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Observation of perfect diamagnetism and interfacial effect on the electronic structures in infinite layer $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ superconductors

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Nickel-based complex oxides have served as a playground for decades in the quest for a copper-oxide analog of the high-temperature (high- T_c) superconductivity. They may provide key points towards understanding the mechanism and an alternative route for high- T_c superconductors. The recent discovery of superconductivity in the infinite-layer nickelate thin films has fulfilled this pursuit¹. Thus far, however, material synthesis remains challenging. The demonstration of perfect diamagnetism is still missing, and understanding of the role of the interface and bulk to the superconducting properties is still lacking²⁻¹⁶. Here, we synthesized high-quality $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films with different thicknesses and investigated the interface and strain effects on the electrical, magnetic and optical properties. Perfect diamagnetism is demonstrated, confirming the occurrence of superconductivity in the thin films. Unlike the thick films in which the normal-state Hall coefficient (R_H) changes signs as the temperature decreases, the R_H of films thinner than 6.1 nm remains negative, suggesting a thickness-driven band structure modification. Moreover, X-ray absorption spectroscopy reveals the Ni-O hybridization nature in doped infinite-layer nickelates, and the hybridization is enhanced as the thickness decreases. Consistent with band structure calculations on the nickelate/ SrTiO_3 heterostructure, the interface and strain effect induce a dominating electron-like band in the ultrathin film, thus causing the sign change of the R_H .

The search for the nickelate superconductivity was enthused by the idea of mimicking the $3d_{x^2-y^2}$ orbital of the single-band high- T_c cuprates¹. However, the recent results indicate the complex multiband structures in doped infinite-layer nickelates, suggesting a new family of superconductivity^{1,2}. Numerous theoretical works have been conducted based on the bulk pictures²; however, the superconductivity has only been observed in epitaxial $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ and $\text{Pr}_{1-x}\text{Sr}_x\text{NiO}_2$ ultrathin films (up to ~ 10 nm) with an infinite-layer structure^{1,3-9}. In contrast, infinite-layer nickelates prepared in bulk form show only insulating behavior¹⁰⁻¹². Moreover, the DC diamagnetic response in the superconducting thin films has never been demonstrated¹. These beg the question of whether the superconductivity occurs in the whole film or at the interface between the nickelate and SrTiO_3 (STO) substrate¹³⁻¹⁶. Theoretical calculation further proposed that the interface/surface-induced Fermi surface modification causes the transformation from a d -wave pairing in bulk into an s -wave pairing at the interface/surface^{2,17}, which might be consistent with the recent observation of two gaps from the tunneling spectrum measurement⁵. This further adds to the puzzle of whether the observed electronic properties are associated with the bulk or the heterostructure interface. Here, we comprehensively investigate the $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ films of various thicknesses to confirm the bulk nature of the superconductivity and reveal the interfacial effects on the multiband picture of the infinite-layer nickelate thin films and demonstrate their perfect DC diamagnetic response.

Figure 1(a) shows the X-ray diffraction (XRD) $\theta-2\theta$ patterns of the $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films of different thicknesses from 5.1 to 17 nm. The XRD characterization of the as-grown perovskite $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_3$ thin films can be found in **Supplementary Data Fig. S1**. The obvious diffraction peak and thickness oscillations in the vicinity of the $(00l)$

infinite-layer peak (l is an integer) indicate the high quality of the films. The $(00l)$ peak positions slightly shift towards a higher angle as the thickness increases, indicating a shrinking of the c -axis, with the lattice constants d change from ~ 3.42 Å for the 5.1-nm film to ~ 3.36 Å for the 17-nm film, as plotted in **Fig. 1(b)**. It has been shown that $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ ($x \leq 0.2$) bulks exhibit an in-plane lattice constant ($3.914 - 3.921$ Å) slightly larger than that (3.905 Å) of STO substrate^{10,11}, suggesting the presence of compressive strain on the films imposed by the substrate. The extent of compressive strain decreases as the film thickness increases and thus causing the shrinking of d . **Figure 1(c)** shows the high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) image of the 11.3-nm film. A clear infinite-layer structure is observed with no obvious defect throughout the layer.

