



Metallic Quantum Well States in Artificial Structures of Strongly Correlated Oxide

K. Yoshimatsu, et al. Science **333**, 319 (2011); DOI: 10.1126/science.1205771

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superficially similar to convective models in some respects (28) but nevertheless fail to explain the origin and energy flux of this structure (4–8, 28) as well as the present observations.

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Acknowledgments: P. Sütterlin provided valuable support during the observations and data analysis, and B. Edvardsson provided advice concerning line blends. J.d.I.C.R. gratefully acknowledges financial support by the European Commission through the SOLAIRE Network (MTRN-CT-2006-035484). CRISP was funded by the Marianne and Marcus Wallenberg Foundation. The Swedish 1-meter Solar Telescope is operated on the island of La Palma by the Institute for Solar Physics of the Royal Swedish Academy of Sciences in the Spanish Observatorio del Roque de los Muchachos of the Instituto de Astrofísica de Canarias.

Supporting Online Material

www.sciencemag.org/cgi/content/full/science.1206429/DC1 Materials and Methods Figs. S1 to S21 Tables S1 to S9 References (31–39)

4 February 2011; accepted 25 May 2011 Published online 2 June 2011; 10.1126/science.1206429

Metallic Quantum Well States in Artificial Structures of Strongly Correlated Oxide

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The quantum confinement of strongly correlated electrons in artificial structures provides a platform for studying the behavior of correlated Fermi-liquid states in reduced dimensions. We report the creation and control of two-dimensional electron-liquid states in ultrathin films of SrVO₃ grown on Nb:SrTiO₃ substrates, which are artificial oxide structures that can be varied in thickness by single monolayers. Angle-resolved photoemission from the SrVO₃/Nb:SrTiO₃ samples shows metallic quantum well states that are adequately described by the well-known phase-shift quantization rule. The observed quantum well states in SrVO₃ ultrathin films exhibit distinctive features—such as orbital-selective quantization originating from the anisotropic orbital character of the V 3d states and unusual band renormalization of the subbands near the Fermi level—that reflect complex interactions in the quantum well.

n electron confined in space by a potential well forms standing waves in the well, or quantum well (QW) states. These states are characterized by the quantum number n, that

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is, the number of half-wavelengths that span the well. QW states are associated with discrete quantized electronic states in thin artificial structures with tunable physical dimensions, in which electron confinement occurs only in the direction perpendicular to the surface, giving rise to twodimensional (2D) QW states. Metallic QW states are similar to those known in semiconductor structures, except that the shorter Fermi wavelength allows spatially narrower confinement of highdensity electrons (1-6). The metallic OW states are realized in QW structures based on metals having nearly free-electron-like sp states, such as Ag/Si (5), Pb/Si (6), and noble metals on metal substrates (1-4). The resultant 2D electronic structures have been intensively studied by using angle-resolved photoemission spectroscopy (ARPES) (1–6).

Depending on the number of conductive layers, layered complex oxides that are both low-dimensional and strongly interacting often exhibit unusual physical properties, such as hightemperature superconductivity in cuprates, triplet superconductivity in ruthenates, and enhancement of colossal magnetoresistance in manganites (7). The properties originate from stacked conductive layers, which strongly correlated electrons, sandwiched by block insulating layers. The lowering of the dimensionality changes the interaction among the spin, charge, and orbital degrees of freedom. However, systematic control of the dimensionality while keeping the fundamental electronic parameters fixed has not yet been conducted, owing to the difficulties in the synthesis of a homologous series of layered complex oxides (7).

The close structural similarities of layered oxides to metallic QW structures have motivated researchers to create low-dimensional systems in a controllable fashion using artificial structures of a strongly correlated oxide. That is, the artificial oxide structures provide a setting in which to study the behavior of strongly correlated electrons in reduced dimensions and to control the extraordinary physical properties of strongly correlated oxides (8). Although the carrier confinement at the interface of oxide semiconductors has been reported (9, 10), the metallic QW states having the dimensional controllability has not been achieved yet in oxide artificial structures.

