

Collective spin excitations in trilayer nickelate $\text{La}_4\text{Ni}_3\text{O}_{10}$

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Ruddlesden-Popper (RP) nickelates have recently emerged as a new family of high-temperature superconductors. In bilayer RP nickelates, magnetic excitations with large exchange couplings have been observed, supporting a spin-mediated pairing mechanism. Whether comparable spin correlations persist in trilayer nickelates, however, remains unknown. Here, we present a Ni L -edge resonant inelastic X-ray scattering (RIXS) study of $\text{La}_4\text{Ni}_3\text{O}_{10}$ single crystals. While the orbital excitations remain similar to those of $\text{La}_3\text{Ni}_2\text{O}_7$, the collective spin excitations in $\text{La}_4\text{Ni}_3\text{O}_{10}$ exhibit a comparable bandwidth of about 60 meV but substantially suppressed spectral weight, implying a weaker electronic correlation in the trilayer compounds. Our results underscore the three-dimensional and multi-orbital electronic character in $\text{La}_4\text{Ni}_3\text{O}_{10}$, highlighting important differences from the bilayer nickelates. These findings provide crucial insights into the evolution of magnetism across the RP nickelate family and its connection to superconductivity.

Understanding the electronic and magnetic interactions is central to the study of unconventional superconductivity, as these interactions may mediate electron pairing. In cuprates, the low-energy physics is largely captured by a single-band Hubbard model derived primarily from the Cu $d_{x^2-y^2}$ orbital hybridized with O $2p$ states, with the essential electronic structure residing in the quasi-two-dimensional CuO_2 planes¹. Within this framework, superconducting pairing is widely believed to arise from strong electronic correlations and antiferromagnetic spin fluctuations inherited from the Mott-insulating parent compounds^{1,2}. However, whether unconventional high-temperature superconductivity can be realized beyond the cuprate paradigm remains an open question.

The recent discovery of high-temperature superconductivity in Ruddlesden-Popper (RP) nickelates provides new insights into this question^{3,4}. Unlike cuprates, the RP phase nickelates $RE_{n+1}\text{Ni}_n\text{O}_{3n+1}$ (RE = rare earth), consist of n -layers of NiO_6 octahedra stacked along the c -axis, and exhibit enhanced three-dimensional character (Fig. 1a). Consequently, multi-band electronic structures are observed for both bilayer ($n = 2$) and trilayer ($n = 3$) nickelates with $d_{x^2-y^2}$ and d_{z^2} orbitals at or near the Fermi level, as revealed by angle-resolved photoemission spectroscopy (ARPES) measurements⁵⁻⁸. Electron hopping between d_{z^2} orbitals across apical oxygens potentially mediates a strong interlayer exchange (J_z) in $\text{La}_3\text{Ni}_2\text{O}_7$, which has been suggested by recent resonant inelastic X-ray scattering (RIXS) and neutron scattering

studies⁹⁻¹¹, revealing collective spin excitations topping around 70 meV. The active involvement of the d_{z^2} orbital also has profound influence on superconductivity, as suggested by different theoretical proposals¹²⁻¹⁷, although its detailed role is still under debate.

Despite the similar structural and orbital motifs, trilayer and bilayer nickelates display significant distinctions. While the nominal valence of Ni ions in both compounds lies between $3d^7$ and $3d^8$, the additional NiO_6 layer in trilayer nickelates introduces a different local crystal-field environment, which inevitably modifies the spin states and magnetic interactions. In fact, an incommensurate spin order has been identified in $\text{La}_4\text{Ni}_3\text{O}_{10}$ ¹⁸, different from the spin-stripe order found in $\text{La}_3\text{Ni}_2\text{O}_7$ ^{9,19,20}. Meanwhile, the maximum superconducting transition temperature (T_c) achieved in bulk $\text{La}_3\text{Ni}_2\text{O}_7$ under pressure (80 K) significantly exceeds that of $\text{La}_4\text{Ni}_3\text{O}_{10}$ (30 K). The similarities and differences between bilayer and trilayer nickelates provide a comparative platform for identifying the key ingredients governing nickelate superconductivity.

