

Ferromagnetism in $\text{LaFeO}_3/\text{LaNiO}_3$ superlattices with high Curie temperature

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Interfacing complex oxides in atomically engineered layered structures can give rise to a wealth of exceptional electronic and magnetic properties that surpass those of the individual building blocks. Herein, we demonstrate a ferromagnetic spin order with a high Curie temperature of 608 K in superlattices consisting of otherwise paramagnetic perovskite LaNiO_3 (LNO) and antiferromagnetic LaFeO_3 (LFO). The ferromagnetism likely results from the covalent exchange due to interfacial charge transfer from Fe to Ni cations. By deliberately controlling the thickness of the LNO sublayers thus the amount of charge transfer, a robust ferromagnetism of $4 \mu_B$ is realized for a stacking periodicity consisting of one single unit cell of both LNO and LFO, an emergent double perovskite phase of $\text{La}_2\text{FeNiO}_6$ with B-site layered ordering configurations. The ferromagnetic LFO/LNO superlattices offer great potential for the search of emergent magnetodielectric and/or multiferroic properties as well as applications in spintronics and electrocatalysts.

Atomically engineered heterostructures of strongly correlated oxides have enabled the rational design of emergent interfacial magnetic and electric properties not present in the constituting layer^{1–3}. The most prominent examples are the two-dimensional (2D) electron gas formed at the interface between two band-gap insulators^{4,5}, superconducting interfaces between non-superconducting cuprates⁶, and the ferromagnetism between two antiferromagnetic materials⁷. Among the magnetically ordered materials, including ferromagnetic (FM), ferrimagnetic, and antiferromagnetic (AFM), FM and ferrimagnetic materials have significant technological importance in diverse applications, such as permanent magnets, microwave devices, sensors, magnetic recording, and memory media⁸. Magnetic interfaces, particularly those based on perovskite oxides, ABO_3 , are of similar technological importance, but the interface-induced changes to electronic structure, orbital occupancy, and charge redistribution in addition to metal–oxygen band hybridization often lead to magnetic exchange interactions across B–O–B bonds different from those of the bulk^{9–11}. Notably, although the charge transfer and the emerging functionalities at oxide heterointerfaces have been

intensively investigated for early transition metal oxides^{9,11–14}, the charge transfer across heterostructures consisting of two late transitional metal oxides remains less investigated^{10,15–17}. Research is extremely limited for cases where the thickness of the component for the heterointerface or the superlattice is precisely controlled down to a single unit cell (uc) of the perovskite oxide ($\sim 0.4 \text{ nm}$)^{18,19}.

Rare-earth nickelate RNiO_3 (with R = rare-earth) is a typical family of late transition metal oxides, which have attracted significant interest in recent years due to their remarkable properties including sharp metal-to-insulator transitions (MIT) tunable with the R radius^{19–21} as well as the emergence of superconductivity in the infinitely layered $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films²². LaNiO_3 (LNO) is the only member of the rare earth perovskite RNiO_3 that is a Pauli paramagnetic (PM) metal with no signature of MIT or magnetic order at any temperature. Electronically, LNO is a charge transfer metal with strong hybridization between Ni $3d$ and oxygen $2p$ states, where the electronic configuration can be described as a superposition form $|\psi\rangle = \alpha|3d^7\rangle + \beta|3d^8\bar{L}\rangle$ ($\bar{L}$ denotes a ligand hole on the oxygen ion)²³. The system shows partial covalence, and the degree of

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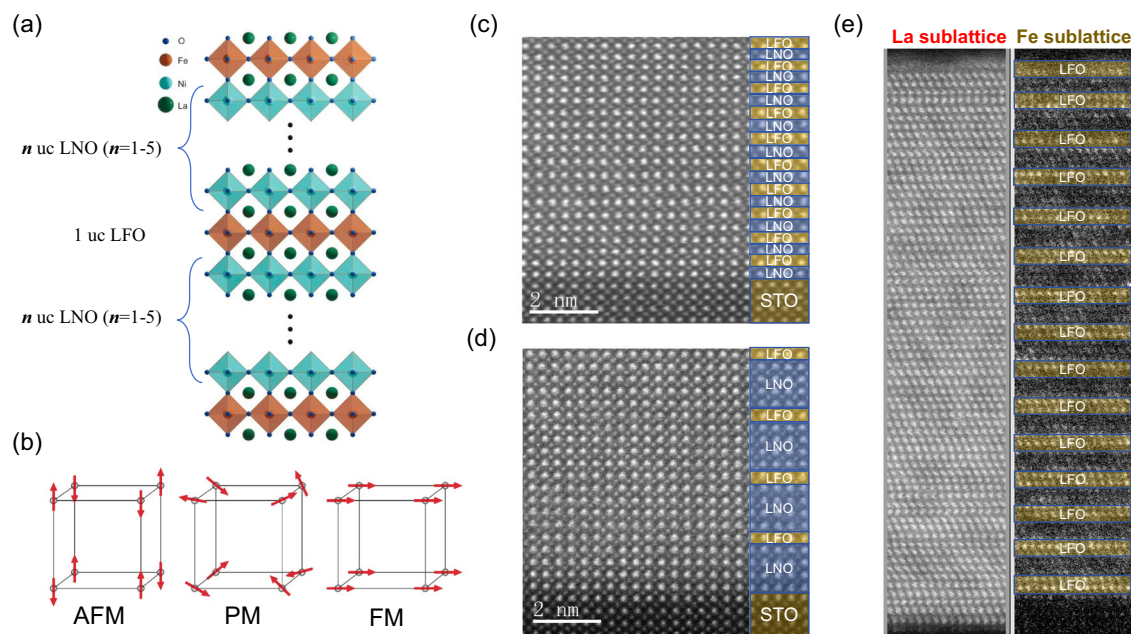


