3 Oxide Heterostructure Project

– Observation and control of novel quantum phenomena in superstructures of strongly-correlated oxides –

Project Leader: Hiroshi Kumigashira

3-1 Introduction

The goal of this project is to design novel physical properties appearing at the heterointerface of strongly correlated oxides. The physical properties arise from strong mutual coupling among spin, charge, and orbital degrees of freedom at the interface between two different oxides [1]. In order to control such properties, it is necessary to clarify the interfacial electronic, magnetic, and orbital structures. Therefore, we are using synchrotron radiation spectroscopic techniques with elemental selectivity to probe these structures in the nanometer-scale region at the oxide heterointerface. For example, the electronic structure at the interface is determined by photoemission spectroscopy (PES) and X-ray absorption spectroscopy (XAS), the magnetic structure by magnetic circular dichroism of XAS, and the orbital structure by linear dichroism of XAS. Furthermore, the electronic band structures near the Fermi level (E_F), which dominate the novel physical properties of oxide nanostructures, are investigated with angle-resolved PES (ARPES). We aim to design and create novel quantum materials by optimally combining sophisticated oxide growth techniques using laser molecular beam epitaxy (MBE) and advanced analysis techniques using quantum beams.

3-2 Construction of a new beamline for surface and interface studies of oxide superstructures

Our developed system ''*in-situ* photoelectron spectrometer – laser MBE'' had been installed at the new undulator beamline MUSASHI (Multiple

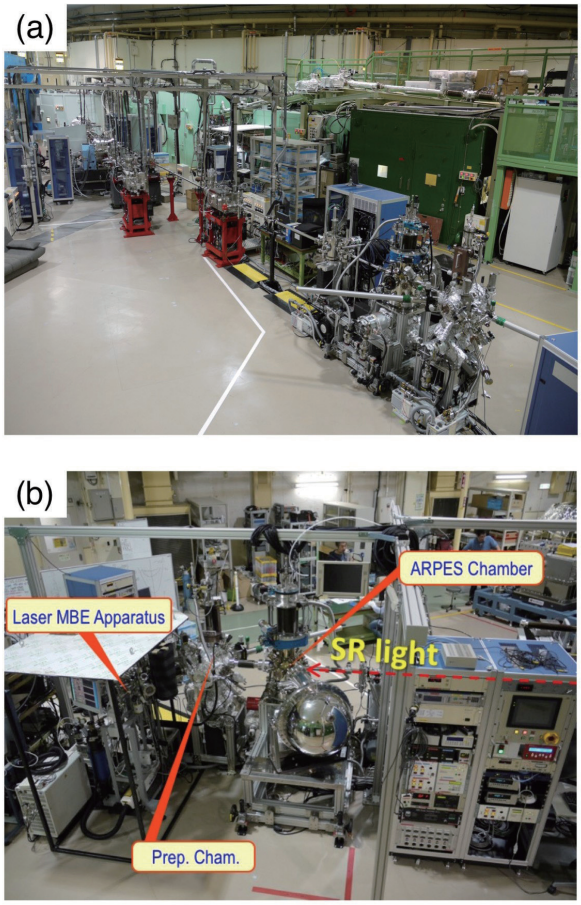

Fig. 1: Photograph of (a) BL-2A MUSASHI beamline and (b) "*in-situ* ARPES – Laser MBE system" which is installed as an end station of BL-2A MUSASHI.

Undulator beamline for Spectroscopic Analysis on Surface and HeteroInterface) as an endstation (Fig. 1). At the new BL-2A MUSASHI, we perform both vacuum ultraviolet (VUV: 30–300 eV) and soft X-ray (SX: 250–2000 eV) spectroscopic measurements at the same time, for the same grown sample, with the same experimental setup. This approach guarantees the quality of the experi-

mental data and offers numerous benefits to the field of surface science. The newly-developed system enables us to obtain complete information on the electronic and chemical structures of oxide nanostructures fabricated by laser MBE with high reproducibility. For example, the chemical states and band lineup are characterized with XAS and PES using SX light, confirming the quality of the heterostructures. Their band structures are then investigated with ARPES using VUV light.