The resistivity versus temperature (ρ - T) curves for the $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films are shown in **Fig. 2(a)**, and the zoomed-in ρ - T curves at temperatures from 50 to 2 K are shown in the inset. All the samples behave like a metal at the normal state and are superconducting at low temperatures. The onset superconducting transition temperature $T_{c,90\%R}$ (defined as the temperature at which the resistivity drops to 90% of the value at 15 K) and zero-resistance $T_{c,zero-R}$ decrease with decreasing thickness (**Fig. 2(f)**). **Figure 2(b)** shows the corresponding temperature dependence of the normal-state Hall coefficients R_H of the $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ films. The R_H for samples with a thickness higher than 6.8 nm shows a negative sign at room temperature and undergoes a smooth transition to a positive sign at a low temperature of ~ 50 K, consistent with previous observation at the doping level $x = 0.2$ ^{1,3,4}. However, as the thickness decreases to 6.8 nm, the R_H sign-change temperature decreases to 22 K. The R_H even remains negative at the whole temperature range below 300 K for the films with thickness lower than 6.8 nm. **Figure 2(e)** presents the thickness dependence of

the R_H at 20 K and 300 K, clearly showing a sign-change from positive to negative with decreasing thickness. This suggests a change of the multiband structures upon reducing thickness.

Figure 2(c) shows the temperature dependence of magnetization (M - T curves) under zero-field cooling (ZFC) mode for $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films with thickness varying from 6.1 to 17 nm. The normal-state magnetization shows temperature independence. Upon cooling down, the magnetization drops steeply to negative values, confirming the occurrence of superconducting transition in the thin films. **Figure 2(d)** shows the zoomed-in M - T curves from 20 down to 2.1 K with ZFC and field cooling (FC) for a representative sample with a thickness of 8.4 nm. Similar to the behavior of typical bulk superconductors, a clear diamagnetic transition is seen, and the onset transitions are the same under ZFC and FC modes, unambiguously confirming the existence of the superconducting phase. The ZFC and FC M - T curves for other thin films are shown in **Supplementary Data Fig. S2**. The $T_{c,\text{dia}}$, defined as the onset transition temperature in diamagnetic response, is shown in **Fig. 2(f)**. It is found that $T_{c,\text{dia}}$ is slightly lower than $T_{c,\text{zero-R}}$, suggesting the presence of inhomogeneity of the superconducting phase in the thin films. Only well below the zero-resistance temperature, the phase coherence occurs in the entire films, and therefore, the Meissner effect is observed.

The unoccupied states of energy bands are crucial to determine the transport properties and could be detected by X-ray absorption spectroscopy (XAS) on oxygen and transition metal edges. To carefully characterize the electronic structures of samples with pure perovskite phase and the resultant infinite-layer phase, we restrict our XAS measurements on thin films with thickness no more than 11.3 nm, as confirmed by XRD measurements. **Figure 3(a)** shows the O K edge XAS of 11.3-nm perovskite $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_3$ and infinite-layer $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films. A prominent

pre-peak at ~ 528.9 eV is observed near the O *K*-edge XAS in $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_3$ film, which is attributed to the presence of a ligand hole in oxygen¹⁸⁻²⁰. In perovskite nickelates, the oxygen *p* to nickel *d* band charge-transfer energy is negative, the electrons spontaneously transfer from oxygen ligands to Ni cations, leaving the holes on the oxygen side even without chemical doping¹⁸. Excitation of oxygen 1s core electrons to such unoccupied states (holes) give rise to the pre-peak in O *K*-edge XAS. The ligand holes in perovskite $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_3$ can also be suggested from the Ni *L*_{2,3} edge XAS (Fig. 3(b)), in which a shoulder is visible at ~ 856 eV corresponding to the electron transition from the Ni core-level *2p* to $3d^8\bar{L}$ state (\bar{L} is ligand hole), beside the main sharp peak at ~ 854.5 eV corresponding to the electron transition from the core-level *2p* to $3d^7$ state¹⁸. As the film is reduced from perovskite to infinite-layer structure, the prominent pre-peak in $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ film disappears, which is consistent with the previous study in underdoped $R\text{NiO}_2$ ($R = \text{La}, \text{Nd}$)^{19,20}. Instead, another pre-peak with less intensity at a higher energy of ~ 530.5 eV is observed, suggesting that the oxygen ligand hole is still present in the infinite-layer film. In the Ni *L*_{2,3} edge XAS (Fig. 3(b)), the main absorption peaks in both films are observed with a position shift to a lower energy for infinite-layer film, consistent with the reduced Ni valence state as the structures evolve from perovskite to infinite layer²¹. The shoulder is still observed in the infinite-layer $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ beside the main peak, even though it shifts to a higher energy position and its intensity is lower compared with that of the perovskite film. This further suggests the presence of oxygen ligand hole state in infinite-layer nickelates.

