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Correlation-driven eightfold magnetic anisotropy in a two-dimensional oxide monolayer

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Engineering magnetic anisotropy in two-dimensional systems has enormous scientific and technological implications. The uniaxial anisotropy universally exhibited by two-dimensional magnets has only two stable spin directions, demanding 180° spin switching between states. We demonstrate a previously unobserved eightfold anisotropy in magnetic SrRuO3 monolayers by inducing a spin reorientation in (SrRuO3)/(SrTiO3)n superlattices, in which the magnetic easy axis of Ru spins is transformed from uniaxial ⟨001⟩ direction (N < 3) to eightfold ⟨111⟩ directions (N ≥ 3). This eightfold anisotropy enables 71° and 109° spin switching in SrRuO3 monolayers, analogous to 71° and 109° polarization switching in ferroelectric BiFeO3. First-principle calculations reveal that increasing the SrTiO3 layer thickness induces an emergent correlation-driven orbital ordering, tuning spin-orbit interactions and reorienting the SrRuO3 monolayer easy axis. Our work demonstrates that correlation effects can be exploited to substantially change spin-orbit interactions, stabilizing unprecedented properties in two-dimensional magnets and opening rich opportunities for low-power, multistate device applications.

RESULTS

Structural characterizations of (SrRuO3)1/(SrTiO3)n superlattices

The (SrRuO3)1/(SrTiO3)n superlattices are shown schematically in Fig. 1A and were fabricated by pulsed laser deposition (PLD) assisted with reflective high-energy electron diffraction (RHEED). X-ray diffraction (XRD) measurements in Fig. 1B reveal superlattice peaks corresponding to the designed periodicity. Layer-by-layer growth and atomically flat surfaces are observed by in situ RHEED and atomic force microscopy, respectively (fig. S1). The x-ray absorption near-edge structures (XANES) of Ru K-edges are measured, which demonstrate similar Ru valences in the superlattices (fig. S2). XRD reciprocal space maps around the (2 0 4) substrate peak are shown in Fig. 1C, demonstrating that all superlattices are coherently strained to the SrTiO3 substrates. The average z-axis lattice constants are calibrated and shown in Fig. 1D. The ideal z-axis lattice constants calculated as \( c_{\text{ideal}} = (N \times c_{\text{STO}} + c_{\text{SRO}})/(N + 1) \) are used to fit \( c_{\text{average}} \), where \( c_{\text{STO}} \) and \( c_{\text{SRO}} \) represent that of SrTiO3 (3.905 Å) and SrRuO3 (3.984 Å), respectively. This comparison shows that, within experimental uncertainty, \( c_{\text{average}} \) matches \( c_{\text{ideal}} \) across all
N, so that the lattice constants and strain states of all superlattices are consistent. Furthermore, we measured half-order diffraction peaks to reveal the oxygen octahedral rotation patterns \((19-21)\). In all superlattices, \(a^{-}a^{-}c^{-}\) rotation patterns are observed (see section S1 for details). Figure 1E shows the \((3/2 1/2 3/2)\) and \((3/2 1/2 5/2)\) half-order peaks with stronger intensities in superlattices of smaller \(N\). In addition, extremely weak \((H/2, H/2, L/2)\) diffraction peaks are observed in all superlattices (fig. S3, A and B), indicating that the residual \(a^{-}\) rotation is much smaller than the \(c^{-}\) rotation. We therefore conclude that all superlattices exhibit tetragonal structural symmetry with \(a^{-}a^{-}c^{-}\) type octahedral rotations, where the \(c^{-}\) rotations are larger than the \(a^{-}\) rotations.

Magnetism and Curie temperatures of (SrRuO\(_3\))\(_{1}/(SrTiO_3)\(_N\) superlattices