3-3 Science topic: Emergence of Quantum Critical Behavior in Metallic Quantum-Well States of Strongly Correlated Oxides

Controlling quantum critical phenomena in strongly correlated electron systems, which emerge in the neighborhood of a quantum phase transition, is a major challenge in modern condensed matter physics. Quantum critical phenomena are generated from the delicate balance between long-range order and its quantum fluctuations [2–4]. So far, the nature of quantum phase transitions has been investigated by changing a limited number of external parameters, such as pressure and magnetic field. Here, we propose a new approach for investigating quantum criticality by changing the strength of quantum fluctuations that are controlled by the crossover from three dimensions (3D) to two dimensions (2D) occurring in the metallic quantum well (QW) structure of strongly correlated oxides [5–7].

The dimensional crossover from 3D to 2D offers a unique opportunity to study quantum critical phenomena [2]. In 3D systems, low-energy electronic states behave as quasiparticles (QPs) owing to the weakness of quantum fluctuations in the order parameter. Consequently, the physical properties are described well within the framework of Fermi liquid (FL) theory or by long-range ordered states with order parameters. In contrast, quantum fluctuations are so strong in one-dimensional systems that even infinitely weak interactions break the QPs into collective excitations, thus preventing long-range order. In a 2D system, the delicate balance between long-range order and quantum fluctuations results in interesting quantum critical phenomena [2–4]. Therefore, dimensional crossover from 3D to 2D is an ideal platform for systematically studying the quantum critical phenomena that emerge in the neighborhood of the quantum phase transition by utilizing the enhancement of the quantum fluctuation driven by dimensional crossover.

In QW structures based on perovskite oxides, which have close structural similarities to the layered oxides [8], we digitally control the number of conductive layers of strongly correlated oxides [5]. Consequently, the competition between long-range order and quantum fluctuations can be precisely investigated as a function of dimensionality (layer thickness t).We have chosen ultrathin films of the conductive oxide $SrVO_3$ (SVO) epitaxially grown on Nb-doped $SrTiO_3$ substrates as the QW structures as a candidate oxide QW structure for this study [5–7].

Figure 2(a) shows a series of ARPES images for the ultrathin SVO films with t = 2–8 ML. Because these band dispersions have been taken along the cut indicated by dashed line in the inset, the ARPES images consist of only quantized d_{zx} bands of V 3d t_{2g} states with quantum numbers n = 1, 2, and 3 from the bottom [5–7]. The occurrence of the thickness-dependent metal-insulator transition (MIT) at the critical layer thickness t_c of 2–3 ML is clearly observed as evidence of the disappearance of a QW subband(s) at 2 ML.

The anomalous spectral changes with approaching the Mott transition are further confirmed by the ARPES spectra near E_F. We show the energy distribution curves (EDCs) and momentum distribution curves (MDCs) corresponding to the ARPES images in Figs. 2(b) and 2(c), respectively [9]. As expected from the ARPES images in Fig. 2(a), sharp QP peaks exist in the vicinity of E_F, and the intensity remains nearly constant down to 6 ML. With a further decrease in t, the QP peaks gradually reduce their spectral intensity in the range of t = 3–5 ML, and finally fades into an energy gap of 0.5–0.7 eV at 2 ML. This value agrees well with that in previous PES results [10]. The dramatic reduction of the QP weight at E_F with approaching the MIT suggests the strongly correlated nature of the QW states. Furthermore, the MDC width Δk gradually increases, as can be seen in Fig. 2(c). These spectral behaviors are associated with the reduction in the QP intensity and imply the emergence of intriguing ground states near the Mott transition.

The strange behavior of QP excitation is reminiscent of that in the vicinity of the quantum phase transition. In order to illuminate the under-

