Quantum Confinement Induced Metal-Insulator Transition in Strongly Correlated Quantum Wells of SrVO₃ Superlattices

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Dynamical mean-field theory (DMFT) has been employed in conjunction with density functional theory (DFT+DMFT) to investigate the metal-insulator transition (MIT) of strongly correlated 3d electrons due to quantum confinement. We shed new light on the microscopic mechanism of the MIT and previously reported anomalous subband mass enhancement, both of which arise as a direct consequence of the quantization of V \( \text{yz}(yz) \) states in the SrVO₃ layers. We therefore show that quantum confinement can sensitively tune the strength of electron correlations, leading the way to applying such approaches in other correlated materials.

In strongly correlated quantum materials, the interaction energy between electrons is comparable to their kinetic energy, leading to many-body behavior and the emergence of qualitatively new phenomena [1]. For example, transition metal oxides host properties as diverse as colossal magnetoresistance, high-temperature superconductivity, and Mott insulating phases, each of which have huge potential for future device and technology applications [2, 3]. More recently, substantial advances in the quality and control of layer-by-layer growth methods have facilitated designed transition metal oxide heterostructures and superlattices (SL), often focusing on emergence at interfaces and/or surfaces [4]. Here, we show that correlated electronic behavior may be delicately tuned by quantum confinement, which narrows the effective bandwidth of the correlated quantum well (QW) subbands, and establishes another means to tune physical properties to suit applications. We illustrate this by driving the prototypical correlated metal, SrVO₃, through a metal-insulator transition (MIT) to a Mott insulating phase, in excellent quantitative agreement with the experimental spectral function extracted from recent spectroscopic measurements [5].

In the bulk, SrVO₃ is a well-characterized correlated metal [6]. Sharp 3d quasiparticle (QP) bands at low excitation energies lead to a well-defined Fermi surface [7], while localized states form incoherent Hubbard sidebands at an energy scale comparable with the Coulomb repulsion parameter, \( U \) [8, 9]. Together, these yield the familiar three-peaked spectral function [3, 10]. While density functional theory (DFT) often adequately describes QP states (once renormalization is accounted for), it is not capable of capturing the many-body behavior, e.g. Hubbard sidebands are completely absent. Dynamical mean-field theory (DMFT), on the other hand, is able to describe all of the on-site local correlations [3, 10], and has been well-tested on SrVO₃ with very good results, including the energetics and spectral weight of Hubbard sidebands and QP renormalization [3, 8, 11, 12].

The importance of electron correlations in a system may be gauged in terms of the ratio, \( U/W \), where \( W \) is the bandwidth. This ratio is known to be significantly enhanced at the SrVO₃ surface [13, 14], and in few layer systems [15, 16]. However, such systems have reduced coordination at the surface, which is often complicated by reconstruction and relaxation [14]. For example, an insulating phase has been observed in SrVO₃ thin films for thicknesses below approximately 6 unit cells, which was attributed to dimensional crossover due to reduced coordination [15, 17]. In bilayer SrVO₃, a DFT+DMFT study has established that a Mott insulator emerges due to crystal field (CF) effects [16]; similar CF-induced insulating phases are predicted in other vanadates [18–21]. Although QW structures are well-known in semiconducting [22] and free electron-like [23] materials, their application to correlated 3d metals has only recently been realized in a select few transition metal oxides [24, 25], including in few-layer SrVO₃ thin films [26]. However, the precise nature of these states, including their unusual subband renormalization [26, 27], is not yet well understood, although this is essential in order to exploit their properties.

In this Letter, we show that quantum confinement can be employed to precisely control strongly correlated electron behavior. We reveal the microscopic mechanism involved using a combination of DFT, DFT+DMFT and tight-binding models applied to the prototypical correlated oxide SrVO₃, which we embed within layers of SrTiO₃ in a SL for direct comparison with previous experimental results [5]. Our results accurately reproduce the trends of the experimental data, and reveal that the microscopic mechanism for the MIT in SrVO₃/SrTiO₃ SLs is due to quantum confinement. Our results also shed light on previous observations of anomalous mass enhancement in SrVO₃ QWs [26, 27], which can be naturally explained as consequences of quantization. These results underline the efficacy of SL engineering in tuning strongly correlated behavior, and leads the way to harnessing functional correlated electron properties in other materials [28].

