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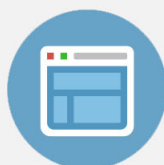
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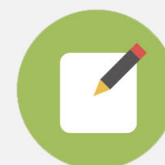


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Photoelectronic behaviors of bilayer ultrathin films manganite-based heterojunctions

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We presented a systematic study on the photoelectronic properties of the $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3$ (20 nm)/ $\text{LaMnO}_3(t)/\text{SrTiO}_3:0.05 \text{ wt. \% Nb}$ (LBMO/LMO(t)/STON) junctions with $0 \leq t \leq 30 \text{ nm}$. The short-circuit photocurrent (I_{ph}) is found to show a complex dependence on the LMO buffer layer. It undergoes first a sharp drop as the layer thickness of LMO increases from 0 to 3 nm and then, after a rigid turn, a slow decrease for further increase in layer thickness. These results indicate that the coupling between LBMO and STON can be effectively depressed by a LMO layer of 3 nm. The photocurrent is further found to be temperature dependent, increasing monotonically upon cooling, and the maximal growth, occurring in the junction of $t = 3 \text{ nm}$, can be as high as 226% when cooled from 320 K to 40 K. Meanwhile, the $I_{\text{ph}}-t$ dependences at different temperatures are similar, which is an indication of temperature independence for the diffusion length of the photocarriers. Analysis of the capacitance-voltage relations indicates that the change of interfacial barrier is the reason for the peculiar photoelectronic behavior observed.

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Manganite-based heterojunctions are typical systems composed of Mott insulators and possess diverse properties absent in conventional junctions, such as excellent rectifying properties,¹ bias-dependent magnetoresistance (MR)^{2,3} and peculiar photoelectronic behaviors.⁴⁻⁶ Among them, the photoelectronic effect is an important feature of heterojunctions. Based on this effect, the dynamic properties of the non-equilibrium charge carriers can be studied.

As well established, the magnetic state of the manganite experiences a complex variation with the decrease of temperature.⁷ Lattice strains, thus, Jahn-Teller distortions, can also have a strong impact on the physical properties of the manganites, leading to, for example, a magnetic and conducting dead layer near the film-substrate interface.^{8,9} The composition distribution from the interface to surface in the manganites is usually different.¹⁰ All these factors will strongly affect the excitation-recombination balance of the photocarriers, thus, the photoelectric effect of the junctions.

Ultrathin film junctions have unique properties that are exclusively determined by interfacial state.^{11,12} There are intensive efforts to modulate the physical properties of the junction by modifying interfacial state, for the purpose of either fundamental research or practical application. As

reported, tensile stress strain in the films can cause an obvious decrease of interfacial barrier height.¹³ Crystallographic orientation also significantly affects the interfacial barrier.¹⁴ The incorporation of a proper intermediate layer even yields an enhanced positive MR effect, which is totally different from the negative MR in simple junctions without buffer layer.^{15,16} In this paper, we will study systematically the influence of buffer layer on photoelectronic properties for the $\text{La}_{0.67}\text{Ba}_{0.33}\text{MnO}_3(20 \text{ nm})/\text{LaMnO}_3(t)/\text{SrTiO}_3:0.05 \text{ wt. \% Nb}$ (LBMO/LMO(t)/STON) junctions.

The LBMO/LMO(t)/STON junctions were fabricated by growing, via the pulsed laser ablation technique, first a LMO layer with a thickness between $t = 0$ and 30 nm and then a LBMO film of $\sim 20 \text{ nm}$ on (001) $\text{SrTiO}_3:0.05 \text{ wt. \% Nb}$ (STON) substrates. The LMO/STON junctions with the corresponding thicknesses were also prepared for comparison study. During the deposition process, temperature of the substrate was kept at 720°C and the oxygen pressure at $\sim 10 \text{ Pa}$, for the LMO film, and $\sim 60 \text{ Pa}$, for the LBMO film.