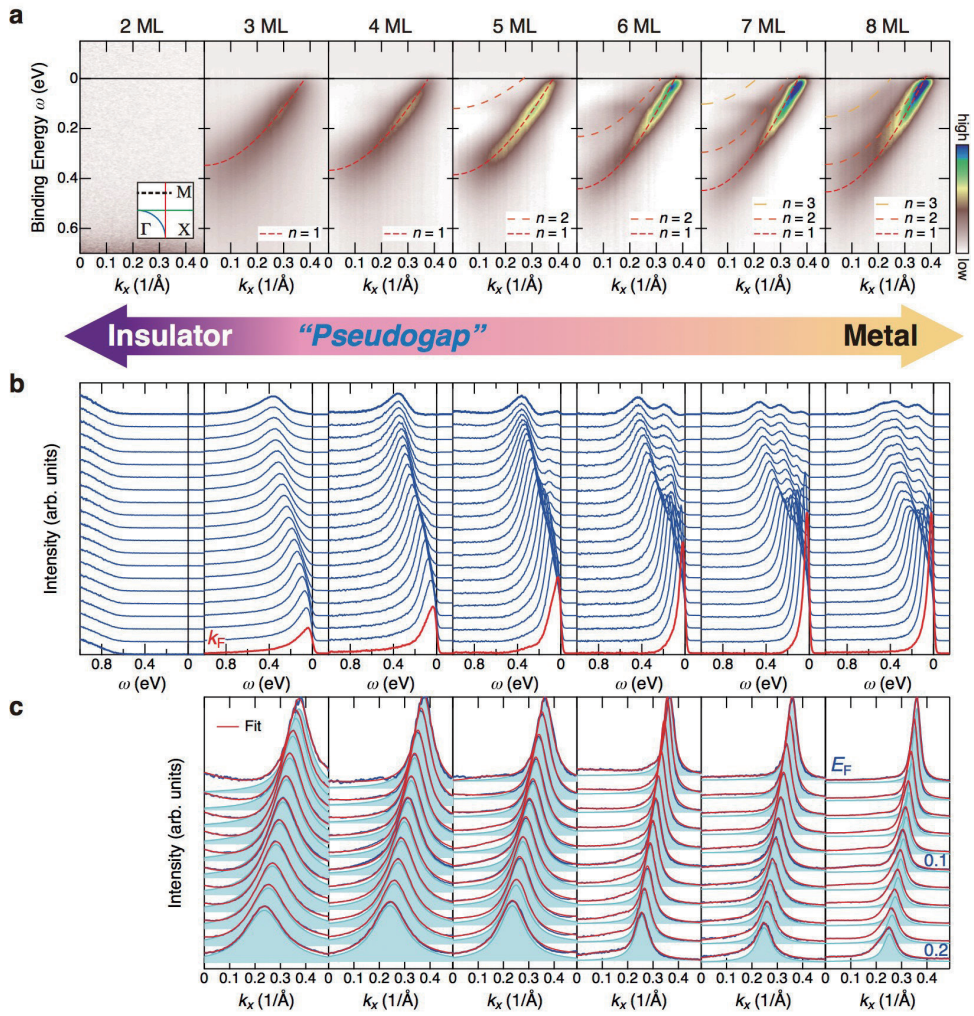

Fig. 2: Series of ARPES spectra for the $SrVO_3$ quantum-well structures with various layer thicknesses. (a) Respective ARPES images of the ultrathin SVO films with t = 2–8 ML. The ARPES data were acquired at a photon energy of $h\nu$ = 88 eV along a k_x slice near the X point (k_y = 0.75π/a), as shown by the dashed line in the inset. The dashed lines show tight-binding fitting results 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 values. The shaded areas indicate Lorentzian functions for the n = 1 states.

lying physics, we have evaluated the self-energy by applying line-shape analysis to the MDCs as a function of the binding energy ω [9]. The obtained imaginary part of the self-energy Im$\Sigma(\omega)$ for the n = 1 state of each QW structure is summarized in Fig. 3a. As can be seen in Fig. 3a, Im$\Sigma(\omega)$ for 6 ML shows parabolic behavior, reflecting the FL ground states of the SVO-QW structure. This has been reported in previous studies [6]. These ω^2 dependences of Im$\Sigma(\omega)$ indicate that the correlated FL ground states in bulk SVO [11] hold for t ≥ 6 ML. However, when film thickness further approaches t_c, the gradient of Im$\Sigma(\omega)$ systematically changes from parabolic to linear. Eventually, the Im$\Sigma(\omega)$ curve becomes linear at a thickness of 3 ML, which is the two-dimensional limit of the metallic SVO-QW structures. The linear dependence of Im$\Sigma(\omega)$ at 3 ML is reminiscent of the marginal FL states in high-T_c cuprates [9].

