Tunnel spectroscopy of superconducting oxide interfaces

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By joining LaAlO₃ and SrTiO₃, a conducting two-dimensional electron liquid (2-DEL) is formed at the contact area where these insulators meet [1]. At low temperatures this liquid is a two-dimensional superconductor [2] in which superconductivity surprisingly coexists with magnetic order [3–5]. Although interface superconductors are rare and have been recognized as candidates for superconductivity at very high temperatures (see, *e.g.*, [6, 7] for recent publications), knowledge of the superconducting phase at the LaAlO₃–SrTiO₃ interface is very limited. To gain direct information about this unusual two-dimensional superconductor, we explored it with the powerful method of tunneling spectroscopy. We hereby were able to measure the spectrum of the longitudinal (LO) optical phonons as well as the value of the superconducting gap.



Figure 1: Cross-sectional sketch of a typical sample. The tunnel junction is provided by the SrTiO₃–LaAlO₃–Au trilayer. The recessed Au/Ti plugs provide ohmic contacts to the 2-DEL. The Ag is used to apply a back-gate voltage to the SrTiO₃ substrate.

Figure 1 shows a cross-sectional sketch of the tunnel junctions that we designed and built for this purpose. The LaAlO₃ layers, which generate the electron liquid and at the same time are used as tunnel barriers, were deposited on TiO₂-terminated SrTiO₃ substrates. To achieve a sizable tunnel current, most of the LaAlO₃ films were grown to comprise only four monolayers of LaAlO₃. This is the minimal thickness necessary to induce the interface electron liquid. On top of this LaAlO₃ film, we deposited metallic layers of Au, AuPd, or YBa₂Cu₃O_{7-x}. Most of the film growth was performed by pulsed laser deposition (LaAlO₃, YBa₂Cu₃O_{7-x}) and rf-sputtering (Au) at Augsburg University. To minimize the density of interface states, all deposition processes were performed in situ. The samples were subsequently patterned via photolithography into ring-shaped tunnel junctions with outer diameters of between 0.4 and 1 mm. A photograph of a tunnel junction is shown in Fig. 2 and a transmission-electron-microscope cross-sectional image of another sample is shown in Fig. 3. In this sample design, electric fields generated by voltages applied to the back of the SrTiO₃ substrates can be used to tune the superconducting state by changing the carrier density of the two-dimensional electron system [8]. The measurements were performed in a dilution refrigerator at the Max Planck Institute for Solid State Research.

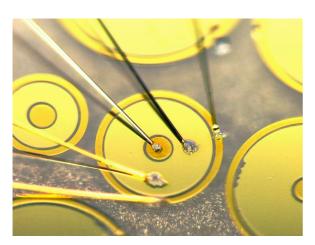


Figure 2: Optical micrograph of wire-bonded ring-shaped Au–LaAlO₃–SrTiO₃ tunnel junction. The narrow outer ring and the inner circle of the device are Au-covered Ti plugs to contact the 2-DEL; the broad golden ring between them is the Au top electrode of the tunnel junction. It has been deposited onto the LaAlO₃ thin film, which serves as a tunnel barrier between the top electrode and the 2-DEL. The outer ring has an outer diameter of 0.7 mm.

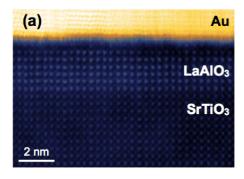




Figure 3: Scanning transmission electron microscopy image (high-angle annular dark field) of a sample comprising six monolayers of LaAlO₃ at high (a) and low (b) resolution (image: Lena Fitting–Kourkoutis, Cornell University).

The Au–LaAlO₃–SrTiO₃ samples with unpatterned LaAlO₃ films had superconducting interfaces. All of the five tunnel junctions grown on two different chips showed good tunnel characteristics. For this reason we will focus in the following on these samples. The origin of the non-superconducting behavior of the patterned samples and of many of the samples with AuPd electrodes remains to be explored.

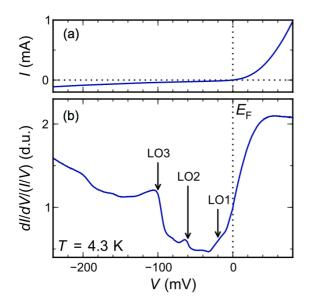


Figure 4: (a) I(V) tunnel characteristic of a typical sample measured at 4.3 K. The voltage characterizes the voltage applied to the interface 2-DEL. (b) Normalized differential conductance dI/dV/(I/V) measured in the normal-conducting state (4.3 K) and given in dimensionless units. The arrows mark the energies of the SrTiO₃ longitudinal optical (LO) phonons.