DFT calculations were performed using the all-electron full potential augmented plane wave ELK package [29] within the local density approximation (LDA). The results are in excellent agreement with previous pseudopotential calculations [5]. The structures of the \((\text{SrVO₃})_p/(\text{SrTiO₃})_q\) SLs are shown in Fig. 1(a) for the three SLs investigated here, with \(pq = 2:7, 3:6\) and \(6:5\). These were chosen for direct comparison with previous experimental results [5, 30]. Experimental lattice parameters were used: the in-plane parameters
were those of the (LaAlO$_3$)$_{0.3}$(Sr$_2$TaAlO$_6$)$_{0.7}$ (LSAT) substrate, $a = b = 3.868$ Å, and out-of-plane parameters were $c_{2:7} = 4.00$ Å, $c_{3:6} = 3.97$ Å and $c_{6:5} = 3.92$ Å. Even in the absence of the SL heterostructure, the different in- and out-of-plane lattice parameters weakly break the V $t_{2g}$ degeneracy into 3d $xy$ and $xz(yz)$ orbitals.

In order to interpret our results, the LDA results for each SL were parameterized using a quantized Bohr-Sommerfeld tight-binding (QTB) model [23, 31]. In this model, the shape of the TB bands was fixed to that of bulk SrVO$_3$, and the free parameters represent the band centers, bandwidths and quantization parameters. In this way, CF splitting and band narrowing between $xy$ and $xz(yz)$ orbitals can be fully captured. The results of fitting this model to the full ELK calculation is shown in Fig. 1(b) for the 2:7 SL, demonstrating the excellent agreement between the two. The quantized nature of the V $xz(yz)$ orbitals is clearly evident, leading to two sub-bands ($n = 0, 1$) originating from the two SrVO$_3$ layers. In Fig. 1(b), we also show the “intrinsic” TB bands, which correspond to bulk-like bands before the quantization conditions are applied and represent the intrinsic 3D electronic structure from which the QW states emerge.

For the DMFT calculation, the TRIQS/CTHYB continuous-time quantum Monte Carlo (CTQMC) solver [32] and the TRIQS library [33] were used with the Hubbard–Kanamori interaction Hamiltonian, $\beta = 40$ eV$^{-1}$ (290 K), $J = 0.75$ eV and the fully localized limit double counting term, as used previously [16, 18, 19]. The DMFT cycle requires multiple impurities depending on the SL structure, as illustrated schematically in Fig. 1(a). The systems investigated correspond to a single impurity for the 2:7 SL and bulk calculations, and two and three impurities for the 3:6 and 6:5 SLs, respectively. In each case, the impurities are considered to be independent of one another. Only the V $t_{2g}$ bands were projected (using Wannier projectors) to construct the LDA Hamiltonian in Wannier space to be used in the DMFT calculation [34]. The results presented use the fully charge self-consistent DFT+DMFT technique as implemented in the TRIQS/DFTTools library [35]. We obtain similar results with the one-shot approach with the exception of the orbital charges [31], which is consistent with other studies [18–20]. For each SL structure, $U$ was varied in the range 5.5 to 6.25 eV, and $\bar{U}_{\text{MIT}}$ was located, corresponding to the $U$ at which the SL becomes insulating. In Fig. 2(a), we show the orbitally-averaged QP residue, $\bar{Z} = (\Sigma Z_i) / N$, for each SL, showing how $\bar{U}_{\text{MIT}}$ increases by $\sim 0.2$ eV for each SL. Here, $Z_i$ was determined from the ith orbital self-energy on the Matsubara frequency axis.

We begin by ensuring that our DFT+DMFT calculations accurately describe the experimental system. In Figs. 2(b–d), we compare quantities extracted from x-ray absorption spectroscopy (XAS) and resonant inelastic x-ray scattering (RIXS) experiments [5] with the corresponding theoretical quantities from our DFT+DMFT calculations for $U = 5.7$ eV, chosen to reproduce the phenomenological behavior of the experiments. As demonstrated by Figs. 2(b–d), our DFT+DMFT calculations not only capture the qualitative behavior, but also yield excellent quantitative agreement with the experiment trends. We note that, although the 2:7 SL is macroscopically insulating, it has a small QP spectral weight in the spectroscopic experiments due to properties of the sample.

Insight into the microscopic mechanism for the MIT can be obtained by analyzing the QTB model for each SL. In Fig. 2(e), the bandwidths of the SLs from the DFT calculations...
are shown alongside those from the QTB model. While the in-plane $xy$ orbitals experience a slight narrowing for thinner SrVO$_3$ SLs, the overall band narrowing of the (quantized) out-of-plane $xz(yz)$ orbitals is substantial, leading to a bandwidth reduction of $\sim 70\%$ for the 2:7 SL [5]. As expected from the quality of the fits in Fig. 1(b), good agreement is observed between the QTB and DFT results. However, this behavior is not captured at all by the “intrinsic” bandwidths, which correspond to the effective 3D bands of the QTB model before quantization. These results demonstrate that the band narrowing in Fig. 2(c) is due to quantization of the $xz(yz)$ orbitals, which has a more pronounced impact for thinner SrVO$_3$ layers. The band narrowing of the thinner SLs leads to a greater $U/W$ ratio, which results in stabilization of the insulating phase, as illustrated by the DFT+DMFT calculations shown in Fig. 2(a).

