Conducting channel at the LaAlO₃/SrTiO₃ interface

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Localization of electrons in the two-dimensional electron gas at the LaAlO₃/SrTiO₃ interface is investigated by varying the channel thickness in order to establish the nature of the conducting channel. Layers of SrTiO₃ were grown on NdGaO₃ (110) substrates and capped with LaAlO₃. When the SrTiO₃ thickness is \leq 6 unit cells, most electrons at the interface are localized, but when the number of SrTiO₃ layers is 8–16, the free carrier density approaches 3.3×10^{14} cm⁻², the value corresponding to charge transfer of 0.5 electrons per unit cell at the interface. The number of delocalized electrons decreases again when the SrTiO₃ thickness is \geq 20 unit cells. The \sim 4 nm conducting channel is therefore located significantly below the interface. The results are explained in terms of Anderson localization and the position of the mobility edge with respect to the Fermi level.

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The two-dimensional electron gas (2DEG) at the interface between the band insulators LaAlO₃ and SrTiO₃ (Ref. 1) continues to stimulate the interest of condensed matter researchers. It exhibits a variety of unexpected properties such as superconductivity, magnetism, spin-orbital coupling, and electronic phase separation.^{5–9} Recent observation of high mobility at low temperatures $(>5 \times 10^4 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1})^{10}$ and the fabrication of millions of transistors on a single chip¹¹ have highlighted the importance of these oxide interfaces both from fundamental and applied perspectives. The 2DEG is thought to result from an electron transfer to the interface between the polar oxide (LaAlO₃) and the nonpolar oxide (SrTiO₃), which is necessary to avoid a divergence of the energy associated with the electric field. 12 A charge transfer of 0.5 electrons per interface unit cell (uc) or 3.3×10^{14} cm⁻² should be required to compensate the electric field in polar LaAlO3 and avert a polar catastrophe. 12 However, a major puzzle is that the experimentally observed carrier densities at low temperatures for the 2DEG in fully oxidized samples^{3,5,13} are an order of magnitude lower than expected. Furthermore, it is unclear where exactly at the interface the conduction electrons are located, since the LaAlO₃ is usually grown on a SrTiO₃ substrate. One proposed explanation is that these "disappearing" electrons are localized within the first SrTiO₃ layers that are closest to the interface, where Ti $3d_{xy}$ subbands have lower energy than the other Ti 3d orbitals. ¹⁴⁻¹⁶ According to the theoretical calculations, the mobile electrons responsible for the transport properties of the 2DEG are farther away (≥4 uc) from the interface. 15 While experimental results 12, 17-19 have shown that the 2DEG can penetrate some distance from the interface into the SrTiO₃ layer, the exact location of the conduction electrons has not been determined. Since the conventional way to fabricate LaAlO₃/SrTiO₃ interfaces is to grow LaAlO₃ layers on SrTiO₃ substrates, it had not been possible to determine the location of the conducting channel.

drawback with conventional LaAlO₃/SrTiO₃ interfaces¹⁻¹³ is that losses in the SrTiO₃ substrate limit the potential application of the 2DEG in high frequency devices.²⁰ In order to overcome this limitation and expand the applicability of 2DEG, LaAlO₃/SrTiO₃ interfaces have been fabricated on other substrates such as silicon, 21 NdGaO3 (110), 22 $(LaAlO_3)_{0.3}(Sr_2AlTaO_6)_{0.7}$ (001), 22-24 and DyScO₃ (110).²² Such an approach may permit a demonstration of the novel topological superconductivity recently predicted to appear in two-layer interacting Rashba systems, which might be fabricated by growing LaAlO₃/SrTiO₃ interfaces on LaAlO₃ substrates. ²⁵ The main aim of the present Rapid Communication is to understand how the physical properties of the 2DEG vary with the thickness of the SrTiO₃ layer at the interface, and to study the transport properties of the electrons involved.

