Observation of transient superconductivity at the LaAlO3/SrTiO3 interface

Gopi Nath Daptary, Shelender Kumar, and Aveek Bid*
Department of Physics, Indian Institute of Science, Bangalore 560012, India

Pramod Kumar, Anjana Dogra, and R. C. Budhani
National Physical Laboratory, New Delhi 110012, India

Dushyant Kumar
Condensed Matter-Low Dimensional Systems Laboratory, Department of Physics, Indian Institute of Technology Kanpur, Kanpur 208016, India

N. Mohanta
Center for Electronic Correlations and Magnetism, Theoretical Physics III, Institute of Physics, University of Augsburg, 86135 Augsburg, Germany

A. Taraphder
Department of Physics, Indian Institute of Technology Kharagpur, West Bengal 721302, India

(Received 26 August 2015; revised manuscript received 3 April 2017; published 1 May 2017)

We report the observation of a magnetic-field-assisted transient superconducting state in the two-dimensional electron gas existing at the interface of LaAlO3/SrTiO3 heterostructures. This metastable state depends critically on the density of charge carriers in the system. It appears concomitantly with a Lifshitz transition as a consequence of the interplay between ferromagnetism and superconductivity and the finite relaxation time of the in-plane magnetization. Our results clearly demonstrate the inherently metastable nature of the superconducting state competing with a magnetic order in these systems. The coexistence of superconductivity and ferromagnetism in the conducting electronic layer formed at the interface of insulating oxides has thrown up several intriguing and as yet unanswered questions. An open question in this field is the energetics of the interplay between these two competing orders and the present observation goes a long way in understanding the underlying mechanism.

DOI: 10.1103/PhysRevB.95.174502

I. INTRODUCTION

The mutual interplay of point group symmetry, charge inversion symmetry, U(1) gauge symmetry, and spin rotation symmetry in heterostructures of complex perovskite oxides [1] leads to the coexistence of a host of intriguing properties: ferroelasticity, ferroelectricity, superconductivity, and ferromagnetism [2,3]. Superconductivity and magnetism are generally considered to be incompatible with each other and hence reports of the observation of a possible coexistence of these two phases in the conducting electronic layer formed at the interface of two insulating oxides LaAlO3 and SrTiO3 [1–6] has opened up a new direction of research in condensed matter physics. In this paper, we report the observation of a magnetic-field-assisted transient superconducting state (TSS) at the interface of LaAlO3 and SrTiO3 at 245 mK. The TSS appears concomitantly with a Lifshitz transition in the system as a consequence of the interplay between ferromagnetism and superconductivity and the finite relaxation time of in-plane magnetization. To the best of our knowledge such a transient superconducting state has not been observed in condensed matter physics. Despite intensive research over the last decade [2] the coexistence of superconducting and ferromagnetic phases in this system is still debatable. There is now overwhelming evidence that superconductivity in LaAlO3/SrTiO3 is mediated by phonons [7] and is conventional-BCS-like [8]. Scanning superconducting quantum interference device (SQUID) measurements have revealed that the superconductivity in these systems is probably spatially inhomogeneous [9] although more recent experiments may suggest otherwise [10]. Direct measurements of the magnetization in this system have yielded contrasting results. On one hand torque measurements show a large in-field magnetization of 0.3–0.4 μB per interfacial Ti ion [11]. On the other hand scanning SQUID experiments show that there are only spatially inhomogeneous patches of local moments with no net magnetization [9]. Although various scenarios have been invoked to reconcile these apparently contradictory experimental observations [12–16] a clear picture of the magnetization behavior of this system is yet to emerge.

When the thickness of the LaAlO3 layer grown over the TiO2-terminated SrTiO3 layer exceeds 4 unit cells, 0.5 electrons per unit cell are transferred from the top layer of LaAlO3 to the Ti3+ ions at the interface to avoid a polar catastrophe [5] resulting in a quasi-2-dimensional electron gas (2DEG) at the interface. Hall measurements suggest that a very small fraction of these electrons actually takes part in transport; it is believed that most of them get localized in the d-t2g orbitals of the Ti atoms at the interface because of strong on-site Hubbard and nearest-neighbor Coulomb repulsive interactions forming the local moments responsible for ferromagnetism [17]. The breaking of mirror inversion symmetry at the interface lifts the degeneracy of the t2g levels of Ti ions at

