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Rutile IrO$_2$/TiO$_2$ superlattices: a hyper-connected analog to the Ruddelsden-Popper structure

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Dimensionality and connectivity among octahedra play important roles in determining the properties, electronic structure, and phase transitions of transition metal oxides. Here we demonstrate the epitaxial growth of (110)-oriented alternating layers of IrO$_2$ and TiO$_2$, both of which have the rutile structure. These (IrO$_2$)$_n$/TiO$_2$$_{2n}$ superlattices consist of IrO$_6$ and TiO$_6$ octahedra tiled in a hyper-connected, edge- and corner-sharing network. Despite the large lattice mismatch between constituent layers ($\Delta d_0 = -2.1\%$ and $\Delta c = +6.6\%$), our reactive molecular-beam epitaxy grown superlattices show high structural quality as determined by x-ray diffraction and sharp interfaces as observed by transmission electron microscopy. The large strain at the interface is accommodated by an ordered interfacial reconstruction. The superlattices show persistent metallic down to $n = 3$ atomic layers, and angle resolved photoemission spectroscopy measurements reveal quantized sub-bands with signatures of IrO$_2$-IrO$_2$ interlayer coupling.

Transition metal oxide superlattices enable the discovery of emergent properties at interfaces and precise manipulation of magnetic$_1$, electronic$_2,3$, and ferroic properties$_4,5$. To date, efforts have focused primarily on superlattices in the perovskite (AMO$_3$/A'MO$_3$)$_n$$_^{1,4-6}$ and related naturally layered Ruddelsden-Popper ($A_{n+1}M_nO_{3n+1}$)$_n$$_^{7-10}$ structures, which consist of MO$_6$ octahedra tiled in all corner-sharing networks. In such superlattices the primary tuning knobs are the chemistry of the transition metal cations $M$ ($M'$), the layer thickness, and epitaxial strain.

More drastic properties changes can, however, be made by altering the connectivity (topology) of the MO$_6$ octahedral network. The rutile polymorph (composition MO$_2$) is one such example, which consists of a mixed edge- and corner-sharing network (Fig. 1). This hyperconnectivity enhances direct $M - M$ interactions along the [001]$_r$ edge-sharing direction ($r$ denotes the conventional rutile unit cell), in contrast with the primarily oxygen mediated $M - O - M$ interactions in corner-sharing perovskites and Ruddelsden-Poppers, and is a key driver in determining the relationships between structure and electronic properties in these materials. For example, the metal-insulator transition in VO$_2$ is driven by direct V-V dimerization in a Mott-Peierls like mechanism$^{11-13}$. The strength of $M - M$ interactions and octahedral connectivity can also tune the effective strength of electron-electron correlations$^{14}$. Finally, the rutile structure belongs to a nonsymmorphic space group, an important symmetry for stabilizing topological states and the spin Hall effect$^{15-18}$. Together, these factors point to octahedral connectivity as a potentially new and important tuning parameter for controlling oxide superlattices. Unfortunately, the combination of large lattice mismatches between different MO$_2$ compounds and high surface energies has made the growth of all-rutile superlattices challenging$^{19,20}$.