These results indicate the occurrence of the a crossover from FL to non-Fermi liquid (NFL) ground states in the vicinity of the MIT. To quantitatively address the crossover of the ground state, the Im$\Sigma(\omega)$ curves have been fitted to the following phenomenological form:

$$|Z\mathrm{Im}\Sigma(\omega)| = \Gamma^{\mathrm{imp}} + \beta'(\omega^2 + (\pi k_B T)^2)^{\alpha/2}, \qquad (1)$$

where Z is the renormalization factor, β' denotes a coefficient reflecting the strength of the electron correlation, k_B is the Boltzmann constant, Γ^{imp} is the inverse lifetime of the QP associated with impurity scattering, and α is the critical exponent. The fitting to Eq. (1) reproduces the ex-

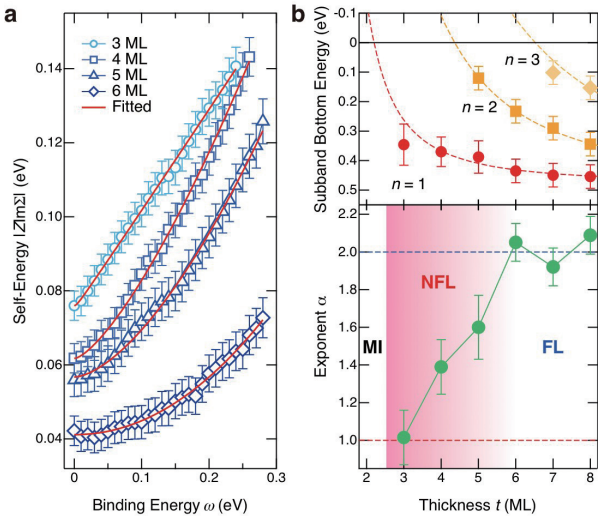

Fig. 3: Self-energy Σ for $SrVO_3$ quantum-well states. (a) Imaginary part of the self-energy ImΣ for the n = 1 state as a function of ω for 3–6 ML. The solid curves represent the fitted curves based on Eq. (1). (b) (Top) Structure plot of the QW states as a function of t. With decreasing t, the number of subbands decreases, and eventually only a single QW subband appears in the two-dimensional limit of metallic SVO-QW structures (3 ML). (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.

perimental ImΣ(ω) curves well, as shown by the solid lines in Fig. 3(a). The estimated values of α are plotted against t in Fig. 3(b) together with the structure plot of the SVO-QW states [5,6]. The crossover from FL to NFL clearly occurs as t is reduced below 6 ML. As t approaches t_c, the value of α gradually reduces from 2 to 1 in the "pseudogap" (dimensional crossover) region of 4–6 ML [10] and then reaches 1 at 3 ML, indicating the existence of a quantum critical point (QCP) around t_c.

From the structure plot of the SVO-QW states in Fig. 3(b), it is clear that the QCP emerges at the two-dimensional limit of the metallic QW structures (3 ML). Because only a single QW subband exists in the occupied states for 3–4 ML, there is no inter-subband interaction between the occupied states. Furthermore, interference between the occupied n = 1 state and an unoccupied n = 2 state is expected to become negligibly weak at 3 ML owing to the largest energy separation between the two quantum states. Thus, in the two-dimensional limit of metallic SVO-QW structures, the QW states verge on the ideal two-dimensional states, suggesting that QCP exists on the borderline between the metallic and Mott insulating phases as a result of the enhancement of quantum fluctuations in 2D. The close relationship between the emergence of the QCP and the energy diagram of the QW states suggests that the strength of the quantum fluctuation can be precisely controlled by tuning the QW structures.

The present experiment strongly suggests the existence of QCP in the close proximity of the thickness-dependent Mott transition. Although the order parameter inducing the QCP is not clear at the moment, our observations have important implications in the search for novel quantum critical phenomena using metallic QW structures. The extraordinary physical properties of strongly correlated systems are usually found around a QCP, such as the ubiquitous formation of a superconducting dome surrounding a QCP in the phase diagram of unconventional superconductors [12,13]. The artificially controllable QW structure of strongly correlated oxides with adjustable physical dimensions will provide a new strategy for designing the quantum critical phenomena emerging around a QCP. It has not escaped our notice that the quantum criticality observed in the two-dimensional limit of the metallic SVO-QW structure immediately suggests the possibility of superconductivity with optimal electron doping, since these SVO-QW structures are the mirror of hole-doped high-T_c cuprates [14].

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