We have grown a LaAlO₃/SrTiO₃ heterostructure on NdGaO₃ (110) substrates. The crystal structure of NdGaO₃ is indexed on an orthorhombic $(\sqrt{2a_0}, \sqrt{2a_0}, 2a_0)$ type cell with a = 5.433 Å, b = 5.503 Å, and c = 7.716 Å. When indexed on the pseudocubic cell (a_0, a_0, a_0) , the in-plane lattice constants a_0 for the pseudocubic lattices of SrTiO₃ (100), NdGaO₃ (110), and LaAlO₃ (100) are 3.905, 3.858, and 3.790 Å, respectively. It is therefore possible to grow epitaxial LaAlO₃/SrTiO₃ (100) interfaces on NdGaO₃ (110) substrates. Ideally, a LaAlO₃ (100) substrate would be the best candidate due to its capacity to reduce the lattice-mismatchinduced strain at the interface, and its low loss tangent at high frequencies.^{20,26} However, problems of crystal twinning²⁷ and unstable surface termination²⁸ in LaAlO₃ set a very stringent limit, as for using a LaAlO₃/SrTiO₃/LaAlO₃ structure for the observation of topological superconductivity,²⁵ and this leads us to choose NdGaO₃ (110) instead. At a conventional LaAlO₃/SrTiO₃ interface, the lattice mismatch between LaAlO₃ and SrTiO₃ is 3%, and it is responsible for the interface strain. However, when the SrTiO3 layer

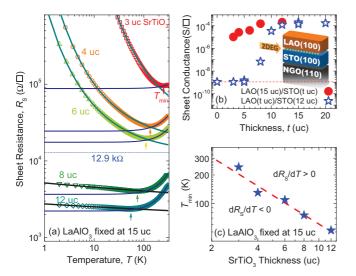


FIG. 1. (Color online) R_S -Tfor (a) curves LaAlO₃/SrTiO₃/NdGaO₃ heterostructures with fixed thickness (15 uc) and different SrTiO₃ thicknesses (from 3 to 12 uc). The blue, dark cyan, and black lines are fits using electron-electron scattering, modified VRH with a two-dimensional Coulomb gap, and weak localization models, respectively. The arrows indicate the upturn temperatures T_{\min} . (b) Room-temperature sheet conductivity as a function of LaAlO₃ thickness for a fixed SrTiO₃ thickness of 12 uc, and also as a function of SrTiO₃ thickness for a fixed LaAlO₃ thickness of 15 uc. The red dashed line is the measurement limitation. The inset is the schematic view for layer structures in a LaAlO₃/SrTiO₃/NdGaO₃ [LAO(100)/STO(100)/NGO(110)] heterostructure, in which the 2DEG exists at the LaAlO₃/SrTiO₃ interface. (c) "Metal-insulator" phase diagram of LaAlO₃/SrTiO₃/NdGaO₃ heterostructure vs SrTiO₃ thickness with a fixed LaAlO₃ thickness of 15 uc. The slope of the red dashed line is -1.

is grown on NdGaO₃ (110), the large mismatch between LaAlO₃ and SrTiO₃ will be partially transferred from the LaAlO₃/SrTiO₃ interface to the SrTiO₃/NdGaO₃ interface. To investigate the depth dependence of the 2DEG, we varied the SrTiO₃ thickness t from 3 to 25 uc during the fabrication of LaAlO₃/SrTiO₃/NdGaO₃ heterostructures, which were capped with 15 uc of LaAlO₃ (Fig. S1 in the Supplemental Material).²⁹ Before deposition, the NdGaO₃ substrates were annealed at 1050 °C in air for 2.5 h to obtain the atomically flat B-site terminated surfaces.³⁰ The growth parameters for both SrTiO₃ and LaAlO₃ layers are as follows: 1.8 J/cm² for laser energy, 760 °C for temperature, and 2×10^{-4} Torr for oxygen partial pressure during the deposition.