As electrodes, two copper (Cu) pads of the size of $1 \times 1 \text{ mm}^2$ were deposited, also by pulsed laser deposition, on the LBMO film and the STON, respectively. Appropriate electric pulses have been applied to the Cu-STON contact to get an Ohmic contact. The contact resistance is $\sim 15 \Omega$ for the Cu-STON and $\sim 200 \Omega$ for the Cu-LBMO contact at ambient temperature. A laser with a wavelength of 633 nm and a

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power of 5 mW was used in the present experiments. The spot size is ~ 0.33 mm in diameter. A Keithley SourceMeter 2611 was used for the acquisition of short-circuit photocurrent (I_{ph}). Temperature of the samples was controlled by a cycling cryostat with the lowest temperature of 20 K. The capacitance was measured by a ZM2353 LCR meter.

The surface topography of manganite films were measured by AFM. Figs. 1(a)–1(c) show the AFM images of the LMO layers with the thicknesses of 1 nm, 3 nm, and 15 nm, respectively. Terrace-structured surface structure with a step of ~ 0.4 nm is observed in all of the LMO layers, signifying a layer by layer growth of the films. The top LBMO film is also quite smooth, with a topography characterized by densely packed grains of 16 nm as shown in Fig. 1(d).

As well established, the J - V relations describe the transport properties of thermally equilibrium charge barriers. The incorporation of buffer layers obviously affects the interfacial state of the bilayer film junctions, modifying the interfacial barrier and depletion width^{13,16} and therefore generation and diffusion of extra charge carriers excited by laser illumination.¹⁷

Typical rectifying behaviors as previously reported are observed (not shown), which confirms the presence of built-in electric field at the interface of the junctions. Electrons in valence band can be excited by laser irradiance to conduction band when photon energy is high enough, and the photocarriers thus produced will be swept to the two sides of the depletion layer by internal field, forming photocurrent. It is obvious that from short-circuit photocurrent and open-circuit photovoltage we can get important information about photocarriers. Considering the fact that the electric leakage may affect the measurement of photovoltage, we will focus on the photocurrent below.

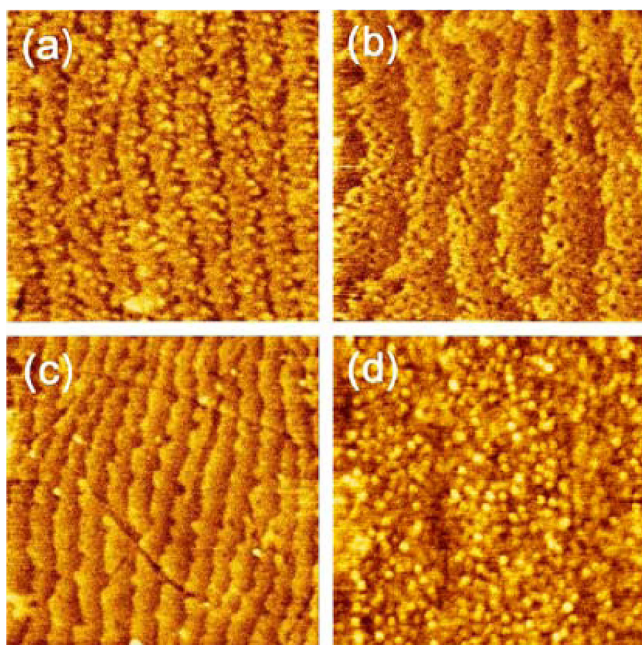


FIG. 1. (a)–(c) Surface topography of the LMO buffer layers with the thickness of 1 nm (a), 3 nm (b), and 15 nm (c). (d) The surface topography of the LBMO(20 nm)/LMO(3 nm) film. The scale of the image is $1 \times 1 \mu\text{m}^2$.

