Spin-triplet superconductivity in Sr$_2$RuO$_4$ 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

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The superconducting gap function of Sr$_2$RuO$_4$ was investigated by means of quasiparticle reflection and transmission at the normal conductor-superconductor interface of Sr$_2$RuO$_4$-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$_2$RuO$_4$.

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 non-phononic 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 [3,4]. The layered superconductor Sr$_2$RuO$_4$ with $T_c$ up to 1.5 K [5] 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$_2$Ru$_2$O$_7$ are both itinerant ferromagnets and spin fluctuations of predominantly ferromagnetic character were inferred from $^{17}$O NMR measurements in Sr$_2$RuO$_4$ [6]. However, antiferromagnetic fluctuations were observed by inelastic neutron scattering [7]. (ii) Below $T \approx 25$ K, Sr$_2$RuO$_4$ 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 3-4 with respect to the free-electron model [8]. (iii) The superconducting state is extremely sensitive to disorder and $T_c$ is strongly suppressed by nonmagnetic as well as magnetic impurities [9]. Furthermore, the specific-heat data indicate a residual density of states in the superconducting state at least in some samples [10]. These hints at unconventional SC suggesting that Sr$_2$RuO$_4$ is an electronic analogue of $^3$He [11]. Indeed, a spin-triplet state has been identified by Knight-shift measurements [12], and the occurrence of a spontaneous magnetic field below $T_c$ as revealed by $\mu$SR measurements [13] 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$_2$RuO$_4$ 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 $k_F$. 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 [14]. However, the fact that the observed ZBA vanishes at $T_c$ renders this possibility unlikely.

The Sr$_2$RuO$_4$ single crystals were grown in air by a modified floating-zone melting process [15]. The superconducting transition temperature $T_c = 1.02$ K and the transition width $\Delta T_c = 35$ 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$_2$RuO$_4$. The setup was mounted inside the mixing chamber of a $^3$He/$^4$He dilution refrigerator. Mechanical feedthroughs allowed establishing and changing of Sr$_2$RuO$_4$-Pt PCs at low T. The spectra, i.e. the differential resistance $dV/dI$ as a function of applied bias $V$, were recorded by standard lock-in technique.
eral behavior is expected for unconventional SC with a $k$-dependent gap function $\Delta = \Delta (k)$, 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.

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

$$d(k) \sim \hat{z}(k_x + ik_y) \quad (A)$$
$$d(k) \sim \hat{x}k_x + \hat{y}k_y \quad (B)$$
$$d(k) \sim (\hat{x} + i\hat{y})(k_x - ik_y) \quad (C)$$

Of these three $p$-wave states, $A$ and $B$ are unitary. State $C$ is a non-unitary 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$_2$RuO$_4$ \cite{14,20}. The key result of the calculations \cite{14,20} is that for current injection into the $ab$-plane two types of spectra can be obtained depending on $Z$, i.e. gap-like 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 ($|\epsilon| \ll \Delta$) Andreev bound states at the surface caused by a sign change of the pair potential \cite{21}. Our observation of two distinct types of spectra strongly supports the existence of an unconventional, probably spin-triplet, OP in Sr$_2$RuO$_4$. 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 non-unitary 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 \cite{14,20} within a Blonder-Tinkham-Klapwijk model \cite{17}. We solved the problem self-consistently for the surface state using weak-coupling quasiclassical theory \cite{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, $T_c/T_p \sim 10^{-2}$ \cite{23} a weak-coupling theory should be a good approximation for Sr$_2$RuO$_4$. A $p$-wave OP $d(k) \sim \hat{z}(\epsilon_x + i\epsilon_y)$ is assumed. The current transport across the PC is modeled allowing a tunable transparency \cite{24}. A phenomenological acceptance cone.

![Diagram](image_url)
$D(\phi) = D_o \exp(-\lambda \sin^2 \phi)$, puts emphasis on quasiparticle transmission through the PC at incidence angles, $\phi$, within $\sin^2 \phi \leq \lambda^{-1}$ of the contact normal. $D_o$ 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.

The measured spectra show SC-related structures up to $T \approx 1.3\, \text{K}$, close to the SC onset determined resistively. This reduction with respect to the optimal $T_c$ of 1.5 K can be attributed to non-magnetic impurities limiting the quasiparticle mean free path to 15 coherence lengths ($\xi_0 = 590\, \text{Å}$) in the superconductor. 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 $\Delta_0$ 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 inset of 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 inset of Fig. 2). Therefore, we first focus on the low-transmission curves 3 and 4 in Fig. 2. If the quasiparticle transmission is low and transport is restricted to small angles of incidence $\phi$, the conductance spectrum is dominated by the Andreev bound states close to the Fermi level. The width and height of the conductance peak depends on the acceptance cone as the position of the bound states disperses with $\phi$ as $\epsilon_a(\phi) \approx \Delta \sin \phi$ and the weight of the state as $w_a(\phi) \approx |\Delta| \cos \phi$. 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 $\Delta_0$. The value $2\Delta_0/e = 2.2 \, \text{mV}$ together with $\lambda = 24$ (reflecting the small angle of incidence) extracted from the calculations fit the measured spectra quite well, see lowest curve in Fig. 2. 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 $2\Delta_0/e$ for $2\Delta_0/e$ (lowest curve in Fig. 2). This value, consistently extracted from both types of spectra, is five 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$_2$RuO$_4$ the average width $2\Delta_0/e = 0.5 \, \text{mV}$ 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. 2.

![Fig. 2](image-url)  
**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 $\Delta = 0.3 \, \text{meV}$ and $Z = 0.2$.

![Fig. 3](image-url)  
**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 $T$. 

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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 $\Delta_0$ 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 $I_{\text{exc}}$ due to AR is proportional to $D$ if one assumes an isotropic $s$-wave OP [26], and therefore the $T$ dependence of $I_{\text{exc}}$ should follow that of $\Delta$. $I_{\text{exc}}(T)$ normalized to the value $I_{\text{exc}}(0)$ at lowest $T$ for the spectra in Fig. 3 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$_2$RuO$_4$.

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$_2$RuO$_4$ 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 hints 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.

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