A previous DMFT study has attributed CF effects as being the principle factor driving the MIT in bilayer SrVO$_3$ on SrTiO$_3$ [16], and it is pertinent to ask what role, if any, the CF plays in our system. In Ref. 16, strain induced by the SrTiO$_3$ substrate led to a lowering of the $xy$ orbitals by 180 meV due to the CF. In contrast, we find a CF splitting of $\leq 51$ meV in favor of the $V xz(yz)$ bands in our SLs, in part owing to the lower strain imparted by the LSAT substrate. To reproduce the effects of this CF, we have calculated strained SrVO$_3$ with a volume-conserving strain of 1%, which leads to similar CF splitting of 53 meV [31]. This 3D system, which reproduces the CF levels of our SLs but without the quantization effects, is shown in Fig. 2(a), and shows very similar behavior to the bulk cubic system. Therefore, we confidently rule out CF effects as a dominant factor in our SLs.

Finally, we discuss the correlated behavior of the quantized electron states, taking the 6:5 SL as an example. Figure 3(a-c) shows the orbital- and layer-resolved occupation number, $n_{\omega}$, QP residue, $Z$, and spectral weight at the Fermi level, $A(\omega = 0)$, as a function of $U$, illustrating the transition to the insulating phase at $U = 6.05$ eV for this SL. As presented in Fig. 3(a), each layer (impurity) exhibits a sizeable orbital polarization in favor of the $xy$ orbitals, which is exaggerated both at the outer (interface) layer and in the insulating phase. This behavior is consistent across all SLs, and originates from a small polarization in the DFT calculations due to the local CF, which is subsequently amplified in the DMFT cycle [31]. The outer layer (impurity 1) is significantly “more correlated”, experiencing a smaller QP residue $Z$ than the other layers [Fig. 3(b)], corresponding to a greater renormalization factor, $1/Z$. Near the MIT, this leads to a collapse in the spectral weight at $\omega = 0$ of the interface layer [Fig. 3(c)]. In this sense, the more correlated interface layer simultaneously triggers the MIT in the remaining layers, in much the same way as suggested for SrVO$_3$ bilayers in Ref. 16.

The DMFT spectral functions, $A(\omega)$ and $A(k,\omega)$, are shown in Fig. 3(d-f) and Fig. 3(i-j) for $U = 5.7$, 6.025 and 6.05 eV, respectively, and were calculated using the maximum entropy method within TRIQS [36]. The renormalization of the V bands increases from a factor of $1/Z \approx 3.5$ at

FIG. 3. Correlated QW electronic structure. (a) Occupation number, $n_{\omega}$, (b) QP residue, $Z$, and (c) spectral function at the Fermi level ($A(\omega = 0)$) of all impurity (imp) correlated orbitals from DFT+DMFT across the MIT for the 6:5 SL. (d-f) The momentum-integrated V $t_{2g}$ spectral function, $A(\omega)$, for $U = 5.7$, 6.025 and 6.05 eV respectively. (g) DFT bandstructure of the 6:5 SL, with V $t_{2g}$ band characters shown. (h) DFT+DMFT momentum-resolved spectral function, $A(k,\omega)$, of the 6:5 SL for $U = 5.7$ eV, showing directly the renormalization of the correlated bands. (i-j) DFT+DMFT spectral function close to the MIT for $U = 6.025$ and 6.05 eV, respectively.
Our results reveal quantization as the primary source of anomalous mass enhancement in embedded (or capped) few-layer SrVO$_3$.

In summary, our results establish quantum confinement engineering as a sensitive method to tune the correlated electron behavior of 3$d$ electron systems. We demonstrate our approach using SLs of few-layer SrVO$_3$ embedded in SrTiO$_3$; this system exhibits an MIT due to a reduction in bandwidth by varying the number of SrVO$_3$ layers. The microscopic mechanism we reveal is a direct consequence of quantum confinement, and is distinct from previous studies invoking CF effects or dimensionality, demonstrating the excellent versatility of the MIT in SrVO$_3$ with respect to different control parameters. Similar to Ref. 16, we find that the outer layers at the interface of the quantum well are more strongly correlated, and trigger the transition in the rest of the layer. Our interpretation is also able to naturally explain the anomalous mass enhancement previously reported in ARPES measurements. Together, these results demonstrate the potential of employing quantum confinement as a tuning parameter for correlated electron behavior in engineered SLs. This approach also has the significant benefits that surface effects are avoided and that large (bulk-like) volumes of the system may respond to external stimuli. Aside from the expected improved performance of such a SrVO$_3$ device as a Mott transistor, quantized SLs made of other correlated materials are likely to show improved properties, e.g. superconducting devices in cuprate SLs, conductivity in nickelate SLs or spintronic devices.

