

Electronic Structure of Compressively Strained Bilayer Nickelate Thin Film

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The discovery of superconductivity in bulk bilayer nickelates under high pressure, and its subsequent stabilization in compressively strained thin films at ambient pressure, has established a new platform for exploring high- T_c superconductivity beyond the cuprates. Central to this development is the prominent role of the $3d_z^2$ orbital in shaping the low-energy electronic structure, imposing constraints on microscopic theories and fueling debate over the superconducting mechanism. Here we report a systematic *in situ* angle-resolved photoemission spectroscopy study of compressively strained bilayer nickelate thin films spanning Ca doping, oxygen stoichiometry and film thickness. Despite variations in oxygen-vacancy disorder and surface termination, the electronic structure remains robust and exhibits a systematic strain-driven evolution consistent with an intermediate-correlation regime. In particular, we demonstrate a Ca-doping-induced electronic structure evolution that is consistent with Fermi-level crossing of the γ band and is decoupled from the presence of superconductivity, suggesting that, while important for the γ band to be near the Fermi level, the debated γ crossing and the resulting Fermi pocket may not be a prerequisite for superconductivity. Together, our results establish key spectroscopic constraints on the minimal fermiology and correlation strength relevant to superconductivity in bilayer nickelates, providing an experimental foundation for microscopic theories of their pairing mechanism.

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I. INTRODUCTION

Since the discovery of high-transition-temperature (T_c) superconductivity in cuprates [1], extensive efforts have sought electronic and structural analogs among transition-metal oxides [2–6]. The recent observations of superconductivity near 80 K in bulk bilayer $\text{La}_3\text{Ni}_2\text{O}_7$ (LNO) and near 30 K in trilayer $\text{La}_4\text{Ni}_3\text{O}_{10}$ under pressure have established a new platform for identifying the essential ingredients of high- T_c superconductivity [7,8]. A growing body of work suggests that straightening the out-of-plane Ni–O–Ni bond is critical for stabilizing superconductivity [9]. However, how this structural change reshapes the

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electronic structure remains unresolved [7,10,11]. Early experimental and theoretical studies of nonsuperconducting bulk LNO revealed a flat γ band with predominantly Ni $3d_{z^2}$ character and a large density of states just below the Fermi level within an otherwise quasi-two-dimensional band structure [12–20]. Under pressure, this γ band has been proposed to cross the Fermi level and promote superconductivity through its large density of states, distinguishing the bilayer nickelate from the $3d_{x^2-y^2}$ -dominated electronic structure of the cuprates [14–24]. Other theoretical studies, however, have suggested that superconductivity may emerge even in the absence of a γ -band Fermi-surface pocket [25–28]. Direct experimental resolution of this issue has remained challenging because superconductivity in bulk LNO emerges only under large hydrostatic pressure.

In this context, the recent stabilization of superconductivity in bilayer nickelate thin films at ambient pressure offers a direct route to probing the superconducting mechanism [11,29,30]. In these systems, superconductivity is realized through large ($\sim 2\%$) compressive epitaxial strain, which has been argued to straighten the apical Ni–O–Ni bond and enhance the crystal symmetry, analogous to the effect of hydrostatic pressure in bulk [31–33]. Comparing the electronic structure of superconducting strained thin films with that of nonsuperconducting ambient-pressure bulk samples therefore offers a means to identifying the key electronic ingredients governing superconductivity and resolving the debate surrounding the necessity of a γ pocket. Theory predicts that compressive epitaxial strain drives systematic modifications of the band structure: at $\sim 2.5\%$ strain, the γ band is expected to move away from the Fermi level by up to ~ 350 meV [32–34], whereas the α and β bands with predominantly in-plane $3d_{x^2-y^2}$ character may shift towards lower binding energy [32].

At the same time, the electronic correlation—parametrized by the ratio between the on-site Coulomb interaction U and the electronic hopping amplitude—may also evolve substantially, as both the straightening of the out-of-plane bond and the reduced in-plane lattice constant modify the underlying hopping parameters. Determining where compressively strained bilayer nickelate thin films reside in terms of correlation strength is therefore central to understanding their superconducting mechanism [35]. In the weak- U limit, the band structure is largely captured by density functional theory (DFT), which generally places the γ band close to or crossing the Fermi level. In this regime, superconducting pairing arises from spin or charge fluctuations [14–17,24,36–43]. In the strong- U limit, by contrast, the $3d_{z^2}$ orbitals lose their bonding–antibonding character and instead form localized moments coupled through interlayer super-exchange interactions. In this regime, the γ band shifts to larger binding energies and superconductivity is then more naturally described by t - J

and related models that incorporate interlayer coupling in the $3d_{z^2}$ channel, together with interorbital Hund’s coupling [17,18,21,22,25–27,44–50]. Because these two limits yield qualitatively distinct electronic structures and pairing scenarios, direct experimental characterization of the electronic structure in superconducting bilayer nickelate thin films is essential for constraining microscopic theories of their superconducting mechanism.

To address this experimental challenge, we synthesized compressively strained superconducting $\text{La}_2\text{PrNi}_2\text{O}_7$ (LPNO) and Ca-doped $(\text{La}, \text{Pr}, \text{Ca})_3\text{Ni}_2\text{O}_7$ (LPCNO) thin films of varying thickness using oxide molecular beam epitaxy (MBE), and characterized their electronic structure *in situ* with a directly coupled synchrotron angle-resolved photoemission spectroscopy (ARPES) end station. To exclude synthesis-specific artifacts, we additionally examined LPNO films of identical composition grown by pulsed laser deposition (PLD) and transferred *ex situ*. All LPNO films were measured before and after *in situ* ozone treatment, which fills oxygen vacancies and stabilizes a superconducting phase with onset $T_c \approx 40$ K; Ca-doped LPCNO films exhibit comparable T_c without additional ozone treatment.

Across growth methods, surface terminations and oxygen-vacancy concentrations, we resolve an electronic structure that is strongly renormalized and systematically shifted by epitaxial strain relative to bulk, with a progressive relaxation upon increasing thickness. A detailed analysis of the γ band reveals its bonding character, placing bilayer nickelates away from the strong- U limit. We further observe a Ca-doping-dependent crossing of the Fermi level, indicating that the presence of a γ pocket is not required for superconductivity, although the proximity of the high density of states associated with the γ band to the Fermi level may still play an important role. Concurrently, we observe a ~ 70 -meV kink in the α -band dispersion, consistent with coupling to oxygen-derived phonons or spin excitations. Together, these results motivate theoretical descriptions of superconductivity rooted in an intermediate-correlation regime and largely independent of the presence of a γ pocket at the Fermi level.

II. METHODS

The MBE-grown bilayer nickelate films investigated in this study were deposited on (001)-oriented SrLaAlO_4 (LSAO) substrates (in-plane lattice constant 3.756 Å). Figure 1(a) shows the bilayer nickelate crystal structure, with arrows indicating the epitaxial compressive strain imposed by the substrate and the resulting elongation along the c axis. Prior to growth, the LSAO substrates were annealed in 5×10^{-6} Torr ozone at 750°C for 10 min. The bilayer nickelate thin films were then deposited layer by layer at 680°C in $2\text{--}7 \times 10^{-6}$ Torr ozone using a shutter-controlled growth mode. During deposition, reflection high-energy electron diffraction with a 15 keV electron

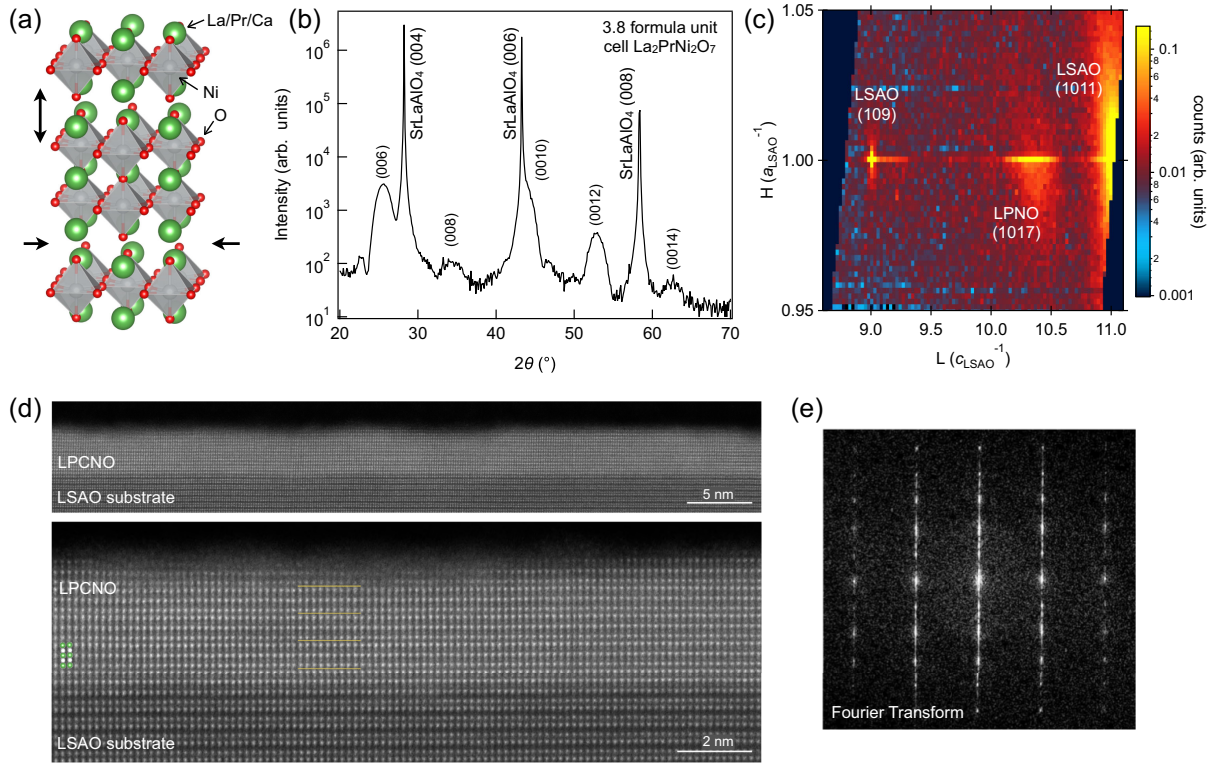


FIG. 1. MBE synthesis of compressively strained bilayer nickelate thin film. (a) The structural schematic of LPCNO with the arrows indicating the epitaxial compressive strain as well as the resulting elongation along the c axis. (b) X-ray diffraction (XRD) along the c axis for a 3.8 f.u.c. thick LPNO film with the substrate and thin film peaks labeled. (c) Reciprocal space map (RSM) of the same thin film sample shown in (b) with the substrate and thin film peaks labeled. (d) High-angle annular dark-field scanning transmission electron microscope (HAADF-STEM) image of a superconducting $\text{La}_{1.84}\text{PrCa}_{0.16}\text{Ni}_2\text{O}_7$ thin film and (e) the corresponding Hann-windowed Fourier transform.