In contrast to the well-characterized magnetic excitations in $\text{La}_3\text{Ni}_2\text{O}_7$, the spin dynamics in trilayer nickelates remain largely unexplored. While recent RIXS studies have reported evidence of spin excitations in trilayer $\text{Nd}_4\text{Ni}_3\text{O}_{10}$ and $\text{La}_4\text{Ni}_3\text{O}_{10}$ ^{22,23}, essential characteristics including the dispersion of the excitation modes have yet to be established. As a result, important questions remain open, particularly regarding the connection between magnetic interactions and T_c . This incomplete

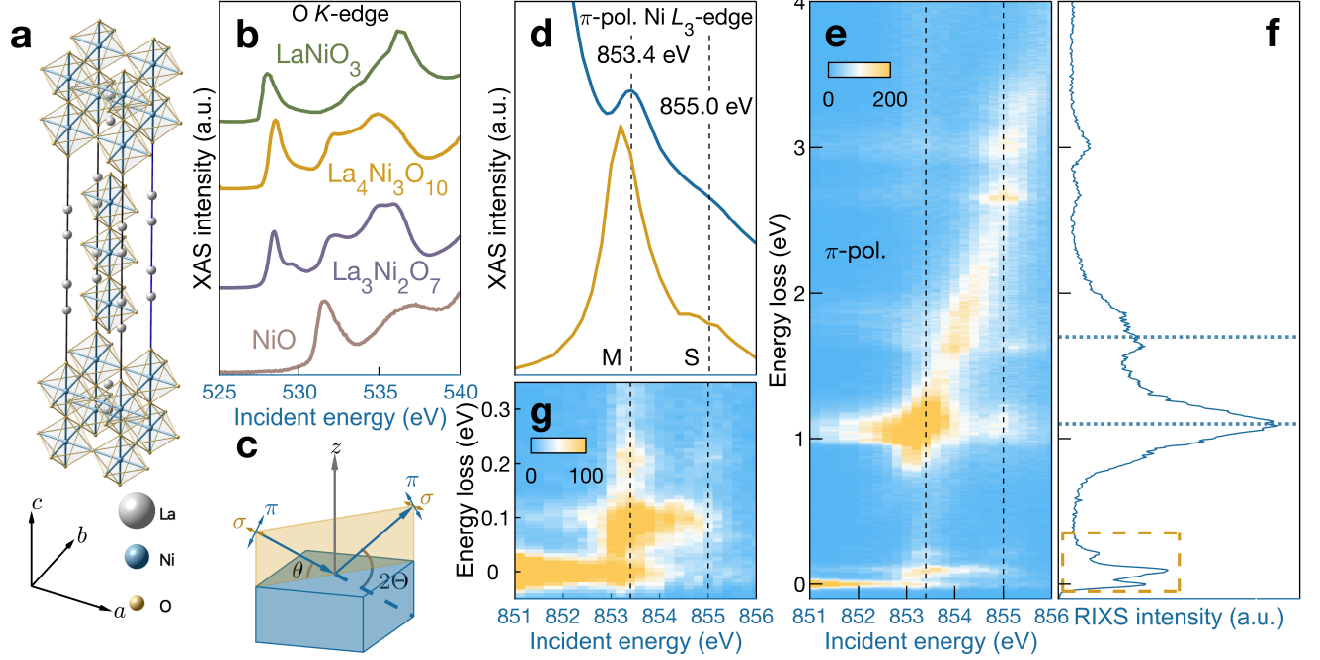


FIG. 1. **XAS and incident energy dependent RIXS.** **a** Crystal structure of $\text{La}_4\text{Ni}_3\text{O}_{10}$. **b** Oxygen K -edge XAS of NiO (pink)⁹, $\text{La}_3\text{Ni}_2\text{O}_7$ (purple)⁹, $\text{La}_4\text{Ni}_3\text{O}_{10}$ (yellow), and LaNiO_3 (green)²¹. **c** Schematic illustration of RIXS scattering geometry. **d** Enlarged XAS spectrum (blue) measured with π -polarised X-rays at $\theta = 20^\circ$, overplotted with the RIXS intensity (yellow) integrated across energy loss range $[-0.1, 0.3]$ eV. **e** RIXS energy map at $\mathbf{Q} = (0.4, 0)$ measured with π -polarised X-rays. **f** Integrated RIXS spectrum across incident energy range $[853, 854]$ eV of panel **e**. **g** RIXS intensity map within $[-0.1, 0.3]$ eV energy loss. Vertical dashed lines mark the energies of the main and satellite resonances.

understanding of magnetism in trilayer nickelates hinders efforts to elucidate the mechanism of nickelate superconductivity.