We chose $SrVO_3$ ultrathin films epitaxially grown on Nb-doped $SrTiO_3$ (Nb:STO) as a possible candidate for creating QW states having 2D electron-liquid states. $SrVO_3$ is a typical Fermiliquid metal with the simple $3d^1$ configuration (7, 11–13), and its electronic structures can be simulated by use of the Hubbard model without

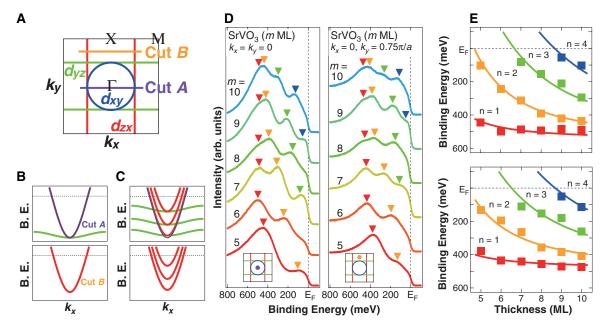


Fig. 1. (**A**) FS cross sections in the ΓXM ($k_z = 0$) plane of SrVO₃. The FS consists of three cylindrical FS sheets originating from three V 3d bands: d_{xy} (blue), d_{yz} (green), and d_{zx} (red) (fig. S2) (1d). The measurement lines (cuts A and B) of the present ARPES are also shown. (**B**) Band dispersions of bulk SrVO₃ predicted from band structure calculations (1T) for cuts A and B. (**C**) Quantization states in SrVO₃ ultrathin films expected from the band structure. Orbital-selective quantization occurs, reflecting the anisotropic character of V 3d states. The resultant

subband dispersions are illustrated for cuts A and B. (**D**) ARPES spectra of SrVO₃ ultrathin films varying the film thickness obtained at points (0, 0) (left) and (0, 0.75 π /a) (right). The peak structures derived from QW states are marked by the colored triangles. (**E**) Structure plots showing the energy positions of corresponding QW-state peaks (data markers) versus the film thickness for points (0, 0) (top) and (0, 0.75 π /a) (bottom). The solid lines are predictions for the QW states (with n = 1 to 4) from the phase-shift quantization model (1 = 6, 16).

explicit consideration of the oxygen p orbital (14). The metallic V 3d states located near the Fermi level ($E_{\rm F}$) in SrVO $_3$ ultrathin films are expected to be highly confined in the QW structures formed between the vacuum (surface) and the Nb:STO substrate (interface) because Nb:STO has a large band gap of 3.2 eV below $E_{\rm F}$ (15, 16). Our previous angle-integrated PES studies on SrVO $_3$ /Nb:STO revealed that the metallic V 3d states near $E_{\rm F}$ are located within the band gap of Nb:STO, although strong electron correlation in the SrVO $_3$ ultrathin films manifests itself in the metal-insulator transition (MIT) that occurs at a critical film thickness of 2 to 3 monolayers (ML) (15).

The Fermi surface (FS) sheets of bulk SrVO₃ with cubic symmetry (fig. S2) (16) are essentially formed from three intersecting cylinders containing the V 3d d_{xy} , d_{yz} , and d_{zx} states (16, 17). Each state has a 2D character in the xy, yz, and zx planes, respectively. The 2D nature of each state causes the FSs in the ΓXM emission plane (the $k_r - k_r$ plane, with $k_z = 0$) to consist essentially of two parallel lines originating from d_{zx} and d_{yz} states and a circular FS from the d_{xy} state (Fig. 1A) (16-20). The band structures along the Γ -Xdirection (cut A) consist of three bands: two degenerate parabolic dispersions derived from the d_{yy} and d_{zx} states and a nearly nondispersive d_{yz} state (16–20). However, the band dispersion along the X-M direction (cut B) is described by one parabolic band from the d_{zx} state (Fig. 1B). When the SrVO₃ film becomes sufficiently thin along the z direction to realize the quantum confinement of V 3d electrons in the film, the bands derived

from d_{zx} and d_{yz} are expected to form quantized states because these orbitals expand along the z direction. On the other hand, the d_{xy} state remains unchanged because of its 2D character in the xy plane. Thus, the quantized d_{yz} and d_{zx} states and one bulk-like d_{xy} state are expected to be observed for cut A, whereas simple quantized states derived from the d_{zx} orbital are expected for cut B (Fig. 1C).