beam was used to monitor the growth process and film quality (see Fig. S2 [51]). After growth, the samples were first cooled at a rate of $80^\circ\text{C}/\text{min}$ to below 80°C while maintaining the ozone pressure, and subsequently transferred into the *in situ*-connected synchrotron ARPES end-station sample stage (8 K) within approximately 20 min. Precise control of stoichiometry (La + Pr + Ca:Ni ratio) and monolayer dosage required for high-quality bilayer nickelate synthesis was achieved through calibration growth of the corresponding $(\text{La}, \text{Pr}, \text{Ca})\text{NiO}_3$ using a co-deposition growth mode [57,58]. The synthesis of the PLD-grown samples has been reported previously [30]. These thin films were exposed to ambient environment and transferred *ex situ* into the load lock chamber ($\sim 1 \times 10^{-9}$ Torr) of the ARPES end station within approximately 30 min after cooling following synthesis.

The structural quality of the as-grown MBE thin films was characterized by XRD, as shown in Fig. 1(b). The diffraction pattern exhibits multiple well-defined (00 l) reflections indexed to the bilayer phase, together with peaks from the LSAO substrate. The extracted c -axis lattice constant is $20.72 \text{ \AA}$, $\sim 1\%$ larger than the bulk value and consistent with previous experimental reports [11,29,30]. The $\sim 2\%$ compressive epitaxial strain in the film is further

confirmed by synchrotron x-ray reciprocal space mapping (RSM), shown in Fig. 1(c). The high crystalline quality of the films is additionally supported by scanning transmission electron microscopy (STEM), which reveals that the bilayer structural order is preserved over a large field of view [Fig. 1(d)]. At the same time, as previously described by Bhatt *et al.*, lower-symmetry crystal structures (for example, *Amam*) can be identified through additional half-order superlattice peaks in the Fourier transforms of high-angle annular dark-field (HAADF) STEM images [31]. In our measurements [Fig. 1(e)], such superlattice peaks are absent in the Fourier transforms, consistent with an $I4/mmm$ symmetry for our thin films imposed by the tetragonal LSAO substrate.

To stabilize superconducting LPNO samples, both MBE- and PLD-grown thin films were transferred to an *in situ*-connected ozone treatment chamber. The samples were annealed under 900 mTorr of flowing ozone at temperatures ranging from 210 to 330°C for 30 min. After treatment, the samples were transferred to the *in situ*-connected synchrotron ARPES end-station sample stage (8 K) through a preparation chamber with base pressure below 1×10^{-10} Torr within approximately 20 min. Transport properties of such superconducting samples were subsequently measured *ex situ*

within 1 h after the ARPES measurements. As-grown samples are measured in a similar procedure without the ozone treatment.

III. RESULTS

A. Overall electronic structure characterization

Assuming relatively weak on-site Coulomb interactions, the electronic structure of bilayer nickelate thin films can be qualitatively inferred from their crystal symmetry and nominal Ni $3d^{7.5}$ valence. Under octahedral coordination, the low-energy electronic manifold is primarily composed of four initially layer-degenerate orbitals with $3d_{z^2}$ and $3d_{x^2-y^2}$ character. Strong interlayer hopping mediated by the central apical oxygen lifts the degeneracy of the two $3d_{z^2}$ orbitals and splits them into bonding and antibonding bands, with the bonding γ band residing just below the Fermi level and exhibiting weak in-plane dispersion characteristic of its orbital character. By contrast, the layer-degenerate $3d_{x^2-y^2}$ orbitals are elevated relative to $3d_{z^2}$ by Jahn-Teller distortion, giving rise to two quarter-filled, itinerant orbitals. Their predominantly in-plane character suppresses direct interlayer hopping and preserves near degeneracy at the Brillouin-zone (BZ) center. However, an indirect $[\cos(k_x) - \cos(k_y)]^2$ interlayer hopping via hybridization with the $3d_{z^2}$ orbitals lifts the degeneracy along the $(\pi, 0)$ and equivalent momentum directions, producing an α pocket at Γ and a cuprate-like β pocket at the BZ corner.

This phenomenology is summarized in Figs. 2(a) and 2(b) using a simple tight-binding (TB) model described in a later section and in the Supplemental Material [51]. The model captures the experimental observations qualitatively well. Figures 2(c)–2(f) present the two-component Fermi surface together with the dispersions of the α , β , and γ bands along three high-symmetry momentum cuts, indicated in Fig. 2(c) by solid lines of corresponding colors, measured by ARPES at 8 K. The photon energies used for each measurement are indicated in the respective panels. Details of the spectral resolution and measurement conditions are provided in Appendix 3. Notably, within our experimental resolution, no discernible differences in the electronic structure are observed between as-grown and ozone-treated superconducting LPNO samples (Fig. S3 [51]). This insensitivity can be understood within the framework established by a recent study of oxygen stoichiometry in our MBE-grown films, in which the roles of distinct oxygen defects were disentangled: oxygen vacancies primarily act as scattering centers, whereas interstitial oxygen predominantly modifies carrier doping [59]. In particular, interstitial oxygen defects become prominent only upon Ca substitution, while ozone treatment of undoped LPNO films mainly suppresses disorder associated with oxygen vacancies present in the as-grown state [59]. This interpretation is further consistent with another recent report on oxygen-stoichiometry tuning [60].

For the in-plane α and β bands, the predicted hybridization-mediated interlayer splitting is resolved along the high-symmetry $X - \Gamma - X$ cut [Fig. 2(d)]. As shown in the lower panel, the α -band dispersion exhibits a pronounced renormalization kink at ~ 70 meV below the Fermi level, indicative of coupling to a collective mode at this energy scale. The momentum-distribution-curve (MDC)-derived α -band dispersion is marked by green circles, which deviates from the solid red lines for bare bands at the kink energy. This feature is consistently observed in as-grown and ozone-treated superconducting LPNO samples, as well as in superconducting LPCNO samples (Fig. S4 [51]), underscoring its robustness across bilayer nickelate thin films. A similar kink has been reported elsewhere, appearing more prominent in thin films than in bulk crystals, an observation that warrants systematic investigation [12,13,61,62]. Its characteristic energy lies within the range of oxygen-related phonons and coincides with the optical magnon energy reported for bulk samples [63,64].

Turning to the γ band, Figs. 2(e) and 2(f) display high-symmetry cuts along the BZ boundary and diagonal, clearly resolving the band at ~ 70 meV below the Fermi level. The statistical distribution of the γ -band maximum found in our LPNO samples is summarized in the histogram in Fig. 2(g). Relative to bulk values (light blue bar), both as-grown (blue) and ozone-treated (red) samples exhibit a deeper band maximum, with a similar mean energy near ~ 70 meV. The broader distribution observed in as-grown samples likely reflects variations in oxygen stoichiometry and strain (see Sec. III C) [32–34,59]. Given its proximity in energy to the α -band kink, the high density of states at the γ band top could participate in the low-energy physics bridged by the same collective modes [65].

B. Effect of Ca doping on the γ band

Much of the theoretical debate surrounding the superconducting mechanism in bilayer nickelates centers on the presence or absence of a γ -band Fermi-surface pocket. While several studies [66], including the present work, place the γ band below the Fermi level, others report it crossing E_F [61,67], pointing to critical sample-dependent variation across experimental groups. Recent studies of oxygen stoichiometry have also found that interstitial oxygen and cation substitution produce comparable hole-doping effects, both capable of shifting the γ band towards the Fermi level [59]. To examine this scenario, we synthesized $\text{La}_{2-x}\text{PrCa}_x\text{Ni}_2\text{O}_7$ thin films with Ca doping up to $x = 0.38$, thereby enhancing both interstitial oxygen solubility and hole doping via cation substitution [59].