Here we demonstrate the epitaxial growth and characterization of the atomic and electronic structure of all-rutile IrO$_2$/TiO$_2$ superlattices. The (110)$_r$, where the $r$ subscript refers to the conventional unit cell of the rutile polymorph, was chosen for the plane of the surface due to its low surface energy to facilitate smooth growth, as opposed to the high surface energy (001)$_r$, orientation$^{21}$. FIG. 1. Comparison between (001)$_r$-oriented Ruddelsden-Popper and perovskite superlattices, with (110)$_r$-oriented rutile superlattices. Transition metal cations $M$ are located at the center of each octahedra, and oxygen is at the corners. Both Ruddelsden-Popper and perovskite superlattices are characterized by four oxygen-mediated $M - O - M$ nearest neighbor hoppings in the (001)$_r$ plane (arrows). In contrast, the rutile structure is characterized by chains of edge-sharing octahedra oriented along [001]$_r$ (into the page). In the (110)$_r$ plane it has four $M - O - M$ hoppings, plus direct $M - M$ hopping along [001]$_r$. Dimensionality and connectivity among octahedra play important roles in determining the properties, electronic structure, and phase transitions of transition metal oxides. Here we demonstrate the epitaxial growth of (110)$_r$-oriented alternating layers of IrO$_2$ and TiO$_2$, both of which have the rutile structure. These (IrO$_2$)$_n$/TiO$_2$$_{2n}$ superlattices consist of IrO$_6$ and TiO$_6$ octahedra tiled in a hyper-connected, edge- and corner-sharing network. Despite the large lattice mismatch between constituent layers ($\Delta d_0 = -2.1\%$ and $\Delta c = +6.6\%$), our reactive molecular-beam epitaxy grown superlattices show high structural quality as determined by x-ray diffraction and sharp interfaces as observed by transmission electron microscopy. The large strain at the interface is accommodated by an ordered interfacial reconstruction. The superlattices show persistent metallic down to $n = 3$ atomic layers, and angle resolved photoemission spectroscopy measurements reveal quantized sub-bands with signatures of IrO$_2$-IrO$_2$ interlayer coupling.

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Here we demonstrate the epitaxial growth and characterization of the atomic and electronic structure of all-rutile IrO$_2$/TiO$_2$ superlattices. The (110)$_r$, where the $r$ subscript refers to the conventional unit cell of the rutile polymorph, was chosen for the plane of the surface due to its low surface energy to facilitate smooth growth, as opposed to the high surface energy (001)$_r$, orientation$^{21}$.
FIG. 2. Growth of (IrO$_2$)$_n$/TiO$_2$ superlattices. (a) Crystal structure of an $n = 5$ superlattice. We define $d_\perp \parallel [110]_r$ as the average out-of-plane atomic spacing, and $d_\parallel \parallel [1\bar{1}0]_r$ and $c \parallel [001]_r$ as the in-plane spacings. (b) RHEED intensity oscillations of the (0,1) spot (upper black curve). The lower red curve shows modulations of the in-plane $c$ lattice spacing, as extracted from horizontal line cuts of the RHEED pattern. (c,d) RHEED patterns for IrO$_2$ and TiO$_2$ along the $[1\bar{1}0]_r$ azimuth. (e) LEED pattern of an IrO$_2$-terminated superlattice at 100 eV. (f) In-plane crystal structure the top layer of IrO$_2$ as viewed down the $[110]_r$ direction.

We choose IrO$_2$ as the electronically active layer due to the delicate interplay of spin-orbit coupling and electron-electron correlations in the iridates, which lead to spin-orbit Mott physics$^{22}$, proposed topological states$^{8,23}$, and possible novel superconductivity in the two dimensional limit$^{24–26}$. More specifically, rutile IrO$_2$ exhibits a large Spin Hall effect$^{27}$ and unusual transport properties in which the sign of the apparent carrier can be tuned via the orientation of the magnetic field$^{28}$. In the rutile polymorph of IrO$_2$, the Ir$^{4+}$ cations have an electron configuration of 5$d^5$, the same as its perovskite and Ruddelsden-Popper cousins SrIrO$_3$ and Sr$_{n+1}$Ir$_n$O$_{3n+1}$. The rutile TiO$_2$, which has configuration 3$d^0$, was chosen as the insulating barrier layer due to its large bandgap. Hence, our all-rutile IrO$_2$/TiO$_2$ (5$d^5$/3$d^0$) superlattices can be viewed as a hyper-connected analogue of (SrIrO$_3$)$_n$/(SrTiO$_3$)$_m$ perovskite and Sr$_{n+1}$Ir$_n$O$_{3n+1}$ Ruddelsden-Popper superlattices. The latter two have exhibited metal-insulator transitions and the onset of magnetic ordering near $n = 3$ perovskite layers$^{6,9}$. Similarly, recent first principles calculations for (001)-oriented rutile IrO$_2$/TiO$_2$ superlattices suggest metal-insulator transitions and magnetic ordering near $n = 2 – 3^{29}$.

