

Surface preparation method for investigating the three-dimensional electronic structure of perovskite nickelates

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The investigation of the electronic structures on perovskite oxides using surface-sensitive spectroscopy techniques is often hindered by their “uncleavable” nature, typically requiring expensive and complex *in situ* experimental setups that integrate the capabilities of sample synthesis and spectroscopy measurement under ultrahigh vacuum condition. Here, we address this challenge by developing an ozone-annealing process that yields atomically flat surfaces on perovskite oxide thin films, making them suitable for high-resolution angle-resolved photoemission spectroscopy measurements. Using this method, we present a three-dimensional electronic structure study of $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_3$ ($x = 0$ and 0.175) thin films with unprecedented accuracy. The experimentally determined low-energy fermiology exhibits quantitative agreements with two-band tight-binding simulations, which is further validated by first-principles calculations considering the material’s actual crystal structure. This work provides an accessible approach for *ex situ* ARPES measurements on perovskite oxides and other strongly correlated oxides, including the recently discovered high- T_c nickelates.

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I. INTRODUCTION

Transition-metal oxides with the perovskite structure (ABO_3) are a fascinating class of materials in condensed matter research, exhibiting a remarkable spectrum of exotic physical properties such as colossal magnetoresistance, multiferroicity, and photovoltaic effect [1–3]. This rich phenomenology arises from the intricate interplay among the spin, charge, and orbital degrees of freedom associated with the transition-metal d electrons [4,5]. Specifically, the local BO_6 octahedron, which serves as the building block of the corner-shared network within the lattice structure, plays a crucial role in determining most of the electronic and magnetic properties in perovskite oxides. Therefore, comprehensive investigations of these materials’ electronic structures are crucial for unraveling the complex interactions that govern their properties and for guiding the strategic design of new materials with tailored functionalities.

Perovskite nickelates or rare-earth nickelates, with the formula RNiO_3 ($R = \text{Pr}, \text{Nd}, \dots, \text{Lu}$), stand as a canonical example

for studying the metal-insulator transition (MIT) in strongly correlated oxides [6]. While the Ni ion is nominally $3d^7$ with one electron in the degenerate e_g orbitals, strong hybridization between O $2p$ and Ni $3d$ orbitals leads to a more accurate electronic description as $3d^8\bar{L}$, where $\bar{L}$ denotes an oxygen ligand hole [7]. The size of the R cation significantly impacts the MIT behavior, with the transition temperature varying by hundreds of degrees [8]. This drastic effect stems from a structural induced electronic property change: a smaller R cation tilts the ideal cubic NiO_6 octahedra into an orthorhombic symmetry [Fig. 1(a)], bending the Ni-O-Ni bond angle θ away from 180° . In perovskite nickelates, the Ni $3d$ orbital bandwidth W is primarily determined by the hopping parameter t_{pd} between the neighboring Ni and O atoms, as described by the relationship $W \sim t_{pd}^2 \cos^2 \theta$. Consequently, a smaller R cation leads to a reduced W , which in turn increases the charge transfer gap and raises the MIT temperature.

Angle-resolved photoemission spectroscopy (ARPES) has proven to be a powerful tool for elucidating the electronic structure of quantum materials [9]. However, the scarcity of large single crystals and the inherently “uncleavable” perovskite structure have made comprehensive ARPES measurements on bulk RNiO_3 samples extremely challenging. To overcome these difficulties, epitaxial thin films have played a vital role to explore the physical properties of RNiO_3 . The transition temperature can be precisely engineered through strain, confinement, and interface effects [10–12]. Additionally, chemical substitution effectively suppresses the MIT on

