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Subband structure of two-dimensional electron gases in SrTiO₃

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Tunneling between two parallel, two-dimensional electron gases (2DEGs) in a complex oxide heterostructure containing a large, mobile electron density of $\sim 3 \times 10^{14} \text{ cm}^{-2}$ is used to probe the subband structure of the 2DEGs. Temperature-dependent current-voltage measurements are performed on SrTiO₃/GdTlO₃/SrTiO₃ junctions, where GdTlO₃ serves as the tunnel barrier, and each interface contains a high-density 2DEG. Resonant tunneling features in the conductance and its derivative occur when subbands on either side of the barrier align in energy as the applied bias is changed, and are used to analyze subband energy spacings in the two 2DEGs. We show that the results agree substantially with recent theoretical predictions for such interfaces. © 2013 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4831976>]

Two-dimensional electron gases (2DEGs) at complex oxide interfaces, such as LaAlO₃/SrTiO₃ and RTiO₃/SrTiO₃ (where *R* is a trivalent rare earth ion such as Gd or La), exhibit remarkable properties such as superconductivity,¹ tunable spin-orbit coupling,² magnetism,^{3–7} and strong electron correlations.^{8–10} These 2DEGs contain high densities of mobile charge carriers that reside in the SrTiO₃.^{11,12} To understand their properties, it is essential to have a quantitative description of the subband structure.

The conduction band edge of SrTiO₃ is derived from Ti 3*d* *t*_{2g} states. In bulk SrTiO₃, the degeneracy of these states at the Γ point is lifted by spin-orbit coupling,¹³ and by the low temperature tetragonal transition. Many recent studies have modeled 2DEGs in SrTiO₃.^{14–25} Most consider interfaces having a carrier density of $\sim 3 \times 10^{14} \text{ cm}^{-2}$, which is the theoretically expected density for polar/non-polar interfaces such as LaAlO₃/SrTiO₃ and RTiO₃/SrTiO₃. While the results from these models differ quantitatively, a consistent qualitative picture has emerged. In particular, the confining electric field gives rise to a low-lying (in energy) *d*_{xy}-derived subband, which has a low in-plane mass and accommodates a large fraction of the total carrier density. Additional, closely spaced (in energy) higher-lying subbands of *d*_{xy} and *d*_{xz,yz} character are also predicted to be occupied. The *d*_{xz,yz} states are less spatially confined than the *d*_{xy}-derived subbands, because of their low out-of-plane mass. The relative role of the different subbands in transport, superconductivity, magnetism, and spin-orbit coupling is currently a subject of active investigation.^{10,20,26–29}

Experimentally, the subband structure of SrTiO₃-based buried (i.e., not at surfaces) 2DEGs has been probed using Shubnikov-de Haas oscillations.^{30–34} These studies indicate that transport occurs in multiple subbands. However, typically, only a small fraction of the carriers gives rise to oscillations. Furthermore, the oscillations tend to be complex, and advances in theoretical understanding are still needed for their complete interpretation and comparison with theory.

A direct method of probing 2DEG subband structures involves out-of-plane transport. In particular, resonant tunneling

between two parallel 2DEGs separated by a tunnel barrier yields structures in *dI/dV* and *d²I/d²V* as a function of applied voltage *V* (*I* is the tunneling current) when the subbands in the two 2DEGs line up (see Fig. 1).^{35–37} The energy minima of the subbands can be obtained directly, as resonant tunneling requires conservation of the in-plane momentum.³⁸

SrTiO₃/GdTlO₃/SrTiO₃ structures are suitable for such 2D-to-2D tunneling experiments, because a high-density 2DEG with the theoretically expected charge density of $3 \times 10^{14} \text{ cm}^{-2}$ forms at each SrTiO₃/GdTlO₃ interface,¹² and the GdTlO₃ can serve as the tunnel barrier (see Fig. 1). Shubnikov-de Haas oscillations obtained from samples containing one such interface were interpreted as arising from either a single, spin-split subband or two subbands.³³ Only about 10%–30% of the carrier density measured in Hall gave rise to oscillations. In this Letter, we report on experimental evidence of resonant tunneling in SrTiO₃/GdTlO₃/SrTiO₃ structures. Subband spacings are obtained and allow for quantitative comparisons with theoretical predictions.