(IrO$_2$)$_n$/TiO$_2$ superlattices with periodicity $n = 3$, 4, and 5 atomic layers were grown by reactive oxide MBE on HF-etched (110)-oriented rutile TiO$_2$ substrates. Growth was performed at 350°C as calibrated by a pyrometer, using a background partial pressure of $10^{-6}$ Torr of distilled ozone as the oxidant. The superlattice structure was defined by alternating shutter-controlled doses of Ir and Ti, each containing the number of atoms in a single monolayer of IrO$_2$ or TiO$_2$. These fluxes were as supplied from an electron beam evaporator and a Ti-ball, respectively$^{30}$. Film growth was characterized in real-time by reflection high-energy electron diffraction (RHEED). After growth, samples were transferred through an ultrahigh vacuum manifold ($p < 3 \times 10^{-10}$ Torr) and characterized by in-situ low-energy electron diffraction (LEED), and in-situ angle resolved photoe-
mission spectroscopy (ARPES) at 20 K using the Iα emission line from a monochromated He lamp ($h\nu = 21.2$ eV) and Scienta R4000 analyzer. Samples were also characterized ex-situ by high-resolution x-ray diffraction (XRD, with Cu Kα radiation), magnetotransport (Quantum Design PPMS), and scanning transmission electron microscopy (STEM).

The shutter timing and RHEED response of our growth method is illustrated in Fig. 2. During growth we observe clear periodic modulations in the RHEED intensity (Fig. 2(b)), where each oscillation corresponds to one IrO$_2$ (TiO$_2$) monolayer (ML). The oscillation periods are in good agreement with flux measurements by a quartz crystal microbalance (QCM) and XRD Kiessig fringes on calibration samples. The corresponding RHEED patterns for the IrO$_2$ and TiO$_2$ are characterized by a sharp arc of spots (Figs. 2(c) and 2(d)), indicating smooth epitaxial growth. The epitaxial alignment is further confirmed by the sharp spots in the final LEED pattern (Fig. 2(e)).

During the first 2 monolayers of IrO$_2$ growth on TiO$_2$, we observe a decaying envelope in the RHEED intensity, with the oscillation amplitude recovering in the third and subsequent layers. Interestingly there is no such decaying envelope for the opposite growth sequence, TiO$_2$ on IrO$_2$. We interpret this asymmetry to result from differences in wetting: first principles calculations suggest that the (110) surface energy for TiO$_2$ (0.9 J/m$^{221}$) is smaller than that of IrO$_2$ (1.4 J/m$^{231}$) implying a greater tendency for TiO$_2$ to wet IrO$_2$ than vice-versa.

We also observe periodic modulations of the in-plane $c \parallel [001]$, lattice spacing during growth, for which there
is a large mismatch of 6.6%. The red curve in Fig. 2(b) plots the changes in c at the surface, as extracted from Gaussian fitted horizontal line cuts of the RHEED intensity (Figs. 2(c) and 2(d)). We find that c expands ($Q_y$ decreases) during growth of IrO$_2$ and contracts ($Q_y$ increases) during growth of TiO$_2$. The magnitude of change from layer to layer is 1.2% and there is also an envelope of increasing c as the growth persists, indicating partial relaxation along the c axis. Along the other in-plane direction, [110]$_c$ ($d_{||}$), for which the mismatch is smaller (-2.1%), we do not observe changes in the Q spacing within a resolution of $\Delta Q/Q \approx 0.3\%$, indicating that $d_{||}$ remains commensurate to the substrate.

