

Three-Dimensional Electronic Structures in Superconducting Ruddlesden-Popper Bilayer Nickelate Films

Yueying Li,^{1,2,*} Lizhi Xu^{1,*}, Wei Lv,^{1,*} Zihao Nie¹, Zechao Wang¹, Yu Miao,³ Jianchang Shen³, Guangdi Zhou,^{1,2} Wenhua Song,¹ Heng Wang^{1,2}, Haoliang Huang^{1,2}, Junfeng He,³ Jin-Feng Jia,^{1,2,4} Peng Li^{1,2,†}, Qi-Kun Xue,^{1,2,5,‡} and Zhuoyu Chen^{1,2,§}

¹State Key Laboratory of Quantum Functional Materials, Department of Physics, Guangdong Provincial Key Laboratory of Topological Matter, Guangdong Basic Research Center of Excellence for Quantum Science, and College of Semiconductors (National Graduate College for Engineers), Southern University of Science and Technology, Shenzhen 518055, China

²Quantum Science Center of Guangdong-Hong Kong-Macao Greater Bay Area, Shenzhen 518045, China

³Department of Physics, University of Science and Technology of China, Hefei 230026, China

⁴State Key Laboratory of Micronano Engineering Science, Tsung-Dao Lee Institute and School of Physics and Astronomy, Key Laboratory of Artificial Structures and Quantum Control (Ministry of Education), Shanghai Jiao Tong University, Shanghai 200240, China

⁵Department of Physics, Tsinghua University, Beijing 100084, China



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Beyond the quasi-two-dimensional (2D) paradigm of cuprates, the role of the third dimension of the Ruddlesden-Popper bilayer nickelates is essential to decoding their superconducting mechanism. Here, using angle-resolved photoemission spectroscopy (ARPES) with varied photon energies, we systematically investigate the electronic band structures in three dimensions for superconducting (La, Pr, Sm)₃Ni₂O₇/SrLaAlO₄ thin films (superconducting onset temperature $T_c^{\text{onset}} \sim 48$ K) transferred via a cryogenic ultrahigh vacuum (UHV) suitcase. We reveal an orbital-dependent dimensionality: While the $d_{x^2-y^2}$ -dominant bands exhibit a quasi-2D character, the d_{z^2} -dominant band displays a finite k_z dispersion. Finite energy gaps are identified on all observed bands across multiple high-symmetry directions. Systematic temperature-dependent analysis characterizes the superconducting nature of the gap on the d_{z^2} -derived band, revealing a large gap $\Delta \sim 18$ meV and a ratio $2\Delta/k_B T_c \sim 8$ exceeding the weak-coupling BCS limit. The suppression of spectral weight near the Fermi level persists above the superconducting transition temperature. Ubiquitous vertical spectral features evidence the presence of electron interactions. These results underscore the role of the d_{z^2} orbital and correlations, placing constraints on theoretical models for nickelate superconductivity.

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The discovery of Ruddlesden-Popper (RP) nickelates superconductors has provided a unique platform to explore the unconventional superconductivity mechanisms. Hosting both $d_{x^2-y^2}$ and d_{z^2} orbitals near the Fermi level, the

RP nickelates represent a distinct multiorbital system that serves as an unprecedented opportunity to bridge the understanding of cuprate and iron-based high-temperature superconductivity [1,2]. Crucially, this physics is tied to the behavior of the d_{z^2} -orbital-derived γ band, which remains theoretically debated [1,3–8] and lacks comprehensive experimental investigations.

Since the discovery of high-temperature superconductivity in RP bilayer bulk crystals under high pressure [9–11], remarkable experimental progress has been achieved in this field. The high-pressure superconductivity is expanded to other RP phase and hybrid RP phase nickelates [12–16]. The ambient-pressure superconductivity is also achieved in RP epitaxial films [17–19], which facilitates the application of some experimental

*These authors contributed equally to this work.

