Dimensional-Crossover-Driven Metal-Insulator Transition in $SrVO_3$ Ultrathin Films Studied by *in situ* Photoemission Spectroscopy

We have investigated the changes occurring in the electronic structure of $SrVO_3$ ultrathin films across the dimensionality-controlled metal-insulator transition (MIT) by *in situ* photoemission spectroscopy. With decreasing film thickness, an energy gap is formed at E_F through spectral weight transfer from the coherent part to the incoherent part, indicating the MIT in the $SrVO_3$ ultrathin film. The observed spectral behavior is reproduced by layer dynamical-mean-field-theory calculations, and it indicates that the observed MIT is a Mott-transition caused by the reduction in the bandwidth due to the dimensional crossover in the presence of electron correlations.

Metal-insulator transition (MIT) is one of the most fundamental phenomena in condensed matter physics [1]. According to the Mott-Hubbard theory [2], MIT can be controlled by varying the relative magnitudes of on-site Coulomb repulsion *U* and bandwidth *W*. Thus, in the case of bulk materials, MIT has been intensively studied by chemical substitution of constituent ions with ones having a smaller ion radius, where *W* is controlled by the resultant changes in the bond angle between transition metal ions and oxygen ions. However, such chemical substitutions always induce randomness in a solid, and consequently this unavoidable chemical disorder has considerably obscured the nature of MIT induced by bandwidth control [3].

Here, we propose an alternative approach to study bandwidth controlled MIT, using dimensional crossover occurring in an artificial structure. Since a decrease in the layer thickness of ultrathin films causes a reduction in the effective coordination number of constituent ions at the interface and surface, the resultant reduction of the effective *W* from a three-dimensional (3D) thick film to the two-dimensional (2D) ultrathin film may drive MIT in the conductive transition metal oxides.

We have applied this new approach to a typical perovskite material with $3d^1$ configuration, $SrVO_3$ (SVO). An SVO ultrathin film is an ideal system for studying the dimensional-crossover-driven MIT, because of the robust metallic states in SVO, irrespective of chemical substitutions. By using a digitally controlled film form, we first observed the MIT induced by the dimensional crossover in SVO ultrathin films by employing *in situ* photoemission spectroscopy [4].

Figure 1 (a) shows the symmetrized V 3*d* spectra of $SrVO_3$ ultrathin films grown onto Nb:STO substrates by digitally controlling the SVO layer thickness. In the case of thicker SVO films, there are two components in V 3d spectra: a peak located at the Fermi level (E_F) and a relatively broad peak centered at about 1.5 eV that corresponds to the coherent (quasiparticle peak) and incoherent (the remnant of the lower Hubbard band) parts, respectively. With decreasing film thickness, the spectral weight is transferred from the coherent to the incoherent part in the layer thickness of 3–6 ML, and finally the V 3*d* spectra for the film thicknesses of 1–2 ML clearly exhibit an energy gap at E_F, which indicates the MIT at a critical film thickness of 2–3 ML. The evolution of the spectral weight at E_F across MIT is more clearly seen in the plot of the quasiparticle intensity as shown in Fig. 1(b).

