Tunnelling Spectroscopy on the Heavy-Fermion Superconductors UPd2Al3 and UNi2Al3 in the Normal State

Aarts, J.; Volodin, A.P.; Menovsky, A.A.; Nieuwenhuys, G.J.; Mydosh, J.A.

Tunnelling Spectroscopy on the Heavy-Fermion Superconductors UPd_2Al_3 and UNi_2Al_3 in the Normal State

EUROPHYSICS LETTERS 20 April 1994

Europhys. Lett., 26 (3), pp. 203-208 (1994)

Tunnelling Spectroscopy on the Heavy-Fermion Superconductors UPd_2Al_3 and UNi_2Al_3 in the Normal State.

J. AARTS, A. P. VOLODIN (*), A. A. MENOVSKY
G. J. NIEUWENHUYS and J. A. MYDOSH

Kamerlingh Onnes Laboratory, Leiden University
P.O.Box 9506, 2300 RA Leiden, The Netherlands

(received 18 October 1993; accepted in final form 24 February 1994)

PACS. 74.70T – Heavy-fermion superconductors.
PACS. 73.40G – Tunnelling: general.
PACS. 74.70H – Magnetic superconductors (inc. reentrant (ferromagnetic) and antiferromagnetic superconductors).

Abstract. – Tunnelling measurements were performed on the new heavy-fermion superconductors UPd_2Al_3 (single crystal, $T_c = 1.8$ K) and UNi_2Al_3 (polycrystal, $T_c = 1.2$ K) and on the magnetic heavy-fermion superconductor URu_2Si_2 above and below the antiferromagnetic-ordering temperatures T_N for $T \geq T_c$. Tunnelling along the (a, b)-planes on UPd_2Al_3 shows that below $T_N \approx 14$ K an energy gap of about 13 meV is formed in the density of states. In contrast, no gap is found along the c-direction. For polycrystalline UNi_2Al_3 a gap of 10 meV is found below $T_N \approx 4.8$ K. Both the observed anisostropy and the values of the gaps of the new compounds prove surprisingly similar to URu_2Si_2. The values of the gaps appear to be set by crystal field excitation energies.

The ground state of heavy-fermion (HF) superconductors is crucially determined by the interplay between antiferromagnetic (AF) correlations and superconductivity [1,2]. Typically, AF ordering of very small moments is encountered at some low temperature T_N, followed by a transition to a superconducting state at a temperature T_c roughly an order of magnitude lower than T_N. Various manifestations of the ensuing coexistence may be found in the experimental behaviour. For instance, in UPt_3, at $T_N \approx 5$ K, hardly any changes occur in normal-state properties such as specific heat or resistance, and neutron scattering or muon procession must be used to detect the magnetic order. Around 0.5 K two transitions to a superconducting state show up in the specific heat, which are assumed to be due to the lifting of the degeneracy of an unconventional multicomponent order parameter by a coupling to the AF ordered state [1]. On the other hand, in URu_2Si_2 at $T_N \approx 17.5$ K, both specific heat and resistance exhibit sharp peaks [3,4]. These effects are related to an energy gap which opens on parts of the Fermi surface, as was indicated in optical-conductivity measurements [5]. Neutron scattering experiments show that at T_N a spin density wave is formed [6], and the

(*) Permanent address: Moscow Institute of Steel and Alloys.

spin wave excitation spectrum also displays a gap. The value of the latter is strikingly similar to the gap found in the optical conductivity (1), which indicates a strong coupling between the spin excitations and the charge degrees of freedom [7]. For URu_2Si_2 only one T_c is found around 1.2 K, although power laws in specific heat, penetration, depth etc. still suggest either a multicomponent or a highly anisotropic order parameter [8, 9].

