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Transportanomaly and itsmodulations via gating effect and lightillumination at the SrNbO$_3$/SrTiO$_3$ interface

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Abstract

LaAlO$_3$/SrTiO$_3$-based two-dimensional electron gas (2DEG) has been extensively studied because of its intriguing physical properties and potential application prospect. However, seldom researches have related their extraordinary macroscopic transport phenomena to the microscopic domain structure of SrTiO$_3$. This requires some unique technique like scanning superconducting quantum interference device (SQUID) microscopy. In this work, we developed a different 2DEG system at the interface of SrNbO$_3$ thin film and SrTiO$_3$. Using only the electrical methods, we found a pronounced hysteresis behavior in the resistance versus temperature curves, marked by the appearance/disappearance of two resistance peaks in the heating/cooling process. In sharp contrast to the conventional gate effect, the resistance peak grows under positive electric biases applied to backgate with conducting interface being grounded. In addition, a weak light (0.04 mW, 405 nm) can completely eliminate the two resistance anomalies. After a systematic analysis, we attribute the resistance anomaly to the cubic-tetragonal transition of bulk SrTiO$_3$ and surface SrTiO$_3$. The present work presents a promising demonstration to get mesoscopic information on oxide interface via transport behaviors.

1. Introduction

Ever since the celebrated interface between LaAlO$_3$ (LAO) and SrTiO$_3$ (STO) where the two-dimensional electron gas (2DEG) was found [1], many STO based 2DEG systems were discovered in succession, boosting this peculiar research fields in physics and material science. For a decade, much effort has been made to study their exotic properties including two-dimensional superconductivity [2], magnetism [3], and enhanced Rashba spin-orbital coupling and so on [4]. Less of them related the properties with the STO surface microstructure. By performing a micrometre-scale study via scanning superconducting quantum interference device (SQUID) on LAO/STO system, current flowed in narrow paths at low temperatures was observed [5]. The STO tetragonal domain structure was implied to affect the configuration of current paths on thermal cycling across the STO structural transition temperature (105K). Furthermore, very recent study has shown that this local domain structure can modulate not only the current flowing behaviour but also the striped magnetism at low temperatures in different 2DEG systems including LAO/STO, γ-Al$_2$O$_3$/STO and bare STO surfaces [6]. This has raised the importance of the STO surface structure over the properties of the 2DEG surface/oxide, bringing forward a new angle of view in both better understanding and possible designing of the 2DEG device.

Although little is known about the impact of the microscopic domain structure from STO on the macroscopic transport behaviors, signatures were indeed captured [7, 8]. In 2007, Siemon et al [9] reported a relationship of resistance versus temperature [R(T)] with obvious hysteretic between the warm-up and cooldown. When cooling down, resistance drops monotonically with temperature but exhibits two clear peaks while warming up in the same thermal cycle. Other groups found even strong effects in mesoscopic devices.

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(micrometers in size) and nano Hall-bars [10, 11]. Preliminary analysis suggests the existence of a close relation between these effect and structural transitions in the interfacial layer of STO [12].

While all of the previous works focused on the relatively mature LAO/STO system, in this letter, we present a different 2DEG system-SrNbO$_3$/SrTiO$_3$ interface which has been reported before but only in a SrTi$_{1−x}$Nb$_x$O$_3$ solid solution system [13]. We found very strong hysteresis phenomenon characterized by two pronounced resistance peaks at ~84 K and ~179 K respectively in the warm-up process. On the contrary to the previous researches on a microscopic scale, we demonstrate a strong hysteresis transport behavior in large samples of millimeters in size, which will smear the domain effect by averaging as generally believed. More than that, a great electric gate tunable effect on this thermal hysteresis was observed, with positive gate voltage amplifying the high temperature peak while negative one suppressing it. Moreover, the hysteresis can be totally eliminated by photo excitation even when incident light is as weak as 0.04 mW (405 nm). All these phenomenon are compatible with the capture and release of charge carries by domain wall in the interfacial layer of STO.

