

Supplementary Information for “Robust Bilayer Electronic Structure and Reduced Magnetic Exchange in $\text{La}_2\text{SmNi}_2\text{O}_7$ ”

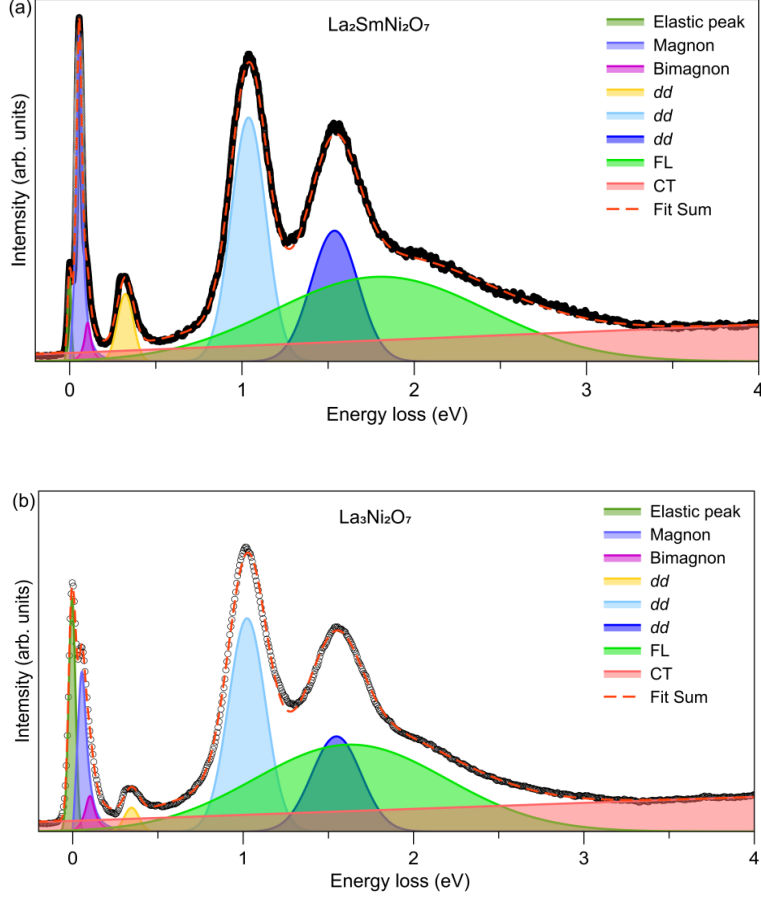


Fig. S1. Wide-range fitting plots of Figs. 1(g) and 1(h). (a),(b) Fitting plots of incident-energy-integrated Ni- L_3 -edge RIXS spectra with π polarization in $\text{La}_2\text{SmNi}_2\text{O}_7$ and $\text{La}_3\text{Ni}_2\text{O}_7$. The measurements of $\text{La}_2\text{SmNi}_2\text{O}_7$ were performed at $T = 20$ K with a scattering angle $2\theta = 90^\circ$ and an incident angle $\theta = 20^\circ$ over the incident-energy range 853.8-856.2 eV. The data of $\text{La}_3\text{Ni}_2\text{O}_7$ are reproduced from Ref. [1]. The spectra were normalized to the peak maximum of the ~ 1.0 eV dd excitation. The elastic scattering (green shaded area) was fitted by Gaussian functions, while magnon (periwinkle) and bimagnon (purple) excitations were fitted by damped-harmonic-oscillator (DHO) functions

$$\chi''(\mathbf{Q}, E) = A \frac{4\gamma E E_0}{(E^2 - E_0^2)^2 + (2\gamma E)^2} \quad (1)$$

The yellow, sky blue, and blue shaded areas represent three types of dd excitations fitted by Gaussian functions. The bright green and pink shaded areas represent the fluorescence and charge transfer, respectively. The red dashed curves denote the fit sum. Three types of dd excitations are observed in $\text{La}_2\text{SmNi}_2\text{O}_7$ with energies of approximately 0.32, 1.04, and 1.54 eV, compared to approximately 0.34, 1.02, and 1.55 eV in $\text{La}_3\text{Ni}_2\text{O}_7$.

