Supplementary Materials for

Unconventional interlayer exchange coupling via chiral phonons in synthetic magnetic oxide heterostructures

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Supplementary Text

Supplementary Text 1. Characterization of the spiral spin structure of the SRO/STO superlattices using a phenomenological model.

To figure out the orientation of the spin vectors in each SRO layer within the superlattices and describe the oscillatory magnetic behavior of $M$ ($2\ K$) values, we modeled synthetic spiral spin structures with different $\gamma$ (in $\text{[SRO|STO]}_{\gamma}$; $\gamma = 2$ (bilayer SRO), 3 (trilayer SRO), 4, and 5) and $t_{\text{STO}}$ (Figs. S8 and S10). Assuming that each SRO layer has a magnetization vector $M_i$, we estimate that neighboring SRO layers have the same amplitude but a relative angle difference ($\phi$) linearly dependent on $t_{\text{STO}}$. In order to calculate the total magnetization of the superlattice, we estimated the sum of projections of $M_i(\phi)$ along the $H$-field for each SRO layer, mimicking the result from MPMS, which only shows an average scalar magnetization value of the whole superlattice along the $H$-field direction. Assuming that the bottommost spin vector is directed along the $H$-field, $M_i(\phi)$ was defined as,

$$\sum_i M_i(\phi) = \sum_i [M_0 + M_1 \cos((\gamma - 1)\phi)],$$

where $M_0$ and $M_1$ are independent of $\phi$. Fig. S10b exhibits a $\phi$-dependent sum of squared error (SSE) values, calculated by $[(\text{Simulated } M) - (\text{Measured } M)]^2$. The spiral spin structure model with $\phi = -160$° and $-200$° have the lowest SSE values for $t_{\text{STO}} = 1.6$ nm (Inset of Fig. S10B). $M_0$ and $M_1$ values are 0.070 and 0.629 $\mu$B/Ru, respectively. While spin models with both $\phi$ values well reproduce the $\gamma$-dependence of $M$ ($2\ K$) (Fig. S10C), the oscillatory magnetic behavior of $M$ ($2\ K$) as a function of $t_{\text{STO}}$ can be better explained by the spiral spin structure with $\phi = \approx 160$° (Fig. S10D). Thus, we conclude that the spiral spin structure with $\phi = \approx 160$° is the most reasonable spin configuration to describe the magnetization results obtained by MPMS. We emphasize that the proposed magnetic configurations do not yield a zero remnant magnetization. See Fig. S11 for clear visualization of the spin configurations. It should also be noted that this spiral spin structure coincides with the PNR measurements as shown below.

Supplementary Text 2. Analysis of PNR data.

We first measured the NR spectra at 300 K without $H$-field to examine the atomic structure of the superlattice (Fig. S12A). The result is highly consistent with the XRR result (Fig. S2 and Table S1). We then estimated the saturation magnetization values of the superlattice by measuring the PNR spectra at 85 K with 1 T ($> H_c$ at 85 K) of in-plane $H$-field (Fig. S12B). The experiment was done without spin-flip polarization analysis, $R^+ = R^{++} + R^-$ and $R^- = R^{--} + R^-$, where non-spin-flip $R^{++}$ ($R^{--}$) is proportional to $M_y = M \sin(\phi)$ and $R^-$ ($R^-$) is proportional to $M_x = M \cos(\phi)$ (34, 41, 46-48, 53). The estimated saturation magnetization value was $\approx 0.4 \mu_B$/Ru, consistent with the MPMS. Assuming that the scalar magnetization value of each SRO layer is $\approx 0.4 \mu_B$/Ru, we carefully fit the PNR spectra at 5 K with a 0.01 T of in-plane $H$-field utilizing three different spin structures, i.e., a collinear FM, a collinear sAFM, and a non-collinear spiral spin model with $\phi = 160$° as shown in Supplementary Text 1. The simulated PNR spectra using collinear spin models have clear distinctions from our PNR data: (1) The strong dip at $Q = \approx 0.8$ nm$^{-1}$ (half of the superlattice peak $Q$ value) in $R^-$ of the collinear sAFM model simulation (Fig. S13B) and (2) the large differences at $Q = \approx 1.6$ nm$^{-1}$ (the superlattice peak $Q$ value) between $R^+$ and $R^-$ of the collinear FM model simulation (Fig. S13C) cannot account for the experimental PNR spectra. On the other hand, the non-collinear spiral spin structure with $\phi = \approx 160 \pm 5$° well-describes our PNR data (Fig. S14), consistent with the analyses from the magnetization results of MPMS as described in Supplementary Text 1 and Fig. S10. In addition, we tested the FM contribution to our PNR spectra by combining the two different spin models, i.e., FM ($M_0$) and
spiral spin structures with $\phi \approx 160^\circ$ ($M_1$) (see Supplementary Text 1, Fig. S15). Table S2 shows that SSE of three different spin models with different ratios between $M_0$ and $M_1$ results in nearly the same SSE values. While this result implies that some FM contribution might exist in our superlattice system at 5 K, the existence of non-collinear spiral spin structure in the SRO/STO superlattices is undeniable, consistently supported by both the MPMS and PNR measurements and analyses.