Figures 3(a) and 3(b) compare high-symmetry cuts along the BZ boundary for undoped and Ca-doped samples. Upon Ca doping, the intense γ -band maximum observed in the undoped compound evolves into two nearly vertical dispersive features that extend up to the Fermi level,

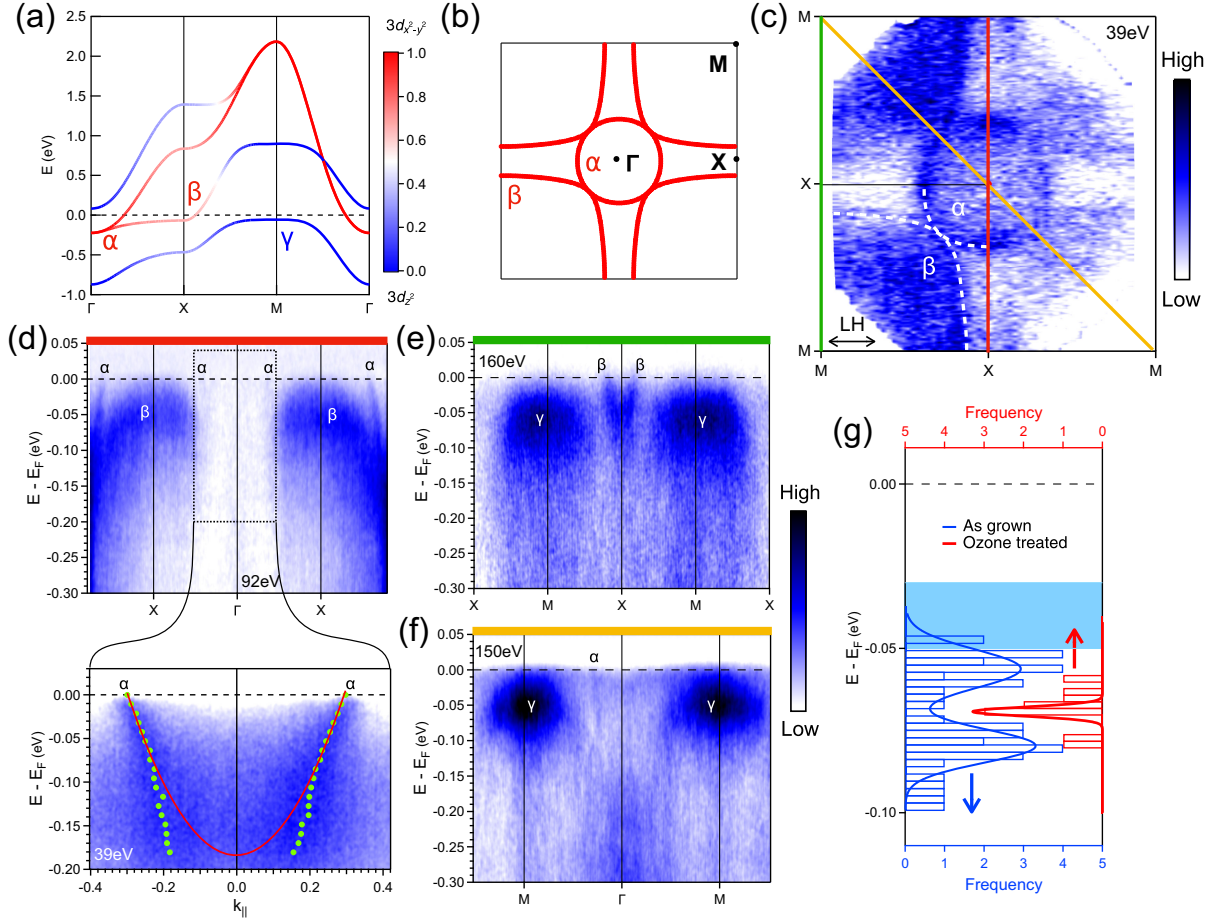


FIG. 2. ARPES characterization of the electronic structure of LPNO thin films. (a),(b) Tight-binding (TB) calculated band dispersion and Fermi surface for a bilayer nickelate thin film under 2% epitaxial compressive strain. The corresponding model and its parameters are discussed in a later section. The color scale of (a) reflects the orbital projection of each band. (c) Fermi-surface map of a representative as-grown MBE LPNO sample acquired with 39 eV linear horizontally (LH) polarized photons at 8 K. White dashed lines indicate the α and β pockets as guides to the eye. (d)–(f) Representative high-symmetry cuts of as-grown MBE samples along the BZ boundary [solid blue line in (c)], BZ diagonal (solid green line), and BZ center (solid red line), measured at 8 K with LH-polarized photons; photon energies are indicated in each panel. The lower panel of (d) shows a magnified view of the α -band dispersion, with green markers extracted from MDC peak fitting and with red lines as guides to the eye highlighting the renormalization kink. (g) Histogram of the γ -band maximum energy for as-grown (blue) and ozone-treated (red) samples. No systematic difference is observed between MBE- and PLD-grown samples. The γ -band energies are primarily determined using 150 eV photons, with three samples measured at 144 and 160 eV. Solid lines denote Gaussian fits, yielding peak positions at -54 , -91 , and -71 meV. The light-blue shaded region indicates the range of γ -band energies reported for bulk LNO samples [12].

accompanied by residual spectral weight at the high-symmetry M point. This evolution is highlighted by MDCs taken at the Fermi level (red dashed line) and at 60 meV below (green dashed line). In the undoped case, both MDCs display a broad single peak, reflecting the flat γ -band maximum. By contrast, the Ca-doped sample exhibits two additional side peaks in both MDCs, indicating that the dispersive features extend to the Fermi level and are spectroscopically distinct from the residual spectral weight centered at the M point. This behavior is observed in superconducting samples with lower Ca dopings as well (Fig. S5; see also the corresponding transport behavior [51]). We note that some oxygen loss may occur in the Ca-doped samples during transfer, especially near the

surface region probed by ARPES. However, the electron doping that could be introduced by oxygen vacancies would shift the electronic bands to higher binding energy, moving them away from the Fermi level. As this behavior is opposite to our observation, the spectral differences are a consequence of Ca doping instead of differing oxygen-vacancy concentrations.

The spectra are also qualitatively consistent with previous reports on undoped LPNO thin films grown by gigantic-oxidative atomically layer-by-layer epitaxy, in which analogous features were interpreted as evidence for a γ -band crossing of the Fermi level [61,67]. However, given the finite signal-to-noise ratio and substantial spectral broadening in the present data—likely partially driven by

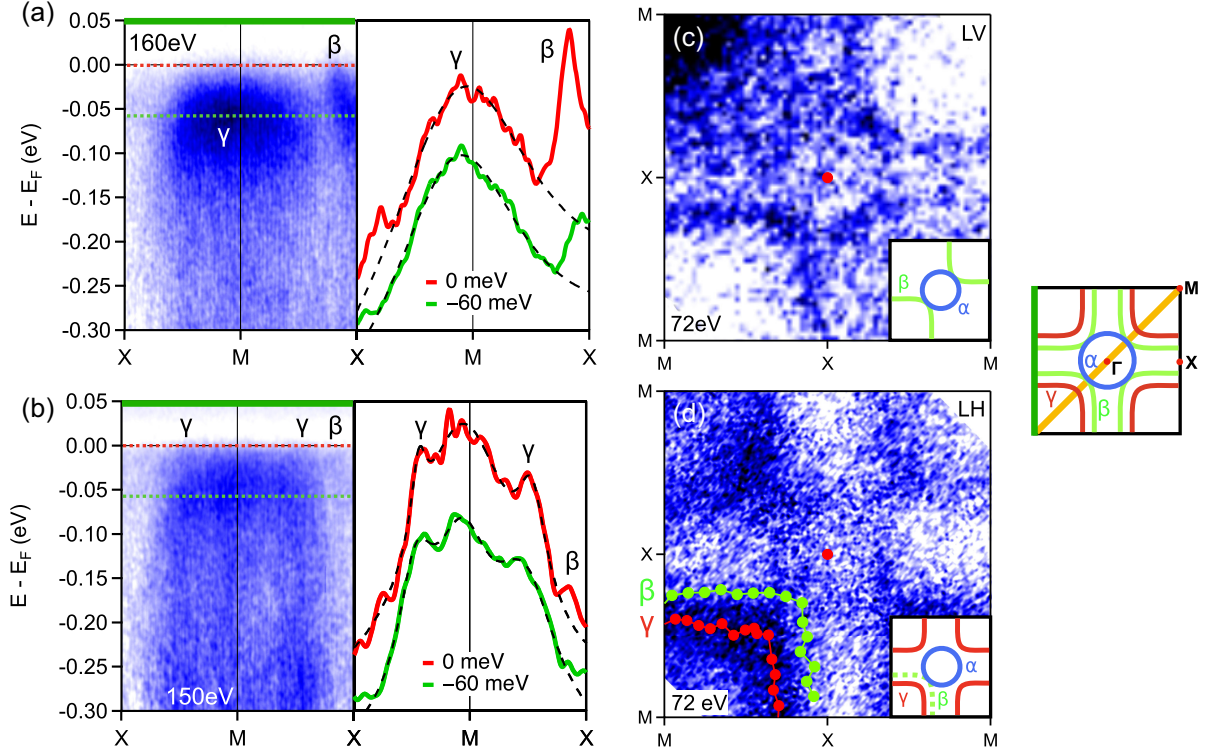


FIG. 3. Ca-doping dependence of the γ band. (a),(b) ARPES spectra acquired at 8 K along the BZ boundary for an as-grown LPNO sample (a) and a superconducting $\text{La}_{1.62}\text{PrCa}_{0.38}\text{Ni}_2\text{O}_7$ sample (b). The corresponding photon energies are indicated in each panel. The right halves show the corresponding MDCs taken at the energies marked by colored dashed lines. The black dashed lines are fits considering one (a) and three (b) Lorentzian peaks. (c),(d) Fermi-surface maps of a $\text{La}_{1.76}\text{PrCa}_{0.24}\text{Ni}_2\text{O}_7$ sample acquired with 72 eV photons under linearly vertical (LV) (c) and LH (d) polarizations. Insets show schematic representations of the resolved Fermi-surface components. Via peak fitting, green markers in (d) reproduce the β pocket extracted from (c) and red markers trace out the γ -like pocket.

