Transport Behaviour of $La_{0.8}Sr_{0.2}AlO_3$ Thin Film on Oxygen Deficient $SrTiO_3$ Substrate [*]

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$La_{0.8}Sr_{0.2}AlO_3$ (LSAO) thin films are grown on $SrTiO_3$ (STO) and MgO substrates by laser molecular beam epitaxy. The LSAO thin film on oxygen deficient STO substrate exhibits metallic behaviour over the temperature range of 80–340 K. The optical transmittance spectrum indicates that the LSAO thin films on MgO substrate are insulating at room temperature. The transport properties of LSAO thin films on STO substrates deposited in different oxygen pressure are compared. Our results indicate that oxygen vacancies in STO substrates should be mainly responsible for the transport behaviour of LSAO thin films.

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Perovskite oxide LaAlO3 (LAO) has been investigated extensively for its significant scientific interests and technological applications.[1−3] LAO is a potential candidate of high-K gate dielectric in metal oxide semiconductor field-effect transistors,[4,5] and also an appropriate substrate for superconducting microwave devices as well as dielectric resonators.[6] In addition, Sr-doped LAO has been investigated as an electrochemical material for sensors and solid oxide fuel cells since it exhibits p-type ionic electrical conduction at high temperature.[7,8] Recently, it is observed that there are polar discontinuity and high electrical conductivity at the interface of LAO/SrTiO₃ (LAO/STO) heterostructure.[9−13] It is worth further noting that a long-term investigation into STO crystal has revealed that a low annealing oxygen pressure greatly influences the transport behaviour and optical transmission of STO substrate.[14−18] Therefore, the characteristic of the LAO-based thin films grown on STO in low oxygen pressure could differ from the insulating bulk due to the change in the property of STO substrate as well as the interface effect.

In this work, we have prepared $La_{0.8}Sr_{0.2}AlO_3$ (LSAO) thin films on STO and MgO substrates, and investigated the transmittance property and transport behaviour of the LSAO thin films. The LSAO films on STO substrates exhibited metalic behaviour. Our results indicate that oxygen vacancies in STO substrates should be mainly responsible for the transport behaviour of the LSAO thin films.

In the experiment, LSAO thin films were grown on (001) STO and MgO substrates in thickness of 0.5 mm by laser molecular beam epitaxy (laser-MBE). After the cleaned STO and MgO wafers were transferred into the epitaxial chamber, XeCl excimer laser beam (about $1.5\,J\cdot cm^{-2}$, 2 Hz, about 20 ns) was irradiated to an LSAO target. During the deposition, the oxygen partial pressure and the substrate temperature was 5×10^{-3} Pa and 610°C, respectively. After the deposition of 220-nm-thick LSAO thin films, the films were *in situ* annealed for 40 min. The LSAO/STO samples became dark grey in colour, while the LSAO/MgO samples maintained transparent colour after the deposition. The crystallization of the LSAO films on the STO substrate was in situ monitored by reflection high-energy electron diffraction (RHEED). The phase formation and crystal quality of the LSAO films on STO substrates were identified by an x-ray diffractometer with Cu Kα radiation. In this work, indium (In) electrodes of about $1.0\,mm^2$ in size were used to measure the transport behaviour of the LSAO thin film and the LSAO/STO structure. The resistance-temperature $(R - T)$ characteristic of the LSAO film was measured by four-probe method. The optical property of the LSAO thin film on the MgO substrate was measured by an automated scanning monochromator (ARC SpectraPro-500I) at room temperature.

Figure 1 shows the x-ray diffraction (XRD) spectrum of the LSAO thin film on (001) STO substrate. Except for the LSAO and STO peaks, no other diffraction peaks from randomly oriented grains or impurity phases were detected, which indicates that the LSAO thin film is in single phase with (001) orientation. The sharp RHEED pattern of the LSAO thin film shown in the inset of Fig. 1 denotes that the LSAO thin film has a smooth surface and a good crystal structure.