Supplementary Text 3. Estimation of the RKKY-induced magnetic oscillation.
We estimated the RKKY interaction induced oscillation wavelength ($\lambda_{RKKY}$), assuming that the STO spacer layer has a finite number of itinerant carriers possibly from unintentionally introduced oxygen vacancies. We note that such speculation is unlikely, as the junction current across the superlattice shows an insulating $T$-dependent behavior as shown in Fig. S16. Junction transport of [6|2]$_{10}$ was performed using Nb-doped (0.5 wt %) STO substrate as the bottom electrode and Au (~500 μm in diameter) as the top electrode, as already mentioned in the method section in our manuscript. A few kΩ of tunneling resistance was observed for the ultrathin STO layer thickness (~8 nm) within the [6|2]$_{10}$ superlattice. If we convert the resistance into resistivity, the value becomes in the order of ~10$^6$ Ω cm, which is compared to that of wood. This is a large enough resistivity considering quantum tunneling is possible in the superlattice. Note that we used high oxygen partial pressure of 100 mTorr to grow the superlattices. Numerous experiments from other groups consistently confirm that 100 mTorr of oxygen partial pressure during growth results in the most stoichiometric and insulating STO thin films. The insulating behavior of the STO layer within our superlattices was further confirmed from optical, XAS, DFT, and STEM-EELS-EDX analyses performed in one of our previous studies on the superlattices (24, 25). Particularly, the Ti $L_3$-edge XAS spectra revealed the prevalence of only the Ti$^{4+}$ valence state, indicating no (unintended) itinerant carriers in the STO layers. We further note that the Ti$^{4+}$ valence state of the superlattice is independent of the STO thickness. Although such speculation is highly unlikely, by considering the carrier density of the STO spacer layer ($n_{STO} \approx 10^{18}$ cm$^{-3}$) (54), the $\lambda_{RKKY}$ was determined by,

$$\lambda_{RKKY} = \frac{\pi}{k_F} = \frac{\pi}{(3\delta_{STO} T)\tau}.$$  

(2)

The estimated $\lambda_{RKKY}$ was ~14.9 nm, which is an order of magnitude larger than the observed magnetic oscillation periods (~2 nm) in the SRO/STO superlattices. Thus, the RKKY interaction cannot account for the magnetic behavior through the NM-I STO spacer.
Supplementary Text 4. Extended discussion of possible IEC through a NM-I layer.

