]}{(\partial/\partial V)\int d^3k\hbar^{-1}(\partial\varepsilon/\partial k_z)[1+A(\varepsilon,\Delta(\hat{\mathbf{k}}))-B(\varepsilon,\Delta(\hat{\mathbf{k}}))]}, \quad (1)$$

where Z is a phenomenological parameter accounting for all processes responsible for a transmission coefficient of the metallic PC less than unity. The integrals in momentum space are restricted to states close to the Fermi energy E_F with energies ε between $E_F-eV/2$ and $E_F+eV/2$ and positive velocity components perpendicular to the interface, $\hbar^{-1}\partial\varepsilon/\partial k_z>0$. In this approximation the proximity effect at the NS interface is not taken into account, although it could have a significant influence on the energy dependence of the probabilities A and B for anisotropic superconductors [14].

Our PC's were produced by pressing a sharp electrochemically etched Ag tip (normal metal) onto the surface of the HF superconductor. The contacts were adjusted at low temperature ($T>30$ mK) inside the mixing chamber of a dilution refrigerator by means of a bimorph stepping motor [15]. This allows the adjustment of many (up to 20) different very stable PC's within one single cooling cycle. A typical PC resistance of 1 Ω corresponds to a radius of the PC of about 200 Å [16]. The results from different PC's pressed onto the c plane of the HF superconductors showed excellent reproducibility. The polycrystalline UPt_3 sample consisted of grains of typical surface size 1 mm^2, whereas single crystals of $CeCu_2Si_2$ and URu_2Si_2 were used with typical surface size in the range of a few mm^2. All the HF's were mechanically cleaved perpendicularly to the c axis ($\hat{\mathbf{n}}\|c$ axis) immediately before mounting in the cryostat. The differential resistance of the PC's was recorded by a modulation technique with an excitation of the order of 1 μV.

Figure 1 shows the normalized resistance curves for PC's on the HF compounds studied here in the superconducting ($B=0$, $T\approx 50$ mK) and in the normal state ($B>B_c$ and/or $T>T_c$). All NS PC's have a reduced resistance at low voltage due to the Andreev-reflection process indicating that a ($\mathbf{k},-\mathbf{k}$) type of Cooper pairing is realized in each of these HF superconductors. In the superconducting state, UPt_3 PC's exhibit characteristics with a structure of nearly triangular shape centered at $V=0$ consistent with other recent observations [4]. $CeCu_2Si_2$ and URu_2Si_2 show a flat structure around $V=0$, a pronounced peaked structure, and a smoothly rising background for increasing voltage. $CeCu_2Si_2$ PC's also show in some cases minima around $V=\pm 50$ μV above which the peaked structure begins to develop.

From Fig. 1 it is obvious that in the case of UPt_3 the excess current I_{exc} is present up to the highest voltage applied ($V=350$ μV). However, for $CeCu_2Si_2$ and URu_2Si_2 the peaked structure corresponds to a vanishing I_{exc} with increasing voltage, i.e., the destruction of superconductivity in the contact area. For UPt_3 the BTK formalism for Andreev reflections can then be applied over the whole studied voltage range, while for the two other compounds it is applicable only for the region below the observed peaks in $R_{NS}(V)$.

In Fig. 2 we compare a typical experimental $R_{NS}/R_{NN}(V)$ characteristic (expt.) obtained on UPt_3 with curves calculated using Eq. (1) for different symmetries of $\Delta(\hat{\mathbf{k}})$ proposed for this system. We have approximated the complicated Fermi surface S_F of UPt_3 [17] by an ellipsoid with a mass anisotropy $m_\perp/m_\parallel\sim 2.3$ determined experimentally by neutron diffraction from the vortex lattice [18]. Here $m_\perp$ and $m_\parallel$ are the effective masses perpendicular and parallel to the c axis. For simplicity the calculations have been performed for $Z=0$ yielding relative resistance changes $R_{NS}/R_{NN}(0)$ of a factor of 2. A broadening of the density of states simulated by a complex term $\Gamma\sim\Delta/2$ in the energy ε of the quasiparticles in the BTK formalism combined with a nonzero value of $Z\sim 0.4$ is able to explain the value of $R_{NS}/R_{NN}(0)$ measured experimentally while leaving practically unchanged