*aveek.bid@physics.iisc.ernet.in
the interface [18] with the $d_{xy}$ level having a lower energy than the $d_{xz}$ and $d_{yz}$ orbitals [19]. At low number densities all the conduction electrons occupy the lower lying $d_{xy}$ orbitals at the interface [2]. It has recently been proposed that when the number density $n_e$ of itinerant electrons exceeds a certain critical value the system undergoes a Lifshitz transition at which point the $d_{xz}/d_{yz}$ bands near the interface begin to get occupied. The system now effectively has two types of carriers: a high-density electron gas residing in the $d_{xy}$ orbital and a lower-density high-mobility electron gas occupying the $d_{xz}/d_{yz}$ orbitals [20–23]. An additional parameter controlling this system is the strong Rashba spin-orbit coupling (SOC) arising due to the broken inversion symmetry at the interface which allows the electronic properties of the system to be modulated over a large range by means of a gate-voltage-induced electric field [24]. Oxygen vacancies, controlled by the O$_2$ partial pressure during the deposition of SrTiO$_3$, are also believed to play a crucial role in determining the magnetic and electrical transport properties of this system [25,26].

II. RESULTS

A. Sample preparation

Our measurements were performed on samples with 10 unit cells of LaAlO$_3$ grown by pulsed-laser deposition (PLD) on TiO$_2$-terminated (001) SrTiO$_3$ single-crystal substrates of thickness 0.5 mm [27]. As-received SrTiO$_3$ substrates were pretreated with standard buffer HF solution [28] in order to achieve uniform TiO$_2$ termination which was confirmed from atomic force microscopy measurements. Prior to deposition, the treated substrates were annealed for an hour at 830 °C in oxygen partial pressure of $7.4 \times 10^{-2}$ mbar to remove any moisture and organic contaminants from the surface and also to reconstruct the surface so that pure TiO$_2$ termination was realized. This was followed by the deposition of 10 unit cells of LaAlO$_3$ at 800 °C at an oxygen partial pressure of $1 \times 10^{-4}$ mbar. Growth with the precision of a single unit cell was monitored by the oscillation count using an in situ RHEED gun. Postdeposition, the samples were cooled at the same O$_2$ partial pressure at the rate of 10 °C/min to the ambient temperature. The epitaxial nature of the films was confirmed by HRXRD performed on a 20 u.c. LaAlO$_3$ film grown under identical conditions on TiO$_2$-terminated SrTiO$_3$ which allowed us to measure the $c$-axis lattice parameter of LaAlO$_3$. The thickness of one unit cell from these measurements came out to be 3.75 Å [27].

A schematic of the device structure is shown in Fig. 1. Electrical contacts were created on top of the LaAlO$_3$ substrate by thermal evaporation of 5 nm Cr followed by 100 nm of Au and were wire bonded to the measurement chip carrier. A gold film deposited on the back side of the SrTiO$_3$ substrate acted as one plate of the capacitor while the conducting layer acted as the other plate of the capacitor for electrostatic gating of the device. The SrTiO$_3$ substrate acted as the gate dielectric material. Measurements were performed on five different samples grown under similar conditions; they differed only in their carrier concentrations at zero gate voltage. All the devices showed qualitatively the same behavior. In this paper we present the results of detailed measurements on two devices, S2 and S5, with S2 having a slightly lower sheet number density of charge carriers ($n_e \approx 1.65 \times 10^{13}$ cm$^{-2}$ at 250 mK) as compared to S5 ($n_e \approx 2 \times 10^{13}$ cm$^{-2}$ at 250 mK). The measurements were performed down to 245 mK in a He-3 refrigerator and down to 10 mK in a dilution refrigerator.

B. Resistance and magneto resistance

The sheet resistance $R_{\text{sheet}}$ of device S2 as a function of temperature at different gate voltages $V_g$ is shown in Fig. 2(a). The superconducting transition temperature $T_C$ and the normal state resistance were both found to depend sensitively on the gate voltage. The $T_C$ (defined as the temperature where resistance drops to 50% of its normal state resistance) increases as the system is progressively electron doped [see Fig. 2(b)] in conformity with previous observations in similar systems [22,29,30].