Fig. 1 | Epitaxial growth of the LNO_n/LFO₁ SLs. **a** The sketch of the SL structure where the thickness of paramagnetic LNO, n , was varied from $n = 1$ to 5 uc, while the G-AFM LFO layer was set to 1 uc. **b** The illustration for magnetic structures of G-AFM, paramagnetic (PM), and FM coupling. **c** and **d** The cross-sectional STEM

HAADF images of the LNO₁/LFO₁ and LNO₄/LFO₁ SL, respectively, were recorded along the [100] zone axis. **e** The EELS maps obtained from the analysis of the La M_{4,5} (left) and Fe L_{2,3} (right) signals reveal the epitaxial growth of the periodic structure and cation intermixing confined within 1–2 uc of LFO (within 1 p.l. of the SL).

covalence is given by the ratio between β^2 and α^2 ($\alpha^2 + \beta^2 = 1$). The hybridization between the 3d band and the oxygen 2p band results in the formation of oxygen holes and a small or negative charge transfer gap²¹, which are key features of high-temperature superconducting cuprates²⁴. Therefore, strain-engineered thin films or multilayers based on nickelates have been intensively investigated to explore electronic structures that would mimic those of cuprate parent materials²⁵, which prefer AFM order. Moreover, the low-lying Ni 3d bands of LNO with partial filling make it prone to electron acceptors. Notably, ferromagnetic-like behavior has been reported in the paramagnetic LNO layer when it is interfaced with another ferromagnetic layer of LaMnO₃ thin films^{15,16} or a ferrimagnetic layer of GdTIO₃¹¹. But most of such charge-transfer-induced ferromagnetism shows Curie temperature below room temperature.

In this manuscript, we report a charge transfer-induced ferromagnetism in the heterostructure of LaFeO₃/LaNiO₃ (LFO/LNO) (Fig. 1a), where LFO is a G-type AFM insulator (Fig. 1b) as electron donor, the amount of the transferred charge is controlled by the thickness of LNO on the scale of 1 uc. In the extreme limit of the single superlattice (SL) stacking periodicity (periodic lattice, p.l.) consisting of one unit cell (uc) pseudo-cubic LNO and one unit cell pseudo-cubic LFO, similar to the theoretically predicted double perovskite (DP) La₂NiFeO₆²⁶, the SL shows not only a high magnetization of 4 μ_B /p.l., but also a high Curie temperature of 608 K. The emergent properties are not observed either in the constituting materials (LFO and LNO) or the mixed compound LaFe_{0.5}Ni_{0.5}O₃²⁷, which could result from the formation of an emergent DP phase of La₂FeNiO₆ with B-site layered ordering configurations along the (001) orientation. The ability to affect interfacial charge transfer and the Ni 3d–O 2p covalency thus the Ni 3d–O 2p–Fe 3d exchange interactions through a precisely controlled approach, not realizable by conventional processing techniques, suggests a new avenue to realizing FM materials.