Figure 2 shows the temperature dependence of the resistance of the LSAO thin film on the STO substrate

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over the temperature range 80–340 K. The increase of resistance with rising temperature indicates a metallic behaviour of the LSAO thin film on the STO substrate.
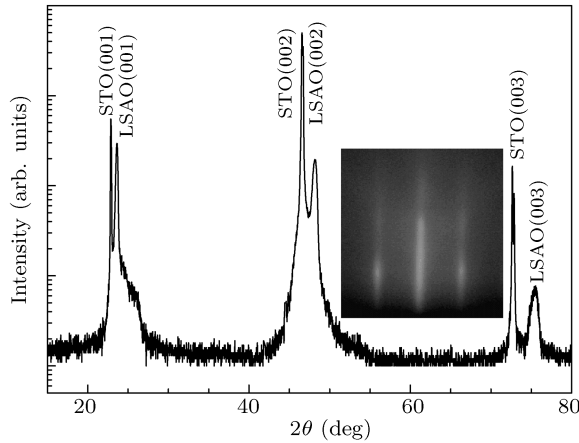

Fig. 1. X-ray diffraction (XRD) spectrum of LSAO films on (001) STO substrates. Inset: RHEED pattern of the LSAO thin film on the (001) STO substrate.
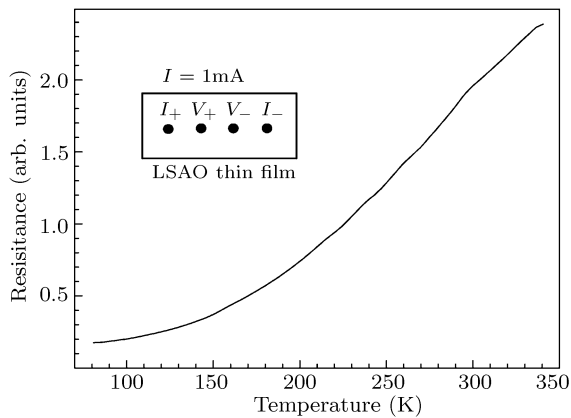

Fig. 2. $R-T$ behaviour of the LSAO film over the temperature range 80–340 K. Inset: the schematic measurement circuit.

Hall measurement of the LSAO thin films on the STO substrate was also carried out at room temperature. The carrier concentration, the resistivity and the Hall coefficient of the LSAO thin film was about 1.97×10^{21} cm^{-3}, 6.65×10^{-5} Ω·cm and -3.12 cm^3/C, respectively. The sign of the hall coefficient indicates an n-type conduction mechanism of the LSAO film at room temperature, which is different from the p-type ionic conductivity of LSAO bulk above 800°C.[8]

It should be mentioned that we have not observed the electrical transport behaviour of the 220-nm-thick LSAO films on MgO substrates because of its poor electrical conduction at room temperature. In order to reveal the property of the LSAO thin film on MgO substrates, we measured the optical transmittance spectrum of the LSAO thin film in the ultra-violet and visible region, and the result is shown in Fig. 3. High optical transmission efficiency up to 90% was obtained in the visible region. The direct allowed band gap of about 5.3 eV was estimated from the plot of $(\alpha h\nu)^2$ against $h\nu$ as shown in the inset of Fig. 3, which is close to that of LAO single crystal (5.5 eV, Ref. [19]). This result indicates that the LSAO thin film on MgO is insulator at room temperature, which is consistent with the electrical measurement.

Based on the above discussion, we can find that the conduction state of the LSAO films drastically changed from the metallic to insulating when grown on different substrates. In order to clarify the transport mechanism of LSAO thin films on STO substrates, we grew LSAO thin films on STO substrates in various oxygen pressures. We did not observe the electrical conduction of the LSAO thin film on the STO substrate deposited at relatively high oxygen partial pressure (above 2.4×10^{-2} Pa) due to high resistance of the LSAO thin films on transparent STO substrates. When the colour of the STO substrate changes from initially transparent white to dark grey after growth of the LSAO thin film, we can observe the electrical conduction behaviour of the LSAO thin film. This could be understood as follows: following the deposition of the LSAO thin film, the colour change of the STO substrate is related to the development of oxygen vacancies in STO.[13,16,18] Oxygen vacancies in the STO substrate are produced in the low oxygen deposition pressure of 5×10^{-3} Pa at a high temperature of 610°C during the growth of LSAO thin films. According to the lattice-charge neutrality condition, a part of Ti^{4+} ions in STO will be reduced into Ti^{3+} ions. The appearance of oxygen vacancies and $3d^1$ electrons of Ti^{3+} ions in STO could be responsible for the observation of the n-type electrical conduction behaviour of the LSAO thin film.
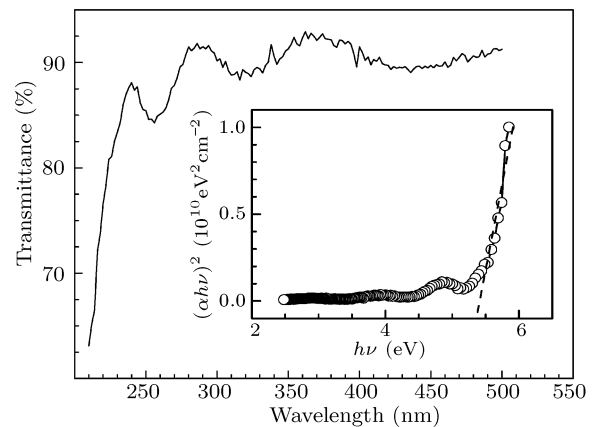