The magneto-resistance data for magnetic fields applied perpendicularly to the interface measured at a few representative values of $V_g$ are shown in Fig. 3(a) for device S2. The measurements were performed at 245 mK where the device is in the normal state at all measured gate voltages [shown by the dotted white line in Fig. 2(b)]. We notice a distinct change in the nature of the magneto-resistance curves as $V_g$ changes from a large negative value to a large positive...
FIG. 3. (a) Magnetoresistance of device S2 at different gate voltages ranging from $-200\,\text{V}$ to $170\,\text{V}$ (in steps of $20\,\text{V}$ from $-200\,\text{V}$ to $-150\,\text{V}$ and subsequently in steps of $10\,\text{V}$ from $-150\,\text{V}$ to $170\,\text{V}$) measured at $245\,\text{mK}$. The TSS state appears for values of gate voltages $V_g > V^*_g$. (b) Magnetoresistance at gate voltage $V_g = -200\,\text{V}$ showing hysteresis at low magnetic fields. The arrows denote the direction of magnetic field sweep. (c) Hysteresis in magnetoresistance as a function of gate voltage and magnetic field at $245\,\text{mK}$. Inset: A log-log plot of the same data as plotted in (b) to show the detailed evolution of the TSS with magnetic field. (d) Magnetoresistance of device S5 at different values of $V_g$ at $245\,\text{mK}$; the data were acquired as the $B$ field was swept down from $8\,\text{T}$ at a rate $dB/dt = 1\,\text{T/min}$. The values of $V_g$ range from $210\,\text{V}$ to $-10\,\text{V}$ in steps of $20\,\text{V}$. Inset: Hall data obtained close to $B = 0\,\text{T}$ at $210\,\text{V}$ as the magnetic field was swept down to $0\,\text{T}$ at a rate $d\mathbf{B}/dt = 1\,\text{T/min}$. The magnetoresistance in the low doping regime has been seen previously in LaAlO$_3$/SrTiO$_3$ heterostructure devices and was taken to indicate the presence of ferromagnetic domains in the system [30,31], there is a growing concern in the community that it might also have contributions from induction effects due to fast magnetic field sweeps. We do not discuss further the data in this region of doping, leaving it for further experimental analysis.

C. Magnetic-field-assisted transient superconductivity

For $V_g > V^*_g$, the magnetoresistance is positive as the magnetic field is swept from $0\,\text{T}$ to $8\,\text{T}$. This change from a positive magnetoresistance to a negative magnetoresistance around a certain value of $V_g$ has been observed before in LaAlO$_3$/SrTiO$_3$ heterostructures and has been interpreted to be due to a transition from weak localization to weak antilocalization mediated by the large Rashba SOC present in this system [24]. As the magnetic field is swept back down towards $0\,\text{T}$, the magnetoresistance curve retraces itself till about $20\,\text{mT}$ below which the sheet resistance jumps down by more than four orders of magnitude and the system goes superconducting with the resistance becoming smaller than our measurement resolution. The data from typical measurements are plotted in Figs. 4(a) and 4(b) for devices S2 and S5, respectively. In the inset of Fig. 4(c) we have replotted the data from device S5 in the low-field regime to emphasize the precipitous drop in the sheet resistance. [The corresponding Hall data are shown in the inset of Fig. 4(d).] Note that in the absence of magnetic field, the devices are in a nonsuperconducting state with $R \approx 425–475\,\Omega$. The superconducting state thus reached is transient and relaxes back to the original zero-field resistive state with a time decrease and eventually vanish at around the critical gate voltage $V_g = 110\,\text{V}$ [see Fig. 3(c)]. Although hysteresis in magnetoresistance in the low doping regime has been observed previously in LaAlO$_3$/SrTiO$_3$ heterostructure devices and was taken to indicate the presence of ferromagnetic domains in the system [30,31], there is a growing concern in the community that it might also have contributions from induction effects due to fast magnetic field sweeps. We do not discuss further the data in this region of doping, leaving it for further experimental analysis.

FIG. 4. (a) Magnetoresistance of device S2 measured at gate voltage $V_g = 150\,\text{V}$ and temperature $245\,\text{mK}$ showing the appearance of the TSS. (b) Magnetoresistance of device S5 measured at gate voltage $V_g = 210\,\text{V}$ and temperature $245\,\text{mK}$ showing the appearance of the TSS. In both (a) and (b) the magnetic field was swept down from $B = 8\,\text{T}$ at a rate $dB/dt = 1\,\text{T/min}$. (c) Time relaxation of the TSS for the two devices (green curve: device S2, $V_g = 135\,\text{V}$; red curve: device S5, $V_g = 210\,\text{V}$) measured after the magnetic field was swept down to $0\,\text{T}$ at a rate $dB/dt = 1\,\text{T/min}$. The measurements were performed at $245\,\text{mK}$. Inset: A log-log plot of the same data as plotted in (b) to show the detailed evolution of the TSS with magnetic field. (d) Magnetoresistance of device S5 at different values of $V_g$ at $245\,\text{mK}$; the data were acquired as the $B$ field was swept down from $8\,\text{T}$ at a rate $dB/dt = 1\,\text{T/min}$. The values of $V_g$ range from $210\,\text{V}$ to $-10\,\text{V}$ in steps of $20\,\text{V}$. Inset: Hall data obtained close to $B = 0\,\text{T}$ at $210\,\text{V}$ as the magnetic field was swept down to $0\,\text{T}$ at a rate $d\mathbf{B}/dt = 1\,\text{T/min}$. 