Figs. 2(a) and 2(b) show the influence of layer thickness of LMO on photocurrent of LMO(t)/STON and LBMO/LMO(t)/STON junctions. Complex dependences of I_{ph} on t were observed. With the increase of the LMO layer thickness, for the LMO(t)/STON junctions, the photocurrent first increases rapidly and then, after a maximum, decreases slowly, similar to the results previously reported.¹⁷

Mechanisms for the peculiar photoelectric behaviors of the bilayer junctions deserve further study. The introduction of a buffer layer will modify the interface structure of the junction, including the electronic structure of films and the interfacial barrier/depletion width of the junction, and therefore, the photoelectric process of junctions. According to the semiconductor theory, the information on the junction interface can be obtained from the capacitance (C)–voltage (V) characteristics. Fig. 3(a) displays the bias voltage dependence of the reciprocal square capacitance for the LBMO/LMO(t)/STON. Considering the effect of LMO, the built-in potential of LBMO/LMO(t)/STON is determined by the $1/n^2 C^2 - V/n$ relation, where n is the ideality factor of the junction.¹⁸ Fig. 3(b) presents the interface barrier as a function of layer thickness of LMO for the LBMO/LMO(t)/STON. With the increase of t , V_d undergoes successively a sharp increase, an abrupt drop, and a smooth variation. The maximal interface barrier is ~ 1.8 eV, occurring at $t \approx 3$ nm. Results obtained under different frequencies are similar. As a supplement, we would like to point out that an essentially similar V_d - t relation is obtained from the analysis of current-voltage characteristics. From Figs. 2(b) and 3(b), the correspondence between I_{ph} and V_d can be clearly seen: low interfacial barrier yields large photocurrent. It is obvious that the abnormal increase of V_d around $t \approx 3$ nm is jointly determined by LBMO, LMO, and STON, and it is an effect arising from the coupling between LBMO and LMO.

It is possible that the I_{ph} is jointly determined by the LBMO and LMO layers when the thickness of the latter is smaller than 3 nm. Photocarriers in the LMO layer can diffuse directly towards the LMO/STON interface, yielding photocurrent. However, for the photocarriers in the LBMO film, the presence of a LMO layer increases their distance from the LMO(t)/STON interface, and a part of them will recombine with holes on their way to the interface, leading to a rapid decrease in photocurrent. This may be the case happened in the thickness range from 0 to 3 nm. When the LMO layer is

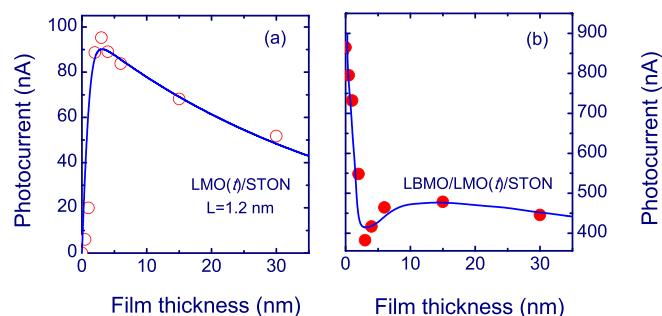


FIG. 2. Photocurrent as a function of LMO layer thickness for the LBMO/LMO(t)/STON (a) and LMO(t)/STON (b) junctions. The solid line in (a) is theoretical result and the solid line in (b) is a guide for the eye.

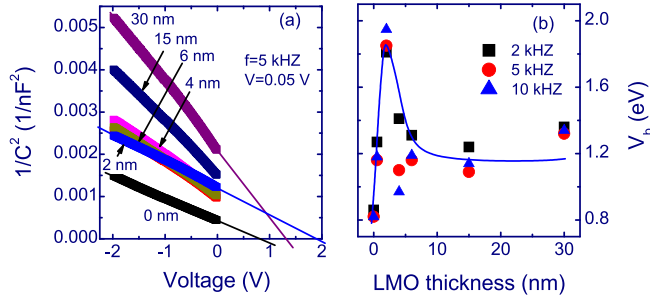


FIG. 3. (a) Reciprocal square capacitance as a function of bias voltage for the LBMO/LMO(*t*)/STON junctions with different LMO buffer layers and (b) built-in potential as a function of LMO layer thickness. Solid lines are guide for the eye.

thick enough, however, all of the photocarriers in LBMO will annihilate before reaching the LMO(*t*)/STON interface, and the I_{ph} - t relation is exclusively determined by the LMO(*t*)/STON junction. This may be the case for $t \gg 3$ nm. These results indicate that a LMO layer thicker than 3 nm completely depresses the LBMO-STON correlation, i.e., the diffusion distance of the photocarriers in LMO cannot exceed 3 nm. According to Fig. 2, the photocurrent of LBMO/LMO(*t*)/STON is obviously larger than that of LMO(*t*)/STON even when the LMO(*t*)/STON interface dominates the photoelectronic behavior for $t \gg 3$ nm. The LMO layer on STON may be different from that sandwiched by LBMO and STON since the latter has been exposed to an oxygen atmosphere of high pressure during the deposition of LBMO.

