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# Gate-tuned superfluid density at the superconducting $LaAlO_3/SrTiO_3$ interface

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The interface between the insulating oxides $LaAlO_3$ and $SrTiO_3$ exhibits a superconducting two-dimensional electron system that can be modulated by a gate voltage. While the conductivity has been probed extensively and gating of the superconducting critical temperature has been demonstrated, the question as to whether, and if so how, the gate tunes the superfluid density and superconducting order parameter needs to be answered. We present local magnetic susceptibility, related to the superfluid density, as a function of temperature, gate voltage, and location. We show that the temperature dependence of the superfluid density at different gate voltages collapses to a single curve that is characteristic of a full superconducting gap. Further, we show that the dipole moments observed in this system are not modulated by the gate voltage.

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Electric field control of conducting channels has allowed great innovation in traditional semiconductor devices.[1] Now heterointerfaces in another class of materials, the complex oxides, have generated significant interest because of their gate tunable properties. Specifically, the conducting interface formed between the band insulators lanthanum aluminate and $TiO_2$ terminated 100 strontium titanate (LAO/STO)[2] exhibits many fascinating properties,[3] suggesting that an electronic reconstruction triggered by the polar/nonpolar interface plays an important role in inducing the conductivity in the STO.[4] At low temperatures this interface displays two-dimensional superconductivity.[5] Additionally, the high dielectric constant of STO at low temperatures[6] makes applying an electric field with a back gate especially effective to tune the properties of this superconducting state.

Caviglia *et al*. showed that, with increasing gate voltage $V_g$, the superconducting critical temperature $T_c$ displayed a dome structure and concurrently the normal state resistance monotonically decreased.[7] Later work showed that the electron mobility and carrier density both increased continuously with $V_g$, with the former dominating the $V_g$ dependence of the conductivity.[8] The evolution of a nonlinearity in the Hall resistivity as a function of $V_g$ (Refs. 8 and 9) has been interpreted by Joshua *et al*. as evidence of electrons populating conduction bands with different mobilities,[10] implying that the ratio of high and low mobility electrons may be tuned by gating.

Notably, the interface breaks spatial inversion symmetry, opening the possibility for spin orbit coupling to impact the electronic properties of the interface gas. Two groups reported tuning of the Rashba spin orbit coupling (RSOC) inferred from magnetoresistance[11,12] and measurements of the in-plane critical fields.[9] They found opposite dependencies for tuning the strength of the spin orbit coupling with $V_g$, making the impact of $V_g$ on the spin orbit coupling unclear, possibly suggesting a peak in the spin orbit coupling.

Moreover, the discovery of magnetic patches that are coexistent with superconductivity[13–15] and the presence of RSOC originating from the noncentrosymmetric nature of the interface have raised the possibility of an unconventional superconducting pairing mechanism or order parameter.[16–18] However, previous measurements studying how gating affects the properties of the interface used electronic transport, which gives limited information about the superconducting state. In this Rapid Communication, we use local magnetic susceptibility to make direct measurements of the superfluid density in LAO/STO and address the question of how the superconducting state evolves with $V_g$.

Measurements were made on a sample with five unit cells (u.c.) of LAO grown at 800 °C and $1.3 \times 10^{-5}$ mbar oxygen partial pressure on a $TiO_2$ terminated STO substrate. The growth was followed by a high pressure oxygen anneal, 600 °C in 0.4 bars. An elemental analysis on samples made with similar growth conditions in the same chamber shows that the magnetism is intrinsic and is not due to external contamination.[19] The sample was silver epoxied to a piece of copper tape, which served as a back gate. $V_g$ was applied between the copper tape and the interface, which was contacted by aluminum wire bonds. Magnetization and susceptibility measurements were made using a scanning superconducting quantum interference device (SQUID),[20] with a 3 $\mu$m diameter pickup loop and a concentric field coil for applying a local ac magnetic field. The pickup loop is sensitive to both the dc static flux and the ac flux resulting from diamagnetic screening currents canceling the field from the field coil. This setup enables simultaneous measurements of ferromagnetism and superconductivity in the sample.[13]