174502-3
constant of around 10 seconds [see Fig. 4(c)]. In Fig. 4(d) we show a plot of the magnetoresistance for device S5 at different values of \( V_g \); the data were acquired as the \( B \) field was swept down from 8 T. It can be seen that the TSS state appears only for values of gate voltage \( V_g > V_g^* \). This condition for the observation of the TSS held true even for temperatures quite close to the \( T_C \); the data obtained at 150 mK are shown in Fig. 5. However, due to technical limitations, the magnetic field sweep rate had to be limited to \( \frac{dB}{dt} = \frac{0.285}{\text{T/min}} \). As shown in Fig. 6, this sweep rate is not enough to take the resistance to zero. We however see a large dip in the resistance at zero field indicative of the TSS state only for \( V_g > V_g^* \); it can be seen that even at temperatures very close to \( T_C \), the data obtained at 150 mK do not exhibit any signatures of TSS for \( V_g < V_g^* \).

The appearance of this TSS depended critically on \( \frac{dB}{dt} \), the rate at which the magnetic field was swept down from its maximum value. For slow sweep rates of the magnetic field, there appeared a dip in the resistance near 0 T, but the resistance remained finite (see Fig. 6). The magnitude of the dip increased as \( \frac{dB}{dt} \) increased and beyond a certain value of \( \frac{dB}{dt} \), the system went into the transient superconducting state.

The TSS was observed up to about 260 mK. Beyond this temperature the TSS does not appear although a dip in the magnetoresistance is seen near zero magnetic field as the field is swept down from 8 T with the magnitude of the dip rapidly decreasing with increasing temperature. The data from a typical measurement at 270 mK and \( V_g = 150 \) V are plotted in Fig. 7(a). The magnetoresistance measurements in the TSS regime were repeated with different dc currents superposed on the measurement ac current of 10 nA; the data are plotted in Fig. 7(c). The critical current extracted from these measurements was about 2 \( \mu \)A which matches well with the critical current measured in similar systems [30]. To the best of our knowledge, a magnetic-field-assisted transient superconducting state has not been observed so far. In a related work a slight reduction in resistance on the insulating side of the superconductor-insulator transition was seen whose magnitude depended on \( \frac{dB}{dt} \) which was interpreted as a signature of the presence of localized cooper pairs in the system in the nonsuperconducting state [32]. The appearance of the TSS depends on the value of the highest magnetic field \( B_{\text{max}} \) to which the system is taken before the field is ramped down. We observed that for \( B_{\text{max}} < 6 \) T the system does not attain the TSS [see Fig. 7(b)]. Interestingly, we also do not observe the TSS when the magnetic field is applied parallel to the interface (see Fig. 8).

There is a due concern about the possible changes in temperature of the sample from changes in spin entropy due to rapid cycling of the magnetic field. We have calculated this change to be on the order of \( \mu \)K owing to the low...
carrier density of the device (see the Appendix). We have also checked for measurement artifacts arising due to any remnant field from the superconducting magnet and have ruled them out through careful measurements. Possible effects arising from magnetocaloric effects of the entire sample holder were ruled out by measuring the temperature changes of a bare device as a function of time. 

As a further check we have also performed similar experiments on SrTiO$_3$ made conducting by Ar$^+$ ion irradiation [33] and on LaTiO$_3$/SrTiO$_3$ heterostructures [34]. Both these systems are known to have a low-temperature superconducting behavior very similar to that of LaAlO$_3$/SrTiO$_3$ but lack the competing ferromagnetic order [21,22,35]. The measurements on these two systems were performed for exactly the same sample dimensions, gate voltage range, temperature, and magnetic field sweep rates as were used for LaAlO$_3$/SrTiO$_3$; the magnetoresistance data are plotted in Fig. 9(a) for reduced SrTiO$_3$ and in Fig. 9(b) for LaTiO$_3$/SrTiO$_3$ heterostructure. We find in both cases that the magnetoresistance plots for increasing and for decreasing magnetic fields fall exactly on top of each other; as expected there is no signature of TSS or hysteresis in the magnetoresistance.

