Photoelectric property of $LaAlO_{3-\delta}$/Si heterojunctions with different oxygen contents*

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Three oxide heterojunctions made of $LaAlO_{3-\delta}$/Si are fabricated under various oxygen pressures by laser molecular-beam epitaxy. They all show nonlinear and rectifying current–voltage characteristics, and the distinct difference in rectification behaviour among them. Their photoelectric properties are examined by a visible HeNe laser and an ultraviolet Hg lamp. We find that their photovoltaic responses are closely related to the oxygen contents in the $LaAlO_{3-\delta}$ films. The junction fabricated under the lower oxygen pressure has a higher photovoltaic sensitivity. The possible mechanism is suggested based on the band structure of the p–n heterojunction.

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1. Introduction

Perovskite oxide thin films have received much attention because of their remarkable dielectric, ferroelectric, piezoelectric, colossal magnetoresistance, nonlinear optics and many other interesting properties.[1−8] Oxygen content in thin film is one of the most important parameters which dramatically affect the physical properties of perovskite oxides.[9−12] For example, the structural, the electrical transport and the magnetic properties of manganite thin films are closely related to the oxygen vacancies.[10,11] $LaAlO_3$ (LAO), as a member of perovskite family, has been intensively investigated due to its promising application in high-k gate dielectric field.[13,14] Stoichiometric $LaAlO_3$ is an insulator, however, its electrical conduction can be significantly enhanced if it is grown in a deoxidization environment. Previously, Xiang *et al.*[14] have already experimentally investigated the effects of oxygen content on the current–voltage (I–V) and capacitance–voltage (C–V) characteristics of $LaAlO_{3-\delta}$/Si heterojunction. Also Huang *et al.*[15] have reported on the electric rectification, ferroelectricity and photovoltaic effects found in $LaAlO_{2.73}$/Si heterojunction. However, to our best knowledge, the dependence of the photoelectric property of $LaAlO_{3-\delta}$/Si heterojunction on oxygen content in LAO thin film has not ever been reported. In this study, we report on the rectificative and photoelectric characteristics of $LaAlO_{3-\delta}$/Si junctions and their dependence on oxygen content in LAO film.

2. Experimental details

$LaAlO_{3-\delta}$ thin films were grown on p-type Si (100) substrates by laser molecular beam epitaxy under different oxygen pressures. The three samples were named as HOP (fabricated under high oxygen pressure), MOP (fabricated under middle oxygen pressure), and LOP (fabricated under low oxygen pressure) according to their fabrication oxygen pressures of 1×10^{-1} Pa (with active oxygen), 2.8×10^{-2} Pa, and 7.8×10^{-4} Pa, respectively. The substrate temperature was kept at 560 °C. The three films were 5×5 mm^2 in size and ∼400 Å in thickness. A detailed deposition procedure can be found in previous reports.[14,15] Both reflection high-energy electron diffraction patterns and x-ray diffraction measurements have confirmed that the LAO films were amorphous. The electrical characteristics were measured using a Keithley 2400 sourcemeter. The photoelectric properties were analysed by using a visible HeNe laser and an ultraviolet (UV) Hg lamp. The photoelectric signals were

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recorded by a 500 MHz digital oscilloscope with an input impedance of 1 MΩ.

3. Results and discussion

Figure 1 shows the I–V characteristics of samples HOP, MOP, and LOP measured in the darkness at room temperature. The I–V curves of the three samples all exhibit asymmetry with respect to the zero bias point, and significant difference in rectification behaviour among them. For sample HOP, the leakage current occurring in the reverse bias region is small (0.77 μA at −3 V bias), and the ratio of forward current to reverse current approaches 230 in the applied voltage region from 3 V to −3 V. In contrast, sample LOP displays a big leakage current at negative bias, reaching 24 μA at −3 V bias. Here the current ratio of 3 V forward to reverse bias is only 3. With the oxygen content in LAO film decreasing, the leakage current of junction increases and the rectification characteristic becomes worse.