Here, we employ high-resolution RIXS at Ni L -edge to study low-energy excitations in bulk $\text{La}_4\text{Ni}_3\text{O}_{10}$. We observe two localized spin-flip excitations at about 100 and 200 meV. In addition, dispersive magnetic excitations — with 60 meV zone boundary energies — are identified, which carry a comparable bandwidth but significantly reduced spectral weight compared to the collective magnetic excitations in its bilayer counterpart $\text{La}_3\text{Ni}_2\text{O}_7$. These features are consistent with a spin-density-wave (SDW) order driven by Fermi surface nesting, suggesting a weaker electronic correlation in the trilayer system. Modeling the collective excitations using linear spin-wave theory with a presumed incommensurate magnetic structure yields effective intra- and inter-layer exchange parameters on similar energy scales, with the inter-layer coupling $SJ_\perp \approx 20$ meV being the strongest, pointing to enhanced three-dimensional magnetism in $\text{La}_4\text{Ni}_3\text{O}_{10}$. Compared to bilayer compounds, the smaller inter-layer exchange interaction is congruent with the lower T_c of trilayer nickelates. The marked differences in their magnetic excitations underscore the importance of correlation strength and dimensionality

effects that should be considered when interpreting the superconducting mechanism.

Results

To investigate the electronic structure of $\text{La}_4\text{Ni}_3\text{O}_{10}$, we first measure its oxygen K -edge X-ray absorption spectrum (XAS) and compare with those of LaNiO_3 , $\text{La}_3\text{Ni}_2\text{O}_7$ and NiO ⁹ (Fig. 1b). Consistent with previous reports²³, a strong pre-edge peak is observed in $\text{La}_4\text{Ni}_3\text{O}_{10}$, which is also present in $\text{La}_3\text{Ni}_2\text{O}_7$ and LaNiO_3 , but absent in NiO , characterizing the existence of ligand holes in RP nickelates. Fig. 1d displays the Ni L_3 -edge XAS of $\text{La}_4\text{Ni}_3\text{O}_{10}$, measured with π -polarization. Despite the partial overlap with the La M_4 -edge (see Supplementary Fig. 1b), two Ni absorption edges are identified and further confirmed by the integrated RIXS intensity in the low-energy loss range — shown in Fig. 1g. The main resonance at 853.4 eV (labeled M) originates from the Ni $2p \rightarrow 3d^8$ excitations, while the higher-energy satellite feature at 855.0 eV (labeled S) corresponds to transitions to excited states with a more pronounced ligand hole character and thereby less efficient core-hole screening. This observation highlights the strong hybridization between the O $2p$ and Ni $3d$ states.

