Spin-Triplet Superconductivity in Sr_2RuO_4 Probed by Andreev Reflection

F. Laube,[1] G. Goll,[1] H. v. Löhneysen,[1] M. Fogelström,[2] and F. Lichtenberg[3]
[1]Physikalisches Institut, Universität Karlsruhe, D-76128 Karlsruhe, Germany
[2]Institut für Theoretische Festkörperphysik, Universität Karlsruhe, D-76128 Karlsruhe, Germany
[3]Experimentalphysik VI, Elektronische Korrelationen und Magnetismus, Institut für Physik, Universität Augsburg, D-86135 Augsburg, Germany
(Received 9 July 1999)

The superconducting gap function of Sr_2RuO_4 was investigated by means of quasiparticle reflection and transmission at the normal conductor-superconductor interface of Sr_2RuO_4-Pt point contacts. We found two distinctly different types of dV/dI vs V spectra either with a double-minimum structure or with a zero-bias conductance anomaly. Both types of spectra are expected in the limit of high and low transparency, respectively, of the interface barrier between a normal metal and a spin-triplet superconductor. Together with the temperature dependence of the spectra this result strongly supports a spin-triplet superconducting order parameter for Sr_2RuO_4.

PACS numbers: 74.70.Dd, 71.20.Lp, 73.40.Jn, 74.80.Fp

A superconductor is denoted as unconventional if below the transition temperature T_c additional symmetries, e.g., the time-reversal symmetry, are broken besides the gauge symmetry. Unconventional superconductivity (SC) is reflected by internal degrees of freedom of the order parameter (OP) which is determined by the probability amplitude for a Cooper pair at a given temperature T, and a nonphononic attractive pair interaction. In superfluid ^{3}He a spin-triplet pairing state is realized with parallel spin $S = 1$ and relative orbital momentum $l = 1$ of the Cooper pairs and the pair interaction is mediated by spin fluctuations [1]. A number of heavy-fermion systems also present evidence for unconventional SC [2]. In the high-T_c cuprates, the OP is predominantly singlet d wave ($l = 2$) with some s-wave admixture [3–5]. The layered superconductor Sr_2RuO_4 with T_c up to 1.5 K [6] is a prime candidate for p-wave SC in an electronic system as supported by remarkable properties of both the normal and superconducting states: (i) The related compounds $SrRuO_3$ and $Sr_3Ru_2O_7$ are both itinerant ferromagnets and spin fluctuations of predominantly ferromagnetic character were inferred from ^{17}O NMR measurements in Sr_2RuO_4 [7]. However, antiferromagnetic fluctuations were observed by inelastic neutron scattering [8]. (ii) Below $T \approx 25$ K, Sr_2RuO_4 reflects Fermi-liquid behavior in the thermodynamic and transport properties: the resistivity exhibits a T^2 dependence and both linear specific heat and Pauli spin susceptibility are enhanced by a factor of 3–4 with respect to the free-electron model [9]. (iii) The superconducting state is extremely sensitive to disorder and T_c is strongly suppressed by nonmagnetic as well as magnetic impurities [10]. (iv) Furthermore, the specific-heat data indicate a residual density of states in the superconducting state at least in some samples [11]. These hint at unconventional SC suggesting that Sr_2RuO_4 is an electronic analog of ^{3}He [12]. Indeed, a spin-triplet state has been identified by Knight-shift measurements [13], and the occurrence of a spontaneous magnetic field below T_c as revealed by muon spin rotation measurements [14] provides direct evidence for time-reversal symmetry broken SC. However, direct experiments to probe the anisotropic gap structure associated with p-wave SC have not yet been performed. Here we report on the investigation of the gap function of Sr_2RuO_4 by means of point-contact (PC) spectroscopy well below T_c. For current injection predominantly parallel to the ab plane we observe two distinctly different types of dV/dI vs V spectra: (i) a typical double-minimum structure seen in many superconductors and (ii) a single-minimum structure centered at $V = 0$, i.e., a zero-bias conductance peak. The zero-bias anomaly (ZBA) is probably caused by an Andreev bound state which is a signature of unconventional SC with a sign change of the pair potential as a function of $\boldsymbol{k}_F$. The different structures in dV/dI vs V might reflect the transition from a metallic to a tunneling PC for a p-wave superconductor. We note that Kondo-type scattering by magnetic impurities in the surface barrier can lead to a ZBA as well [15]. However, the fact that the observed ZBA vanishes at T_c renders this possibility unlikely.