Knowledge of gap formation in the normal state is crucial for a correct description of the superconducting state, since this bears on the important issues of the anisotropy and the symmetry of the superconducting order parameter (see, *e.g.*, ref. [10]). In the recently discovered HF superconductors UPd_2Al_3 ($T_N \approx 14.5$ K, $T_c \approx 2$ K) and UNi_2Al_3 ($T_N \approx \approx 4.6$ K, $T_c \approx 1$ K) [11, 12] coexistence of AF ordering and superconductivity also occurs, but it is not clear as yet whether the antiferromagnetism is accompanied by the formation of spin or charge density waves. Neutron scattering studies of UPd_2Al_3 [13] found a commensurate AF structure consisting of large U moments ($0.9\mu_B$) ferromagnetically aligned in the basal plane with the successive planes AF coupled. UNi_2Al_3 shows a similar structure with AF coupled planes, but with smaller moments in the plane ($0.24\mu_B$), which are incommensurate with the nuclear lattice [14]. Resistivity measurements on UPd_2Al_3 only show a downward kink at T_N instead of a rise and a maximum as in the case of URu_2Si_2.

A direct way to obtain information on the electronic density of states (DOS) is by tunnelling spectroscopy. Tunnelling has the advantage that mainly k-vectors perpendicular to the surface are probed, so that a high sensitivity to anisotropy can be expected. In URu_2Si_2, measurements based on STM (Scanning Tunnelling Microscopy) with the tip along the (a, b)-planes found a DOS gap with zero conductivity opening up below $T_N \approx 17.5$ K, but for the tip along the c-direction the conductivity was metallic, thereby for the first time clearly demonstrating the anisotropic nature of this gap [15]. We note here that (low resistance) point contact spectroscopy appears much less sensitive to the anisotropy. In URu_2Si_2, a strong dip in the differential conductance of point contacts was found below T_N around zero bias, with a temperature dependence similar to the spin wave gap seen in neutron scattering [16, 17]. However, this dip is not very sensitive to the crystal direction, and the superconducting gap was found to open within this dip. In this letter we present tunnelling measurements on single crystals of UPd_2Al_3 and URu_2Si_2, and on polycrystalline UNi_2Al_3. In all cases we find the opening of a gap below the antiferromagnetic transition, but for the single crystals only when tunnelling along the ab-direction, not along the c-direction. The maximum gap values are all of the order 10–15 mV.

Both single crystals were grown in a tri-arc furnace, as described in ref. [18] for UPd_2Al_3 and in ref. [19] for URu_2Si_2. The UPd_2Al_3 sample had a T_c of 2.0 K. Measurements were performed using an STM-based adjustable point contact with a tungsten tip which could be cooled down to 1.8 K. The samples were oriented with X-ray diffraction, cut by spark erosion, and sputter-etched by Ar ions in a UHV environment until the carbon Auger signal had disappeared. After mounting, measurements in air at room temperature typically showed a semiconducting-like gap of about 2 V for both crystal directions. Stabilizing the tip with a current of 1 nA at 3 V bias proved to be suitable for vacuum tunnelling, *i.e.* scanning over the surface was possible. We assume that the semiconducting property of the surface is due to its oxidation. For spectra near zero bias, the tip was stabilized with a much lower bias voltage, of the order of 50 mV, for which scanning was not possible. Probably, the tip now is in (weak) mechanical contact with the surface, making the contact equivalent to a small

---

(1) The values are equal and about 55 cm^{-1} = 1.67 THz = 80 K when comparing the optical conductivity with the mean gap in the spin wave density of states. The *minimum* spin wave excitation energy is much smaller, 0.5 THz = 24 K.

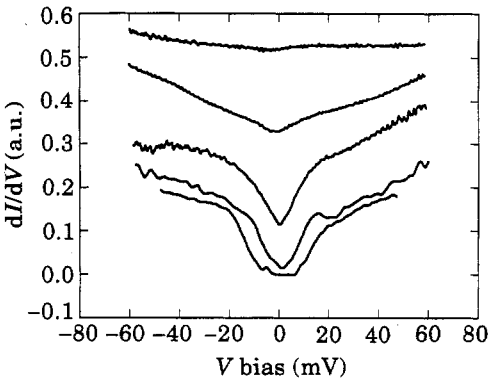

Fig. 1.