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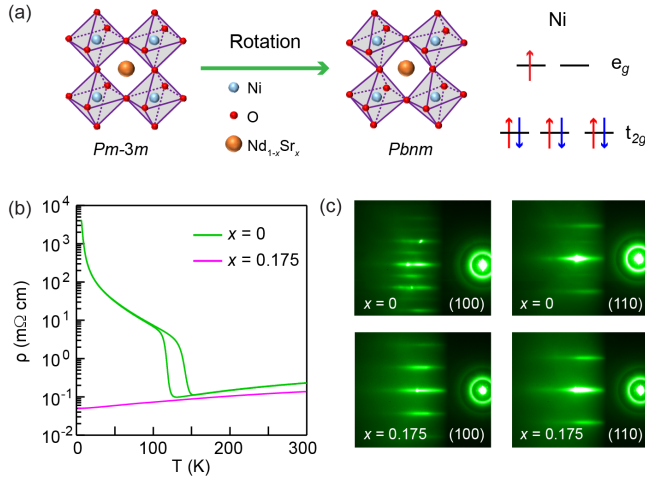


FIG. 1. Metal-insulator transition (MIT) in perovskite nickelates $RNiO_3$. (a) The smaller R cation drives a structural distortion from cubic symmetry ($Pm\bar{3}m$) to orthorhombic symmetry ($Pbnm$) in the metallic state. The Ni ion has a nominal $3d^7$ configuration with partially filled e_g orbitals. (b) Transport data for $Nd_{1-x}Sr_xNiO_3$ ($x = 0, 0.175$) thin films. (c) RHEED patterns of ozone-annealed $Nd_{1-x}Sr_xNiO_3$ ($x = 0, 0.175$) thin films. The 2×2 reconstruction on the undoped sample is possibly related to a polarized surface termination.

both electron and hole doping sides, providing a versatile platform to modulate the physical properties of $RNiO_3$ by tuning d -electron correlation strength [13]. While the three-dimensional electronic structure of $RNiO_3$ thin films has been reported in previous literature [14–16], a quantitative experiment-theory comparison remains elusive, especially for the doped samples. Hence, a detailed study of the fermiology and electron dynamics is urgently required to fully unravel the rich physics of the perovskite nickelates.

In this article, we present an ozone-annealing process developed to achieve atomically flat oxide surfaces, a crucial step for *ex situ* ARPES measurements. Applying this method on perovskite nickelates, we conducted a high-resolution ARPES study of the hole-doped $Nd_{1-x}Sr_xNiO_3$ ($x = 0$ and 0.175) thin films. The three-dimensional Fermi surface was clearly mapped by performing photon-energy dependent measurements. We quantitatively reproduce the observed Fermi surface topology and low-energy band structure using a two-band tight binding model, suggesting that the metallic phase of $Nd_{1-x}Sr_xNiO_3$ is essentially captured by Fermi liquid theory. Furthermore, these findings are complemented by first-principles calculations that demonstrate the strong hybridization between O $2p$ and Ni $3d$ orbitals, supporting the negative charge transfer scenario for this system.

II. METHODOLOGY

$Nd_{1-x}Sr_xNiO_3$ ($x = 0, 0.175$) epitaxial thin films were synthesized by the pulse-laser deposition method on LSAT (001) substrates [17]. The film thickness is about 6 nm. Figure 1(b) displays the transport measurements on $Nd_{1-x}Sr_xNiO_3$ thin films. The resistivity curve of the $x = 0$ sample shows a thermally driven MIT with a hysteretic loop,

confirming the first-order phase transition. Chemical substitution of Nd with Sr suppresses the insulating ground state, providing a new knob to control the transition at low temperature regime. After the growth, the samples were transferred in air to a commercial oxide molecular beam epitaxy system (Veeco GEN930), which is *in situ* connected to the ARPES end station at Stanford Synchrotron Radiation Lightsource beamline 5-2 equipped with a Scienta DA30 analyzer. We annealed the thin films at 500 °C for 20 min under a base pressure of 1×10^{-5} Torr purified ozone. Considering the surface sensitivity of ARPES technique, we used reflection high-energy electron diffraction (RHEED) to monitor the surface quality during the ozone-annealing process [Fig. 1(c)]. The 2×2 surface reconstruction on the undoped sample arises from the catastrophe effect of a polarized termination [18]. Hole dopants neutralize this charge imbalance, recovering the 1×1 RHEED pattern for the $x = 0.175$ sample. After ozone annealing, all samples have sharp RHEED streaks, which are very important to obtain high-quality data in the following ARPES measurements. The base pressure during the ARPES measurements was better than 3×10^{-11} Torr. Linear-horizontally polarized light within a photon energy range 60–180 eV was used to explore the three-dimensional electronic structure. The measurement temperature was 10 K on the $x = 0.175$ sample, and 180 K on the $x = 0$ sample.