The resulting superlattices show high structural quality as measured by x-ray diffraction. This is quite remarkable given the large mismatch between the IrO$_2$ and TiO$_2$ constituent layers ($\Delta d_{\perp} = -2.1\%$ and $\Delta c = +6.6\%$). Figures 3(a) and 3(b) show symmetric $\theta - 2\theta$ scans for $n = 3, 4$ and 5 superlattices (16, 12, and 24 repeat units, respectively). We observe all of the expected superlattice reflections out to $2\theta = 100^\circ$ as well as Kiessig finite thickness fringes indicating sharp interfaces (Figs. 3(a) and 3(b)). Rocking curves are similarly sharp, with full width at half maxima (FWHM) of 23, 21, and 28 arc sec for the $n = 3, 4$, and 5 samples, respectively, comparable to the substrate, indicating high structural quality despite the large lattice mismatch between IrO$_2$ and TiO$_2$. These rocking curve widths are comparable to MBE-grown perovskite and Ruddlesden-Popper superlattices. The in-plane epitaxial alignment to TiO$_2$ (110) is confirmed by $\phi$ scans of the film 222, and 400$_r$ reflections, which show two-fold rotation and no in-plane rotation twin variants.

In Fig. 3(e) we plot the measured in-plane (red) and average out-of-plane (black) lattice spacings for the $n = 3, 4$, and 5 superlattices and for a 50 monolayer thick IrO$_2$ film. The average out-of-plane spacing $d_{\perp}$, determined from Nelson-Riley extrapolations of the on-axis $2\theta$ scans, decreases with monotonically with $n$ as expected from Vegard’s Law. The in-plane lattice spacings $d_{||}$ and $c$ were extracted from projections of the off-axis 400$_r$ and 222$_r$ reflections, respectively. We find that while $d_{\perp}$ remains commensurate to the substrate, $c$ partially relaxes. Both trends in $c$ and $d_{||}$ are in agreement with the lattice parameters extracted from RHEED (Fig. 2(b)).

To further explore the interface structure and partial relaxation we perform cross-sectional annular dark field (ADF) STEM measurements of the $n = 4$ sample (Fig. 4). Here, due to the higher atomic mass of Ir, columns of Ir atoms appear as bright regions while columns of Ti appear as the darker regions. The frame averaged line cut along the [110]$_r$ growth direction confirms the stacking sequence of 4 monolayers of IrO$_2$ and 2 monolayers of TiO$_2$.

Higher resolution imaging of the IrO$_2$ on TiO$_2$ interface (insert) and layer-resolved horizontal line cuts (bottom) reveal a periodic modulation of the STEM intensity in the top-most TiO$_2$ layer (blue curve), with periodicity twice that of the atomic [001]$_r$ spacing. No obvious superstructure appears in the bottom most IrO$_2$ layer (red curve). We interpret this modulation to result from an interfacial reconstruction that partially relieves stresses at the IrO$_2$/TiO$_2$ interface due to the large lattice mismatch. Such a reconstruction may explain why well-defined IrO$_2$/TiO$_2$ superlattices are able to form, despite the mismatch. This interfacial reconstruction may also be responsible for the decay in the RHEED intensity during the initial growth of IrO$_2$ on TiO$_2$ (Fig. 2(b)).

Temperature dependent transport measurements reveal that all superlattices remain metallic ($\partial\rho/\partial T > 0$) (Fig. 5(a)) down to $n = 3$ layers. For the $n = 4$ and $n = 5$ superlattices, the zero-field resistivity follows a temperature squared dependence from 2 K to 300 K, consistent with Fermi liquid behavior. For $n = 3$ there is a small upturn in the low-temperature resistivity, which we attribute to weak antilocalization in the two-dimensional limit, but the overall temperature dependence indicates metallic behavior.

