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Coexistence of superconductivity and ferromagnetism in two dimensions

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Ferromagnetism is usually considered to be incompatible with conventional superconductivity, as it destroys the singlet correlations responsible for the pairing interaction. Superconductivity and ferromagnetism are known to coexist in only a few bulk rare-earth materials. Here we report evidence for their coexistence in a two-dimensional system: the interface between two bulk insulators, LaAlO3 (LAO) and SrTiO3 (STO), a system that has been studied intensively recently. Magnetoresistance, Hall and electric-field dependence measurements suggest that there are two distinct bands of charge carriers that contribute to the interface conductivity. The sensitivity of properties of the interface to an electric field make this a fascinating system for the study of the interplay between superconductivity and magnetism.

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There has been much interest recently in the conducting interface that forms between the two band insulators, SrTiO3 (STO) and LaAlO3 (LAO) [1–13]. This interface shows a rich variety of behavior, including superconductivity [3, 9, 10], magnetism [4, 8, 11, 13], and electric field controlled metal-insulator [2, 6] and superconductor-insulator transitions [7, 9]. Two important mechanisms have been proposed for the creation of the conducting layer at the interface [5, 8, 14–17]: charge transfer from the LAO to the Ti+2 ions at the interface (the so-called “polar catastrophe” mechanism); and conduction due to oxygen vacancies, which can be controlled by growth or post-growth annealing conditions. The Ti bands are also thought to contribute to the magnetism seen in some samples [14].

The electronic characteristics are very sensitive to the growth conditions: generally, it is found that samples grown in an environment with low oxygen partial pressure $P_{O_2}$ have more oxygen vacancies and are consequently more conducting; the conductivity is reduced if the samples are grown in high $P_{O_2}$, or subjected to a post-growth oxygen anneal [8, 13]. It is not only the conductivity that is sensitive to growth conditions: superconductivity is observed in samples grown in intermediate $P_{O_2}$ [8], and signatures of ferromagnetism are observed for samples grown in high $P_{O_2}$ [4, 13], although both phenomena have not been observed until now in the same sample. Here we report measurements on LAO/STO interface structures where both phases are seen simultaneously at low temperatures. Magnetoresistance and Hall measurements indicate that there are at least two bands of charge carriers in the system. Earlier theoretical calculations [18] have pointed to a ferromagnetic ground state of the system due to the multivariant nature of the Ti ions at the interface, but the origin of superconductivity is still not clear. This system joins only a few other bulk materials in which superconductivity and ferromagnetism have been observed simultaneously [19–22], with two critical differences: both the superconductivity and the magnetism are confined to a two-dimensional interface, and the electronic properties of this interface can be tuned over a wide range by means of an electric field. Consequently, it forms a unique system for the investigation of the interplay between superconductivity and magnetism.

The films in this work had 10 unit cells (uc) of LAO grown by pulsed laser deposition at $P_{O_2} = 10^{-3}$ mbar on TiO$_2$ terminated (001) STO single crystal substrates [23]. For electrical measurements, a Hall bar geometry was patterned by photolithography and etched using argon ion milling that removed the LAO layers and a few layers of STO. Measurements confirmed that the bare etched STO was not conducting from room temperature down to millikelvin temperatures. A gate voltage $V_g$ applied to the back of the substrate was used to modulate the conductance of the devices. While the qualitative behavior of all the samples was the same, the devices showed small sample-to-sample variations that were only evident at millikelvin temperatures. The origin of these variations is not clear at the moment. We shall concentrate in this paper on data from a single longitudinal section and its adjacent Hall configurations that showed the sharpest superconducting transitions.

The normal state and superconducting characteristics and their dependence on $V_g$ and temperature $T$ are similar to that seen by other groups [3, 7, 9] (Fig. 1(a)). The current-voltage curves also show a characteristic superconducting signature with a critical current $I_c$ that vanishes when $V_g < -20$V (Fig. 1(b)). However, $I_c$ and the transition temperature $T_c$ do not increase monotonically with increasing $V_g$, but show maxima at $V_g = 80$V (Fig. 1(c)).