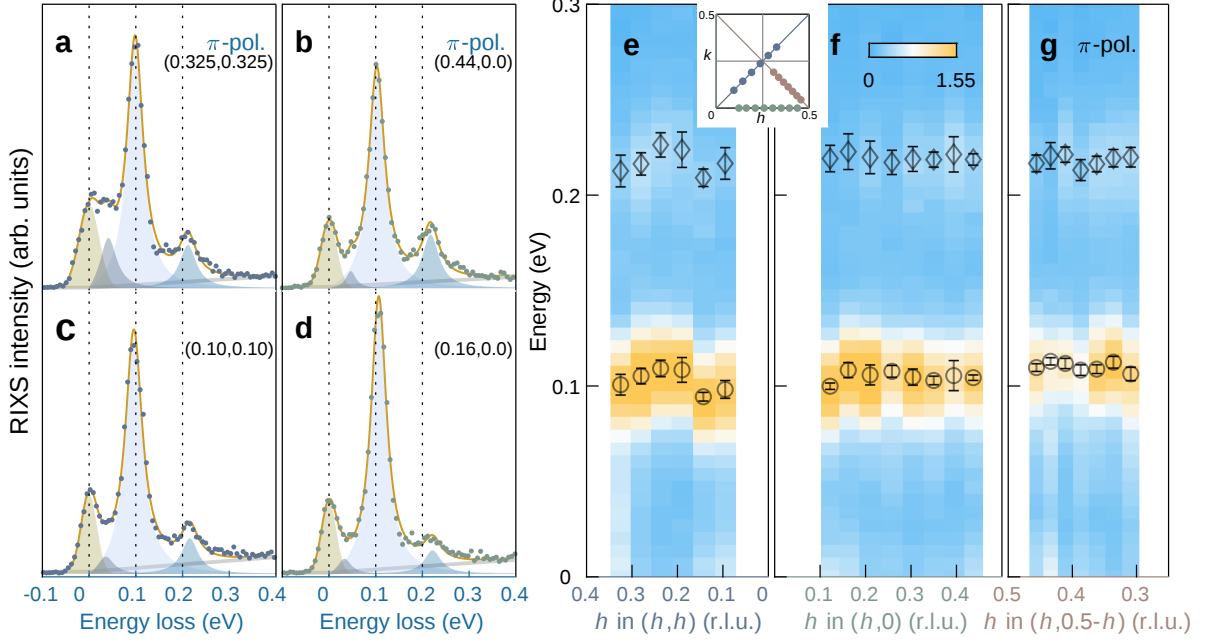


FIG. 2. **Low-energy excitations and their spectral components.** **a-d** Representative RIXS spectra and the fits. The yellow fitting curve are composed of three components: elastic scattering (dark yellow), magnetic excitations (shades of blue), and high-energy background (gray). **e-g** RIXS intensity maps under variations of momentum and energy loss measured by π -polarized light following three paths in the reciprocal space labeled with circles in the inset. Data were taken at 23 K. Error bars represent one standard deviation.

Figure 1e displays the incident energy dependence of RIXS intensities across Ni L_3 resonance measured at 23 K. Fluorescence-like signals are observed between 1 eV and 4 eV, originating from particle-hole excitations within the Ni-O charge continuum. Within this energy range, dd excitations are identified and selectively enhanced at the main and satellite resonances. The integrated intensity across the main resonance energy in Fig. 1f reveals rich orbital excitations, with the most pronounced features appearing around 1.0 and 1.7 eV, corresponding respectively to the t_{2g} -to- e_g dd excitations, and transitions involving multiple d orbitals, as commonly observed in RP nickelates^{9,22}.

At lower energy, prominent excitations are observed around ~ 0.1 eV, at both the main and satellite absorption peaks (Fig. 1e-g). Their spectral weight, integrated over $[-0.1, 0.3]$ eV, follows the trend of the Ni L_3 XAS (Fig. 1d), indicating a resonant electronic origin. A closer inspection of the low-energy spectra at the main resonance (Fig. 2a) reveals a rich set of excitations, including a pronounced sharp mode near 100 meV, a second distinct mode at about 200 meV, and a weaker excitation around 60 meV.

Next, we investigated the momentum dependence of the low-energy excitations at the main resonance energy (853.4 eV). The RIXS intensity as a function of energy loss and in-plane momentum transfer along high-

symmetry directions reveals negligible dispersion for the excitations near 100 meV and 200 meV (Fig. 2e-g). For quantitative analysis, we fitted the low-energy line shape using five components: a Gaussian profile for the elastic line, three damped harmonic oscillators (DHOs)^{24,25} for the excitation modes, and a linear function for the background.

The absence of dispersion and resolution-limited linewidth of the ~ 100 - and 200-meV modes reveal their localized characters. To further elucidate their origin, we performed polarimetric RIXS measurements at selective wave vectors to decompose the polarization of the outgoing X-rays. As revealed in Fig. 3a-d, negligible spectral weight near 100 and 200 meV appears in the $\sigma - \sigma'$ channel, indicating a predominantly magnetic origin, as will be discussed later.

Although much weaker than the ~ 100 -meV mode, the excitation near 60 meV is clearly resolved in the raw spectra thanks to the high energy resolution, and is found to be dispersive — see Fig. 4a,b. To determine its dispersion, we subtract the fitted contributions of other spectral components and plot the resulting intensity as a function of energy and momentum in Fig. 4d-f. The intensity exhibits a strong modulation that increases away from the Γ point and toward the magnetic ordering wave vector $\mathbf{Q}_{\text{spin}} = (0.31, 0.31)$ determined in a previous neutron diffraction study¹⁸, corroborating the