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**Supplementary Information for “Quantum Confinement Induced Metal-Insulator Transition in Strongly Correlated Quantum Wells of SrVO₃ Superlattices”**

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**COMPARISON WITH EXPERIMENTAL QUANTITIES**

Figure S1 illustrates the main experimental spectroscopic results on the superlattices (SL) [1, 2], in which transport measurements established that the 2:7 and 3:6 SLs were insulating, whereas the 13:4 SL was metallic. The 6:5 SL was found to be metallic at room temperature, with an metal-insulator transition (MIT) at low temperature. The experimental results (performed at room temperature) show the evolution in correlated electron behavior extracted from x-ray absorption spectroscopy (XAS) and resonant inelastic x-ray scattering (RIXS).

For completeness, we briefly outline the experimental properties and how they were extracted here. The metallicity [Fig. S1(a)] was extracted from both XAS and RIXS as the leading edge of the O K edge XAS and from the intensity of the quasi-elastic peak in V L edge RIXS. The quasiparticle (QP) bandwidth [Fig. S1(b)] was extracted from the SrVO₃ layer contribution to the O K edge XAS as the full-width at half-maximum of the QP peak. The QP spectral weight [Fig. S1(c)] was also extracted from the SrVO₃ layer contribution to the O K edge XAS as the ratio of the area under the QP peak to the total area under unoccupied V 3d states (i.e., the sum of QP, upper Hubbard band (UHB) and ε_g spectral weights). Occupied states are not accessible in O K edge XAS. Finally, the UHB energy [Fig. S1(d)] is accessible to both XAS and RIXS. From O K edge XAS, the UHB peak is directly observed, and its center is shown here. From V L edge RIXS, the UHB energy is available from transitions from occupied QP states to the unoccupied UHB. Both show equivalent evolution with SL structure.

Figure S2 shows a schematic illustration comparing the extracted experimental quantities with their DFT+DMFT (density functional theory with dynamical mean-field theory) definitions. The QP bandwidth has been extracted from the DMFT spectral function by obtaining the width defined by the minima around the central QP peak. The QP ratio was determined by taking the ratio of the the quasiparticle weight (labeled Q in Fig. S2) and the UHB weight (labeled U). Finally, the energy of the UHB was obtained by locating the peak in the DMFT spectral function, referenced to ω = 0.

In the experimental RIXS process, the UHB peak energy represents the peak in the joint QP and UHB density of states, and therefore is referenced to an energy ω < 0. To compare the theoretical and experimental quantities, we therefore shift all theoretical quantities to match for the 2:7 SL (the shift is −0.584 eV).

**FIG. S1.** Evolution of correlated electron behavior from experimental x-ray absorption spectroscopy (XAS) and resonant inelastic x-ray scattering (RIXS) measurements of SrVO₃/SrTiO₃ superlattices, reproduced from Ref. 2. From top to bottom, evolution in (a) metallicity of SLs, (b) quasiparticle (QP) bandwidth, (c) QP spectral weight and (d) the energy of the upper Hubbard band are shown.

**FIG. S2.** A schematic illustration showing how the variables in the theory-experimental comparison were determined.
FIG. S3. The DFT $t_{2g}$ partial density of states (PDOS) of the bulk SrVO$_3$, 2:7, 3:6 and 6:5 SLs. The dashed borders outline which plots belong to the corresponding structure. Each panel shows the PDOS of the inequivalent V atoms in each structure, labeled by their impurity number in the DMFT cycles (imp 1 refers to the interface). The arrows indicate the contributions of quantized states to each inequivalent V atom. The greatest contribution are from the relatively flat bands along Γ-X.

DENSITY FUNCTIONAL THEORY CALCULATIONS

DFT calculations were performed using the E不知 FP-LAPW (full potential linearized augmented plane wave) code within the local density approximation (LDA) [3]. The results are in excellent agreement with previous pseudopotential calculations within the generalized gradient approximation (GGA) of the same SLs [2]. Previous PES work [4], show how the dimensionality of SrVO$_3$ influences the MIT. In their results, ten SrVO$_3$ layers closely resembles bulk behavior. From this, we approximate the 13:4 SL in Ref. 2 with bulk DFT+DMFT calculations.

Self-consistency was achieved on a 12 × 12 × 4 mesh in the full Brillouin zone (BZ) for relatively low computational cost with sufficient sampling, corresponding to 84 k-points in the irreducible (1/16$_{th}$) BZ. To stabilize the DFT self-consistent cycles, small values of mixing of the new potentials was used, at the cost of computational time. For bulk SrVO$_3$, a k-mesh of 12 × 12 × 12 was used (84 k-points in the 1/48$_{th}$ irreducible BZ).