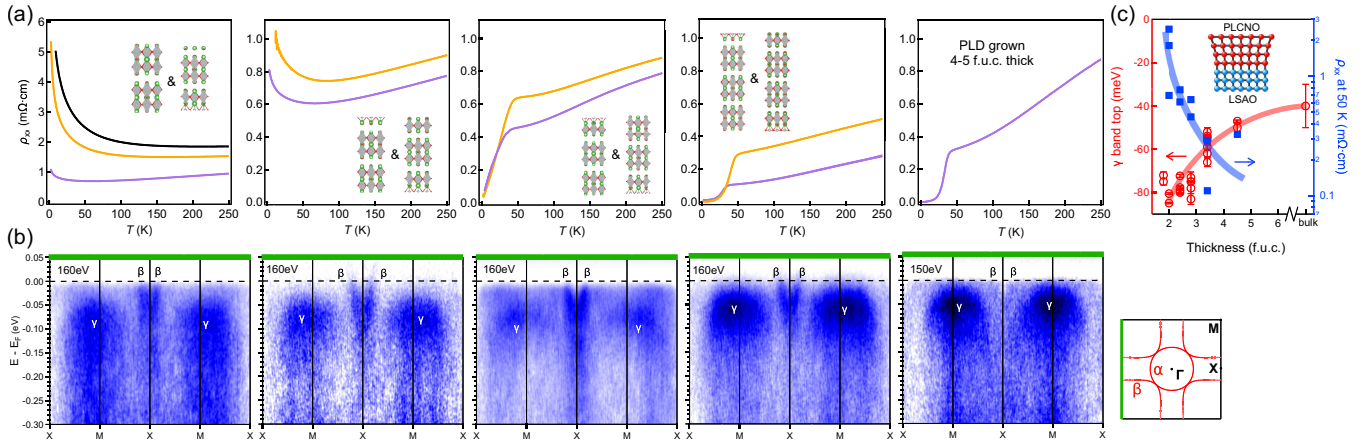


FIG. 4. Effect of epitaxial strain on the electronic structure. (a) Temperature-dependent resistivity for representative superconducting MBE-grown films with thicknesses ranging from 2 to 3.4 f.u.c., together with a 4–5 f.u.c. PLD-grown film. Different colors denote distinct samples. The corresponding film thickness in each case is indicated by the structural schematic insets (two configurations are shown to account for mixed substrate termination) or labelled directly. (b) Representative high-symmetry cuts along the BZ boundary (red solid line in the right inset) for as-grown samples of thickness corresponding to (a) panels above, measured with LH-polarized photons. Photon energies are indicated in the upper-left corner of each panel. (c) Energy position of the γ -band maximum relative to E_F and the thin film resistivity at 50 K as a function of film thickness. The inset schematically illustrates the progressive relaxation of epitaxial strain with increasing thickness.

disorder or inhomogeneous regions introduced through Ca substitution—we refrain from making a definitive claim of a γ -band crossing. Instead, we adopt this phenomenological framework as a basis for the following discussion.

Returning to the additional spectral weight at the M point in Ca-doped samples, one possible interpretation is the opening of a superconducting gap in the γ band. However, we do not observe a temperature dependent shift of its energy position or concomitant gap in the α or β bands within our experimental energy resolution, rendering this scenario unlikely. Instead, the feature may reflect extrinsic factors such as sample inhomogeneity or an intrinsic many-body effect, such as spin polarons—bound states of quasiparticles and paramagnons—arising from magnetic fluctuations associated with the large density of states of the γ band [68].

Additional support for the γ -band-crossing scenario is provided by polarization-dependent measurements. Figures 3(c) and 3(d) present ARPES Fermi-surfaces maps acquired on another doped sample using 72 eV photons, where matrix-element effects under different polarization geometries enable a clear separation of the β pocket, enhanced under LV polarization, from the additional γ -like pocket observed under LH polarization at the Brillouin-zone corner. The solid green markers in Fig. 3(d) trace the β pocket extracted from the LV data in Fig. 3(c), highlighting its clear separation from the γ -like pocket marked by the solid red markers. The corresponding Fermi-surface contours resolved under each polarization geometry are illustrated schematically in the insets, while the rightmost inset summarizes the complete Fermi-surface topology.

This systematic evolution with Ca doping suggests that experimental discrepancies in the literature may arise from unintentional hole doping, for example via interfacial Sr incorporation or variations in growth and ozone treatment conditions [29,67]. Importantly, within a single experimental platform, we are able to reproduce both classes of electronic structure reported previously while independently confirming superconductivity through transport measurements in each case. These results demonstrate that the presence of a γ -band-like Fermi-surface pocket is not a prerequisite for superconductivity in bilayer nickelate thin films, although the proximity of the γ band's high density of states to the Fermi level can remain relevant to the superconducting mechanism.

C. Shift of γ band by compressive epitaxial strain

Beyond capturing the general electronic structure outlined above, theory further predicts a systematic strain-induced band shift in bilayer nickelate thin films relative to bulk. DFT + U calculations with intermediate $U \approx 3.5$ eV indicate that epitaxial compressive strain shifts the in-plane α and β bands towards lower binding energy, while driving the out-of-plane γ band to substantially higher binding energy [32–34]. Figure 4(a) presents a series of LPNO films with

increasing thickness from 2 to 4–5 formula-unit cells (see Methods), where this strain-driven evolution is directly observed; the corresponding thicknesses are indicated by structural schematics or annotations in Fig. 4(a).

A pronounced thickness dependence is first evident in transport. As shown in Fig. 4(a), with increasing thickness, the normal-state conductivity is systematically enhanced, evolving from strongly insulating behavior in the 2 f.u. limit to a fully metallic temperature dependence. Superconductivity emerges once the low-temperature resistivity falls below ~ 0.6 m $\Omega \cdot$ cm, corresponding to ~ 6.7 k Ω /sq per bilayer. This value is remarkably close to the quantum critical resistance $h/(2e)^2 = 6.45$ k Ω /sq associated with the insulator-superconductor transitions [69]. Such behavior is widely observed in superconducting oxide thin films, where a critical thickness is required for superconductivity to develop [70,71].

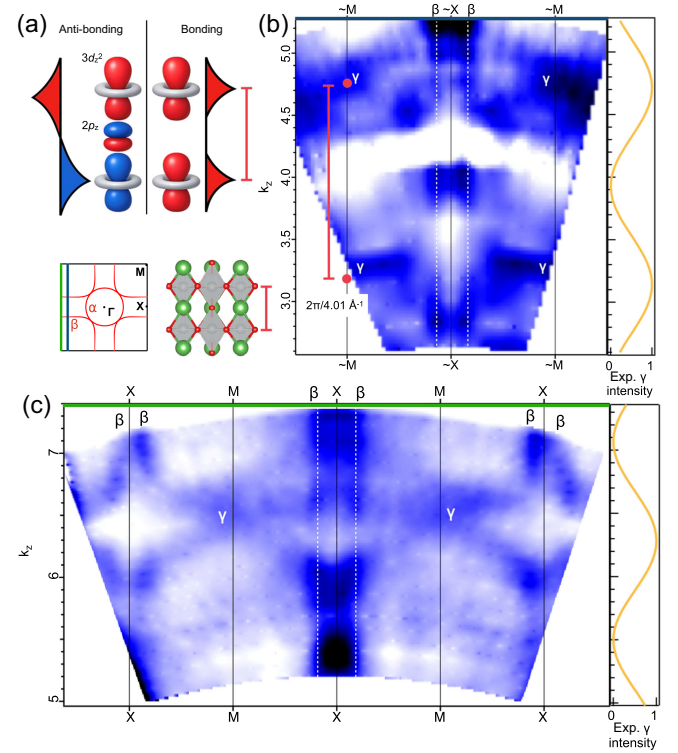


FIG. 5. Bonding nature of the γ band. (a) Schematic illustration of the orbital composition of the antibonding and bonding states formed by the two Ni $3d_{z^2}$ orbitals, together with the corresponding phase structure of their wavefunctions. (b),(c) Out-of-plane Fermi-surface maps acquired along the trajectories indicated by the solid dark blue (b) and green (c) lines in the Fermi-surface schematic to the left of (b). Photon energies are varied from 20 to 100 eV in (b) and from 96 to 200 eV in (c). The panels on the right plot the expected oscillatory γ band spectral intensity. White dashed lines serve as guides to the eye, marking the β band. The schematic on left side of (b) illustrates the real-space interlayer spacing corresponding to the periodicity indicated by the solid red line in momentum space.

Concomitantly, ARPES, which is more sensitive to the surface layers, reveals a systematic shift of the γ band towards lower, bulklike binding energy with increasing thickness, as seen in the high-symmetry cuts along the BZ boundary [Fig. 4(b)]. This evolution is summarized in Fig. 4(c), where the γ -band maximum shifts from ~ -85 meV in the thinnest films to ~ -45 meV in thicker samples, while the thin film resistivity at 50 K drops from above 1 to 0.1-0.3 m Ω ·cm in parallel. This pronounced downward shift in 2 f.u.c. films relative to bulk, followed by a gradual recovery towards bulk values with increasing thickness, reflects the progressive relaxation of epitaxial

strain and provides qualitative confirmation of the theoretically predicted strain-driven band evolution. At the same time, the persistent observation of the γ band below E_F in undoped LPNO thin films across all thicknesses indicates that the measured electronic structure is intrinsic to the bilayer nickelate phase, rather than arising from intergrowths of secondary Ruddlesden-Popper phases at the surface, which only become appreciable in thicker films.

