Quantum Critical Behavior in Metallic Quantum Well of Strongly Correlated Oxides

Control of quantum critical behavior emerging near the quantum phase transition in strongly correlated electron systems is a major challenge in recent condensed matter physics. The origin of quantum phase transition has been studied by changing limited external fields such as magnetic field and pressure. Here, we have developed a new way to investigate quantum critical phenomena by varying the strength of quantum fluctuation due to dimensional crossover in the metallic quantum-well structure of strongly correlated oxides. The result indicates that an artificially controllable quantum-well structure provides an ideal platform to study quantum phenomena in strongly correlated oxides.

Reduction of dimensionality affects complex interactions among charge, spin, and orbital degrees of freedom of electrons in materials, leading to changes of the electronic properties [1, 2]. Generally, the quantum fluctuation of order parameter is too weak in three-dimensional (3D) systems and too strong in one-dimensional systems. In two-dimensional (2D) systems, intermediate between 3D and 1D, the delicate balance between long-range ordering and quantum fluctuation results in quantum critical phenomena [3]. Control of dimensionality from 3D to 2D in single crystals has been investigated in layered oxides such as the Ruddlesden-Popper series $A_{t+1}B_tO_{3t+1}$ (*A*: alkaline-earth elements, *B*: transition-metal elements) where ABO_3 conductive layers are sandwiched by insulating block layers *AO* [4]. These layered oxides involve both low dimensionality and strong correlation and show extraordinary physical properties derived from the quantum fluctuation such as high-T_c superconductivity [5]. However, systematic growth of layered oxides having different numbers of conductive layers is quite difficult, and it is a challenge to control the dimensionality from 3D to 2D under fixed electronic structure parameters. To address this issue, we propose a new way to investigate quantum critical phenomena accompanied with the dimensional crossover from 3D to 2D using the quantum well of strongly correlated oxides. Strongly correlated oxide $SrVO_3$ (SVO) thin films with the thickness of several monolayers (MLs) show quantum well (QW) states, which have been confirmed by *in situ* angle-resolved photoemission spectroscopy (ARPES) [6]. With decreasing film thickness, metal-insulator transition (MIT) on the SVO-QW states occurs when the thickness falls below 3 ML [7]. Therefore, the SVO-QW structure is an ideal platform to investigate quantum critical phenomena originating from the increase of quantum fluctuation due to dimensional crossover.

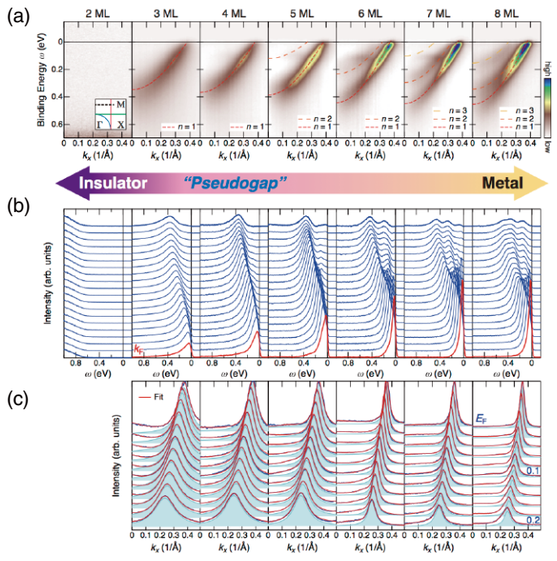

Figure 1: A series of ARPES spectra of $SrVO_3$ quantum-well structures with various layer thicknesses. **(a)** Respective ARPES images of ultrathin SVO films with t = 2–8 ML. The ARPES data were acquired with the present photon energy of $h\nu$ = 88 eV along a k_x slice near the X point (k_y = 0.75 π/a) as shown by the dashed line in the inset, which includes only the quantized d_{xy} band. The dashed lines show the results of the tight-binding fitting for each QW state. The inset shows the in-plane Fermi surface and the ARPES measured cut. **(b)** EDCs corresponding to the respective ARPES images. **(c)** Line-shape analysis for the MDCs at various ω. The shaded areas indicate the Lorentzian functions for the n = 1 states.

Figure 1 shows a series of ARPES images observed in the SVO ultrathin films with thicknesses from 2 to 8 MLs. These spectra have been obtained along a cut crossing only the d_{xy} band (see the inset), and so the ARPES images consist of only the quantized d_{xy} band. The number of bands depends on the film thickness, and subband structures corresponding to quantum number n = 1, 2, and 3 have been observed. In the energy distribution curves (EDCs) shown in **Fig. 1(b)**, the peak intensity near the Fermi level (E_F), that is, quasiparticle weight, gradually decreases with decreasing thickness below 6 ML and finally disappears at 2 ML, indicating that MIT depends on thickness and the critical thickness t_c is 3 ML. Since the quasiparticle weight is proportional to renormalization factor Z reflecting the strength of electron correlation, the MIT in SVO QW structures originates from the electron–electron interaction.