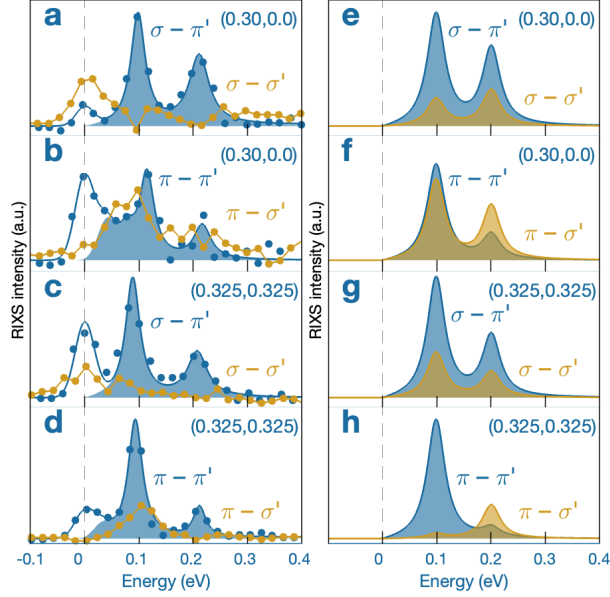


FIG. 3. **Polarimetric RIXS and exact diagonalization calculations.** **a-d** Polarimetric RIXS spectra recorded with in-plane momenta as indicated with σ - and π - incident X-rays. All spectra were decomposed into components of σ -out (yellow) and π -out (blue) channels. **e-h** ED calculation results for in-plane momentum transfers and incident X-ray polarization as indicated. A life-time broadening is applied to mimic the experimental damped harmonic oscillator profile.

propagating nature of this excitation. This assignment is also consistent with a recent O K -edge RIXS and Raman spectroscopy study on $\text{La}_4\text{Ni}_3\text{O}_{10}$ ²³, which identified a bimagnon mode at an energy approximately twice that of the dispersive mode observed here.

Discussion

Previous X-ray diffraction (XRD) measurements have shown that NiO_6 octahedra in the inner layer of trilayer nickelates possess more isotropic Ni-O bond lengths than those in the bilayer compounds^{4,26}. In line with this structural trend, a recent transport study revealed three-dimensional superconductivity in trilayer $\text{La}_4\text{Ni}_3\text{O}_{10}$ and $\text{Pr}_4\text{Ni}_3\text{O}_{10}$, evidenced by the extremely small anisotropy of the upper critical field, which is in stark contrast with bilayer nickelates and cuprates²⁷. Considering the dominant $3d^8\bar{L}$ (where $\bar{L}$ denotes a ligand hole) character of the Ni ions, we model the polarimetric RIXS results by considering a single Ni^{2+} ion under a three-dimensional effective exchange field. As shown in Fig. 3e-h, exact diagonalization (ED) calculations assuming an effective exchange field of $J^* \approx 45$ meV qualitatively reproduced both localized excitations (see Supplementary Information for details). Within this picture, the 0.1 eV and 0.2 eV modes originate from the local dipolar and quadrupolar spin excitations, respectively. This assign-

ment accounts for the two-fold excitation energy of the 0.2 eV mode and is consistent with the observed polarization dependence at different wave vectors.

We hypothesize that the localized excitations originate from Ni ions adopting a high-spin configuration but without ordered moment. Indeed, a previous neutron diffraction study on $\text{La}_4\text{Ni}_3\text{O}_{10}$ proposed a magnetic structure in which the inner layer is nonmagnetic¹⁸. Furthermore, the inner-layer NiO_6 octahedra deviate only slightly from the ideal octahedral geometry^{4,26}, compared with those in the bilayer structure. These observations justify our interpretation of the localized spin excitations.

To describe the dispersive low-energy magnetic excitations, we adopt a Heisenberg spin Hamiltonian motivated by earlier neutron diffraction results, which revealed an incommensurate in-plane ordering wave vector $\mathbf{Q}_{\text{spin}} = (0.31, 0.31)$, close to $(1/3, 1/3)$, with a node at the inner layer. We therefore consider a magnetic structure consistent with this pattern, shown in Fig. 4g,h, and include two in-plane exchange couplings, J_1 and J_2 , and an interlayer coupling $J_{\perp}$. The linear-spin-wave-theory (LSWT) calculations (Fig. 4i-k) generally capture the observed dispersion and the momentum-dependent intensity modulation, with $SJ_1 = 12$ meV, $SJ_2 = 8$ meV, and $SJ_{\perp} = 20$ meV providing the best agreement with the experiment.

