Ferroelectric Control of the Conduction at the $LaAlO_3/SrTiO_3$ Hetero-interface

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Abstract

The $LaAlO_3/SrTiO_3$ (LAO/STO) interface serves as a model system in which a highly mobile quasi-two-dimensional electron gas (2DEG) forms between two band insulators [1,2], exhibiting 2D superconductivity [3] and unusual magnetotransport properties [4]. In the study reported here, we added a ferroelectric $Pb(Zr_{0.2}Ti_{0.8})O_3$ (PZT) layer near the LAO/STO interface. The ferroelectric polarization of the PZT layer serves as a control parameter to modulate the 2DEG conducting behavior. The as-grown polarization (P_{up} state) leads to charge depletion and consequently low conduction. Switching the polarization direction (P_{down} state) results in charge accumulation and enhances the conduction at the LAO/STO interface. The origin of this modulation is attributed to a change in the electronic structure due to the ferroelectric polarization states, evidenced by X-ray photoelectron spectroscopy (XPS). Control of the conduction at this oxide interface suggests that the concept can be generalized for other oxide systems to design functional interfaces.

Keywords – $LaAlO_3/SrTiO_3$, 2DEG, ferroelectric $Pb(Zr_{0.2}Ti_{0.8})O_3$, transport, XPS.

Introduction

The LAO/STO interface is served as a model system in which a highly mobile quasi-two dimensional electron gas (2DEG) forms between two band insulators [1,2], exhibiting 2D superconductivity [3]. The ferroelectric polarization of PZT layer serves as a control parameter to modulate the 2DEG conducting behaviors. The as-grown polarization (P_{up} state) leads to charge depletion and consequently a low conduction. Switching the polarization direction (P_{down} state) results in a charge accumulation and enhances the conduction at the interface of LAO/STO. The origin of this modulation is attributed to a change in the electronic structure due to the ferroelectric polarization states, evidenced by x-ray photoelectron spectroscopy (XPS). Control of the conduction at this oxide interface suggests that the concept can be generalized for other oxide systems to design functional interfaces.

Experiments

In order to understand the influence of ferroelectricity on the LAO/STO interface experimentally, we firstly carried out electrical transport and XPS measurements on the heterostructure. In the ferroelectric PZT/LAO/STO heterojunction with a naturally upward PZT layer, the polarization was reversed by the scanning probe technique (probe voltage set to 8 V) to obtain a downward polarization area of about 1 mm^2 that is larger than x-ray beam size of about 400 μm^2. This ferroelectric-pattern assisted spectral technique makes us to study the polarization reversal using the same sample, not having to compare the polarization state in two different samples, avoiding all the discussions regarding sample quality and other differences.

Results

We firstly carried out electrical transport measurements on the heterostructure (Figure 1a), in which the PZT layer with the spontaneous polarization P_{PZT} functions as the polarized dielectric slab to modulate the conduction of LAO/STO heterointerface. Figure 1b shows the sheet resistance versus temperature (*R-T* curves) of the PZT/LAO/STO samples with various PZT layer thicknesses (0~40 nm), while the LAO thickness was fixed (6 u.c.). The R-T curves for these samples show that the sheet resistance at room temperature is low (50 KΩ/sq) and decreased with temperature, showing the metallic behavior. For the samples with PZT on top, the ferroelectric effect sets in and the sheet resistance starts increasing with PZT thickness, showing the strong impact of the intrinsic polarization (P_{up}) of PZT on electron conduction at the interface. This is anticipated since the ferroelectric field effect provides one more degree of freedom to compensate the charge unbalance at the interface[15]. We also conducted transport measurements on the samples with different LAO thickness, while PZT thickness was kept constant (20 nm) (Figure 1c). Several studies suggested a critical LAO thickness (4 u.c.) for the formation of 2DEG at this interface[7]. Under such a circumstance, all the samples with LAO above a critical thickness behave like a metal.

The sheet resistance is decreased with lowering the temperature and increased when the thickness of LAO is reduced.

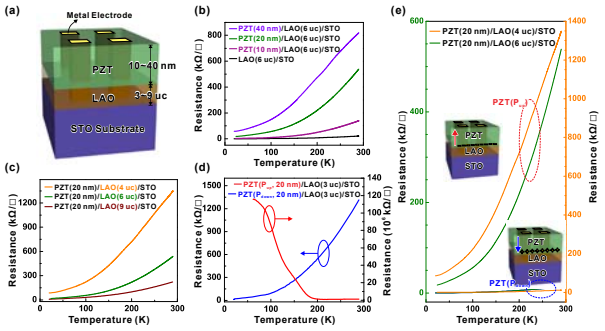

Fig. 1. Transport measurement of PZT/LAO/STO devices.