Results

Creation of LNO_n/LFO₁ superlattices with layered ordering of cations

We have grown LNO_n/LFO_m superlattices (SLs) by alternately stacking LNO and LFO layers using pulsed laser deposition on SrTiO₃ (STO) (001)

substrates (see the “Methods” section), where n and m represent the layer thickness of LNO and LFO in pseudo-cubic uc, respectively. Here n was varied from $n = 1$ to 5 uc, while the LFO layer was set to 1 uc thick. The LNO_n/LFO₁ bilayer was repeated 10–20 times to form the SL. The SL growth was monitored in-situ by reflection high-energy electron diffraction (RHEED). Streaked RHEED patterns with clear intensity oscillations were observed during the growth of both LNO and LFO films, implying a 2D layer-by-layer film growth mode (Supplementary Fig. S1). The epitaxial growth of the high-quality LNO_n/LFO₁ SLs was also confirmed by X-ray diffraction experiments and high-resolution scanning transmission electron microscopy (STEM). Notably, although clear Laue fringes are visible for all the SLs from high-resolution XRD θ – 2θ scans (Supplementary Fig. S2) due to their atomically smooth surfaces as confirmed by atomic force microscopy (Supplementary Fig. S3), the satellite peaks indicating the chemically modulated structure with well-defined interfaces were only visible for $n \geq 3$, consistent with the previous results on similar structure¹⁷. Moreover, for $n \leq 3$, the SL is under compressive strain with $c \geq 3.905$ Å. While as $n > 3$, the SL is under tensile strain. Such decrease of out of plane c with increasing n is consistent with the smaller lattice constant of LNO compared to LFO (LFO is orthorhombic with a pseudo-cubic lattice constant $a_0 = 3.93$ Å¹⁷, and LNO is rhombohedral with pseudo-cubic $a_0 = 3.838$ Å). Figure 1c and d show the atomically resolved high-angle annular dark-field (HAADF) image of the cross-section of the LNO₁/LFO₁ SL and LNO₄/LFO₁ SL samples, respectively, recorded along the [100] zone axis. As the intensity of HAADF-STEM images is sensitive to atomic number (Z), both SLs show coherent epitaxial lattice as demonstrated by the brightest sublattice of the La ions. No dislocations, defects, or any nano-scale phase-separated microstructures such as Fe or Ni metal clusters were observed. The compositional electron energy-loss spectroscopy (EELS) maps obtained from the analysis of the La M_{4,5} (Fig. 1e left), Fe L_{2,3} (Fig. 1e right), and Ni L_{2,3} signals reveal the epitaxial growth of the periodic layered structure, which is flat and continuous over long lateral distances. Cation intermixing of Fe and Ni ions when occurs is found to be confined within 1–2 uc of LFO (within 1 p.l. of the SL). Shortly, high-quality LNO_n/LFO₁ superlattices with layered ordering of configurations for Fe and Ni cations are successfully fabricated.