The results on temperature-dependent sheet resistance (R_S-T) are shown in Fig. 1(a). Although at high temperatures the resistance of all the LaAlO₃/SrTiO₃/NdGaO₃ samples is dominated by electron-electron scattering with an $R_S \propto T^2$, an upturn of sheet resistance invariably occurs below a temperature T_{\min} (where R_S is minimum). This upturn in R_S -T has been also reported at LaAlO₃/SrTiO₃ interfaces grown on other substrates, $^{21,23,24}_{21,23,24}$ and it depends on the stoichiometry of LaAlO₃. Reducing the thickness of the SrTiO₃ layer raises T_{\min} and R_S at the same time. For the samples with 3, 4, and 6 uc SrTiO₃ layers, the R_S at 2 K is far above the quantum of resistance (12.9 k Ω , including spin

degeneracy). The R_S -T curves for these samples diverge as the temperature is decreased, and they can be well fitted to $R_S \propto \exp[(T_0/T)^{1/2}]$, which suggests carrier localization in these heterostructures and modified variable-range hopping (VRH) with a soft two-dimensional Coulomb gap. For comparison, the samples with a thicker SrTiO₃ (8 and 12 uc) have R_S less than half of 12.9 k Ω , and the R_S -T curves are of the form $1/R_S \propto A + B \ln T$ (A and B are constants) below $T_{\rm min}$, suggesting weak localization in two dimensions. Given our experimental result that the SrTiO₃/NdGaO₃ heterostructures prepared in 10^{-4} Torr are insulating ($R_S > 10^7 \Omega$), the conducting behavior ($dR_S/dT > 0$ and only weak localization at low temperatures) seen in Fig. 1(a) must be due to the electrons at the LaAlO₃/SrTiO₃ interface.

This insulating SrTiO₃/NdGaO₃ interface is different from the conducting one, which is found when NdGaO3 is grown on a SrTiO₃ substrate.³⁴ In the former case, there is no observable conductivity, similar to the insulating interface, where the SrTiO₃ layers are grown on a LaAlO₃ substrate.³⁵ This can be ascribed to the loss of polar discontinuity at the interface. When NdGaO₃ or LaAlO₃ is not a freshly deposited polar layer, but the substrate itself which has been exposed to the ambient atmosphere, the surface charge is compensated by some external charge centers, as the surface of the NdGaO₃ or LaAlO₃ substrate becomes neutral. Hence, the polar discontinuity cannot be easily established at the interface when polar oxides are used as substrates. Similar results to those in Fig. 1(a) are observed when the NdGaO₃ (110) substrates are replaced by (LaAlO₃)_{0.3}(Sr₂AlTaO₆)_{0.7} (001), which shows that a band-gap mismatch with the substrate is not a critical factor.

Moreover, the conducting behavior LaAlO₃/SrTiO₃/NdGaO₃ samples also depends greatly on the LaAlO₃ thickness, as shown in Fig. 1(b), where it is seen that 10-12 uc of LaAlO₃ are needed to make 12 uc of SrTiO₃ conducting. But this critical thickness of LaAlO₃ is larger than 4 uc that are commonly observed at conventional LaAlO₃/SrTiO₃ interfaces. Given that a higher critical thickness for the LaAlO₃/SrTiO₃ interfaces grown on other substrates is also observed by Bark et al., who found that with 50 uc SrTiO₃ the critical LaAlO₃ thickness is around 15 uc,²² this result could imply an important role of the strain in this phenomenon. Figure 1(c) shows the variation of T_{min} with $SrTiO_3$ thickness t. The temperature T_{min} separates the regions with $dR_S/dT > 0$ for the higher temperatures and thicker SrTiO₃ layers, and $dR_S/dT < 0$ on the opposite side. The dependence of T_{\min} on SrTiO₃ thickness can be described by $T_{\rm min} \sim 1/t$, which is consistent with a temperature-dependent mean free path or relaxation time signifying small energy transfer scattering in the 2DEG.^{36–38}