The parent compound of the canonical cuprate superconductor is a charge-transfer insulator according to the Zaanen-Sawatzky-Allen scheme²². The doped holes reside at the oxygen sites due to the strong hybridization of $\text{Cu-}3d_{x^2-y^2}$ and O-*2p* orbitals

forming the $3d^9L$ states, and therefore, the pre-peak of O K edge emerges upon doping²³. The spin of the doped holes in oxygen sites and spin in Cu site form the Zhang-Rice singlet state, reducing the cuprate to be an effective single-band system²⁴. Whether a similar situation occurs in infinite-layer nickelate, however, is far from clear. The theoretical model suggests that the parent compound NdNiO_2 is a Mott-Hubbard insulator, in which the O $2p$ band is below Ni $3d$ lower Hubbard Band^{25,26}. This has been suggested by the EELS and XAS measurements that the pre-peak near the O K edge is completely suppressed in undoped NdNiO_2 and LaNiO_2 ^{19,20}. The pre-peak nature in doped $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ has not been fully explored by XAS since the STO capping layer obscures the absorption from the underlying nickelate films²⁷. Our observation of the obvious pre-peak in O K edge and shoulder in Ni $L_{2,3}$ edge in $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ films, which is similar to the cuprate²⁸, suggests the Ni-O orbital hybridization in infinite-layer nickelate. Note that the EELS measurements have also shown the pre-peak feature in $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ with increasing doping, although the intensity is weak²⁰. Moreover, a prominent pre-peak has also been seen in trilayer nickelates $R_4\text{Ni}_3\text{O}_8$ ($R = \text{La}$ and Pr), which possess the same NiO_2 square plane as in the infinite-layer nickelates and an effective $1/3$ hole doping^{21,29}.

Figures 3(c) and 3(d) show O K edge and Ni $L_{2,3}$ edge XAS of infinite-layer $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ films with different thicknesses. The intensity of pre-peak in O K edge generally increases with decreasing thickness due to the decreasing occupancy (more empty states), suggesting the increase of Ni-O hybridization. The Ni $L_{2,3}$ peak intensity increases and peak position shifts systematically to higher energies as the thickness decreases. This indicates that with decreasing thickness, the Ni $2p$ core-level electrons have enhanced binding energy, implying a gradual increase of the oxidation state of Ni. The evolution of XAS may be due to the interface and strain effects with decreasing

thickness and could be related to the transport properties as shown above. It has been constructed by the bulk electronic structure calculations that the infinite-layer nickelate possesses multiband structures which show the hole pocket originating from a Ni- $3d_{x^2-y^2}$ orbital and the electron pockets from the rare-earth element $5d_{xy}$ and $5d_{3z^2-r^2}$ orbitals². Such multiband pictures are consistent with the observed change of R_H in Sr-doped NdNiO₂ and PrNiO₂^{1,3,4,6}. Our observation is that the R_H of Nd_{0.8}Sr_{0.2}NiO₂ also changes with the thickness (Fig. 2(b)), possibly related to the alteration of band structure due to the interface and strain effects.