The Sr_2RuO_4 single crystals were grown in air by a modified floating-zone melting process [16]. The superconducting transition temperature $T_c = 1.02$ K and the transition width $\Delta T_c^{10\%-90\%} = 35$ mK were determined from bulk resistivity measurements. No ferromagnetic impurity phases have been detected by x-ray powder diffraction and magnetization measurements with a SQUID magnetometer within the resolution of both methods. Heterocontacts between the superconducting sample (S) and a normal-metal (N) counterelectrode Pt were established with preferred direction of current injection parallel to the ab plane of tetragonal Sr_2RuO_4. The setup was mounted inside the mixing chamber of a ^{3}He/^{4}He dilution refrigerator. Mechanical feedthroughs allowed establishing and changing of Sr_2RuO_4-Pt PCs at low T.

The spectra, i.e., the differential resistance dV/dI as a function of applied bias V, were recorded by the standard lock-in technique.

Figure 1 shows four representative zero-field spectra at low $T \leq 0.2$ K $\ll T_c$. For comparison, the spectra have been normalized to the zero-bias resistance R_0. Almost 20 contacts were investigated. Typical R_0 values for stable contacts range from 0.1 to 40Ω. The main result is that two distinctly different types of spectra are observed. Curves 1 and 2 represent spectra which exhibit a double-minimum structure (type 1); curves 3 and 4 represent spectra which show a single minimum in dV/dI centered at $V = 0$, and shoulders symmetric in V at higher bias (type 2). The normal-state background of both types of curves is almost flat with no additional structure (see the inset in Fig. 1). Minima at finite V in the differential resistance are expected for N/S PCs due to Andreev reflection (AR) at the N/S interface for both conventional [17] and unconventional superconductors [18]. This scattering process where an electron is injected and a hole is retroreflected with probability R_A leads to a minimum in dV/dI vs V of width $\approx 2\Delta/e$. A finite but small probability $1 - R_A$ of normal reflection due to an interface barrier increases the zero-bias resistance and leads to the characteristic double-minimum feature. Phenomenologically, the barrier strength is modeled by a parameter Z which assumes a δ-functional barrier potential at the interface. $Z = 0$ corresponds to a pure metallic PC without interface barrier. With increasing Z gradually the tunneling limit is approached. Qualitatively, the same general behavior is expected for unconventional SC with a k-dependent gap function $\Delta = \Delta(\boldsymbol{k})$, although the "transparency" of the junction for AR has to be determined self-consistently to take into account that the interface itself might be pairbreaking for some OP symmetries.

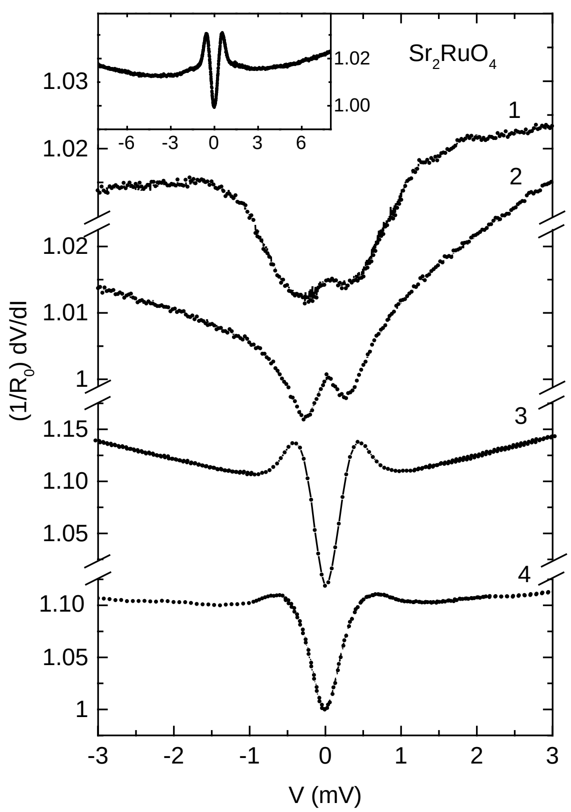

FIG. 1. Differential resistance dV/dI vs bias voltage V normalized to the zero-bias resistance R_0 measured at low $T = 0.2$ K $\ll T_c$ with $R_0 = 4.4$ (curve 1), 5.0 (curve 2), 5.3 (curve 3), and 3.6Ω (curve 4). The inset shows a dV/dI curve with $R_0 = 7.6\Omega$ to higher bias.