A superconductor will generate screening currents to screen an applied field. The currents extend into a bulk superconductor by the penetration depth $\lambda$. The temperature dependence of $\lambda$ is a probe of the superconducting state. For a thin superconductor of thickness $d$, the screening distance is given

 

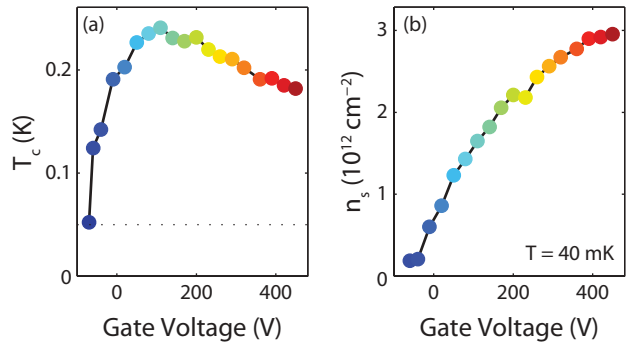


FIG. 1. (Color) (a) The critical temperature as a function of gate voltage forms a dome. The dashed line represents our lowest measurement temperature. (b) The superfluid density at our lowest temperature as a function of gate voltage. The superfluid density increases monotonically throughout the dome. The color scale represents gate voltage and is repeated in Fig. 2.

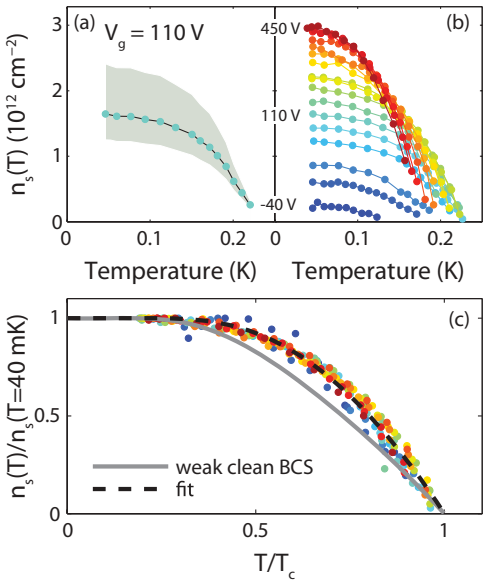


FIG. 2. (Color) (a) Superfluid density vs temperature for $V_g = 110$ V, the peak of the superconducting dome. The gray area shows a systematic error. (b) Superfluid density vs temperature for every gate voltage. The colors represent the same $V_g$ from Fig. 1. (c) Normalized curves from (b). The gray line shows the temperature dependence of a weakly interacting clean BCS $s$-wave superconductor ($\Delta = 1.76$ and $a = 1$). The black dashed line is a fit to the data ($\Delta = 2.2$ and $a = 1.4$).

by the Pearl length $\Lambda = 2\lambda^2/d$.[21] Using a model by Kogan,[22] we extract $\Lambda$ from measurements of the screening currents as a function of the distance between the sensor and the sample. $\Lambda$ is related to the superfluid density, $n_s = 2m^*/\mu_0 e^2 \Lambda$, where $e$ is the elementary charge, $\mu_0$ the permeability of free space, and $m^* = 1.46 m_e$ the effective electron mass measured by Ref. 23 from Shubnikov–de Haas on LAO/STO interfaces. We repeat these measurements at multiple temperatures and gate voltages to map out the superconducting state (Fig. 1). We define $T_c$ as the temperature at which the diamagnetic screening drops below our noise level of $0.01\Phi_0$/A, corresponding to a minimum detectable $n_s$ of 4–14 $\times 10^{10}$ cm$^{-2}$. The statistical errors were smaller than the systematic errors, outlined in gray in Fig. 2(a), from imprecise knowledge of our measurement geometry.[22,24] The systematic errors are fixed for a single cooldown and represent an overall scaling of $n_s$ which would be the same for every measurement.

$T_c$ vs $V_g$ [Fig. 1(a)] has a maximum $T_c = 240$ mK. In the range of applied $V_g$ superconductivity can only be eliminated on the underdoped side of the dome, and $n_s$ grows monotonically with $V_g$, with $n_s = 3.0 \times 10^{12}$ cm$^{-2}$ at the largest $V_g$ [Fig. 1(b)]. The carrier density and mobility were measured in a separate cooldown with no back gate. At 2 K the mobility was $1.02 \times 10^3$ cm$^2$/V s and the density was $2.05 \times 10^{13}$ cm$^{-2}$, ten times larger than the largest $n_s$ we observed.