D. Number density extracted from Hall measurement

To understand the origin of TSS it is first necessary to understand the nature of the mobile charge carriers in the system. In Fig. 10(a) we plot $n_e$ extracted from the Hall measurement data assuming a single type of charge carrier in the system. We note that for $V_g > V_c^*$, $n$ appears to decrease with increase in $V_g$; simultaneously the Hall voltage $V_H$ develops a slight nonlinearity with $B$. The charge carriers being electrons in this case, applying a positive gate voltage
is expected to enhance the carrier density $n_z$, as can be seen from the plot of resistance vs $V_g$ in Fig. 10(b). In Fig. 10(a) we also plot the estimated carrier density $n_{calc}$ that would be induced in the system by the gate voltage; the estimate takes into account the electric field dependence of the dielectric constant of the SrTiO$_3$ substrate [36]. We find that $n_{calc}$ and $n_z$ match very well (to within a geometric factor) for $V_g < V'_g$. For values of gate voltage beyond $V'_g$, $n_z$ begins to drop below the expected range showing that the apparent decrease of $n_z$ with increasing gate voltage cannot be accounted for by the electric field dependence of the dielectric constant of the SrTiO$_3$ substrate. The fact that $n_z$ seemingly decreases with increase in $V_g$ beyond $V'_g$ indicates that the transport in this regime is best described by a multiband model [12]. It is known for LaAlO$_3$/SrTiO$_3$ heterostructures that at a certain number density, the system undergoes a Lifshitz transition between light and heavy subbands having different symmetries [37]. The additional carriers introduced are believed to occupy a higher mobility $d_{cz}/d_{cz}$ band near the interface and are responsible for the appearance of superconductivity in the system [12,38].

III. THEORY

There exist now indications, both experimental [9,11,26] and theoretical [13,39], that superconductivity at the interface coexists with (in-plane) magnetization in phase-segregated regions [40]. At low gate voltages our particular device is deep inside the ferromagnetic regime as seen from the large hysteresis in the magnetoresistance. Beyond a certain critical density the system is in a metastable state; the itinerant electrons in the $d_{xz}/d_{yz}$ orbitals favor a superconducting ground state while the in-plane magnetization [14], which originates from the localized magnetic moments at the interface, opposes superconductivity, suppressing superconducting $T_c$. On the application of a perpendicular magnetic field, magnetization of the (in-plane) FM-aligned domains reduces while the out-of-plane component of magnetization $m_z$ takes on a finite value.

To understand quantitatively the origin of the TSS, we have computed the three components of magnetization and the superconducting gap parameter at each instant of time when the perpendicular magnetic field is ramped linearly with time. The data are shown in Fig. 11. We start with a situation where the in-plane magnetization (taken along the x axis) has completely destroyed the superconducting order. While increasing magnetic field, the dynamics of the three components of magnetization is described by the following set of Bloch’s equations:

$$\frac{dm_x}{dt} = \gamma B_z(t)m_y - \frac{m_z}{T_1},$$

$$\frac{dm_y}{dt} = -\gamma B_z(t)m_x - \frac{m_y}{T_2},$$

$$\frac{dm_z}{dt} = -m_z - m_{zs}/T_1,$$

where $\gamma$ is called the gyromagnetic ratio, and $T_1$ and $T_2$ are the time scales for the spin-lattice and spin-spin relaxation, respectively. $B_z(t)$ is increased at the rate $dB/dt$ so as to reach the final value $B_{max}$. With the initial conditions $m_x(t=0) = m_{z0}, m_y(t=0) = 0, m_z(t=0) = 0$, the solutions to the above equations are

$$m_x(t) = m_{z0} \cos[\gamma B_z(t)t]e^{-t/T_1},$$

$$m_y(t) = -m_{z0} \sin[\gamma B_z(t)t]e^{-t/T_2},$$

$$m_z(t) = m_{zs}(1 - e^{-t/T_1}).$$

Therefore, the in-plane magnetization $m_x$ decreases exponentially from its initial value $m_{z0}$ while the out-of-plane magnetization $m_z$ grows up to its saturation value $m_{zs}$. Even though the magnetization $m_z$ along $y$ direction was zero initially, it attains a finite value and oscillates over a large range further degrading the electron pairing. Since perpendicular magnetization is much more detrimental to superconductivity than an in-plane one, it is not possible for the superconductivity to appear in this case.

