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Growth Control of the Electronic Properties of the $LaAlO_3/SrTiO_3$ Heterointerface

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Introduction

There is increasing recent interest in oxides and their heterostructures, because of the rich variety of physical properties displayed by them. The perovskite $SrTiO_3$ is a key material among the oxides. It is a wide band gap semiconductor and becomes metallic and even superconducting when slightly electron-doped. The mobility can exceed 10^4 $cm^2V^{-1}s^{-1}$ at low temperatures in bulk, motivating researchers to study quantum transport in this material, particularly in the two-dimensional (2D) limit.

The $LaAlO_3/SrTiO_3$ heterointerface is one of the most actively studied systems among the $SrTiO_3$-based heterostructures, since a quasi-2D, high mobility electron gas forms at the interface [1]. A potential advantage of this system is the presence of modulation doping, as schematically shown in Fig. 1. The unreconstructed $LaAlO_3/SrTiO_3$ heterostructure has a *polar discontinuity*, which creates a diverging electrostatic potential [Fig. 1(b)]. This discontinuity can be resolved by an electronic reconstruction [Fig. 1(c)]. This electrostatic doping may enhance the mobility compared to chemical doping, since there is no scattering by ionized impurities. However, the doping mechanism at the $LaAlO_3/SrTiO_3$ interface is still controversial, and therefore it is currently unclear whether the potential advantage mentioned can be exploited or not. It is also not clear how to control the carrier density in this system, which is essential, since the electronic properties of $SrTiO_3$ are strongly sensitive to such changes.

It has been reported that the properties of this system can be modulated by growth parameters; for example, $LaAlO_3$ film thickness [2], oxygen pressure during growth and postannealing treatment [3]. However, systematic studies are still very limited. In particular, the effect of the laser parameters in pulsed laser deposition has not been discussed in the literature, although it is known to strongly influence properties in oxide materials [4]. In this study, we investigate the impact of growth parameters on the electronic properties of this system in detail.

Experimental details

$LaAlO_3$ films were grown on TiO_2-terminated $SrTiO_3$ (001) substrates by pulsed laser deposition using a KrF excimer laser. The variable growth parameters were: the $LaAlO_3$ film thickness, the laser spot size, the laser fluence, and the growth temperature (T_g). Before growth the substrates were preannealed at 950 °C in an oxygen pressure of 5×10^{-6} Torr for 30 mins. Following this anneal, the substrates were cooled to T_g (variable) and the oxygen pressure was increased to 1×10^{-5} Torr, the film growth conditions. The $LaAlO_3$ thickness was monitored using *in situ* reflection high-energy electron diffraction. After growth the samples were cooled to room temperature with the chamber filled by 300 Torr of oxygen, with a 1 hour pause at 600 °C (laser parameter series) or 500 °C (growth temperature series).

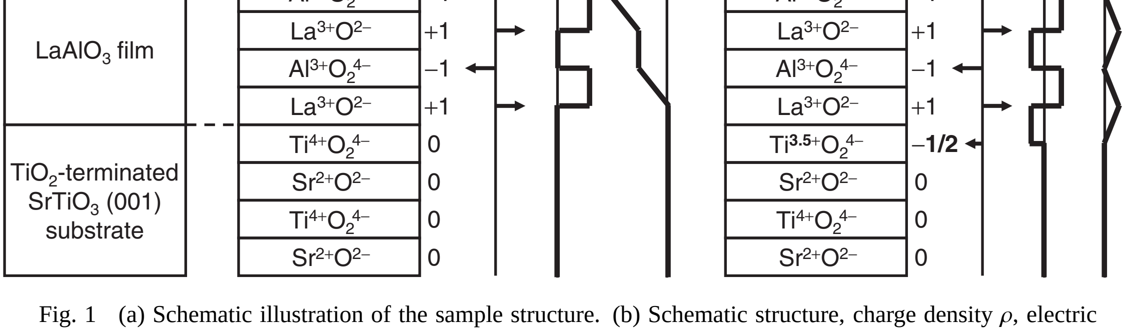

Fig. 1 (a) Schematic illustration of the sample structure. (b) Schematic structure, charge density ρ, electric field E and electrostatic potential V of the unreconstructed $LaAlO_3/SrTiO_3$ heterostructure. (c) Those of the reconstructed $LaAlO_3/SrTiO_3$ heterostructure via charge transfer. Adapted from Nakagawa *et al.* [5].

