I. FURTHER DETAILS OF THE SINGLE-CRYSTAL SYNTHESIS

Single-crystal growth of $R_4\text{Ni}_3\text{O}_{10}$ ($R=$La, Pr) was performed as described Refs. [1, 2]. The parent Ruddlesden-Popper phases were prepared in a floating zone furnace (HKZ-1, SciDre GmbH) with 20 bar $O_2$ for $R=$La and 140 bar for $R=$Pr. Oxygen was flowed at a rate of 0.1 l/min during growth and the feed and seed rods were counter-rotated at 30 r.p.m. and 27 r.p.m., respectively, to improve zone homogeneity. The traveling speed of the seed was 4 mm/h and the growth time was 30 hours. 438-phase crystals were obtained by reducing the 4310-specimens in 4 mol % $H_2$/$Ar$ gas at $350^\circ C$ for five days. The resulting samples have appreciable residual strain and are very brittle, so they were mounted on copper plates for transport.

II. RESONANT BEHAVIOR

In Fig. S1 we show the resonant behavior of the magnon. The magnon peak is visible at several energies from 851.5 to 853.1 eV exhibiting a Raman-like behavior in which it appears at a constant energy loss, rather than a constant final x-ray energy. We find that the magnon is strongest at 852.7 eV (as emphasized by the dashed line at this energy). This is well above the La $M_4$-edge at 849 eV, further confirming the magnetic origin of the magnon.

III. FITTING OF THE RIXS DATA

The spectra were fitted with a Gaussian function for the elastic peak, a damped harmonic oscillator model for the paramagnon and a quadratic background. The inelastic peak was convoluted with a Gaussian function to account for the energy resolution. This lineshape is described by nine parameters, but only six parameters are free to vary in the fit. For the Gaussian lineshape describing the elastic peak, the center and the width are fixed by measurements of a graphite elastic reference sample, only the amplitude is free to vary. For the inelastic mode, the temperature is fixed, and the center, width and amplitude are free. For the quadratic background, we use a function $f(x) = b$ for $x < 0$.

![Graph of RIXS spectra of La$_4$Ni$_3$O$_{10}$ at (-0.44, 0) as a function of incident energy around the Ni $L_3$ edge. The chosen working energy that optimizes the magnon intensity, 852.7 eV, is highlighted using a dashed line and occurs above the maximum in the elastic line resonance, further confirming the magnetic nature of the observed inelastic excitations.](image)
FIG. S2. RIXS spectra of Pr$_4$Ni$_3$O$_8$ as a function of $Q$ at the resonant energy of the magnon 852.7 eV. Data are shown as red points and the fit is shown as a black line, which is composed of the magnetic excitation in orange and the elastic line in blue. The in-plane $Q$ of the measured spectrum is denoted in the top right of each panel. Note that the scale of the y-axis is half of that in Fig. 2 of the main text.

and $f(x) = a * x^2 + b$ for $x > 0$, $a$ and $b$ are free parameters. Prior to computing the final fit, we performed an initial fit in which the elastic energy was allowed to vary, which we used to shift the spectra in energy such that the elastic energy is set to exactly zero.

IV. COMPARISON OF La$_4$Ni$_3$O$_8$ AND Pr$_4$Ni$_3$O$_8$

The difference between La$_4$Ni$_3$O$_8$ and Pr$_4$Ni$_3$O$_8$ has been studied in prior x-ray absorption and density functional theory (DFT)-based work [2]. This study concluded that both materials are rather similar regarding their high- and medium-energy physics such as spin states, orbital polarization, etc. The primary difference is that stripe order opens a small insulating gap in La$_4$Ni$_3$O$_8$, whereas Pr$_4$Ni$_3$O$_8$ remains metallic without long-range order. Pr$_4$Ni$_3$O$_8$ was later reported to have spin-glass behavior likely coming from short-range stripe correlations [4]. Since the more ordered and insulating nature of La$_4$Ni$_3$O$_8$ compared to Pr$_4$Ni$_3$O$_8$ is expected to give sharper magnetic RIXS spectra, we focused on the former material for this paper, but we also took data on Pr$_4$Ni$_3$O$_8$ as shown in Fig. S2. A similar energy peak is observed which is slightly broadened and less intense compared to La$_4$Ni$_3$O$_8$. Fitting the spectra for Pr$_4$Ni$_3$O$_8$ in the same way as was done for La$_4$Ni$_3$O$_8$ yields a value of the near-neighbor exchange perhaps 10% lower, but overall the two materials are very similar (Fig. S3).