2. Methods

SrNbO$_3$ (SNO) thin films were deposited on TiO$_2$-terminated STO (001) substrate (5 × 5 × 0.5 mm$^3$ in size) by pulsed laser deposition (PLD). The STO (001) substrate before growth was chemically etched by buffer HF and subsequently annealed at 1000 °C to obtain a typical step-and-terrace surface with TiO$_2$-termination. A KrF Excimer laser (wavelength 248 nm) with the laser fluence of 1.5 J cm$^{-2}$ and a repetition rate of 1 Hz was adopted. In the growth process, the substrate temperature was kept at 700 °C in an oxygen pressure of 1.2 × 10$^{-3}$ mbar. After growth, the sample was directly cooled down to room temperature in the same oxygen ambience without oxygen annealing. The film thickness was monitored by reflection-high-energy electron diffraction (RHEED). After the preparation of the sample, surface morphology measurement was conducted via an atomic force microscope (AFM) (SPI 3800N, Seiko), and the crystal structure was measured using a thick SNO film deposited in same conditions by a Bruker diffractometer (D8 Discover, Cu Ka radiation). The temperature dependence of the out-of-plane lattice parameter for STO was detected by powder x-ray diffractometer (Rigaku, smart Lab). Resistance was measured using a Quantum-Design physical property measurement system (PPMS) in the temperature range from 2 K to 300 K and the field range up to 7 T, with an applied current of 10 μA. During the temperature sweep for both cool-down and warm-up processes, 5 K/minute’s sweep rate was adopted. Ultrasonic Al wire bonding (20 μm in diameter) with the four-point van der Pauw geometry was used for electrode connection. When performing a gating experiment, back gate was applied and the SNO/STO 2DEG interface was grounded. The maximal leakage current is lower than 10 nA. For the light illumination experiments, lasers with the wavelengths of 405 nm and 532 nm were introduced though an optical fiber into the PPMS vacuum chamber. The spot size of the light was ~2 mm in diameter, focusing on the middle space of the samples.

3. Results and discussion

Figure 1(a) shows the time-dependent RHEED intensity oscillations of the specular spot during growth of the SNO film. RHEED oscillation periodicities for 10 U.C. film thickness were observed. The deposition rate is around 68 pulses per layer. In spite of the damping of the oscillation intensity at the third unit cell, the following regular intensity indicates that a layer-by-layer growth mode still dominate. The RHEED patterns are shown in the insets and the streaky pattern after the 10 U.C. deposition suggests a smooth and island free film surface. Figure 1(b) exhibits the AFM topographic images after the 10 U.C. growth, corroborating the surface flatness with step-and-terrace morphology and a root-mean-squared (RMS) roughness less than 1 nm. To identify the structure phase of the SNO film, out-of-plane x-ray diffraction (XRD) is conducted on a thick film as shown in figure 1(c). The (002) Bragg reflections at 44.74° is well-defined for the SNO film, corresponding to the lattice constant of 4.048 Å. Taking the different substrate strain into consideration, the reported value of 4.1 Å on KTaO$_3$ is very close to our SNO film [14]. Accordingly, the lattice mismatch is deduced to be -3.5% using the STO lattice constant of 3.905 Å. The full width at half maxima (FWHM) of the SNO (002) peak is 0.76°.

Although the lattice mismatch between SNO and STO is large (~3.5%), we still get a good quality film as indicated by the data of RHEED and XRD with the clear oscillation of the RHEED intensity and a well-defined crystal structure. All these reveal epitaxial growth as well as very high crystallinity of our films.

Figure 2(a) records the temperature (T) dependence of the sheet resistance ($R_s$) of the SNO film. The overall trends of both cool-down (blue arrow) and warm-up (red arrow) process, from 300 K down to 18 K, are metallic. The resistance upturn below 18 K is attributed to weak localization. During cool-down, the sheet resistance decreases monotonically from 300 K down to 18 K. Twenty-fold reduction of the $R_s$ from 39 471 Ω/$\square$ at 300 K to 1997 Ω/$\square$ at 18 K is experienced. Surprisingly, hysteresis behavior is observed in the warm-up process.
between 10 K and 190 K, resulting in two resistance kinks at $\sim$84 K and $\sim$179 K respectively. The insert plot in figure 2(a) is obtained by analyzing the antisymmetrized Hall resistance $R_{xy}$ versus magnetic field adopting a two-band model [15]. $n_1$ and $n_2$ are coexisted two species of charge carriers. The first and second species are respectively high-density-low-mobility and low-density-high-mobility charge carriers. The total carrier density is $1.16 \times 10^{14}$ cm$^{-2}$ at room temperature and $9.17 \times 10^{12}$ cm$^{-2}$ at 2 K, in the reasonable range for STO-based
2DEG. Obviously, despite of the unusual thermal hysteresis, an immediate conclusion can be drawn that SNO/STO shares many features with LAO/STO.