Recently, the conductance spectra of N/S junctions have been calculated for unitary and nonunitary spin-triplet pairing states [19,20]. Taking the quasi-two-dimensionality of the system into account the gap function parametrized by a vector function $\boldsymbol{d}(\boldsymbol{k})$ is given by one of the following functions:

$$\boldsymbol{d}(\boldsymbol{k}) \sim \hat{\boldsymbol{z}}(k_x + ik_y), \qquad (A)$$

$$\boldsymbol{d}(\boldsymbol{k}) \sim \hat{\boldsymbol{x}}k_x + \hat{\boldsymbol{y}}k_y, \qquad (B)$$

$$\boldsymbol{d}(\boldsymbol{k}) \sim (\hat{\boldsymbol{x}} + i\hat{\boldsymbol{y}})(k_x - ik_y). \qquad (C)$$

Of these three p-wave states, A and B are unitary. State C is a nonunitary state with the consequence that the excitation spectrum is gapped only for one spin direction. The pairing states A, B, and C are realized in the three superfluid phases A, B, and A_1 of ^{3}He, respectively, and are being discussed as possible candidates for the OP of Sr_2RuO_4 [12]. The key result of the calculations [19,20] is that for current injection into the ab plane two types of spectra can be obtained depending on Z, i.e., gaplike structures for low-Z contacts as found for many superconductors (type 1) and spectra with a ZBA for high-Z contacts (type 2). The ZBA is due to low-lying ($|\epsilon_b| \ll \Delta$) Andreev bound states at the surface caused by a sign change of the pair potential [21]. Our observation of two distinct types of spectra strongly supports the existence of an unconventional, probably spin-triplet, OP in Sr_2RuO_4. However, these angle- and spin-averaged curves gave no unambiguous criterion to discriminate between the different spin-triplet pairing states, since for all OP symmetries A, B, and C the theoretically predicted spectra look qualitatively the same, while quantitatively the gapless channel of the nonunitary state leads to a reduction of the SC-related features.

In order to quantitatively compare the theoretical spectra with our data and to extract the magnitude and the T dependence of the gap, $\Delta(T)$, one has to perform a more detailed calculation than the calculations [19,20] within a Blonder-Tinkham-Klapwijk model [17]. We solved the problem self-consistently for the surface state using weak-coupling quasiclassical theory [22], which allows conveniently to self-consistently include effects on the calculated spectra of bulk disorder, surface suppression of the OP, and surface quality. Given the smallness of the ratios of T_c to the Fermi temperature, $T_c/T_F \sim 10^{-4}$, and to the paramagnon temperature, $T_c/T_P \sim 10^{-2}$ [23], a weak-coupling theory should be a good approximation for

Sr_2RuO_4. A p-wave OP $\boldsymbol{d}(\boldsymbol{k}) \sim \hat{z}(k_x + ik_y)$ is assumed. The current transport across the PC is modeled allowing a tunable transparency [24]. A phenomenological acceptance cone, $\mathcal{D}(\phi) = \mathcal{D}_o \exp(-\lambda \sin^2\phi)$, puts emphasis on quasiparticle transmission through the PC at incidence angles, ϕ, within $\sin^2\phi \leq \lambda^{-1}$ of the contact normal. $\mathcal{D}_o$ is the transmission probability for quasiparticles along the contact normal. The signal of the measurements is only 1%–5% of the background conductance; hence the overall amplitude of the calculated spectra must be rescaled for the comparison to be made [25].