First-principles calculations for electronic structures were performed in the framework of density functional theory (DFT) using projected augmented wave (PAW) pseudopotentials [19] as implemented within the Vienna *ab initio* simulation package (VASP) [20]. The calculations employed the generalized gradient approximation (GGA) using the PBE exchange-correlation functional [21], as well as the DFT + U approach using the spherically averaged form of the rotationally invariant effective U parameter [22]. The electronic structure calculation as shown in the main text is implemented at A-type AFM structure with $U = 2.0$ eV, 4.0 eV, 6.0 eV, and 8.0 eV on the correlated Ni $3d$ orbitals. A $24 \times 24 \times 12$ Monkhorst-Pack grid centered at the Γ point is used in the self-consistent calculation [23]. We implemented the method of Methfessel-Paxton of order four for smearing, with $\sigma = 0.2$ eV. For the cubic structure calculation, we chose a $24 \times 24 \times 24$ Monkhorst-Pack grid centered at the Γ point and $U = 1.5$ eV for the DFT + U calculation.

III. THREE-DIMENSIONAL FERMI SURFACE

Figure 2(a) presents a schematic diagram of the three-dimensional Fermi surface for perovskite nickelates in the reference cubic structure, consisting of an electron pocket centered at the Γ point and a hole pocket centered at the A point. Experimentally, obtaining the full four-dimensional information (k_x, k_y, k_z, E) is crucial for constructing the three-dimensional electronic structure. In ARPES measurements, the in-plane momentum $k_{\parallel}$ and the out-of-plane momentum k_z are determined by the following relations:

$$|k_{\parallel}| = \frac{\sqrt{2m(h\nu - \phi - E_B)} \sin \theta}{\hbar}, \quad (1)$$

$$k_z = \frac{\sqrt{2m[(h\nu - \phi - E_B) \cos^2 \theta + V_0]}}{\hbar}, \quad (2)$$

