Key to the Metallic Conduction Found in an Insulator-Insulator Interface

Two kinds of transition metal oxide ultrathin film, one which exhibits anomalous conductivity and another which is insulating, were examined by the crystal truncation rod scattering technique combined with holographic analysis. Successive least squares structure refinements revealed the difference in atomic intermixing at the interface as well as the local polarization. The conductive interface has a wide polarized region, whereas the insulating interface has a small polarized region. This difference in polarization gives different amounts of band bending, which accounts for the conductive properties.

Physical properties that emerge only at interfaces have long been the focus of great interest. A classic example is the semiconductor diode, and a modern example is metal oxide heterostructures. The interfaces between transition metal oxides appear to be promising for device application because of the wide variety of properties of metal oxides and the availability of highly precise film fabrication techniques.

Interfaces between $LaAlO_3$ (LAO) and $SrTiO_3$ (STO) show anomalous conductivity. Although both LAO and STO are band insulators, the interface between TiO_2-terminated STO and LAO (n-type interface) shows metallic conductivity [1]. Interestingly, the anomalous conductivity disappears when the substrate is changed to SrO-terminated STO (p-type interface). This fact was first reported in 2004 [1], and attracted great interest. Within a few years, the sample growth technique had matured and various groups reported consistent transport properties: for example, the carriers are distributed in STO within ~10 nm from the interface [2]. It is now clear that the difference in structure, n-type and p-type, causes the difference in conductivity. Therefore, the key to conduction is likely to be found in the structure. The detailed structures of both types of interface were analyzed by surface X-ray diffraction experiments performed at BL-3A [3].

Five-unit-cell thick LAO ultrathin films were fabricated by means of pulsed laser deposition (PLD). Four inequivalent crystal truncation rods (CTRs) were measured for each sample with 12-keV X-ray at room temperature in air. Examples of the observed CTRs for the n-type and the p-type samples are shown in Fig. 1 [3]. Other than the substrate Bragg reflections, broad undulations reflecting the thin LAO structures are clearly seen. The total scattering amplitude reflects the Fourier transform of the electron density near the surface.

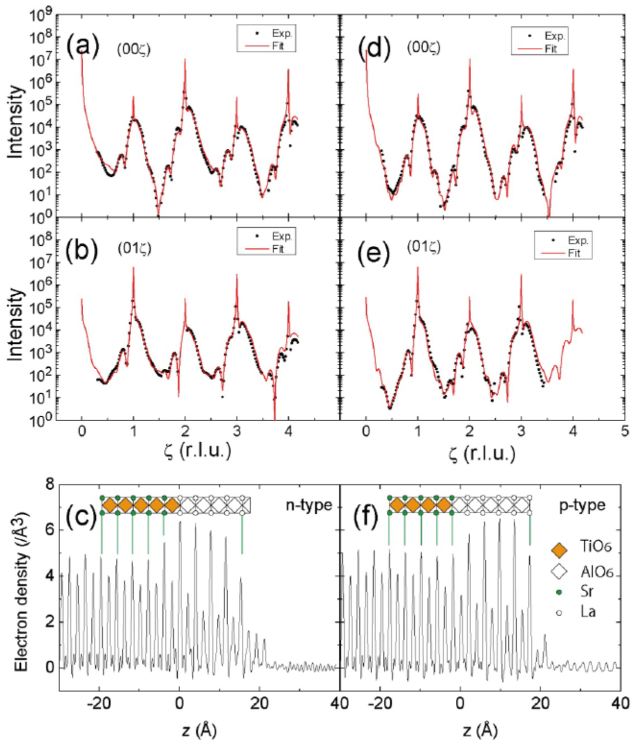

Figure 1
X-ray scattering intensity profiles along (a) the (00 ζ)-line and (b) the (01 ζ)-line for the n-type sample. The depth profile of the electron density, obtained from the electron density analysis performed on the (00 ζ) rod, is shown in (c). Panels (d)-(f) show those for the p-type sample. Figure taken from [3], copyright (2011) by the American Physical Society.

