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### Anisotropy of the superconducting energy gap in  $URu_2Si_2$ **studied by point-contact spectroscopy**

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PACS. 74.70Tx – Heavy-fermion superconductors. PACS. 73.40Jn – Metal-to-metal contacts. PACS. 74.70−b – Superconducting materials (excluding high- $T_c$  cuprates).

**Abstract.** – The differential resistance  $dV/dI$  vs. applied voltage V for  $URu_2Si_2$ –normal-metal point-contacts (PCs) is investigated. For some PCs a clear double-minimum structure is observed in  $dV/dI$  vs. V at zero bias well below the superconducting critical temperature  $T_c$  as expected for the Andreev reflection. The double minimum occurs only for PCs with suppressed antiferromagnetic (AF) features in  $dV/dI$ , in support of a recent model of the gap depression by AF order in the  $(a, b)$ -plane of tetragonal URu<sub>2</sub>Si<sub>2</sub>. A strong anisotropy of the reduced gap  $2\Delta/kT_c$  and of  $B_{c2}$  for PCs along the a and the c axis is also observed.

Introduction. – The possibility of unconventional superconductivity with strongly anisotropic order parameter in heavy-fermion superconductors (HFS) caused by  $p$ - or d-wave pairing has been under intense scrutiny during the last years [1]. Here the order parameter  $\Delta$  is expected to vanish on lines or points on the Fermi surface, which can explain the non-exponential temperature dependences of many physical properties of HFS in the superconducting (SC) state. The well-established power law behaviour of specific heat in URu2Si<sup>2</sup> [2] likewise indicates strong SC gap anisotropy. Direct information about the SC order parameter is accessible by means of point-contact (PC) spectroscopy via the mechanism of Andreev reflection (AR) at a normal-metal–superconductor (N-S) interface [3]. However, PC measurements on  $URu_2Si_2$  reported up to now are contradictory and not entirely conclusive. Hasselbach *et al.* and Samuely *et al.* [4], [5] found that their PC data were described slightly better with d-wave symmetry, while De Wilde et al. [6] from their data suggest the absence of zeros in the gap. These measurements either were carried out only for the c-direction in a single crystal [4], [6] or on a polycrystalline sample [5], which does not allow an unequivocal conclusion concerning gap anisotropy. Furthermore, these measurements did not show the unmistakable signature of AR, namely a double-minimum structure in the differential resistance  $dV/dI$ , which should be present whenever besides AR there is a finite probability of ordinary reflection [3].

As suggested by Brison *et al.* [7] the antiferromagnetic  $(AF)$  order in  $URu<sub>2</sub>Si<sub>2</sub>$  may lead to a line of zeros for ∆, independent of the pairing mechanism. In this case the gap vanishes for electrons with a wave vector **k** normal to the AF wave vector **Q** because of the pair breaking





Fig. 1. – Temperature (a)) and magnetic-field dependence at  $T = 50 \text{ mK}$  (b)) of normalized  $(dV/dI)^{\text{sym}}$  vs. V of a URu2Si2-Pt PC with  $R(0) = 2 \Omega$  established along the c-axis. The dashed line shows the difference between curves at lowest T and  $T > T_c$  to show that the double-minimum structure is not due to the broad maximum observed at  $T > T_c$ . The data have been symmetrized (see text). Insets: a) measured  $dV/dI$  curves below and above  $T_c$ , b)  $dV/dI$  (solid line) and its symmetric part (dashed line) on a larger voltage scale.

by the exchange field. Hence the SC gap should be maximal along the c-axis and vanish in the  $(a, b)$ -plane of the tetragonal URu<sub>2</sub>Si<sub>2</sub> lattice.

On the other hand, PC and tunnelling measurements of  $URu<sub>2</sub>Si<sub>2</sub>$  below the Néel temperature  $T_N = 17.5 \text{ K}$  were interpreted [8]-[10] in terms of an SDW gap opening in the AF state mainly in the  $(a, b)$ -plane. This should likewise hinder an SC gap formation in the tetragonal plane. The aim of this paper is to search for the anisotropy of the SC order parameter in a single crystal of  $URu<sub>2</sub>Si<sub>2</sub>$  and to investigate its relation with the AF order. In order to obtain reliable results, we focus on the few PC spectra where a clear double-minimum feature could be resolved. Here a strong anisotropy is found for directions along and perpendicular to the c-axis.