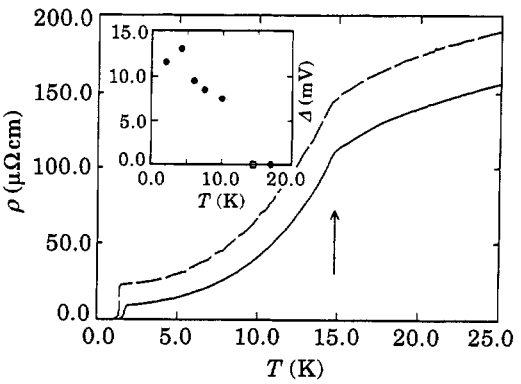

Fig. 2.

Fig. 1. – Differential-conductance traces of UPd_2Al_3 with the tip along the (a, b)-planes at temperatures (from top to bottom) $T = 30$ K, 17 K, 10 K, 7.5 K, 4.2 K.

Fig. 2. – Bulk resistivity of UPd_2Al_3 measured along ab (drawn line) and along c (dashed line). The arrow marks T_N. The inset shows $\Delta(T)$ as determined from the tunnelling measurements, with the open symbol denoting T_N.

planar junction. We still refer to these «tunnelling point contacts» as tunnelling, and define the resistance of the contact from the current at 100 mV. The differential-conductance traces presented below were obtained by numerical differentiation. Upon cooling to He temperature, scanning was usually not possible, but tunnelling-like resistance in the range 100 kΩ–100 MΩ could be attained controllably using the mechanical coarse approach and the piezo offset.

Typical differential-conductance traces (dI/dV *vs.* V) with the tip along the (a, b)-planes of the single crystal UPd_2Al_3 are shown in fig. 1. They were taken in different experiments at temperatures of 30 K, 17 K, 10 K, 7.5 K and 4.2 K and a contact resistance of approximately 10 MΩ. The temperature was unstabilized above 4.2 K and a slow drift was present, so that the above values are to within 0.5 K. In fig. 2 we show the temperature dependence of the bulk resistivity of the crystal for both directions up to 25 K; T_N is indicated by an arrow. The conductance spectra are V-shaped with finite conductance around zero bias down to 17 K. At 10 K, below T_N, downward kinks appear in the conductance, while at 4.2 K a clear plateau with a residual conductance of (nearly) zero is found. In fig. 3*a*) a comparison is made with URu_2Si_2 measured with the tip along the a-direction, also at 4.2 K. Figure 3*b*) shows measurements on the polycrystalline sample UNi_2Al_3 at 4.5 K and 1.8 K. The result on URu_2Si_2 also shows a zero-conductance plateau, while for UNi_2Al_3 it is clear that a gap starts to open around $T_N = 4.7$ K. A plateau is found at 1.8 K, although with larger residual conductance, which may well be due to the fact that the tunnelling direction is now undefined.

It must be remarked here that the shape of the spectrum depends on the contact resistance. Generally speaking, and in agreement with the report on URu_2Si_2 in ref. [15], at 100 kΩ we do not see a plateau, but only a strong dip with finite conductance at zero bias. In the range 1–20 MΩ we observe a zero-conductance plateau. At very high resistances the spectrum becomes parabolic and featureless. This diverse behaviour will be discussed elsewhere [20], but we believe that the window 1–10 MΩ gives a reliable representation of the energy gap. If we define the gap Δ as half of the spectrum width at half-height between the downward kink and the plateau, we find $\Delta(T = 0) = 9.5$ mV (equivalent to 110 K) for

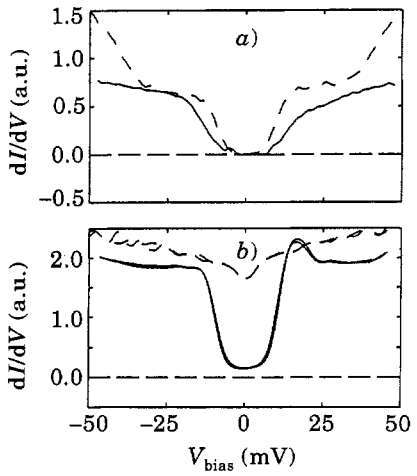

Fig. 3.