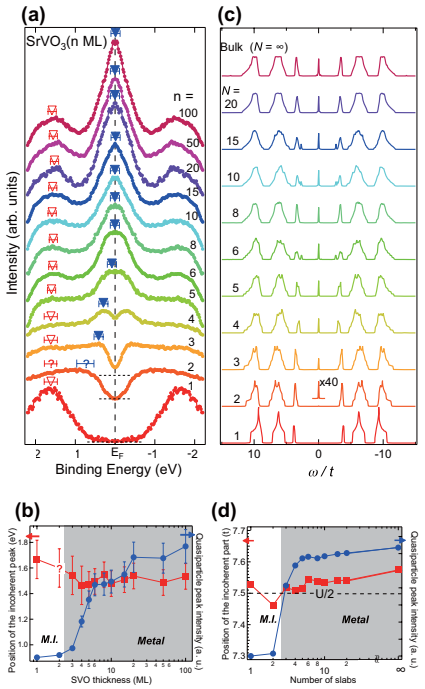

Figure 1
(a) Symmetrized V 3*d* spectra for thickness-dependent $SrVO_3$ ultrathin films. The filled and open triangles represent the energy positions of the coherent and incoherent parts, respectively.
(b) Plots of the quasiparticle peak intensity and the peak positions of the incoherent parts in V 3*d* spectra of SVO ultrathin films as a function of the film thickness.
(c) Spectral functions calculated from layer DMFT.
(d) Plots of the quasiparticle peak intensity and the centroid of the incoherent part in the calculated spectra as a function of the number of slabs. MI denotes the Mott insulator region.

The observed thickness-dependent MIT may be explained in terms of dimensional crossover, as illustrated in Fig. 2. As shown in Fig. 1(b), the peak position of the incoherent part remains unchanged in all film thicknesses, indicating that *U* does not change as a function of the film thickness. This means that the MIT in SVO ultrathin films originates from the reduction of bandwidth *W*. The reduction of *W* with decreasing film thickness is naturally explained by the decrease in the effective coordination number of ions due to the lack of neighboring V ions at the interface and surface: *W* is scaled with *t* as $W \sim 2Nt$ ($W \sim 8t$ in 2D and $12t$ in 3D, respectively) in the Hubbard model, where *N* is the coordination number and *t* characterizes the energy scale of electron hopping between nearest-neighbor sites. The conduction electrons in SVO are confined in the in-plane direction.

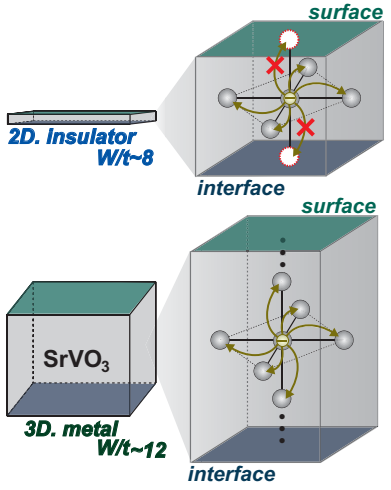

Figure 2
Schematic illustration for explaining the dimensional-crossover-induced metal-insulator transition in $SrVO_3$ thin films.

The resultant reduction of the effective W from 3D to 2D may drive a Mott MIT in SVO ultrathin films.

In order to confirm that the observed thickness-dependent MIT can be understood by dimensional crossover, we compared the symmetrized V 3*d* spectra with the spectral function obtained from layer dynamical-mean-field-theory (DMFT) calculations [5]. The numerical results are shown in Figs. 1(c) and (d). The layer DMFT calculation was performed using the multi-layered single-band Hubbard model, in which slabs are sandwiched between vacuums. For simulating the dimensional crossover derived MIT in the calculation, we selected an appropriate value for *U* so that the system is insulating in 2D but metallic in 3D, and interpolates the two limits by changing the slab number (N) corresponding to the change in the effective bandwidth. The observed thickness-dependent MIT is well reproduced by the layer DMFT calculations. This indicates that the observed MIT is a Mott-transition caused by the reduction in bandwidth due to the dimensional crossover.

REFERENCES

[1] M. Imada, A. Fujimori and Y. Tokura, *Rev. Mod. Phys.* **70** (1998) 1039.
[2] J. Hubbard, *Proc. R. Soc. A* **276** (1963) 238 .
[3] D.D. Sarma, O. Rader, T. Kachel, A. Chainani, M. Mathew, K. Holldack, W. Gudat and W. Eberhardt, P*hys. Rev. B* **49** (1994) 14238.
[4] K. Yoshimatsu, T. Okabe, H. Kumigashira, S. Okamoto, S. Aizaki, A. Fujimori and M. Oshima, *Phys. Rev. Lett.* **104** (2010) 147601.
[5] S. Okamoto and A.J. Millis, *Phys. Rev. B* **70** (2004) 241104(R).

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