Figure 2(a) shows the superconducting transition at $V_g = 80$V at a few different values of magnetic field $H$ applied perpendicular to the film plane. Defining $T_c$ as the temperature corresponding to half the normal state...
FIG. 1: (a) Superconducting transition at different gate voltages \( V_g \). (b) Current voltage characteristics of the sample at 15 mK. (c) Transition temperature \( T_c \) (defined as the midpoint of the resistive transition) and critical current (defined as the current at which the sample switches to the resistive state on ramping the current up from zero) as a function of \( V_g \). Both measures indicate that the maximal superconducting properties are obtained for \( V_g \approx 80 \, \text{V} \). Error bars indicate the difference in \( T_c \) measured between cooling and warming traces.

resistance, a plot of \( T_c(H) \) is shown as an inset to the figure. For two dimensional superconductors in this field orientation, \( T_c(H) \) should be linear, its slope proportional to the Ginzburg-Landau coherence length \( \xi_0 \) [24]. A linear fit gives \( \xi_0 = 64 \, \text{nm} \), close to that reported previously [3, 10].

However, this method of determining \( T_c(H) \) misses some of the more novel and interesting behavior of this system. To show this behavior, we map \( T_c(H) \) continuously by controlling the sample temperature so that the sample resistance \( R_S \) remains fixed as we ramp the magnetic field. Figure 2(b) shows \( T_c(H) \) for three different resistance bias points at \( V_g = 80 \, \text{V} \). The most striking aspect of the data is that the behavior is hysteretic as a function of \( H \). Consider the curve corresponding to the bias point \( R_S = 208 \, \Omega \), slightly lower than the midpoint of the transition. Increasing the magnetic field from negative values towards \( H = 0 \), \( T_c \) shows a smooth increase that is almost linear. The slope of this linear curve gives \( \xi_0 = 71 \, \text{nm} \), close to the value obtained from Fig. 2(a). On ramping \( H \) beyond zero, however, the behavior becomes non-monotonic: in particular, there is a local minimum at \( H \approx 7 \, \text{mT} \). Increasing \( H \) further, \( T_c(H) \) becomes monotonic again. Reversing the ramp direction results in a mirror image of the first trace, giving rise to a characteristic “butterfly” curve. Similar behavior is seen at other gate voltages for which the sample goes superconducting (Fig. 2(c)).

Hysteretic behavior and butterfly curves in the magnetic field dependence of electrical characteristics are signatures of underlying ferromagnetic order in a sample. Such behavior has already been reported in LAO/STO interface samples: Brinkman et al. [4] demonstrated that hysteretic behavior is observed at low temperatures in 26 uc thick LAO films grown under high \( P_{O_2} \); more recently, Ariando et al. [13] were able to observe hysteretic magnetization curves coexisting with paramagnetic and diamagnetic behavior in samples also grown in high \( P_{O_2} \) that persisted to room temperature. For low \( P_{O_2} \) during growth, Huijben et al. [8] note that a low sheet resistance and metallic behavior is observed; for intermediate \( P_{O_2} \), the samples go superconducting; while for high \( P_{O_2} \) magnetic behavior is seen. These experiments suggest to us that the magnetism is associated with the polar catastrophe mechanism, while the superconductivity is...
associated with the presence of oxygen vacancies. However, in all previous experiments, the magnetic and superconducting regimes were quite distinct. In contrast, our samples, which are grown under high $P_{O_2}$ and have $R_S$ consistent with the superconducting samples measured by others [8], show a coexistence of superconductivity and ferromagnetism. The electronic properties of LAO/STO interfaces are extremely sensitive to growth conditions: samples grown by different groups with nominally identical growth conditions show some variations in electronic properties. Our samples appear to be in a growth regime where both phenomena coexist.

Caviglia et al. [12] have reported weak (anti-)localization magnetoresistance (MR) in LAO/STO samples. At first sight, the data from our samples appear to be very similar to their data. Figure 3(a) shows the MR of the sample at a few different temperatures at $V_g = -100$ V, not in the superconducting regime. The magnitude of the resistance change and the sharpness of the resistance dip near zero field increase with decreasing temperature, consistent with a phase coherence length that increases with decreasing temperature. However, a closer look at the low field MR, shown in Fig. 3(b), reveals some significant differences from [12]. In addition to the nonmonotonic MR over a large field scale (Fig. 3(a)), the samples show an additional resistance dip near zero field whose magnitude increases and width decreases with decreasing temperature. This behavior, which is characteristic of weak localization (WL) [25], suggests that there are two independent carrier gases that contribute to the conductance of the device in parallel, each with its own WL contribution. The MR also shows a hysteretic “butterfly” pattern similar to that seen in $T_c(H)$ (Fig. 2(b,c)), indicating that local magnetic fields arising from magnetic order also modulate quantum interference in the carrier gases.