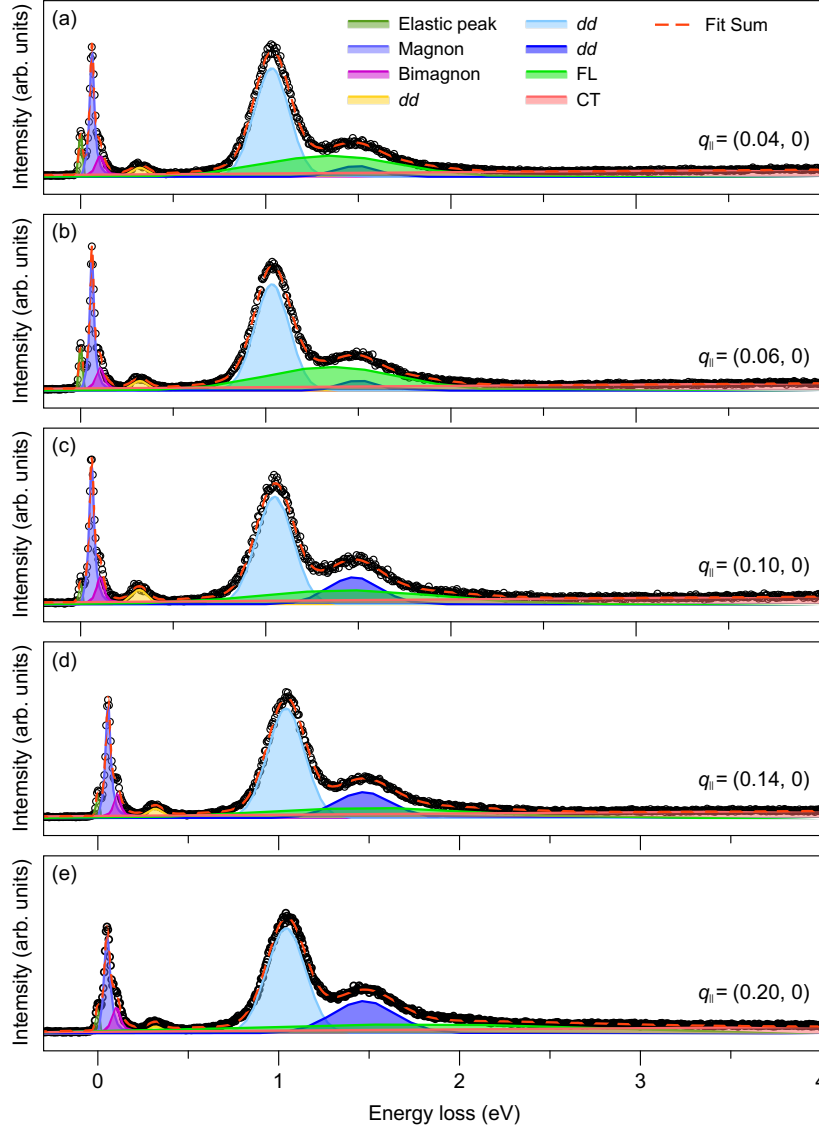


Fig. S2. Wide-range fitting plots of RIXS spectra in $\text{La}_2\text{SmNi}_2\text{O}_7$. (a)-(e), Fitting plots of momentum-dependent Ni- L_3 -edge RIXS spectra in $\text{La}_2\text{SmNi}_2\text{O}_7$ at $q_{||} = (0.04, 0)$, $(0.06, 0)$, $(0.10, 0)$, $(0.14, 0)$, $(0.20, 0)$, respectively. The spectra were measured along the $[H, 0]$ direction using $E_i = 854.65$ eV with π polarization at 20 K with a scattering angle $2\theta = 90^\circ$. The RIXS spectra were normalized to the ~ 1.0 eV dd excitation and fitted using the same functions and color scheme as in Fig. S1. Well-defined dispersive magnon excitations are observed.