The partial densities of states (PDOS) of $t_{2g}$ orbitals are shown in Fig. S3 for the bulk and 2:7, 3:6 and 6:5 SLs. Sharp peaks in the PDOS reflect the quantized electronic structure along c. For the inner layers of the thicker SLs, the PDOS more closely resembles that of bulk SrVO$_3$, e.g. impurity 3 of the 6:5 SL. Near the interface, the $xz(yz)$ PDOS extends to higher energies as a result of mixing of these states with Ti states in the SrTiO$_3$ layer.

The characters of the subbands are shown in Fig. S4 for each of the different V and Ti sites, using the 6:5 SL as an example. As expected, the V bands dominate the character at the Fermi level, with weak contribution from interfacial Ti ions. The spatial distribution of the subband wavefunctions of the SrVO$_3$ quantum wells can be seen directly in the characters. The V $n = 0$ subband, with greatest amplitude in the centre of the well, has strong character in the central V ion and weak character at the interface. Correspondingly, the V $n = 2$ subband has strongest character at the interface and is almost absent in the second layer close to where a node is expected in the quantum well wavefunction. At higher energies, the quantized V $e_g$ subbands appear above 1 eV.
FIG. S4. The DFT V and Ti $t_{2g}$ band characters of the 6:5 superlattice (SL). The thickness of the lines indicates the total character of each V (left) and Ti (right) site. The top row shows the band characters at the interface, while the bottom row shows the character in the center of each layer. On the right, a schematic illustration of the real-space probability distribution of the quantized subbands in the out-of-plane direction of the SrTiO$_3$ layers is shown. The edge of the box does not coincide with the interface Ti ion or its neighboring SrO layer due to the finite phase accumulated at the interface.

The Ti band characters shed light on the broadening of the Ti bands in the DFT+DMFT calculations shown in Fig. 3 of the main manuscript. As above, the central Ti ion contributes strongly to the Ti $n = 0$ and $n = 2$ subbands. On the other hand, the interfacial Ti ion contributes significantly to the Ti $n = 1$, 2 and 3 subbands, with the largest contribution to the Ti $n = 2$ subband. The interfacial Ti ion mixes most strongly with the V orbitals, which are the correlated orbitals in the subsequent DMFT cycle. This demonstrates how the spatial penetration of the Ti $n = 1$ and $n = 2$ subbands into the correlated SrVO$_3$ layers leads to substantial broadening of these subbands in the subsequent DMFT cycle. In contrast, the Ti $n = 0$ subband is spatially deep within the SrTiO$_3$ layer and does not feel the effects of the correlated SrVO$_3$ orbitals very strongly, remaining reasonably sharp even in the insulating phase [Fig. 3(j) of the main manuscript].

QUANTIZED TIGHT-BINDING MODEL

Bulk tight-binding bands

The tight-binding (TB) model was constructed up to 12$^{th}$ nearest neighbors, consisting of 24 hopping terms, $t_i$, up to $[l, m, n] = [2, 2, 2]$. For the $xy$ band the TB dispersion, $\varepsilon_{xy}$, is given by,

$$\varepsilon_{xy} = E_{xy}^0 + \sum_{lmn} t_{xy}^{lmn} \cos (lk_x + mk_y + nk_z),$$

where the band energy, $E_{xy}^0$, corresponds to the crystal field energy. Since the purpose of our model is to accurately describe the bulk 3D DFT band structure, we do not attempt to analyze individual parameters, as has been done before [5]. Although terms corresponding to the 5$^{th}$ nearest neighbor and higher had a magnitude of less than 10 meV, these terms were found to be necessary to adequately describe the FP-LAPW band structure. After fitting this model to the bulk LDA band structure in the full cubic Brillouin zone, we find the r.m.s.
Quantum confinement

In order to account for the effects of quantum confinement of the V 3d electrons in the SrVO$_3$ layers, we apply the Bohr-Sommerfeld phase accumulation model [6],

\[ 2k_z^n(E)L + \delta(E) = 2\pi n, \]

where \( n = 0, 1, 2, \ldots \) is the quantum number, \( 2k_z^n(E)L \) is the total phase accumulated in traveling through the SrVO$_3$ layer and back, \( k_z^n(E) \) is the quantized out-of-plane wavevector, \( L = mc \) is the SrVO$_3$ layer thickness (\( m \) and \( c \) are the number of SrVO$_3$ layers and c-axis lattice parameter of the SrVO$_3$ layers, respectively). \( \delta(E) \) is the total phase acquired due to reflection at both SrVO$_3$/SrTiO$_3$ interfaces. For asymmetric quantum wells, e.g. thin overlayers with a vacuum interface, \( \delta = \phi_1 + \phi_2 \) is composed of different individual phase shifts at each reflection; in our case of symmetric barriers, \( \delta = 2\phi \), where \( \phi \) is the phase at a single SrVO$_3$/SrTiO$_3$ interface. In general, \( \delta = \delta(E) \) is explicitly dependent on the energy of the confined state. However, in order to simplify the fitting, and avoid unnecessary degrees of freedom, we instead implicitly include the energy dependence through different phases for each quantum number, \( \delta = \delta_n \). With this, the quantization condition reduces to,

\[ k_z^n = \frac{2\pi n - \delta_n}{2mc}, \]

from which the quantized TB dispersion, \( E_n(k_x, k_y, k_z^n) \), may be evaluated.