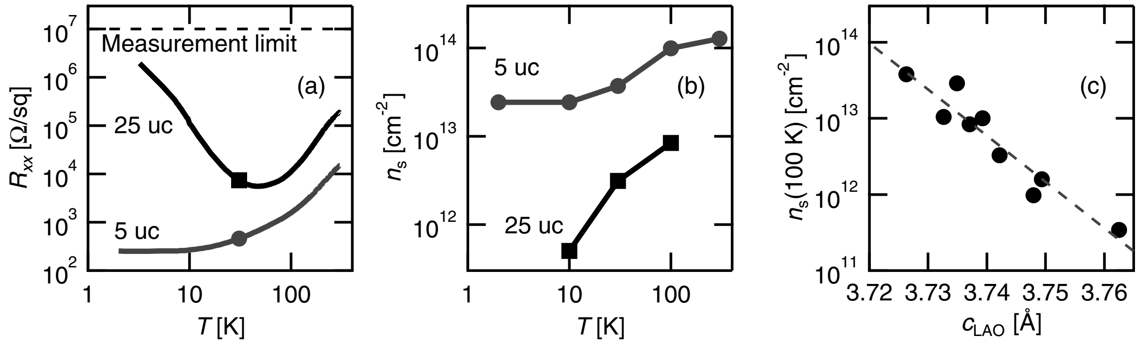

Fig. 2 Temperature dependence of (a) the sheet resistance and (b) the sheet carrier density of 5 and 25 uc samples grown at 0.7 J/cm^2 fluence with 5.6 mm^2 spot area. Lines in (b) are guides to the eye. (c) Sheet carrier density at 100 K of 25 uc samples grown at different laser conditions, as a function of the out-of-plane lattice constant of the $LaAlO_3$ films. Dashed line is a guide to the eye.

Transport measurements were carried out in a Physical Property Measurement System using a standard Hall bar geometry. The samples were electrically contacted using an ultrasonic wirebonder with Al wire. Film lattice constants were evaluated using the (002) peak of the x-ray diffraction θ–2θ patterns.

Results and discussions

I. Laser parameter dependence of the interface properties

First we examined the effect of the laser parameters at constant T_g = 800 °C. Figure 2(a) shows the temperature dependence of the sheet resistance of the samples with $LaAlO_3$ thicknesses of 5 and 25 unit cells (uc), grown at a fluence of 0.7 J/cm^2 with a spot area of 5.6 mm^2. The thinner sample shows metallic behavior down to 2 K, whereas the thicker sample shows an upturn in the resistance at low temperature. The high resistivity of the thicker sample is found to be caused mainly by the decrease of the carrier density [Fig. 2(b)].

To examine the possible carrier density modulation by the laser parameters, 25 uc $LaAlO_3/SrTiO_3$ samples were grown in 9 different laser conditions, varying the total laser energy and the spot size. We also examined the possible modulation of the cation stoichiometry of the $LaAlO_3$ film, similar to that reported in Ref. [4] for $SrTiO_3$. The film lattice constant was used as a parameter of the film stoichiometry, since the cation vacancies in the insulator are known to cause lattice expansion due to Coulomb repulsion [4].

Figure 2(c) shows the sheet carrier density at 100 K of the 25 uc samples, as a function of the out-of-plane lattice constant of the $LaAlO_3$ films. A strong modulation of the carrier density is observed, and the samples with larger film lattice constants are found to have lower carrier densities. These results suggest that the $LaAlO_3$ film stoichiometry is an important control parameter of the conductivity at the $LaAlO_3/SrTiO_3$ interface, which can be tuned by the laser parameters.

The 25 uc samples are found to be often highly resistive at low temperatures, as seen in Fig. 2(a), especially when the carrier density is low. Correspondingly the mobility at the lowest temperature is low, consistent with the previous report [2], and therefore the thinner samples are more suitable to investigate quantum transport in this system. To control the carrier density and the mobility of thinner $LaAlO_3/SrTiO_3$ samples, we next focused on the effect of the growth temperature.

II. Growth temperature dependence of the interface properties

Quite recently, it has been reported that the low-temperature growth enhances the mobility at the $LaAlO_3/SrTiO_3$ interface [6]. Motivated by this report, we examined the effect of the growth temperature on the interface properties. The film thickness, the laser spot size and the fluence were fixed at 10 uc, 5.6 mm^2 and 0.7 J/cm^2, respectively.

Figures 3(a) and (b) shows the temperature dependence the sheet carrier density and the Hall mobility of the samples grown at various T_g. The reduced growth temperature decreases the carrier density and increases the mobility at low temperature. The mobility of the sample grown at T_g = 600 °C is found to be high enough to observe the Shubnikov-de Haas oscillations, as shown in Fig. 3(c), qualitatively consistent with Ref. [6].