SrTiO₃/GdTlO₃/SrTiO₃ heterostructures were grown on (001) (LaAlO₃)_{0.3}(Sr₂AlTaO₆)_{0.7} (LSAT) single crystals, using hybrid molecular beam epitaxy (MBE). The growth procedure and detailed structural characterization have been reported elsewhere.^{12,39} A schematic of the heterostructure is shown in Fig. 1(a). An 80 nm thick Gd-doped SrTiO₃ layer serves as the contact to the bottom 2DEG. Its carrier density ($\sim 1 \times 10^{19} \text{ cm}^{-3}$) is an order of magnitude lower than the effective 3D charge density in the 2DEG ($\sim 3 \times 10^{14} \text{ cm}^{-2}$) in the 5-nm-wide undoped SrTiO₃ layer. The GdTlO₃ barrier thickness was 20 nm. Devices were fabricated using contact lithography. Al/Ni/Pt/Au layers with thicknesses of 40/20/50/300 nm, respectively, were used as Ohmic contacts. The top Ohmic contact (dimensions: $10 \times 10 \mu\text{m}^2$) was deposited on the top-most SrTiO₃ layer using e-beam evaporation. It served as the hard mask for an inductively coupled plasma (ICP) assisted dry etch with BCl₃ gas to form square vertical structures as shown in Fig. 1(a) and that exposed the Gd-doped SrTiO₃ layer for depositing the bottom Ohmic contact. The entire device was covered with a 400 nm SiO₂ layer (not shown in the schematic), deposited using e-beam evaporation, followed by an ICP dry etch step containing

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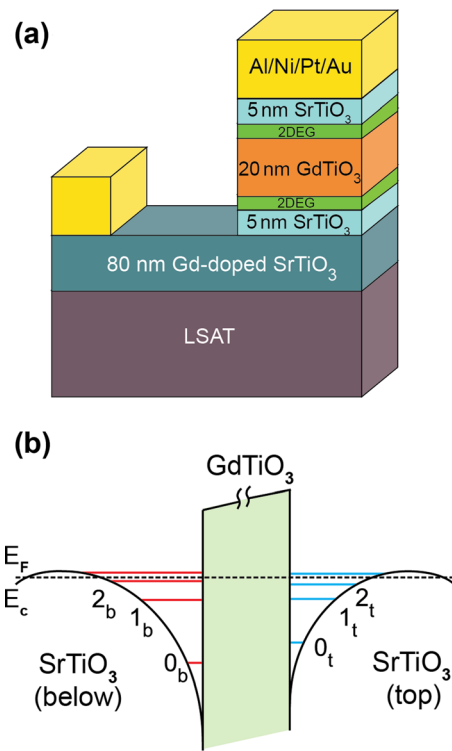


FIG. 1. (a) Schematic of the tunnel device. (b) Schematic energy diagram of the subband configuration at a SrTiO₃/GdTiO₃/SrTiO₃ heterostructure at zero applied bias and low temperature. A slight asymmetry in the carrier concentration and resulting subband spacing between the two 2DEGs is assumed. The number of subbands and band bending shown is for illustrative purposes only and the schematic is not meant to be quantitative. E_c indicates the conduction band edge and E_F (dashed line) is the Fermi level. The numbers (0, 1, 2) refer to the subbands indices, and the subscripts to top and bottom 2DEG, respectively.

CF₄ gas to vertically etch through the SiO₂ and expose the Ohmic contacts. Thick Au pads (~300 nm) were deposited directly atop the exposed Ohmic contacts using e-beam evaporation to create larger contacts on top of the SiO₂ layer to facilitate wire bonding. I-V measurements were carried out using a Keithley 2400 Broad Purpose Source Meter using voltage-controlled measurements with the bias applied to the top Ohmic contact. I-V curves were collected at different voltage sweep rates, and from different devices to ensure that the data was consistent. Measurements as a function of temperature were performed in a Physical Property Measurement System (Quantum Design PPMS Dynacool).

Figure 2 shows dI/dV and d^2I/d^2V characteristics obtained from I-V measurements at different temperatures. The tunnel spectra consist of an exponentially increasing background current that is due to electrons that tunnel non-resonantly (caused by processes that do not conserve in-plane momentum or involve phonon assisted transport) on which features that are due to subband resonant tunneling are superimposed (arrows in Fig. 2). The fact that resonant tunneling features can be observed indicates that the GdTiO₃ tunnel barrier is of reasonable quality, i.e., loss of momentum conservation due to defects in the barrier is insufficient to broaden the resonant tunneling features to a point where they become indiscernible from the background.