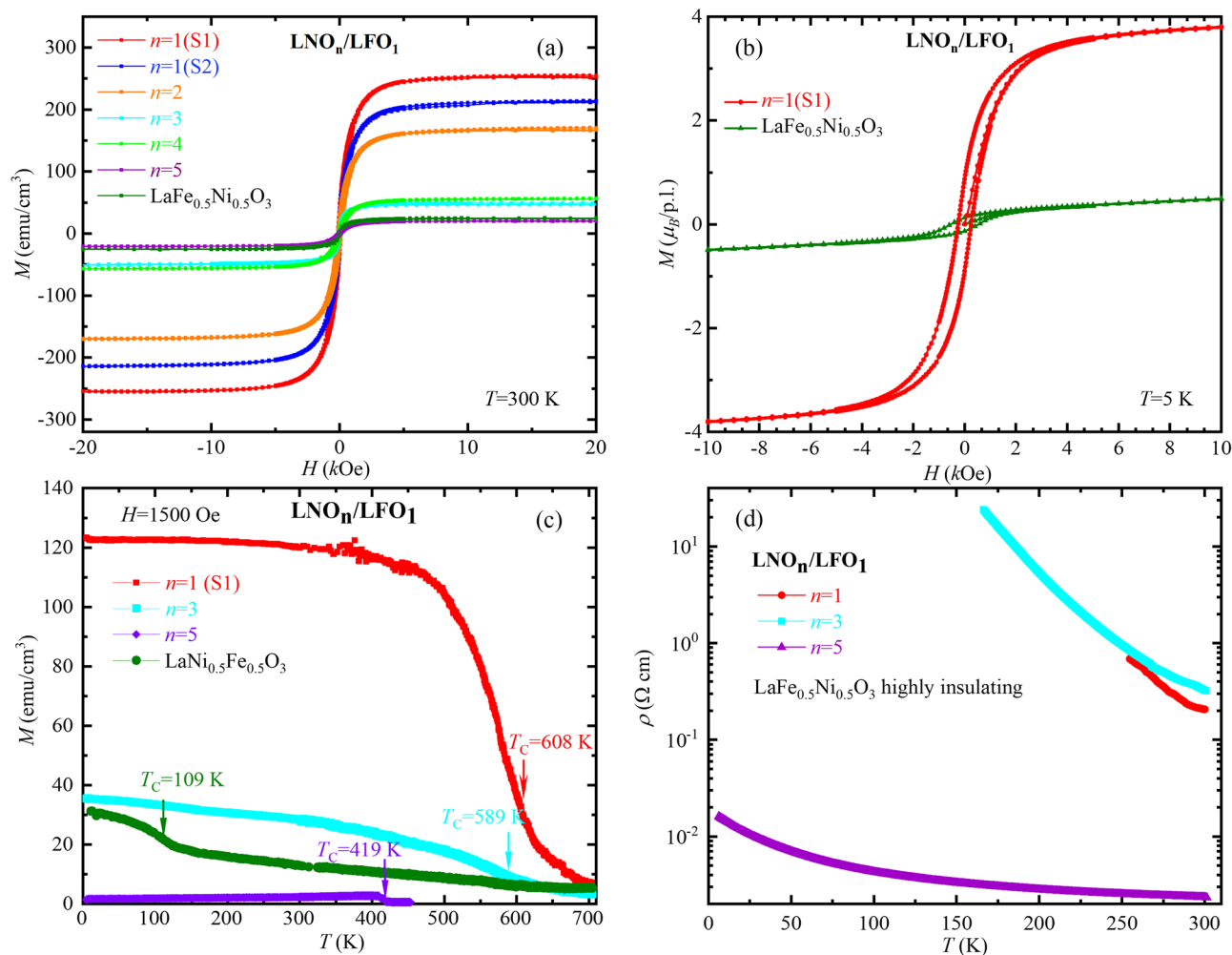


Fig. 2 | Magnetic and transport properties of the atomically engineered LNO_n/LFO₁ SLs. **a** The hysteresis curves of the LNO_n/LFO₁ SLs measured at 300 K, for comparison the results of LaFe_{0.5}Ni_{0.5}O₃ thin films, is also presented. **b** The hysteresis curve of the LNO₁/LFO₁ SL measured at 5 K in comparison to the LaFe_{0.5}Ni_{0.5}O₃ thin film. **c** The temperature dependence of magnetization for the LNO_n/LFO₁ ($n = 1, 3$ and 5) SLs and the LaNi_{0.5}Fe_{0.5}O₃ film. **d** The temperature-dependent resistivity, ρ , of the LNO_n/LFO₁ ($n = 1, 3$, and 5) SLs, showing semi-conductive behavior.

Magnetic and transport properties of the emergent ferromagnetic superlattices

LNO is a paramagnetic metal, and LFO is a G-type AFM insulator with intralayer antiparallel spin alignments, which shows one of the highest ordering temperatures of perovskite oxide ($T_N \approx 740$ K). Remarkably, when they were epitaxially grown along the (001) directions but absent for the (110) and (111) directions (Supplementary Fig. S4), the LNO_n/LFO₁ SLs show strong signatures of ferromagnetism, which depends critically on the stacking thickness, n , of LNO. Figure 2a shows the hysteresis curves of the LNO_n/LFO₁ SLs measured at 300 K, i.e. the dependence of the magnetization (M) as a function of the magnetic field B , which is saturated under the applied field above 3.5 kOe. All samples exhibited key characteristics of FM behavior, including hysteresis and remnant magnetization. At $n = 5$, the SL shows weak saturation ferromagnetism similar to LaFe_{0.5}Ni_{0.5}O₃ thin films. Remarkably, as n decreases, the highest saturation magnetization increases significantly, reaching 252.8 emu/cm³ for $n = 1$, $-4.0 \mu_B/\text{p.l.}$, comparable to the $4.96 \mu_B/\text{f.u.}$ (f.u. = formula unit) for the FM DP La₂NiMnO₆²⁸, and much larger than that of the LaFe_{0.5}Ni_{0.5}O₃ thin films (Fig. 2b). Figure 2c shows the temperature dependence of magnetization (M - T curve) for the typical LNO_n/LFO₁ ($n = 1, 3$ and 5) SL samples in addition to the LaNi_{0.5}Fe_{0.5}O₃ film. The Curie temperature, T_C , could be determined based on the minimum value of the temperature coefficient TCM, defined as $\text{TCM} = 1/M(dM/dT)$. Remarkably, all the SL

Table 1 | The emerging ferromagnetic SLs and the typical double perovskites (DP) with ferromagnetism near and above room temperature

