Transport properties of embedded LaTiO₃ layers

(LaTiO₃ 界面における輸送特性)

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1. Introduction

Interfaces have many interesting physical properties, such as the formation of quantum wells and are therefore at the center of attention as subjects for research. Most interface studies have been done with Si and other semiconductors. Interfaces are widely used by the semiconductor industry in electronic devices. Recently limitations of size and performance have moved interest away from Si to other, more versatile materials, such as oxides.

Many of the interesting properties of oxide materials are related to various forms of ordering, such as magnetic order, charge order or orbital order and are studied by many research groups. However, making perfect oxide heterostructures is difficult because of oxygen loss and the presence of various crystal defects that have a large effect on material properties. Only a few research groups have succeeded in making truly 2-dimensional oxide heterostructures, e.g. LaAlO₃ / SrTiO₃ [1].

The appearance of electronic phases in oxides is often closely related to the number of carriers in the material. Various techniques exist to control the carrier density, with chemical doping being the most common. In addition to bulk doping, however, there are various other ways to change the carrier density. For example, field effect, photocarrier injection, charge transfer at interfaces, and nanoscale carrier confinement. It is therefore very interesting to study the effect of variable carrier density on the properties of oxide heterostructure with the aim of controlling electronic ordering.

In this study, the focus is on the LaTiO₃ / SrTiO₃ system. Recent theoretical works have shown that this type of heterostructure may support various forms of electronic ordering [2, 3]. Experimental evidence, however, is still lacking.

The aim is to study the possibility of tuning the carrier density and electronic phases in LaTiO₃ / SrTiO₃ heterostructures by changing temperature or applying magnetic or electric fields.

2. Experiment

All heterostructures studied in this work were grown on SrTiO $_3$ (100) substrates. SrTiO $_3$ (STO: a_0 =3.91) is one of the most common substrates used for atomic-level growth of oxide thin films, mainly because of good quality of commercially available single crystals. All SrTiO $_3$ substrates were 0.2 $^\circ$ miscut and wet etched to obtain a regular atomic-scale step-and-terrace surface which is predominantly terminated by the TiO $_2$ atomic layer of the perovskite structure.

Lanthanum titanate (LaTiO₃) was deposited on the SrTiO₃ substrates. Since both SrTiO₃ and LaTiO₃ (LTO: a_0 =3.97) have the perovskite structure and the lattice mismatch is small, it is possible to grow epitaxial LaTiO₃ films on SrTiO₃. Non-doped LaTiO₃ is a Mott-insulator but it shows metallic behaviour when doped with Sr.

Thin films were grown by Pulsed Laser Deposition (PLD). This is a physical vapor deposition method that can be used to fabricate high-quality oxide thin films when the laser ablation conditions are under precise control. The 0.2° miscut SrTiO₃ (100) substrates were annealed at 900 °C for 30 min to obtain straight and parallel surface steps before deposition. After that, LaTiO₃ films were deposited on the substrate at 500 °C and 10⁻⁶ Torr of oxygen. Under these conditions, a pure LaTiO₃ phase

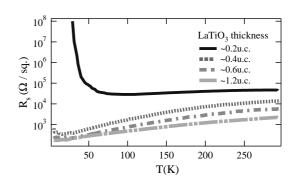


Figure 1: Sheet resistance of fractional LaTiO₃ layers in SrTiO₃.

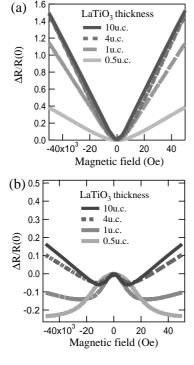


Figure 2: Magnetoresistance for various LaTiO₃ layer thicknesses with the magnetic field applied (a) perpendicular or (b) parallel to the film surface at 5 K.

can be grown without the appearance of the La₂Ti₂O₇ phase [4], which is a band insulator. The LaTiO₃ deposition rate was 46 pulses per unit cell using a KrF excimer laser ($\lambda = 248$ nm) at 1 Hz. The film thickness was estimated by deposition rate and RHEED oscillations. In order to study how the interface morphology changes with LaTiO₃ coverage, several samples which had different LaTiO3 thicknesses were made. After LaTiO3 deposition, SrTiO₃ (100) was deposited at 500 °C and 10⁻⁶ Torr oxygen at a deposition rate of approximately 26 pulses per unit cell at 2 Hz as a capping layer to prevent surface depletion layers from affecting the carrier density in the LaTiO₃ layer. Such crystal growth conditions resulted in a large density of oxygen vacancies in the heterostructures. In order to refill the oxygen vacancies in the SrTiO₃ capping layer and the substrate, samples were annealed for 6 hours at 400 °C in a furnace in air.

The physical properties of these samples were characterized as follows: surface morphology was observed by Atomic Force Microscopy (AFM), resistance was measured from 300 K to 5 K. Magnetoresistance and Hall effect measurements were carried out in a PPMS to determine the magnetic field response of the samples. The Hall bars were prepared by mechanical milling.

In back-gating bias experiments, silver paste was used to obtain an Ohmic contact with the substrate. The leak current was less than 1 nA.

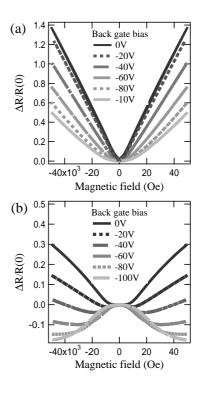


Figure 3: Magnetoresistance of a 10 unit cell LaTiO₃ layer in $SrTiO_3$ under various back-gate biases with the magnetic field applied (a) perpendicular or (b) parallel to the film at 5 K.

