

Distinct orbital contributions to electronic and magnetic structures in $\text{La}_4\text{Ni}_3\text{O}_{10}$ Shilong Zhang^{1,*}, Hengyuang Zhang^{2,*}, Zehao Dong³, Jie Li⁴, Qian Xiao^{1,5}, Mengwu Huo², Hsiao-Yu Huang⁶, Di-Jing Huang⁶, Yayu Wang³, Yi Lu⁴, Zhen Chen⁷, Meng Wang^{2,†}, and Yingying Peng^{1,8,‡}¹International Center for Quantum Materials, School of Physics, *Peking University*, Beijing 100871, China²Guangdong Provincial Key Laboratory of Magnetoelectric Physics and Devices, School of Physics, *Sun Yat-Sen University*, Guangzhou 510275, China³State Key Laboratory of Low Dimensional Quantum Physics, Department of Physics, *Tsinghua University*, Beijing 100084, China⁴National Laboratory of Solid State Microstructures and Department of Physics, *Nanjing University*, Nanjing 210093, China⁵School of Physics and Information Engineering, *Guangdong University of Education*, Guangzhou 510303, China⁶National Synchrotron Radiation Research Center, Hsinchu 30076, Taiwan⁷Beijing National Laboratory for Condensed Matter Physics, *Institute of Physics, Chinese Academy of Sciences*, Beijing 100190, China⁸Collaborative Innovation Center of Quantum Matter, Beijing 100871, China

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High- T_c superconductivity has recently been discovered in Ruddlesden-Popper (RP)-phase nickelates under pressure, where the low-energy electronic structure is dominated by Ni $d_{x^2-y^2}$ and d_{z^2} orbitals. However, the respective roles of these orbitals in superconductivity remain unclear. Here, by combining x-ray absorption, electron energy loss spectroscopy, and density functional theory calculations on $\text{La}_4\text{Ni}_3\text{O}_{10}$ single crystals, we identify ligand holes in the $p_{x,y}$ orbitals of planar oxygen and the p_z orbitals of apical oxygen, which hybridize with the Ni $d_{x^2-y^2}$ and d_{z^2} orbitals, respectively. These ligand holes enable orbital-selective O K -edge resonant inelastic x-ray scattering (RIXS) study, which reveals that $d_{x^2-y^2}$ states dominate the low-energy charge excitations and are more itinerant. We also observe an ~ 0.1 eV bimagnon through RIXS and Raman spectroscopy. Our results reveal distinct contributions of Ni $d_{x^2-y^2}$ and d_{z^2} orbitals to the electronic and magnetic structure and provide direct experimental insights to understand the RP-phase nickelate superconductors.

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Recently, superconductivity has been discovered in bilayer and trilayer Ruddlesden-Popper (RP) nickelate single crystals under pressure, with maximum transition temperatures T_c of about 80 and 30 K, respectively [1–7]. Remarkably, ambient-pressure superconductivity ($T_c \approx 40$ K) has been realized in ultrathin $(\text{La}, \text{Pr})_3\text{Ni}_2\text{O}_7$ (LPNO) films via strain engineering [8,9]. These RP-phase nickelates possess $3d^7$ to $3d^8$ electron occupancy and additional interlayer apical oxygen atoms, which distinguish them from the infinite-layer nickelate superconductors RNiO_2 [10–13]. To understand the superconducting mechanism, extensive research on RP-phase nickelates has investigated their macroscopic and microscopic electronic and magnetic structures. Angle-resolved photoelectron spectroscopy reveals a multiband structure, with density functional theory (DFT) calculations attributing these bands to Ni $3d_{x^2-y^2}$ and $3d_{z^2}$ orbitals [14–16]. Experiments indicate that RP-phase nickelates exhibit a spin density wave (SDW) state at ambient pressure, which vanishes under pressure [17–21].