The measured spectra show SC-related structures up to $T \approx 1.3$ K, close to the SC onset determined resistively. This reduction with respect to the optimal T_c of 1.5 K can be attributed to nonmagnetic impurities limiting the quasiparticle mean free path to 15 coherence lengths ($\xi_o = 590$ Å) in the superconductor [10]. We first discuss the spectra for $T \ll T_c$. The existence of two types of spectra is instrumental in determining the zero-temperature energy gap, $\Delta(0)$. In this respect, it is reassuring for our assignment of p-wave SC that these two different types of spectra yield the same Δ_0 as will be discussed now. In a simple s-wave picture $\Delta(T)$ can be determined from the position of the minima in dV/dI (see the inset in Fig. 2). Independently of Z, the minima occur at $V \approx \pm\Delta/e$. However, the spectra of type 1 are much wider than expected for the s-wave case, although the double-minimum feature is reproduced quite well (see the inset in Fig. 2). Therefore, we first focus on the low-transmission curves 3 and 4 in Fig. 1. If the quasiparticle transmission is low and transport is restricted to small angles of incidence ϕ, the conductance spectrum is dominated by the Andreev bound states close to the Fermi level [21]. The width and height of the conductance peak depends on the acceptance cone as the position of the bound states disperses with ϕ as $\epsilon_b(\phi) \approx \Delta \sin\phi$ and the weight of the state as $w_b(\phi) \approx |\Delta| \cos\phi$. The position of the shoulders where dV/dI levels off towards larger V, on the other hand, does not depend on details of the acceptance cone. It is here where we can read off directly the magnitude of the energy gap Δ_0. The value $2\Delta_0/e = 2.2$ mV together with $\lambda = 24$ (reflecting the small angle of incidence) extracted from the calculations fit the measured spectra quite well; see the lowest curve in Fig. 3. Returning to the double-minima spectra, a fit with a high transmission ($D_0 \approx 1$) and a large acceptance cone ($\lambda = 1$) describes the data quite well, with the same value of 2.2 mV for $2\Delta_0/e$ (lowest curve in Fig. 2). This value, consistently extracted from both types of spectra, is 5 times the value expected from a weak-coupling theory. We remark that employing the double-minimum criterion for all spectra of type 1 on Sr_2RuO_4 the average width $2\Delta_0/e =$ 0.5 mV is inconsistent with the large value obtained. In addition, s-wave SC cannot account for the ZBA seen in spectra 3 and 4 in Fig. 1.

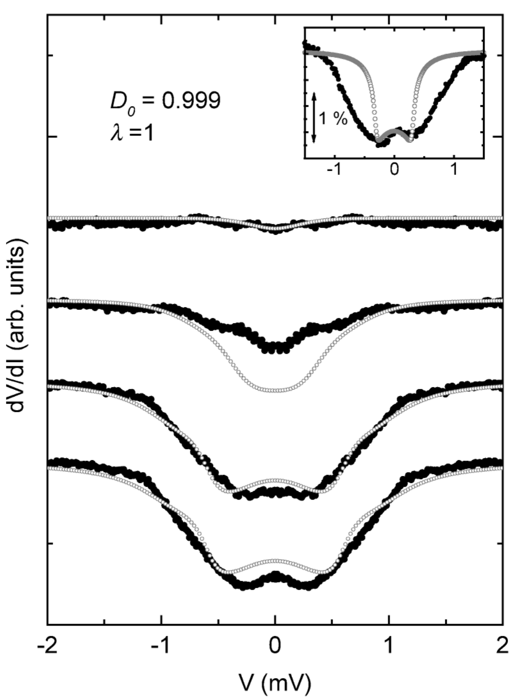

FIG. 2. Temperature dependence of the spectra with a double-minimum structure. Closed (open) symbols denote the measured (calculated) spectra. The temperatures T (in K, from bottom to top) are $T = 0.23, 0.40, 1.02$, and 1.25. For clarity, the curves at higher T are shifted with respect to the curve at lowest T. The inset shows the comparison of the curve at lowest T to a calculation within the Blonder-Tinkham-Klapwijk model with an isotropic gap $\Delta = 0.3$ meV and $Z = 0.2$.