In Fig. 2 (A and B), we show the Ti \(L\)-edge x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) of the superlattices measured at a temperature \(T\) of 10 K in an applied magnetic field \(H\) of 4 T. As the absorption energy of the Ru \(M\)-edge and Ti \(L\)-edge overlap, the XAS and XMCD are completely dominated by Ti so that it is not possible to distinguish the Ru \(M\)-edge signal in the superlattices (22). More XMCD data with both normal and grazing incidence beam of the other superlattices are shown in fig. S4. No measurable valence change or magnetic dichroism was observed on the Ti edge in all superlattices, excluding any magnetic contribution from the SrTiO\(_3\) and indicating that the magnetization \((M)\) is confined purely within the SrRuO\(_3\) layers. The magnetism and MA of the SrRuO\(_3\) monolayer are further revealed by superconducting quantum interference device (SQUID) magnetometer and magneto-optic Kerr effect (MOKE) measurements. The former detects the overall magnetism from the film and possible artificial backgrounds, while the latter only detects the film with an optical penetration depth (~30 nm) less than the film thickness. Note that the cooling fields to orient the magnetic domains are 0.05 and 0.5 T for the MOKE and SQUID measurements, respectively. The lower cooling field yielded the low-temperature peak features in some of the MOKE measurements. The temperature-dependent Kerr rotation (Fig. 2C, right axis, and fig. S5A) reveals Curie temperatures \((T_C)\) of approximately 100 K for the \(N = 1\) and 70 K for the \(N = 2\) to 5 superlattices. Thus, the magnetic transition can be confirmed to be intrinsic to the films.

The quantitative magnetizations of the superlattices are studied using SQUID \(M\)-versus-\(H\) measurements (Fig. 2, D to F, and fig. S5, B and C), which reveals a saturation magnetization between 0.5 \(\mu_B/\text{Ru}\) and 0.7 \(\mu_B/\text{Ru}\) for the \(N = 1\) and 2 superlattices and approximately 0.4 \(\mu_B/\text{Ru}\) for the \(N \geq 3\) superlattices. Interestingly, the MA of the superlattices exhibits a notable dependence on the SrTiO\(_3\) layer number. The \(N \leq 2\) superlattices are similar to the bulk, remaining uniaxial with the easy axis along [001] (see Fig. 2D and fig. S5B). However, the magnetic hysteresis of the \(N \geq 3\) superlattices indicates the easy axis transition to be along the [111] direction [see Fig. 2 (E and F) and fig. S5C].
Since the Ru M-edge XMCD was not detectable, we have probed the magnetization distribution in the $N = 3$ superlattice with polarized neutron reflectometry (PNR), as shown in Fig. 3A. Although the superlattice repeat length is extremely thin, so that the first-order Bragg reflection appears at approximately 4 nm $- 1$, the extremely sharp interfaces and sample uniformity allow the observation of a clear superlattice peak in the expected location, as shown in Fig. 3B. Because the neutron spin provides sensitivity to the magnetic scattering length densities, analysis and model fitting of the PNR data allow a depth-dependent picture of the magnetization distribution to be extracted, shown in Fig. 3D. Specifically, we note that a nonzero splitting (see Fig. 3C) is observed between the $(++)$ and $(- -)$ reflectivities near the critical edge, which indicates an intrinsic net magnetization within the film of at least $0.24 \mu_B$/Ru and up to $0.37 \mu_B$/Ru. The PNR-detected magnetization is slightly smaller than the SQUID magnetization but agrees reasonably well given possible background contributions to the SQUID value.

**Depth-dependent magnetization distribution**

Since the Ru M-edge XMCD was not detectable, we have probed the magnetization distribution in the $N = 3$ superlattice with polarized neutron reflectometry (PNR), as shown in Fig. 3A. Although the superlattice repeat length is extremely thin, so that the first-order Bragg reflection appears at approximately 4 nm $- 1$, the extremely sharp interfaces and sample uniformity allow the observation of a clear superlattice peak in the expected location, as shown in Fig. 3B. Because the neutron spin provides sensitivity to the magnetic scattering length densities, analysis and model fitting of the PNR data allow a depth-dependent picture of the magnetization distribution to be extracted, shown in Fig. 3D. Specifically, we note that a nonzero splitting (see Fig. 3C) is observed between the $(++)$ and $(- -)$ reflectivities near the critical edge, which indicates an intrinsic net magnetization within the film of at least $0.24 \mu_B$/Ru and up to $0.37 \mu_B$/Ru. The PNR-detected magnetization is slightly smaller than the SQUID magnetization but agrees reasonably well given possible background contributions to the SQUID value. Furthermore, a small but statistically notable spin asymmetry (SA), defined as $(R_{++} - R_{--})/(R_{++} + R_{--})$, of $0.167 \pm 0.045$ was observed at the first-order Bragg reflection. Modeling indicates that the SA of this feature is highly dependent on which layer the net magnetization originates in, with magnetic SrRuO$_3$ yielding a positive SA and magnetic SrTiO$_3$ yielding a negative SA. Since the observed SA is clearly positive, we conclude with high confidence that the observed magnetism originates from the SrRuO$_3$ monolayers as expected. Model fitting of the data supports this interpretation, with an approximate fitted magnetic moment of $0.004 \mu_B$/Ti $\pm 0.055 \mu_B$/Ti in the SrTiO$_3$ layers. We therefore conclude that PNR reveals net magnetization originating from the SrRuO$_3$ monolayers in excellent agreement with the SQUID, MOKE, and Ti XMCD measurements.