D. Intermediate-correlation character of the γ band

So far, the electronic structure is qualitatively consistent with an intermediate- U regime. The observations are not

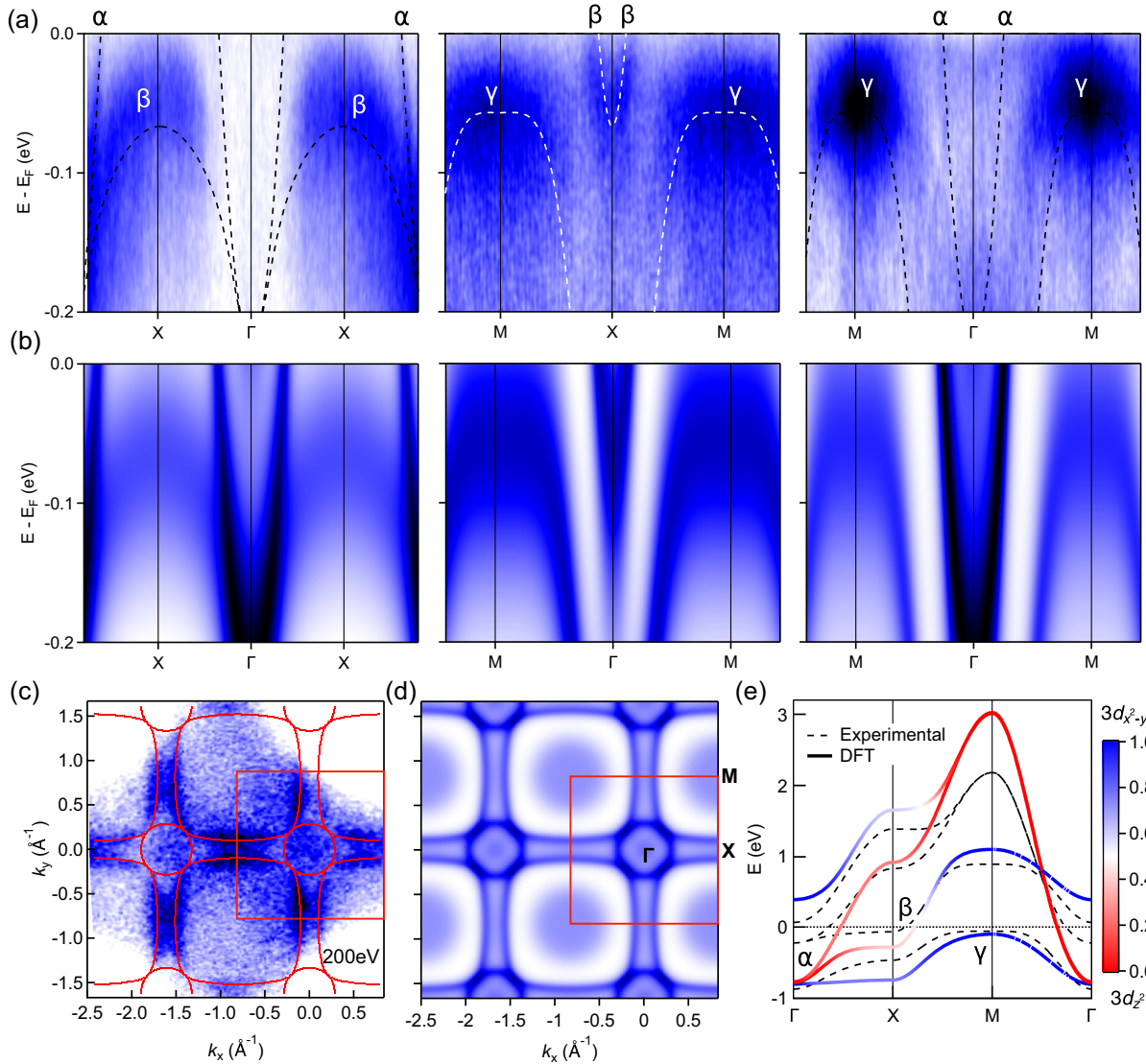


FIG. 6. Correlation effects on the electronic structure. (a) High-symmetry cuts of LPNO samples along the BZ boundary, diagonal, and center directions, as already shown in Figs. 2(d) and 2(e). Black and white dashed lines denote optimized TB fits to the measured dispersions. (b) Simulated electronic dispersions based on the optimal TB model, with each panel corresponding to the experimental cut shown above. (c) Measured Fermi-surface map of an LPNO sample with 200 eV photons and (d) the corresponding simulated map derived from the optimal TB fit. Solid red lines in (c) indicate the Fermi surface obtained from the TB model. (e) DFT band dispersions calculated with $U = 0$ eV and using the same lattice constants as the thin-film samples. The color scale denotes the orbital character of each band. Black dashed lines indicate the dispersions obtained from the TB fit to experimental observations shown in (a),(b).

satisfactorily captured either by a strong-coupling picture with localized and singly occupied $3d_{z^2}$ orbitals or by calculations assuming minimal correlation effects. In contrast to the large- U limit, the clearly resolved $\alpha - \beta$ band splitting implies substantial interlayer hopping mediated by the $3d_{z^2}$ orbitals. Conversely, the absence of a γ -band crossing at E_F in undoped LPNO films disfavors the weak-correlation limit, in which the γ band is generally predicted to lie at or above the Fermi level [41]. Instead, the observed electronic structure is more consistent with nontrivial correlation effects that shift the γ band to higher binding energy [41,72]. Furthermore, the thickness-dependent evolution of the band structure is well reproduced by DFT + U calculations in the intermediate-correlation regime. Beyond these observations, we next highlight two additional spectroscopic signatures that further support the placement of bilayer nickelate thin films within an intermediate-correlation framework.

In bilayer nickelates with relatively weak U , hybridization through the $2p_z$ orbital introduces an additional minus sign in the effective hopping, such that the bonding state (γ band) acquires a wave function symmetric about the bilayer midplane, in contrast to the antisymmetric antibonding state [Fig. 5(a)]. This phase structure gives rise to a characteristic oscillatory modulation of the photoemission matrix element as a function of photon energy, with a periodicity set by the interlayer spacing when expressed in k_z momentum [73] (see Supplemental Material [51]). In particular, the symmetric bonding state exhibits a cosine-like dependence, with maximal intensity at $k_z = 0$. This matrix-element modulation can substantially affect the apparent spectral weight and visibility of the γ band, potentially contributing to the differing interpretations reported in the literature.

This behavior is borne out in the bilayer nickelate thin films. Figures 5(b) and 5(c) show the k_z -dependent Fermi surface along two cuts spanning photon energies from 20 to 200 eV near the BZ boundary (assuming an inner potential of 13 eV), with the corresponding cut trajectories indicated in the schematic above Fig. 5(c). The spectral weight near the M point serves as a proxy for the γ -band spectral intensity, reflecting its large density of states extending to the Fermi level. The red lines denote the expected k_z periodicity set by the interlayer spacing, $\sim 1.57 \text{ \AA}^{-1}$, in excellent agreement with the observed oscillation of the γ -band intensity, which extrapolates to a maximum at $k_z = 0$. Notably, our observations are also qualitatively consistent with a recent independent study that reported enhanced γ -band intensity near $k_z \approx 4.7$ and $6.5 \text{ \AA}^{-1}$, despite arriving at a different interpretation of the underlying electronic structure [74].

In the strong- U limit, bonding and antibonding states no longer constitute the appropriate eigenbasis, and the γ band is expected to acquire mixed bonding and antibonding character. Consequently, additional spectral weight

associated with the antibonding component would be expected at photon energies where a purely bonding state exhibits minimal intensity. However, no such additional spectral weight is observed over binding energies from E_F to approximately -150 meV . Together, these results place bilayer nickelate thin films away from the strong-correlation regime.

All three low-energy bands, particularly the γ band, exhibit pronounced renormalization, indicative of substantial electronic correlations. To quantify this effect, we fit the measured dispersions using a two-orbital four-band TB model optimized through a machine-learning-assisted procedure; details are provided in Supplemental Material [51]. As shown in Figs. 6(a)–6(d), the resulting simulations [Figs. 6(b) and 6(d)] reproduce the key features of both the experimental band dispersions [Fig. 6(a)] and Fermi surfaces [Fig. 6(c)]. We note that matrix-element effects are not included in the TB simulations, and therefore the weak α -band intensity observed at 150 eV photon energy in the first BZ of the rightmost panel in Fig. 6(a) is not captured, despite the otherwise excellent agreement in higher BZs and at other photon energies. The extracted TB parameters, summarized in Table I, provide a quantitative theoretical description of the electronic structure and establish a foundation for microscopic modelling. In particular, the interlayer hopping amplitude of the Ni $3d_{z^2}$ orbitals ($t_{\perp z}$) substantially exceeds the nearest-neighbor in-plane hopping of the Ni $3d_{x^2-y^2}$ orbitals (t_{1x}), underscoring the central role of interlayer coupling in shaping the low-energy electronic structure.

Comparison between the experimentally extracted hopping parameters and DFT calculations performed with

TABLE I. Parameters of the four-orbital tight-binding model describing the experimentally measured band structure of compressively strained bilayer nickelate thin films and the corresponding DFT band structure calculated with $U = 0 \text{ eV}$. The DFT calculations use the same lattice constants as the thin-film samples. Definitions of the fitting parameters are provided in the Supplemental Material [51]. The two dominant hopping amplitudes, t_{1x} and $t_{\perp z}$, are highlighted in gray.

