Origin of Metallic States at the Heterointerface between the Band Insulators $LaAlO_3$ and $SrTiO_3$ Using *in situ* Photoemission Spectroscopy

We have studied the electronic structure at the heterointerface between the band insulators $LaAlO_3$ and $SrTiO_3$ using *in situ* photoemission spectroscopy. Our experimental results clearly reveal the formation of a notched structure on the $SrTiO_3$ side due to band bending at the metallic $LaAlO_3/TiO_2$-$SrTiO_3$ interface. The structure, however, is absent at the insulating $LaAlO_3$/SrO-$SrTiO_3$ interface. The present results indicate that the metallic states originate not from short-range charge transfer through the interface, but from the long-range accumulation of carriers.

Recently, Ohtomo and Hwang have discovered the appearance of metallic conductivity with extremely high mobility at the interface between the band insulators $LaAlO_3$ (LAO) and $SrTiO_3$ (STO), depending on the terminating layer of the interface [1]. Although there have been many intensive theoretical and experimental studies [2-4], the mechanism of this metallic-state formation is still unclear, and a large number of discussions exist on the fundamental origin of the high electrical conductivity and mobility. One of the scenarios proposed for describing the physics of the metallic states is the "charge transfer" that originates from the charge discontinuity at the interface between polar LAO and nonpolar STO [1-3]. According to this scenario, in order to prevent the potential divergence in LAO films, electrons are transferred from the LaO layer into the TiO_2 bonds of the STO layer through the interface. Consequently, the "electron-doped" STO layer at the heterointerface becomes conducting.

Here, we show spectral evidence that the metallic states originate not from charge transfer, but from the accumulation of carriers on a notched structure formed at the STO in the interfacial region. The interfacial band structure, which is determined by in situ photoemission spectroscopy (PES), provides a comprehensive understanding of the appearance of metallic conductivity at the interface, depending on its terminating layer.

LAO/STO multilayers with an LAO/TiO_2-STO or LAO/SrO-STO interface were fabricated on atomically flat TiO_2-terminated Nb:STO (001) substrates by the pulsed laser deposition method [5]. Photoemission spectra were recorded using a Scienta SES-100 electron energy analyzer.

Figure 1 shows Ti 2*p* core level spectra of (a) a TiO_2-terminated STO and a metallic LAO/TiO_2-STO interface and (b) a SrO-terminated STO and an insulating LAO/SrO-STO interface. An absence of reduced Ti^{3+} states is observed in the Ti 2*p* core level, indicating that the electron confinement proposed by the charge transfer scenario does not occur at the interface. The shift of Ti 2*p* core-level peak as a function of LAO overlayer thickness is plotted in Fig. 1(c). Interestingly, the metallic LAO/TiO_2-STO interface clearly shows a peak shift toward higher binding energy as the LAO overlayer thickness increases. In contrast, the insulating interface does not show any detectable shift.

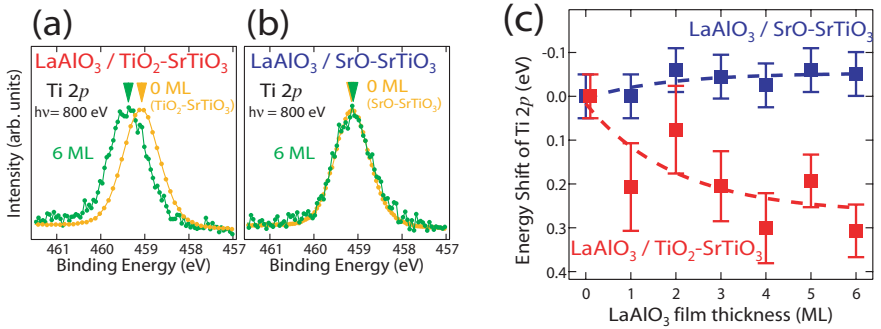

Figure 1
Ti 2*p* core level spectra of (a) a TiO_2-terminated $SrTiO_3$ and a metallic $LaAlO_3/TiO_2$-$SrTiO_3$ interface, and (b) of a SrO-terminated $SrTiO_3$ and an insulating $LaAlO_3$/SrO-$SrTiO_3$ interface. (c) Plots of the energy shift of the Ti 2*p* core-level peaks for metallic (red) and insulating (blue) $LaAlO_3/SrTiO_3$ interfaces as a function of $LaAlO_3$ overlayer thickness.