When the magnetic field is decreased at the rate $dB/dt$ from the value $B_{max}$ at which the final magnetizations are $[m_x, m_y, m_z]$, the set of equations describing the dynamics is

$$\frac{dm_x}{dt} = \gamma B_z(t)m_y - \frac{m_x}{T_1},$$

$$\frac{dm_y}{dt} = -\gamma B_z(t)m_x - \frac{m_y}{T_2},$$

$$\frac{dm_z}{dt} = -m_z - m_{zs}/T_1.$$

FIG. 11. (a) Profile of the perpendicular magnetic field varied at the rate $dB/dt = 1$ T/min up to a maximum $B_{max} = 7$ T. (b) Time variation of the three components of normalized magnetization, $m_x$, $m_y$, and $m_z$. (c) Time variation of the mean-field pairing gap. The shaded regions in all the figures represent the time window over which superconductivity appears. Inset of panel (c) is the magnified view of the region of nonzero superconducting order parameter. Model parameters used were hopping amplitude $t' = 0.277$ eV, chemical potential $\mu = 0$, strength of the Rashba spin-orbit interaction $\alpha = 20$ meV, strength of the attractive pairwise electron-electron interaction $U = t'$, sweep rate $dB/dt = 1$ T/min, maximum applied field $B_{max} = 7$ T, the gyromagnetic ratio $\gamma = 1$, $T_1$ (B increasing) $= 200$ sec, $T_2$ (B increasing) $= 100$ sec, $T_1$ (B decreasing) $= 400$ sec, $T_2$ (B decreasing) $= 200$ sec, and $T_3$ $= 550$ sec ($T_1, T_2$, and $T_3$ are the relaxation times).

174502-6
The solutions to the above equation, with the initial conditions \( m_x(t = 0) = m_{sf} \), \( m_y(t = 0) = m_{sf} \), \( m_z(t = 0) = m_{sf} \), are

\[
m_x(t) = \{m_{sf} \cos[\gamma B_z(t)t] + m_{sf} \sin[\gamma B_z(t)t]\} e^{-t/T_z},
m_y(t) = \{m_{sf} \cos[\gamma B_z(t)t] - m_{sf} \sin[\gamma B_z(t)t]\} e^{-t/T_z},
m_z(t) = m_{sf}(1 - e^{-t/T_z}).
\]

While decreasing magnetic field, the localized moments at the interface start establishing the in-plane magnetization again to its initial value \( m_{so} \) according to

\[
m_{si}(t) = m_{so}(1 - e^{-t/T_z}) + m_{sf} e^{-t/T_z},
\]
which accompanies \( m_z(t) \) in the above equation. Therefore when the field is ramped down, \( m_z \) starts to decay and the in-plane components of magnetization begin to grow towards its zero-field value. However, a finite relaxation time of \( m_z \) implies a finite time for the in-plane magnetization to come back to this value. This creates a narrow time window when the net magnetization is small enough for the superconducting state to be the lower energy state facilitating the emergence of this TSS. Therefore, at 245 mK, superconductivity is a hidden order \([41]\) and is masked by the in-plane magnetization, appearing only when the net magnetization is sufficiently low. The fact that no TSS is seen for magnetic field applied parallel to the interface supports this picture. Our calculations confirm the net magnetization is small enough for the superconducting state to be the lower energy state allowing the superconducting state to manifest. The lifetime of this TSS obtained from our calculations is about 12 seconds allowing the superconducting state to manifest. The lifetime decreased beyond a certain rate, there appears a slice of time where superconductivity manifests as a result of electron pairing becomes energetically favorable. The calculated maximum magnetic field \((\approx 6 \text{ T})\), below which the transient superconductivity does not appear, also comes out naturally from our calculations.

IV. CONCLUSION

To conclude, in this paper we report the observation of a transient superconducting state which appears when a relaxing normal magnetic field reduces the magnetization of the system to a value such that electron pairing becomes energetically favorable. This shows the inherently metastable nature of the superconducting state competing with a magnetic order. The results may have significant impact in understanding the nature of superconductivity in diverse systems such as high-\( T_c \) superconductors and iron pnictide superconductors where superconductivity manifests as a result of electron doping a parent magnetic compound. The coexistence of superconductivity and magnetic order and their controlled tunability using external field open up a regime of investigation with potential in device applications.

ACKNOWLEDGMENTS

A.B. acknowledges funding from Nanomission, Department of Science and Technology (DST), Government of India, and IISc, Bangalore. A.T. and N.M. acknowledge IIT, Kharagpur, for support. A.D. and R.C.B. acknowledge funding from the IFCPAR (Project No. IFCPAR 4704-I) and also extend acknowledgment to DST for the J. C. Bose Fellowship of R.C.B. and to CSIR-India for financial support through the AQuaRIUS Project. A.B. acknowledges fruitful discussions with H. R. Krishnamurthy, Vijay Shenoy, and Manish Jain.