III. Discussions

It should be noted that the $LaAlO_3$ film itself is a robust insulator, and therefore the effect of the film growth parameters on the conductivity is far from obvious. For example, no strong stoichiometry dependence of the carrier density can easily be explained purely within the polar discontinuity picture, since it is based on the very fundamental physics of this system. On the other hand, other possible origins actively discussed, for example that the $LaAlO_3$ film growth provides some chemical dopants, is likely to fail to explain the thickness dependence.

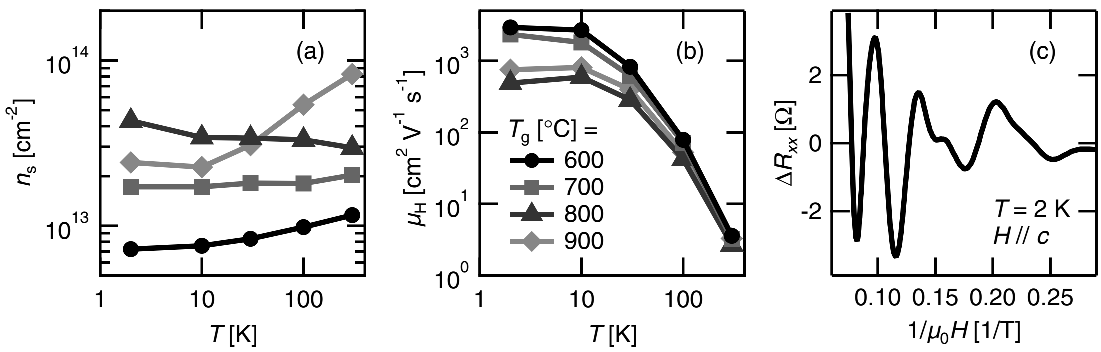

Fig. 3 Temperature dependence of (a) the sheet carrier density and (b) the Hall mobility of 10 uc samples grown at various T_g. Lines are guides to the eye. (c) Magnetoresistance of the sample grown at 600 °C, after background subtraction, versus reciprocal magnetic field. Measured at 2 K with magnetic field applied parallel to the substrate normal.

Such doping mechanisms are potentially enhanced by growing the film thicker, which is contradictory to the experimental results. Another possible explanation is that there is an increase of the numbers of traps with the film thickness and/or stoichiometry, rather than a suppression of the doping mechanism itself. A candidate for the origin the traps might be dislocations near the interface, induced by the strain energy in the $LaAlO_3$ film. This could explain the thickness dependence and the importance of the film lattice constant.

While the laser parameters decrease the carrier density and the mobility at the same time, the reduced growth temperature increases the mobility with decreasing the carrier density. This suggests that the underlying mechanisms of the conductivity modulation are different. A possible explanation of the enhancement of the mobility is the suppression of intermixing, which can be thermodynamically reasonable. This scenario still has problems, however, especially that this does not answer how the polar problem is solved. More detailed structural studies near the interface, by surface x-ray diffraction, for example, should give more insight into these open questions.

Conclusion and perspectives

We demonstrated a strong impact of the growth parameters on the electronic properties of the $LaAlO_3/SrTiO_3$ heterointerface. By controlling the laser parameters and the growth temperature, the electronic properties of this system were modulated over a wide range. Further reduction of the carrier density and the enhancement of the mobility, with careful optimization of these parameters, may enable the observation of the quantum Hall effect. This would give access to 2D electron physics in an unexplored regime, since this system has many characteristics that are absent in conventional semiconductors, especially notable being low carrier density superconductivity [7].

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Presentations and publication

A) H. Sato, T. Higuchi, Y. Hikita, and H. Y. Hwang, JSAP 70th Autumn Meeting, Toyama, Japan (2009).
B) H. Sato, T. Higuchi, C. Bell, Y. Hikita, and H. Y. Hwang, JSAP 57th Spring Meeting, Kanagawa, Japan (2010).
C) H. Sato, T. Higuchi, Y. Hikita, and H. Y. Hwang, 2010 MRS Spring Meeting, San Francisco CA, USA (2010).
D) H. Sato, J. A. Mundy, T. Higuchi, Y. Hikita, C. Bell, D. A. Muller, and H. Y. Hwang, 'Fabrication of bulk-like single crystal $LaAlO_3$ thin films on $SrTiO_3$ (001) using "$SrAlO_x$" buffer layers,' in preparation.
* Two more presentations (1 international, 1 domestic) as a coauthor.
** Two more presentations (1 first author, 1 coauthor) to be presented at the JSAP 58th Spring Meeting, Kanagawa, Japan (2011).