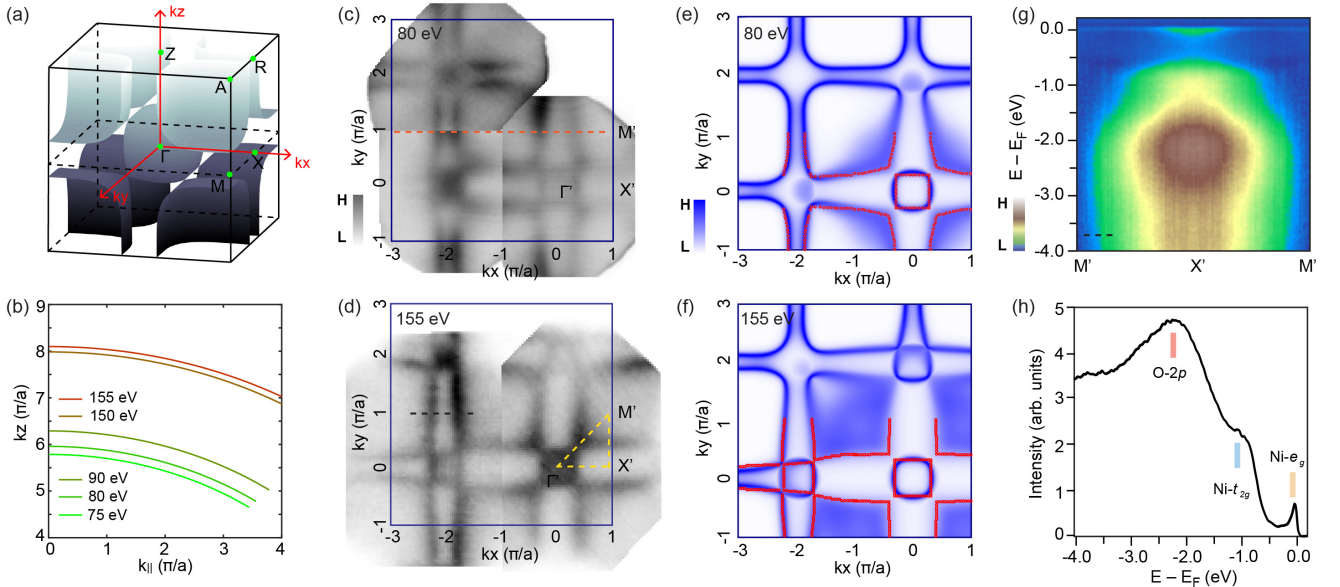


FIG. 2. Three-dimensional Fermi surface of $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_3$ ($x = 0.175$) thin film measured at 10 K. (a) Schematic representation of the Fermi surface with pockets centered at Γ and A. (b) $k_z - k_{\parallel}$ relations at different photon energies $h\nu = 75$ eV, 80 eV, 90 eV, 150 eV, and 155 eV. (c), (d) Experimental Fermi surfaces at $h\nu = 80$ eV and 155 eV, integrated over a 20 meV window around E_F . (e), (f) Simulated Fermi surfaces for comparison with the experimental data in (c) and (d). Red circles denote experimental k_F values from MDC analysis. (g) $E - k$ dispersion of the valence band along the second BZ boundary as indicated by the black dashed line in (d). (h) EDC spectrum of the valence band, showing contributions from $\text{Ni-}e_g$ (near E_F), $\text{Ni-}t_{2g}$ (~ 0.8 eV), and $\text{O-}2p$ (~ 2.3 eV) orbitals.

where m is the free electron mass, $h\nu$ is the incident photon energy, ϕ is the analyzer work function, E_B is the binding energy, and θ is the polar angle with respect to the surface normal. V_0 is the inner potential, which was determined to be 12 eV in this study by analyzing the experimental data and considering the periodicity of the Brillouin zone (BZ).

By varying the incident photon energy, we can probe different slices of the three-dimensional Fermi surface [Fig. 2(b)]. The variation of the k_z value across neighboring BZs can be as large as $0.2\pi/a$, where a is the in-plane lattice constant. Figures 2(c) and 2(d) display the two-dimensional projected Fermi surfaces obtained at $h\nu = 80$ eV and 155 eV, respectively. For clarity, we label the high symmetry points in these projected planes as Γ' , X' , and M' to distinguish them from the points in the $k_z = 0$ plane. Benefitting from the high-resolution spectroscopy, we can now obtain many new aspects of the Fermi surface structure that were not reported in previous studies [14–16]. For example, we can clearly map the electron pocket around the Γ point, as well as the asymmetric Fermi surface features at higher-order BZs, which arise from the variation of the k_z value along the $k_{\parallel}$ direction.

To parametrize the Fermi surface, we employ a two-band tight binding model [24], described by the Hamiltonian

$$\mathcal{H}_{tb} = - \sum_{i,j} \sum_{a,b} t_{ij}^{ab} c_{ia}^\dagger c_{jb} - \mu, \quad (3)$$

where i, j are site indices, $a, b = 1, 2$ are orbital indices corresponding to the two e_g states ($d_{x^2-y^2}$ and d_{z^2} orbitals), and $c_{ia}^\dagger$ and c_{jb} are creation and annihilation operators, and μ is the chemical potential. Our model considers only nearest-neighbor ($t_{\pm ax}, t_{\pm ay}, t_{\pm az}$) and next-nearest-neighbor ($t_{\pm ax\pm ay},$