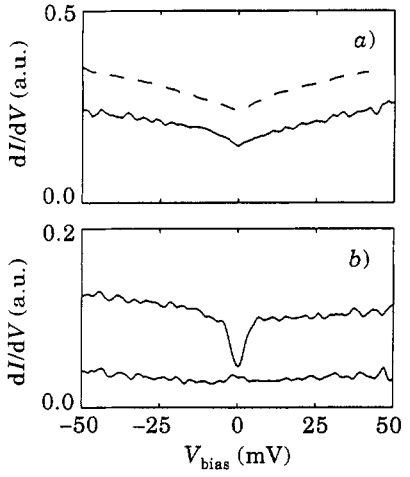

Fig. 4.

Fig. 3. – Differential conductance of *a*) UPd_2Al_3 (continuous line) and URu_2Si_2 (dashed line) at 4.2 K along the (*a*, *b*)-planes; *b*) polycrystalline UNi_2Al_3 at 4.5 K (dashed line) and 1.8 K (continuous line).

Fig. 4. – *a*) Differential conductance of UPd_2Al_3 (lower curve) and URu_2Si_2 (upper curve) at 4.2 K along the *c*-direction. The curve for URu_2Si_2 has been shifted upward by 0.1 unit for clarity. *b*) Two typical conductance traces on UPd_2Al_3 along the *c*-direction at 1.8 K.

URu_2Si_2, somewhat lower than the value of 15 mV estimated from the data given by Aliev *et al.* [15], but very similar to the mean gap in the spin wave spectrum found in neutron scattering ($\Delta = 80$ K [6, 7]) or resistivity ($\Delta = 90$ K [3]), and to the DOS gap inferred from specific heat ($\Delta = 129$ K [4]). The same analysis yields $\Delta(0) = 13$ mV (or 151 K) for UPd_2Al_3, which is now larger than the value of 40 K for the spin wave gap, extracted from resistivity measurements [21]. For UNi_2Al_3 at 1.8 K, $\Delta = 10$ mV (or 116 K). Finally, the insert of fig. 2 shows the temperature dependence of the gap for UPd_2Al_3.

In fig. 4*a*), spectra are shown with the tip along the *c*-direction at 4.2 K, for UPd_2Al_3 and for URu_2Si_2. The spectra are similar and V-shaped, and the conductance stays finite. Also, the spectra are very similar to those taken on UPd_2Al_3 along the *ab*-direction at temperatures higher than the ordering temperature (and for roughly the same contact resistance), as shown in fig. 2. Finally, the shape remains similar down to the lowest measured contact resistance of 100 kΩ. It is therefore reasonable to conclude that bulk properties are probed both along *ab* and along *c*, from which it follows that the DOS gap in UPd_2Al_3 is anisotropic, as it is in the case of URu_2Si_2. It is worth mentioning here that point-contact measurements in the low-Ohmic regime also give the same picture as for URu_2Si_2: a gap is found along *ab*, and a rather weaker gap structure is found along *c* [20], presumably because the point contact also samples other directions than the *c*-axis. Figure 4*b*) displays two conductance traces taken along the *c*-axis of UPd_2Al_3 at our lowest obtainable temperature of 1.8 K. They were recorded in different approaches and on different parts of the surface. One trace hardly shows any anomaly at zero bias, but the other exhibits a distinct and strong dip. Both types of curves were encountered several times. We assume that the dip signifies the onset of superconductivity, but that this is not found everywhere on the surface because we are too close to $T_c \approx 2$ K, which may vary for different parts of the surface.