Theoretical calculations suggested the presence of electronic and atomic reconstructions at NdNiO₂/SrTiO₃ interfaces and the resultant alteration of band structure at the interface¹³⁻¹⁶. At the surface and interface, the NiO₂ layer bends and Ni is displaced vertically due to atomic reconstruction and/or the presence of residual apical oxygen at the NdO plane. It is expected that as the thickness decreases, the interface and/or strain effects are more pronounced. Indeed, the bending of the NiO₂ layer causes the overall increase of the *c*-axis lattice constant, as indicated by the XRD measurement (Fig. 1(b)). The bending of the NiO₂ layer and the resultant tilt of Ni-O bonding causes the extra extraction of the electron from Ni to O. This is consistent with our XAS measurement that the oxidation state of Ni increases as the thickness decreases (Figs. 3(b) and 3(d)). Moreover, it has been revealed that the multiband structures become more pronounced at the interface, for example, the mixture of $d_{x^2-y^2}$ and d_{z^2} states are present near Fermi level at the interface, causing extra electron pockets¹³⁻¹⁶. Therefore, as the thickness decreases, the R_H remains negative below 300 K (Fig. 2(b)). It was proposed that the pairing state of Nd_{1-x}Sr_xNiO₂ changes from (*d*+*is*)-wave to *d*-wave pairing as the doping increases³⁰, in line with the crossover of R_H from negative to positive sign^{3,4}. Interestingly, the interface/surface effect, which

caused the negative R_H sign in our result, also induced dominant s -wave pairing¹⁷ as opposed to d -wave symmetry for the bulk^{2,31}. Overall, even though the Meissner effect is confirmed in the nickelate thin films, the interfacial effects due to the atomic reconstruction play an important role in the modification of multiband structures.

Methods

The perovskite $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_3$ thin films with different thicknesses were grown on a TiO_2 -terminated (001) SrTiO_3 (STO) substrate using a pulsed laser deposition (PLD) technique with a 248-nm KrF excimer laser. No capping layer is introduced for all samples. The deposition temperature and oxygen partial pressure P_{O_2} for all samples were 600 °C and 150 mTorr, respectively. The laser energy density on the target surface was set to be 1.8 Jcm^{-2} . After deposition, the samples were annealed for 10 min at 600 °C and 150 mTorr and then cooled down to room temperature at a rate of 8 °C/min. The as-grown samples were cut into pieces with a size of around $2.5 \times 5 \text{ mm}^2$. The pieces were then embedded with about 0.15 g of CaH_2 powder and wrapped in aluminum foil, and then placed into the PLD chamber for reduction. Using the PLD heater, the wrapped samples were heated to 340 – 360 °C at a rate of 25 °C/min and kept for 80 minutes, and then cooled down to room temperature at a rate of 25 °C/min. The wire connection for the electrical transport measurement was made by Al ultrasonic wire bonding. The electrical transport and magnetization measurements were performed using a Quantum Design Physical Property Measurement System and Superconducting Quantum Interference Device Magnetometer, respectively. The X-ray diffraction (XRD) measurement was done in the X-ray Diffraction and Development (XDD) beamline at Singapore Synchrotron Light Source (SSLS) with an X-ray wavelength of $\lambda = 1.5404 \text{ \AA}$. The XAS measurements were performed using

linearly polarized X-ray from the Soft X-ray-ultraviolet (SUV) beamline at SSLS, using a total electron yield (TEY) detection method. The incidence angle $(90 - \theta)^\circ$ of X-rays refers to the normal of the sample surface, which was varied by rotating the polar angle of the sample. The spectra were measured in a grazing-incident alignment ($\theta = 20^\circ$) to obtain better sample signals. The spectra were normalized to the integrated intensity at the tail of the spectra after subtracting an energy-independent background. The high-angle annular dark-field scanning transmission electron microscopy (HAADF-STEM) imaging was carried out at 200 kV using a JEOL ARM200F microscope, and the cross-sectional TEM specimens were prepared by a focused ion beam machine (FEI Versa 3D).

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Author Contributions

SWZ and AA conceived the project. SWZ prepared the thin films and conducted the electrical and magnetic measurements with the assistance from KH, ZH, YC, LEC, DYW, ZTZ and ZSL. SWZ, PY, CST and XMY conducted the XRD measurements. XMY, CZD, CST and ATSW conducted the XAS measurements. CJL and SJP conducted the STEM measurements. SWZ and AA wrote the manuscript with contributions from all authors. All authors have discussed the results and the interpretations.