$t_{\pm ax\pm az}, t_{\pm ay\pm az}$) hopping processes. The hopping matrices are given by

$$t_{\pm ax} = \frac{t_1}{4} \begin{pmatrix} 1 & -\sqrt{3} \\ -\sqrt{3} & 3 \end{pmatrix}, \quad t_{\pm ax\pm ay} = \frac{t_2}{2} \begin{pmatrix} 1 & 0 \\ 0 & -3 \end{pmatrix}, \quad (4a)$$

$$t_{\pm ay} = \frac{t_1}{4} \begin{pmatrix} 1 & \sqrt{3} \\ \sqrt{3} & 3 \end{pmatrix}, \quad t_{\pm ax\pm az} = \frac{t_2}{2} \begin{pmatrix} -2 & \sqrt{3} \\ \sqrt{3} & 0 \end{pmatrix}, \quad (4b)$$

$$t_{\pm az} = t_1 \begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}, \quad t_{\pm ay\pm az} = \frac{t_2}{2} \begin{pmatrix} -2 & -\sqrt{3} \\ -\sqrt{3} & 0 \end{pmatrix}. \quad (4c)$$

Here, t_1 and t_2 are the hopping parameters. The basis vectors $[1, 0]$ and $[0, 1]$ represent the d_{z^2} and $d_{x^2-y^2}$ orbitals, respectively. We compute the ARPES intensity $I(E, k_x, k_y)$ by averaging over k_z using a Lorentzian convolution with a full width at half-maximum (FWHM) given by $\Delta k_z = 1/\lambda$:

$$I(E, k_x, k_y) = \int_{-\Delta k_z/2}^{\Delta k_z/2} f(E) \frac{A(E, k)}{(k_z)^2 + (\Delta k_z/2)^2} dk_z. \quad (5)$$

Here, $f(E)$ is the Fermi-Dirac function, $\lambda = 5 \text{ \AA}$ is the photoelectron mean free path, and $A(E, k)$ is the spectral function:

$$A(E, k) = \frac{1}{\pi} \frac{\Sigma''(E)}{(E - \epsilon(k) - \Sigma'(E))^2 + (\Sigma''(E))^2}, \quad (6)$$

where $\epsilon(k)$ is the electron band energy at momentum k , and $\Sigma'(E)$ and $\Sigma''(E)$ are the real and imaginary parts of the self-energy, respectively. For our simulations, we chose $\Sigma''(E) = 50$ meV. All parameters used in the model are listed in Table I.

The simulated Fermi surfaces at $h\nu = 80$ eV and 155 eV are displayed in Figs. 2(e) and 2(f). To better benchmark the theoretical model with the experiment, we performed

TABLE I. Tight-binding parameters.

t_1	t_2	μ ($x = 0.175$)	μ ($x = 0$)
0.18 eV	$0.1 t_1$	$3.7 t_1$	$3.44 t_1$

momentum-distribution-curve (MDC) analysis to extract the experimental Fermi momenta k_F , which are indicated by the red circles in Figs. 2(e) and 2(f). A good consistency is observed between the model simulations and the experimental data in the low-energy range. Furthermore, we provide additional Fermi surface simulations at $h\nu = 75$ eV, 90 eV, and 150 eV in Fig. 3. These simulations also show good agreements with the corresponding experimental results. Figures 2(g) and 2(h) show the energy-momentum ($E - k$) image plot and the corresponding energy-distribution curve (EDC) of the valence band, respectively. The sharp peak near the Fermi level is primarily contributed by the Ni- e_g states, while the shoulder feature around ~ 0.8 eV and the broad peak with an onset at ~ 2.3 eV originate from the Ni- t_{2g} and the O- $2p$ states, respectively [25].