A typical I(V) characteristic taken at 4.3 K is shown in Fig. 4a and the corresponding characteristic of the normalized differential conductance dI/dV/(I/V) (V) in Fig. 4b. The polarity of the applied voltage was chosen to characterize the sign of the voltage applied to the interface. Thus, at positive voltages electrons tunnel into unoccupied states of the n-type 2-DEL. For V < 0 (electrons tunneling out of interface states) the dI/dV/(I/V) (V) characteristic is shaped by inelastic tunneling processes. In this case, the electron tunneling is assisted by SrTiO₃ longitudinal optical phonons, which generate peaks in the dI/dV/V curves at energies with large phonon density of states (DOS) as marked in Fig. 4b [9]. For V > 0 elastic tunneling prevails. This part of the spectrum reflects the 2-DEL's density of unoccupied states. The 2-DEL's DOS shows a strong energy dependence around the Fermi energy E_F , which resides only ~ 10 meV above of the conduction-band edge.

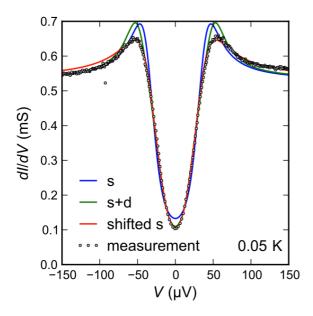
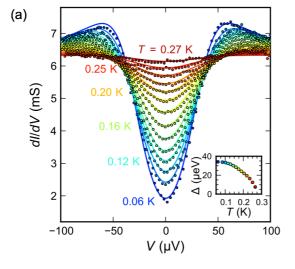


Figure 5: Typical dI/dV(V) characteristic of a Au-LaAlO₃-SrTiO₃ tunnel junction measured at low temperatures (50 mK). The blue and green lines show best fits to the characteristics expected for BCS-type s-wave superconductors and s+d wave superconductors. The best of the three fits is obtained using the s-wave BCS DOS plus a conductance shift of -0.35 mS.



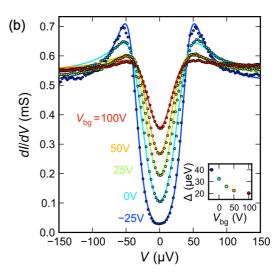


Figure 6 (a): Measured tunnel spectra as a function of temperature (circles) and best fits using the "shifted s" model (lines). The inset shows the temperature dependence of the superconducting gap Δ (T) obtained from the fits. The resistive T_c of this sample equals ~150 mK. The superconducting gap is still clearly visible at 250 mK. (b): tunnel spectra measured as a function of gate voltage applied to the back (bottom) side of the 1-mm-thick SrTiO₃ substrate (50 mK). The solid lines show best fits to the measured data (circles). The inset shows the gate-voltage dependence of the superconducting gap Δ (V_{bg}) obtained from the fits. Positive voltages enhance the 2-DEL's electron density, but reduce the gap Δ .

At low temperatures the tunneling provides direct information on the superconducting state. Figures 5 and 6 show a dI/dV(V) tunneling characteristic at 50 mK, well below the superconducting transition. The characteristic reveals a clear superconducting gap and allows the gap of the 2-DEL to be measured for the first time: it equals $\Delta (50 \text{ mK}) \sim 35 \,\mu\text{V}$. This value is derived from fitting the data with a modified BCS s-wave quasiparticle density of states as described below. The gap's temperature dependence is shown in the inset of Fig. 6a. The gap vanishes only at 280 mK and is therefore found well above the resistive T_c of 150 mK. This behavior agrees with the predicted behavior of a 2-D superconductor for which the transition into the superconducting state is a Berezinskii–Kosterlitz–Thouless (BKT) transition. Above the resistive T_c given by the BKT transition temperature and below the critical pair-condensation temperature $T_c \sim 280 \,\text{mK}$ the electrons are paired and a gap exists. However, fluctuating vortex–antivortex pairs cause the transport to be dissipative.

Surprisingly, we did not succeed in achieving a good fit of the $\mathrm{d}I/\mathrm{d}V(V)$ characteristics and of the $\Delta(T)$ dependence with the canonical BCS model for any order parameter symmetry. We are currently exploring whether this problem can be attributed to mundane effects such as measurement noise, whether it is caused by parallel tunneling into superconducting and normal domains, or whether it points to even more fundamental aspects of the 2-DEL's superconducting properties.

The device architecture developed offers the advantage that the tunneling spectra can be measured while applying a gate voltage $V_{\rm bg}$ to the back (bottom) side of the 1-mm-thick SrTiO₃ substrate. Because the gate voltage changes the carrier density of the 2-DEL, the dependence of the superconducting properties on the carrier density in the 2-DEL can be measured. Figure 6b shows the result of such a study done at 50 mK. Enhancing the carrier density in the 2-DEL by applying positive gate voltages causes a reduction of the superconducting gap Δ .

In summary, we studied the superconducting properties of the 2-D electron liquid at LaAlO₃–SrTiO₃ interfaces by means of tunneling spectroscopy. Our studies reveal a clear superconducting gap with a low temperature value of $\sim 35 \, \mu V$. The gap is present up to 280 mK, well above the resistive transition temperature of 150 mK. We have not succeeded in using the standard BCS theory to describe all of the data.

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