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Figures and Captions:

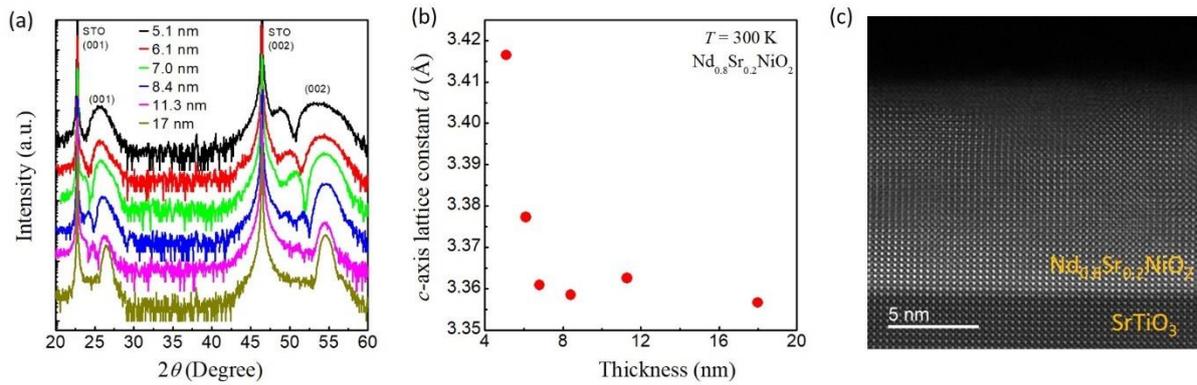


Figure 1: Thickness dependence of the infinite layer structure. (a) The XRD θ - 2θ scan patterns of the $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films with different thicknesses on SrTiO_3 substrates. The intensity is vertically displaced for clarity. (b) The room-temperature c -axis lattice constants, d , as a function of thickness, as calculated from the (001) peak positions. (c) The HAADF-STEM image of the 11.3-nm $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ on SrTiO_3 substrate.