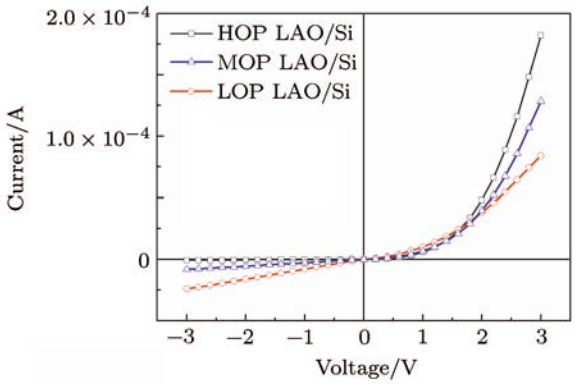


Fig. 1. (Colour online) I–V curves of samples HOP, MOP, and LOP measured in the darkness at room temperature.

Figure 2 displays the current–voltage behaviours of the three LAO/Si junctions with/without light illumination at room temperature in semi-logarithmic scale. Open and solid symbols denote the data measured in the darkness and under illumination, respectively. In the measurement, the HeNe laser with a wavelength of 632.8 nm and a light power density of 1 mW/mm^2 was adopted as an illumination light source. For samples LOP and MOP, a high photocurrent in the reverse bias under illumination can be clearly observed on the I–V curves, while the corresponding photocurrent for sample HOP is much lower. It indicates that the photoelectric responses of samples LOP and MOP to HeNe laser illumination are more sensitive than that of sample HOP.

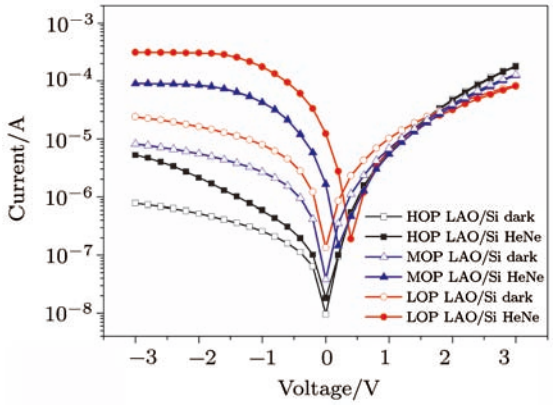


Fig. 2. (Colour online) I–V curves of samples HOP, MOP, and LOP compared between under illumination (solid symbols) and in the darkness (open symbols).

To make a clearer comparison of the photoelectric response among the three samples, the open-circuit photovoltage (PV) was recorded directly by an oscilloscope. Figure 3 shows the open-circuit photovoltaic signals under illumination of the HeNe laser. The area of laser spot was 7 mm^2. As can be seen from this figure, the PVs are 55, 136, and 432 mV for samples HOP, MOP, and LOP, respectively. With the decrease of oxygen content in the LAO thin film, the PV of the LAO/Si junction increases evidently, which is consistent with the result from I–V measurement as shown in Fig. 2. To further verify the photoelectric property, we used a UV Hg lamp with a wavelength of 253.65 nm to illuminate the samples. The UV light intensity was 0.53 mW/cm^2 and the light spot area was 25 mm^2. In this case, as displayed in Fig. 4, the PVs of 19, 78, and 200 mV were obtained for samples HOP, MOP and LOP, respectively.

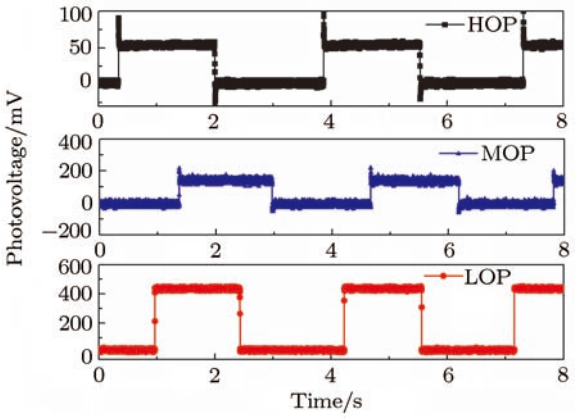


Fig. 3. (Colour online) Photovoltages of samples HOP, MOP, and LOP under illumination of a visible HeNe laser.