In addition to the $x = 0.175$ sample, we also investigated the Fermi surface contours of the $x = 0$ sample, as shown in Fig. 4. The basic topology is nearly the same for both samples, consisting of a small electron pocket at the Γ point and a large hole pocket around the A point. However, subtle differences exist in the sizes of these pockets between the two samples. Applying the Luttinger theorem, we can estimate the doping levels by counting the filled states enclosed by the Fermi surface pockets. Our calculations yield doping levels of $x_L = 0.006$ for the parent compound and $x_L = 0.172$ for the doped sample (see Table II for more details). These calculated values show good consistency with the nominal doping concentrations. Furthermore, our analysis reveals that the hole

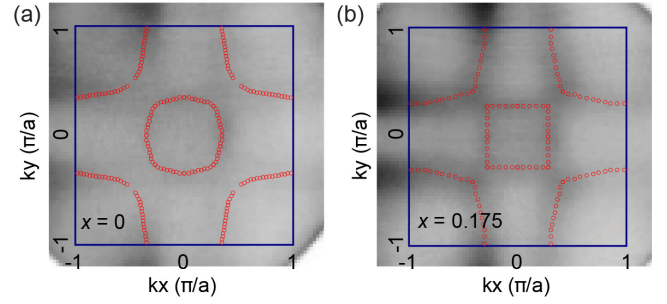


FIG. 4. Fermi surface contours for $x = 0$ (a) and $x = 0.175$ (b) samples at $h\nu = 80$ eV. Red circles denote experimental k_F values from MDC analysis.

pocket is significantly affected by Sr doping, whereas the electron pocket appears less sensitive to carrier concentration.

Thermodynamic measurements, such as the negative Seebeck coefficients, imply the prevalence of electron-type charge transport in the metallic phases of perovskite nickelates [26]. Specifically, our ARPES measurements determined an effective electron concentration of 0.05–0.06 e/Ni atoms in the $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_3$ thin films, which is consistent with the characteristics of a bad metal [27]. It is important to note that epitaxial strain can effectively change the electronic properties of perovskite nickelates. For instance, a previous study on the NdNiO_3 thin film grown on LaAlO_3 substrates reported a larger hole pocket compared to our findings, along with an additional small hole pocket near the M point [15]. The mechanism of the compressive strain in tuning the MIT requires more investigations in the future.

IV. LOW-ENERGY ELECTRONIC STRUCTURE

The intrinsic three-dimensionality of the perovskite structure significantly affects the band dispersions at different k_z values. For instance, Fig. 5(a) displays the experimental $E - k$ image plot along the BZ boundary at $h\nu = 80$ eV. Although the variation of k_z substantially changes the bandwidths in the first and second BZs, the tight binding simulation impressively reproduces the essential features of the band structures with quantitative consistency [Fig. 5(b)], implying the feasibility of the two-band model to capture the low energy physics in perovskite nickelates. Moreover, the high-resolution spectroscopy cuts at $h\nu = 150$ eV allow us to simultaneously observe the degenerate e_g bands along the diagonal direction of the BZ, as shown in Fig. 5(c). Again, most of the essential features in the experimental data are well described by the model simulations [Fig. 5(d)]. For example, the d_{z^2} orbital contributes predominantly to the hole pocket, while the electron pocket comes from the hybridization of the two e_g orbitals. The residual intensity in the high energy regime, often referred to as the “waterfall” feature, is suggested to