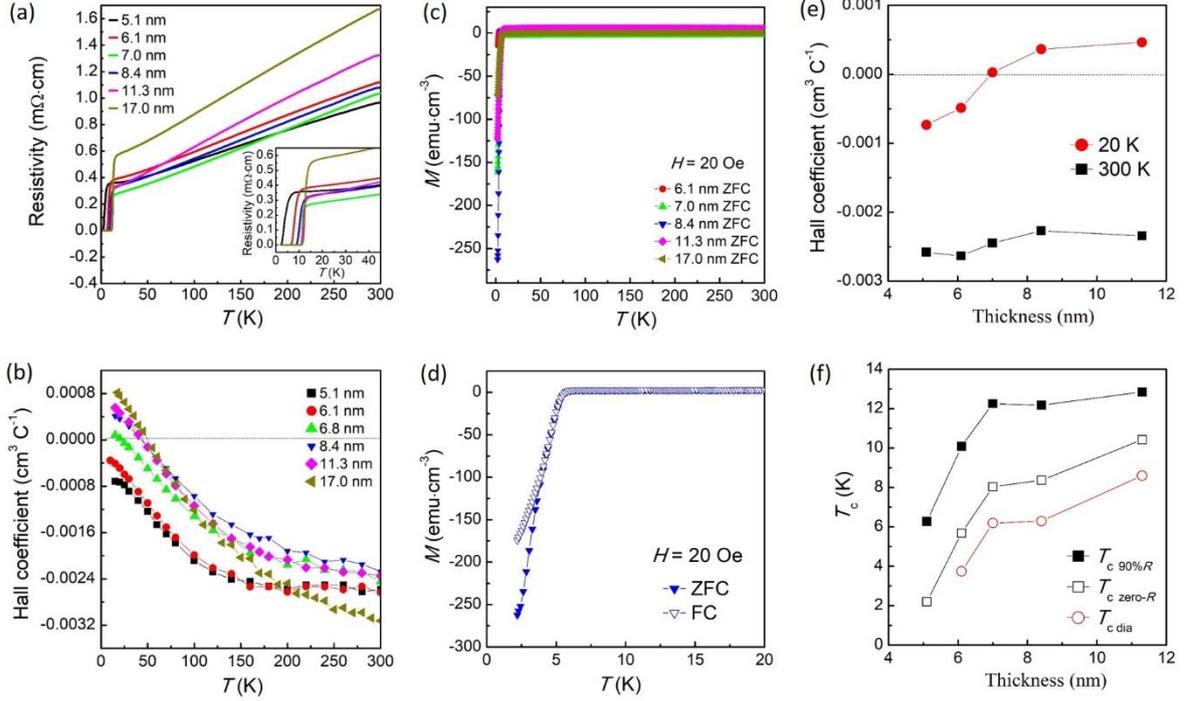


Figure 2: Thickness dependence of the transition temperature, diamagnetic response, and Hall coefficient. (a) The resistivity versus temperature (ρ - T) curves of the $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films with different thicknesses from 5.1 to 17.0 nm. The inset shows the zoomed-in ρ - T curves at temperatures from 50 to 2 K. (b) The temperature dependence of the normal-state Hall coefficients R_H . (c) The temperature dependence of magnetization (M - T curve) with zero-field cooling (ZFC) for $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films with different thicknesses from 6.1 to 17.0 nm. (d) The zoomed-in M - T curves at temperatures from 20 to 2.1 K with field cooling (FC) and ZFC for the sample with a thickness of 8.4 nm. The measurement and cooling fields are 20 Oe. The magnetic field is applied perpendicularly to the a - b plane. (e) The R_H at $T = 300$ and 20 K as a function of thickness. (f) The critical temperature, T_c , as a function of thickness. The $T_{c,90\%R}$ is defined as the temperature at which the resistivity drops to 90% of the value at 15 K (the onset of the superconductivity). The $T_{c,\text{zero-R}}$, is defined as the temperature at which the resistivity drops to be zero and $T_{c,\text{dia}}$ is defined as the temperature at the onset of the diamagnetic response. The dash lines in (b) and (e) show the position where the R_H is zero.