Certain aspects of 2D OW states in metal films have been studied by use of ARPES (1-6). By measuring ARPES spectra, the energy position of each quantization state and the in-plane energy dispersion of the confined electrons were obtained directly. Figure 1D shows the ARPES spectra for SrVO₃ ultrathin films varying in overlayer thickness obtained at the (0, 0) and $(0, 0.75\pi/a)$ points in the ΓXM emission plane. The energy positions of the several quasiperiodic peaks evolved as a function of overlayer thickness: With increasing thickness, one additional peak appears after another in the ARPES spectra at $E_{\rm F}$, and their peak positions shift to higher binding energies. The peak shift apparently converged around 500 meV, which corresponds to the bottom of the V 3d conduction bands (18-20). The spectral changes suggest the formation of metallic QW states in SrVO₃/Nb:STO.

In order to show the dependence of the quantized electronic states on the film thickness more clearly, we plotted the binding energies as a function of the $SrVO_3$ film thickness (Fig. 1E). For quantitative analysis, we invoked the usual phase-shift quantization rule, which has been used successfully in the interpretation of metallic QW states in noble metal systems (I-6, I6). The

solid lines in Fig. 1E represent the calculated results for the QW states. Comparing the experimental and calculated data, the experimental binding energies are fairly well reproduced by calculations based on the phase-shift quantization rule. This good agreement indicates that the metallic V 3d states located near $E_{\rm F}$ in SrVO $_3$ are confined in the QW structures.

The orbital-selective quantization of the OW states could be seen in the in-plane band dispersion determined with ARPES. The band dispersion of the subbands formed for an 8-ML SrVO₃ ultrathin film is shown in Fig. 2. As expected from the illustration in Fig. 1C, two types of subbands were observed for the band dispersion along cut A (Fig. 2A); one is the parabolic band, which approaches and eventually crosses $E_{\rm F}$ with increasing distance from the Γ point, whereas the other is the nearly flat band. These two bands are degenerate at the Γ point. However, only parabolic subbands were observed along cut B (Fig. 2B). Considering the anisotropic orbital nature of the 3d states, the flat bands seen only in Fig. 2A and the parabolic band seen in both cases originate from the quantized d_{vz} and d_{zx} states, respectively. From the structure plots shown in Fig. 1E, the quantum number of each subband is n = 1, 2, and 3 fromthe bottom. Because the original d_{zx} and d_{xy} states are degenerate along the Γ -X direction (Fig. 1B), the band dispersion of the first quantized d_{zx} state almost overlaps with that of the bulk-like d_{xy} state because their bottom energies are nearly equal. The existence of the original d_{xy}

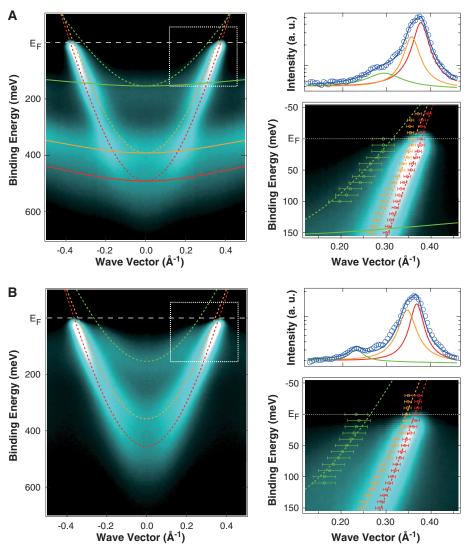


Fig. 2. Orbital-selective quantization of V 3d states in SrVO₃ ultrathin films. (**A** and **B**) Intensity plots of ARPES spectra for 8-ML SrVO₃ ultrathin films for cuts A (A) and B (B). The intensity plots are symmetrized with respect to the center line and averaged (figs. S5 to S9) (1G). The dashed and solid lines present the fitted curves to the dispersion using Eq. 1 for the d_{zx} and d_{yz} quantization states, respectively. Red, orange, and green lines correspond to states in which n = 1, 2, and 3, respectively. Magnifications of the boxes to the left are shown correspondingly to the right, together with the momentum distribution curve (MDC) at E_F with an energy window of ± 5 meV. The blue line for each MDC curve is the sum of three Lorentzians corresponding to the respective subbands. The peak positions determined from MDCs are shown by the open circles (figs. S10 to S12) (1G).

band irrespective of the quantization of d_{yz} and d_{zx} bands was confirmed through ARPES measurements by using dichroic effects along different cuts where the degeneracy was lifted (16).