The temperature dependence of sheet carrier density (n_S) and mobility (μ_H) are plotted in Figs. 2(a) and 2(b), respectively. Compared to conventional LaAlO₃/SrTiO₃ interfaces grown on SrTiO₃ substrates,^{3,5} the interfaces grown epitaxially on NdGaO₃ with an 8–16 uc SrTiO₃ layer show an n_S independent of temperature below 100 K that is one order of magnitude larger, and close to the value of 3.3×10^{14} cm⁻², which corresponds to 0.5 electrons per unit cell at the interface. It can be argued that the reason for the temperature independence is the clamping effect of the NdGaO₃ substrate,

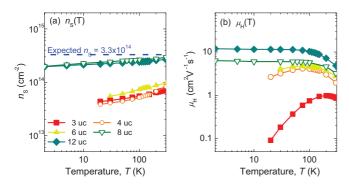


FIG. 2. (Color online) (a) The carrier density $n_{\rm S}$ and (b) mobility $\mu_{\rm H}$ as a function of temperature for samples with different SrTiO₃ thicknesses, from 3 to 12 uc, keeping the LaAlO₃ thickness at 15 uc.

which prevents the SrTiO₃ layer from undergoing the structural transitions that occur at low temperatures, 5,39 thus avoiding the strong localization of carriers at low temperatures, which is widely observed in the 2DEG on SrTiO₃ substrates. This result supports the view that the SrTiO₃ phase transitions are important for determining the low temperature 2DEG properties, carrier localization in particular. ⁴⁰ Also the absence of temperature dependence in mobility when the free carrier concentration is high suggests that electron-electron scattering is dominant. The main point here is that these characteristics, the temperature-independent value of n_S and the high density of free carriers approaching the ideal value predicted by the polar catastrophe model, are observed only when the number of $SrTiO_3$ monolayers is $\geqslant 8$. This implies that the formation of a mobile 2DEG requires at least 8 uc (~3 nm) of SrTiO₃ for the conducting channel, which should be regarded as the minimal propagating depth for the 2DEG and is consistent with previous experiments performed on conventional LaAlO₃/SrTiO₃ heterostructures, ^{12,17} but this time the opposite limit, i.e., carrier localization induced in SrTiO₃ layers ≤6 uc thick, has been directly observed.

In order to illustrate further the effect of SrTiO₃ thickness on the 2DEG, room-temperature n_S and μ_H as a function of $SrTiO_3$ thickness t are plotted in Figs. 3(a) and 3(b). The abrupt enhancement of $n_{\rm S}$ from 0.9×10^{14} to 2.9×10^{14} cm⁻² is observed in a very small window of SrTiO₃ thicknesses, i.e., from 6 to 8 uc. The value of n_S seems to saturate for t =8–16 uc, and then falls to 0.8×10^{14} and 0.6×10^{14} cm⁻² at 20 and 25 uc. This is also consistent with the results of Bark et al., who observed a low value of $n_S = 0.5 \times 10^{14} \text{ cm}^{-2}$ at 50 uc.²² On the other hand, in Fig. 3(b), a linear increase of room-temperature μ_H with SrTiO₃ thickness is observed from 3 to 12 uc, which proves that the SrTiO₃ layer is truly the conducting channel for the 2DEG. On further increasing the SrTiO₃ thickness to 16 and 25 uc, a nearly constant $\mu_{\rm H}$ (close to that for the conventional LaAlO₃/SrTiO₃ interface at room temperature) is attained. According to the above data, the SrTiO₃ thickness required for a mobile 2DEG is around 8-16 uc.

A rough estimate of the width t of the 2DEG at the interface can be obtained by considering its energy per unit area [Fig. 3(c), and Fig. S9 in the Supplemental Material]. Generally, the electrons can lower their energy by spreading out deeper into the SrTiO₃, provided the states are available.