The retained metallicity of our rutile superlattices is quite notable, as in purely corner-sharing (SrIrO$_3$)$_n$/SrTiO$_3$ perovskite superlattices, the $n = 3$
member is insulating. For \(\mathrm{Sr}_{n+1}\mathrm{Ir}_n\mathrm{O}_{3n+1}\) Ruddelsden-Popper superlattices, a metal-insulator transition is expected in the vicinity of \(n = 2 - 3\) atomic layers.\(^8\)\(^9\) The retained metallicity for our rutile superlattices results from the higher degree of octahedral connectivity and wider bandwidths in the rutile structure, as discussed in recent ARPES studies of \(\mathrm{IrO}_2\).\(^{14}\)

Magnetoresistance measurements at 2 K confirm the weak antilocalization (Fig. 5(b), insert), as evidenced by a narrow local minimum in the magnetoresistance superimposed on a broader maximum at zero field. Fitting to a Hikami-Larkin-Nagaoka model\(^{33}\) we extract a magnetic dephasing length of approximately 100 nm. For the thicker \(n = 4\) and 5 superlattices we observe a linear dependence of the magnetoresistance on field, consistent with a low density semimetal.

ARPES measurements on the superlattices reveal quantized sub-bands with signatures of interlayer \(\mathrm{IrO}_2/\mathrm{IrO}_2\) coupling. In Fig. 6 we compare measurements for an \(n = 4\) superlattice (\(\mathrm{IrO}_2\)-terminated, \((\mathrm{IrO}_2)_4/(\mathrm{TiO}_2)_2\) x12), with that of a 4 ML ultrathin \(\mathrm{IrO}_2\) film and a bulk-like, 50 ML thick \(\mathrm{IrO}_2\) film. All three structures are characterized by a broad band of oxygen 2\(\rho\) states from binding energies of 10 to 3 eV, and a band of iridium 5\(d\) states extending from 3 eV to the Fermi energy (Fig. 6(h)), in agreement with previous ARPES and density functional theory studies of bulk \(\mathrm{IrO}_2\).\(^{14}\) Zooming in to the Fermi level (Fig. 6(a-c)), the bulk-like sample is characterized by two highly dispersive bands (bandwidth greater than 1.5 eV) that are well described by density functional theory calculations (solid curves)\(^{14}\). For the 4 ML thin film, we observe additional electron-like bands that we attribute to metallic quantum well states, resulting from strong spatial confinement in the out-of-plane direction, which cross through the Fermi energy. These sub-bands are highly reminiscent of the sharp quantum well sub-bands observed in ultrathin films of \(\mathrm{SrVO}_3\) with perovskite structure.\(^{35}\) We emphasize two important differences between our \(\mathrm{IrO}_2/\mathrm{TiO}_2\) superlattices and the \(\mathrm{SrVO}_3\) ultrathin films: (1) our \(\mathrm{IrO}_2/\mathrm{TiO}_2\) crystallize in the rutile structure as opposed to the perovskite structure of \(\mathrm{SrVO}_3\), and (2) our \(\mathrm{IrO}_2/\mathrm{TiO}_2\) samples are in superlattice form with a finite number of repeats, as opposed to a singly layered \(\mathrm{SrVO}_3\) thin films, and are thus expected to be more robust to external perturbations such as surface contamination.

Comparing the 4 ML ultrathin film to the \(n = 4\) superlattice, we also observe three occupied sub-bands; however, the sub-bands in the superlattice differ from that of the ultrathin film in two respects: (1) their binding energies are shallower (shifted upwards in energy towards the Fermi energy by \(\sim 100\) meV) and (2) they are broadened in energy, as shown in the energy dispersion curves (EDCs, Fig. 6(d)). We interpret these differences to arise from \(\mathrm{IrO}_2/\mathrm{IrO}_2\) interlayer coupling, in which tunneling across the \(\mathrm{TiO}_2\) barriers causes the quantized states residing within each \(\mathrm{IrO}_2\) quantum well to hybridize with states in neighboring quantum wells, resulting in a finite out-of-plane dispersion. The \(\mathrm{TiO}_2\) barrier thickness provides a degree of freedom for tuning the electronic structure and the effective \(k_z\) dispersion of the superlattice sub-bands, that does not exist for an ultrathin film. The sharpness of the barrier, i.e., the sharpness of the \(\mathrm{IrO}_2/\mathrm{TiO}_2\) interfaces, as well its structure, e.g. an ordered interfacial reconstruction, are expected to be additional parameters for tuning the sub-band dispersions. The full thickness dependent evolution of the quantum well states for ultrathin films is the subject of a concurrent publication.\(^{34}\) In addition to the quantum well states, in some samples we also observe subtle kinks in the dispersion at characteristic energy of approximately 80 meV, comparable to the Debye temperature of \(\mathrm{IrO}_2\) (700K \(\approx 60\) meV\(^{36}\)). Therefor we speculate the kink may arise from electron-phonon coupling.