APPENDIX

1. Estimate of adiabatic temperature change due to sweeping of magnetic field

The change in isothermal magnetic entropy change due to magnetic field variation is accompanied by an adiabatic temperature change \( \Delta T_{ad} \) given by \([42]\)

\[
\Delta T_{ad} = -\mu_0 \int_0^H \frac{T}{C_p} \left( \frac{\partial M}{\partial T} \right)_H dH,
\]
where \( H \) is the magnetic field, \( M \) is the magnetization, \( T \) is the temperature, and \( C_p \) is the zero-field heat capacity.

We have calculated the \( \left( \frac{\partial M}{\partial T} \right)_H \) at a given field \( H \) using the values of magnetization \( M(H,T) = M_{sf} \) obtained by a mean-field (Curie) calculation for \( s = 1/2 \) systems, with FM \( T_c \) set at 200 K and the saturation magnetization \( 0.3 \mu_B \) as seen from experiments.

The saturation magnetization \((\sim 0.3 \mu_B)\) for the LaAlO\(_3\)/SrTiO\(_3\) sample is given by \( M_0 = 0.3n g\mu_B J' \), where \( n \approx 10^{17} \text{ m}^{-2} \) is the carrier density, \( g = 2 \), \( \mu_B = 9.27 \times 10^{-24} \text{ J/T K} \), and \( J' = 1/2 \).

The heat capacity is given by

\[
C_p = A t + (B/\alpha) |t|^{-\alpha} + C,
\]
where \( t = T/T_c - 1 \) and the parameters \( A = 5, B = 18, \alpha = -0.8, \) and \( C = 27.6 \) are used to obtain the temperature dependence of \( C_p \) close the typical value for SrTiO\(_3\) \([43]\). The calculated values of the temperature variations of the normalized magnetization \( M/M_0 \) and the zero-field specific heat \( C_p \) are plotted in Fig. 12.

The resultant adiabatic temperature change is plotted for different values of maximum magnetic field \( B_{max} \) and sweep rate \( dB/dt \) in Fig. 13. The nature of the temperature variation is similar to experimental data in other ferromagnetic systems \([44]\). The result obtained gives an estimate of the temperature variation due to sweeping of magnetic field and shows that the change in temperature is insignificantly small due to the small

FIG. 12. Temperature variation of the (a) normalized magnetization \( M/M_0 \) and (b) the specific heat \( C_p \).
carrier density of LaAlO₃/SrTiO₃ interface 2DEG. It shows a peak near the Curie temperature (where maximum entropy is lost) as expected and the temperature variation below 50 K is almost undetectable (see Fig. 13).

The entropy lost due to superconductivity (changes therein due to magnetic field) is exceedingly small since we are deep inside the SC region and the SC Tc actually increases as found experimentally (throwing us deeper in the SC phase).

### 2. Possible effect of remnant field of the superconducting magnet

We have checked very carefully for the effect of remnant field from the superconducting magnets and have ruled out its effect on the phase diagram and on the observed transient superconducting state by the following arguments:

1. The cryogen-free dilution refrigerator and the magnet in it are frequently warmed up to room temperature. The measurements of Tc reported here have been performed after such a warm up before the magnetic field was turned on.

2. Any effect of remnant magnetic field would result in an asymmetric magnetoresistance scan with the peak of magnetoresistance shifted away from zero magnetic field. As shown in the data in Fig. 3(a), the magnetoresistance data always peak at the zero magnetic field showing that the effect of trapped fluxes in the superconducting magnet coil is negligibly small.

3. The measurements reported here have been performed on two different cryostats: a cryogen-free dilution refrigerator equipped with a 16 T magnet which operates down to 10 mK, and a wet He-3 system equipped with an 8 T magnet which operates down to 250 mK. The magnets in these two systems are very different in size, inductance, and construction. The data obtained in both these cryostats could be compared down to 245 mK (the base temperature of the He-3 system); to this temperature the data obtained from both superposed on each other show that the effect of trapped field, if any, is negligibly small.

We have also considered the effects of dB/dt on the copper sample holder. There will be some currents induced due to Faraday effect but a simple estimate showed these to be negligibly small.

### 3. Model Hamiltonian and Bogoliubov–de Gennes treatment

We consider the following tight-binding Hamiltonian, to describe electron pairing at the interface:

\[
H = -t' \sum_{\langle ij \rangle, \sigma} (\hat{c}_i^\dagger \hat{c}_j + \text{H.c.}) - \mu \sum_{i,\sigma} \hat{c}_i^\dagger \hat{c}_i \sigma - B \sum_{i,\sigma} \hat{c}_i^{\dagger} \sigma \cdot \hat{s}_{\sigma} \cdot \hat{s}_{\sigma} \cdot \hat{d}_{ij} \cdot \hat{c}_j \sigma \nonumber
\]