Resolution-convolved analysis of the low-energy spectra

The broad-range fits in Figs. S1 and S2 are retained to describe the electronic excitations. The quantitative low-energy magnetic parameters reported below and in the main text are instead obtained from resolution-convolved fits. The magnon and bimagnon responses use the DHO form in Eq. (1), with undamped energy E_0 and damping γ . The measured spectrum is modeled as

$$I_q(E) = I_{\text{el},q}(E) + \sum_v [R_q * F_{v,q}](E) + B_q(E) \quad (2)$$

where $I_{\text{el},q}$ is the observed elastic line, B_q is an additive background, and $F_{v,q}$ denotes an intrinsic inelastic component. The symbol $*$ denotes convolution in energy with the zero-centered, unit-area resolution function R_q . The elastic line is not convolved a second time. Numerical magnetic profiles use the positive-energy DHO response; thermal occupation corrections are negligible in the 40–90 meV peak region at 20 K ($k_B T \approx 1.72$ meV).

La₂SmNi₂O₇: independently fixed momentum-dependent resolution

For each π -polarized La₂SmNi₂O₇ spectrum, the energy-gain side and apex of the elastic line were first fitted independently over -80 to $+5$ meV using a Pearson VII profile plus a constant background. The fitted elastic position, FWHM and shape parameter were then fixed in the full-spectrum fit; only the elastic amplitude was refitted. The resolution kernel uses the same FWHM and shape, centered at zero. The respective FWHM values are 22.19, 20.35, 20.60, 20.18 and 29.07 meV at $h = 0.04, 0.06, 0.10, 0.14$ and 0.20 .

The second-stage fit covers -100 to 500 meV and contains the elastic peak, magnon and bimagnon DHO responses, a Gaussian dd excitation near 0.32 eV, and a quadratic background. Each inelastic component is convolved with the fixed resolution kernel. Least squares uses equal point weights, with no extra weighting of the 40–90 meV interval. Taking the elastic profile as the instrumental response assumes that it is resolution limited.

Bulk La₃Ni₂O₇: reanalysis without an additional zero correction

The published σ -polarized spectra of Ref. [1] were taken from its public source data (Zenodo, DOI: 10.5281/zenodo.13955595; Fig2a_1.txt and Fig2a_2.txt). Seventeen spectra were reanalyzed: six along (H, H) and eleven along $(H, 0)$. Their calibrated energy axes were left unchanged, and the elastic center was fixed at 0 meV for every fit. Within -100 to 300 meV, the component model comprises a Gaussian elastic line, magnon and bimagnon/high-energy DHO responses, and a nonnegative linear background. The elastic Gaussian and convolution kernel both have the reported 36.5 meV FWHM. This retains the components shown in Supplementary Note 4 and Figs. S7–S9 of Ref. [1], while explicitly convolving both magnetic responses. A weak nonnegative linear background follows the plotted supplementary fits of Ref. [1].

Relation between the intrinsic DHO energy, damping and peak position

For the positive-energy DHO in Eq. (1), let E_p denote the maximum of the intrinsic $\chi''(E)$ before convolution. Setting its energy derivative to zero gives $3E_p^4 + (4\gamma^2 - 2E_0^2)E_p^2 - E_0^4 = 0$, hence

$$E_p^2 = \frac{E_0^2 - 2\gamma^2 + \sqrt{(E_0^2 - 2\gamma^2)^2 + 3E_0^4}}{3} \quad (3)$$

Thus E_p is lower than E_0 for finite damping. In the weak-damping limit ($\gamma/E_0 \ll 1$), the intrinsic peak becomes approximately Lorentzian and

$$E_p \approx E_0 - \frac{\gamma^2}{2E_0}, \quad \Delta E_{\text{int}} \approx 2\gamma \quad (4)$$

where ΔE_{int} is its intrinsic FWHM. The latter relation is approximate; γ is the DHO damping parameter, not the full linewidth. The real part of the underdamped response pole, $(E_0^2 - \gamma^2)^{1/2}$, is also not exactly the maximum in Eq. (3). For $\text{La}_2\text{SmNi}_2\text{O}_7$, the fitted $\gamma = 6.29\text{--}8.86$ meV and $\gamma/E_0 = 0.114\text{--}0.155$ give $E_0 - E_p = 0.39\text{--}0.69$ meV. For $\text{La}_3\text{Ni}_2\text{O}_7$ at (0.06, 0), (0.10, 0) and (0.14, 0), the corresponding differences are 0.24, 0.35 and 0.30 meV.