**MA of (SrRuO$_3$)$_1$/(SrTiO$_3$)$_N$ superlattices**

To reveal the exact symmetry of the MA, we perform transverse MR and magnetic field angle-dependent resistance (MAR) measurements. The MR was measured at 5 K with the current driven along the [100] direction (Fig. 4, A and B). The MR of $N = 1$ and 2 superlattices with magnetic field $H // [001]$ shows a two-peak structure with lobes reflecting the magnetic hysteresis loops. In contrast, the hysteresis loops are suppressed in the MR with $H // [010]$, consistent with the weaker in-plane magnetization of the $N = 1$ and 2 superlattices. The MR measurements of the $N = 3$ to 5 superlattices all show similar behavior in the $H // [001]$ and $H // [010]$ measurements, indicating symmetric in-plane and out-of-plane spin alignments. Figure 4C presents the polar plots of the MAR of (SrRuO$_3$)$_1$/(SrTiO$_3$)$_N$ superlattices measured at $H = 9$ T and at temperatures of 5, 25, and 50 K. The MAR of $N = 3$ and 5 superlattices with 5-K temperature steps are shown in fig. S6 (A and B). Here, we define MAR as $MAR = (\rho(\Theta) - \rho(0^\circ))/\rho(90^\circ)$, where $\rho$ is the resistivity and $\Theta$ represents the angle between the magnetic field $H$ and the film surface normal (see the inset of Fig. 4B). $H$ was rotated in the (100) plane with the electric current maintained perpendicular to the field. The $N = 1$ and 2 samples...
exhibit perpendicular MA at all measured temperatures, as do all other samples at \( T > 25 \) K, consistent with the perpendicular MA identified in the SQUID measurements. For \( N \geq 3 \), we observe a transition from twofold perpendicular MA to fourfold MA with decreasing temperature. The magnetic easy axes at low temperatures are along the \([011], [0T1], [01T], \) and \([0TT]\) directions. The MAR at 5 K with \( H \) rotating in the \((010)\) and \((001)\) planes is similar with that of the \((100)\) plane (fig. S6, C and D), as expected given the pseudocubic structure of SrRuO\(_3\) crystal lattice in the superlattices. More comprehensive MAR measurements at 5 K of the \( N = 3 \) superlattice with \( H \) rotating in the \((110)\) plane (Fig. 4D) reveal an angle of \(-71^\circ/109^\circ\) between the two magnetic easy axes within the \((110)\) plane. Thus, the observed MAR symmetry identifies magnetic easy axes along the eightfold \((111)\) directions of the SrRuO\(_3\) pseudocubic lattice. These results confirm that perpendicular MA exists in SrRuO\(_3\) monolayers for \( N \leq 2 \) superlattices at all temperatures and in \( N \geq 3 \) superlattices above \(-25 \) K. Below approximately 25 K (±5 K), the SrRuO\(_3\) monolayers in \( N \geq 3 \) superlattices exhibits eightfold MA.

**First-principle calculations of the MA**

To understand why MA of \((\text{SrRuO}_3)_{1}/(\text{SrTiO}_3)_{N}\) superlattices changes with the thickness of SrTiO\(_3\) at low temperatures, we perform first-principle calculations. The in-plane lattice constants (along \( x \) and \( y \) axes) of all superlattices are constrained to match the theoretical lattice constant of the SrTiO\(_3\) substrate. We start from a crystal structure with the experimentally observed \( a \a c \) rotation pattern (space group no. 14, \( P2_1/c \)). After atomic relaxation, density functional theory (DFT) calculations find a large rotation angle \( \gamma \) about the \( z \) axis but a very small rotation angle \( \alpha \) about the \( x \) and \( y \) axes (\( \approx 0.5^\circ \)) in both \( N = 1 \) and \( N = 3 \) superlattices, consistent with the XRD results. The layer-resolved rotation angles \( \alpha \) and \( \gamma \) of each oxygen octahedron are shown in Fig. 5 (A and D). We note that the calculated \( \gamma \) angles from our calculations are very similar to those reported in a previous study (5).