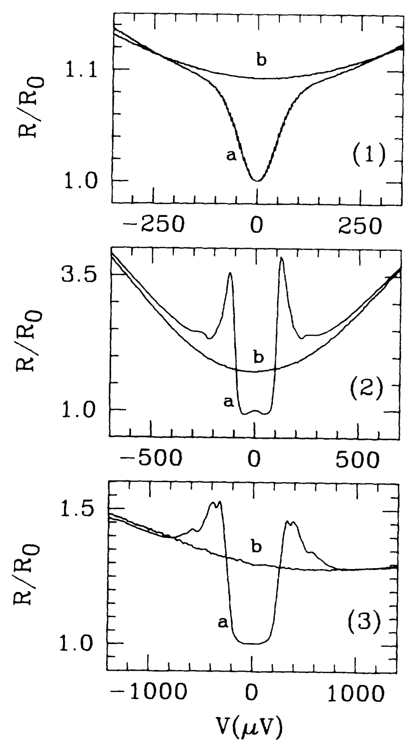

FIG. 1. Normalized differential resistance $R(V)/R_0$ of PC's constituted by an Ag tip pressed on the c plane of UPt_3 (1, $R_0=1.4$ Ω), $CeCu_2Si_2$ (2, $R_0=0.7$ Ω), and URu_2Si_2 (3, $R_0=0.1$ Ω). Curves a correspond to the NS characteristics ($B=0, T\sim 50$ mK) and curves b to the NN characteristics ($B\geq B_c, T\sim 50$ mK). The latter are not significantly different from characteristics recorded at $B=0$ for T just above T_c. The arrows in (2) and (3) indicate V_{thr}.

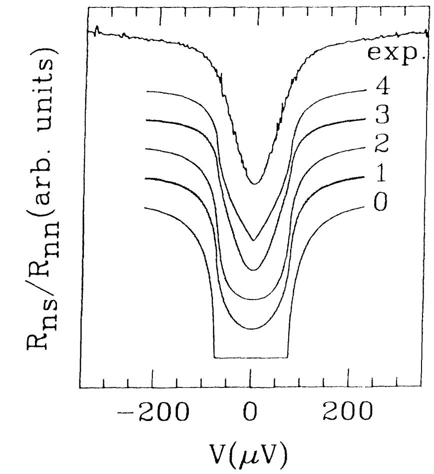

FIG. 2. Experimental (expt) R_{NS}/R_{NN} curve vs voltage V for the same Ag/UPt_3 PC as in Fig. 1.1 compared with computed curves for different symmetries. Curve 0: $\Delta=\Delta_0$ (isotropic case). Curves 1,3: $\Delta=2\Delta_0\hat{v}_z(\hat{v}_x+i\hat{v}_y)$. Curve 2: $\Delta=\Delta_0\hat{v}_z$. Curve 4: $\Delta=(3\sqrt{3}/2)\Delta_0\hat{v}_z(\hat{v}_x^2+\hat{v}_y^2)$. Curves 0,1,2,4: $\Delta_0=75$ μV, $Z=0$, and $m_\perp/m_\parallel=2.25$. Curve 3: $\Delta_0=75$ μV, $Z=0$, and $m_\perp/m_\parallel=10$.

the shape of the calculated characteristics compared to the case $Z=0$ [19].