As reported, the physical state of the manganite usually experiences a variety of variation upon cooling. The effect of temperature on the photoelectronic properties of manganite junctions is therefore worthwhile. Fig. 4(a) shows the temperature-dependent photocurrent for the LBMO/LMO(*t*)/STON. The photocurrent of different junctions exhibits essentially similar dependence on temperature, growing monotonically with the decrease of temperature. For the temperature variation from 320 K to 40 K, the general increase in I_{ph} is $\sim 226\%$. In fact, there are two possible processes in the junctions under the irradiance of laser, i.e., photocarrier excitation and recombination. Based on the technique described in Ref. 17, a theoretical I_{ph} - t can be deduced after considering the decay of laser in the film and the diffusion of the photocarriers. The corresponding results are shown by the solid line in Fig. 2(a). The diffusion distance of the photocarriers deduced is ~ 1.2 nm for the LMO film. The photocurrent as a function of the LMO thickness is shown in Fig. 4(b). The I_{ph} dependences on t at different temperatures are similar, which may be an indication for the temperature independence of the diffusion distance of photocarriers.

To our knowledge, this is the first observation of the distinctive photoelectronic properties of bilayer manganite junctions, totally different from that of monolayer junctions.¹⁷ Incorporation of the LMO layer has considerably modified the interface state of the junction, giving non-equilibrium charge carriers' unique dynamic characteristics.

In conclusion, the photoelectronic properties of the LBMO/LMO(*t*)/STON with t of 0–30 nm have been studied

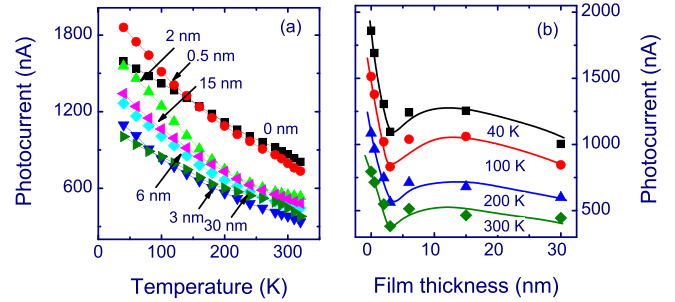


FIG. 4. Photocurrent as a function of temperature (a) or the LMO thickness (b) for the LBMO/LMO(*t*)/STON junctions. Solid lines are guide for the eye.

systematically. After introducing a LMO buffer layer, a complex dependence of photocurrent on LMO thickness was observed. For the LMO(*t*)/STON junctions, the photocurrent first increases rapidly then, after a maximum, decreases slowly with the increase of the LMO layer. In contrast, the LBMO/LMO(*t*)/STON junctions show completely different behaviors. The I_{ph} is maximal without buffer layer and decreases rapidly as t increases from 0 to 3 nm. Further increase in t yields a relatively rigid turn in the I_{ph} - t curve leading to a slow decrease of the photocurrent. These results indicate that a LMO layer above the t of 3 nm can effectively depress the coupling between LBMO and STON.

The photocurrent of the junctions displays similar dependence on temperature, regardless of the thickness of LMO. With the decrease of temperature, I_{ph} increases rapidly. For the temperature variation from 320 K to 40 K, the maximal increase is up to $\sim 226\%$. Besides, the I_{ph} dependences on t at different fixed temperature are similar, which indicates that diffusion distance of non-equilibrium carrier in films is independent of temperature. Based on the analysis of the C - V relations, the change of interfacial barrier may be the reason of peculiar photoelectronic behavior of junctions.

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