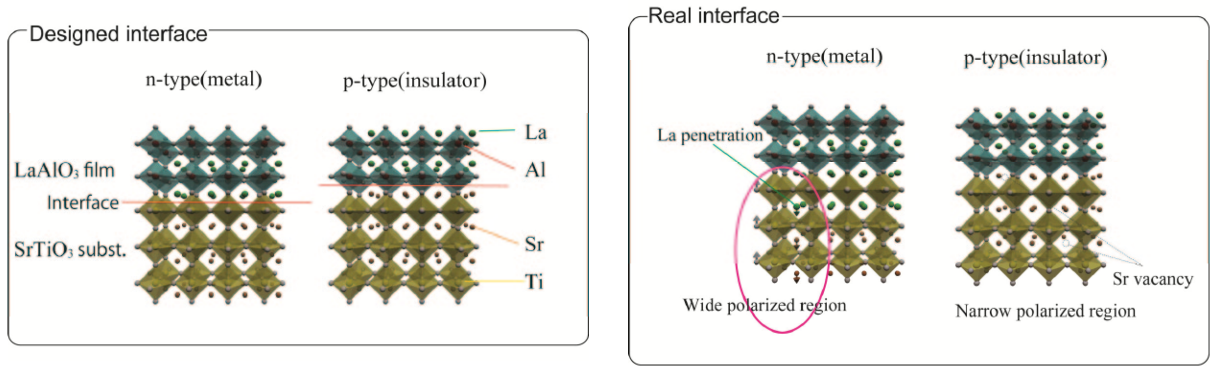

Figure 2
Schematic view of the designed and the observed structures of n-type and p-type interfaces.

The electron density profiles obtained by a holographic analysis applied to 00ζ profiles are shown in Figs. 1 (c) and (f). The structural parameters were refined by a least squares fitting using the peak positions and areas in Figs. 1 (c) and (f) as the starting models. Note that the number of structural parameters of an ultrathin film is large, and the holographic analysis played a significant role to obtain a physically reasonable structure. As a result, we found a structure difference between the two types of interface as shown in Fig. 2. The observed interfacial structure was different from the designed structure in some aspects, particularly in the degree of La inter-diffusion and the polarization. The former corresponds to the formation of the conductive $(La,Sr)TiO_3$ for one monolayer only in the n-type interface [4]. While it appears to be the origin of the anomalous conduction, the formation of one monolayer $(La,Sr)TiO_3$ cannot account for the thickness of the carrier distribution [2]. The latter feature is more important. Significant polarization was found only in STO in the n-type sample. This polarization causes large band bending, which can make the interface conductive. In the insulating p-type interface, by contrast, detectable polarization was suppressed within 1 nm. This screening of the polarization caused by the cation deficiency makes the band bending small, resulting in the difference in conductivity.

REFERENCES

[1] A. Ohtomo and H.Y. Hwang, *Nature,* **427** (2004) 423.
[2] M. Basletic, J.-L. Maurice, C. Carretero, G. Herranz, O. Copie, M. Bibes, E. Jacquet, K. Bouzehouane, S. Fusil and A. Barthelemy, *Nature Materials,* **7** (2008) 621; C. Bell, S. Harashima, Y. Kozuka, M. Kim, B.G. Kim, Y.Hikita and H.Y. Hwang, *Phys. Rev. Lett.*, **103** (2009) 226802.
[3] R. Yamamoto, C. Bell, Y. Hikita, H.Y. Hwang, H. Nakamura, T. Kimura and Y. Wakabayashi, *Phys. Rev. Lett.*, **107** (2011) 036104.
[4] P.R. Willmott, S.A. Pauli, R. Herger, C.M. Schlepütz, D. Martoccia, B.D. Patterson, B. Delley, R. Clarke, D. Kumah, C. Cionca and Y. Yacoby, *Phys. Rev. Lett.*, **99** (2007) 155502.

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