The intensity of the dispersive magnetic excitations is nearly an order of magnitude weaker compared to that observed in $\text{La}_3\text{Ni}_2\text{O}_7$ ⁹ (Fig. 4c). The marked suppression of the spectral weight is consistent with an itinerant SDW state. Recent ARPES studies on La- or Pr-based trilayer nickelates have revealed Fermi-surface nesting, band folding, and band splitting related to the SDW state^{28,29}. Our RIXS results thus are consistent with such an itinerant origin of the collective magnetic excitations. Moreover, ARPES measurements also revealed significant orbital selective correlation effects in trilayer nickelates³⁰. Under the combined influence of orbital selective electronic correlations and nesting induced instability, a complex magnetic ground state¹⁸ and excitation spectrum are therefore expected, consistent with the observed coexistence of localized and collective magnetic excitations.

Compared with bilayer $\text{La}_3\text{Ni}_2\text{O}_7$, the collective magnetic excitations in $\text{La}_4\text{Ni}_3\text{O}_{10}$ exhibit a comparable bandwidth but a more pronounced three-dimensional character, as demonstrated by intra- and interlayer exchange couplings of comparable magnitude. This is consistent with the more three-dimensional electronic structure reported for trilayer nickelates²⁷. The remarkable differences between the magnetism of the two systems reveal the importance of three-dimensionality, correlation strength, and multiorbital characters in defining the magnetic interactions and possibly the superconducting pairing mechanism in RP nickelates.

Note added. Upon completion of this work, we became aware of an independent RIXS study of magnetic and

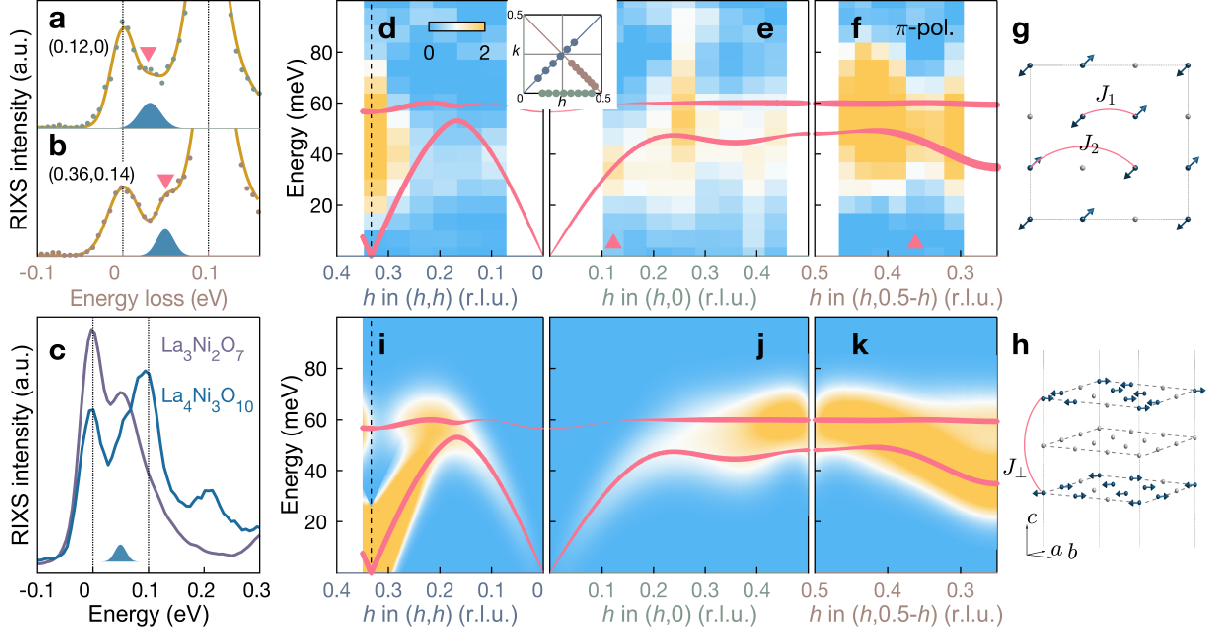


FIG. 4. **Dispersion and spectral weight of the collective magnetic excitations.** **a-b** Selected low-energy RIXS spectra — for momenta as indicated — with fitted yellow curve and low-energy gaussian component filled by shades of blue. **c** Normalized integrated RIXS intensities of $\text{La}_3\text{Ni}_2\text{O}_7$ (purple) and $\text{La}_4\text{Ni}_3\text{O}_{10}$ (blue) with the indication of scaled gaussian component from panel **b** filled in blue⁹. **d-f** Low-energy RIXS intensity map with components of elastic, 0.1 eV, and 0.2 eV excitations subtracted. A Gaussian smoothing has been applied. The pink lines are simulated dispersion using LSWT. Downward triangles indicate the position of spectra in panels **a** and **b**. **g,h** Magnetic structure adopted for the LSWT calculation in **i-k**. Blue and gray circles denote the Ni sites with and without ordered moment, respectively. The arrows indicate the spin orientations.

electronic excitations in $\text{La}_4\text{Ni}_3\text{O}_{10}$ ³¹.

