Evidence for superconducting phase coherence in the metallic/insulating regime of the LaAlO$_3$–SrTiO$_3$ interface electron system

E Fillis-Tsirakis$^1$, C Richter$^{1,2}$, J Mannhart$^1$ and H Boschker$^1$

$^1$ Max Planck Institute for Solid State Research, D-70569 Stuttgart, Germany
$^2$ Center for Electronic Correlations and Magnetism, Augsburg University, D-86135 Augsburg, Germany

E-mail: h.boschker@fkf.mpg.de

Keywords: superconductivity, tunneling spectroscopy, two-dimensional electron system

Abstract

A superconducting phase with an extremely low carrier density of the order of $10^{13}$ cm$^{-2}$ is present at LaAlO$_3$–SrTiO$_3$ interfaces. If depleted from charge carriers by means of a gate field, this superconducting phase undergoes a transition into a metallic/insulating state that is still characterized by a gap in the spectral density of states. Measuring and analyzing the critical field of this gap, we provide evidence that macroscopically phase–coherent Cooper pairs are present in the metallic/insulating state. This is characterized by fluctuating vortex–antivortex pairs, and not by individual, immobile Cooper pairs. The measurements furthermore yield the carrier–density dependence of the superconducting coherence length of the two-dimensional system.

1. Introduction

Two-dimensional (2D) electron systems are fascinating, including phenomena such as the quantum Hall effect [1, 2], ferromagnetism [3, 4] and superconductivity [5–9]. The 2D-superconducting state yields substantial critical temperatures despite the presence of phase and amplitude fluctuations of the order parameter. The large susceptibility to fluctuations results in many cases in a bosonic 2D superconductor-to-insulator (SIT) transition [9–15] or 2D superconductor-to-metal-to-insulator transition (SMIT) [16–26] with Cooper pairs existing in the insulating state [27]. These transitions are generally induced by tuning disorder or by changing the carrier density [28–41]. The LaAlO$_3$–SrTiO$_3$ interface 2D electron liquid (2DEL) [42, 43] is a 2D-superconductor that can be driven into an insulating state by depleting charge carriers with an electric field [44–48]. This superconductor is of great interest because the superconducting state exists at extremely small carrier densities.

The transition between the superconducting state and the metallic/insulating state in the LaAlO$_3$–SrTiO$_3$ 2DEL has been well established by transport measurements [45–53], however, with a different shape of the gate-voltage dependent $R(T)$ characteristics. Here $R$ is the sheet resistivity and $T$ is the temperature. Caviglia et al presented characteristics with $dR/dT < 0$ for gate voltages below a critical value and characteristics with $dR/dT > 0$ or a well defined zero resistivity state for gate voltages above this critical value. Intriguingly, at the critical value of the gate voltage, where $dR/dT = 0$, the sheet resistivity matches the quantum resistance of paired electrons $\hbar/4e^2$, [45, 49]. This indicates a bosonic SIT, with Cooper pairs present in the insulating state. Other experiments, however, offer a less clear picture. One experiment observed an SIT at a resistance value equal to one third of $\hbar/4e^2$, [50], and others observed characteristics with $dR/dT = 0$ for a large range of gate voltages [44, 51–53]. In these experiments there is no single separatrix between the superconducting and insulating state, similar to the SMIT. We therefore refer to the non-superconducting phase as the metallic/insulating phase. These metallic phases have been argued to be bosonic in nature [23].