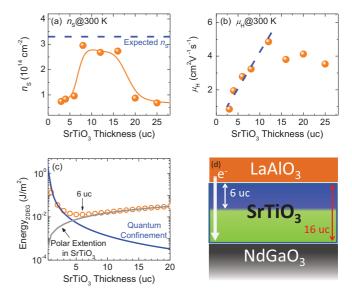


FIG. 3. (Color online) The SrTiO₃ thickness-dependent $n_{\rm S}$ and $\mu_{\rm H}$ at 300 K are shown in (a) and (b), respectively. (c) The calculation of 2DEG energy per unit area of interface. (d) Schematic view of the LaAlO₃/SrTiO₃/NdGaO₃ heterostructure showing at low temperatures the SrTiO₃ thickness range for a mobile 2DEG is around 8–16 uc (green area), and below 6 uc the carriers are localized (blue area).

However, there is an energy penalty to be paid because the polarization fields extend into the SrTiO₃, hence the total energy to confine the 2DEG in SrTiO₃ can be written as

$$E_{\rm 2DEG} \approx n\hbar^2/2mt^2 + \sigma^2 t/24\varepsilon_0\varepsilon,\tag{1}$$

where n is the electron density per unit area, m is the electron mass, σ is the sheet charge density, and ε is the dielectric constant of SrTiO₃. Minimizing this energy with respect to t, we find the minimum at

$$t = [24\varepsilon_0 \varepsilon n\hbar^2 / m\sigma^2]^{1/3}, \tag{2}$$

with $n=3.3\times10^{14}~\rm cm^{-2}$, $\sigma=50~\mu \rm C\,cm^{-2}$, and $\varepsilon=300$, a value that depends little on temperature in SrTiO₃ thin films, ⁴² and this gives us a SrTiO₃ thickness of 2.2 nm or 6 uc, which is close to our minimal SrTiO₃ thickness of 8 uc for a delocalized 2DEG.

We propose that the electron transport is dominated by Anderson localization, which is related to the two-dimensional nature of the channel. The delocalized carrier density depends on the effective position of the mobility edge $(E_{\rm M})$ with respect to the Fermi level (E_F) . When the SrTiO₃ layer is very thin, the electrons are localized because of the potential fluctuations induced by charge disorder due to ionic interdiffusion at the interface. This ionic interdiffusion usually involves the first 1-2 uc from the interface, and it will create a mobility edge, localizing the states at the bottom of the band. As the SrTiO₃ thickness increases, these potential fluctuations are screened, and the mobility edge drops rapidly to a lower energy. The Fermi energy falls less rapidly with thickness, varying as 1/t for a constant density of states. At about 8 uc of $SrTiO_3$, E_F exceeds E_M and we see the onset of electron delocalization, and metallic conductivity at low temperatures. Our experiments clearly prove that the conducting electrons are located appreciably below the LaAlO₃/SrTiO₃ interface of our films. The reduction in carrier density again in SrTiO₃ layers thicker than 16 uc could be understood in several ways. As the 0.5 electrons per Ti extend into SrTiO₃ layer farther from the interface, the Fermi level $E_{\rm F}$ falls towards the bottom of the band due to the increase of available electronic states and it approaches the t_{2g} band mobility edge for the bulk. This arises from defects such as oxygen vacancies that are distributed throughout the SrTiO₃, which is more disordered when it is an epitaxially grown layer rather than a single-crystal substrate. Also, there is a tendency for strain relaxation and associated defect production in these thicker SrTiO₃ layers, which can raise the mobility edge again and localize the carriers. A third possibility is the polarization of the distorted SrTiO₃ layer, which can compensate the polarization catastrophe at the interface.²²

We can model the number of mobile electrons with thickness quite nicely with a single band having a constant density of states. The total number of available states increases in proportion to the number of layers, leading to a Fermi level (relative to the bottom of the rectangular conduction band)

$$E_{\rm F} = a/t,\tag{3}$$

where t is the SrTiO₃ thickness and a is a constant. Then we model the mobility edge to vary due to two effects—one falling off exponentially with t, due to the disorder in the interface layer, and the other a constant low-energy mobility edge due to residual disorder in the SrTiO₃, giving

$$E_{\rm M} = b \exp(-t/t_0) + c, \tag{4}$$

where b, t_0 , and c are all constants. At room temperature, we roughly assume that there is a baseline for the delocalized carrier density $n_{\rm S}$ due to the thermal activation. Hence, $n_{\rm S}$ can be evaluated by