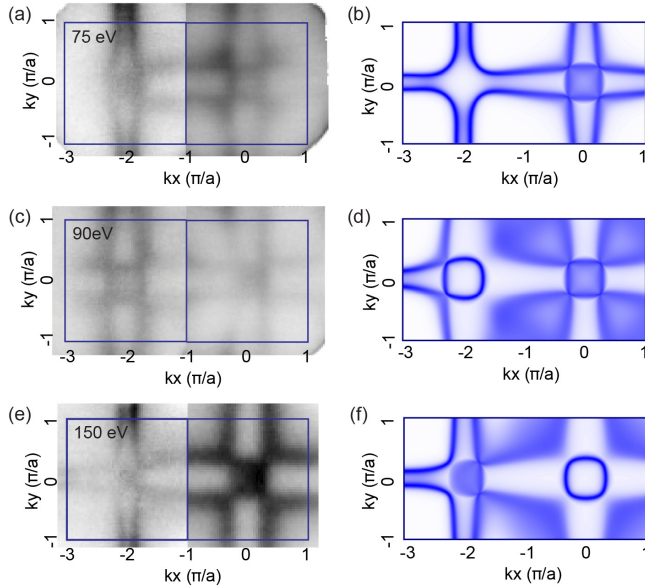


FIG. 3. Experimental and simulated Fermi surfaces at various photon energies. (a), (b) Experimental and simulated Fermi surfaces at $h\nu = 75$ eV. (c), (d) Experimental and simulated Fermi surfaces at $h\nu = 90$ eV. (e), (f) Experimental and simulated Fermi surfaces at $h\nu = 150$ eV.

TABLE II. Luttinger theorem analysis.

Sample	Hole pocket	Electron pocket	Total
$x = 0.175$	21.74%	4.48%	17.26%
$x = 0$	6.71%	6.15%	0.56%

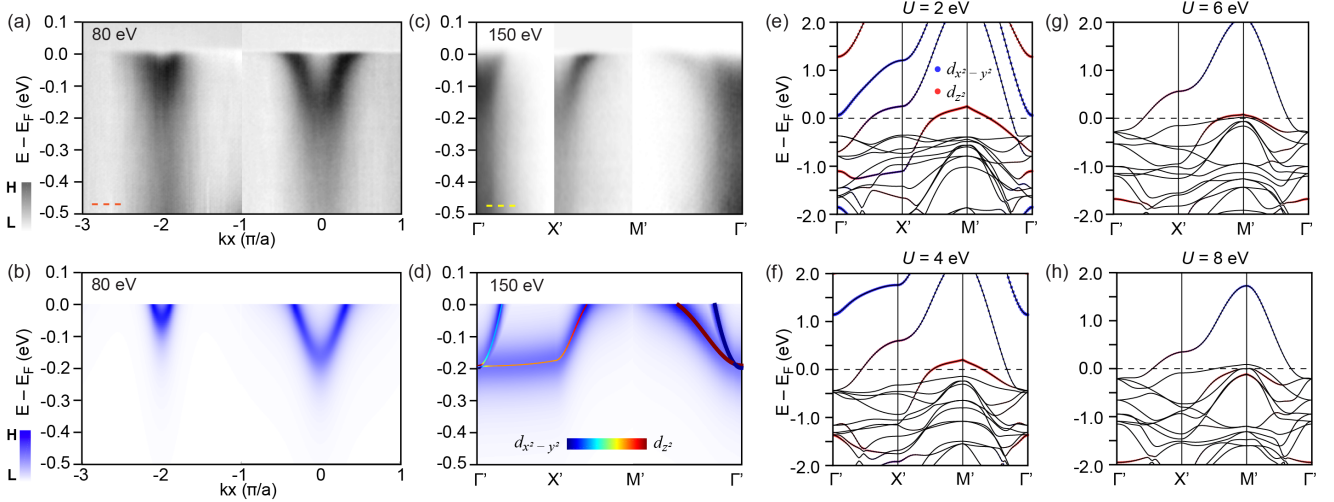


FIG. 5. Low-energy electronic structure of $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_3$ ($x = 0.175$) thin film measured at 10 K. The momentum directions correspond to the same color lines indicated in Fig. 2. (a), (b) Experimental and simulated $E - k$ plots along the BZ boundary as shown by the orange dashed line in Fig. 2(c). (c) Experimental $E - k$ plot along high-symmetry cuts as shown by the yellow dashed line in Fig. 2(d). (d) Simulated $E - k$ plot along the same cuts as in (c). The color scale represents the relative contributions of d_{z^2} and $d_{x^2-y^2}$ orbitals. (e)–(h) DFT band structure calculations along the cuts in (c) for varying Hubbard U values: $U = 2$ eV (e), $U = 4$ eV (f), $U = 6$ eV (g), and $U = 8$ eV (h).

arise from many-body effects, a phenomenon extensively discussed in studies of high- T_c cuprates and other correlated oxides, although a full consensus on its nature has not yet been reached [28,29].