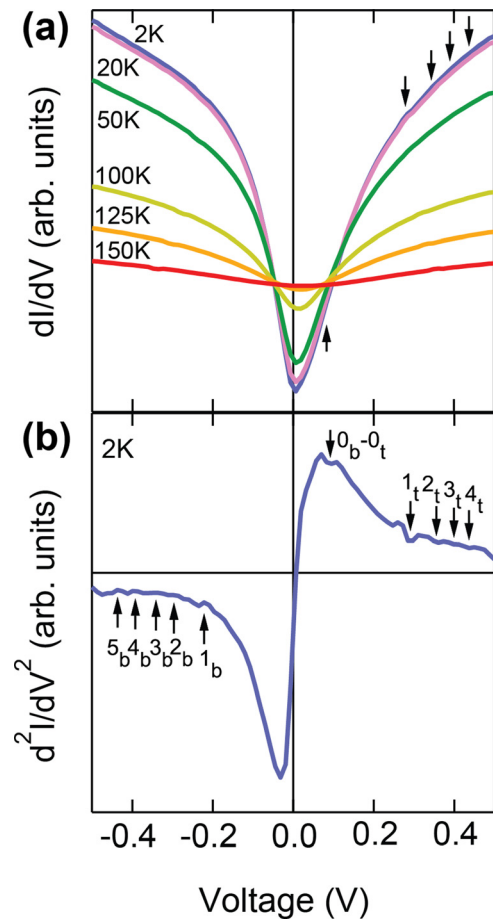


FIG. 2. (a) dI/dV characteristics of the device at different temperatures and (b) d^2I/d^2V characteristics of the device at 2K. The I-V data collected had 80 data points in between -0.5 V and $+0.5$ V.

Similar to 2DEGs in III-V heterostructures^{40,41} there exists an inherent asymmetry between the 2DEGs formed at SrTiO₃/GdTiO₃ and GdTiO₃/SrTiO₃ interfaces, respectively. Specifically, when SrTiO₃ is grown on GdTiO₃, the 2DEG shows lower mobility and slightly lower carrier concentration ($\sim 2 \times 10^{14} \text{ cm}^{-2}$).⁴² Differences in the atomic structure between these interfaces were detected⁴³ and are the likely origin of the slightly different interface charge. The band diagram shown in Fig. 1(b) accounts for the asymmetry at zero bias, with the top 2DEG having a reduced carrier density compared to the bottom 2DEG.⁴² The asymmetry prevents the lowest lying d_{xy} states on either side (denoted 0_t and 0_b here) from lining up in energy at zero bias. When a positive gate bias is applied to the top 2DEG, the subbands in the top 2DEG are lowered in energy with respect to the bottom 2DEG. This causes the 0_t and 0_b subbands to align, allowing for resonant tunneling between them. This gives rise to the first feature at positive bias in the d^2I/d^2V characteristics shown in Fig. 2(b) and occurs at a bias of 89 mV. It indicates that the separation in energy between 0_t and 0_b levels of the two 2DEGs is 89 meV.

With increasing positive bias, additional features are observed and correspond to resonant tunneling from the lowest subband of the bottom 2DEG into various subbands of the top 2DEG. When the bias is reversed, similar features are seen, and correspond to resonant tunneling into subbands of the bottom 2DEG. From the voltages at which the features

appear in the d^2I/d^2V , the subband spacings in the top and bottom 2DEGs can be obtained. They are summarized in Table I, using the data at 2 K. The resonance position was taken as the voltage of the minimum (dip) in the d^2I/d^2V . The results show a relatively large separation between the lowest-lying subband and the second lowest subband of 210 and 310 meV, for the top and bottom 2DEG, respectively. The subband spacing is very sensitive to the carrier density,^{14,17} the smaller spacing for the top 2DEG is consistent with its lower carrier density, and consequently, reduced confinement. Theoretical calculations predict that the lowest two subbands have d_{xy} character for carrier concentrations in the range of 10^{14} cm^{-2} . The theoretically predicted spacing for these two subbands ranges between ~ 160 and 270 meV for a carrier density of $\sim 3 \times 10^{14} \text{ cm}^{-2}$,^{14,16,18,19,21,42} i.e., the density of the bottom 2DEG. The larger subband spacing in the experiments may be due to a larger confinement (out-of-plane) mass than is commonly assumed in theory,⁴⁴ and/or the compressive in-plane epitaxial strain from the lattice mismatch with the LSAT substrate, which lowers the energy of the d_{xy} subbands,^{45,46} Considering this, experiments and theory are in excellent agreement.