$$n_{\rm S}(E_{\rm F}) = \begin{cases} d & (E_{\rm F} < E_{\rm M}), \\ d + n \left[\frac{E_{\rm F} - E_{\rm M}}{E_{\rm c}}\right] & (E_{\rm F} \geqslant E_{\rm F}), \end{cases}$$
(5)

where d is a constant baseline [assumed to be 0.5×10^{14} cm⁻², which is the carrier density for 50 uc SrTiO₃ (Ref. 22)] and n is the carrier density for the ideal 2DEG, 3.3×10^{14} cm⁻². Based on these, we show the fit to both $E_{\rm F}$ and $E_{\rm M}$ as a function of thickness in Fig. 4(a). Because $E_{\rm F}$ falls much more slowly with thickness and $E_{\rm M}$ is higher than $E_{\rm F}$ at low thickness, there are two crossover points of t_1 and t_2 , indicating free carriers in the thickness range $t_1 < t < t_2$. The fitted curve for the delocalized carriers is also quite consistent with the experimentally observed values in Fig. 4(b). A reasonable screening length t_0 of about 1–2 unit cells is seen for the mobility edge, the details are of which is given in Fig. S10 in the Supplemental Material.⁴³

In conclusion, we have shown that the thickness of the SrTiO₃ layer in epitaxially grown LaAlO₃/SrTiO₃ heterostructures is critical for determining the 2DEG transport properties. Samples with the thinnest SrTiO₃ exhibit a low carrier density at room temperature, a robust insulating ground state, and modified variable-range hopping transport behavior at low

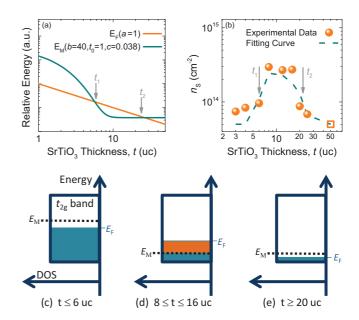


FIG. 4. (Color online) (a) Fitted $E_{\rm F}$ and $E_{\rm M}$ as a function of SrTiO₃ thickness. (b) The comparison on experimentally observed $n_{\rm S}$ and fitted $n_{\rm S}$ as a function of SrTiO₃ thickness. The open square in (b) is taken from Ref. 20 as a comparison. The details on controlling the fitting parameters are discussed in Fig. S10 (Ref. 43). Schematic 2D-density of states (DOS) (number of available electronic states per unit interface area) vs energy for the $t_{\rm 2g}$ band are shown with SrTiO₃ thickness \leq 6 uc in (c), 8–16 uc in (d), and \geq 20 uc in (e). Electrons below $E_{\rm M}$ are localized and denoted by blue, while electrons which are below $E_{\rm F}$ but beyond $E_{\rm M}$ are delocalized and denoted by orange. In these sketches, the total area below $E_{\rm F}$ is fixed for each thickness, indicating the total number of carriers at the LaAlO₃/SrTiO₃ interface is fixed.

temperatures, due to the Anderson localization of the 2DEG arising from the random interface potential with ionic interdiffusion. When the SrTiO₃ thickness reaches 8–16 uc, the carrier density increases to almost the expected 0.5 electrons per interface Ti site, and the mobility saturates at the conventional room-temperature value. However, the carrier density falls again for the thicker layers to the conventional value found for single-crystal SrTiO₃ substrates. Most of the Ti 3d electrons are localized at the very bottom of the 3d band, below the mobility edge. We are therefore able to explain the observed behavior on a localization model where the position of the Fermi level and the mobility edge depend on the SrTiO₃ layer thickness. The study of these thin epitaxially grown layers has enabled us to describe the role of localization and to define the extent of the conducting region at the LaAlO₃/SrTiO₃ interface. It shows how to tailor the oxide interface to optimize the two-dimensional conduction, which will be of importance for oxide electronics.

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