Both scenarios suggest that Cooper pairs are present in the metallic/insulating state of the LaAlO$_3$–SrTiO$_3$ 2DEL. Furthermore, tunneling measurements find a superconducting gap in the density of states (DOS) across the transition [51]. Depending on their interaction, the Cooper pairs possibly form a macroscopic quantum-state characterized by an order parameter, a coherence length, and a critical magnetic field. It is also possible that the Cooper pairs act as single particles if their interaction is non-existent or weak. Which of the two scenarios
occurs in a superconductor with small carrier density is unknown, yet important for the general understanding of superconductivity. The two scenarios can be fundamentally different, as exemplified by their response to applied (perpendicular) magnetic fields $H$. If the Cooper pairs are phase coherent, the perpendicular upper critical field $H_c$ is determined by vortex behavior, whereas if the Cooper pairs are localized without phase coherence, $H_c$ is determined by pair breaking due to the Zeeman energy. In the above cases, $H_c$ differs considerably and for LaAlO$_3$–SrTiO$_3$ it equals 0.3 and 0.8 T, respectively, as discussed below. This difference opens a route to determine unequivocally the Cooper-pair nature of the metallic/insulating state, as $H_c$ can be well measured. To measure $H_c$ precisely, we use magnetic-field-dependent tunneling spectroscopy. Resistivity measurements also allow us to determine $H_c$, but are less stringent for interpreting in the metallic/insulating state. In tunneling spectroscopy measurements, the disappearance of the superconducting gap in the spectra quantitatively yields $H_c$ for the superconducting as well as for the metallic/insulating sides of the SMIT. As described in this letter, we observe $H_c$ values across the SMIT that are in clear agreement with the vortex-physics scenario. The data therefore provide conclusive evidence that the Cooper pairs in the metallic/insulating side are phase coherent on a length scale of at least the vortex size.

2. Experimental

To perform the tunneling spectroscopy measurements, planar junctions were realized, with device structures that are discussed in detail elsewhere [51]. A schematic of the junction structure is shown in figure 1. A Au top electrode is deposited on top of the four unit cell thick LaAlO$_3$ layer that simultaneously generates the 2DEL and acts as a tunnel barrier. Different Ti contacts to the 2DEL allow for four-point measurements of the tunneling current. The device area is approximately 1 mm. Tunnel spectra were obtained by applying a current between the top electrode and the interface and by simultaneously recording the tunneling voltage as well as the ac conductance using a standard lock-in technique. The polarity of the voltage characterizes the sign of the interface voltage with respect to the top electrode bias; for $V_0 < 0$ electrons tunnel out of the 2DEL. Measurements with these junctions resolved the superconducting gap of the 2D-state. The carrier concentration of the 2DEL was tuned electrically by a back-gate voltage $V_G$, allowing tunnel measurements across the entire superconducting dome (maximum $T_c \approx 300$ mK) as well as in the metallic/insulating state. Even in the insulating state, the minimal tunneling resistance significantly exceeds the maximal resistance of the 2DEL. All measurements were done at a temperature of $\sim 60$ mK.

3. Results

To measure $H_c$, the disappearance of the superconducting gap was analyzed as a function of magnetic field $H$. Figure 2 shows tunnel spectra as a function of $H$, with $V_G = 0$. In these tunnel junctions, the differential conductance reflects the DOS of the 2DEL, and the superconducting gap $\Delta$ is observed with a value of $\sim 60$ $\mu$V. The suppression of the DOS at $V = 0$ and the quasiparticle peaks disappear gradually with increasing $H$, as expected for type-II superconductors. The measured tunnel spectra represent a spatial average of the

\[ V_G \]

\[ I^+ \]

\[ I^- \]

\[ V^- \]

\[ V^+ \]

Figure 1. The schematic of a tunnel junction on the measured sample. In detail: the four unit-cell thick LaAlO$_3$ layer is shown in red, the millimeter-thick SrTiO$_3$ layer in light blue, the 2DEL in light green, the gold layers in yellow, dark green is for the titanium contacts to the 2DEL and gray is the silver backgate electrode. The device current and the gate voltage were applied with respect to the ground contact of the 2DEL and the device voltage was measured between the top-electrode and another 2DEL contact.