Resonant inelastic x-ray scattering (RIXS) at the Ni L edge has observed dispersive magnon up to 70 and 50 meV in bilayer and trilayer RP-phase nickelates, respectively [17,22]. Various theoretical models have been proposed to explain the superconducting pairing mechanism [23–35]. These theories suggest that interlayer hopping is essential to the pairing mechanism, but there remains a key open question as to whether superconducting pairing originates from the d_{z^2} or the $d_{x^2-y^2}$ orbitals. Therefore, it is essential to experimentally identify the electronic states of the d_{z^2} and $d_{x^2-y^2}$ orbitals to understand RP-phase nickelate superconductors.

However, distinguishing the contributions of the Ni $3d_{z^2}$ and $3d_{x^2-y^2}$ orbitals remains challenging, as both exhibit a substantial density of states near the Fermi energy. In Ni L_3 -edge RIXS measurements, electrons can be excited into the Ni $3d_{z^2}$ and $3d_{x^2-y^2}$ orbitals simultaneously, obscuring the orbital origin of the observed magnetic excitations. Previous electron energy loss spectroscopy (EELS) studies on $\text{La}_3\text{Ni}_2\text{O}_7$ have revealed ligand holes in both planar and apical oxygens [36,37]. These states can hybridize separately to the Ni $3d_{x^2-y^2}$ and $3d_{z^2}$ orbitals, offering an indirect route to probe these orbitals individually. Moreover, employing different polarizations in the RIXS study, it is possible to excite electrons into different O $2p$ orbitals [38], providing orbital-sensitive information in the energy loss spectra [39].

In this work, we combine O K -edge x-ray absorption spectroscopy (XAS), RIXS, EELS, and Raman spectroscopy to investigate a trilayer RP-phase nickelate $\text{La}_4\text{Ni}_3\text{O}_{10}$ single

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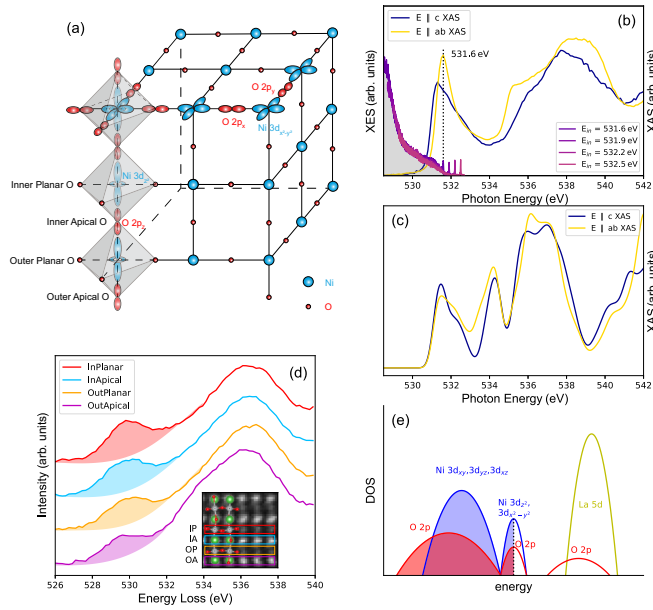


FIG. 1. (a) Schematic illustration of the La₄Ni₃O₁₀ Ruddlesden-Popper structure, highlighting the outer-apical, inner-apical, and planar oxygen sites, together with the atomic orbitals contributing near the Fermi level. (b) The XAS spectra at O *K* edge along in-plane *ab* direction and *c* direction and XES spectra obtained from the RIXS spectra. The dashed line marks the Fermi level obtained by XES. These indicate that both O *p_{x,y}* and *p_z* orbitals have ligand holes at the Fermi energy. (c) Calculated XAS spectra along in-plane and out-of-plane directions, which indicate that the holes are distributed in the orbitals shown in panel (a). (d) Occupancy of oxygen holes at different positions obtained by EELS. The small energy discrepancy in the prepeak compared with the XAS feature is due to different instrumental calibrations. (e) Schematic plot of DOS in La₄Ni₃O₁₀, showing that the oxygen 2*p* orbital is partially filled.