To demonstrate the feasibility of this picture, we have simulated the WL contribution of two parallel two-dimensional gases in the presence of an external magnetic field and a magnetic field arising from a hysteretic intrinsic magnetization. To calculate the WL contribution in the presence of spin-orbit scattering, we use the formalism of Santhanam et al. [26]. The resulting curve is shown in Fig. 3(c). The simulation qualitatively reproduces the experimental features: a non-monotonic MR over a large field scale, and a smaller non-monotonic MR over a smaller field scale, with hysteresis due to intrinsic magnetic order. In the simulation, the larger contribution (88%) is due to a carrier gas that has short phase coherence ($L_φ$) and spin-orbit scattering ($L_{so}$) lengths ($L_φ = 0.17 \ \mu m$, $L_{so} = 0.03 \ \mu m$), while the remaining contribution is due to a carrier gas with longer $L_φ$ and $L_{so}$ ($L_φ = 4.5 \ \mu m$, $L_{so} = 0.45 \ \mu m$). Experimentally, we observe an increase in the amplitude of the low-field MR as the sample approaches the superconducting transition (by changing $V_g$, for example), which is probably due to the presence of oxygen vacancies. Howev...

Further evidence for two parallel conduction channels can be seen in the Hall resistance data. Figure 3(d) shows the Hall resistance $R_{Hall}$ of the sample at $V_g = -100$ V at three different temperatures. The sign of the slope of $R_{Hall}(H)$, $R_H$, corresponds to negatively charged carriers with a density $n \approx 4.4 \times 10^{13} / \text{cm}^2$, assuming a single carrier band. If there were only one electron gas at the interface, increasing $V_g$ should increase $n$, decreasing both $R_S$ and $R_H$. As shown in Fig. 3(e), increasing $V_g$ does indeed decrease $R_S$, but increases $R_H$. This behavior is only possible if there are at least two types of carriers involved in electrical transport, with different dependences of densities and mobilities on gate voltage. Evidence for multiple charge carriers has also been observed by other...
of the system by means of a gate voltage makes this a fascinating system for studying competing cooperative phenomena in two dimensions.

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FIG. 4: (a) Magnetoresistance (top curve) and Hall resistance (bottom curve) at $V_g = 20$ V at 50 mK. (b) Same data as in (a), for one direction of the magnetic field sweep, except the background linear component is subtracted from the Hall resistance.

For conventional itinerant ferromagnets, it is well known that a finite magnetization should manifest itself as a contribution to $R_{\text{Hall}}$ through the anomalous Hall effect [28]. Figure 4(a) shows the longitudinal and Hall MR at $V_g = 20$ V and 50 mK, where $R_S = 0$ when $H = 0$ (Fig. 1(a)). Figure 4(b) shows the same data with the field sweep in one direction. Here a linear contribution determined by fitting $R_{\text{Hall}}(H)$ at high magnetic fields has been subtracted. It can be seen that some of the structure in $R_S(H)$ also has corresponding signatures in $R_{\text{Hall}}(H)$ (this structure is not due to misalignment of the Hall contacts, which is very small). The structure in $R_S(H)$ and $R_{\text{Hall}}(H)$ for $H > 0$ (for this field sweep direction) is also seen at other gate voltages, further from the superconducting transition. We believe that this structure in the Hall resistance is due to an anomalous Hall effect arising from the interaction of the charge carriers with the magnetic moments at the interface, but the structure seen in Fig. 4(b) indicates that this interaction is more complicated than that in simple itinerant ferromagnets, and warrants further investigation. The features in $R_{\text{Hall}}(H)$ change with $V_g$. The sharp dip in both the longitudinal and the Hall resistance for $H < 0$ appears only close to the superconducting transition, and may be associated with vortex flow in the superconductor [29].

In summary, the interface between LAO and STO shows a rich variety of behavior, including interacting ferromagnetism and superconductivity. Our measurements indicate that there are two different types of charge carriers in the system. The ability to tune the properties of the system by means of a gate voltage makes this a interesting and complex phenomena.