Figure 5B shows the near–Fermi-level density of states (DOS) of the \( N = 1 \) superlattice. Ru in SrRuO\(_3\) has a formal \( d^4 \) occupancy, with three electrons occupying the majority spin state (upper half of the panel) and the fourth electron in the minority spin state (lower half of the panel). The SrRuO\(_3\)/SrTiO\(_3\) interfaces remove the degeneracy between Ru \( d_{xy} \) and \( d_{yz} \), so that the fourth (minority spin) electron is evenly shared by Ru \( d_{xz} \) and Ru \( d_{yz} \) orbitals. This electronic structure is consistent with previous results (5, 15). Turning on SOC to induce MA, we test three different magnetic moment orientations: along \((001), (100), \) and \((111)\) directions. We find that in the \( N = 1 \) superlattice, the \((001)\) state has the lowest total energy among the three magnetic orientations (Fig. 5C), in agreement with the SQUID and magnetotransport measurements. The twofold \((001)\) MA is explicitly shown in the inset of Fig. 5C.

However, in the \( N = 3 \) superlattice, we find a completely new correlated state with different electronic, magnetic, and orbital properties. Figure 5E shows the near–Fermi-level DOS of the \( N = 3 \) superlattice, which indicates semiconducting behavior with a small bandgap of about 0.1 eV, in agreement with the transport measurements (fig. S7A). In the \( N = 3 \) superlattice, in the minority spin channel, Ru \( d_{xz} \) and Ru \( d_{yz} \) orbitals hybridize into a pair of new orbitals Ru \( \alpha \left| xz \right\rangle + \beta \left| yz \right\rangle \) orbital [referred to as Ru (\(+\) state)] and Ru \( \beta \left| xz \right\rangle - \alpha \left| yz \right\rangle \) orbital [referred to as Ru (\(-\) state)], where \( \alpha^2 + \beta^2 = 1 \). From our DFT + \( U \) calculations, we find \( \alpha \sim \beta / \sqrt{2} \). In each RuO\(_2\) plane, there are two distinct Ru atoms: On one Ru atom, the fourth electron fills Ru (\(+\) state and leaves Ru (\(-\) state empty; on the other Ru atom, the fourth electron fills Ru (\(-\) state and leaves Ru (\(+\) state empty. The filled new orbital is referred to as a lower Hubbard band, which is just below the Fermi level; the empty new orbital is referred to as an upper Hubbard band, which is about 2 eV above the Fermi level. Such an
orbital ordering is very similar to what is found in layered perovskite
K$_2$CuF$_4$, in which the hole orbitals $|x^2 - r^2\rangle$ and $|y^2 - r^2\rangle$ alternate in
a basal plane (23). This orbital ordering results in a ferromagnetic in-
sulating state in the CuF$_2$ plane according to Goodenough-Kanamori-
Anderson rule (24–26). The emergence of the new orbital ordering
in the $N = 3$ superlattice is corroborated with the fact that in each
RuO$_2$ layer, Ru has one pair of long Ru-O bond and one pair of short
Ru-O bond (2.06 and 1.97 Å, respectively) in our DFT calculation.
Such a bond disproportionation has also been observed in our cal-
culated results of $N = 5$ superlattice and in K$_2$CuF$_4$ (23). On the other
hand, in the $N = 1$ superlattice in which the new orbital ordering
does not occur, our calculation shows that Ru has four equal Ru-O
bonds in the RuO$_2$ plane (2.00 Å).

It is precisely this new orbital ordering that changes MA. To
demonstrate this, we turn on SOC and find that in the $N = 3$ super-
lattice, the (001) state does not have the lowest energy but rather,
the (111) state becomes the most stable among the three magnetic
orientations considered (Fig. 5F), which is consistent with the key
experimental discovery as described above. The eightfold (111) MA is
explicitly shown in the inset of Fig. 5F. The DFT calculation of
$N = 5$ superlattice is similar to that of $N = 3$, and the results are shown
in fig. S8. The reason a new correlated state emerges in the $N = 3$
and 5 superlattices is that with the RuO$_2$ layers further separated,
interplanar Ru-Ru hopping is suppressed, decreasing the band width
of Ru antibonding states (Fig. 5, B and E) and increasing correlation
effects on the Ru sites. Furthermore, the rotations of oxygen octahedra
reduce the crystal symmetry, contributing to the removal of the orbital
degeneracy (fig. S9). The two factors combined lead to a hybridization
of Ru $d_{xz}$ and Ru $d_{yz}$ orbitals and a split into a pair of lower and
upper Hubbard bands. The role of oxygen octahedral tilts on the
electronic structure is discussed in section S2. The new correlation-
driven orbital ordering and the resulting eightfold (111) MA of
(SrRuO$_3$)$_N$/(SrTiO$_3$)$_N$ ($N \geq 3$) superlattices are different from those
of magnetic interfaces in previous studies (27–31).

