Close Relationship between the Two Characteristic Features of Metallic Quantum Well States in $SrVO_3$ Ultrathin Films

Metallic quantum well (QW) structures based on transition metal oxides provide a platform for studying the behavior of strongly correlated electrons under quantum confinement. Metallic QW states observed in $SrVO_3$ ultrathin films show two distinctive features that have not been observed in conventional metallic QW, i.e., orbital-selective quantization and anomalous subband-dependent mass enhancement. Here, we report the existence of a close relationship between the two characteristic features in terms of *in-situ* angle resolved photoemission spectroscopy (ARPES) measurements and detailed line-shape analysis of ARPES spectra.

Quantum confinement of strongly correlated electrons in oxide heterostructures has attracted a lot of interest for not only its promising technological applications in future oxide electronics [1], but also understanding the fundamental low-dimensional physics of strongly correlated electron systems [2]. Recently, metallic quantum well (QW) states have been discovered in $SrVO_3$ (SVO) ultrathin films grown on Nb:$SrTiO_3$(001) substrates [3]. The observed QW states exhibit two distinctive features. The first is *orbital-selective quantization* originating from the anisotropic orbital character of the V 3*d* t_{2g} (d_{xy}, d_{yz} and d_{zx}) band states, as shown in **Fig. 1a**. Each orbital essentially has a two-dimensional (2D) character in the respective plane. Consequently, the d_{yz}/d_{zx} bands are subject to quantization [from 2D to one-dimensional (1D)], while the d_{xy} band remains unchanged (maintaining its 2D character). The other characteristic behavior is *anomalous subband-dependent mass enhancement*: the subband dispersion becomes considerably narrower as its bottom energy (quantization energy) E_n approaches the Fermi level (E_F) (see **Fig. 1b**). Such anomalous mass enhancement (narrowing) in subbands has not been observed in the conventional metallic QW structure based on metals having nearly free-electron-like *sp* states [4]. Therefore, the anomalous mass enhancement, as well as the orbital selective quantization, is a characteristic behavior of confined strongly correlated electrons. Despite intense theoretical and experimental studies, however, the physical origin of this behavior is not yet understood. To address this issue, we performed *in-situ* angle-resolved photoemission spectroscopy (ARPES) measurements on SVO ultrathin films and analyzed the line shape of ARPES spectra, particularly focusing on the width of momentum distribution curves (MDCs) [5].

Figure 1b shows the dispersion of each subband $\varepsilon_n(\boldsymbol{k})$ determined from the ARPES measurements, together with the MDC at E_F. It is evident that the MDC width Δk becomes wider with increasing quantum number *n* for each film thickness and as E_n approaches E_F. From a Δk value, one can estimate the imaginary part of self-energy ImΣ reflecting electron–electron correlation [6]. The obtained ImΣ values for each subband are plotted in **Figs. 2a-c** as a function of binding energy ω. As can be seen in **Figs. 2a-c**, the rate of increase of Im$\Sigma(\omega)$ tends to be larger with increasing *n* and/or decreasing thickness, namely as E_n approaches E_F.

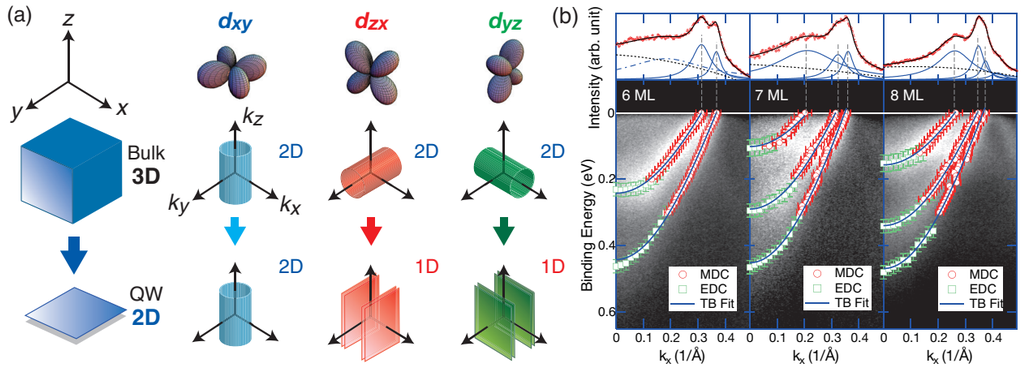

Figure 1: Quantum well states in $SrVO_3$ ultrathin films. **(a)** Schematic illustration of the QW structure and quantization of V-3*d*-derived states. Owing to the 2D character of each orbital in the respective plane, the d_{yz}/d_{zx}-derived electronic structures are subject to quantization (from 2D to 1D), while that of the d_{xy} orbital remains unchanged (from 2D to 2D). **(b)** ARPES intensity plots for d_{zx} subbands of the 6-ML, 7-ML, and 8-ML $SrVO_3$. The top panels show the corresponding MDCs at E_F.