In our tight binding simulations, we initially assumed ideal cubic symmetry for the perovskite nickelates. However, the thin films experimentally investigated possess a distorted orthorhombic structure. To account for the materials' actual crystal symmetry, we performed first principles calculations using the DFT + U method. Figures 5(e)–5(h) display the DFT results along the same momentum cuts as in Fig. 5(c), calculated with different values of the on-site Hubbard U parameters. Although qualitative agreement is achieved between the DFT calculations and the experimental data, the bandwidths obtained from DFT are notably larger than both the experimental measurements and the tight binding simulations.

To determine an appropriate value for U , we compared the positions of the experimental Fermi momenta along different cuts with the DFT results. This method allows us to identify $U = 4$ eV as a suitable value. Using this determined U , we further analyzed the orbital-dependent correlation effects by examining the bandwidth renormalizations. We find that the measured electron-type and hole-type carriers are renormalized by factors of approximately 2.2 and 4.6, respectively, to match the calculated band dispersions. The results are consistent with a previous study on LaNiO_3 , which reported a renormalization factor of about three [25].

V. DISCUSSION AND CONCLUSION

In perovskite nickelates, the strong hybridization between the Ni- $3d$ and O- $2p$ orbitals leads to a situation where the charge transfer energy is small or even effectively “negative.” This energetic alignment implies that the O- $2p$ levels are close to or above the Ni- $3d$ levels, resulting in significant covalent mixing. Consequently, the ground state is a mixture of the purely ionic $3d^7$ configuration and the

ligand-hole $3d^8\bar{L}$ configuration. Despite the presence of ligand and holes residing on the oxygen orbitals, the low-energy physics of perovskite nickelates can be quantitatively described by a minimal effective two-band model that retains the degenerate Ni- e_g character. The oxygen states should be considered for the high-energy related questions in this system.

It should be noticed that we did not experimentally observe the nondispersive band along the Γ' to X' direction in Fig. 5(d), a feature predicted by both tight-binding simulations and DFT calculations. There are two possible reasons for this discrepancy. First, the tight binding model assumes cubic symmetry, whereas the real material has orthorhombic symmetry. The reduced lattice symmetry could potentially shift this nondispersive band away from the Fermi level. Indeed, the DFT calculations on a cubic structure show degenerate nondispersive bands from Γ to X in Fig. 6, which aligns well with the tight-binding simulations. Second, the nondispersive band in DFT calculations is mostly contributed by the O- $2p$ orbital, which is not sensitive to the experimental photon energies $h\nu = 60 - 180$ eV. To definitively illustrate the contribution of oxygen orbitals, future investigations employing soft x-ray ARPES would be beneficial.

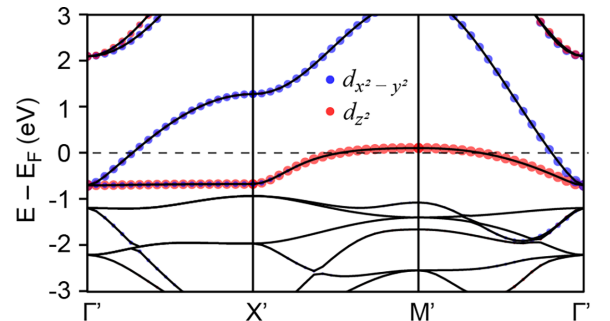


FIG. 6. DFT calculations in the cubic structure.

In summary, we quantitatively determined the three-dimensional low-energy electronic structure of perovskite nickelates by simulating the experimental data using a two-band model. A key contribution of this work is the development of a simple and reliable method for preparing clean surfaces, which is essential for surface-sensitive spectroscopic techniques. This proven recipe significantly broadens the possibilities for *ex situ* ARPES studies of other transition metal oxides, including perovskite oxides like SrFeO_3 , $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$, and the recently discovered high- T_c nickelates.

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DATA AVAILABILITY

The data that support the findings of this article are openly available [30].

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