Experimental. – The URu<sub>2</sub>Si<sub>2</sub> single crystal of the present study was grown as described elsewhere [2]. The PCs were realized by pressing a 50 µm Pt wire against the freshly polished surface of the  $URu_2Si_2$  single crystal at room temperature, with the surface normal along the  $c$  or a axes. Then the PCs were mounted inside the mixing chamber of a  ${}^{3}$ He- ${}^{4}$ He dilution refrigerator to cool down. (In addition, a large number of PCs with Ag established at low T were investigated which, however, did not show a double minimum in the  $dV/dI$  curve.) The differential resistance  $dV/dI$  vs. voltage V was measured by the usual lock-in technique in the temperature range between 0.05 and 1.7 K. The applied magnetic field was parallel to the Pt tip, *i.e.* parallel to the c or a axes of the crystals. We assume that the preferred current direction is perpendicular to the interface, *i.e.* parallel to the "contact axis".

Results. – The vast majority of the dV/dI curves ( $\sim 95\%$ ) showed only one more or less pronounced minimum at zero bias, which vanished for temperature or magnetic field exceeding  $T_c$  or  $B_{c2}$  of URu<sub>2</sub>Si<sub>2</sub>, similar to previous studies [4]-[6], [11]. Of course, this



Fig. 2. – Temperature (a)) and magnetic-field dependence at  $T = 80 \text{ mK}$  (b)) of normalized  $dV/dI$ vs. V of URu<sub>2</sub>Si<sub>2</sub>-Pt PC with  $R(0) = 0.86 \Omega$  established along the a-axis. The data are not symmetrized. Insets: a) T-dependence of  $R(0)$  of this PC, b) symmetric part of  $dV/dI$  well below and above  $T_c$  on a larger voltage scale.

feature is related to the superconductivity of  $URu<sub>2</sub>Si<sub>2</sub>$ . However, there is no direct way to relate a zero-bias minimum to AR. Therefore, we focus on the few ( $\sim$  5%) PCs where a double-minimum structure is observed. The inset of fig. 1a) shows an example of  $dV/dI$ vs. V curves for  $T \geq T_c$  and  $T \leq T_c$  of a URu<sub>2</sub>Si<sub>2</sub>-Pt heterocontact with preferred current flow parallel to the c-axis, normalized to the zero-bias resistance  $R(0)$ . In general, the curves are asymmetric. The asymmetry has been discussed in detail elsewhere [8], [11] and will be disregarded in the following discussion. It will be subtracted from the data yielding  $(dV/dI)^{\text{sym}} = 1/2[(dV^+/dI) + (dV^-/dI)].$  At low temperatures and zero magnetic field a double-minimum structure about  $V = 0$  is clearly resolved in  $(dV/dI)^{sym}$ . The double minimum vanishes above  $T_c$  or  $B_{c2}$  (along the c-axis) of URu<sub>2</sub>Si<sub>2</sub> (fig. 1), showing that the structure is connected to superconductivity in  $URu<sub>2</sub>Si<sub>2</sub>$ .

It must be emphasized that this structure was observed only for PCs which did not exhibit a pronounced (up to ~ 10 % of  $R(0)$ ) maximum of  $(dV/dI)^{sym}$  between  $\pm 10$  mV, usually associated with an SDW gap formed on part of the Fermi surface of  $URu_2Si_2$  below the onset of AF order [8], [11]. This is illustrated in the inset of fig. 1 b) and fig. 2 b) where  $dV/dI$  is shown up to  $\pm 10$  mV. Conversely, when we found an AF maximum in  $(dV/dI)^{sym}$  at  $V = 0$  we never observed a double-minimum feature. Hence the double-minimum structure is very likely caused by AR at the N-S boundary with non-zero barrier strength  $Z$  at the N-S interface [3], under the conditions (not yet clear) when the SDW gap is suppressed in the contact region.