†Contact author: lipeng@quantumsc.cn

‡Contact author: xueqk@sustech.edu.cn

§Contact author: chenzhuoyu@sustech.edu.cn

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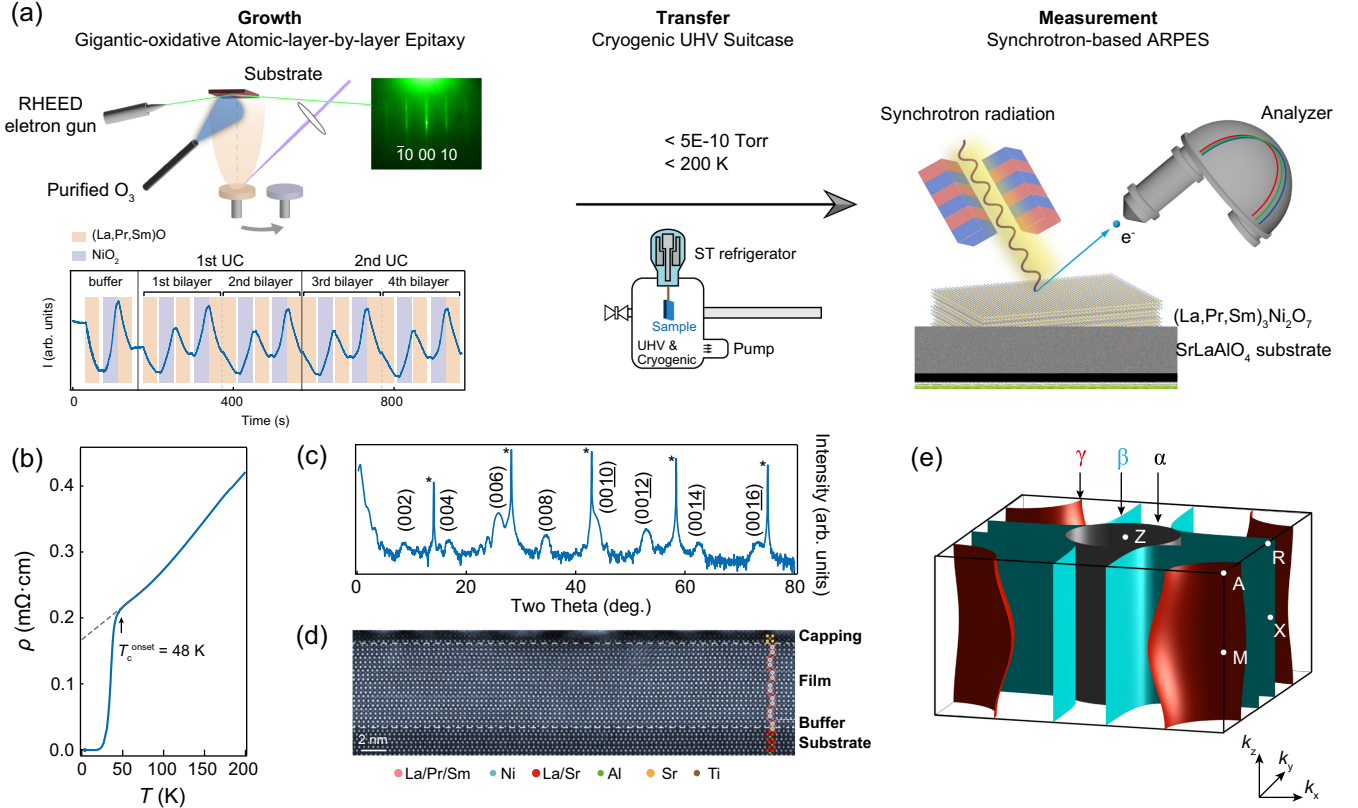