crystal [40]. Our results reveal that the ligand holes reside in the planar O *p_{x,y}* orbitals and the apical O *p_z* orbitals via hybridization with the Ni 3*d_{x²-y²}* and 3*d_{z²}* orbitals, respectively. Further investigation of excited states reveals that the *d_{x²-y²}* orbitals dominate low-energy charge excitations and are more itinerant than the *d_{z²}* orbitals. We also observed an ~ 0.1 eV bimagnon from the intralayer spin exchange, which places an upper bound of $J_z \sim 50$ meV on the interlayer superexchange interaction. Raman measurements at varying temperatures show that the bimagnon weakens significantly with increasing temperature but does not disappear above T_{SDW} , indicating robust short-range magnetic fluctuations. These results reveal the distinct orbital contributions to the electronic and magnetic structure and advance our understanding of the RP-phase nickelate superconductors.

To probe the oxygen states in La₄Ni₃O₁₀ near the Fermi level, we performed O *K*-edge XAS and RIXS measurements. The trilayer structure is illustrated in Fig. 1(a), using the *P1*/mmm lattice [40] with $a \approx b \approx 3.9$ Å and $c \approx 27.9$ Å; reciprocal lattice units (r.l.u.) are defined accordingly, and the scattering plane lies in the *ac* plane. By measuring the x-ray absorption spectra at different incident angles and polarizations, we decomposed the absorption spectra of La₄Ni₃O₁₀ into in-plane *ab* and out-of-plane *c* directions, as shown in

Fig. 1(b). The XAS spectra show strong prepeak in both directions, indicating that holes are present in the O *p_x*, *p_y*, and *p_z* orbitals. We measured O *K*-edge RIXS spectra at different incident x-ray energies and obtained fluorescence excitation spectra, which are analogous to x-ray emission spectroscopy (XES) spectra and provide information on occupied states. The cutoff energy of these fluorescence excitations identifies the Fermi energy and aligns with the XAS prepeak, suggesting that the oxygen holes are located at the Fermi level [41].

We also performed XAS calculations for in-plane and out-of-plane directions [Fig. 1(c)]. It generally reproduces the experimental line shapes, with the discrepancy reflecting the well-known tendency of DFT-based O *K*-edge simulations to produce a compressed energy scale relative to the experiment [42,43]. In the calculations, the lowest-energy prepeak in the oxygen *K*-edge XAS primarily involves planar O 2*p_{x,y}* hybridized with Ni *d_{x²-y²}* for electric field *E*//*ab* and apical O 2*p_z* hybridized with Ni *d_{3z²-r²}* for *E*//*c*, while higher-energy features include additional contributions from La 5*d* states. The orbital-resolved decomposition of the simulated XAS spectra is shown in the Fig. S5 of the Supplemental Material [44]. Our O *K*-edge EELS measurements on La₄Ni₃O₁₀ reveal the spatial distribution of oxygen holes, as shown in Fig. 1(d). Prepeaks are observed in both planar and inner apical oxygens, consistent with previous results on La₃Ni₂O₇ [36]. A weaker and lower-energy prepeak appears in the outer apical oxygen, differing from La₃Ni₂O₇. Based on these results, we constructed a schematic diagram of the density of states (DOS) in Fig. 1(e). Near the Fermi energy, in addition to the Ni *d_{x²-y²}* and *d_{z²}* orbitals, there are also ligand hole states in the oxygen 2*p* orbitals. These ligand hole states are located on the planar oxygen *p_{x,y}* orbitals and the apical oxygen *p_z* orbitals, which hybridize with the intralayer Ni 3*d_{x²-y²}* orbitals and the interlayer Ni 3*d_{z²}* orbitals, respectively. These ligand holes enable the independent probing of the 3*d_{x²-y²}* and 3*d_{z²}* orbitals by separately exciting distinct oxygen atomic orbitals.