Full quantization parameters

For each SL, four parameters were fitted to describe the “intrinsic” band structure, and \( n \) parameters described the confined bands. The quantized TB dispersion was fitted to the FP-LAPW ELK band structure of the SLs. The four intrinsic parameters consist of band centers (\( E_0^n \) in Eqn. 1) and band widths for the \( xy \) and \( xz(yz) \) bands. The band width parameter, \( W_i \), is a multiplicative factor to the hopping terms, \( t_i \) (the hopping terms themselves were fixed to the cubic bulk parameters determined above, effectively fixing the shape of the band). In addition to the intrinsic parameters, the phase shifts for each confined state, \( \delta_n \), were also fitted. The fitted phases are shown in Fig. S5 against the mean energy of each state, and closely follow the same roughly linear relationship with energy for all SLs.

The results of fitting the FP-LAPW bands to the quantized TB model are shown in Table I, separated into contributions from the underlying bulk “intrinsic” bands and after quantizing these bands. An example of the fitted band structure is shown in Fig. 1 of the main paper for the 2:7 SL. Since its wavefunction is perpendicular to the quantization axis, the \( xy \) bandwidth is hardly affected by confinement, but the \( xz(yz) \) bands are significantly narrowed compared with their intrinsic (bulk-like) counterparts. The confinement leads to the preferential filling of the quantized \( xz(yz) \) out-of-plane bands as their \( k_z \) dispersion is suppressed and they become 1D-like, which also pulls the Fermi level down slightly.

We note that confinement alone is capable of reproducing the SL band structure to a large extent, correctly describing the narrowing of the quantized bandwidth and its variation with SrVO$_3$ layer thickness. This has been checked by restricting the “intrinsic” bands in the fit to the bulk bands (i.e. setting \( E_0^n \) and \( W_i \) to the bulk ones). This provides additional support that the band narrowing that eventually drives the MIT is primarily due to quantization effects rather than crystal field (CF) effects.

**DYNAMICAL MEAN-FIELD THEORY CALCULATIONS**

The output from the ELK DFT calculation was imported to the TRIQS library [7] via an in-house interface with the dmftproj [8] application. As in the literature [9–11], only the \( V \ t_{2g} \) bands were projected (using Wannier projectors) [12] to construct the LDA Hamiltonian in Wannier space. These projectors were constructed in the following correlated energy

| SL | CF splitting (meV) | Intrinsic \( W_{xy} \) anis. | Quantized bands \( W_{xy} \) anis. |
|----|-------------------|--------------------------|-------------------------------|
| 6:5 | 33 | 0.966 0.959 0.993 | 0.950 0.900 0.948 |
| 3:6 | 40 | 0.971 0.960 0.989 | 0.935 0.798 0.853 |
| 2:7 | 51 | 0.963 0.952 0.988 | 0.911 0.713 0.782 |

**TABLE I.** Results of fitting the FP-LAPW ELK bands to a quantized tight-binding model. The crystal field (CF) splitting is the energy difference, \( E_0^n - E_0^{0} \). The bandwidth (relative to bulk SrVO$_3$), \( W_i \), of the \( xy \) and \( yz \) bands are shown for both intrinsic bands (before quantization) and for the quantized bands, alongside their anisotropy \( (W_{yz}/W_{xy}) \).
FIG. S6. The effect of $U$ on the orbital charge $n_e$ (top), quasiparticle residue $Z$ (middle) and the spectral function around the Fermi level (bottom) for each SL one-shot and fully charge self-consistent (FCSC) DFT+DMFT calculation. The dashed line represents the bulk degenerate orbital charge.

windows: 2:7 SL: $[-1.36, 2.0]$ eV, 3:6 SL: $[-1.29, 2.0]$ eV, 6:5 SL: $[-1.29, 2.0]$ eV and bulk: $[-1.50, 1.90]$ eV. These
windows were constructed such that all of the V t_{2g} bands are included and the valence charge above the lower bound, corresponding to the charge in the V t_{2g} orbitals, is equal to 1 per V impurity. Each DMFT cycle calculation used $84 \times 10^6$ Monte Carlo sweeps.