In summary, we have demonstrated the epitaxial growth of all-rutile \(\mathrm{IrO}_2/\mathrm{TiO}_2\) (110), superlattices by reactive oxide MBE. This layered rutile structure can be viewed as a hyper-connected, non-symmorphic alternative to the perovskite and Ruddelsden-Popper oxide structures that are more commonly studied. Our superlattices show high structural quality as determined by x-ray diffraction, with all of the expected superlattice reflections out to \(2\theta = 100^\circ\), and rocking curve widths comparable to the best perovskite and Ruddelsden-Popper superlattices. This is quite remarkable given the large mismatch between the \(\mathrm{IrO}_2\) and \(\mathrm{TiO}_2\) constituent layers (\(\Delta d_j = -2.1\%\) and \(\Delta c = +6.6\%\)). Cross sectional transmission electron microscopy suggests that the large strain is accommodated by an interfacial reconstruction. The higher octahedral connectivity in the rutile structure provides a greater tendency towards retained metallicity, as compared to the insulating behavior in perovskite and Ruddelsden-Popper iridate superlattices. ARPES measurements reveal quant-
FIG. 6. ARPES measurements of (a) a 4 ML ultrathin IrO$_2$ film, (b) the $n = 4$ superlattice (IrO$_2$-terminated, [(IrO$_2$)$_4$/(TiO$_2$)$_2$] x12), and (c) a bulk-like 50 ML thick film, using He I$\alpha$ photons ($h\nu = 21.2$ eV). The dispersion is through the zone center $(0,0)$, $k_x \parallel [1\bar{1}0]$. All samples were grown on TiO$_2$ (110). The left hand side shows the raw photoemission intensity (color scale) and peak positions from fitting of the energy and momentum distribution curves (EDC/MDC, circles). The right hand side shows the second derivative of the intensity $-\partial^2 I/\partial E^2$. Solid curves in (a) and (b) are guides to the eye, curves in (c) are the calculated dispersion from density functional theory, using the generalized gradient approximation and including spin-orbit coupling (GGA+SO)$^{14}$. (d) Energy dispersion curve (EDC) at $k_0 = (0,0)$, showing a shift in the quantum well binding energies (arrows) and energy broadening for the superlattice. (e-g) Cartoon structures for the ultrathin film, superlattice, and bulk-like samples. (h) Angle integrated measurements of the valence bands using He II$\alpha$ photons ($h\nu = 40.8$ eV).

tum confined, metallic sub-bands with interlayer coupling. Due to the highly anisotropic crystal structure of rutile, with chains of edge-sharing octahedra oriented along [001], we expect the electronic properties of rutile superlattices to be highly dependent on orientation. For example, while our (110)$_r$, oriented superlattices exhibit retained metallicity, (001)$_r$ oriented superlattices to be more likely to yield metal-insulator transitions, as interfaces in this orientation break the continuity of edge-sharing IrO$_6$ chains$^{29}$. Realizing high quality superlattices in the high surface energy (001)$_r$ orientation, however, remains an outstanding challenge$^{19,20}$. This interplay of anisotropy, topology, octahedral connectivity, and reduced dimensionality in the rutile structure presents a new platform for tuning the properties of oxide superlattices.

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