Methods

Sample synthesis and characterization

$\text{La}_4\text{Ni}_3\text{O}_{10}$ single crystals were grown using the high-pressure optical floating zone method, as described in ref. 4. The phase purity and single crystallinity were characterized by X-ray diffraction measurements both before and after the cleave. The magnetic phase transition was characterized by transport and susceptibility measurements. (see Supplementary Information).

RIXS experiments

Ni L -edge RIXS experiments were carried out at the ID32 beamline at the European Synchrotron Radiation Facility (ESRF)³². Samples were aligned *ex-situ* using Laue diffraction (see Supplementary Fig. S1), and cleaved *in-situ* in the load-lock chamber. The wave vector $\mathbf{Q}$ in (q_x, q_y, q_z) is defined as $(h, k, l) = (2\pi/a, 2\pi/b, 2\pi/c)$ in reciprocal lattice units (r.l.u.), where $a = b = 3.86$ Å and $c = 27.97$ Å are the lattice parameters of the pseudo-tetragonal unit cell. The scattering angle is fixed to $2\Theta = 149.5^\circ$. The energy resolution is characterized by the full-width-at-half-maximum (FWHM) of the

elastic scattering profile of silver paste or amorphous carbon which was set to 38 meV. RIXS intensities were normalized to the weight of the dd excitations between 0.8 eV and 2.4 eV, as in refs. 33 and 34 (See Supplementary Note 2). Data were collected at 23 K (30 K) at ID32 (41A1). Direct and indirect RIXS measurements were performed consecutively to resolve outgoing X-rays in π' - and σ' -polarization channels. Under the limited reflectivities of the mirror ($R_\sigma \approx 0.137$ and $R_\pi \approx 0.0703$), to attain the same photon statistics of polarimetric measurements, an increase of from six- to ten-fold exposure time is required, compared to a direct RIXS measurement. Additional RIXS experiments were carried out at the 41A1 beamline of National Synchrotron Radiation Research Center (NSRRC)³⁵, yielding consistent results from those obtained at ID32 (see Supplementary Information).

Single-ion and spin-wave calculations

Exact diagonalization calculations based on a single-ion model were performed using the EDRIXS package³⁶ to interpret the localized excitation modes. Detailed parameters for the calculation are provided in Supplementary Note 4. We employ an effective Heisenberg Hamiltonian

to model the dispersive low-energy excitations and evaluate the magnetic interaction strengths, including two in-plane exchange couplings J_1 and J_2 and one interlayer coupling $J_\perp$:

$$H = J_1 \sum_{\langle i,j \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J_2 \sum_{\langle i,i' \rangle} \mathbf{S}_i \cdot \mathbf{S}_{i'} + J_\perp \sum_{\langle i,j' \rangle} \mathbf{S}_i \cdot \mathbf{S}_{j'} \quad (1)$$

where $\langle i, j \rangle$, $\langle i, i' \rangle$, and $\langle i, j' \rangle$ denote pairs of coupled spin sites as indicated in Fig. 4g,h, and $\mathbf{S}_i$ denotes the spin operator at the lattice site i . The exchange couplings were determined by comparing the momentum dependent RIXS intensity with the calculated dynamic spin structure factor, with its modulation due to the varying l component in the RIXS geometry accounted. The SpinW package was used for the spin-wave calculations³⁷.

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Author contributions

Q.W. conceived the project. Y.Z., E.Z., L.C., and J.Z. grew the $\text{La}_4\text{Ni}_3\text{O}_{10}$ single crystals, and characterized the samples with Y.C. and T.W. Y.C., Y.Y., X.H., M.R.C. and N.B.B. carried out the RIXS experiments at ID32. Y.C., Y.L., J.O., H.Y.H. and D.J.H. carried out the RIXS experiments at 41A1. Y.C., Y. L., Y.S. and Q.W. analyzed the RIXS data. Y.C., J.C. and Q.W. wrote the manuscript with input from other authors.

Data availability

Data supporting the findings of this study are available from the corresponding authors upon request.

Competing interests

The authors declare no competing interests.

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