3 The SrTiO$_3$ substrate had previously undergone an isotope exchange, with $^{18}$O substituting for $^{16}$O. We did not see an effect of the exchange on the transport properties of the 2DEL [51, 54].
superconducting gap because the tunnel junctions are much larger than the vortex size. Therefore, for $H \ll H_c$, the tunnel spectra are magnetic-field independent as the volume fraction of the vortices is negligible. For larger values of $H$ that are still smaller than $H_c$, the measured spectra have reduced and broadened coherence peaks and a large conductance at $V = 0$, owing to a reduced superconducting gap across a significant fraction of the device area. Close to $H_c$, the vortices overlap and the maximum gap is reduced. Finally, for $H > H_c$, the spectra are weakly dependent on the magnetic field and the conductance has only a small reduction when $V \rightarrow 0$. This reduction is commonly attributed to the Altshuler–Aronov correction \cite{55} that accounts for electron–electron interactions. The Altshuler–Aronov correction can be easily distinguished from the superconducting gap, because it has a different energy scale (it persists up to approximately 1 meV) and a different $T$ and $H$ dependence.

We now turn to the carrier-density dependence of the critical field. The $dI/dV(V = 0)$ spectra for four different values of gate voltage are shown in figure 3. The four panels cover the entire density range, from the overdoped to the metallic/insulating side, as shown by the $R(T)$ characteristics of the 2DEL. In all cases the superconducting gap is present at 0 T. The gap increases with decreasing $V_G$, i.e., decreasing carrier concentration $n$. In addition, with the decrease of $n$, the normal-state conductance is also reduced. Even in the metallic/insulating state, clear quasiparticle coherence peaks are observed in the spectra. The applied magnetic field increases the conductivity at $V = 0$ and reduces the quasiparticle peaks. The superconducting gap features are no longer observed at fields larger than $T_0$ for all values of the gate voltage. The doping level also controls the $dI/dV(V = 0, H)$ behavior. With decreasing carrier density, larger magnetic fields are required to increase the $dI/dV(V = 0)$ conductance towards the normal-state level.

Figure 4 shows the $dI/dV(V = 0)$ as a function of $H$, in order to illustrate the transition to the normal state more precisely. For all gate voltages, $dI/dV(V = 0)$ increases until a plateau is reached. The plateau reflects the absence of the superconducting gap. By linearly extrapolating the curves at the steepest point of $dI/dV(V = 0, H)$ we determine $H_c$, as shown in figure 4. We now discuss the carrier density dependence of $H_c$. In figure 5(a) the measured values for $H_c$ are given as a function of the backgate voltage. The low temperature resistivity, at $H = 0$, of the 2DEL is shown as well, indicating the SMIT. $H_c$ monotonically increases with decreasing charge carrier density. Starting at 80 mT in the overdoped range, $H_c(V_G)$ reaches 300 mT in the underdoped range. Also in the metallic/insulating regime ($-200$ and $-300$ V), $H_c \approx 300$ mT.

![Figure 2](image_url)

Figure 2. The $H$ dependence of the $dI/dV(V)$ characteristics at zero back-gate field. Tunnel spectra were obtained at several values of $H$, with an incremental step of 0.025 T for $H < 0.2$ T; the larger values are 0.25, 0.3, 0.4, 0.5, 0.75 and 1 T.
4. Discussion

Now we compare the critical fields with the Chandrasekhar–Clogston critical field \( H_{cc} \) and the one induced by vortices. For the latter case, \( H_c \) is obtained from the Ginzburg Landau coherence length \( \xi \) by

\[
H_c = \frac{\Phi_0}{2\pi \xi^2},
\]