Formula	Structure	Saturation magnetization M_s ($\mu_B/\text{p.l.}$ or f.u.)	Curie temperature, T_C (K)
LaFeO ₃ /LaCrO ₃ ⁷	1:1 SL	3.0	375
LaNiO ₃ /LaMnO ₃ ¹⁶	7:7 SL	3.7.	175
LaNiO ₃ /LaFeO ₃	1:1 SL	4.0	608
LaNiO ₃ /LaFeO ₃	3:1 SL	1.3	589
La ₂ NiMnO ₆ ²⁸	DP	4.96	280
Sr ₂ FeMoO ₆ ²⁹	DP	3.98	345
Sr ₂ CrReO ₆ ²⁹	DP	2.6	625

samples show a T_C much higher than room temperature, around $T_C = 608, 589$, and 419 K, respectively, for $n = 1, 3$, and 5 . It is also noteworthy that the LaNi_{0.5}Fe_{0.5}O₃ film shows a $T_C = 109$ K, much lower than those of the SL but higher than the bulk spin glassy temperature of 83 K²⁷. The results strongly suggest that a unique FM order is accessed in the SLs structures with the strongest magnetism obtained at $n = 1$, an emergent La₂NiFeO₆ DP structure with B -site layered ordering configuration along (001) direction. As illustrated in Table 1

its T_C is close to the highest value ($T_C = 625$ K) reported for $\text{Sr}_2\text{CrReO}_6$ among the DP compounds²⁹. The temperature-dependent resistivity, ρ , of the $\text{LNO}_n/\text{LFO}_1$ ($n=1, 3$ and 5) SL sample was also measured between 2 and 300 K, as shown in Fig. 2d. All the SLs show a semi-conductive behavior with small polaron conduction mechanism ($n \leq 4$) (Supplementary Fig. S5), although the $\text{LaFe}_{0.5}\text{Ni}_{0.5}\text{O}_3$ thin film is found to be highly insulating. This strong conduction difference, in addition to the strong difference in magnetism, may be related to the difference in the ordered arrangement of the Fe and Ni cation atoms over the B -sites, where the higher level of cations ordering prefers higher magnetization and higher conductivity¹⁹. We thus obtained an emergent $\text{La}_2\text{NiFeO}_6$ DP with Curie temperature much higher than room temperature, which has been predicted to be a half-metallic ferromagnet²⁶.

Magnetic domain microstructure characterized by magnetic force microscopy

The magnetic domain microstructure and its evolution with respect to the applied field for the $\text{LNO}_1/\text{LFO}_1$ SL were also investigated by magnetic force microscopy (MFM). Generally, the magnetograms obtained by MFM provide information on the effective magnetization along the z direction of the local domains. Figure 3 shows the magnetograms obtained at 2 K in different out-of-plane magnetic fields corresponding to the critical points on the M - H loop in Fig. 2b. Here the red or blue-colored regions represent net-magnetization oriented up ($+M_z$) or down ($-M_z$), respectively, and the green color represents the zero M_z signal. As shown in Fig. 3, the as-grown $\text{LNO}_1/\text{LFO}_1$ SL first demonstrates nearly zero-magnetization along the z direction after the zero-field-cooling (ZFC) process. By applying a positive magnetic field along the out-of-plane direction, discrete upward domains begin to appear and grow continuously (+900 Oe), eventually reaching an upward single-domain state at the saturation field of +20 kOe. By applying a negative magnetic field, the single-domain state turns to fragments of upward and downward domains at the coercive field of approximately -250 Oe, corresponding to the zero net magnetization in the M - H loop. The typical size of the fragment domains after magnetization (measured at +250 Oe) is about 300–500 nm (Supplementary Fig. S6). The magnetic domain evolution is repeatable during the field-sweeping process, as further evidenced by the downward single-domain state at -20 kOe and then the fragment-domain state at +250 Oe (the positive coercive field). The domain structures after ZFC and their evolution upon applying magnetic fields are consistent with the direct magnetization measurements. This further confirms that the strong magnetism is intrinsic to our SL samples, where the plausible magnetic contribution from the STO bulk substrate is negligible.

Discussion

The magnetic exchange interactions in $3d$ perovskite oxides occur through d orbitals (dominated mainly by e_g orbitals) centered at neighboring B cations bridged by an oxide ion, referred to as “super-exchange interaction”. The guidelines referred to as the Goodenough–Kanamori–Anderson rules can be used to predict the ferromagnetic and antiferromagnetic states for transition-metal oxides³⁰. The nominal Fe^{3+} ion in LFO has a $3d^5$ ($t_{2g}^3e_g^2$) configuration. It is AFM in accordance with the Goodenough–Kanamori rules. The nominal Ni^{3+} ion has a $3d^7$ ($t_{2g}^6e_g^1$) configuration with filled t_{2g} orbitals and one electron residing in the doubly degenerate e_g orbitals. Due to the electron-acceptor nature of LNO as illustrated in Fig. 4c, the strong ferromagnetism could originate from the electron transfer from the LFO to the LNO layer, which fills not only $\alpha|3d^7\rangle$ but also $\beta|3d^8L\rangle$ states, where the ferromagnetism at the interface could emerge from the covalent exchange coupling between Ni^{2+} and Fe^{4+} ions through the hybridization between the oxygen $2p$ band and the $3d$ band³¹.