These relations concern an isolated intrinsic DHO, not the maximum of the measured spectrum. Resolution convolution and overlap with the elastic line, bimagnon and background can further alter the observed peak position. Omitting the instrumental response allows resolution broadening to enter the fitted γ and, through correlated line-shape parameters, can also bias E_0 . We therefore fit the convolved spectrum directly, rather than infer intrinsic damping by quadratically subtracting an instrumental FWHM. A raw spectral-peak separation at one momentum is not the same quantity as the difference between fitted dispersion band tops.

Dispersion comparison using the convolved mode energies

An unweighted $J_c\text{--}J_1\text{--}J_2$ fit to the seventeen $\text{La}_3\text{Ni}_2\text{O}_7$ energies above 40 meV gives $SJ_c = 36.35 \pm 4.77$ meV, $SJ_1 = 1.53 \pm 0.37$ meV and $SJ_2 = 7.27 \pm 0.68$ meV, with a Γ -point band top of 68.37 ± 1.76 meV. Scaling all three parameters to fit the five $\text{La}_2\text{SmNi}_2\text{O}_7$ energies gives $s = 0.9004 \pm 0.0027$ and a band top of 61.56 meV; the scale error is conditional on the $\text{La}_3\text{Ni}_2\text{O}_7$ reference curve. A three-parameter free fit gives $SJ_c = 31.02 \pm 1.07$ meV, $SJ_1 = 2.70 \pm 0.42$ meV and $SJ_2 = 6.33 \pm 0.07$ meV, with a band top of 61.72 meV, but has only two residual degrees of freedom. The respective $\text{La}_3\text{Ni}_2\text{O}_7\text{--}\text{La}_2\text{SmNi}_2\text{O}_7$ band-top differences are 6.81 and 6.65 meV. All curves satisfy $E(0.25, 0.25) = 0$ identically; this model condition is not an additional measured point or degree of freedom.