A small ratio of the superfluid density to the normal density is expected in the dirty limit, in which the elastic scattering time $\tau$ is much shorter than the superconducting gap $\Delta_0$ ($\hbar/\tau \gg \Delta_0$). $\hbar$ is the reduced Plank's constant. Above $T_c$ the normal density of electrons $n$ is given by the optical sum rule $n \propto \int_0^\infty \sigma_1(\omega) d\omega$, where $\sigma_1$ is the real part of the conductivity and $\omega$ the frequency. For a metal $\sigma$ is sharply peaked near zero frequency, so scattering moves the spectral weight to higher frequencies. Below $T_c$, a gap opens at $\omega = 2\Delta_0/\hbar$ and the spectral weight within that gap collapses to a delta function at the origin whose amplitude is proportional to $n_s$.[25] Therefore, in the dirty limit, only a fraction of carriers enter the superconducting state, $n_s/n = 2\Delta_0/(\hbar/\tau)$. Using the gate tuned mobilities reported by Bell *et al.*, 100–1000 cm$^2$/V s,[8] we expect the ratio $n_s/n$ to be 0.01–0.1, consistent with our measured $n_s$.

We now look at the temperature dependence of the superfluid density. Figure 2(b) plots $n_s$ vs $T$ for all $V_g$ across the dome. Strikingly, when normalizing the curves they collapse [see Fig. 2(c)], showing that within our experimental errors there is no change in the superconducting gap structure with electrostatic doping. Furthermore, the collapse is reproducible over multiple positions, sweeps of $V_g$, and samples.[26]

The temperature dependence of the superfluid density is a direct probe of the superconducting order parameter. It can be used to distinguish BCS superconductors from unconventional superconductivity. We fit the normalized curves to a phenomenological BCS model with two parameters $\Delta$ and $a$.[28] $\Delta$ scales the superconducting gap $\Delta_0 = \Delta k_B T_c$. $a$ is a shape parameter that determines how rapidly the gap opens below $T_c$, $n_s \propto 1 - (T/Tc)^{2a}$,[27–29] $\Delta = 1.76$ and $a = 1$ for a clean $s$-wave BCS superconductor with weak coupling,[28] plotted as the gray line in Fig. 2(c). The fit to our data gives $\Delta = 2.2$ and $a = 1.4$. This is consistent with a BCS description with increased coupling or disorder. Both will theoretically increase the gap and the $a$ parameter,[30] shifting the curve up and to the right.

The flattening at low temperature indicates a fully gapped behavior with a gap that is larger than BCS weak-coupling $s$ wave. Our lowest measurement temperature is 1/6 of $T_c^{\max}$, and $n_s$ remains flat (within 3%) up to 35% of $T_c$. A full gap indicates the absence of low energy quasiparticle excitations, ruling out order parameters with nodes in the Fermi surface. Furthermore, the steep rise of $n_s$ near $T_c$ and the absence of a kink in the functional form rule out most weak-coupling

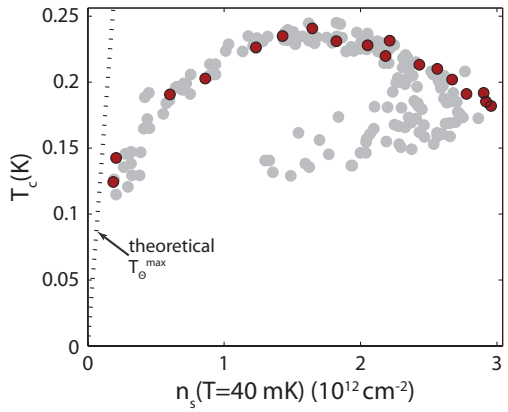


FIG. 3. (Color) Critical temperature vs the superfluid density at lowest temperature ($T \sim 40$ mK). The red points are the data from Fig. 1 and the gray dots represent additional data sets. The dotted line is the theoretical phase fluctuation temperature from Ref. 32, which may be limiting the critical temperature on the underdoped side of the dome. The bimodal distribution on the overdoped side is due to inhomogeneity that locally suppresses $n_s$ in different regions of the sample while the $T_c$ remains the same. See also Fig. 4.

two-band models,[31] because a second smaller gap will slow the onset of superconductivity near $T_c$. Two gaps of similar size, both larger than the BCS gap or a dominant single large gap with a second smaller amplitude gap, could reproduce the data.