We then performed RIXS measurement with different incident x-ray polarizations to identify excitations from in-plane and out-of-plane O *p* orbitals. The experimental geometry is shown in Fig. 2(a) and polarizations are shown in Fig. 2(b). The σ -polarization direction (perpendicular to the scattering plane) always aligned within the *ab* plane of the sample, while the π -polarization direction (parallel to the scattering plane) has an out-of-plane projection depending on the incident angle. By scanning the incident x-ray energy at π polarization, we identify several features with constant energy loss: two high-energy excitations at ~ 0.5 and ~ 1.0 eV [Fig. 2(c)], and two low-energy modes at ~ 50 and ~ 100 meV [Fig. 2(d)].

To determine their orbital character, we then performed *H*-dependent and 2θ -dependent RIXS measurements. For *H*-dependent studies, the scattering angle 2θ was fixed at 150° , while varying the in-plane momentum transfer *H* along *a*-axis direction. For 2θ -dependent measurements, we fixed $H = -0.05$ r.l.u. while scanning 2θ . We first focus on the high-energy spectra and found a pronounce 0.5 eV excitation with distinct polarization dependence. Its intensity remains constant under σ polarization [Figs. 3(a) and 3(b)] but shows significant changes under π polarization [Figs. 3(c) and 3(d)]. The integration of the π -polarized spectral weight reveals an angular dependence well described by $A \sin^2 \alpha \sin^2 \beta + c$

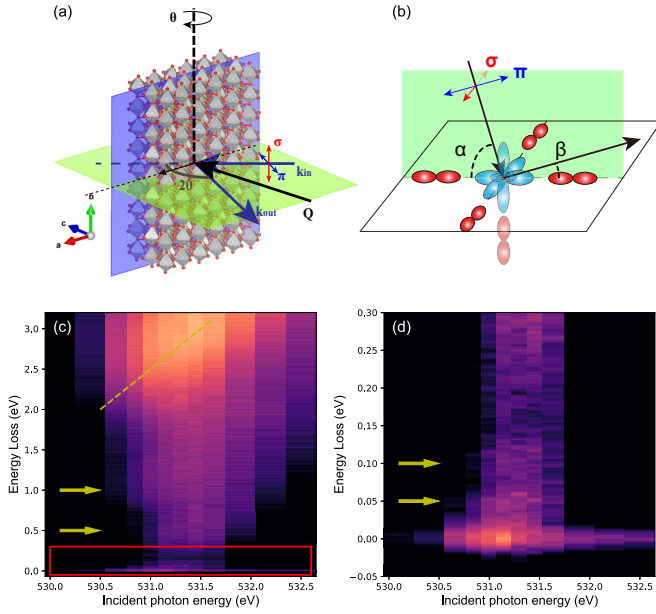


FIG. 2. (a) Schematic plot of scattering geometry with lattice orientation. We define the ab plane as the planar Ni–O plane and a direction along the Ni–O–Ni direction, while c denotes the direction perpendicular to the planar Ni–O planes. (b) Schematic plot of scattering plane with different polarizations. (c) Incident energy detuning map of RIXS, where two Raman features are found at about 0.5 and 1 eV. (d) Close-up of the red boxed region in panel (c) showing low-energy Raman excitations at around 50 and 100 meV.

[Figs. 3(e) and 3(f)], where α and β represent the incoming and outgoing angles [see Fig. 2(b)]. We notice that for charge excitations originating from in-plane orbitals, the π – π' channel will exhibit a polarization factor of $\sin^2 \alpha \sin^2 \beta$, while the σ – σ' channel remains constant, reflecting the projection of the electric field onto the orbital plane [45,46]. This polarization dependence is consistent with our experimental observations, and considering the atomic orbitals near the Fermi energy presented in Fig. 1(a), we then assign this excitation to a charge excitation involving planar O $p_{x,y}$ and Ni $d_{x^2-y^2}$ orbitals. We observed a less pronounced feature above 1 eV, which can be observed only under grazing incidence angle with π -polarization conditions [Fig. 3(c)], suggesting its origin from out-of-plane orbitals. The different behaviors between the in-plane components and the out-of-plane components suggest that these two orbitals do not hybridize together.