Tight-binding parameters	Optimal values for measured dispersions	DFT-derived values ($U = 0 \text{ eV}$)
t_{1x}	−0.301	−0.474
t_{1z}	−0.102	−0.0878
t_{2x}	0.07	0.0769
t_{2z}	−0.052	−0.015
t_{3xz}	0.162	0.209
$t_{\perp x}$	0	0.006
$t_{\perp z}$	−0.478	−0.603
t_{4xz}	−0.01	−0.029
e^x	0.7	0.814
e^z	0.222	0.212

TABLE II. Energy positions, renormalization factors, and c -axis lattice expansion of the α , β , and γ bands. The energy positions correspond to the α -band bottom, the β -band saddle point at the BZ X point, and the γ -band top. For the thin-film samples, these values are extracted from the tight-binding fits described in Fig. 6. Renormalization factors are obtained by rescaling the thin-film dispersions to the bulk band structure at ambient pressure, as described in Fig. S6 [51]. The bulk band positions, c -axis lattice constants, and renormalization factors are taken from Refs. [7,12]. Uncertainties in the thin-film renormalization factors arise from the renormalization kink in the α -band dispersion and the weak dispersion of the γ band.

Band position (meV) (α , β : saddle point; γ : band top)			c -lattice constant ($\text{\AA}$)		Renormalization factor	
Band	Ambient pressure bulk	LPNO thin film	LPNO thin film	Ambient pressure bulk	Ambient pressure bulk	LPNO thin film
α	-248	-223	20.72	20.52	1.8	2.1 ± 0.2
β	-80	-66			2.6	3.1 ± 0.9
γ	-50	-57			5.5	7.3 ± 1.9

$U = 0$ eV further reveals the magnitude of correlation-driven renormalization. The DFT calculations use the experimentally determined lattice constants of the compressively strained thin films. The resulting relaxed structure closely approximates the $I4/mmm$ structure, with the out-of-plane Ni-O-Ni bond angle $\sim 180^\circ$. Its TB parameters are obtained via Wannier downfolding (see Supplemental Material [51]). As summarized in Table I, the two dominant energy scales, $t_{\perp z}$ and t_{1x} , are reduced by approximately 20%–40% relative to their DFT values. This suppression is directly reflected in the flatter experimental dispersions shown in Fig. 6(e), where DFT-derived bands (solid-colored lines) are compared with experimentally extracted dispersions (black dashed lines). The observed renormalization is notably consistent with previous dynamical mean-field theory calculations, which predict similarly flattened dispersions for $U > 3.6$ eV (Ref. [41]). Such a correlation energy scale is comparable to the electronic bandwidth, emphasizing the nontrivial correlation within the material.

The strength of the renormalization can also be evaluated experimentally by comparing the thin-film dispersions with those reported for bulk bilayer nickelates at ambient pressure, where band-dependent mass renormalization factors have previously been established [12]. As shown in Fig. S6 [51], the $3d_{x^2-y^2}$ -derived bands exhibit relatively modest renormalization factors of ~ 2 – 3 , whereas the $3d_{z^2}$ -derived γ band displays a much stronger enhancement of ~ 7 . Table II summarizes these renormalization factors together with the corresponding orbital-dependent energy positions and lattice parameters of the compressively strained LPNO thin films, providing a minimal experimental description of the material complementary to Table I. Together, these results establish the presence of substantial electronic correlations in compressively strained bilayer nickelate thin films and identify the system as residing in an intermediate-correlation regime.

IV. CONCLUSIONS

As a high- T_c superconductor, the bilayer nickelate differs fundamentally from the cuprates in that the $3d_{z^2}$

orbital plays a central role in shaping the low-energy electronic structure and therefore cannot be neglected in microscopic descriptions of superconductivity. Identifying the key electronic features associated with this orbital is thus essential for understanding the bilayer nickelates and, more broadly, for elucidating the ingredients required for high- T_c superconductivity. Our measurements reveal an electronic structure governed by strong interlayer hybridization of the $3d_{z^2}$ orbitals, which drives the pronounced splitting between the α and β bands along the $(\pi, 0)$ and symmetry-equivalent momentum directions.

Focusing on the $3d_{z^2}$ -derived γ band, our observations further suggest that a γ -like pocket at the Fermi level is not a prerequisite for superconductivity. Instead, its apparent emergence likely reflects additional hole doping, introduced here through Ca substitution but potentially also attainable via excess interstitial oxygen. This observation constrains theoretical scenarios in which a γ -like pocket is required to stabilize superconductivity, suggesting instead that such models represent a limiting rather than generic case. Moreover, the observation of photon-energy-dependent intensity modulations characteristic of a symmetric bonding wave function, together with the strong renormalization of the γ -band dispersion relative to DFT calculations, places these thin films within an intermediate-correlation regime. Taken together, our results establish key experimental constraints on band-selective pairing scenarios and the microscopic origin of superconductivity in bilayer nickelates.

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B. Y. W. and Z. X. S. conceived and designed the project. B. Y. W. synthesized the MBE grown films with help from S. A., Y. Z., and R. W. Y. L. and E. K. K. synthesized the PLD grown films with the supervision from H. Y. H. B. Y. W., Y. L., Y. Z., Y. Y., J. L., V. T., R. W., and Y. T. performed the XRD and transport characterizations with the supervision from Y. S. L. and H. Y. H. B. Y. W. and Y. Y. performed the ozone annealing study. B. H. G. performed the STEM measurements. B. Y. W. and S. A. performed the ARPES measurement with help from Y. Z., R. W., C. L., M. H., and D. L. Y. Z., X. Z., Y. W., T. P. D., and C. J. performed the DFT calculation and tight-binding model analysis. B. Y. W., S. A., and Z. X. S. analyzed the data and wrote the manuscript with input from all authors.

DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

APPENDIX

1. Sample preparation and XRD characterization

For both MBE and PLD grown LPNO thin films, the SrLaAlO₄(001) substrates (MTI Corporation) were sonicated in acetone and IPA. The different thickness of MBE grown sample is achieved through the layer-by-layer shutter-controlled growth. Here, 2, 2.4, and 2.8 f.u.c. thicknesses correspond to stopping the growth after depositing 2 complete f.u.c., after additional one layer of LaO and NiO₂ each, and after two layers of LaO and NiO₂ each. The PLD grown LPNO thin films were deposited with a KrF excimer laser (wavelength 248 nm). During the growth, the substrate temperature was maintained at 660 °C, under oxygen partial pressure $p(\text{O}_2)$ of 150 mTorr. The pulsed laser repetition rate during the deposition was 5 Hz. The laser fluence was 0.56 J/cm² with a spot size of 3.3 mm². After growth, XRD data were measured using a monochromated Cu K α 1 source ($\lambda = 1.5406 \text{ \AA}$). The thickness of the PLD grown film is obtained through Scherrer fitting to the XRD spectrum. Additional XRD and RSM measurement was performed at beamline BL17-2 of Stanford Synchrotron Radiation Lightsource, using a photon energy of 8.33 keV at room temperature. Diffraction

signals were collected using a PILATUS3 100 K-M detector.

2. Transport measurements

The thin film samples were contacted with aluminum wires by an ultrasonic wire bonder and measured in 4He cryostats (base temperature $\sim 1.8 \text{ K}$). The 4-terminal measurements were carried out with 10 μA current.

3. ARPES measurements

The ARPES measurements were performed at beamline BL5-2 of Stanford Synchrotron Radiation Lightsource with a Scienta DA30 analyzer. The energy resolution is 26 meV at 200 eV photon energy and better at lower photon energies. The base pressure during the ARPES measurements was better than 3×10^{-11} Torr and the measurement temperature of the sample stage was maintained at 8 K. Before measurement, samples are transferred at room temperature through a preparation chamber with a base pressure better than 1×10^{-10} Torr. To minimize beam damage during the measurement, we performed a raster scan of the film surface, spending roughly 10 minutes per measurement location. Otherwise, negligible sample degradation is observed during our measurement period.

4. STEM measurements

Cross-sectional lamellae for scanning transmission electron microscopy investigations were prepared using the standard focused ion beam (FIB) lift-out procedure on a Thermo Fisher Scientific Helios G5 X FIB. High-angle annular dark-field (HAADF) STEM images were on a double aberration-corrected JEOL ARM300F operating at 300 kV with a probe convergence semiangle of 30 mrad, with 68 (200) mrad inner (outer) collection angle.

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- [1] J. G. Bednorz and K. A. Muller, *Possible high T_c superconductivity in the Ba-La-Cu-O system*, *Z. Phys. B* **64**, 189 (1986).
 - [2] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J. G. Bednorz, and F. Lichtenberg, *Superconductivity in a layered perovskite without copper*, *Nature (London)* **372**, 532 (1994).
 - [3] V. I. Anisimov, D. Bukhvalov, and T. M. Rice, *Electronic structure of possible nickelate analogs to the cuprates*, *Phys. Rev. B* **59**, 7901 (1999).
 - [4] D. Li, K. Lee, B. Y. Wang, M. Osada, S. Crossley, H. R. Lee, Y. Cui, Y. Hikita, and H. Y. Hwang, *Superconductivity in an infinite-layer nickelate*, *Nature (London)* **572**, 624 (2019).
 - [5] Y. Kamihara, T. Watanabe, M. Hirano, and H. Hosono, *Iron-based layered superconductor $\text{La}[\text{O}_{1-x}\text{F}_x]\text{FeAs}$ ($x = 0.05\text{--}0.12$) with $T_c = 26 \text{ K}$* , *J. Am. Chem. Soc.* **130**, 3296 (2008).