The low $n_s$ in the underdoped region may result in the suppression of $T_c$ by thermal phase fluctuations. Such fluctuations would result in a linear temperature dependence of $n_s$ in the underdoped region. Following Ref. 32, we calculate a phase ordering temperature, $T_\theta^{\max} = A\hbar^2 n_s(0)/4m^*$, where $A = 0.9$ in two-dimensional (2D) systems. Figure 3 shows $T_c$ vs $n_s$ (40 mK), and additionally $T_\theta^{\max}$ is plotted as a linear function of $n_s$: The line does not suggest a fit to our data. We have insufficient data at the lowest superfluid densities to make any statement about the functional form of $T_c(n_s)$ in the region where phase fluctuations may be limiting $T_c$. Nevertheless, the proximity of the phase ordering line to the underdoped data suggests that phase fluctuations may drive the abrupt decrease of $T_c$.

Given the 2D nature of the superconducting system we expect a Berezinsky-Kosterlitz-Thouless (BKT) transition, where unbinding of vortex-antivortex pairs suppresses superconductivity and results in a discontinuous jump in $n_s$ near $T_c$. The jump should occur at a finite superfluid density $n_s = 2m^* T_c/\pi\hbar^2$.[33] For the maximum $T_c = 240$ mK a BKT transition should occur at $5 \times 10^{10}$ cm$^{-2}$, which is too close to our measurement threshold to establish a BKT jump in our $n_s$ vs $T$ curves.

Are our observations consistent with a simple $s$-wave order parameter from doped STO[34] or a two-gap mixed state induced by symmetry breaking at the interface? Rashba spin orbit coupling (RSOC), induced by the structural inversion asymmetry, is expected to lift the spin degeneracy and split the energy bands.[35] Additionally, RSOC breaks parity and consequently mixes singlet and triplet states, resulting in an $s$-wave component $\Delta_s$ mixed with a triplet induced $d$ vector, $\mathbf{d}(\mathbf{k}) = \hat{x}k_y - \hat{y}k_x$.[18,36] Mixing results in two gaps, $\Delta = \Delta_s \pm |d_k|$, whose magnitudes depend on the weights of the singlet and triplet components. Varying the relative weights changes the density of states, but always results in two fully gapped Fermi surfaces except for the special case where the $s$-wave singlet and triplet gaps are the same and accidental line nodes form on one band.[18]

Other reports[9,11] have demonstrated significant tuning of the strength of RSOC with $V_g$. An open question, of particular importance to testing this two-gap picture, is how do the weight of the two components change with $V_g$. Our results, showing a consistent functional form for $n_s$ vs $T$ across all $V_g$, show that the superconducting gap structure does not change with $V_g$ and consequently the relative gap weights do not