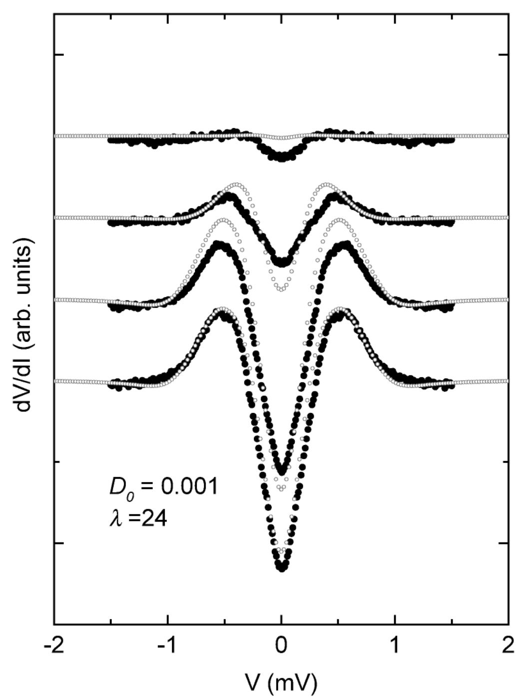

FIG. 3. Temperature dependence of the spectra with a single-minimum. Closed (open) symbols denote the measured (calculated) spectra. The temperatures T (in K, from bottom to top) are $T = 0.3, 0.41, 1.01$, and 1.26. For clarity, the curves at higher T are shifted with respect to the curve at lowest T.

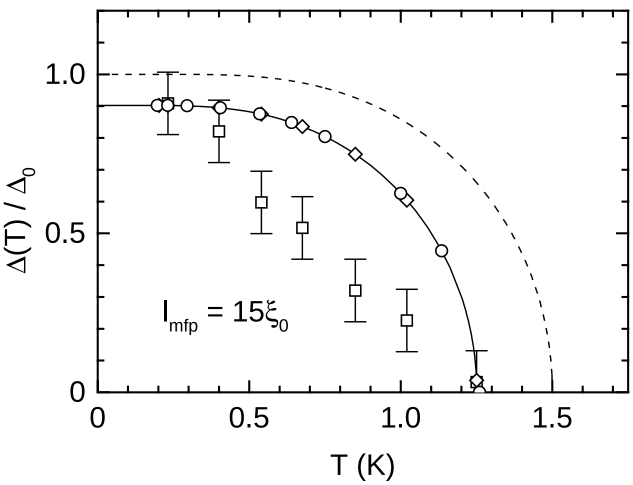

FIG. 4. Temperature dependence of the energy gap Δ assuming a p-wave state for curves of type 1 (diamonds) and type 2 (circles) in comparison with a T dependence extracted from the excess current (squares). The T dependence of a clean p-wave gap is indicated by the dashed line, the p-wave gap modified by impurities by the solid line.

The T dependence of both types of spectra is shown in Figs. 2 and 3 together with calculated spectra. In both cases, the SC-related features become weaker with increasing T and vanish near T_c. $\Delta(T)$ is calculated without additional parameter once Δ_0 has been determined by fitting to the spectra at the lowest T. The extracted $\Delta(T)$ behaves as the anisotropic gap modified by the influence of impurities (solid line), as displayed in Fig. 4. Note that the extracted $\Delta(T)$ is identical for both types of contacts. We mention that we have chosen the extreme cases $D_0 \approx 1$ and 0.001 as fitting parameter. Equally good fits are obtained with $D_0 \approx 0.9$ and 0.1, respectively. For a metallic PC, the excess current I_{exc} due to AR is proportional to Δ if one assumes an isotropic s-wave OP [26], and therefore the T dependence of I_{exc} should follow that of Δ. $I_{\mathrm{exc}}(T)$ normalized to the value $I_{\mathrm{exc}}(0)$ at lowest T for the spectra in Fig. 2 vanishes much faster than expected in a weak-coupling theory for an isotropic gap (Fig. 4) and again demonstrates the failure of an s-wave model for the SC in Sr_2RuO_4.