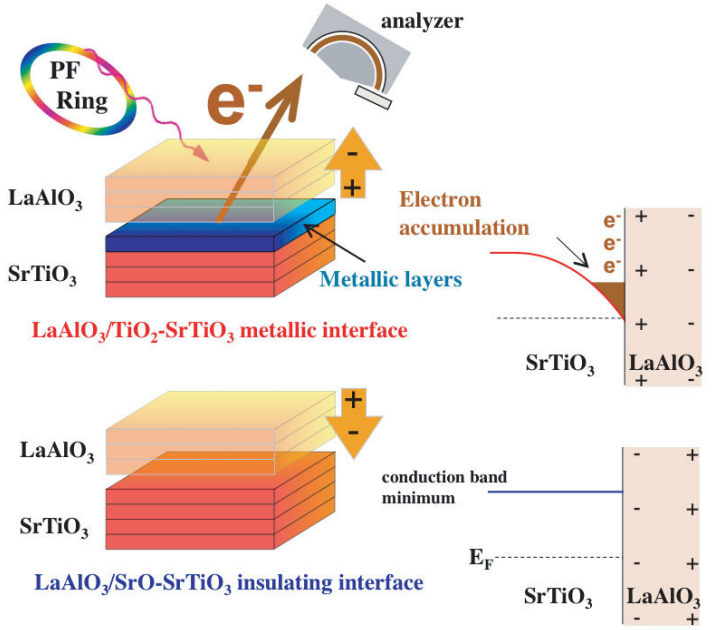

Figure 2
Band diagrams of the metallic and insulating $LaAlO_3/SrTiO_3$ interfaces determined by the present experiments. As a consequence of downward band bending, a notched structure is formed in the $SrTiO_3$ layer only at the metallic interface.

The band diagrams of the metallic and insulating LAO/STO heterointerfaces deduced from the present PES results are illustrated in Fig. 2 [5, 6]. The existence of the notched structure within the STO layers at the metallic interface suggests a mechanism for the formation of the metallic states while still avoiding the potential divergence in the polar LAO layers. Since the polar LAO layers have alternating $\pm e$ charge sheets, where e is the electron charge, the stacking of the LAO layer on nonpolar STO produces a positive electric field. This in turn leads to an electric potential that diverges with increasing LAO overlayer thickness. The divergence catastrophe [2] can be avoided by the formation of a long-range electric potential inside the STO with a spatial variation governed by the carriers in the STO layers, instead of the charge transfer through the interface. In other words, the accumulation of electrons in the notched structure, where the electrons may be generated by oxygen vacancies in the STO layers [4, 5], produces the metallic states at the LAO/STO heterointerface.

REFERENCES

[1] A. Ohtomo and H.Y. Hwang, *Nature* **427** (2004) 423.
[2] N. Nakagawa, H.Y. Hwang and D.A. Muller, *Nature Materials* **5** (2006) 204.
[3] M. Huijben, G. Rijnders, D.H.A. Blank, S. Bals, S.V. Aert, J. Verbeeck, G.V. Tendeloo, A. Brinkman and H. Hilgenkamp, *Nature Materials* **5** (2006) 556.
[4] G. Herranz, M. Basletić, M. Bibes, C. Carrétéro, E. Tafra, E. Jacquet, K. Bouzehouane, C. Deranlot, A. Hamzić, J.-M. Broto, A. Barthélémy and A. Fert, *Phys. Rev. Lett.* **98** (2007) 216803.
[5] K. Yoshimatsu, R. Yasuhara, H. Kumigashira and M. Oshima, *Phys. Rev. Lett.* **101** (2008) 026802.
[6] K. Yoshimatsu, R. Yasuhara, H. Kumigashira and M. Oshima, *Phys. Rev. Lett.* **102** (2009) 199704.

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