Figure 2 shows a corresponding series of measurements on a PC with the preferred current flow parallel to the  $a$ -axis. A clear anisotropy is observed. One can estimate roughly the magnitude of the SC gap  $\Delta$  from the location of the minima ( $\sim 0.5 \,\text{mV}$  for I  $\parallel$  c and  $\sim 0.2 \,\text{mV}$ for  $I \parallel a$ ) or, more accurately, from the fit procedure according to the modified BTK model invoking a phenomenological finite-lifetime broadening parameter  $\Gamma$  [12]. Such an analysis has been performed repeatedly, quite recently for  $UPt_3$  [13]. In the present case it yields  $2\Delta \sim 1.4$  meV or  $2\Delta/kT_c \sim 12$  for the c-direction and, correspondingly,  $2\Delta \sim 0.5$  meV or a



Fig. 3. – Comparison of the symmetrized  $dV/dI$  curves (data points) with the modifed BTK model (solid lines),  $T \simeq 50$ –80 mK. a) and b) fit of  $dV/dI$  curves for two PCs established along the c-axis with  $R(0) = 2 \Omega$  (PC from fig. 1) and 0.41  $\Omega$ , respectively. The fit parameters are: a)  $\Delta = 0.75$  meV,  $Z = 0.33$ ,  $\Gamma = 0.2$  meV, b)  $\Delta = 0.6$  meV,  $Z = 0.43$ ,  $\Gamma = 0.65$  meV (note that we need to scale curve a) by a factor of 28 to fit with the BTK model). c)  $dV/dI$  curve of the PC from fig. 2 established along the a-axis. The fit parameters are:  $\Delta = 0.25 \,\text{meV}$ ,  $Z = 0.33$ ,  $\Gamma = 0.25 \,\text{meV}$ .

reduced gap  $2\Delta/kT_c \sim 5$  for PCs along the a-axis. In both cases,  $T_c \sim 1.3$  K was assumed as determined from the vanishing of the SC structure in  $dV/dI$  or from the midpoint of the resistive transition for the PCs (see,  $e.g.,$  inset of fig.  $1\,a),\,2\,a).$ 

The B-dependence of the  $dV/dI$  curves for two directions in the  $URu_2Si_2$  single crystal shows a well-pronounced critical field anisotropy. Along the a-axis the SC structure is observed at least up to 8 T (fig. 2b), for other PCs up to 11 T), while  $B_{c2}$  along the c-axis is only about 2 T (fig. 1b), for other PCs up to 2.5-3 T). This is in agreement with the  $B_{c2}$  anisotropy for bulk samples, *i.e.*  $B_{c2} \simeq 14$  and 3 T along the a and the c axis, respectively [14].

Discussion. – In order to obtain the SC gap from  $dV/dI$  of an N-S contact using the BTK model, one should ascertain that AR is the dominant process responsible for the observed SC features. The correlation between the magnitude of the minimum at  $V = 0$  in the  $dV/dI$ curves of PCs with HFS and their electric resistivity reported by Gloos  $et \ al.$  [15] led these authors to suggest that the SC structure in  $dV/dI$  might be due to resistive transport of electrons through the PC. Indeed, a calculation of the  $dV/dI$  curves of S-S contacts assuming a growth of normal-metal regions in the PC with increasing dissipation when increasing V yields a minimum  $dV/dI$  at  $V = 0$  [16]. Thus, it is unclear which processes are responsible for the smooth SC minimum at  $V = 0$  below  $T_c$ . The peaked structure often observed at higher voltage above  $\Delta/e$  in N-S contacts for both classical and high- $T_c$  SCs, are not related to AR or other spectroscopic features and are more likely connected with critical current effects. In any case, one would expect that AR does occur at the N-S interface, with its relative contribution to the  $dV/dI$  curve compared to other effects being somewhat unpredictable. Therefore, the double-minimum structure at voltages corresponding to the expected SC gap is taken as the hallmark for AR at N-S boundary, since these features cannot be derived in any simple way from thermal or diffusive electron transport through the PC. Our  $dV/dI$  curves are indeed described quite well by the modified BTK model (fig. 3). While the contact resistance varies up to a factor of five in fig. 3*a*) and *b*), the measured gap differs by only  $\approx 20\%$ . This supports that the double-minimum structure does not originate from the resistive transport (or critical current effects) through the PC. The maxima above the double minimum suggest that not only AR affects the  $dV/dI$  curves but with increasing I other processes start to play an important role. The vanishing of the double-minimum structure with increasing  $T$  or  $B$ , *i.e.* transformation into a triangularly shaped minimum (fig. 2), might likewise suggest the destruction of superconductivity in the PC by a current with increasing T or B.

