Supporting Information

Dimensionality-controlled evolution of charge-transfer energy in digital nickelates superlattices

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Figure S1. (a)(b)(c)(d)(e)(f)(g) RHEED patterns of superlattices.
**Figure S2.** (a)(b)(c)(d)(e)(f)(g) AFM image of superlattices. The lower right corner is a color scale. The average surface roughness Ra of the m = 1, 2, 3, 4, 5, 7, 9 superlattices is 0.12 nm, 0.1 nm, 0.12 nm, 0.11 nm, 0.13 nm, 0.15 nm, 0.15 nm, respectively.

**Figure S3.** (a)(b)(c)(d)(e)(f)(g) Rocking curve of superlattices with XRD. The full width at half maxima (FWHM) of the m = 1, 2, 3, 4, 5, 7, 9 superlattices is 0.078°, 0.066°, 0.065°, 0.065°, 0.071°, 0.058°, 0.067°, through Gaussian fitting, respectively.
**Figure S4.** Comparison of RHEED intensity integral curves of SrTiO$_3$ and superlattice films. It clearly shows that the RHEED streak spacing of the SLs match well with those of the SrTiO$_3$ substrates, indicating the coherent growth of the films on SrTiO$_3$ substrates.

![RHEED intensity integral curves](image)

**Figure S5.** The RSM around (103) reflections for m = 4 SL. Along the horizontal axis, the film is in-plane lattice matched to the SrTiO$_3$ substrate, which confirms that the SL film grown on SrTiO$_3$ is fully strained.

![RSM](image)
Figure S6. Linear fit (red line) to two-dimensional variable range hopping, small polaron hopping, and activated conduction model for $m = 3$, 4, 5 SLs, respectively. It is difficult to find a suitable single model to fit well due to the complex resistance-temperature curve in the insulating regions for these samples.
Figure S7. (a) Resistivity versus temperature during warming and cooling for m = 5, 9 SLs. (b) The $T_{\text{MIT}}$ of the SLs series obtained by derivation $d\rho/dT = 0$ during warming and cooling.

Figure S8. (a) XAS at the Ti L edge for the SLs series. No charge transfer happened at the STO/NNO interface for the Ti cation strongly prefers the +4 oxidation state regardless of the thickness of NNO slab.