Higher-lying subbands are visible as a series of closely spaced features in the d^2I/d^2V . We observe four (three) additional subbands for the bottom (top) 2DEG. Although their close spacing makes the dips somewhat difficult to discern, the separation between these higher-lying energy states is much smaller, on the order of a few tens of meV (see Table I). These findings are also in accordance with the theoretical models, which show a series of closely spaced subbands at higher energy. The theoretical energy spacings for higher-lying d_{xy} bands are on the order of 10–80 meV,^{14,16,18,19,21,42} similar to what is observed here. Theory also predicts higher-lying subbands with $d_{xz,yz}$ character. In 2D resonant tunneling, the energy and in-plane (parallel to the barrier) momentum of the electrons is conserved,³⁸ Thus, only d_{xy} -to- d_{xy} tunneling should be observed, as the first subband (d_{xy}) lines up with other subbands with increasing bias. We therefore conclude that at least four of the higher-lying subbands have sufficient d_{xy} character for resonant tunneling to be allowed. A recent theoretical study of tunnel junctions consisting of a SrTiO₃ tunnel barrier with a La-delta-doped layer in its center indicates that resonant tunneling in this structure occurs preferentially via states that have $d_{xz,yz}$ character.⁴⁷ The present study, which has a different structure (purely 2D-to-2D tunneling),

TABLE I. Subband spacings in the top and bottom of 2DEG, respectively, derived from the dip positions in the d^2I/d^2V characteristics at 2 K. The dip positions were obtained from a set of I-V curves that had twice as many data points as those shown in Fig. 2(b), to maximize the accuracy with which the dip positions could be obtained. Given the number of data points, the accuracy is no better than 6.25 meV.

Subbands (top 2DEG)	Subband spacing (meV)	Subbands (bottom 2DEG)	Subband spacing (meV)
0 _t -1 _t	200	0 _b -1 _b	310
1 _t -2 _t	65	1 _b -2 _b	75
2 _t -3 _t	44	2 _b -3 _b	38
3 _t -4 _t	38	3 _b -4 _b	58
		4 _b -5 _b	44

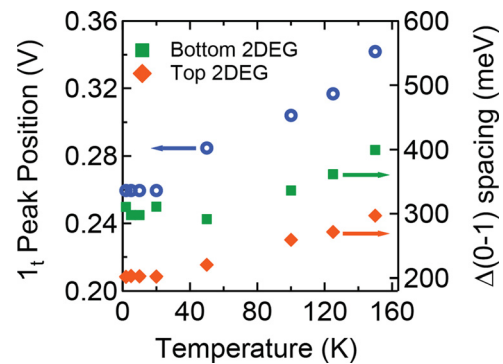


FIG. 3. Temperature dependence of the 1_t feature (left axis) and of the 0-1 subband spacing (right axis) of the bottom and top 2DEG, respectively.

shows that d_{xy} -to- d_{xy} tunneling has sufficient transmission probability to give rise to resonant tunneling features. The results also show that it will be difficult to observe Shubnikov-de Haas oscillations from any subband except the lowest-lying, d_{xy} -derived subband, due to the very close spacings of the higher-lying subbands, and is consistent with the observation of oscillations from a single, spin-split subband.³³

As the temperature is increased the dip position shifts to larger bias (Fig. 3) and subband spacings increase. Similar shifts (in the same direction) have been observed in tunnel studies of Si and III-V subbands, respectively, and have been explained with thermal activation to higher-lying, less spatially confined subbands, which changes the potential profile in a manner that subband spacings increase.^{35,48} The change in subband spacing is about 47% and 28% between 2 and 150 K for the top and bottom 2DEGs, respectively.

Finally, we comment on inelastic processes like phonon emission and absorption that could take place. These mechanisms cannot explain the prominent features (1_t, 0_b-0_t, 1_b). The highest frequency phonons in the oxide structures are $\sim 0.1 \text{ eV}$ —this is substantially smaller than the positions of 1_t and 1_b. The position of feature 0_b-0_t could, on energetic basis, be assigned to an inelastic process. But, if so, it should appear with a companion at negative voltages—it does not. The temperature dependence (Fig. 3) is also not compatible with an inelastic phonon process, as phonon frequencies would not increase by $\sim 28\%$ – 47% between 2 and 150 K.

In summary, the study demonstrates the utility of resonant tunneling to probe subband spacings in 2DEGs at complex oxide interfaces, where the mobility is too low and the subband spacing too small to resolve a significant fraction of subbands via quantum oscillations. The studies provide evidence of an inherent asymmetry of 2DEGs on either side of the barrier, the effect of carrier concentration and confinement on subband spacings, and confirm the predictions of theoretical models. Resonant tunneling studies should be useful to study the effects of epitaxial strain and modulation of carrier density on the subband structure, thus further advancing the theoretical understanding of 2DEGs in complex oxides.

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