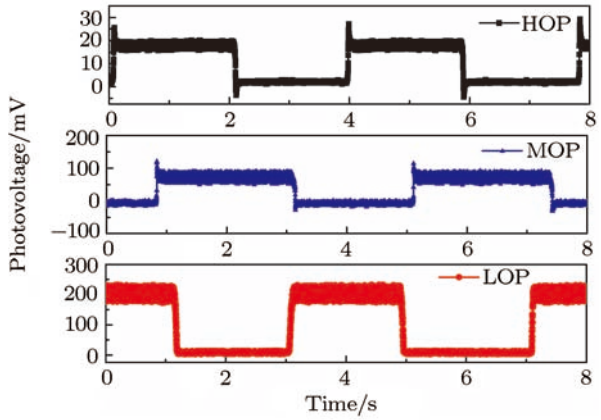


Fig. 4. (Colour online) Photovoltages of samples HOP, MOP, and LOP under illumination of a UV Hg lamp.

From Figs. 3 and 4, we can ascertain that the photoelectric efficiency of sample LOP is higher than that of HOP. Similar results have been reported in our previous papers on $BaTiO_{3-\delta}$/Si and $SrTiO_{3-\delta}$/Si.[12,16] Besides, a more interesting thing is that the PV signals of samples HOP and MOP each have a shoulder-like transient transition when the light is turned on or off. However, the PV signal of sample LOP does not show such a feature at all. In order to understand the involved mechanism, we constructed a schematic band structure for samples HOP and LOP as presented in Fig. 5. As we all know, samples fabricated at lower oxygen pressure usually have more oxygen vacancies and so sample LOP has the most oxygen vacancies and sample HOP has the least oxygen vacancies. In forming a LAO/Si heterojunction, electrons with higher density in n-type LAO film than in Si should diffuse into Si, whereas holes with a higher density in Si than in LAO should diffuse into LAO. The diffusion causes a built-in electric field in the space charge region around the interface, which is how a barrier is built up at the interface as shown in Fig. 5.[17] An energy spike is also established at the interface due to the mismatch between the Fermi levels and band structures of the two semiconductors.

As mentioned above, sample HOP was deposited under a higher pressure of 1×10^{-1} Pa (with active oxygen). As a result, the oxygen content in sample HOP is nearly stoichiometric and the carrier concentration is very low.[14] While sample LOP is quite different from sample HOP, as it was fabricated at 7.8×10^{-4} Pa and its electron concentration is about $6.43\times10^{20}/\mathrm{cm}^3$. The impurity levels introduced by the oxygen vacancies are distributed in the bandgap of conduction band and valence band of LAO, as schematically plotted as the shadow parts in Fig. 5. According to the semiconductor theory, the energy spike width of sample LOP should be narrower due to its higher carrier concentration.