As depicted in Figure 2a, positive (P_{down} state) and negative (P_{up} state) bound charge sheets at bottom PZT layer affected at the conducting LAO/STO hetero-interface, leading to an accumulation or depletion of free electrons in STO layer. The schematics (Figure 2a) also shows the ferroelectric-pattern-assisted XPS technique on PZT (3.5 nm)/LAO (4 u.c)/STO heterjunction, the binding energy of the Sr core-level from buried layer thus decreases ($E_B^{Sr} - eV_{up}$) or increases ($E_B^{Sr} + eV_{up}$) depending on the downward and upward polarization directions, respectively, by comparing the binding energy of the Sr core-level (E_B^{Sr}) from LAO/STO heterojunction. Evidently, the concept of that interface capacitor is also confirmed from the lack of change of the core-level emissions from the top PZT (Pb 4*f* core-level) and LAO (La 4*d* and Al 2*s* core-levels) layers. Figure 2b shows the characteristic core-level photoelectron spectra in the ferroelectric PZT/LAO/STO and conducting LAO/STO heterojunction samples. The binding energy of Sr $3d_{5/2}$ core-level shifts associated with the potential drops of the PZT-modulated LAO/STO interface capacitors were obtained at eV values of +0.15 eV (P_{up} state) and −0.1 eV (P_{down} state), and the core-level difference between Sr $3d_{5/2}$ and La $4d_{5/2}$ (ΔE_{CL}) were determined of 31.15 eV (P_{up} state) and 30.90 eV (P_{down} state).

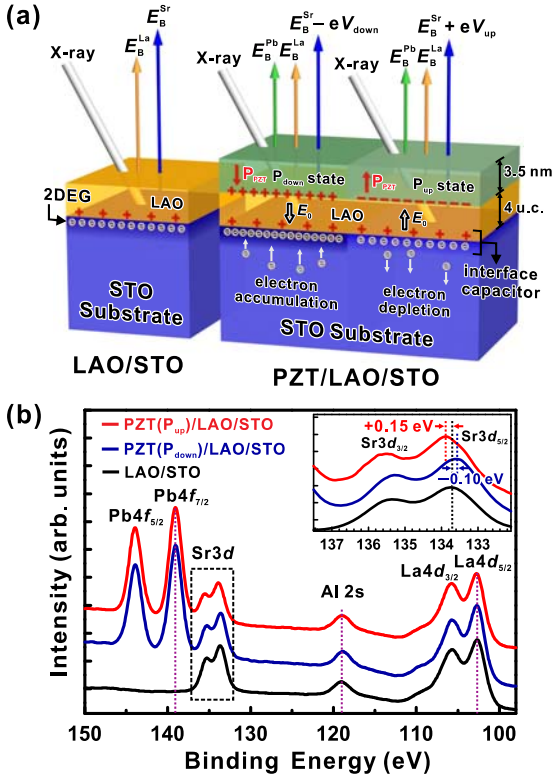

Figure 2 X-ray Photoemission spectroscopy study on the upward / downward ferroelectric polarization.

In conclusion, we have demonstrated that the ferroelectric polarization can tune and modulate the conduction at the LAO/STO hetero-interface. The XPS results reveal the electrostatic predictions of the conducting state modulation, and demonstrate the possibility of nonvolatile control and provide compelling evidence in favor of ferroelectric doping at the conducting polar-nonpolar oxide heterointerface.

Acknowledgments

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References

[1] A. Ohtomo, H. Y. Hwang, A high-mobility electron gas at the LaAlO3/SrTiO3 heterointerface. *Nature* 427, 423–426 (2004).

[2] Nakagawa, N.; Hwang, H. Y.; Muller, D. A. Why some interfaces cannot be sharp. *Nature Mater.* 5, 204–209 (2006).

[3] Reyren, N.; Theil, S.; Caviglia, A. D.; Kourkoutis, L.; Hammerl, G.; Richter, C.; Schneider, C. W.; Koop, T.; Rüetschi, J. -M.; Jaccard, D.; , Gabay, M.; Muller, D.A.; Triscone, J. -M.; Mannhart, J. Superconducting interfaces between insulating oxides. *Science*. 317, 1196–1199 *(*2007*)*.

[4] Brinkman, A.; Huijben, M.; Van Zalk, M.; Huijben, J.; Zeitler, U.; Mann, J. C.; Van Der Wiel, W. G.; Rijders, G.; Blank, D. H. A.; Hilgenkamp, H. Magnetic effects at the interface between non-magnetic oxides. *Nature Mater.* 6, 493–496 (2007).