For a nonspherical S_F, the dependence of Δ on $\hat{\mathbf{k}}$ given by group theory has to be replaced by a similar dependence on the components of the velocity $\hat{\mathbf{v}}_\mathbf{k}$ in order to include the structure in S_F [20]. Besides the isotropic symmetry $\Delta=\Delta_0$ (curve 0) we consider three simple possible gap symmetries for UPt_3 with nodes in the plane $\hat{k}_z=0$. The often proposed 2D even parity representation E_{1g} with symmetry $\Delta=2\Delta_0\hat{v}_z(\hat{v}_x+i\hat{v}_y)$ [4,21] with an equatorial line of nodes and two nodes at the poles, and two recently discussed 1D odd symmetries [9] belonging to the A_{2u} representation with one single equatorial line of nodes $\Delta=\Delta_0\hat{v}_z$, or combined with two additional nodes at each pole $\Delta=(3\sqrt{3}/2)\Delta_0\hat{v}_z(\hat{v}_x^2+\hat{v}_y^2)$. In each of these expressions, the maximum gap amplitude is equal to Δ_0. The best agreement with the experimental curve is obtained for $\Delta_0=75$ μV yielding a ratio $2\Delta_0/k_BT_c=3.96$ ($T_c\sim440$ mK) somewhat larger than the BCS weak coupling value 3.53. The symmetries given by $\Delta=2\Delta_0\hat{v}_z\times(\hat{v}_x+i\hat{v}_y)$ and $\Delta=\Delta_0\hat{v}_z$ yield bell shaped characteristics (curves 1 and 2) which are difficult to reconcile with the experimental curve with $m_\perp/m_\parallel\sim2.3$. A better agreement can only be obtained with $\Delta=2\Delta_0\hat{v}_z(\hat{v}_x+i\hat{v}_y)$ (curve 3) if $m_\perp/m_\parallel\geq10$, which seems hard to justify. The second 1D symmetry $\Delta=(3/\sqrt{3}/2)\Delta_0\hat{v}_z(\hat{v}_x^2+\hat{v}_y^2)$ yields a sharp structure around $V=0$ (curve 4) and gives a better qualitative agreement with the experimental curve although it would imply an inflection at $V=75$ μV which is not observed experimentally. In the real form for Δ in UPt_3 one has to include a function $f(\hat{\mathbf{k}})$ with the full hexagonal symmetry of the crystal [20], assumed here to be 1. An exact knowledge of $f(\hat{\mathbf{k}})$ is therefore necessary for a more conclusive determination of the gap symmetry.

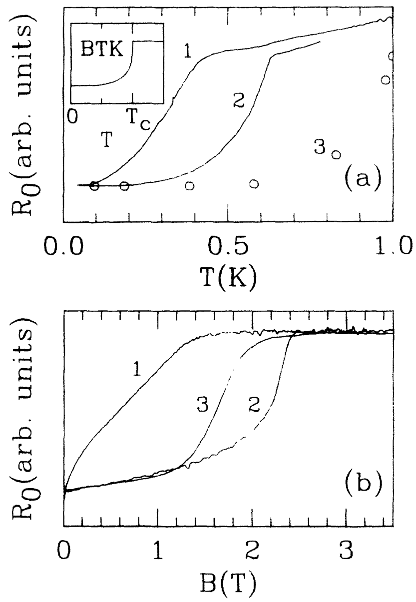

FIG. 3. Temperature (a) and magnetic field (b) dependence of the zero-bias resistance R_0 of PC's of Ag on UPt_3 (1, $R_0=1.4$ Ω, $T_c\approx440$ mK), $CeCu_2Si_2$ (2, $R_0=0.8$ Ω, $T_c\approx630$ mK), and URu_2Si_2 (3, $R_0=0.1$ Ω, $T_c\approx1$ K). The inset shows the temperature dependence of R_0 in the BTK theory for isotropic superconductors. The curves have relative scaling: $X_1=1.09$, $X_2=1.72$, $X_3=1.29$, $Y_1=1.10$, $Y_2=1.53$, $Y_3=1.14$.

The existence of thermally excited quasiparticles at E_F is also believed to cause the rise in the temperature dependence observed in the zero-bias resistance $R_0(T)$ for UPt_3 shown in Fig. 3(a), which begins at much lower temperatures than expected for an isotropic superconductor [13] whose $R_0(T)$ behavior is shown in the inset of Fig. 3(a). When T increases, the number of thermally excited quasiparticles at E_F also increases, which leads to a decrease in the probability of Andreev reflection and therefore to an increase in R_0.

The usual theory of isotropic type-2 superconductors predicts that the pair breaking effect of an external magnetic field B leads to a region of gapless superconductivity beyond a finite threshold field [22]. In the case of UPt_3 [Fig. 3(b)] the steep increase of R_0 starting at $B=0$ due the decrease of the Andreev reflection probability suggests that a regime of gapless superconductivity is directly reached once $B\neq0$.