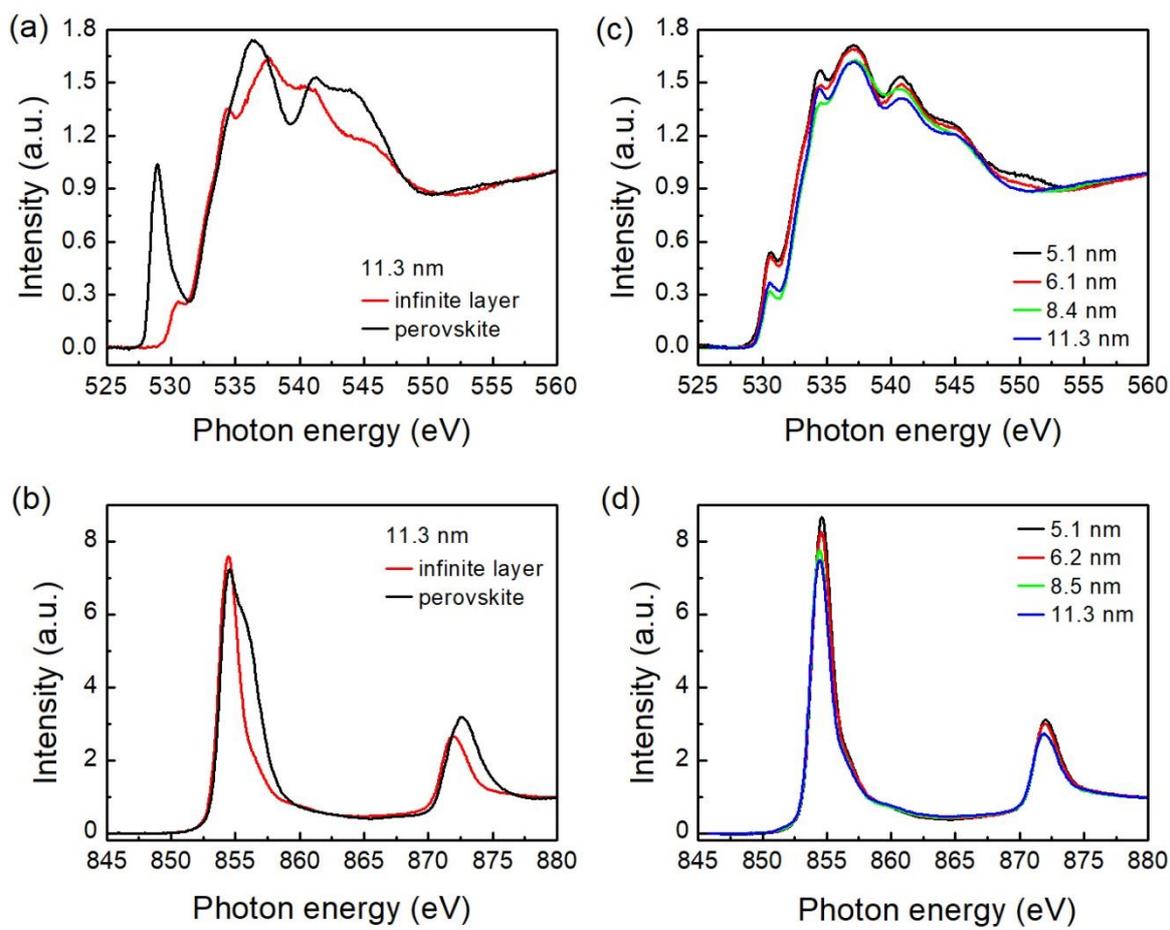
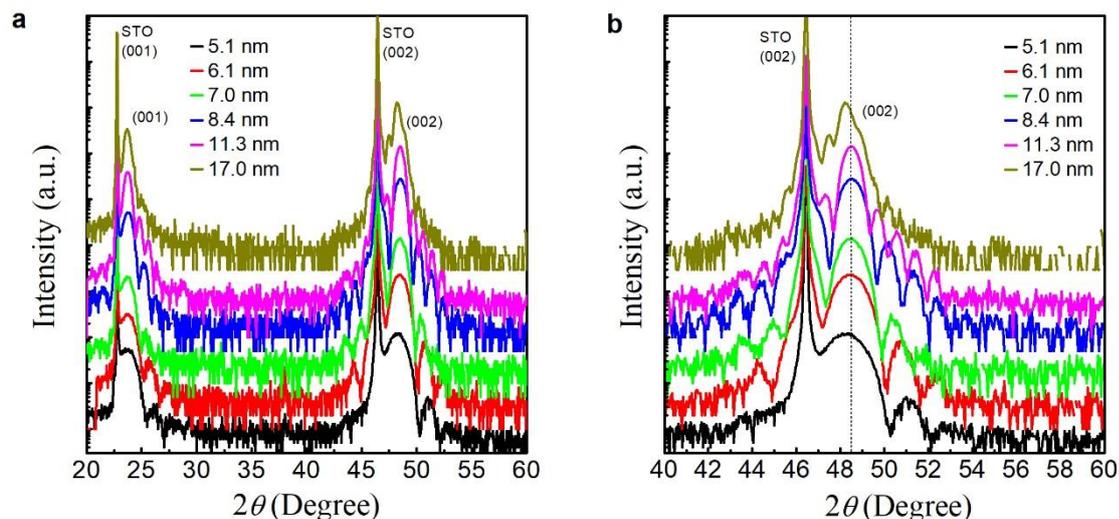
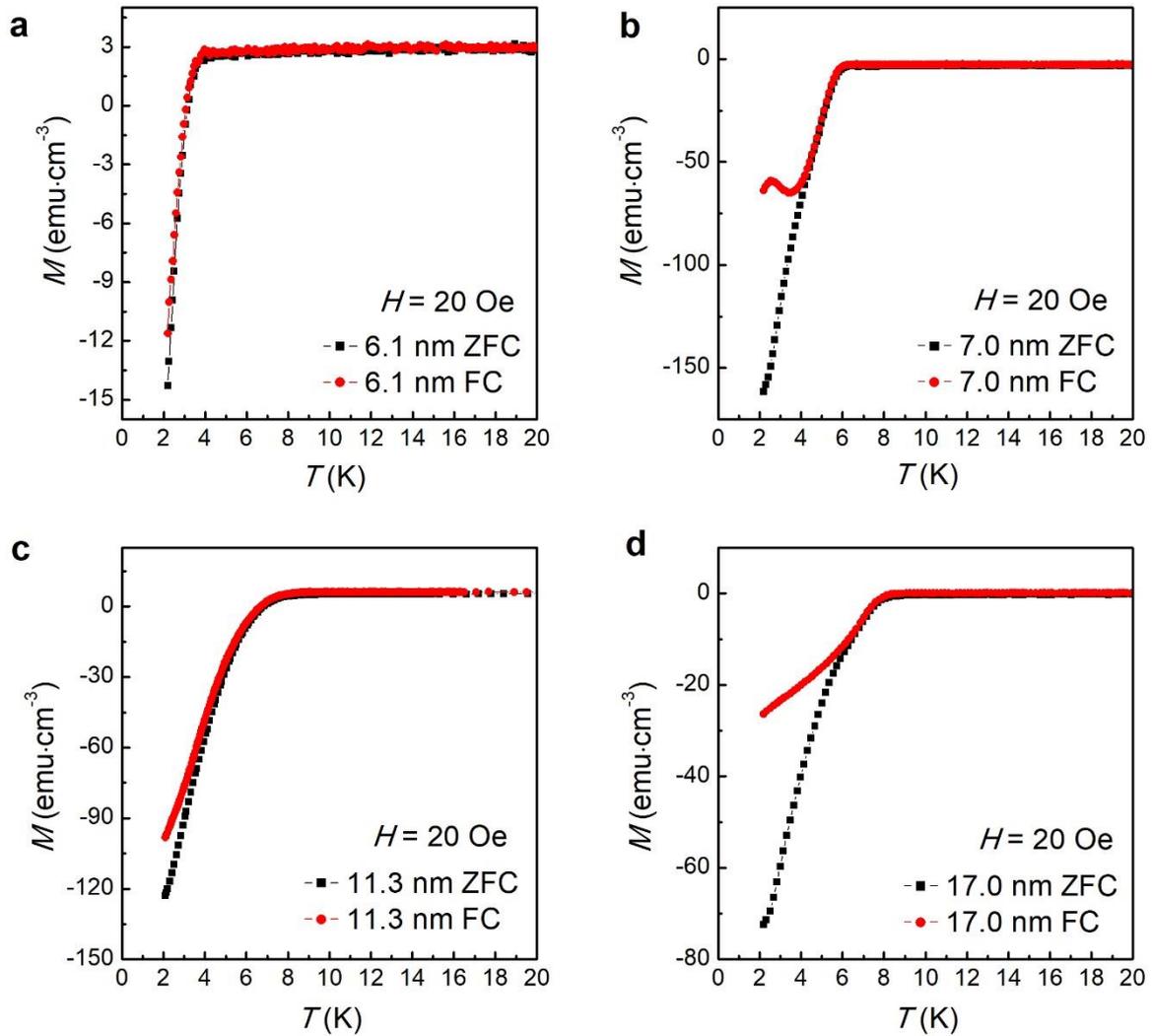