Although the quantization reflecting the anisotropic V 3d states itself is expected from band structure calculation, orbital-selective quantization has important implications for thickness-dependent MIT in SrVO₃ ultrathin films (15). SrVO₃ ultrathin films underwent a thickness-dependent phase transition from metal to insulator at a film thickness of less than 4 ML. In the structure plot (fig. S4) (16), the quantization states did not exist in occupied states below 2 ML. This thickness corresponds to the critical thickness at which MIT occurs in SrVO₃ thin films, suggesting that the thickness-dependent MIT in SrVO₃

ultrathin films is related to the orbital-selective quantization effect.

Lastly, there is the band renormalization of subbands located near $E_{\rm F}$. In the in-plane dispersion of the subbands shown in Fig. 2, the subband dispersion becomes narrower with approaching its bottom energy to $E_{\rm F}$. For quantitative discussion, the band renormalization factor ($Z_{\rm sub}$) is evaluated with a least-squares fitting of the subband dispersion by using the following equation

$$E_{\rm sub}(k_{/\!/}) = Z_{\rm sub}E^{\rm TB}(k_{/\!/}) + \epsilon^*$$
 (1)

where $E^{\rm TB}(k)$ is the band dispersion calculated by using the tight-binding parameters (16, 17). Only $Z_{\rm sub}$ and ε^* are adjustable parameters so as to fit

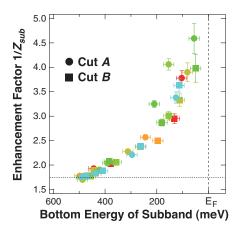


Fig. 3. Plots of the enhancement factor (1/Z_{sub}) for respective QW subbands as a function of their bottom energy for various SrVO₃ film thicknesses. The data marker colors correspond to those of the respective ARPES spectra shown in Fig. 1D.

the in-plane dispersion of the subbands. As can be seen in Fig. 2, the fitted curves by using Eq. 1 adequately reproduce the in-plane dispersion of the subbands.

The relation between the enhancement factor $(1/Z_{\rm sub})$ and the binding energy of the quantization states is summarized in Fig. 3. As the binding energy of quantization states approaches $E_{\rm F}$, the effective mass of the subbands is considerably enhanced. Such a mass enhancement has not been observed in QW structures based on metals having nearly free-electron-like states (1-6). We conclude that the mass enhancement in the subbands is associated with strong interaction among V 3d electrons confined in the QW structures (8, 11-13). There is one feature of the band renormalization that seems to contradict with the Fermi-liquid picture: If each subband can be described as a simple Fermi liquid, subbands located at lower binding energies should have weaker band narrowing owing to the reduction of correlation effects with reduced band filling (21). The unusual band renormalization may be related to the complicated interactions induced by the reduction of dimensionality, such as orbital selective Mott transition (22) and orbital and/or electronic reconstruction at the surface and the interface, but this issue remains to be resolved.

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Acknowledgments: The authors are very grateful to I. Matsuda,

S. Okamoto, and M. J. Rozenberg for useful discussions and K. Ono and A. Yagishita for their support in the experiment at the Photon Factory (PF), KEK. This work was supported by a Grant-in-Aid for Scientific Research (A19684010 and S22224005) from the Japan Society for the Promotion of Science (JSPS) and the JST PRESTO program. K.Y. acknowledges the financial support from JSPS

for Young Scientists. This work at KEK-PF was done under the approval of the Program Advisory Committee (proposals 08S2-003 and 09S2-005) at the Institute of Materials Structure Science, KEK.

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17 March 2011; accepted 25 May 2011 10.1126/science.1205771

Explosive Percolation Is Continuous

Oliver Riordan*† and Lutz Warnke*

"Explosive percolation" is said to occur in an evolving network when a macroscopic connected component emerges in a number of steps that is much smaller than the system size. Recent predictions based on simulations suggested that certain Achlioptas processes (much-studied local modifications of the classical mean-field growth model of Erdős and Rényi) exhibit this phenomenon, undergoing a phase transition that is discontinuous in the scaling limit. We show that, in fact, all Achlioptas processes have continuous phase transitions, although related models in which the number of nodes sampled may grow with the network size can indeed exhibit explosive percolation.