FIG. 1. Experimental procedures of this study. (a) Schematic diagram of the workflow. The samples were grown and optimized by the GAE technique, then *in situ* transferred via a UHV cryogenic suitcase with the film preserved below 200 K, and finally measured using synchrotron radiation ARPES. (b) Temperature-dependent resistivity curve of the (La, Pr, Sm)₃Ni₂O₇ film (thickness of 3 UC). The T_c^{onset} is defined as the temperature where resistivity deviated from the linear fit, shown as the dashed line. (c) X-ray diffraction 2θ - ω scan along the c axis of the (La, Pr, Sm)₃Ni₂O₇ film (thickness of 3 UC) grown on a SrLaAlO₄ substrate (diffraction peaks marked by asterisks). (d) STEM-HAADF image of the (La, Pr, Sm)₃Ni₂O₇ film. The sample is capped with a SrTiO₃ layer to prevent damage during specimen preparation by a focused ion beam. (e) Schematic of the 3D Fermi surface observed in (La, Pr, Sm)₃Ni₂O₇ films. The high-symmetry points are labeled in the Brillouin zone.

technologies, like angle-resolved photoemission spectroscopy (ARPES), to directly probe the electronic structures of the superconducting state [20–24]. However, fundamental questions regarding the dimensionality of the electronic states remain unresolved due to the lack of k_z -resolved spectroscopic evidence. Clarifying these aspects is pivotal for distinguishing between competing theoretical models [3,4,25–32].

In this work, we systematically resolve the three-dimensional electronic structure of superconducting bilayer nickelate films using synchrotron-based ARPES. Crucial to this experiment is the use of a cryogenic ultrahigh vacuum suitcase for sample transfer, which minimizes surface oxygen loss and preserves the intrinsic nature of the surface-sensitive d_{z^2} -derived γ band. Enabled by the pristine sample quality, we reveal an orbital-dependent dimensionality: The quasi-2D $d_{x^2-y^2}$ bands contrast with the γ band, which exhibits a distinct k_z dispersion. Critically, the large superconducting gap on the γ band unambiguously confirms its contribution to the superconductivity. This large gap, together with waterfall features, implies the

role of electron correlations. These findings highlight the shared correlation physics with cuprates while demarcating the unique three-dimensional, multiorbital nature of the nickelates.

Figure 1 summarizes the experimental workflow of this study. As shown in Fig. 1(a), using the gigantic-oxidative atomic-layer-by-layer epitaxy (GAE) method [33], superconducting (La, Pr, Sm)₃Ni₂O₇ films [(La: Pr : Sm = 4:1:1)] are grown under ultrastrong oxidation, monitored in real time by reflection high-energy electron diffraction (RHEED) (see Supplemental Material for details of sample optimization [34]). The RHEED pattern displays sharp spots, and the corresponding diffraction intensities exhibit periodic oscillations with negligible decay, signifying highly crystalline surface and atomic layer-by-layer growth. X-ray diffraction (XRD) with a series of well-defined diffraction peaks and scanning transmission electron microscopy high-angle annular dark-field (STEM-HAADF) images confirm the absence of impurity phases across macroscopic and microscopic scales, respectively [Figs. 1(c) and 1(d)]. Transport measurements identify