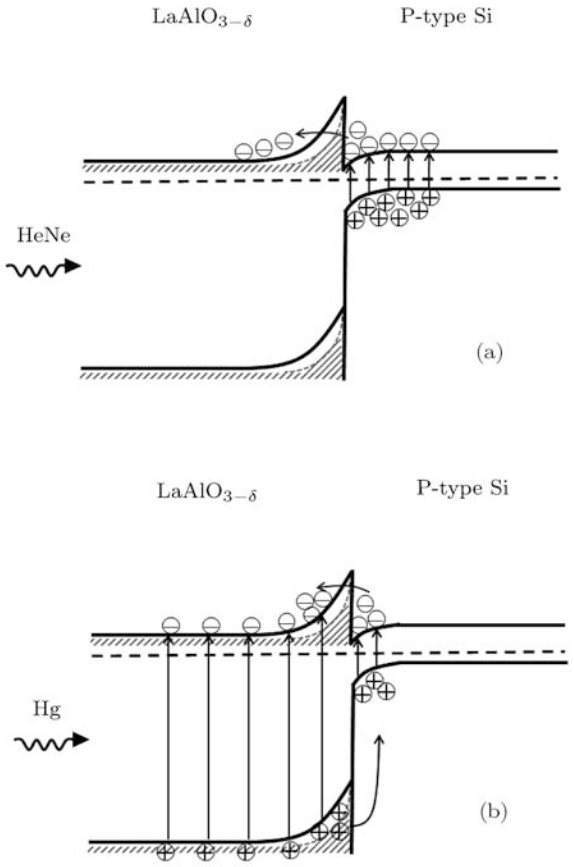


Fig. 5. Schematic band structures and carrier movement pictures for samples HOP and LOP under illumination of (a) a HeNe laser and (b) a Hg lamp. The conduction band and the valence band of sample HOP are denoted by black solid lines and those bands of sample LOP are indicated by gray dashed lines.

When a HeNe laser is shone on the film surface of sample LOP, the LAO thin film is transparent due to its wide bandgap. Therefore the HeNe laser photons can directly pass through the thin film to enter the underlying Si substrate. On the Si side, the photons are absorbed completely to generate electron–hole pairs. The photogenerated electrons will drift to the interface driven by the built-in field and tunnel through the interfacial energy spike into the LAO film, while the photogenerated holes will be confined on the Si side (see Fig. 5(a)). Thus a photovoltage between the two electrodes is formed.[18] While, under the irradiation of a Hg lamp, the photon-induced carriers can be created not only in LAO thin film but also in Si substrate, as the photon energy is larger than the bandgaps of both LAO and Si. In this case, the photoinduced holes generated in the LAO film will drift

into the Si substrate by the built-in potential, and the photoinduced electrons generated in Si substrate will travel to LAO film. Bidirectional motion and separation of the electrons and the holes contribute to the photovoltage (see Fig. 5(b)). In comparison with the case of the HeNe laser irradiation, it is easy to understand that the smaller PV is induced by the Hg lamp, since the photon flux of the Hg lamp is much lower than that of the HeNe laser.

As mentioned above, our results indicate that the heterojunction of LAO/Si with more oxygen vacancies in LAO film always has a bigger PV, with either a HeNe laser or a Hg lamp used as a light source. We speculate that this may be related to the number of impurity levels in the band gap of LAO. More oxygen vacancies in sample LOP should produce more impurity levels.[19] These impurity levels act as trap centres which can trap electrons and deter their recombination with holes. Hence, the lifetimes of photogenerated electrons and holes in sample LOP are much longer than those in sample HOP. In sample LOP, the electrons and holes have much more time to diffuse or drift into the space charge region before recombining. This conjecture has been further verified by the open-circuit PV signals recorded by an oscilloscope. As shown in Figs. 3 and 4, the PV signals of sample HOP each have a pulsed transient process when the light is switched on or off. This transient process actually reflects a recombination process of photoinduced electrons and holes. The lifetime of carriers in sample HOP is very short, so the majority of carriers are recombined quickly upon their generation. Naturally, the shorter the photocarrier lifetime is, the fewer the carriers that are separated by the built-in field in space charge region will be, and then the lower the obtained PV will be. Therefore sample HOP has a lower PV than sample LOP. Another factor, i.e., tunneling possibility, should also be considered in the process of charge transferring at the interface. As stated above, the width of energy spike of sample LOP is much narrower due to its higher carrier concentration. As a result, the photo-generated electrons in Si substrate have a larger possibility to tunnel through the energy barrier. It makes the carrier transport easier in sample LOP than in sample HOP.

4. Conclusions

The photoelectric properties of the three LAO/Si heterojunctions with different oxygen content in LAO films are investigated in detail. It is found that the LAO/Si heterostructure with a lower oxygen content has a higher PV and better photoconductivity We believe that this kind of oxygen content dependence is related to the lifetime of photocarriers. The impurity levels of LAO film should play an important role in the process of charge transferring at the interface of the LAO/Si heterojunctions.

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