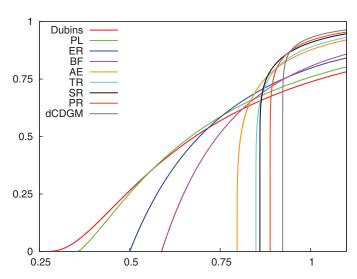
he quintessential example of a phase transition in statistical physics is the emergence of the giant component in the mean-field random graph: Erdős and Rényi (1) discovered that in a graph or network constructed in a completely (uniformly) random manner, as the ratio between the number m of edges (also called links) and the number n of vertices (nodes) passes ½, there is a dramatic change in the component (connectivity) structure. More precisely, if $m \sim tn$, where t represents time, for $t < \frac{1}{2}$, the number C of nodes in the largest connected component is logarithmic in n, whereas for $t > \frac{1}{2}$, C is of order n: A macroscopic component emerges at the critical time $t_c = \frac{1}{2}$; this component is known as the giant component, even when its size is a small constant times n. Taking the natural scaling limit, there is a function $\rho(t) = \rho^{ER}(t)$ given by a simple formula such that $C \sim \rho(t)n$ when $m \sim tn$, with $\rho(t) > 0$ if and only if $t > \frac{1}{2}$.

The Erdős-Rényi (ER) model and its phase transition are of fundamental importance for two main reasons. Mathematically, the properties of this phase transition are extremely well understood, including the fine details of the behavior at and near the critical point, and the dynamics of the transition [see, for example, (I-5)]. This model serves as the natural reference point when studying a wide range of phase transitions in mathematics and statistical physics: Similarities to and differences from the mean-field model are key to understanding more complex and perhaps less tractable models.

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Second, random graphs are the natural mathematical models for complex or disordered networks in the real world. The mean-field model is unlikely to be appropriate itself for any particular application, but intuition from its study is the starting point for understanding more recent network models, such as the scale-free models of Barabási and Albert (6) and many others. Studying the emergence of large-scale connectedness as an edge-density parameter is varied corresponds to varying the proportion of links in an existing network that fail, and asking whether it remains connected on a large scale (7, 8). Alternatively, thinking of the links as contacts that may spread a disease, the emergence of a giant component corresponds to epidemic spread of a disease rather than localized outbreaks (9, 10).

Fig. 1. The scaling limit of the giant component size, for various random graph models. The ER, power-law (PL), and Dubins models fall in the BJR family. The BF rule, SR, and PR are Achlioptas processes; the adjacent edge (AE) rule (23), triangle rule (TR) (23), and dCDGM (26) are Achlioptas-like processes. The figure is based on simulations with n =109 (PL and Dubins) or $n = 10^{12}$ vertices. The last five curves appear to be discontinuous at the critical point, but this is not the case.



For the reasons above, phase transitions in random graph models of many types have been widely studied [see the many references in (11)]. A key question is, how do the transitions in the different models relate? Which features are specific to the model, and which are universal? In 2007, Bollobás, Janson, and Riordan (BJR) (11) introduced a very general family of models that includes many special cases previously studied. They showed that for each model M in this family, there is a function $\rho^{M}(t)$ describing the rescaled size of the giant component; this function is zero up to some critical value t_c (which may be 0) and positive for $t > t_c$. For all models in the BJR family, $\rho^{M}(t)$ is continuous, but a huge range of different behaviors near t_c is possible (Fig. 1). For any critical exponent $\gamma \ge 1$, examples are given in (11) with $\rho^{M}(t_{c}+\varepsilon) \sim A\varepsilon^{\gamma}$ as $\varepsilon \to 0$, where A is a constant. Infinite order transitions also occur in natural models, with $\rho^{M}(t_{c} + \varepsilon) \approx \exp(-1/\sqrt{\varepsilon})$ [see (12) and the references therein].

With continuity of the function p seeming to be the only property that does not change from one model to the next, it was extremely exciting when Achlioptas, D'Souza, and Spencer (13) announced that a rather simple dynamical modification of the ER model seemed to undergo a discontinuous phase transition, a phenomenon now known as explosive percolation. Here, we show that this is not the case. Our argument is relatively

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