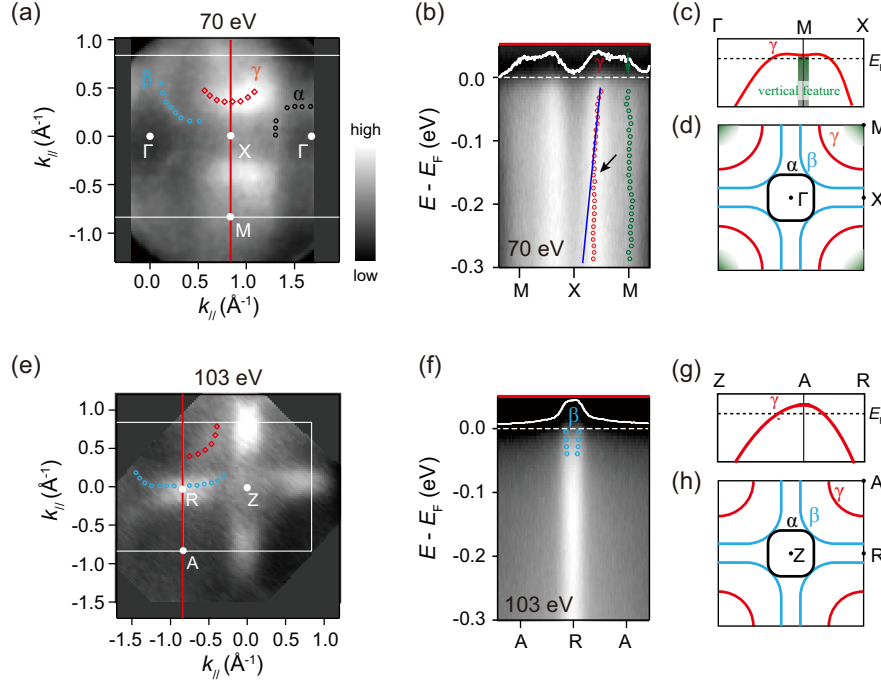


FIG. 2. Fermi surfaces and high-symmetry cuts at $k_z \sim 0$ and $k_z \sim \pi$. (a),(e) Fermi surface maps measured at 70 eV ($k_z \sim 0$) and 103 eV ($k_z \sim \pi$), respectively. The black, light blue, and red dots indicate the peak positions of the α , β , and γ pockets, respectively. (b),(f) ARPES spectra along the red lines in panels (a) and (e) (along the $\bar{M}$ - $\bar{X}$ direction) measured at 70 eV and 103 eV, respectively. The black arrow in panel (b) shows the kink position at around 150 meV below Fermi energy, which is the waterfall electronic structure at the γ band caused by electron correlation. The blue line is the extended dispersion from the low-energy part of the γ band (d_{z^2} orbital). The green circles present the vertical feature at the M point. (c),(g) Schematic illustration of the γ band dispersion at $k_z \sim 0$ and $k_z \sim \pi$, respectively. (d),(h) Schematics of the fermiology at $k_z \sim 0$ and $k_z \sim \pi$, respectively. The color bar reflects the intensity of ARPES spectra. The film thickness of this $(\text{La, Pr, Sm})_3\text{Ni}_2\text{O}_7$ sample is 2 UC.

a superconducting onset temperature (T_c^{onset}) of 48 K [Fig. 1(b)]. Crucially, the as-grown superconducting samples were quenched below 200 K in a UHV cryogenic suitcase and then transferred to an ARPES endstation. This low-temperature protocol effectively suppresses the oxygen loss during transit [37], enabling a reliable and systematic 3D probe of the electronic structure via photo-energy-dependent ARPES.

To characterize the Fermi surface topography, two representative photon energies of 70 eV ($k_z \sim 0$) and 103 eV ($k_z \sim \pi$) were selected, as shown in Figs. 2(a) and 2(e). For both photon energies, the γ band (red diamonds) consistently crosses the Fermi level, forming a pocket centered at the $\bar{M}$ point, in agreement with our previous reports and theoretical calculations [6,20,24]. Notably, the spectral intensity exhibits strong photon-energy dependence: The γ band appears intense at 70 eV, whereas the β band becomes more pronounced at 103 eV. This contrast is attributed to matrix element effects and distinct photoemission cross sections of the d_{z^2} and $d_{x^2-y^2}$ orbitals at different photon energies [38]. The intensity variation is further elucidated by spectra along the $\bar{M}$ - $\bar{X}$ direction [Figs. 2(b) and 2(f)].