To determine the cation valence ($\text{Fe}^{3+}/\text{Fe}^{4+}$, $\text{Ni}^{3+}/\text{Ni}^{2+}$) thus the charge transfer in addition to their spatial distribution of the Fe–Ni system³², X-ray absorption spectroscopy (XAS) measurements were

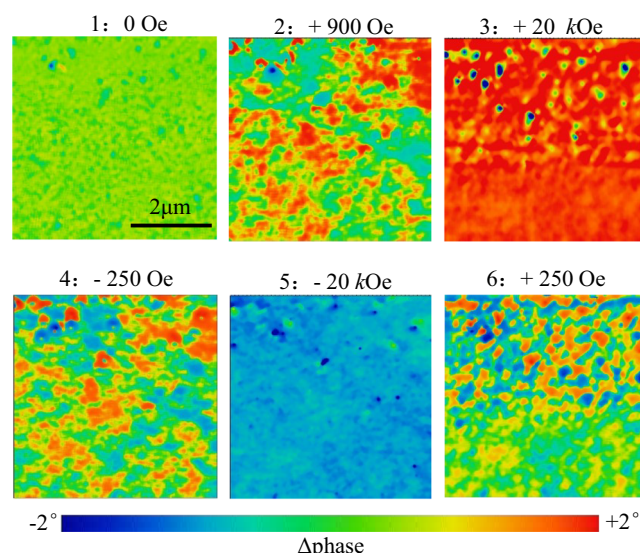


Fig. 3 | Magnetic force microscopy (MFM) domain structure of the $\text{LNO}_1/\text{LFO}_1$ SL sample obtained at 2 K. The magnetic contrasts ($\Delta\phi$) are represented by colors: the red (blue) colored area with positive (negative) $\Delta\phi$ represents the net magnetization projecting along the upward (downward) z direction, and the green colored area with nearly zero $\Delta\phi$ represents for domain walls or the grains with zero-magnetization.

performed on our $\text{LNO}_n/\text{LFO}_1$ ($n=1$ and 4) SL samples. The signal of the $\text{Fe } L_{2,3}$ edge was found to show roughly similar features between $\text{LNO}_1/\text{LFO}_1$ and $\text{LNO}_4/\text{LFO}_1$ samples, except that the splitting between the two $\text{Fe } L_3$ peaks of the $\text{LNO}_1/\text{LFO}_1$ was slightly wider than that of the $\text{LNO}_4/\text{LFO}_1$ sample (Supplementary Fig. S7). In contrast, the XAS signal at the $\text{Ni } L_{2,3}$ edge, after correction for the overlap of the $\text{La } M_{4,5}$ signal, shows a distinct difference between $\text{LNO}_1/\text{LFO}_1$ and $\text{LNO}_4/\text{LFO}_1$ (Supplementary Fig. S7). The shape of the spectra in our SL samples clearly deviates from that of the LNO film³³, which readily indicates a different formal Ni oxidation state. Particularly, as shown in Fig. 4a and b, we compared the $\text{Ni } L_2$ -edge region for the $\text{LNO}_1/\text{LFO}_1$ and $\text{LNO}_4/\text{LFO}_1$ samples. This region has no overlap with La edges and shows quite distinct features between Ni^{3+} and Ni^{2+} thus can gain insight into the Ni valence in our samples as reported previously¹¹. The $\text{Ni } L_2$ experimental spectra for the $\text{LNO}_1/\text{LFO}_1$ and $\text{LNO}_4/\text{LFO}_1$ samples were fitted with two Gaussian peaks labeled as ‘a’ and ‘b’. The changes in the spectral shape, particularly the sharpening of the low-energy feature (labeled ‘a’), are reminiscent of the signature of Ni^{2+} in the NiO ³⁴. By comparing the relative intensity of the two features (‘a’ and ‘b’), it is clear that the Ni valence is more strongly reduced towards $2+$ for the $\text{LNO}_1/\text{LFO}_1$ sample than the $\text{LNO}_4/\text{LFO}_1$ sample. These results not only confirm the charge transfer from Fe to Ni ions in the $\text{LNO}_n/\text{LFO}_1$ SL samples but also reveal a strong dependence of the number of transferred electrons on the thickness of the nickelate, where the charge transfer length is in a short-range nature and strongly confined to the proximity of the interface.