The hysteretic transport behaviors of \( R_\text{s}(T) \) are believed to be a consequence of the phase transition of STO. As is well established, STO experiences a cubic to tetragonal transition at approximately 105 K \[16\], resulting in unusual phenomena. When the sample is cooling across 105K, as revealed by the results of low-temperature scanning electron microscopy, the originally homogeneous interface conductivity starts to break into highly resistant domain areas, leaving network-like conducting domain walls \[17\]. When a warm-up procedure melts domains, the current paths will reconfigure, resulting further resistance changes. As driving forces, over cooling and over heating are generally required by phase transition. This may be the reason for the appearance of thermal hysteresis. The resistance peak in the warming process implies a sudden release of the confined charge carriers in domain walls. In contrast, the carrier accumulation toward domain walls in the cooling process may be slow seen from the absence of resistance anomaly in the cooling branch of the \( R(T) \) curve.

The explanation for the resistance peak at \( \sim 179 \) K is not simple. To get further structure information, we conducted a x-ray diffraction investigation for STO in the temperature interval from 4 K to 300 K. Shown in figure 2(b) is the temperature dependence of the out-of-plane lattice parameter for STO. In addition to an visible shrinkage slightly below 105 K, which corresponds to the well known cubic-tetragonal transition, no other anomaly is observed. But there is report on the phase transition from tetragonal to orthorhombic which occurs at around 65 K \[18\]. Unfortunately, our powder x-ray diffractometer with variable temperatures (Rigaku, smart Lab) has no enough resolution to detect a splitting of the substrate peak at low temperatures. Obviously, the resistance peak at 180K has nothing to do with bulk phase transition. Fortunately, we found evidences for the occurrence of STO surface phase transition at a temperature well above 105 K. As reported by Salman et al \[19\], a cubic-tetragonal transition takes place at \( \sim 150K \) when the surface layer of STO is 150 nm in thickness. Moreover, the critical temperature will monotonically increase as surface layer thickness decreases. It is therefore a reasonable inference that the outmost STO layers undergo a phase transition at temperatures \( \sim 179 \) K when warmed up, releasing confined charge carriers.

In general, the charge carriers of the 2DEG display a distribution along out-of-plane direction. Most charge carriers reside in a very thin layer in the vicinity of the STO surface (\( \sim 10 \) nm), and only a tiny minority of charge carriers locate in very interior of STO. The two resistance peaks in figure 2(a) are the respect responses of shallow and deep charge carriers to phase transition.

Goble et al reported a study of \( R(T) \) in mesoscopic LAO/STO devices combined with the polarized light microscopy of structural domains. They found no any detectable features by polarized microscopy at the higher temperature at \( \sim 179 \) K \[10\]. Obviously, if domain structure appears, they will situate in an ultrathin layer underneath the STO surface, and cannot be detected by the conventional optical technique.

To shed further light on the anomalous phenomenon, we investigated the effect of electric gating: a gate field will tune the distribution of 2DEG in conduction channels. Indeed, we observed strong changes in \( R(T) \) curves when gated. As schemed in figure 3, when the gate voltages from -20 V to 150 V are imposed on the \( R_\text{s}(T) \) curves, the cool-down plots keep monotonically decreasing with temperature (figure 3(a)), and the resistance change occurs in the normal way: resistance increases or decreases according to the the depletion or accumulation of charge carriers.