where \( \Phi_0 = h/2e \). In the BCS model, \( \xi \) is related to the superconducting gap by \( \xi = \frac{\hbar v_f}{\pi \Delta} \). Here, \( v_f \) is the Fermi velocity. The Ginzburg Landau coherence length is equal to the BCS coherence length for superconductors that are in the clean limit. In the case of the LaAlO\(_3\)–SrTiO\(_3\) superconductor, the coherence length is of similar magnitude to the transport scattering length (20–100 nm \([58–60]\)) \( \approx 100 \text{ nm} \). We therefore expect the BCS coherence length to be similar to the Ginzburg Landau coherence length. To calculate \( \xi \), we determine \( H_c \) from the tunnel spectra at zero applied magnetic field. It ranges from 37 to 66 \( \mu \text{eV} \). Equating \( H_c \) to the measured \( H_c \) in the superconducting region \( \approx 100 \text{ V} \leq V_G \leq 300 \text{ V} \), we determine \( v_f \) to be \( 1.1 \times 10^4 \text{ m s}^{-1} \), thus it is found to be independent of the gate voltage. This value is smaller than the theoretically predicted \( 7 \times 10^4 - 5 \times 10^5 \text{ m s}^{-1} \) \([65]\). We note that the chemical potential changes only by a small amount in this gate voltage range \([54]\), thus \( v_f \) is also expected to show only small changes. The good agreement between \( H_c \) and measured \( H_c \) also holds in the metallic/insulating state for which no resistive superconducting transition is observed.

4.1. Gate-voltage dependence of the tunnel spectra measured at 60 mK, as a function of magnetic field \( H \). The superconducting gap for gate voltages of −200, −100, 0, 200 V is suppressed at \( H = 300 \pm 50 \text{ mT} \), \( 300 \pm 50 \text{ mT} \), \( 215 \pm 30 \text{ mT} \) and \( 90 \pm 20 \text{ mT} \), respectively. The error margins were derived by analyzing a series of spectra spaced from 20-50 mT (not shown here). In the metallic/insulating state the superconducting gap and the coherence peaks are visible even at gate voltages for which no resistive superconducting transition is observed.

4.2. The \( dI/dV(V = 0, H) \) characteristics for \( V_G \) ranging from 300 to 300 \( \mu \text{V} \). \( H_c \) is derived as illustrated.
standard g-factor of 2 is assumed. With $\Delta = 66 \mu eV$ (the constant value of the gap in the metallic/insulating region) this ratio yields $H^\text{CC}_{c} = 0.8$ T. This value does not include the spin–orbit coupling [66, 67] and is therefore a lower-limit estimate of the upper critical field due to depairing.

The previous analysis yields an accurate determination of the in-plane coherence length $\xi$ as a function of gate voltage. It ranges from 34 to 65 nm across the phase diagram, figure 5(b). These values for $\xi$ are lower than those reported in previously published work [50, 52, 61–64], which are typically around 70 nm, increasing with increasing carrier density [63]. The main reason is due to the difference of the methods used to determine $H^\text{BCS}_{c}$. In the present work, the disappearance of the superconducting gap is used as a criterion to identify $H^\text{BCS}_{c}$. In earlier work, $H^\text{BCS}_{c}$ has been determined as the magnetic field at which the sample resistance equals 50 % of the normal state resistance. In electron systems with a broad superconducting transition, the latter criterion underestimates $H^\text{BCS}_{c}$, resulting in correspondingly larger values of the coherence length.

5. Conclusion

The measured critical field $H_{c} \approx 0.28$ T is consistent only with the value expected for the suppression of a vortex phase; the localized Cooper pair scenario does not match the measured data. Because the coherence length $\xi$ also evolves gradually from the macroscopically phase-coherent, superconducting state to the metallic/insulating state, the measurements provide evidence that the electron system in the metallic/insulating state consists of one or more ensembles of macroscopically phase-coherent Cooper pairs. This understanding is corroborated by the existence of the coherence peaks in the tunnel spectra in the metallic/insulating state [68]. The data exclude an electron system consisting solely of superconducting puddles with a length scale of $l < \xi$.

Acknowledgments

We thank M Etzkorn for helpful discussions.