For the low-energy spectra, we resolved three phonons, a broad excitation near ~ 100 meV, and a tail from high-energy excitations [see Figs. 4(a) and 4(b)]; fitting details are provided in the Supplemental Material [44]. The phonon energies are consistent with previous Raman studies [52]. Here, we focus on the broad dispersionless peak at ~ 100 meV and assign it to a bimagnon excitation for several reasons. First, its energy is approximately twice the magnon energy of $\text{Nd}_4\text{Ni}_3\text{O}_{10}$ observed by Ni L -edge RIXS [22]. Second, although the absence of spin-orbit coupling in the K -shell orbitals prevents direct single-magnon excitation [53], previous studies in cuprates have shown that O K -edge RIXS can probe bimagnons [54]. As illustrated in Fig. 4(d), the intensity of this mode exhibits

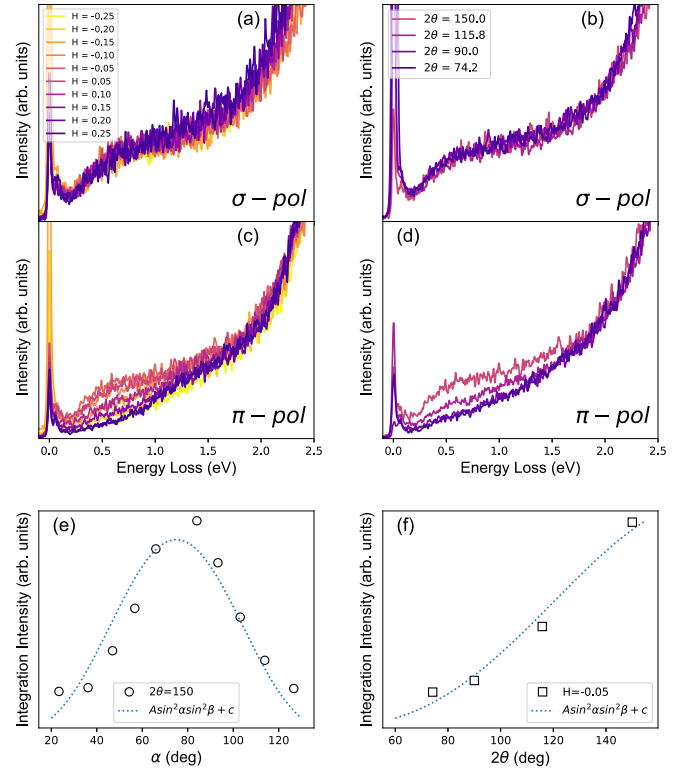


FIG. 3. (a), (c) H -dependent RIXS spectra at σ and π incident polarization, where the scattering angle 2θ is fixed to 150° . (b), (d) 2θ -dependent RIXS spectra at σ and π incident polarization, where the H is fixed to 0.05 r.l.u. (e), (f) The integrated intensity near 0.5 eV, extracted from the RIXS spectra in panels (c) and (d), which follow $A \sin^2 \alpha \sin^2 \beta + c$ behavior.

a pronounced polarization dependence: It increases with momentum in σ polarization but decreases with momentum in π polarization. At grazing incidence or emission angles in the π -polarization geometry (i.e., at larger momentum $|H|$), the electric field becomes increasingly aligned with the out-of-plane direction. Such cross-section behavior indicates that it is difficult to excite the mode with out-of-plane polarization, in sharp contrast to the phonon response (see Supplemental Material [44]) and consistent with the bimagnon behavior reported in cuprates [54]. We also performed temperature-dependent Raman measurements in Fig. 4(c). We found a dominant peak at ~ 113 meV, consistent with the bimagnon observed in RIXS. Two weaker and lower-energy peaks at 90 and 105 meV correspond to biphonons, each linked to single-phonon modes in the low-frequency Raman spectrum (see the Supplemental Material [44]). As shown in Fig. 4(e), the biphonon intensities are essentially temperature independent, whereas the bimagnon peak decreases markedly with increasing temperature but persists above T_{SDW} of 137 K, indicating the presence of short-range magnetic fluctuations above the spin-density-wave transition.