Figure 3: Thickness dependence of the X-ray Absorption Spectroscopy. (a) The O K edge and (b) Ni $L_{2,3}$ edge XAS of 11.3-nm perovskite $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_3$ and infinite-layer $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films. (c) The O K and (d) Ni $L_{2,3}$ edge of infinite-layer $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ films with different thicknesses.

Supplementary Data



Supplementary Figure S1: (a) The XRD θ - 2θ scan patterns of the as-grown $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_3$ thin films with different thicknesses on STO substrates. The intensity is vertically displaced for clarity. Only the $(00l)$ perovskite peaks are observed, where l is an integer, confirming the c -axis oriented epitaxial growth. (b) The zoomed-in XRD θ - 2θ scan patterns at angles from 40 to 60 degrees. The black dash line shows the position of the (002) perovskite peaks for thin films with different thicknesses. One can see that for thin films with thicknesses from 5.1 to 11.3 nm, the (002) peaks are at the same position, indicating the same c -lattice constant. For the film with a thickness of 17 nm, the presence of multiple peaks is seen at (002) position, suggesting the presence of mixed phases as the film thickness is increased, consistent with previous reports. The thickness is calculated through the period of thickness oscillations in the vicinity of the (002) peak.



Supplementary Figure S2: Temperature dependence of magnetization (M - T curve) with zero-field cooling (ZFC) and field cooling (FC) for $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ thin films with different thickness of (a) 6.1 nm, (b) 7 nm, (c) 11.3 nm and (d) 17 nm. The M - T curves are shown at temperatures from 20 to 2.1 K. The measurement and cooling fields are 20 Oe. The magnetic field is applied perpendicularly to the a - b plane.