- [6] K. Takada, H. Sakurai, E. Takayama-Muromachi, F. Izumi, R. A. Dilanian, and T. Sasaki, *Superconductivity in two-dimensional CoO₂ layers*, *Nature (London)* **422**, 53 (2003).
- [7] H. Sun, M. Huo, X. Hu, J. Li, Z. Liu, Y. Han, L. Tang, Z. Mao, P. Yang, B. Wang *et al.*, *Signatures of superconductivity near 80 K in a nickelate under high pressure*, *Nature (London)* **621**, 493 (2023).
- [8] Y. Zhu, D. Peng, E. Zhang, B. Pan, X. Chen, L. Chen, H. Ren, F. Liu, Y. Hao, N. Li *et al.*, *Superconductivity in pressurized trilayer La₄Ni₃O_{10-δ} single crystals*, *Nature (London)* **631**, 531 (2024).
- [9] L. Wang, Y. Li, S.-Y. Xie, F. Liu, H. Sun, C. Huang, Y. Gao, T. Nakagawa, B. Fu, B. Dong *et al.*, *Structure responsible for the superconducting state in La₃Ni₂O₇ at high-pressure and low-temperature conditions*, *J. Am. Chem. Soc.* **146**, 7506 (2024).
- [10] J. Li, D. Peng, P. Ma, H. Zhang, Z. Xing, X. Huang, C. Huang, M. Huo, D. Hu, Z. Dong *et al.*, *Identification of superconductivity in bilayer nickelate La₃Ni₂O₇ under high pressure up to 100 GPa*, *Natl. Sci. Rev.* **12**, nwaf220 (2025).
- [11] E. K. Ko, Y. Yu, Y. Liu, L. Bhatt, J. Li, V. Thampy, C. T. Kuo, B. Y. Wang, Y. Lee, K. Lee *et al.*, *Signatures of ambient pressure superconductivity in thin film La₃Ni₂O₇*, *Nature (London)* **638**, 935 (2025).
- [12] J. Yang, H. Sun, X. Hu, Y. Xie, T. Miao, H. Luo, H. Chen, B. Liang, W. Zhu, G. Qu *et al.*, *Orbital-dependent electron correlation in double-layer nickelate La₃Ni₂O₇*, *Nat. Commun.* **15**, 4374 (2024).
- [13] C. C. Au-Yeung, X. Chen, S. Smit, M. Bluschke, V. Zimmermann, M. Michiardi, P. C. Moen, J. Kraan, C. S. B. Pang, C. T. Suen *et al.*, *Oxygen-centred planar orbitals in the electronic structure and spin-density-wave reconstruction of multilayer nickelates*, *Nat. Phys.* (2026), 10.1038/s41567-026-03286-4.
- [14] Z. Luo, X. Hu, M. Wang, W. Wú, and D.-X. Yao, *Bilayer two-orbital model of La₃Ni₂O₇ under pressure*, *Phys. Rev. Lett.* **131**, 126001 (2023).
- [15] Y. Zhang, L.-F. Lin, A. Moreo, and E. Dagotto, *Electronic structure, dimer physics, orbital-selective behavior, and magnetic tendencies in the bilayer nickelate superconductor La₃Ni₂O₇ under pressure*, *Phys. Rev. B* **108**, L180510 (2023).
- [16] F. Lechermann, J. Gondolf, S. Bötzel, and I. M. Eremin, *Electronic correlations and superconducting instability in La₃Ni₂O₇ under high pressure*, *Phys. Rev. B* **108**, L201121 (2023).
- [17] Q.-G. Yang, D. Wang, and Q.-H. Wang, *Possible s_±-wave superconductivity in La₃Ni₂O₇*, *Phys. Rev. B* **108**, L140505 (2023).
- [18] Y. Shen, M. Qin, and G.-M. Zhang, *Effective bi-layer model hamiltonian and density-matrix renormalization group study for the high-T_c superconductivity in La₃Ni₂O₇ under high pressure*, *Chin. Phys. Lett.* **40**, 127401 (2023).
- [19] Z. Ouyang, M. Gao, and Z.-Y. Lu, *Absence of electron-phonon coupling superconductivity in the bilayer phase of La₃Ni₂O₇ under pressure*, *npj Quantum Mater.* **9**, 80 (2024).
- [20] L. Bleys, N. Corkill, Y. F. Zhao, G. L. Pascut, H. LaBollita, A. S. Botana, and K. F. Quader, *Role of correlations in Ruddlesden-Popper bilayer nickelates under compressive strain*, *arXiv:2509.00940*.
- [21] Q. Qin and Y.-F. Yang, *High-T_c superconductivity by mobilizing local spin singlets and possible route to higher T_c in pressurized La₃Ni₂O₇*, *Phys. Rev. B* **108**, L140504 (2023).
- [22] Y.-F. Yang, G.-M. Zhang, and F.-C. Zhang, *Interlayer valence bonds and two-component theory for high-T_c superconductivity of La₃Ni₂O₇ under pressure*, *Phys. Rev. B* **108**, L201108 (2023).
- [23] V. Christiansson, F. Petocchi, and P. Werner, *Correlated electronic structure of La₃Ni₂O₇ under pressure*, *Phys. Rev. Lett.* **131**, 206501 (2023).
- [24] Y.-B. Liu, J.-W. Mei, F. Ye, W.-Q. Chen, and F. Yang, *s_±-wave pairing and the destructive role of apical-oxygen deficiencies in La₃Ni₂O₇ under pressure*, *Phys. Rev. Lett.* **131**, 236002 (2023).
- [25] C. Lu, Z. Pan, F. Yang, and C. Wu, *Interlayer-coupling-driven high-temperature superconductivity in La₃Ni₂O₇ under pressure*, *Phys. Rev. Lett.* **132**, 146002 (2024).
- [26] H. Oh and Y. H. Zhang, *Type-II t-J model and shared superexchange coupling from Hund's rule in superconducting La₃Ni₂O₇*, *Phys. Rev. B* **108**, 174511 (2023).
- [27] K. Jiang, Z. Wang, and F.-C. Zhang, *High-temperature superconductivity in La₃Ni₂O₇*, *Chin. Phys. Lett.* **41**, 017402 (2024).
- [28] Y. Wang, K. Jiang, Z. Wang, F.-C. Zhang, and J. Hu, *Electronic and magnetic structures of bilayer La₃Ni₂O₇ at ambient pressure*, *Phys. Rev. B* **110**, 205122 (2024).
- [29] G. Zhou, W. Lv, H. Wang, Z. Nie, Y. Chen, Y. Li, H. Huang, W. Q. Chen, Y. J. Sun, Q. K. Xue *et al.*, *Ambient-pressure superconductivity onset above 40 K in (La, Pr)₃Ni₂O₇ films*, *Nature (London)* **640**, 641 (2025).
- [30] Y. Liu, E. K. Ko, Y. Tam, L. Bhatt, J. Li, V. Thampy, B. H. Goodge, D. A. Muller, S. Raghu, Y. Yu *et al.*, *Superconductivity and normal-state transport in compressively strained La₂PrNi₂O₇ thin films*, *Nat. Mater.* **24**, 1221 (2025).
- [31] L. Bhatt, E. Abarca Morales, A. Y. Jiang, E. K. Ko, Y. F. Zhao, N. Schnitzer, G. A. Pan, D. Ferenc Segedin, Y. Liu, Y. Yu *et al.*, *Structural modifications in strain-engineered bilayer nickelate thin films*, *Nature (London)* **653**, 76 (2026).
- [32] H. C. R. B. Bhatta, X. Zhang, Y. Zhong, and C. Jia, *Structural and electronic evolution of bilayer nickelates under biaxial strain*, *arXiv:2502.01624*.
- [33] Y.-F. Zhao and A. S. Botana, *Electronic structure of Ruddlesden-Popper nickelates: Strain to mimic the effects pressure*, *Phys. Rev. B* **111**, 115154 (2025).
- [34] B. Geisler, J. J. Hamlin, G. R. Stewart, R. G. Hennig, and P. J. Hirschfeld, *Fermi surface reconstruction and enhanced spin fluctuations in strained La₃Ni₂O₇ on LaAlO₃ (001) and SrTiO₃ (001)*, *Phys. Rev. B* **112**, L100506 (2025).
- [35] Y. Wang, K. Jiang, J. Ying, T. Wu, J. Cheng, J. Hu, and X. Chen, *Recent progress in nickelate superconductors*, *Natl. Sci. Rev.* **12**, nwaf373 (2025).
- [36] W. Xi, S. L. Yu, and J. X. Li, *Transition from s_±-wave to d_{x²-y²}-wave superconductivity driven by interlayer interaction in the bilayer two-orbital model of La₃Ni₂O₇*, *Phys. Rev. B* **111**, 104505 (2025).
- [37] H. Sakakibara, N. Kitamine, M. Ochi, and K. Kuroki, *Possible high T_c superconductivity in La₃Ni₂O₇ under high*