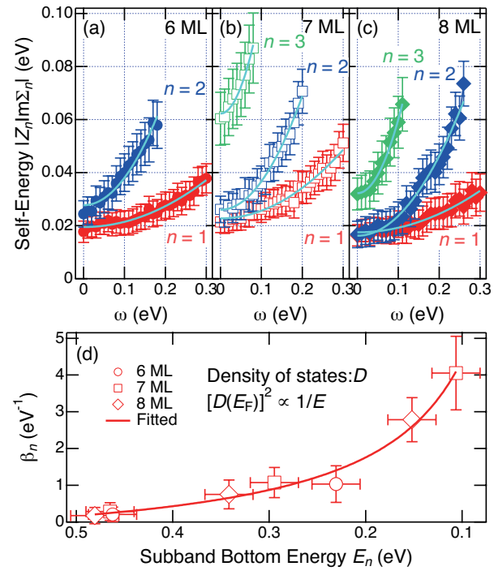

Figure 2: Self-energy for $SrVO_3$ QW states. **(a)-(c)** ImΣ_n of each subband plotted as a function of ω for 6-, 7-, and 8-ML $SrVO_3$ ultrathin films. **(d)** E_n dependence of β_n estimated from the curve fitting. The solid curve is the result of fitting assuming the quasi-1D DOS.

In order to evaluate the strength of electron correlation, Im$\Sigma(\omega)$ curves are fitted to the following equation, which assumes the Fermi liquid ground state [5]:

$$\left|Z_n \operatorname{Im}\Sigma(\omega)\right| = \Gamma_n^{\mathrm{imp}} + \beta_n\left(\omega^2 + (\pi k_B T)^2\right) \qquad (1)$$

where β_n denotes a coefficient reflecting the strength of electron correlation, k_B is the Boltzmann constant, and Γ_n^{imp} is the inverse lifetime of quasiparticles associated with impurity scattering. The curve fitting closely reproduces the experimental results, as shown in **Figs. 2a-c**. The estimated β_n is plotted against E_n in **Fig. 2d**. As E_n approaches E_F, the value of β_n is considerably enhanced, indicating that the electron correlation significantly contributes to the anomalous subband-dependent mass enhancement in SVO QW states.

The trend of the observed enhancement of electron correlation seems to be opposite to what would be expected from the Fermi-liquid behavior. In general, the strength of the electron–electron interaction is proportional to the square of the density of states (DOS) *D* at E_F. Therefore, based on the behavior of bulk SVO [7], a weaker correlation effect is expected in the subbands located at lower binding energies owing to the reduction of band filling [8]. The seeming contradiction can be reconciled by considering the orbital-selective quantization as shown in **Fig. 1a**. In the present case, the quasi-1D electronic states are realized in the d_{yz}/d_{zx}-derived subbands by the further reduction of dimensionality (from 2D to 1D) due to the quantum confinement. DOS in 1D systems is proportional to $1/\sqrt{E}$, where *E* is the energy measured from the band edge. Thus, the β_n within the quasi-1D d_{yz}/d_{zx} subbands will be proportional to $[D(E_F)]^2 \propto 1/E_n$, which is the opposite behavior of the three-dimensional case ($[D(E_F)]^2 \propto E_n$). Indeed, the experimental results are adequately reproduced by the curve fitting assuming the quasi-1D state as shown in **Fig. 2d**.

These results indicate the close relationship between the two characteristic behaviors of metallic QW states in $SrVO_3$ ultrathin films: the *anomalous subband-dependent mass enhancement* originates from the quasi-1D nature of the quantized V 3*d* states as a result of the *orbital-selective quantization*. The present study demonstrates that the QW structure of strongly correlated oxide provides an ideal platform both for investigating the behavior of correlated electrons and for manipulating novel quantum phenomena in reduced dimensions.

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BEAMLINES

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