**DISCUSSION**
Our study reveals a novel eightfold (111) MA in SrRuO$_3$ monolayers
in (SrRuO$_3$)$_N$/(SrTiO$_3$)$_N$ superlattices ($N \geq 3$). Theoretically, our first-
principle calculations demonstrate that the enhanced correlation

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**Fig. 4.** Magnetotransport properties of (SrRuO$_3$)$_N$/(SrTiO$_3$)$_N$ superlattices. The MR at $T = 5$ K of $N = 1$ to 5 superlattices with the magnetic field applied parallel to
(A) [001] and (B) [010] directions. The color correspondences are the same in (A) and (B). (C) Polar plots of MAR of $N = 1$ to 5 superlattices measured under a magnetic field
of 9 T and at temperatures of 5, 25, and 50 K. The geometry of the MAR measurement is shown in the inset of (B). The sample rotates around the [100] direction, and the
current is along the [001] direction, always being perpendicular to the magnetic field. $\theta$ is between the [001] direction and the field direction within the (100) plane.
(D) Polar plots of MAR of $N = 3$ superlattice measured under a magnetic field of 9 T and at temperature of 5 K. The sample rotates around the [110] direction, and the
current is along the [001] direction. $\theta$ is between the [001] direction and the field direction within the (110) plane. Both MAR with the sample rotating clockwise and anti-
clockwise are shown.
strength on Ru atoms leads to a metal-to-semiconductor transition and induces an orbital ordering that is different from that of \( N = 1 \) superlattice but is similar to ferromagnetic insulator \( K_2\text{CuF}_4 \). The emergent orbital ordering changes the underlying spin-orbit interaction, reorienting the Ru magnetic easy axis. Experimentally, we performed four independent measurements (SQUID, MOKE, PNR, and MR) to understand the magnetic property of \((\text{SrRuO}_3)_{1}\!/\!(\text{SrTiO}_3)\) superlattices. Near–Fermi-level DOS of \( \text{Ru} = 4 \text{ eV} \). \( \langle 001 \rangle \), \( \langle 100 \rangle \), and \( \langle 111 \rangle \) refer to the orientation of Ru magnetic moments. The energy of the \( \langle 001 \rangle \) state is used as the reference. The twofold \( \langle 001 \rangle \) MA is explicitly shown in the inset of (C). The eightfold \( \langle 111 \rangle \) MA is explicitly shown in the inset of (F).

**MATERIALS AND METHODS**

**Preparation and structural characterizations of \((\text{SrRuO}_3)_{1}\!/\!(\text{SrTiO}_3)_{N}\) superlattices**

The \((\text{SrRuO}_3)_{1}\!/\!(\text{SrTiO}_3)_{N} (N = 1 \text{ to } 5)\) superlattices were fabricated on \( \langle 001 \rangle \) \( \text{SrTiO}_3 \) substrates using single-crystalline \( \text{SrTiO}_3 \) and ceramic \( \text{SrRuO}_3 \) targets by PLD assisted with RHEED. The (001) substrates with atomically flat TiO\(_2\) termination were obtained via buffered hydrofluoric acid etching and annealing. The RHEED system was used to monitor the layer-by-layer growth of the films, and the total repetitions of the \( N = 1 \) to 5 superlattices are all 50. The thicknesses of both the \( \text{SrRuO}_3 \) layer and the \( \text{SrTiO}_3 \) layer are precisely controlled at a single molecular level by RHEED. All films were grown at a substrate temperature around 700°C and under an oxygen pressure of 10 Pa. During the growth, the laser frequency and energy density were 2 Hz and ~1 J/cm\(^2\), respectively. After the deposition, all films were in situ annealed at 500°C for an hour in an oxygen environment of \( 5 \times 10^{19} \text{ Pa} \) to remove oxygen vacancies.