References

[1] Klitzing K v, Dorda G and Pepper M 1980 Phys. Rev. Lett. 45 494
[2] Tsukazaki A, Ohtomo A, Kita T, Ohno Y, Ohno H and Kawasaki M 2007 Science 385 1388
[3] Oja R et al 2012 Phys. Rev. Lett. 109 127207
[1] Vaz C A F, Bland J A C and Laudoff G 2008 Rep. Prog. Phys. 71 056051
[2] Gozar A, Logvenov G, Fitting Kourkoutis L, Bollinger A T, Gianuzzi I A, Muller D A and Bozovic I 2008 Nature 455 782
[3] Reyren N et al 2007 Science 317 1196
[4] Ge J, Liu Z-L, Liu C, Gao C-L, Qian D, Xue Q-K, Liu Y and Jia J-F 2014 Nat. Mater. 14 285
[5] Zhang W H et al 2014 Chin. Phys. Lett. 31 017401
[6] Goldman A M and Markovic N 1998 Phys. Today 51 39
[7] Shalimov A I 1938 Nature 142 74
[8] Haviland D B, Liu Y and Goldman A M 1989 Phys. Rev. Lett. 62 2180
[9] Valles J M, Dynes R C and Garner J P 1992 Phys. Rev. Lett. 69 5567
[10] Yazdani A and Kaptitulnik A 1995 Phys. Rev. Lett. 74 3037
[11] Fisher M P A, Grinstein G and Girvin S M 1990 Phys. Rev. Lett. 64 587
[12] Lee P A and Ramakrishnan TV 1985 Rev. Mod. Phys. 57 287
[13] Ephron D, Yazdani A, Kaptitulnik A and Beasley M R 1996 Phys. Rev. Lett. 76 1529
[14] Mason N and Kaptitulnik A 1999 Phys. Rev. Lett. 82 5341
[15] Seo Y, Qin Y, Vicente C L, Choi K S and Yoon J 2006 Phys. Rev. Lett. 97 057005
[16] Qin Y, Vicente C L and Yoon J 2006 Phys. Rev. B 73 100505(R)
[17] Humbert V, Couedo F, Crauste O, Berge L, Drillon A, Marrache-Kikuchi C A and Dumoulin L 2014 J. Phys.: Cond. Mat. 26 456205
[18] Li Y, Vicente C L and Yoon J 2010 Phys. Rev. B 81 020505(R)
[19] Sacepe B and Kim E 2014 arXiv:1401.3947
[20] Phillips P and Dalidovich D 2003 Science 302 243
[21] Jaeger H M, Haviland D B, Goldman A M and Orr B G 1986 Phys. Rev. B 34 17
[22] Mason N and Kaptitulnik A 2001 Phys. Rev. B 64 080504(R)
[23] Christiansen C, Herran I M and Goldman A M 2002 Phys. Rev. Lett. 88 037004
[24] Fisher M P, A, Weichman P B, Grinstein G and Fisher D S 1989 Phys. Rev. B 40 546
[25] Baturina T, Mironov A Y, Vinokur V M, Balakonov M R and Strunk C 2007 Phys. Rev. Lett. 99 257003
[26] Sacepe B, Chapelier C, Baturina T, Vinokur V M, Balakonov M R and Sanquer M 2010 Nat. Commun. 1 140
[27] Dubi Y, Meir Y and Avishai Y 2007 Nature 449 876
[28] Biscaras J, Bergeal N, Hurand S, Feuillet-Palma C, Rastogi A, Budhani R C, Grilli M, Caprara S and Lesqueur J 2013 Nat. Mater. 12 542
[29] Stewart M D Jr, Yin A, Xu J M and Valles J M Jr 2007 Science 318 1273
[30] Barber R P Jr, Merchant L M, La Porta A and Dynes R C 1994 Phys. Rev. B 49 3409
[31] White A E, Dynes R C and Garner J P 1986 Phys. Rev. B 33 3549
[32] Holten S M, Nguyen H Q, Rudisuela E, Stewart M D Jr, Shainline J, Xu J M and Valles J M Jr 2011 Phys. Rev. B 84 045128
[33] Eshoven D S, Slazouhi E and Froynan A 2011 Phys. Rev. B 84 014529