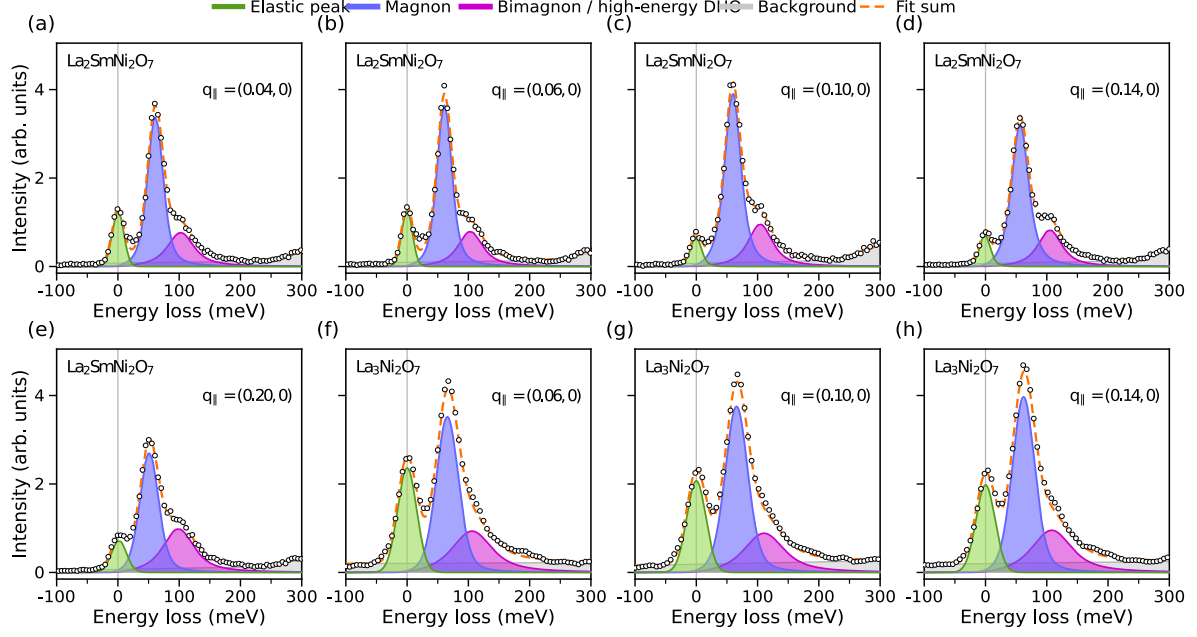


Fig. S3. Resolution-convolved fits of the low-energy Ni- L_3 -edge RIXS spectra. Panels (a)–(e) show the π -polarized $\text{La}_2\text{SmNi}_2\text{O}_7$ spectra at $q_{\parallel} = (0.04, 0)$, $(0.06, 0)$, $(0.10, 0)$, $(0.14, 0)$ and $(0.20, 0)$. Panels (f)–(h) show the published σ -polarized $\text{La}_3\text{Ni}_2\text{O}_7$ spectra [1] at the matched momenta $(0.06, 0)$, $(0.10, 0)$ and $(0.14, 0)$. Black open circles are data and orange dashed curves are total fits. Green, periwinkle and purple components denote elastic scattering, magnon and bimagnon/high-energy DHO responses. Gray denotes the background, including the Gaussian dd contribution for $\text{La}_2\text{SmNi}_2\text{O}_7$. Only -100 to 300 meV is displayed; the $\text{La}_2\text{SmNi}_2\text{O}_7$ fits extend to 500 meV. The $\text{La}_2\text{SmNi}_2\text{O}_7$ resolution function is fixed by independent elastic-line fits, whereas $\text{La}_3\text{Ni}_2\text{O}_7$ uses the reported 36.5 meV Gaussian resolution with its elastic center fixed at zero and no additional energy-axis correction. The convolved magnon contributes at least 80% of the combined magnon plus bimagnon intensity from 40 to 90 meV. The plotted parameters for $\text{La}_2\text{SmNi}_2\text{O}_7$ are intrinsic DHO quantities before convolution and are listed in Tables S1.

Table S1. Parameters of the resolution-convolved $\text{La}_2\text{SmNi}_2\text{O}_7$ fits. ΔE_{res} is the independently determined Pearson VII FWHM. $E_{0,M}$, γ_M and $E_{0,Bi}$ are the intrinsic magnon energy, damping and bimagnon energy. The three broad dd energies are unchanged from Fig. S2.

$q_{\parallel}$	ΔE_{res} (meV)	$E_{0,M}$ (meV)	γ_M (meV)	$E_{0,Bi}$ (meV)	dd 0.3 (eV)	dd 1.0 (eV)	dd 1.5 (eV)
$(0.04, 0)$	22.19	61.46	7.00	104.49	0.32	1.03	1.49
$(0.06, 0)$	20.35	61.21	6.95	104.72	0.32	1.03	1.50
$(0.10, 0)$	20.60	59.98	7.77	105.73	0.32	1.05	1.48
$(0.14, 0)$	20.18	57.10	8.86	106.42	0.32	1.04	1.47
$(0.20, 0)$	29.07	51.37	6.29	101.22	0.32	1.04	1.47

Reference

- [1] X. Chen, J. Choi, Z. Jiang, J. Mei, K. Jiang, J. Li, S. Agrestini, M. Garcia-Fernandez, H. Sun, X. Huang, D. Shen, M. Wang, J. Hu, Y. Lu, K.-J. Zhou, and D. Feng, Electronic and magnetic excitations in $\text{La}_3\text{Ni}_2\text{O}_7$, *Nat. Commun.* **15**, 9597 (2024).