Our results elucidate distinct contributions of the Ni $d_{x^2-y^2}$ and d_{z^2} to electronic properties in $\text{La}_4\text{Ni}_3\text{O}_{10}$. The RIXS spectra in Fig. 3(c) reveal the absence of low-energy excitations below 1 eV under grazing-incidence π polarization, which selectively probes out-of-plane orbitals, supporting the

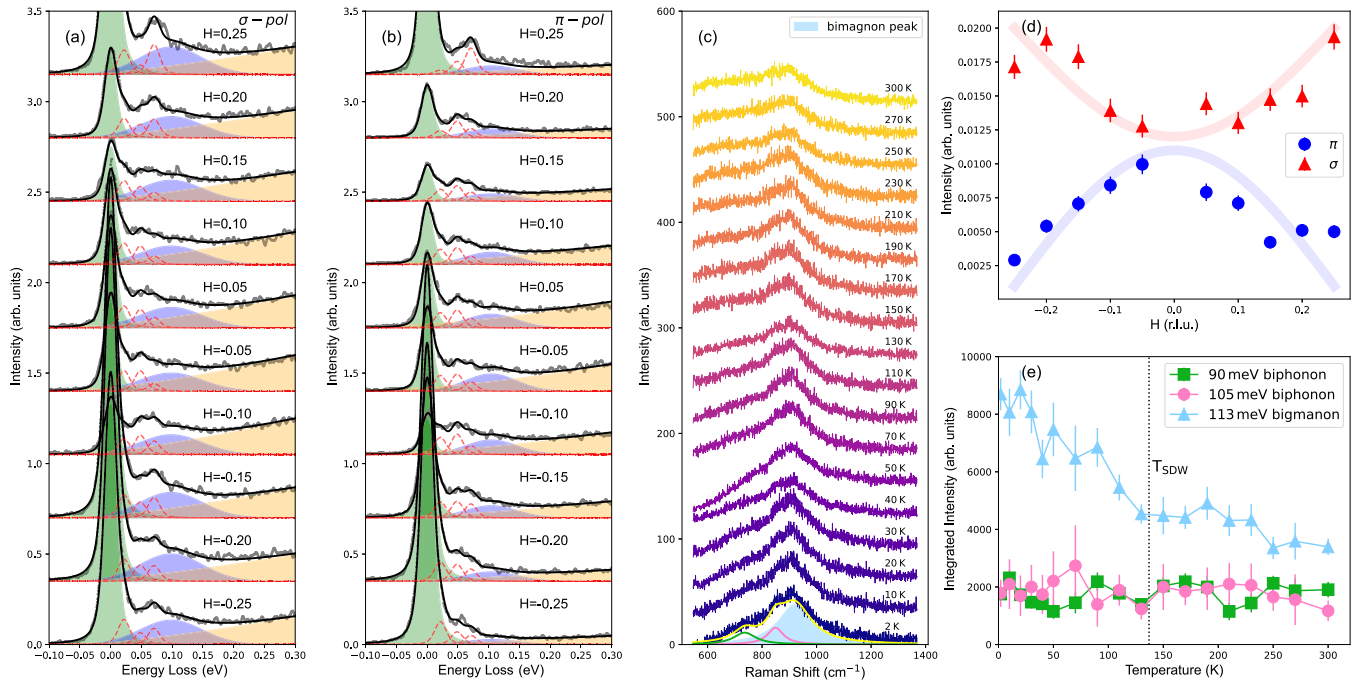


FIG. 4. (a), (b) Fitting of low-energy RIXS spectra, including elastic peak (green peak), phonons (dashed line), bimagnon (blue peak), and high-energy background (yellow tail). Each spectrum is shifted vertically for clarity. (c) Raman spectra at different temperatures, which can be fitted by three peaks, including biphonons (solid lines) and bimagnons (shaded area). Each spectrum is shifted vertically for clarity. (d) H -dependent behavior of bimagnon intensity at different polarizations extracted by fitting RIXS spectra. (e) Temperature-dependent behavior of three Raman peaks. Biphonon do not change with temperature, whereas the bimagnon weakens with increasing temperature.