The most striking observation is the anomalous variation of the resistance peaks at 179 K (figure 3(b)), appearing in the warming curves: The peak resistance monotonically grows from \( \sim 5800 \Omega/\square \) without being gated to \( \sim 13000 \Omega/\square \) under a \( V_\text{G} \) of 150 V. This observation conflicts with our consensus. In general, a positive bias will introduce more electrons into 2DEG, depressing sheet resistance. As an analogy with the situation of 105 K, the anomalous gating effect may be the consequence of surface phase transition of STO. Below 179 K, the surface layer of STO may mainly takes the tetragonal structure. Earlier first-principles study has indicated the tendency of oxygen vacancies to migrate and pin in the domain walls \[20\]. For this phase, gate field will break large domain into stripe-shaped domains, yielding more domain walls. Therefore, as the amplitude of the positive voltage increases, trapping centers like oxygen vacancies in domain walls increase, thus the resistance grows. Although additional electrons are injected into the conduction channel by positive biases, they may be unable to compensate the effects of enhanced charge trapping. In this scenario, we can understand the enhancement of the resistance peak at 179K. When temperature exceeds 179 K, the surface tetragonal-cubic transition takes place, sheet resistance exhibits an abrupt drop due to the release of trapped carriers. As documented by recent literature, a gate voltage indeed favors fine domains \[17\].

According to the resistance change (figure 4(a)), we simply estimated the released charge carriers at phase transition. During the estimation, it has been assumed that \( \Delta n/\sigma = \Delta \sigma/\sigma = \Delta R/R \) and the mobility takes the value at 180 K. As shown in figure 4(b), nearly half charge carriers are trapped by domain walls and defects there. As for the resistance kink at 84 K, it shows weak gating effect. This is because the phase transition is almost completely finished in the interior of STO.
Further evidence comes from the conducting light illumination experiments, showing that the defects at domain boundaries are responsible for the thermal hysteresis and the abnormal gating effect (illustrated in figure 5). Because of the cool-down plots coincide with the warm-up ones, we only show illumination effects for warming curves. The most significant feature is that the hysteresis effect can be totally suppressed by light illumination. Take the 405 nm and 0.04 mW light as an example. Not only the two resistance anomalies disappear, the resistance upturn below 18 K is also suppressed. It means that the resistance humps are indeed caused charge trapping. Without gate field, the density of in-gap states may be low and is completely eliminated by a light as weak as 0.04 mW (corresponds to a power density of 1.27 mW/cm$^2$). As expected, further increasing light power causes a strong depression of the R(T) curve, especially at high temperatures. 532 nm

![Figure 3](image1.png)

**Figure 3.** Cooling (a) and warm-up (b) $R_s(T)$ curves taken at different back-gate-voltages from -20 V to 150 V.

![Figure 4](image2.png)

**Figure 4.** (a) The warm-up $R_s(T)$ curve at a back-gate-voltage of 150 V. $\Delta R$ is the resistance difference between the kink peak and a linear extension from the high temperature part. (b) Estimation of $\Delta n/n = \Delta R/R$ versus positive gate voltages taking the mobility value at 180 K.
lights was also applied and has similar effect on killing the abnormity. Obviously, the trapped states locate in the band gap of STO.

Under the light illumination, the trapped electrons in oxygen vacancy states in the domain walls are certainly excited, so that the carrier density is increased, resulting in higher conductivity as well as an enhanced mobility. Moreover, in our previous work, light illumination can reinforce the electromigration of oxygen vacancies [21]. Consequently, the trapped electrons at the domain boundaries are released and migrate beyond the walls, smoothing all the hysteretic transport behaviors.

4. Conclusion

In conclusion, two-dimensional electron gas with unusual transport behavior has been fabricated by growing SrNbO$_3$ on SrTiO$_3$. A strong thermal hysteresis of the resistance-temperature plots is observed, characterized by the appearance/disappearance of two resistance peaks in the heating/cooling process. Moreover, electric gate causes an opposite variation of the resistance peaks, on the contrary to the conventional gate effect. However, a weak light (0.04 mW, 405 nm) can completely eliminate the two resistance anomalies, resulting in smooth resistance-temperature relation. We found signatures for a relation between resistance anomaly structure transition of SrTiO$_3$, Bulk and surface SrTiO$_3$ under cubic-tetragonal transition at different temperatures, resulting in charge trapping/releasing thus resistance abnormity. The present work demonstrates the macroscopic responses to mesoscopic domain structure.

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