The correlation effects manifest in the band dispersion along the high-symmetry directions where the γ band

displays vertical intensity tails at higher binding energies [black arrow in Fig. 2(b)] (see Fig. S2 for self-energy analysis [34]). The experimental band dispersions are compared with the DFT calculations [6] to extract the band renormalization factor of approximately 3 corresponding to the previous reports [23,39,40] (Fig. S3 [34]). Beyond selective visibility of the γ and β bands in the 70- and 103-eV spectra, a pronounced difference appears near the Brillouin zone corner: The 70-eV data exhibit an intense vertical feature at the M point, whereas this feature is absent at the A point for the 103-eV spectra. This vertical spectral feature may be interpreted as “waterfalls” associated with the γ band top, as schematically illustrated in the right panels of Figs. 2(c) and 2(g). Theoretical calculations suggest the existence of a local electronlike dip at the γ band top for $k_z \sim 0$ [41–43], which was also recently observed experimentally [23]. In the presence of electron correlations, such a flat band with a local electronlike curvature could manifest waterfall-like redistributions of spectral weights [44,45], while the conventional parabolic band dispersion at $k_z \sim \pi$ precludes such a feature.

Figure 3 presents the $k_z - k_{\parallel}$ Fermi surface map and momentum distribution curves at the Fermi level (E_F) obtained from photon-energy-dependent measurements along $\bar{M}$ - $\bar{X}$, overlaid with the fitted peak positions of the

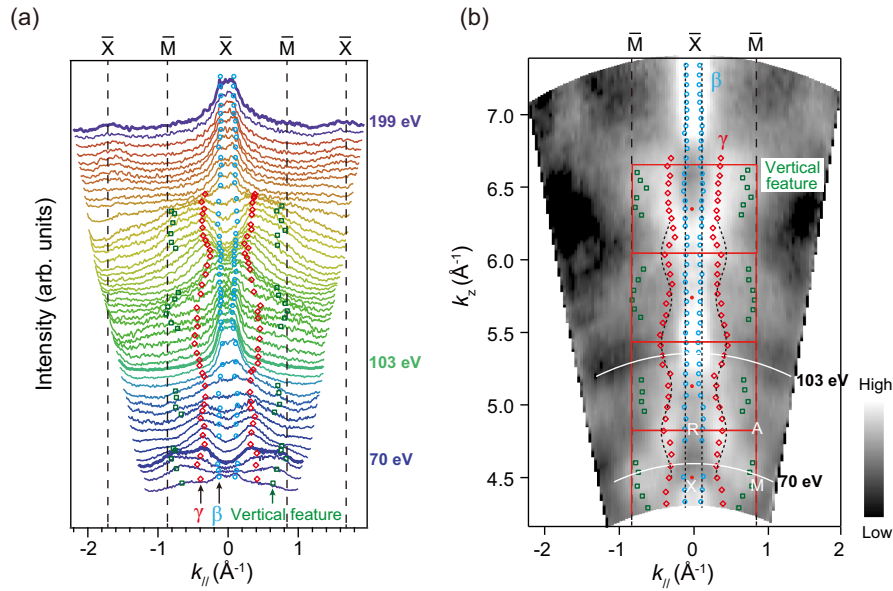


FIG. 3. Three-dimensional electronic structure of superconducting $(\text{La, Pr, Sm})_3\text{Ni}_2\text{O}_7$ films with a thickness of 2UC. (a) Momentum distribution curves at E_F of the photon-energy-dependent measurements in the $\bar{M}$ - $\bar{X}$ plane. The integrated energy range is from -30 meV to the Fermi level. The light blue, red, and green dots represent the positions of the β band, the γ band, and the vertical feature, respectively. (b) Corresponding k_z - $k_{\parallel}$ map at E_F in the $\bar{M}$ - $\bar{X}$ plane. The overlapped white curves indicate the k_z positions of 103 eV and 70 eV. The color bar reflects the intensity of ARPES spectra.