3. Results and discussion

As shown in Fig. 1, LaTiO₃ layers down to 0.7 u.c. coverage showed metallic resistance behavior as the temperature decreased to 5 K. In contrast, a sample in which the LaTiO₃ coverage was approximately 0.3 u.c., showed a metal-insulator transition at around 50 K. It is obvious that the carrier density in this fractional-layer sample was low, possibly resulting in the localization of carriers at that temperature. This insulator phase could not be broken down by applying a magnetic field. This means that for sub-monolayer La doping at the interface, no evidence of magnetic ordering was seen by transport measurements. However, for obtaining a quantum well structure with unit-cell-scale confinement, this type of sample showed the greatest promise.

The conductivity of the heterostructures was also measured in a magnetic field. Fig. 2 shows the LaTiO₃ thickness dependence of magnetoresistance (MR). It is clear that thinner LaTiO₃ layers in SrTiO₃ do not contain high-mobility carriers because the strong positive magnetoresistance, which is caused by high-mobility carriers in SrTiO₃ substrate, was reduced for sub-monolayer LaTiO₃ coverage. When the magnetic field was applied parallel to the film, a large negative MR was observed in thinner LaTiO₃ samples. This means that the negative MR originates in the heavily-doped SrTiO₃ interface layer.

Several mechanisms that can be responsible for the observed MR were considered. There is a possibility that the negative in-plane magnetoresistance might be caused by charge ordering, considering the temperature dependence of fractional LaTiO₃ layer conductivity [Fig. 1]. However, from the magnetoresitance results, other possibilities also arise, including scattering between two conducting layer that have a large difference in carrier mobilities. The presence of multiple layers was studied by measuring substrate step edge direction effect on MR. Negative MR was only observed when the current was directed along the terraces. Since step edges work as strong scatterers, it is clear that inter-layer scattering is important in this system.

Further evidence was obtained from field-effect and Hall resistance measurements. Fig. 3 shows the MR behavior under electric field applied from a back gate electrode for a 10 u.c. LaTiO₃ sample. By applying a negative gate voltage, it is possible to tune the potential at the interface, depleting high-mobility carriers from the SrTiO₃ substrate. With increasing gate bias, magnetoresitance behavior started to resemble the magnetic field dependence seen in thinner LaTiO₃ sample that has fewer carriers distributed in SrTiO₃.

Non-linear Hall resistance results [Fig. 4] is another evidence of the existence of multiple conductive layers. Multiple conducting layers were approximated by a two-layer model that includes a high-carrier-density, low-mobility layer and a low-carrier-density high-mobility layer. The Hall resistance data were fitted by this model[5]. The fitting results showed that mobility in SrTiO₃ dropped as the LaTiO₃ layer thickness was reduced or when a negative bias was applied.

4. Conclusion

SrTiO₃ / LaTiO₃ / SrTiO₃ heterostructures were grown with different LaTiO₃ thicknesses and transport properties were studied under magnetic and electric fields. Thinner LaTiO₃ layer showed strong negative MR, and a nonlinear Hall resistance. No direct evidence of ordered phases was seen in MR measurements. In this study, quasi-2D behavior was seen, but it was also recognized that spilling of carriers into SrTiO₃ limits the carrier confinement.

References

- [1] A. Ohtomo and H. Y. Hwang *Nature* **427**, 423 (2004).
- [2] S. Okamoto, A. J. Mills and N. A. Spaldin *Physical Review Letters* **97**, 056802 (2006).

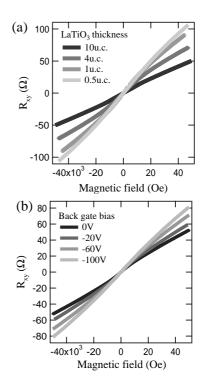


Figure 4: Non-linear Hall resistance of LaTiO₃ layers in SrTiO₃, (a) LaTiO₃ thickness dependence and (b) applied back-gate bias dependence for a 10 u.c. LaTiO₃ sample at 5 K.

- [3] R. Pentcheva and W. E. Pickett *Physical Review Letters* **99**, 016802 (2007).
- [4] A. Ohtomo, D. A. Muller, J. L. Grazul and H. Y. Hwang *Applied Physics Letters* **80**, 3922 (2002).
- [5] D. K. Schroder SEMICONDUCTOR MATERIAL AND DEVICE CHARACTERIZATION, 2nd ed (1998).

Conferences

- JSAP 70th Autumn meeting 2009 [8p-H-5], "Magnetoresistance of SrTiO₃ / LaTiO₃ / SrTiO₃ quantum well" (presentation), <u>R. Ohtsuka</u>, K. Nishio, M. Matvejeff and M. Lippmaa
 - (Another poster presentation at JSAP and one more presentation at next JSAP meeting)
- 16th International Workshop on Oxide Electronics
 (WOE 16) 2009 "Interface and transport properties of embedded, sub-unit cell LaTiO₃ and (La, Sr)O layers" (poster), <u>R. Ohtsuka</u>, M. Matvejeff° and M. Lippmaa

(Another presentation at 基盤 A 「遷移金属酸化物界面における新規強相関電子状態の放射光分光と探索」 2009)