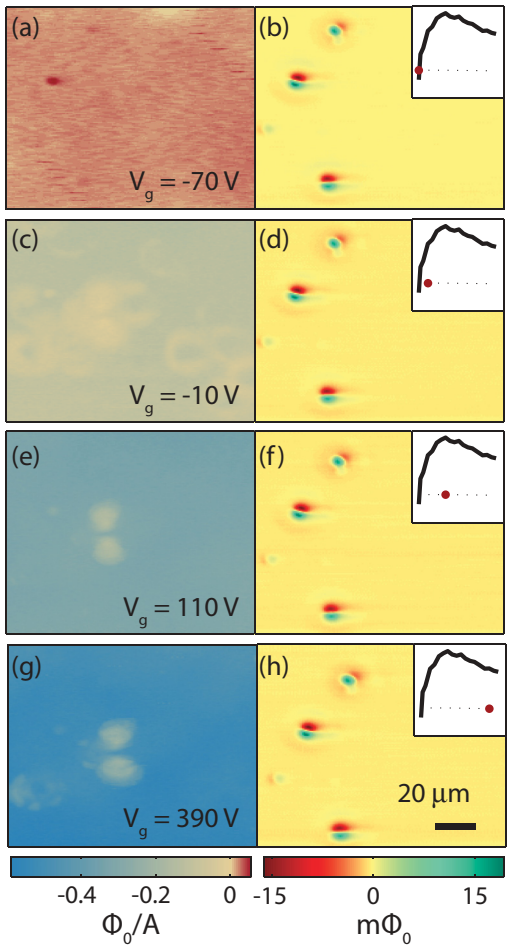


FIG. 4. (Color) Susceptometry (left) and magnetometry (right) at 80 mK at different gate voltages. Inset: Reproduction of the $T_c$ dome from Fig. 1 showing the relative location of $V_g$ in each panel. (a), (b) The sample is no longer superconducting and has a paramagnetic response. Individual ferromagnetic dipoles are also visible in the paramagnetic image. (c), (d) Superconductivity appears and the landscape is relatively inhomogeneous. (e), (f) Peak of the superconducting dome, most inhomogeneity disappears. (g), (h) Excess inhomogeneity returns on the overdoped side of the dome. The ring shapes in the inhomogeneous response match the dimensions of the SQUID's field coil and are likely due to disperse pointlike defects which interact strongly with the field coil. The ferromagnetic patches do not change with $V_g$ and remain when superconductivity is gone.

change with $V_g$. The effect of RSOC on the band structure may depend on the chemical potential which is also tuned by the gate. Therefore the insensitivity of superconductivity to $V_g$ cannot completely rule out a RSOC induced two-gap scenario. Yet, our second observation of the fast opening of the gap near $T_c$ and the compatibility of the data with a single-gap BCS model limits two-gap models. Both gaps must be larger than the BCS $s$-wave gap to capture both the fast rise and flat low temperature dependence of the data.[37,38]

Finally, disorder may play a role in washing out the triplet component. As stated above, the LAO/STO system is a dirty superconductor, with $\hbar/\tau \gg \Delta$. Disorder averaging has very little impact on the isotropic $s$-wave component but may eliminate the triplet component.

In short, our data is most consistent with a single gap. We cannot rule out the presence of two gaps, but our observations limit their size and $V_g$ dependence.

Our scanning SQUID system allows two-dimensional mapping of superconductivity and magnetism at different $V_g$. Figure 4 shows simultaneously imaged susceptometry and magnetometry scans of the same region at 80 mK for four different $V_g$. The inhomogeneity in the diamagnetic screening is very large in the underdoped region ($V_g = -10$ V) and reenters the image in the overdoped region ($V_g = 390$ V). The least inhomogeneity is observed at optimal doping, although it does not disappear. In contrast, the ferromagnetic patches are insensitive to $V_g$ with a constant magnitude and orientation for all $V_g$. This behavior was also observed on 15, 10, and 3.3 u.c. samples, showing the electron density that is modified by $V_g$ does not appear to influence the ferromagnetism.

In conclusion, we presented measurements of the superfluid density as a function of temperature at multiple gate voltages throughout the superconducting dome in LAO/STO heterostructures. The temperature dependence of $n_s$ is well described by a fully gapped BCS model. Moreover, the normalized $n_s$ vs $T$ curves collapse to a single functional form, indicating there is no change in the gap structure with $V_g$. Although we cannot rule out a two-gap mixed singlet/triplet model, the insensitivity of the superconducting state to $V_g$ and the large slope near $T_c$ limits two-gap scenarios. Specifically, both gaps must be larger than the BCS $s$-wave gap and their relative size cannot change throughout the dome. A future experiment to distinguish between these two scenarios may be to gate the superconductivity in the presence of an in-plane field, which can change the relative magnitude of triplet and singlet gaps. Alternatively, samples in the clean limit may reveal a clearer two-gap structure. Additionally, we found that the magnitude and orientation of the ferromagnetic patches that coexist with superconductivity are unchanged by $V_g$, while at the same time $n_s$ goes from zero to $3.0 \times 10^{12}$ cm$^{-2}$. This shows the population of electrons that is modified by the gate is separate from the electrons that contribute to the ferromagnetic order.

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