β , γ bands, and the vertical feature. Detailed photon-energy-dependent spectra are provided in Fig. S4 [34]. These results reveal a contrast in dimensionality: The β band, with predominantly $d_{x^2-y^2}$ character, exhibits negligible k_z dispersion, whereas the d_{z^2} -derived γ band displays a distinct k_z dispersion while consistently crossing E_F . These observations align with theoretical calculations [46]. The vertical feature near the M point displays a discontinuous evolution along the k_z direction. It should be noted that the periodicity of the γ band along the k_z direction follows the spatial periodicity of the bilayer structure (i.e., half the unit cell). Consequently, the 2 UC film investigated here encompasses four complete dispersion periods along the out-of-plane direction, providing sufficient thickness to establish a quasicontinuous k_z dispersion, which is further confirmed in another 3 UC film exhibiting quite similar behavior (Fig. S5 [34]). This periodicity is consistent with a physical picture involving bonding and antibonding d_{z^2} states within a bilayer, coupled with relatively weak interactions between adjacent bilayers. Analogous k_z periodicity has been reported in other bilayer systems [47,48].

Furthermore, the momentum distribution of superconducting gaps is systematically detected. As shown in Figs. 4(a) and 4(b), finite gaps are observed on all bands (α , β , and γ) along high-symmetry directions. We perform detailed temperature-dependent analysis specifically on the γ band along the $\bar{M}$ - $\bar{X}$ - $\bar{M}$ direction using 70-eV photons. The symmetrized energy distribution curves (EDCs) show that a gap opens in the superconducting state [Fig. 4(c)]. The gap size is determined to be $\Delta \sim 18$ meV at 9.5 K by

fitting the symmetrized EDCs with a Bardeen-Cooper-Schrieffer (BCS)-based phenomenological model convolved with an experimental energy resolution of around 9 meV and the corresponding measurement temperature [49]. This case yields a ratio of $2\Delta/k_B T_c \sim 8$, which significantly exceeds the weak-coupling BCS value of 3.5 and corresponds well with the theoretical result [50]. The temperature evolution of the gap is detailed in Figs. 4(d) and 4(e). Upon increasing temperature, the superconducting gap gradually decreases. However, the variation of the dip intensity at the Fermi level (I_{dip}) does not fully saturate at T_c^{onset} . This suppression of spectral weight near the Fermi level persists up to approximately 90 K for the γ band, suggesting possible pseudogap behavior similar to that previously reported in the β band [21]. These temperature-dependent phenomena were consistently observed across two independent samples (#S1 and #S2). To further validate the findings, transport measurements were performed on #S2 immediately after the ARPES experiments, confirming a macroscopic superconducting transition onset at around 45 K, demonstrating that the samples maintain their robust superconductivity throughout the measurement process.

The systematic mapping of the 3D electronic structure in bilayer RP nickelates allows us to discuss two key implications regarding their microscopic physics. First, regarding the electronic dimensionality, the distinct k_z dispersion of the d_{z^2} -derived γ band underscores the non-negligible interlayer orbital coupling. In this context, the apparent disappearance of the γ band at specific photon energies