[34] Sherman D, Koppov G, Shadir G and Dynes A M 2012 Phys. Rev. Lett. 108 177006
[35] Noat Y et al 2013 Phys. Rev. B 88 014503
[36] Bollinger A T, Dubuis G, Yoon J, Pavuna D, Misiewich J and Bozovic I 2011 Nature 472 458
[37] Shi X, Logvenov G, Bollinger A T, Bozovic I, Panagopoulos C and Popovic D 2012 Nat. Mater. 11 47
[38] Chand M, Saraswat G, Kamnapure A, Mondal M, Kumar S, Jusdasan J, Bagwe V, Benfatto L, Tripathi V and Raychaudhuri P 2012 Phys. Rev. B 85 014508
[39] Ohtomo J, Hwang H Y 2004 Nature 427 423
[40] Breitschaft M et al 2010 Phys. Rev. B 81 153414
[41] Hurand S et al 2015 Sci. Rep. 5 12571
[42] Caviglia A D, Gariglio S, Reyren N, Jaccard D, Schneider T, Gabay M, Thiel S, Hammerl G, Mannhart J and Triscone J-M 2008 Nature 456 624
[43] Schneider T, Caviglia A D, Reyren N, Jaccard D, Schneider T and Triscone J-M 2009 Phys. Rev. B 79 184502
[44] Mehta M, Dikin D A, Bark C W, Ryu S, Folkman C M, Eom C B and Chandrasekhar V 2014 Phys. Rev. B 90 100506(R)
[45] Caprara S, Bucheli D, Scopigno N, Biscaras J, Hurand S, Lesueur J and Grilli M 2015 Solid State Commun. 246 84
[46] Lin Y-H, Nelson J and Goldman A M 2015 Physica C 514 130
[47] Dikin D A, Mehta M, Bark C W, Folkman C M, Eom C B and Chandrasekhar V 2011 Phys. Rev. Lett. 108 177006
[48] Richter et al 2013 Nature 502 528
[49] Herranz G et al 2015 Nat. Commun. 6 6028
[50] Bell C, Harashima S, Kozuka Y, Kim M, Kim B G, Hikita Y and Hwang H Y 2009 Phys. Rev. Lett. 103 226802
[51] Boschke H, Richter C, Fillis-Tsirakis E, Schneider C W and Mannhart J 2015 Sci. Rep. 5 12309
[52] Altshuler B and Avron A 1971 Solid State Commun. 10 115
[53] Chandrasekhar B S 1962 Appl. Phys. Lett. 17
[54] Clugston A M 1962 Phys. Rev. Lett. 9 266
[55] Mannhart J and Scholl D G 2010 Science 327 1607
[56] Ben Shalom M, Tai C W, Lereah Y, Sachs M, Levy E, Rakhmilevich D, Palevski A and Dagan Y 2009 Phys. Rev. B 80 140403
[57] Huijben M, Rijnders G, Blank D H A, Bals S, van Aert S, Verbeeck J, van Tendeloo G, Brinkman A and Hilgenkamp H 2006 Nat. Mater. 5 566
[58] Reyren N, Gariglio S, Caviglia A D, Jaccard D, Schneider T and Triscone J-M 2009 Appl. Phys. Lett. 94 112506
[59] Gariglio S, Reyren N, Caviglia A D and Triscone J-M 2009 J. Phys. Condens. Matter 21 164213
[60] Ben Shalom M, Sachs M, Rakhmilevich D, Palevski A and Dagan Y 2010 Phys. Rev. Lett. 104 126802
[61] Mehta M M, Dikin D A, Bark C W, Ryu S, Folkman C M, Eom C B and Chandrasekhar V 2012 Nat. Commun. 3 955
[62] Nakamura Y and Yanase Y 2013 J. Phys. Soc. Japan 82 083701
[63] Caviglia A D, Gabay M, Gariglio S, Reyren N, Caviglia A D and Triscone J-M 2010 Phys. Rev. Lett. 104 126803
[64] Kim M, Kozuka Y, Bell C, Hikita Y and Hwang H Y 2012 Phys. Rev. B 86 085121
[65] Sacepe B, Dubouchet T, Chapelier C, Sanquer M, Ovadia M, Shahrar D, Feigelman M and Ioffe I 2011 Nat. Phys. 7 239