Figure 1(c) shows the momentum distribution curves (MDCs). The width of the MCD peak increases with decreasing thickness of the QW structure. The MDC width Δk is proportional to the mean free path of conduction electron l and imaginary part of the self-energy ImΣ:

$$\hbar v_k \Delta k = \hbar v_k / l \approx |2\mathrm{Im}\Sigma(\boldsymbol{k}, \omega)|, \qquad (1)$$

where $\hbar$ is the reduced Planck constant and v_k is the group velocity. **Figure 2(a)** shows ImΣ converted from the estimated Δk using Eq. (1) plotted as a function of binding energy ω. The observed ω^2 dependence of ImΣ of the 6 ML film suggests that the Fermi liquid (FL) ground state of single crystalline (or bulk) SVO [8, 9] is maintained in QW structures with thickness above 6 ML as previously reported [10]. The gradient of the ImΣ curve gradually changes from parabolic to linear as the thickness approaches t_c, and ImΣ becomes linear at the critical thickness of 3 ML. To elucidate this change quantitatively, the ImΣ curves are fitted to the following phenomenological form:

$$|Z\mathrm{Im}\Sigma(\omega)| = \Gamma^{\mathrm{imp}} + \beta'(\omega^2 + (\pi k_B T)^2)^{\alpha/2}, \qquad (2)$$

where β' is a coefficient reflecting the strength of electron correlation, k_B is the Boltzmann constant, Γ^{imp} is the inverse lifetime of the quasiparticle, and α is the critical index. As shown in **Fig. 2(a)**, the fitting using Eq. (2) well reproduces the experimental ImΣ curves. The value of α is 2 (parabolic) above 6 ML and gradually decreases with decreasing thickness in the 4–6 ML region and becomes 1 (linear) at 3 ML, as shown in **Fig. 2(b)**. These results suggest that crossover of the ground state of the QW from FL to non-Fermi liquid (NFL) occurs in the vicinity of MIT.

Since only one subband exists in the occupied states in the 3 ML QW structure as shown in the structure plot of **Fig. 2(b)**, the QW state of the 3 ML film is considered to be two-dimensional. Therefore, the emergence of NFL states near MIT is considered to be a consequence of the enhancement of quantum fluctuation near the 2D limit. This result strongly suggests the existence of a quantum critical point (QCP) in close proximity to the thickness-dependent MIT. Based on these findings, we conclude that the artificially controllable QW structures of strongly correlated oxides with adjustable physical dimensions will provide a new strategy for investigating quantum critical phenomena emerging near a QCP.

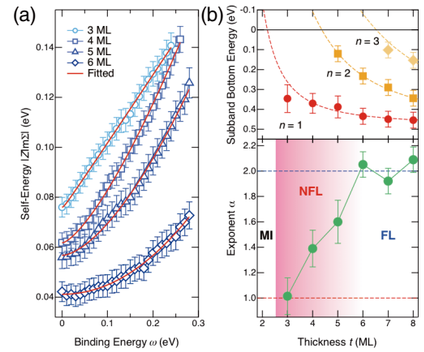

Figure 2: Self-energy Σ for $SrVO_3$ quantum-well states. **(a)** Imaginary part of the self-energy ImΣ for the n = 1 state as a function of ω for 3–6 ML. The solid curves represent the fitted curves based on Eq. (2). **(b)** (Top) Structure plot of the QW states as a function of t. The dashed lines represent the simulation results based on the phase shift quantization rule [9]. (Bottom) Plot of evaluated exponent α with respect to t. Here, FL, NFL, and MI denote Fermi liquid, non-Fermi liquid, and Mott-insulating states, respectively.

REFERENCES

[1] P. D. C. King, H. I. Wei, Y. F. Nie, M. Uchida, C. Adamo, S. Zhu, X. He, I. Božovic, D. G. Schlom and K. M. Shen, *Nat. Nanotech.* **9**, 443 (2014).
[2] H. Y. Hwang, Y. Iwasa, M. Kawasaki, B. Keimer, N. Nagaosa and Y. Tokura, *Nat. Mater.* **11**, 103 (2012).
[3] S. Sachdev, *Science* **288**, 475 (2000).
[4] A. Nozaki, H. Yoshikawa, T. Wada, H. Yamauchi, and S. Tanaka, *Phys. Rev. B* **43**, 181 (1991).
[5] M. Imada, A. Fujimori and Y. Tokura, *Rev. Mod. Phys.* **70**, 1039 (1998).
[6] K. Yoshimatsu, K. Horiba, H. Kumigashira, T. Yoshida, A. Fujimori, M. Oshima, *Science* **333**, 319 (2011).
[7] K. Yoshimatsu, T. Okabe, H. Kumigashira, S. Okamoto, *S. Aizaki*, A. Fujimori and M. Oshima, *Phys. Rev. Lett.* **104**, 147601 (2010).
[8] S. Aizaki, T. Yoshida, K. Yoshimatsu, M. Takizawa, M. Minohara, S. Ideta, A. Fujimori, K. Gupta, P. Mahadevan, K. Horiba, H. Kumigashira and M. Oshima, *Phys. Rev. Lett.* **109**, 056401 (2012).
[9] I. H. Inoue, O. Goto, H. Makino, N. E. Hussey and M. Ishikawa, *Phys. Rev. B* **58**, 4372 (1998).
[10] M. Kobayashi, K. Yoshimatsu, E. Sakai, M. Kitamura, K. Horiba, A. Fujimori and H. Kumigashira, *Phys. Rev. Lett.* **115**, 076801 (2015).

BEAMLINES

BL-2A and BL-28A

M. Kobayashi[1], K. Yoshimatsu[1,2], T. Mitsuhashi[1,3], M. Kitamura[1], E. Sakai[1], R. Yukawa[1], M. Minohara[1], A. Fujimori[2], K. Horiba[1] and H. Kumigashira[1,3] ([1]KEK-IMSS-PF, [2]The Univ. of Tokyo, [3]Tohoku Univ.)