\[
+ \sum_i \Delta_i \hat{c}_i^\dagger \hat{c}_i + \text{H.c.},
\]

where t' is the kinetic hopping amplitude of electrons, μ is the chemical potential, μB is the Bohr magneton, \( h = (m_x, m_y, m_z) \) represents the exchange fields due to the different components of magnetization, \( \alpha \) is the strength of the Rashba spin-orbit interaction, \( \hat{d}_{ij} \) is a unit vector between sites i and j, and

\[
\Delta_i = -\alpha B \cdot \hat{s}_{\sigma} \cdot \hat{s}_{\sigma} \cdot \hat{d}_{ij} \cdot \hat{c}_j \sigma - \mu \sigma \cdot \hat{s}_{\sigma} \cdot \hat{d}_{ij} \cdot \hat{c}_j \sigma.
\]

FIG. 13. Temperature variation of the adiabatic temperature change \( \Delta T_{ad} \) (a) for different values of the maximum magnetic field \( B_{\text{max}} \) and constant sweep rate \( \frac{dB}{dt} = 1 \text{T/min} \) and (b) for different \( \frac{dB}{dt} \) and constant \( B_{\text{max}} = 8 \text{T} \).

FIG. 14. (a) Relaxation of the magnetoresistance with time. The magnetic field was initially ramped up from 0 T to 0.45 T at the rate 1 T/min. The field was then held at 0.45 T and the resistance monitored. (b) Correlation of the magnetic field at different temperatures at \( V_g = -200 \text{V} \). (c) Resistance \( R_{\text{corr}} \) as a function of magnetic field at different temperatures at \( V_g = -200 \text{V} \). (d) Plot of the hysteresis in magnetoresistance as a function of magnetic field at different temperatures at \( V_g = -200 \text{V} \); the hysteresis is seen to decrease sharply as the temperature increases.
\[ \Delta_i = -U \langle c_{i\uparrow} c_{i\downarrow} \rangle \] is the on-site pairing amplitude with the attractive pair potential \( U \).

The above Hamiltonian is diagonalized via a spin-generalized Bogoliubov-Valatin transformation \( \hat{\epsilon}_n(\vec{r}_i) = \sum_{\sigma, \sigma'} u_{n\sigma}(\vec{r}_i) \hat{f}_{\sigma} + v^*_{n\sigma}(\vec{r}_i) \hat{f}_{\sigma}^\dagger \), and the quasiparticle amplitudes \( u_{n\sigma}(\vec{r}_i) \) and \( v_{n\sigma}(\vec{r}_i) \) are determined by solving the Bogoliubov–de Gennes equations

\[
H \phi_n(\vec{r}_i) = \epsilon_n \phi_n(\vec{r}_i),
\]

where \( \phi_n(\vec{r}_i) = [u_{n\sigma}(\vec{r}_i), u_{n\sigma'}(\vec{r}_i), v_{n\sigma}(\vec{r}_i), v_{n\sigma'}(\vec{r}_i)] \). The local pairing gap \( \Delta_i \) is obtained using the following relation:

\[
\Delta_i = -U \sum_n [u_{n\uparrow}(\vec{r}_i) v^*_{n\downarrow}(\vec{r}_i)[1 - f(\epsilon_n)]
\]
\[
+ u_{n\downarrow}(\vec{r}_i) v^*_{n\uparrow}(\vec{r}_i) f(\epsilon_n)],
\]

where \( f(x) = 1/[1 + \exp(x/k_B T)] \) is the Fermi-Dirac distribution function at temperature \( T \) with \( k_B \) the Boltzmann constant. At any instant of time \( t \), the components of magnetization are calculated and then inserted into the above Hamiltonian to solve the mean-field pairing gap self-consistently.

4. Relaxation of the magnetoresistance in perpendicular field

Figure 14(a) shows a plot of the relaxation of the resistance \( R_{\text{sheet}} \) as a function of time at 0.45 T magnetic field. To obtain this data, the magnetic field was initially ramped up from 0 T to 0.45 T at the rate 1 T/min. The magnet was then held constant at 0.45 T and the resistance monitored as a function of time. It was seen that \( R_{\text{sheet}} \) relaxes to a higher value over a couple of minutes. In a separate experiment, the magnetic field was ramped down starting from 8 T at the rate 1 T/min to 0.45 T, the field was held at 0.45 T, and the resistance monitored as a function of time. It can be seen that in both cases \( R_{\text{sheet}} \) relaxes to the same value, although with slightly different time constants. Figure 14(d) shows the temperature evolution of the hysteresis in magnetoresistance. The hysteresis in magnetoresistance weakens as the temperature is increased and eventually dies out by 1.8 K.