In order to avoid potential complications from the ill-posed problem of analytic continuation, quantities were determined from the Green’s function and self-energy on the imaginary time ($\tau$) or frequency axis as much as possible. The charge of each orbital ($n_e$) was determined by,

$$n_e = \sum_n G(i\omega_n)e^{i\omega_n0^+}$$  \hspace{1cm} (4)

within the TRIQS library. As there is negligible inter orbital-orbital overlap on the impurity, $n_e$ is diagonal. The spectral function at the Fermi level, $A(\omega = 0)$, is an averaged quantity over a frequency window approximately equal to $T$ [9]. Here, $A(\omega = 0)$ is determined directly from the imaginary time Green’s function by,

$$A(\omega = 0) = \frac{\beta G(\tau = \frac{1}{2}\beta)}{\pi},$$  \hspace{1cm} (5)

where $\beta$ is the inverse temperature in natural units. The value of QP residue $Z$ was determined by

$$Z = \left(1 - \frac{\partial \text{Im}[\Sigma(i\omega_n)]}{\partial i\omega_n} \bigg|_{i\omega_n \to 0^+}\right)^{-1},$$  \hspace{1cm} (6)

where the $Z$ is evaluated from the differential of the imaginary part of the Matsubara self-energy at $i\omega_n \to 0^+$. For $U$ values far from the Fermi liquid regime (namely for the 6:5 $Z$ values close to the MIT), the $Z$ values were approximated by using the differential of the interpolated self-energies at $i\omega_n = 0$.

There are two ways to realize the insulating solution. First, by a divergence in $\text{Im}[\Sigma(i\omega_n)]$, which comes naturally with $Z = 0$. Second, the combination of the $\text{Re}[\Sigma(i\omega_n)]$ and the chemical potential might move the pole position outside of the non-interacting bandwidth, meaning that no QP peak is possible in the Green’s function. In the latter case, we have $A(\omega = 0)$ vanishing with non-diverging $\text{Im}[\Sigma(i\omega_n)]$. In that case, that we also see here, we set $Z$ to zero manually. From this, the MIT $U$ value ($U_{\text{MIT}}$) is defined as the lowest $U$ value in which $A(\omega = 0) = 0$.

The spectral functions, $A(\omega)$, for each impurity were calculated from $G(\tau)$ using the LineFitAnalyzer technique of the maximum entropy analytic continuation method implemented within the MaxEnt application of TRIQS [13]. The k-resolved spectral functions $A(k,\omega)$ were calculated from the analytically continued self-energy.

The effective and correlation subband mass enhancement factors, $1/Z_{\nu}$ and $1/Z_{\nu}'$, were calculated from the ratios of the Fermi velocities using

$$Z_{\nu} = \frac{v_F^\nu}{v_{QTB}^\nu},$$  \hspace{1cm} (7)

and

$$Z_{\nu}' = \frac{v_{F,\nu}^c}{v_F^c}.$$  \hspace{1cm} (8)

Here, the Fermi velocities were determined from the gradient of the linearly expanded band dispersions along M-X around $k_F$ of the DFT+DMFT subbands ($v_F^\nu$), the quantized bands from QTB ($v_{QTB}^\nu$) and the intrinsic (bulk-like) TB bands ($v_{F,\nu}^c$). The intrinsic bands were used as they incorporate the effect of renormalization due to strain. Therefore, $Z_{\nu}^c$ and $Z_{\nu}$ describe the effect of renormalization from correlations and the combination of correlations and confinement (band) effects respectively.

The DFT+DMFT subband energy centers, $E_{\nu,k}$, were calculated by using

$$E_{\nu,k} = \epsilon_{\nu,k} - \mu + \text{Re}[\Sigma_{\nu}(k,\omega = E_{\nu,k})],$$  \hspace{1cm} (9)

where $\epsilon_{\nu,k}$ is the DFT energy, $\mu$ is the chemical potential and $\text{Re}[\Sigma_{\nu}(k,\omega)]$ is the real part of the diagonal unfolded self-energy elements on the real frequency axis. The QP lifetime in the inset of Fig. 4 of the main manuscript was determined from the inverse imaginary part of the analytically continued unfolded self-energy. Finally, the subband energies at the $\Gamma$ high symmetry point in Fig. 4 of the main manuscript were determined from Eqn. 9.

FIG. S7. Spectral functions of the 2:7 (top), 3:6 (middle) and 6:5 (bottom) SLs from fully charge self-consistent calculations, showing $xy$ (left) and $xz(yz)$ (right) orbitals. The spectra of the insulating 2:7 SL have been shifted such that the Fermi level lies at the center of the band gap of the $xy$ spectrum.
One-shot and FCSC DFT+DMFT results

The main manuscript presents fully charge self-consistent (FCSC) DFT+DMFT calculations. Here, we present the results of one-shot (OS) DFT+DMFT for comparison. Overall, charge self-consistency slightly adjusts some details of the results, but the main conclusions of our study are already present in one-shot calculations.