(1) Schottky defects-induced IEC
It is noteworthy that there are a few recent experimental reports on magnetic oscillations across insulating spacers (31-34). However, interpretations of the magnetic oscillation are often controversial, and most of them conceive unintentional charge carriers remaining in the NM-I spacer layer. More importantly, previous observations rely on the change in the macroscopic exchange bias, stressing the role of extrinsic magnetic pinning layer. In particular, a recent report speculated that Schottky defects in NM-I spacer within a polar heterostructure can induce an oscillation of exchange bias via defect-induced electron hopping, by showing a defect-induced Raman active mode for the NM-I spacer, with strong magnetic field dependence (34). Note that the reference does not provide any rigorous origin of the behavior, and it does not indicate any phonons playing a role. Our SRO/STO superlattices are clearly different. SRO/STO superlattices exhibit highly suppressed charge transfer at the interfaces, evidenced by various experiments and theoretical calculations (23-25). There is no exchange bias effect in the $M(H)$ curves of the SRO/STO superlattices, indicating the absence of extrinsic magnetic pinning layers (Numerous experiments consistently showed no exchange bias in SRO/STO heterostructures (21, 24-27)). Furthermore, observed phonon modes in the SRO/STO superlattices are octahedral distortion-related intrinsic phonon modes in the SRO layer, and not defect-induced modes of the NM-I spacer. Finally, our system is nominally non-polar, and hence, cannot be understood in terms of Schottky defect-induced IEC (34). The identical $A$-site (Sr) ions in our SRO/STO superlattices lead to a nominally symmetric interface.