**Magnetic and magnetotransport characterizations**

The magnetic properties of the superlattices were probed using SQUID and MOKE techniques. The temperature-dependent magnetization and Kerr rotation measurements of the superlattices were done during warming up under a smaller field of 0.05 T after the samples were
first cooled down to 4 K under a field of 0.5 T for SQUID and 0.05 T for MOKE. The transport properties were measured using a standard linear four-probe method by a physical property measurement system equipped with a sample rotator. Au electrodes were deposited using Ar ion sputtering on top of the superlattices. During the transport measurements, the dc current of around 10 µA was applied to the films, and the direction of the current was maintained to be perpendicular to the magnetic field.

PNR measurements

PNR measurements were performed using the polarized beam reflectometer instrument at the National Institute of Standards and Technology Center for Neutron Research. Samples were cooled to 6 K in an applied field of 3 T. Full polarization analysis was performed using both a spin polarizer and spin analyzer. The spin-dependent reflectivity was measured as a function of the scattering vector Q along the film normal. Data were reduced with the Reducct softwre package (42) and analyzed with the Refl1D software package for reflectometry modeling (43). Uncertainties in fitted parameters were extracted using a Markov chain Monte Carlo algorithm Differential Evolution Adaptive Metropolis (DREAM) as implemented in the Bumps python package. We note that PNR is sensitive only to the net in-plane components of the magnetization within the film, so that any out-of-plane component, for example, canted toward [111] axes will not be observed. For that reason, the reported magnetization values have been adjusted to account for the fact that SQUID magnetometry indicates that the films are approximately 10% below saturation value at 3 T. We also note that since no in-plane perpendicular magnetization component is expected in an applied field of 3 T, the spin-flip reflectivities \( R^+ \) and \( R^- \) are expected to be zero and were not collected. Only the non-spin-flip scattering cross sections \( R^+ \) and \( R^- \) were measured.

X-ray spectroscopic measurements

The Ti L-edge XAS and XMCD measurements were performed on beamline 4.0.2 at the Advanced Light Source (ALS) of Lawrence Berkeley National Laboratory at a temperature of 10 K and under the vacuum pressure of \( \approx 1 \times 10^{-6} \) Pa. The XAS spectra were recorded in total electron yield (TEY) mode (sample-to-ground drain current) and normalized by the incident photon flux determined from the photocurrent of an upstream Au mesh. The samples were measured with alternating left-polarized (\( \mu^- \)) and right-polarized (\( \mu^+ \)) photons at 10 K cooled by liquid helium in an applied field of 4 T. During the XMCD measurement, the incident beam was perpendicular or inclined with a grazing angle of 20° to the sample surface and the spectra were collected in both TEY and luminescence yield mode. Preliminary room temperature XAS measurements have been performed on beamline 8.0.1 at ALS and beamline BL12B-a at the National Synchrotron Radiation Laboratory of China. The XANES measurements at Ru K-edge were performed at the beamline 12-BM-B, and the x-ray linear dichroism measurements at Ru L3-edge were carried out at the beamline 4-ID-D at the Advanced Photon Source of Argonne National Laboratory.

First-principle calculations

We perform DFT calculations using a plane wave basis set and projector-augmented wave method (44), as implemented in the Vienna Ab initio Simulation Package (45). We use Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation as exchange correlation functional (46). An energy cutoff of 600 eV is used throughout the calculations. The Brillouin zone integration is performed with a Gaussian smearing of 0.05 eV over a Γ-centered \( k \)-mesh of 12 \( \times \) 12 \( \times \) 12 for the \( N = 1 \) superlattice and a Γ-centered \( k \)-mesh of 12 \( \times \) 12 \( \times \) 6 for the \( N = 3 \) and 5 superlattices. The threshold of self-consistent calculations is \( 10^{-6} \) eV. Crystal structure is relaxed until each force component is smaller than 0.01 eV/Å. The in-plane lattice constant is fixed to be 3.93 Å, which is the theoretical lattice constant of SrTiO3 calculated by DFT-PBE method. Correlation effects on Ru atoms are taken into account (47) by using the rotationally invariant Hubbard U method in DFT calculations (DFT + U method) (48). Following the previous study, we use \( U_{Ru} = 4 \) eV (5). The key results do not qualitatively change for \( U_{Ru} \geq 3 \) eV. SOC is turned on to study MA in DFT + U + SOC calculations.

SUPPLEMENTARY MATERIALS

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/6/15/eaya0114/DC1

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