localized character of Ni d_{z^2} and O p_z orbitals. In contrast, the in-plane orbitals formed by Ni $d_{x^2-y^2}$ and O $p_{x,y}$ exhibit a broad charge excitation centered around 0.5 eV (Fig. 3). The lower excitation energy of these in-plane orbitals facilitates electronic transitions between neighboring Ni $d_{x^2-y^2}$ sites via O $p_{x,y}$ orbitals, indicating that the in-plane states are more itinerant than the out-of-plane d_{z^2} and p_z orbitals. Our findings demonstrate that the $d_{x^2-y^2}$ and d_{z^2} orbitals behave independently and possess distinct electronic behaviors.

The difficulty of exciting the bimagnon with out-of-plane polarization, as observed in Fig. 4(d), provides important insight into its microscopic origin. In cuprates, the bimagnon observed in O K -edge RIXS arises from spin exchange between two Cu $d_{x^2-y^2}$ orbitals [54,55]. Considering the similar form factors between the nickelates and cuprates, one naive understanding is that the spin in nickelates also resides in the $d_{x^2-y^2}$ orbitals. However, such a simple interpretation ignores the multilayer nature of $\text{La}_4\text{Ni}_3\text{O}_{10}$. In addition to the bonding and antibonding states in a bilayer nickelate, the trilayer configuration also involves nonbonding states on the outer Ni sites [40,56]. Motivated by neutron scattering results on $\text{La}_4\text{Ni}_3\text{O}_{10}$ showing magnetism confined to the outer Ni layers [18], a more plausible scenario is that the magnetism originates from nonbonding d_{z^2} orbitals at the outer layers. They can couple to $d_{x^2-y^2}$ orbitals via a strong on-site Hund's coupling J_H on the order of 1 eV [57]. In this framework, out-of-plane polarized excitation at an inner apical oxygen site cannot generate bimagnons, while in-plane polarization can flip two neighboring $d_{x^2-y^2}$ spins while simultaneously aligning their on-site d_{z^2} spins. In our RIXS

measurements, excitation of planar O $p_{x,y}$ orbitals mediates spin exchange between intralayer Ni atoms, producing a bimagnon whose energy reflects the underlying superexchange interactions. If magnetic interactions in $\text{La}_4\text{Ni}_3\text{O}_{10}$ are likewise dominated by interlayer coupling J_z , as proposed in $\text{La}_3\text{Ni}_2\text{O}_7$ [17,58], such excitations would break two J_z bonds. Under this assumption, the observed bimagnon energy would correspond to an effective energy scale of $SJ_z \sim 56$ meV, which should be regarded as an upper bound, as the excitation likely involves both interlayer and in-plane interactions and thus requires a model that includes both contributions.

In conclusion, we uncover distinct and orbital-selective contributions to the electronic and magnetic structures of the RP-phase nickelate $\text{La}_4\text{Ni}_3\text{O}_{10}$. We demonstrate that ligand and holes occupy planar O $p_{x,y}$ and apical O p_z orbitals, hybridized predominantly with Ni $d_{x^2-y^2}$ and d_{z^2} states, respectively. Despite the multiorbital nature of this system, low-energy charge excitations are governed by the more itinerant $d_{x^2-y^2}$ electrons, while the d_{z^2} orbitals remain largely localized, exhibiting negligible spectral weight below 1 eV. By combining RIXS and Raman spectroscopy, we further identify a bimagnon mode at ~ 0.1 eV that persists well above T_{SDW} , revealing robust short-range spin correlations beyond long-range magnetic order. Together, our results provide the first direct experimental disentanglement of orbital-specific electronic and magnetic roles in RP-phase nickelates, offering essential constraints for microscopic theories of nickelate superconductivity and establishing a broadly applicable approach for resolving orbital contributions in complex multiorbital materials.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of this research project. The data are available from the authors upon reasonable request.

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