Fig. 3. Transmittance spectra of the 220-nm-thick LSAO thin film on the (001) MgO substrate.

Taking into account the conductivity of the STO substrate, the electrical behaviour of the LSAO thin

film would give a gross estimate of LSAO/STO structure. When two oxide semiconductors with different Fermi levels contact, carrier electrons will transfer from the high-Fermi-level material to the low-Fermi-level material, and carrier holes transfer in the opposite direction. Then the space charge region will be built near the interface of heterostructures to maintain charge equilibrium, which can result in some interesting transport behaviour in oxide heterostructures.[20]
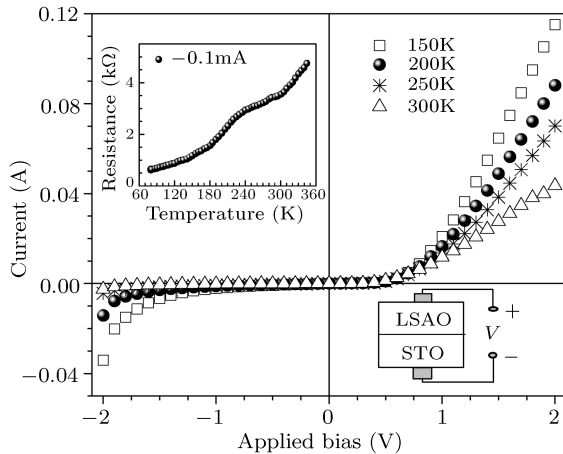

Fig. 4. The rectifying property of the LSAO/STO heterostructure over the temperature range 150–300 K. The schematic measurement circuit and the $R - T$ behaviour of the LSAO/STO heterostructure are shown in the top bottom insets, respectively.

Thus we also measured the current-voltage $(I - V)$ relationship of the LSAO/STO heterostructure over the temperature range of 150–300 K with a pulse-modulated voltage source as shown in Fig. 4. The schematical measurement circuit is shown in the bottom inset of Fig. 4. The LSAO/STO heterostructure exhibits a rectifying property. Interestingly, the current of LSAO/STO heterostructure decreases with increasing temperature, which is different from the previous observations in other oxide junction.[21] We have also measured the $R - T$ curve of LSAO/STO heterostructure under an applied reverse current of 0.1 mA over the temperature range of 140–340 K, as shown in the top inset of Fig. 4. The metallic behaviour observed in the LSAO/STO heterostructure could be responsible for the temperature-dependent $I - V$ relationship. In order to clarify the origin of the rectifying behaviour of LSAO/STO heterostructure, we measured the $I - V$ characteristic of the STO substrate annealed at the same preparation condition as for preparing LSAO thin film, and observed a linear relationship at room temperature (not shown here). This result indicates that the rectifying property of the LSAO/STO heterostructure actually originates from the junction interface rather than the contact between the metallic indium electrode and the induced STO substrate.

In summary, we have prepared LSAO thin films on (001) STO and MgO substrates by laser-MBE. The LSAO thin film grown on a MgO substrate exhibits an insulting behaviour, which is identified with a direct allowed band gap of about 5.3 eV obtained from optical transmittance spectrum. This result indicates the insulator characteristic of LSAO on the MgO substrate. However, the LSAO thin film on an STO substrate shows a metallic behaviour. The comparison of the transport property of the LSAO thin films on STO substrates prepared in different oxygen pressures reveals that oxygen vacancies in STO substrates should be responsible for the electrical conduction behaviour of the LSAO thin film. In addition, the LSAO/STO heterostructure exhibits a rectifying behaviour over the temperature range 150–300 K due to the interface effect.

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