Thickness-dependent electronic structure of LaNiO₃ ultrathin films

Masatomo TAMAMITSU¹, Enju SAKAI¹,2, Kohei YOSHIMATSU¹, Koji HORIBA¹,2,3
Hiroshi KUMIGASHIRA¹,3,4, and Masaharu OSHIMA¹,2,3
¹Department of Applied Chemistry, The Univ. of Tokyo, Tokyo 113-8656, Japan
²CREST, Japan Science and Technology Agency, Tokyo 113-8656, Japan
³Synchrotron Radiation Research Organization, The Univ. of Tokyo, Tokyo 113-8656, Japan
⁴PRESTO, Japan Science and Technology Agency, Saitama 332-0012, Japan

Introduction
Immediately after predicting the similarity in electronic structures of LaNiO₃(LNO)/LaAlO₃(LAO) superlattices to high-Tc cuprate [1], a number of the experimental efforts have been devoted to explore the possible high-Tc superconductivity in the heterostructures based on LNO [2]. The theory predicted the emergence of the two-dimensional electron liquid states in the LNO layer as results of the quantum confinement of Ni 3d electrons and the e₉ orbital ordering. However, almost all experiments reported that the heterostructures underwent the transition from metal to insulator (MIT) at a critical LNO-layer thickness of 3-5 ML [2,3], strongly implying the intrinsic insulating ground states of the LNO layer with a few ML thicknesses. In order to investigate how the electronic structure changes as a function of layer thickness, we have performed in-situ photoemission spectroscopy (PES) on LNO ultrathin films grown onto LAO substrates with varying film thickness.

Experiment
LNO ultrathin films were grown onto the LAO (100) substrates in a laser molecular-beam epitaxy chamber connected to a synchrotron-radiation photoemission system at BL2C. LNO films were deposited at the substrate temperature of 450 °C under an oxygen partial pressure of 10⁻³ Torr. The atomically flat surface and chemically abrupt interface were confirmed by atomic force microscope and cross-section transmission electron microscopy, respectively. The PES spectra were recorded at room temperature using an SES 2002 electron energy analyzer with the total energy resolution of 120 meV at the photon energy of 600 eV.

Results and Discussion
Figure 1 shows the in-situ PES spectra near the Fermi level (E₀) of digitally-controlled LNO ultrathin films. These spectra exhibit remarkable and systematic changes as a function of the LNO film thickness. The spectra consist of structures located just at E₀ and around 1.5 eV, which are assigned to e₉ and t₂g states, respectively. As a result, a pseudogap seems to be formed at E₀ below 4 ML. With a further decrease in the film thickness, the pseudogap finally evolves to an energy gap at Eᵣ, indicating that thickness-dependent MIT occurs at a critical film thickness of 3–5 ML. The observed spectral behavior across the MIT is quite similar to that for bandwidth controlled MIT in RNiO₃. These results suggest that dimensional-driven MIT [4] occurs in LNO ultrathin films as a consequence of the reduction of bandwidth due to a decrease in the effective coordination number of constituent ions at the interface and surface.

References

*tamamitsu@sr.t.u-tokyo.ac.jp

Figure 1: In-situ PES spectra near Eᵣ of LaNiO₃ (n ML)/LaAlO₃.