Upon charge transfer, the filling of the $\text{Ni}^{3+} \alpha|3d^7\rangle$ increases $\text{Ni}^{2+} \alpha|3d^8\rangle$ and the level of the corresponding subband will decrease. As a result, the overlapping or the hybridization between the $3d$ band and the oxygen $2p$ band $\beta|3d^8L\rangle$ will also increase, as illustrated in Fig. 4d. This rehybridization process requires a redistribution of electrons between the $|3d^7\rangle$ and the $|3d^8L\rangle$ states, which occurs at an energy cost $|\Delta|$ for each electron transferred between $|3d^8L\rangle$ and $\alpha|3d^7\rangle$, but thanks to the presence of $\alpha|3d^8\rangle$, the $|\Delta|$ is expected to be decreased after charge transfer and the rehybridization is more prone to occur. On the other hand, due to the strong covalent character of LNO with a large rare-earth size, which increases the cost of rehybridization, the competition between the charge transfer, controlled by the energy gain

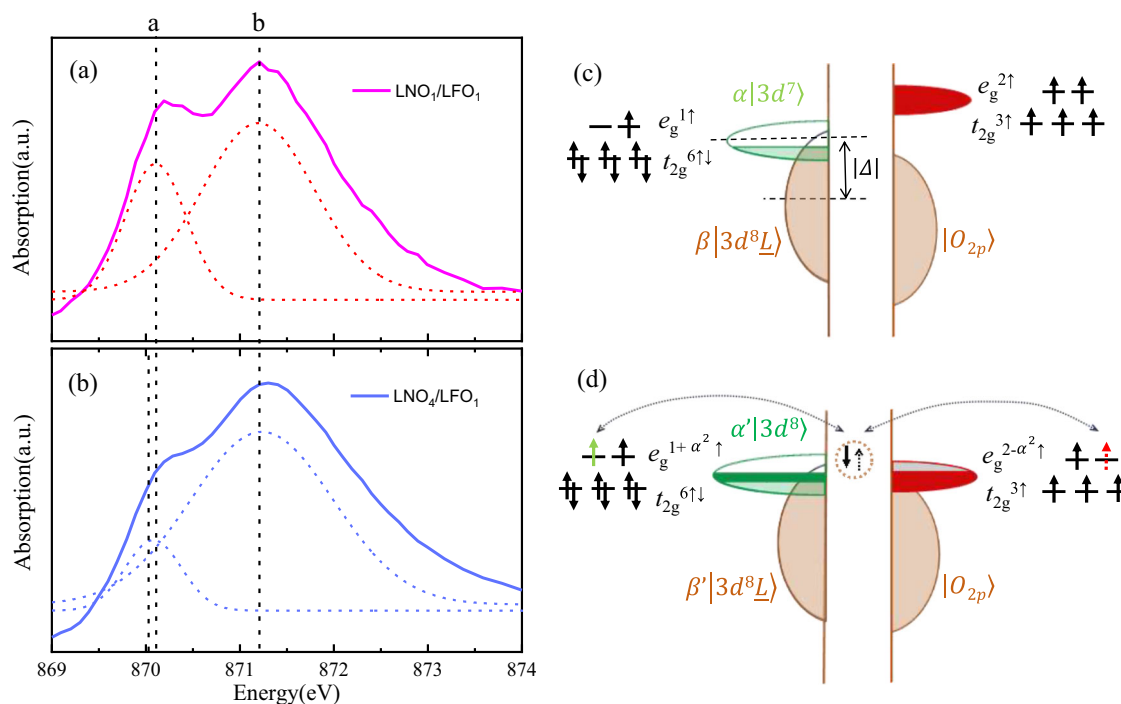


Fig. 4 | Charge transfer at the LNO_n/LFO₁ SL and a schematic representation of the covalent ferromagnetism in the LNO₁/LFO₁ SL. a and b The XAS signal of LNO₁/LFO₁ and LNO₄/LFO₁ near the Ni L₂ edge, respectively. All curves were fitted with two Gaussian functions, labeled as “a” and “b”. The fitting data were shown as

thin dashed lines. **c and d** The schematic representation of the charge state before and after the charge transfer induced rehybridization process, respectively, from LFO to LNO.