From the above measurements, two facts clearly emerge. Firstly, both URu_2Si_2 and $U(Pd/Ni)_2Al_3$, with different crystal and magnetic structures, show the same gap anisotropy. Secondly, the zero-temperature values of the gaps are nearly equal. The underlying physics must therefore be similar, and the occurrence of gaps should be discussed in the framework of what is known for URu_2Si_2. With respect to the directionality, it is difficult to comment. In principle, the different periodicity of a spin density wave (SDW) or an AF ordered structure with respect to the periodicity of the crystal can cause gaps in the DOS due to the formation of superzone boundaries. The directionality of these gaps, however, will depend on the topology of the Fermi surface, knowledge of which is still incomplete [11, 22]. Comparing to the neutron scattering measurements of the spin wave gap, the situation is also complicated. In URu_2Si_2, a spin wave gap is found for all propagation directions. The excitations become strongly damped for momentum transfer along the *c*-direction [6], which is interpreted as a sign of strong coupling to the conduction electrons. If the tunnelling results indirectly show the spin wave gap, it is at the moment not clear why a gap is absent along *c*. UPd_2Al_3 has not been investigated as extensively as URu_2Si_2, but recent results indicate a spin wave excitation spectrum without a gap at the antiferromagnetic zone centre [23].

The values of our measured gaps are significantly larger than those expected for purely itinerant spin density waves, where the ratio $\Delta/k_B T_N$ is about 1.75 [24]. This ratio becomes 6.3, 10.5 and 24.2, for URu_2Si_2, UPd_2Al_3 and UNi_2Al_3, respectively. The gap is, therefore, set by another energy scale. In URu_2Si_2 the DOS gap appears equal to the spin wave gap and the latter is determined by the energy difference between the lowest crystal field level and the next higher level, connected by a non-zero matrix element for J_z (since the polarization is along *c*). These levels are two singlets with an energy difference of 115 K, very close to the value we extract for $\Delta(0)$. For UPd_2Al_3 it was recently found that the magnetic behaviour can also be described by tetravalent U and a crystal field splitting [25]. In this case the spin wave polarization is in-plane [26], and the relevant energy difference is for non-zero matrix elements of $J_{x,y}$, which is about 150 K and again surprisingly close to our value for $\Delta(0)$. In view of this, we would still suggest that spin and charge are strongly coupled, and that in an indirect way the zero-conductance gap in the *IV* spectra is a measure for the spin wave and, therefore, crystal field excitations.

We conclude by noting that the tunnelling technique, in contrast to the point-contact method, comprises a simple and direct method for measuring energy gaps in the density of states in different directions, which until now have remained elusive. Apart from the points discussed above, the results indicate that the superconducting order parameter for both URu_2Si_2 and UPd_2Al_3 is strongly anisotropic, since no superconducting gap can exist in the basal plane. This will be the subject of future low-temperature investigations.

* * *

We would like to thank H. NAKOTTE for help with the UNi_2Al_3 sample, S. A. M. MENTINK for characterisation and discussions, and J. ZAANEN and T. E. MASON for discussions. This work was supported by the Dutch Foundation for Fundamental Research of Matter (FOM) and the Dutch Foundation STW.

REFERENCES

[1] SIGRIST M. and UEDA K., *Rev. Mod. Phys.*, **63** (1991) 239.
[2] TAILLEFER L., FLOUQUET J. and LONZARICH G. G., *Physica B*, **169** (1991) 257.