(2) Interfacial magnetic coupling
The absence of exchange bias in $M(H)$ curve of the SRO/STO superlattice indicates that there is no extrinsic pinning layer in the superlattices, especially near the interfaces. Moreover, the $t_{STO}$-dependent oscillation of magnetization cannot be understood from any interfacial magnetic coupling. $M(H)$ curve for the superlattices with thick STO layer (i.e., [6]18)10 superlattice, $t_{STO} = \sim$7 nm) returns to conventional single FM hysteresis loop with almost the same saturation magnetization of [6]10 superlattice. It strongly supports that the interfacial magnetic coupling cannot account for the unconventional IEC in SRO/STO superlattices.
Fig. S1. XRD $\theta$-2$\theta$ scans of $[\alpha|\beta]_{10}$ superlattices with different $\alpha$ and $\beta$. Clear superlattice Bragg peaks show the atomically well-defined periodicities of superlattices. The asterisk (*) indicates the STO (001) substrate peak.
Fig. S2. X-ray reflectivity (XRR) of [6/β]10 superlattices with different β. The XRR result shows atomically controlled periodicities of the superlattices with a small deviation of the thickness $< 0.1$ nm. Thus, we believe magnetic inhomogeneity would be small, if any, and further, it would not disrupt the NM-I spacer thickness-dependent unconventional magnetic behavior. The symbols and solid lines are the experimental data and fit of the XRR data.
Fig. S3. Atomically sharp interface and surface of SRO/STO heterostructures (not a superlattice). Cross-sectional HAADF-STEM images of the SRO/STO heterostructures (not a superlattice) in (A) low and (B) high magnifications. The number of atomic layers was clearly visualized to confirm the designed structure, and the interfaces were proven to be atomically sharp. The bright layers enclosed by orange rectangles indicate the SRO layers. The STEM images manifest a coherent superlattice with a fully-strained state (24, 25). (C) Surface topography of SRO/STO heterostructures with an atomically flat step-and-terrace structure recorded using atomic force microscopy. This image was obtained in $5 \times 5 \, \mu m^2$ scales using contact mode.
Fig. S4. \( M(H) \) and \( M(T) \) curves of an SRO single film. (A) In-plane and (B) out-of-plane \( M(T) \) curves of an SRO single film measured with 0.01 T of \( H \)-field using a field-cooled method. The inset shows \( H \)-dependent in-plane magnetization of an SRO single film. The arrows indicate the direction of the \( H \)-field. This result shows the nearly second-order FM transition of SRO, consistent with previous studies (52), and that the magnetic easy axis of SRO single films lies along the out-of-plane direction. We also note that the magnetic anisotropy field of SRO is in the order of 10 T, indicating out-of-plane magnetic moment cannot rotate along the in-plane direction under a few T of the in-plane magnetic field applied.
Fig. S5. Anisotropic $M (T)$ curves of an SRO/STO superlattice. (A) In-plane and (B) out-of-plane $M (T)$ curves of [6|4]$_{10}$ superlattice measured with 0.01 T of $H$-field using a field-cooled method. We note that the magnetic easy-axis of the superlattice is still out-of-plane, but the $t_{STO}$-dependent magnetic oscillation was observed only along the in-plane direction.
Fig. S6. $M(T)$ curves of an SRO/STO superlattice at different $H$-field. (A) $M(T)$ curves of a [6|4]$_{10}$ superlattice recorded at various in-plane $H$-field. The field-cooled method was used. Because the diamagnetic of STO with a high magnetic field show a large negative offset of the $M$ value, we displayed $M(T) - M(300 \text{ K})$ value. (B) In-plane $M(H)$ curves measured at 5 and 85 K. The arrows indicate the direction of $H$-field.
Fig. S7. STO-thickness dependent oscillations in [8β]10 and [4β]10 superlattices. (A) $M(T)$ curves of the [8β]10 superlattices. (B-D) Results from sub-u.c. controlled [4β]10 superlattices. (B) XRD $\theta$-2$\theta$ scans and (C) $M(T)$ curves of sub-u.c. controlled [4β]10 superlattices. (D) $M(2K)$ values extracted from (C). These results support the oscillatory magnetization behavior in the SRO/STO superlattices. Note that this sample set with 4 u.c. SRO thickness shows lower absolute magnetization values compared to other superlattices shown in the main text, possibly originating from the thickness deviation of the SRO layer.
Fig. S8. Reproducibility of the magnetic behavior of the superlattices. (A) and (B) Two different sample sets of the superlattices with the same configuration. Both consistently show the $t_{\text{STO}}$-dependence, indicating good reproducibility of the oscillatory result. $M(T)$ curves were measured with 0.01 T of in-plane $H$-field using the field-cooled method.
Fig. S9. Estimated PNR spectra with spin-flip polarizations. Estimated PNR spectra with spin-flip polarizations ($R^+$ and $R^-$) show the small PNR signal.
Fig. S10. Characterization of the spin vectors for the SRO/STO superlattices. (A) $M(T)$ curves of $[6|4]_\gamma$ superlattices with different $\gamma$. $M(T)$ curves were measured with 0.01 T of in-plane $H$-field using the field-cooled method. (B) $\phi$-dependent SSE values (see Supplementary Text 1). The dotted vertical line indicates $\phi = 180^\circ$, indicating the sAFM configuration. The inset shows a magnified region near $\phi = 180^\circ$. The arrows indicate the lowest SSE values at $\phi = 160^\circ$ and $200^\circ$. (C) $\gamma$- and (D) $t_{STO}$-dependent $M(2\,K)$ values and the estimated magnetization values using the model described in Supplementary Text 1. The error bars indicate the deviation from the measurements.
Fig. S11. Schematic representation of the in-plane spin vector configurations and net magnetization in the SRO/STO superlattices. $S_i$ represents the in-plane spin of the SRO layer starting from the bottommost layer ($S_1$). Assuming that each SRO layer has an in-plane magnetization vector $M_i$, we estimate that neighboring SRO layers have the same amplitude, but different orientation (see Supplementary Text 1). The red arrows denote the summation of vectors, $S_{\text{sum}}$, indicating the non-zero net in-plane magnetization that oscillates with $t_{\text{STO}}$. (1) Repetition number- (γ-) and (2) $t_{\text{STO}}$- (β-) dependent $S_{\text{sum}}$ are shown for comparison.
Fig. S12. **PNR spectra were measured at different $T$.** (A) Unpolarized neutron reflectivity (NR) spectra at 300 K. (B) PNR spectra at 85 K with an in-plane $H$-field of 1 T, where $R^+$ and $R^-$ indicate the PNR spectra for spin-up and spin-down neutrons, respectively. We effectively reduced the number of fitting parameters in our PNR fitting. Initially, the structural fitting parameters were fixed based on our XRR and NR measurements. Then, we estimated the saturation magnetization values of the superlattice by measuring the PNR spectra at 85 K with 1 T ($> H_c$ at 85 K) of in-plane $H$-field.
**Fig. S13. Analysis of the PNR spectra.** (A) PNR spectra with S. A. results for three models corresponding to three different scenarios for spin configuration (see Supplementary Text 2). Magnified PNR spectra for (B) $Q = 0.5–1.0$ nm$^{-1}$ and (C), $Q = 1.4–1.8$ nm$^{-1}$ region.
Fig. S14. Simulation of non-collinear spiral spin structure with varying $\phi$. To confirm the sensitivity of $\phi$ in the spiral spin structures of the superlattices, we performed the simulation with three different values of $\phi$ ($\phi = 150^\circ$, $160^\circ$, and $170^\circ$). The bottom panels show the enlarged portion near $Q = 0.8$ nm$^{-1}$. The result from spin model with $\phi = 150^\circ$ shows a large discrepancy to the experimental spectra for the $R^+$ polarization (blue), whereas the result from spin model with $\phi = 170^\circ$ shows a large discrepancy to the experimental spectra for the $R^-$ polarization (red). The result from spin model with $\phi \approx 160^\circ$ best describes our experimental PNR data, consistent with the magnetization analyses based on the results of MPMS.
Fig. S15. Partial FM contribution in the PNR spectra. (A-C) PNR spectra and S. A. results for three different spin models, including different contributions of the FM component, $M_0$ (see Supplementary Text 2).
Fig. S16. Junction transport property of the superlattice with atomically thin STO layer. $T$-dependent resistance measured using junction transport geometry of a $[6|2]_{10}$ superlattice. 0.3 $\mu$A of the excitation current was used. The superlattice even with the thin STO layers (~0.8 nm) clearly shows an overall insulating $T$-dependence. Since SRO layer is metallic, the STO layer should be insulating to show the overall insulating $T$-dependence. The inset schematically shows the measurement configurations. See Supplementary Text 3 for more discussion.
**Fig. S17.** *T*-dependent confocal Raman spectra in $z(x)xz$ polarization of a [6|8] superlattice. The symbols and solid lines represent experimental data and fit for the Raman spectra, respectively. Pink and yellow solid lines correspond to the $A$ and $B$ phonons. $T$-dependent $\omega$ splitting below $T_c$. The dotted horizontal lines indicate (up) $T_c$ of SRO and (down) the lowest $T$ (10 K) of the measurement.
Table S1. Thickness analyses of the atomically controlled SRO/STO superlattices. The total thickness of superlattice samples, obtained by XRR, PNR (data not shown for [6|6]10 and [6|8]10 superlattices), and STEM (25), consistently show the atomically designed SRO/STO superlattices with a small deviation in thickness below ~1 nm.