- pressure through manifestation of a nearly half-filled bilayer hubbard model, *Phys. Rev. Lett.* **132**, 106002 (2024).
- [38] K. Y. Jiang, Y. H. Cao, Q. G. Yang, H. Y. Lu, and Q. H. Wang, *Theory of pressure dependence of superconductivity in bilayer nickelate $\text{La}_3\text{Ni}_2\text{O}_7$* , *Phys. Rev. Lett.* **134**, 076001 (2025).
- [39] Y. Gu, C. Le, Z. Yang, X. Wu, and J. Hu, *Effective model and pairing tendency in the bilayer Ni-based superconductor $\text{La}_3\text{Ni}_2\text{O}_7$* , *Phys. Rev. B* **111**, 174506 (2025).
- [40] Y. B. Liu, H. Sun, M. Zhang, Q. Liu, W. Q. Chen, and F. Yang, *Origin of the diagonal double-stripe spin density wave and potential superconductivity in bulk $\text{La}_3\text{Ni}_2\text{O}_7$ at ambient pressure*, *Phys. Rev. B* **112**, 014510 (2025).
- [41] C. Yue, J. J. Miao, H. Huang, Y. Hua, P. Li, Y. Li, G. Zhou, W. Lv, Q. Yang, F. Yang *et al.*, *Correlated electronic structures and unconventional superconductivity in bilayer nickelate heterostructures*, *Natl. Sci. Rev.* **12**, nwaf253 (2025).
- [42] G. Heier, K. Park, and S. Y. Savrasov, *Competing d_{xy} and $s_{\pm}$ pairing symmetries in superconducting $\text{La}_3\text{Ni}_2\text{O}_7$: LDA + FLEX calculations*, *Phys. Rev. B* **109**, 104508 (2024).
- [43] C. Xia, H. Liu, S. Zhou, and H. Chen, *Sensitive dependence of pairing symmetry on Ni- e_g crystal field splitting in the nickelate superconductor $\text{La}_3\text{Ni}_2\text{O}_7$* , *Nat. Commun.* **16**, 1054 (2025).
- [44] Y. H. Tian, Y. Chen, J. M. Wang, R. Q. He, and Z. Y. Lu, *Correlation effects and concomitant two-orbital $s_{\pm}$ -wave superconductivity in $\text{La}_3\text{Ni}_2\text{O}_7$ under high pressure*, *Phys. Rev. B* **109**, 165154 (2024).
- [45] Z. Luo, B. Lv, M. Wang, W. Wú, and D.-X. Yao, *High- T_c superconductivity in $\text{La}_3\text{Ni}_2\text{O}_7$ based on the bilayer two-orbital t - J model*, *npj Quantum Mater.* **9**, 61 (2024).
- [46] Z. Wang, H.-J. Zhang, K. Jiang, and F.-C. Zhang, *Self-doped molecular Mott insulator for bilayer high-temperature superconducting $\text{La}_3\text{Ni}_2\text{O}_7$* , *Natl. Sci. Rev.* **12**, nwaf353 (2025).
- [47] Y.-Y. Zheng and W. Wú, *$s_{\pm}$ -wave superconductivity in the bilayer two-orbital Hubbard model*, *Phys. Rev. B* **111**, 035108 (2025).
- [48] Z. Fan, J. F. Zhang, B. Zhan, D. Lv, X. Y. Jiang, B. Normand, and T. Xiang, *Superconductivity in nickelate and cuprate superconductors with strong bilayer coupling*, *Phys. Rev. B* **110**, 024514 (2024).
- [49] Z. Liao, L. Chen, G. Duan, Y. Wang, C. Liu, R. Yu, and Q. Si, *Electron correlations and superconductivity in $\text{La}_3\text{Ni}_2\text{O}_7$ under pressure tuning*, *Phys. Rev. B* **108**, 214522 (2023).
- [50] X.-Z. Qu, D.-W. Qu, J. Chen, C. Wu, F. Yang, W. Li, and G. Su, *Bilayer t - J - $J_{\perp}$ model and magnetically mediated pairing in the pressurized nickelate $\text{La}_3\text{Ni}_2\text{O}_7$* , *Phys. Rev. Lett.* **132**, 036502 (2024).
- [51] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/4h5x-btvx> for details on theoretical modeling, sample characterization, and ARPES analysis, which includes Refs. [14,52–56].
- [52] P. Giannozzi, O. Andreussi, T. Brumme, O. Bunau, M. Buongiorno Nardelli, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, M. Cococcioni *et al.*, *Advanced capabilities for materials modelling with Quantum ESPRESSO*, *J. Phys. Condens. Matter* **29**, 465901 (2017).
- [53] J. P. Perdew, K. Burke, and M. Ernzerhof, *Generalized gradient approximation made simple*, *Phys. Rev. Lett.* **77**, 3865 (1996).
- [54] H. J. Monkhorst and J. D. Pack, *Special points for Brillouin-zone integrations*, *Phys. Rev. B* **13**, 5188 (1976).
- [55] G. Pizzi, V. Vitale, R. Arita, S. Blügel, F. Freimuth, G. Géranton, M. Gibertini, D. Gresch, C. Johnson, T. Koretsune, and J. Ibañez-Azpiroz, *Wannier90 as a community code: new features and applications*, *J. Phys. Condens. Matter* **32**, 165902 (2020).
- [56] Y. Zhang, Y. Zhong, N. H. Tran, S. Li, K. Lee, Y. Lee, T. C. Wang, H. Y. Hwang, Z. X. Shen, and C. Jia, *Machine learning reconstruction of high-dimensional electronic structure from angle-resolved photoemission spectroscopy*, *arXiv:2603.16725*.
- [57] G. A. Pan, Q. Song, D. Ferenc Segedin, M. C. Jung, H. El-Sherif, E. E. Fleck, B. H. Goodge, S. Doyle, D. Córdoba Carrizales, A. T. N'Diaye *et al.*, *Synthesis and electronic properties of $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$ Ruddlesden-Popper nickelate thin films*, *Phys. Rev. Mater.* **6**, 055003 (2022).
- [58] Z. Li, W. Guo, T. Zhang, J. Song, T. Gao, Z. Gu, and Y. Nie, *Epitaxial growth and electronic structure of Ruddlesden-Popper nickelates ($\text{La}_{n+1}\text{Ni}_n\text{O}_{3n+1}$, $n = 1-5$)*, *APL Mater.* **8**, 091112 (2020).
- [59] Y. Liu, B. Y. Wang, J. Li, Y. Tarn, L. Bhatt, M. Colletta, Y. M. Wu, C. T. Kuo, J. S. Lee, B. H. Goodge *et al.*, *A superconducting half-dome in bilayer nickelates*, *arXiv:2603.12196*.
- [60] Y. Miao, R. Luan, Y. Chen, Z. Ou, G. Zhou, J. Shen, H. Wang, H. Huang, X. Wu, H. Sun *et al.*, *Electronic structures across superconductor-insulator transition in Ruddlesden-Popper bilayer nickelate films*, *arXiv:2502.18068*.
- [61] J. Shen, G. Zhou, Y. Miao, P. Li, Z. Ou, Y. Chen, Z. Wang, R. Luan, H. Sun, Z. Feng *et al.*, *Nodeless superconducting gap and electron-boson coupling in $(\text{La}, \text{Pr}, \text{Sm})_3\text{Ni}_2\text{O}_7$ films*, *Science* eadw8329 (2026).
- [62] S. N. Abadi, K.-J. Xu, E. G. Lomeli, P. Puphal, M. Isobe, Y. Zhong, A. V. Fedorov, S.-K. Mo, M. Hashimoto, D.-H. Lu, B. Moritz, B. Keimer, T. P. Devereaux, M. Hepting, and Z. X. Shen, *Electronic structure of the alternating monolayer-trilayer phase of $\text{La}_3\text{Ni}_2\text{O}_7$* , *Phys. Rev. Lett.* **134**, 126001 (2025).
- [63] T. Xie, M. Huo, X. Ni, F. Shen, X. Huang, H. Sun, H. C. Walker, D. Adroja, D. Yu, B. Shen *et al.*, *Strong interlayer magnetic exchange coupling in $\text{La}_3\text{Ni}_2\text{O}_{7-\delta}$ revealed by inelastic neutron scattering*, *Sci. Bull.* **69**, 3221 (2024).
- [64] X. Chen, J. Choi, Z. Jiang, J. Mei, K. Jiang, J. Li, S. Agrestini, M. Garcia-Fernandez, H. Sun, X. Huang *et al.*, *Electronic and magnetic excitations in $\text{La}_3\text{Ni}_2\text{O}_7$* , *Nat. Commun.* **15**, 9597 (2024).
- [65] X. Chen, S. Maiti, A. Linscheid, and P. J. Hirschfeld, *Electron pairing in the presence of incipient bands in iron-based superconductors*, *Phys. Rev. B* **92**, 224514 (2015).
- [66] W. Sun, Z. Jiang, B. Hao, S. Yan, H. Zhang, M. Wang, Y. Yang, H. Sun, Z. Liu, D. Ji *et al.*, *Observation of superconductivity-induced leading-edge gap in Sr-doped $\text{La}_3\text{Ni}_2\text{O}_7$ thin films*, *arXiv:2507.07409*.
- [67] P. Li, G. Zhou, W. Lv, Y. Li, C. Yue, H. Huang, L. Xu, J. Shen, Y. Miao, W. Song *et al.*, *Angle-resolved photoemission*

- spectroscopy of superconducting (La, Pr)₃Ni₂O₇/SrLaAlO₄ heterostructures*, *Natl. Sci. Rev.* **12**, nwaf205 (2025).
- [68] E. A. Stepanov, S. Bötzel, I. M. Eremin, and F. Lechermann, *Co-operating multi-orbital and nonlocal correlations in bilayer nickelate*, [arXiv:2604.08221](#).
- [69] M. P. A. Fisher, G. Grinstein, and S. M. Girvin, *Presence of quantum diffusion in two dimensions: Universal resistance at the superconductor-insulator transition*, *Phys. Rev. Lett.* **64**, 587 (1990).
- [70] W. H. Tang, C. Y. Ng, C. Y. Yau, and J. Gao, *Thickness dependence of superconductivity for YBa₂Cu₃O_y ultra-thin films*, *Supercond. Sci. Technol.* **13**, 580 (2000).
- [71] H. Sato, *Thickness dependence of superconductivity and resistivity in La_{1.85}Sr_{0.15}CuO₄ films*, *Physica C* **468**, 991 (2008).
- [72] Y. Y. Zong, S. L. Yu, and J. X. Li, *Correlation-driven orbital-selective fermiology and superconductivity in the bilayer nickelate La₃Ni₂O₇*, [arXiv:2605.10101](#).
- [73] S. Moser, *An experimentalist's guide to the matrix element in angle resolved photoemission*, *J. Electron Spectrosc. Relat. Phenom.* **214**, 29 (2017).
- [74] Y. Li, L. Xu, W. Lv, Z. Nie, Z. Wang, Y. Miao, J. Shen, G. Zhou, W. Song, H. Wang *et al.*, *Three-dimensional electronic structures in superconducting Ruddlesden-Popper bilayer nickelate films*, [arXiv:2604.08430](#).