While we achieve a consistent description within p-wave pairing, we note that d-wave pairing as in cuprate superconductors can qualitatively account for a ZBA as well [27]. dV/dI spectra calculated in a $d_{x^2-y^2}$ scenario show a strong dependence on the relative crystal-to-junction orientation. In particular, junctions established mainly along the [100] crystal axis should show a largely increased dV/dI at low voltage compared to a sharp minimum along [110]. With the experimental technique employed by us the current is preferentially injected along different crystal axes depending on where the contact is made. However, we never observed a large $d_{x^2-y^2}$-derived dV/dI feature associated with the [100] direction. In order to further discriminate between a d- and p-wave OP, measurements in a magnetic field are underway.

In summary, directly probing the superconducting energy gap of Sr_2RuO_4 by means of PC spectroscopy gives strong support of an unconventional pairing state. The most convincing indication clearly comes from the occurrence of a zero-bias anomaly for contacts with a weakly transparent interface, in addition to the more conventional double-minimum features occurring for highly transparent interface. However, even for the latter type of contacts the shape of the spectra and their temperature dependence hint at unconventional superconductivity. A consistent zero-temperature gap and T dependence for both types of spectra is obtained within a model of p-wave pairing.

This work was supported by the Deutsche Forschungsgemeinschaft through Graduiertenkolleg “Anwendungen der Supraleitung.” The work of M. F. was financed by the EU TMR Network ERBFMRXCT96-0042. F. Li. acknowledges support by the BMBF Project No. 13N6918/1.

[1] D. Vollhardt and P. Wölfle, *The Superfluid Phases of ^{3}He* (Taylor&Francis, London, 1990).
[2] R. Heffner and M. Norman, Comments Condens. Matter Phys. **17**, 361 (1996).
[3] D. J. van Harlingen, Rev. Mod. Phys. **67**, 515 (1995).
[4] K. A. Kouznetsov *et al.*, Phys. Rev. Lett. **79**, 3050 (1997).
[5] J. Y. T. Wei *et al.*, Phys. Rev. Lett. **81**, 2542 (1998).
[6] Y. Maeno *et al.*, Nature (London) **372**, 532 (1994); Y. Maeno *et al.*, Phys. Rev. Lett. **81**, 3765 (1998).
[7] T. Imai, A. W. Hunt, K. R. Thurber, and F. C. Chou, Phys. Rev. Lett. **81**, 3006 (1998).
[8] Y. Sidis *et al.*, Phys. Rev. Lett. **83**, 3320 (1999).
[9] Y. Maeno and K. Yoshida, Czech. J. Phys. S6 **46**, 3097 (1996).
[10] A. P. Mackenzie *et al.*, Phys. Rev. Lett. **80**, 161 (1998).
[11] Y. Maeno, J. Low Temp. Phys. **105**, 1577 (1997).
[12] T. M. Rice and M. Sigrist, J. Phys. Condens. Matter **7**, L643 (1995).
[13] K. Ishida *et al.*, Nature (London) **396**, 658 (1998).
[14] G. M. Luke *et al.*, Nature (London) **394**, 558 (1998).
[15] E. L. Wolf, *Principles of Electron Tunneling Spectroscopy* (Oxford University Press, New York, 1985).
[16] F. Lichtenberg *et al.*, Appl. Phys. Lett. **60**, 1138 (1992).
[17] G. E. Blonder, M. Tinkham, and T. M. Klapwijk, Phys. Rev. B **25**, 4515 (1982).
[18] Chr. Bruder, Phys. Rev. B **41**, 4017 (1990).
[19] C. Honerkamp and M. Sigrist, J. Low Temp. Phys. **111**, 895 (1998).
[20] M. Yamashiro, Y. Tanaka, and S. Kashiwaya, Phys. Rev. B **56**, 7847 (1997).
[21] L. J. Buchholtz and G. Zwicknagl, Phys. Rev. B **23**, 5788 (1981).
[22] J. W. Serene and D. Rainer, Phys. Rep. **101**, 221 (1983).
[23] I. I. Mazin and D. J. Singh, Phys. Rev. Lett. **79**, 733 (1997).
[24] A. V. Zaitsev, Sov. Phys. JETP **59**, 1015 (1984).
[25] G. Goll, C. Bruder, and H. v. Löhneysen, Phys. Rev. B **52**, 6801 (1995).
[26] I. O. Kulik, Sov. J. Low Temp. Phys. **18**, 302 (1992).
[27] L. H. Greene *et al.*, Physica B (Amsterdam) (to be published).