associated with the difference in electron affinity, and the cost of rehybridization, limits the number of electrons that can be transferred across the interface. Notably, changes in the multiplet splitting of the Ni L₃ absorption edge can be used to estimate the level of covalence³⁵. This splitting corresponds to the energy separation between t_{2g} and e_g levels resulting from the interplay between hybridization and Coulomb repulsion, which is both stronger for e_g levels. Previous studies have shown that the larger splitting of the L₃ peaks indicates a decrease in covalence³⁴. The fitted XAS at the Ni L₃ edge also indicates that peak splitting increases from LNO₁/LFO₁ to the LNO₄/LFO₁ samples, reflecting a decrease in covalence, where a weakened ferromagnetism is also observed. These results suggest that the charge transfer from Fe³⁺ to Ni³⁺ indeed results in an increase of covalence, favoring the ferromagnetic Ni²⁺-O²⁻-Fe⁴⁺ interactions³¹. The hybridization and charge transfer along the interfacial Fe–O–Ni bonds are mainly proposed to explain the ferromagnetism across the interface and on the Ni site. As for the magnetic nature of the LFO single layer, in contrast to the bulk G-AFM configuration, it probably exhibits a helicoidal magnetic (HM) ordering due to the negative charge transfer energy and the enhanced oxygen–oxygen hopping amplitude when it is interfaced with LNO upon charge transfer³⁶. It is noteworthy that X-ray magnetic circular dichroism (XMCD) measurements showed negligible local Fe or Ni moments at the Fe L_{2,3} and Ni L_{2,3}, although they were found to be coupled ferromagnetically to each other. As for the fact that the estimated Fe and Ni moment from XMCD is significantly lower than that measured by the SQUID magnetometry, it might result from the presence of a surface layer with a strongly reduced FM signal as the XMCD measurement was performed in total electron yield configuration, which is very surface sensitive and probes only a few nano-meter deep. Similar huge deviations have been also observed in BaFeO₃ thin films³⁷ and for LaMnO₃/LaNiO₃ superlattices¹⁶. Therefore, the exact origin of the strong ferromagnetism remains under investigation.

In summary, we have created a late transition metal oxide superlattice composed of LNO_n/LFO₁ exhibiting robust ferromagnetism with

high magnetization and Curie temperature significantly larger than room temperature by precisely controlling the charge transfer as well as the interfacial orbital hybridization. The latter is viable for tuning the strength of the exchange interactions in all-oxide heterostructures. The semiconducting and FM LNO₁/LFO₁ SL, which can be regarded as an emergent DP phase of La₂FeNiO₆ with B-site layered ordering configurations along the (001) orientation, can have a wide spectrum of applications for spintronics.

Methods

Sample fabrication and characterization

High-quality LNO_n/LFO₁ SLs were epitaxially grown on (001)-oriented STO substrates by pulsed laser deposition (KrF, $\lambda = 248$ nm) with 10–20 repetitions. During film growth, the substrate temperature was kept at 620 °C and the oxygen pressure was set to 3.4×10^{-2} mbar. The fluency of the laser pulse was 1.2 J/cm², and the repetition rate was 2 Hz. The SL growth was monitored in situ by reflection high-energy electron diffraction (RHEED). Layer-by-layer epitaxial growth was achieved for both the LNO and LFO layers. After deposition, the sample was cooled to room temperature under the oxygen pressure of 0.1 mbar. The crystal structure of the superlattice was observed by X-ray diffraction (XRD).

The magnetic properties were measured by a quantum-designed vibrating sample magnetometer (VSM-SQUID). The M – T measurements were performed in two temperature ranges from 2–300 and 300–750 K using the same system with an oven option. The transport measurement was measured by a physical properties measurement system (PPMS) at temperatures from 5 to 300 K.

MFM measurements were performed in a variable temperature system from attocube equipped with a superconducting magnet (attoDRY2100). All measurements were conducted in a vacuum at 2 K, based on a phase modulation technique in noncontact mode, with a cantilever (PPP-MFMR) with a spring constant $k = 2.8$ N/m and a resonant frequency $f = 75$ kHz.

Scanning transmission electron microscopy (STEM)

For cross-sectional microscopy, the sample was prepared by using focused ion beam (FIB) milling. Cross-sectional lamellas were thinned down to 60 nm thick at an accelerating voltage of 30 kV with a decreasing current from the maximum of 2.5 nA, followed by fine polish at an accelerating voltage of 2 kV with a small current of 40 pA. STEM measurements were conducted by a double Cs-corrected JEOL JEM-ARM200CF operated at 200 kV with a CEOS Cs corrector (CEOS GmbH, Heidelberg, Germany).

X-ray absorption spectroscopy (XAS)

Resonant X-ray reflectometry (RXR) experiments were carried out at the REIXS beamline of the Canadian Light Source at 300 K in an ultrahigh vacuum environment.

Data availability

All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data related to this paper may be requested from the authors.

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Author contributions

Y.Z.C. designed the concept and experiments. T.L.Z., F.G., and S.J.Q. fabricated the samples and performed magneto-transport characterization. Y.S.C. performed the MFM experiment and data analysis. Y.Z.H. and Q.H.Z. performed the STEM measurements and analysis. B.G.S., J.R.S., F.X.H., X.Z.H., Y.C.Z., M.H.L., and J.J.L. discussed the results and interpretations. All authors wrote the manuscript together.

Competing interests

The authors declare no competing interests.

Additional information

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