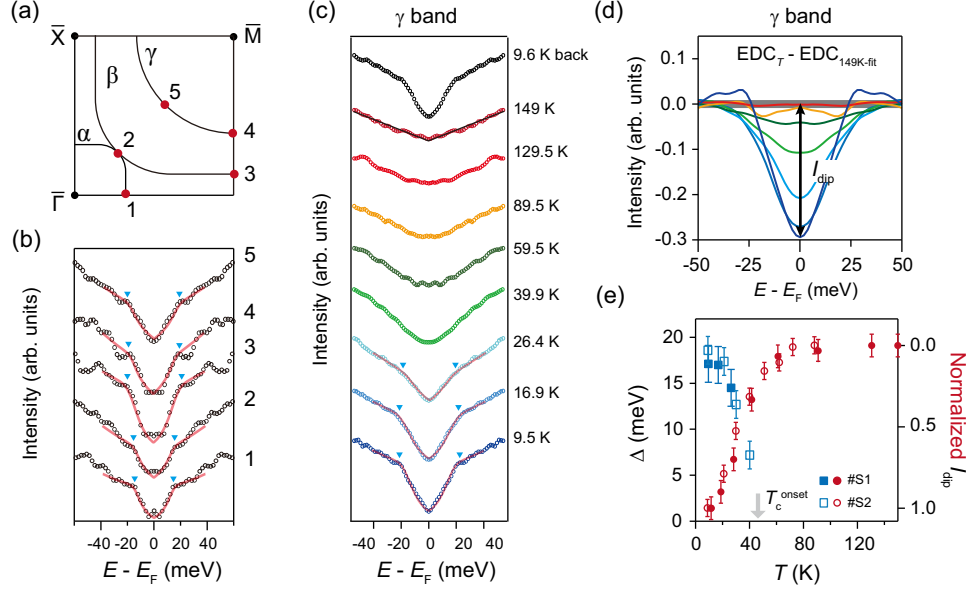


FIG. 4. Superconducting gap analysis. (a) Schematic Fermi surface map showing the high-symmetry directions and labeled momentum positions. (b) Momentum-dependent symmetrized EDCs at k_F for the positions 1–5 indicated in panel (a). (c) Temperature evolution of the symmetrized EDCs at k_F for the γ band at point 4. Overlaid red curves represent fits using a phenomenological BCS-based model, and black lines at 149 K indicate the linear-fit background. (d) Overlapped spectra from panel (c) after subtracting the 149-K linear-fit background, allowing clearer identification of the gap opening and coherence peaks. The black arrow denotes the spectral dips. (e) Extracted superconducting gap size (Δ) and the normalized dip intensity (I_{dip}) as a function of temperature. The gray arrow indicates T_c^{onset} . Data in panel (b) are from sample #S2; data in panels (c) and (d) are from sample #S1; solid and open symbols in panel (e) represent independent measurements from samples #S1 and #S2, respectively.

should be interpreted as a consequence of strong k_z -dependent matrix element effects rather than an intrinsic absence from the Fermi surface. This finding confirms that the electronic structure possesses non-negligible three-dimensional character, necessitating a framework beyond the strictly two-dimensional limit. Second, regarding the electron correlations, our results reveal significant deviations from standard mean-field descriptions. The ubiquitous vertical features, combined with a large superconducting gap ratio, indicate non-negligible correlation effects. This sizable gap identifies the d_{z^2} orbital as an active participant in the superconducting pairing, suggesting that a multiorbital physics incorporating electron correlations is essential for understanding the pairing mechanism. While our observations establish the critical contribution of the γ band to superconductivity in $(\text{La, Pr, Sm})_3\text{Ni}_2\text{O}_7$ films, its universality as a prerequisite for superconductivity in the broader nickelate family warrants further investigation across different phases. Nevertheless, recent systematic studies on a series of Ruddlesden-Popper nickelate thin-film superstructures suggest that the presence of the γ band at the Fermi level is correlated with the emergence of superconductivity [24].

In summary, by employing a cryogenic ultrahigh vacuum suitcase to preserve pristine sample surfaces, we have systematically resolved the three-dimensional electronic structure of pure-phase superconducting RP bilayer

nickelate films. Our findings reveal an orbital-selective dimensionality, where the d_{z^2} -dominant band exhibits significant k_z dispersion in contrast with the quasi-two-dimensional character of the $d_{x^2-y^2}$ -dominant bands. The identification of finite energy gaps across all observed bands suggests a nodeless pairing scenario, while the large gap ratio ($2\Delta/k_B T_c \sim 8$) and vertical or waterfall features provide evidence for electron correlations. These findings highlight the indispensability roles of the third dimension and the d_{z^2} orbital in the mechanism of nickelate superconductivity.

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Q. K. X. supervised the entire project. Z. C. and P. L. initiated the study and coordinated all the research efforts. Y. L., L. X., and P. L. conducted the ARPES measurements, with assistance from Y. M., J. S., and W. S., under supervision from P. L., Z. C., and J. H. W. L. performed thin-film growth, with assistance from Z. N. and Y. C., under supervision from G. Z. and Z. C. Z. W. performed STEM measurements. W. L. and H. W. performed the transport properties measurements. J.-F. J. and all other authors participated in discussions. P. L., Y. L., and Z. C. wrote the manuscript with input from all other authors. Y. L., L. X., and W. L. contributed equally.

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.

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