We now turn to the difference in the AR-related  $dV/dI$  features presented in fig. 1 and fig. 2 for the  $c$  and  $a$  axes, which suggests a sizable gap anisotropy. As mentioned above, the fitting of the  $dV/dI$  curves in fig. 3 yields the SC gap for the c-axis to be a few times higher than for the a-axis. This is in agreement with the idea [7] that the SC gap vanishes for quasiparticles moving in the tetragonal plane perpendicular to the AF wave vector **Q** because they experience a constant exchange field caused by AF order. For quasiparticles moving along the c-direction, *i.e.* along  $Q$ , the spatially oscillating exchange field suppresses the SC order parameter less because this field averages out on the scale of the SC coherence length. Thus, the SC gap is expected to be maximal for the c-direction. Furthermore, this theory predicts  $2\Delta/kT_c$  to be up to a few times larger than the usual BCS value 3.5 which is indeed observed. The question is why we observe the gap-like structure in the tetragonal plane at all (fig. 2). A possible explanation is found in the proposed model of the gap zero in  $URu_2Si_2$ . Clearly, the gap vanishes (in this model) only for quasiparticles moving almost exactly in the  $(a, b)$ -plane, *i.e.* their trajectory should not cross an adjacent plane (with antiparallel moments to the original plane) within a distance of roughly one SC coherence length. This is, of course, never fulfilled for a realistic PC. The situation is to be contrasted to that in  $UPt_3$ , where a symmetry-enforced line of the nodes in the hexagonal plane is actually widened to a 'belt' when impurities are present, hence AR is very small for current injection into the basal plane [17].

Of course, one has to consider the real structure of HFS–normal-metal interface, for instance surface roughness. This might also lead to a contact which is macroscopically aligned in one direction picking up features of other directions. Hence one can speak only about a preferred direction of the current flow with respect to the crystallographic axes. Furthermore, if the actual N-S boundary is located in the interior of  $URu<sub>2</sub>Si<sub>2</sub>$  somewhat remote from the constriction because of suppression of the superconductivity at the surface, the amount of backscattered holes through the constriction decreases, especially in a magnetic field [18]. This might be the reason for the observed small (only a few per cent) amplitude of the double-minimum AR structure and its vanishing in a magnetic field  $B \leq B_{c2}$ , see, e.g., fig. 2b). Another possibly related feature worth noting but not readily explained is that the AR feature (depth of the minimum) is actually smaller for the c-direction with larger  $\Delta$ .

The possibility of the N-S boundary being "buried" in the interior of  $URu<sub>2</sub>Si<sub>2</sub>$ , somewhat remote from the physical HFS–normal-metal boundary where SC and AF order are suppressed, might also account for the fact that the SDW feature is strongly suppressed for the present contacts, exhibiting the AR feature. This might be attributed to the different mechanism leading to features in the  $dV/dI$  characteristics: AR in the case of superconductivity and activation of charge carriers over the SDW gap. Of course, the corresponding length scales, roughly the SC coherence length and the mean free path, are also quite different, which will certainly affect the  $dV/dI$  characteristic of such a PC. Thus, the information about the SDW gap due to the quasiparticle tunnelling-like fraction of the total current appears to be lost, whereas AR from the N-S boundary, though diminished, is still visible, carrying the unperturbed information about the SC order parameter anisotropy.

Summary. – A pronounced anisotropy of the  $dV/dI$  characteristics for PCs of URu<sub>2</sub>Si<sub>2</sub> with the normal metal Pt for two directions, along the  $c$ -axis and along the  $a$ -axis, has been found. For PCs along the  $c$ -axis, the SC double-minimum structure is broader than for the perpendicular direction. This hints at a strong anisotropy of the SC gap in  $URu<sub>2</sub>Si<sub>2</sub>$ , caused probably by suppression of superconductivity in the  $(a, b)$ -plane because of the exchange field due to the ferromagnetically aligned layers of the U magnetic moments in the basal plane. First of all, the SC structure in the  $dV/dI$  curves disappears at  $B \approx 2.5$  T for B k c and above 11 T for B || a in accord with bulk  $B_{c2}$  measurements [14]. The absence of SDW-related features for the contacts exhibiting an AR double-minimum structure is possibly related to the different underlying mechanisms affecting the  $dV/dI$  curves.

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