For $CeCu_2Si_2$ and URu_2Si_2, the peaked structure in the NS differential resistance (Fig. 1) corresponds to the destruction of superconductivity in the contact area at high V. The peaked structure never develops below a finite threshold voltage V_{thr}. The observed flat structure in $R/R_0(V)$ for $V<V_{thr}$ would correspond to a nonvanishing Δ of minimum value eV_{thr}. For $E<eV_{thr}$, the Andreev-reflection process then permits only a nondissi-

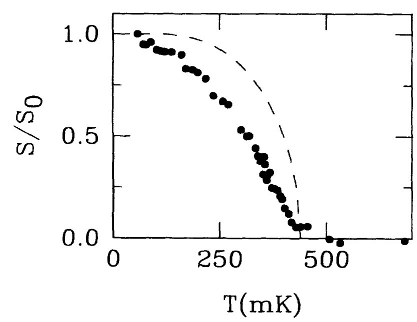

FIG. 4. Temperature dependence of the area S enclosed between $R(V)$ characteristics in the NS case and the corresponding NN background for UPt_3 normalized to its value S_0 obtained at the lowest temperature. $R_0=0.6\ \Omega$. The dashed curve represents the behavior of $\Delta(T)$ in the BCS theory.

pative current carried by Cooper pairs to flow in the superconductor. For $E>eV_{thr}$, inelastic scattering of the quasiparticle excitations within the PC leads to the local dissipation of excess electron energy with the destruction of the NS interface indicated by the peaks in $R_{NS}/R_{NN}(V)$. Such a mechanism for these two compounds is due to the shorter (inelastic) mean free path compared to UPt_3 for which energy dissipation occurs far from the PC.

For $CeCu_2Si_2$ and URu_2Si_2, $R_0(T)$ is shown in Fig. 3(a) and is in better agreement than for UPt_3 with both theoretical predictions [13], presented in the inset, and experimental observations [23] on isotropic superconductors. This also supports that in this case the injected electrons are probing a nonzero $\Delta(\hat{\mathbf{k}})$. A similar behavior is observed for $R_0(B)$ where the small slope at low fields compared with the case of UPt_3 suggests the absence of quasiparticles at E_F for $B=0$.

According to BTK theory R_{NS}/R_{NN} only depends on the ratio Δ/E. Therefore the area S enclosed between $R_{NS}(V)$ and the corresponding normal state background is directly proportional to the superconducting gap. Figure 4 shows the T dependence of the normalized area $S(T)/S(T=57\text{ mK})$ obtained from the $R_{NS}(V)$ characteristics of a PC on UPt_3 at different temperatures. $S(T)/S(T=57\text{ mK})$ decreases much more rapidly than expected for the superconducting gap of an isotropic BCS superconductor represented by the dashed curve. This can again be assigned to the presence of nodes in $\Delta(\hat{\mathbf{k}})$ as in the case of $R_0(T)$. For $CeCu_2Si_2$ and URu_2Si_2 the area $S(T)$ cannot be interpreted easily because of the normal state developing beyond V_{thr} in the superconductor.

In summary, we have observed the Andreev-reflection process by means of NS PC's produced on the c surface of the three HF superconductors UPt_3, URu_2Si_2, and for the first time $CeCu_2Si_2$. For UPt_3, the anomalous triangular shape of the $R(V)$ characteristics, the steep increase in the T and B dependence of the contact resistance R_0, and the rapid falloff of the deduced gap point very consistently to the existence of nodes in $\Delta(\hat{\mathbf{k}})$. Within the framework of a generalized BTK theory for anisotropic superconductors, the $R(V)$ curves observed in UPt_3 have been compared with three possible symmetries of the gap among which the 1D symmetry $\Delta=(3\sqrt{3}/2)\times\Delta_0\hat{v}_z(\hat{v}_x^2+\hat{v}_y^2)$ gave qualitatively the best agreement with $\Delta_0=75\ \mu\text{eV}$. However, the weighted averaging over k space in the expression for the current at the NS interface limits a conclusive determination of the anisotropy of Δ. For $CeCu_2Si_2$ and URu_2Si_2, the flat region in $R(V)$ around zero bias, without trace of dissipation, followed by the sudden destruction of the excess current I_{exc} at finite voltage as well as the behavior of $R_0(T)$ and $R_0(B)$ suggest that the injected electrons are probing a superconducting gap without nodes. Minimum values of 80 μeV for $CeCu_2Si_2$ and 170 μeV for URu_2Si_2 are deduced for Δ.

L. Taillefer would like to acknowledge the support of the Canadian Institute for Advanced Research.

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