Figure S6 shows the $U$-dependent MIT for each SL and different DFT+DMFT methods. The behavior of the OS and FCSC calculations is very similar, exhibiting a similar $U_{\text{MIT}}$ with similar characteristics, e.g. $A(\omega = 0)$ and $Z$. Some differences are observed in the orbital polarization between the two methods, whereby the polarization is somewhat suppressed in the FCSC calculation compared with OS. This behavior, most notable for the 2:7 SL, is consistent with other studies [10, 11, 14], and is caused by the charge redistribution with the rest of the system at the DFT stage. This trend from 2:7 to bulk is also seen in Fig. S9 for the orbitally-averaged values of $Z$, where there are some differences in $\bar{Z}$ for the 2:7 SL between OS and FCSC, but the bulk values are very similar.

An important note to make about Fig. S6 is that $Z$ at the interface (imp 1) for the $xz(yz)$ orbitals tends to zero first for each SL. This suggests that the weight from the $xz(yz)$ QP peak depletes first. Therefore, when the interface $xz(yz)$ QP state has been fully depleted, this causes the SL to transition into the insulating state. From this, the interface between the oxides has a strong influence on the MIT. The $A(\omega = 0)$ for imp 1 of the 6:5 also tends to zero which strengthens the argument for at least that SL.

FIG. S8. The averaged orbital charge over all layers from the DFT and DFT+DMFT Wannier $V_{xy}$ and $xz(yz)$ orbitals for each SL and bulk. This includes charges from DFT, DFT+DMFT and average orbital charge per layer (orb. ave.) for each SL and bulk.

FIG. S9. The comparison of the orbitally-averaged quasiparticle residue, $Z$, between the one-shot (OS) and fully charge self-consistent (FCSC) DFT+DMFT methods. The plot lines are guides to the eye.

FIG. S10. The orbital $A(\omega)$ comparisons between the one shot and fully charge self consistent (FCSC) DFT+DMFT methods from different input DFT codes for mono layer SrVO$_3$.

OTHER SIGNIFICANT SL FCSC RESULTS

Fig. S7 shows the $A(\omega)$ of each correlated impurity orbital in each SL at $U = 5.7$ eV (the value used in the theoretical-experimental comparisons). It is evident that the 2:7 is insulating and the 3:6 and 6:5 is metallic from the absence/presence of the QP peak at the Fermi level. There are sharp features in the QP peaks around the Fermi level for the 3:6 and 6:5 SLs. These features are often attributed to spurious noise from the analytic continuation procedure, however, it may not be the case here due to the quantized bands being present around the Fermi level. The peak position of the Hubbard bands (notably the UHB) are closer in energy to the Fermi level. These features are often attributed to spurious noise from the analytic continuation procedure, however, it may not be the case here due to the quantized bands being present around the Fermi level. The peak position of the Hubbard bands (notably the UHB) are closer in energy to the Fermi level for the 3:6 and 6:5 SLs. This is another indication that the interface layer is more correlated compared to the other layers.

The splitting of the orbital degeneracy strongly affects the polarization of the orbital charge as shown in Fig. S8. It is interesting to note that the reduction of the number of layers sig-
significantly increases the charge in the xz(yz) orbitals, which appear to tend to half filling (whereas the xy orbitals are tending towards zero charge). This is a likely consequence of these orbitals trying to reduce the potential energy, analogous to what is seen in the previous mono layer calculations. The reduction in the orbitally-averaged DFT charge with lower SrVO$_3$ layers is likely due to hybridization with Ti at the interface.

**Elk-TRIQS interface test: monolayer SrVO$_3$**

The results presented used an in-house interface between Elk and TRIQS, so this section presents comparison results between Elk and Wien2k inputs into TRIQS to show that the interface works for a similar system, namely monolayer SrVO$_3$. The monolayer SrVO$_3$ calculation was set up in the same way and using the same parameters as in Ref. 11. Figure S10 shows the comparison between $A(\omega)$ calculated from the different DFT code inputs. This comparison shows excellent agreement between the different inputs for the different DFT+DMFT methods. This test shows that the interface used between Elk and TRIQS is able to reliably perform DFT+DMFT calculations.

**EFFECTS OF STRAIN**

We performed volume conserving strain calculations on bulk SrVO$_3$ to investigate the effect of CF has on the MIT while the bandwidths of the $t_{2g}$ orbitals are approximately unchanged. Compressive strain of 1% was applied along the $c$-axis; the other axes were tensively strained to conserve volume compared with the bulk. This strain was chosen to yield a CF splitting of 53 meV, slightly larger than but comparable to the CF splitting of the 2:7 SL. The strained FCSC $U_{\text{MIT}}$ is approximately 6.525 eV, the same as for the bulk. The OS strained calculation had a slightly lower $U_{\text{MIT}}$ of 6.475 eV. Due to the small change on $U_{\text{MIT}}$, the CF splitting is insufficient to cause the MIT in these SLs.

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