[3] PALSTRA T. T. M., MENOVSKY A. A., V. D. BERG J., DIRKMAAT A. J., KES P. H., NIEUWENHUYS G. J. and MYDOSH J. A., *Phys. Rev. Lett.*, **55** (1988) 2727; PALSTRA T. T. M., MENOVSKY A. A. and MYDOSH J. A., *Phys. Rev. B*, **33** (1986) 6528.
[4] MAPLE M. B., CHEN J. W., DALICHAOUCH Y., KOHARA T., ROSSEL C., TORIKACHVILI M., MCELFRESH M. W. and THOMPSON J. D., *Phys. Rev. Lett.*, **56** (1986) 185.
[5] BONN D. A., GARRETT J. D. and TIMUSK T., *Phys. Rev. Lett.*, **61** (1988) 1305.
[6] BROHOLM C., LIN H., MATTHEWS P. T., MASON T. E., BUYERS W. J. L., COLLINS M. F., MENOVSKY A. A., MYDOSH J. A. and KJEMS J. K., *Phys. Rev. B*, **43** (1991) 12809.
[7] MASON T. E. and BUYERS W. J. L., *Phys. Rev. B*, **43** (1991) 11471.
[8] SCHLABITZ W., BAUMANN J., POLLIT B., RAUCHSCHWALBE U., MAYER H. M., AHLHEIM U. and BREDL C. D., *Z. Phys. B*, **62** (1986) 171.
[9] KNETSCH E. A., MENOVSKY A. A., PETERSEN J. J., NIEUWENHUYS G. J., MYDOSH J. A., SIGNORE P. J. C., BROWN S. E. and MEISEL M. W., *J. Magn. & Magn. Mater.*, **108** (1992) 71.
[10] OZAKI M. and MACHIDA K., *Phys. Rev. B*, **39** (1989) 4145.
[11] GEIBEL C., SCHANK C., THIES S., KITAZAWA H., BREDL C. D., BÖHM A., RAU M., GRAUEL A., CASPARY R., HELFRICH R., AHLHEIM U., WEBER G. and STEGLICH F., *Z. Phys. B*, **84** (1992) 1.
[12] GEIBEL C., THIES S., KACZOROWSKI D., MEHNER A., GRAUEL A., SEIDEL B., AHLHEIM U., HELFRICH R., PETERSON K., BREDL C. D. and STEGLICH F., *Z. Phys. B*, **83** (1991) 305.
[13] KRIMMEL A., FISCHER P., ROESSLI B., GEIBEL C., SCHANK C., GRAUEL A., LOIDL A. and STEGLICH F., *Z. Phys. B*, **86** (1992) 161.
[14] SCHRÖDER A., LUSSIER J. G., GAULIN B. D., GARRETT J. D., BRUYERS W. J. L., REBELSKY L. and SHAPIRO S. M., *Phys. Rev. Lett.*, **72** (1994) 136.
[15] ALIEV F. G., KOVACHIK V., MOSHCHALKOV V. V., PRYADUN V. V., ALEKSEEVSKII N. E., MITIN A. V., AGRAIT N., VIEIRA S. and VILLAR R., *J. Low Temp. Phys.*, **85** (1991) 359.
[16] HASSELBACH K., KIRTLEY J. R. and LEJAY P., *Phys. Rev. B*, **46** (1992) 5826.
[17] NOWACK A., NAIDYUK Y. G., CHUBOV P. C., YANSON I. K. and MENOVSKY A. A., *Z. Phys. B*, **88** (1992) 295.
[18] DE VISSER A., NAKOTTE H., TAI L. T., MENOVSKY A. A., MENTINK S. A. M., NIEUWENHUYS G. J. and MYDOSH J. A., *Physica B*, **179** (1992) 84.
[19] KNETSCH E. A., PETERSEN J. J., MENOVSKY A. A., MEISEL M. W., NIEUWENHUYS G. J. and MYDOSH J. A., *Europhys. Lett.*, **19** (1992) 637.
[20] VOLODIN A. P., AARTS J., MENOVSKY A. A., NIEUWENHUYS G. J. and MYDOSH J. A., *Physica B*, **194-196** (1994) 2033.
[21] DALICHAOUCH Y., DE ANDRADE M. C. and MAPLE M. B., *Phys. Rev. B*, **46** (1992) 8671.
[22] NORMAN M. R., OGUCHI T. and FREEMAN A. J., *Phys. Rev. B*, **38** (1988) 11193.
[23] MASON T. E., PETERSEN T., AEPPLI G., RAMIREZ A. P., BÜCHER E. and HUFNAGL J., *Proceedings of the International Conference on Strongly Correlated Electron Systems, San Diego 1993*, to be published.
[24] OVERHAUSER A. W., *Phys. Rev.*, **128** (1962) 1437.
[25] GRAUEL A., BÖHM A., FISCHER H., GEIBEL C., KÖHLER R., KOMATSUBARA T., MODLER R., SATO N., SCHANK C., STEGLICH F. and WEBER G., *Phys. Rev. B*, **46** (1992) 5818.
[26] MASON T. E., private communication.