| Samples  | Designed thickness (nm) |
|----------|-------------------------|
| [6|2]10   | 31.366                  |
|          | 31.256                  |
|          | XRR                     |
| [6|4]10   | 39.176                  |
|          | 39.306                  |
|          | 39.134                  |
|          | PNR                     |
| [6|6]10   | 46.986                  |
|          | 46.756                  |
|          | 47.430                  |
| [6|8]10   | 54.796                  |
|          | 54.356                  |
|          | 53.930                  |
| [6|8]10   | 54.930                  |
Table S2. SSE values of PNR spectra. We estimated the partial FM contribution in the PNR measurement at 5 K with 0.01 T of in-plane $H$-field via comparing the sum of squared error (SSE) values. The SSE of three different spin structures (A to C spin model) is calculated by $[(\text{Simulated } M) - (\text{Measured } M)]^2$.

| Spin model |         | B      | C      |
|------------|---------|--------|--------|
| $M_0$ ($\mu B$/Ru) | 0.4     | 0.3    | 0.2    |
| $M_1$ ($\mu B$/Ru) | 0       | 0.1    | 0.2    |
| SSE        | A       |        |        |
| R$^+$      | 0.001118| 0.001115| 0.001110|
| R$^-$      | 0.001163| 0.001167| 0.001218|
| Average    | 0.001141| 0.001141| 0.001164|
| S. A.      | 0.020479| 0.020023| 0.023138|
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