V. THEORY OF MAGNETIC EXCITATIONS IN THE STRIPE-ORDERED STATE

In this Section, we compute the dispersion relation and RIXS intensity for the magnons in a diagonal stripe state. To reproduce the stripe order shown in Fig. 1(b) of the main text, we consider a model with the following interactions illustrated in Fig. S4. $J$ couples nearest-neighbor spins within the same stripe, $J_1$ couples spins across the stripes in the [1, 0, 0] direction, and $J_2$ couples spins between layers within the trilayer in the [0, 0, 1] direction. A further $J_2$ coupling across the stripes along the [1, 1, 0] direction was also considered, but its effect could not be distinguished in the measured RIXS spectra, so it was omitted. This is expected as this super-superexchange contribution is weak given the 90 degree Ni-Ni-Ni pathway that is involved. We also ignore any single-ion anisotropy, again because it would be difficult to detect given the width of the elastic line. The in-plane lattice vectors for the structural unit cell are

$$a_1 = (a, 0, 0), \quad a_2 = (0, a, 0)$$

and for the magnetic unit cell are

$$a_1^{mag} = (3a, 0, 0), \quad a_2^{mag} = (-a, a, 0).$$

(1)

(2)
where, for simplicity, we have assumed a primitive magnetic cell with non-orthogonal lattice vectors. Along the $c$ direction, we have trilayers separated by the body-centered translation $(a/2, a/2, c/2)$. Since there are no observable correlations between trilayers [5], we consider only a single trilayer here. The layers within a trilayer are in registry along $c$, with each layer separated by the interlayer distance $d \approx c/8$. The scattering vector $Q$ is presented in normalized units with $a = c = 1$.

### A. Dispersion relation

We proceed to calculate the magnon dispersion for the diagonal stripe state by generalizing the torque equation formalism of Carlson et al. [6] to the trilayer case (equivalent results can be obtained using the less transparent Holstein-Primakoff treatment [6]). According to neutron scattering data, the spins in the ground state are oriented...
along $c$. Therefore, the generalized torque equations for the spins reduce to:

\[
\begin{align*}
\frac{dS_{r,i}^x}{dt} &= -\frac{1}{\hbar} \left( S_{r,i}^y \sum_{r',j} J^{ij}_{rr'} S_{r',j}^z - S_{r,i}^z \sum_{r',j} J^{ij}_{rr'} S_{r',j}^y \right), \\
\frac{dS_{r,i}^y}{dt} &= -\frac{1}{\hbar} \left( S_{r,i}^z \sum_{r',j} J^{ij}_{rr'} S_{r',j}^z - S_{r,i}^z \sum_{r',j} J^{ij}_{rr'} S_{r',j}^y \right), \\
\frac{dS_{r,i}^z}{dt} &\approx 0,
\end{align*}
\]

where $r, r'$ label the positions of the spins in different magnetic unit cells and the indices $i, j$ label the spins within each magnetic unit cell ($i, j = 1, \ldots, 6$) as shown in Fig. S2. We seek sinusoidal solutions of the form

\[
S_{r,i}^x = S_i^x \exp[i(Q \cdot r - \omega t)], \quad S_{r,i}^y = S_i^y \exp[i(Q \cdot r - \omega t)],
\]

and we set $S_{r,i}^z = \pm S$ with the sign given by the orientation of the spin in the ground state. To start, we identify the couplings $J^{ij}_{rr'}$ that connect the spins at different lattice positions, with the origin taken to be the location of spin 1. We distinguish two groups of spins, $S_1, S_3, S_5$ and $S_2, S_4, S_6$, with each group having equivalent in-plane locations due to the $c$-axis translational symmetry. Their couplings are

- $S_1$ ($r = 0$) couples to $S_2$ twice with $J$ ($r' = a_1, r'' = a_2$) and twice with $J_1$ ($r' = -2a_1, r'' = -2a_2$). The same applies for $S_3$ ($S_5$) coupled to $S_4$ ($S_6$).
- $S_2$ ($r = a_1$) couples to $S_1$ twice with $J$ ($r' = 0, r'' = a_1 - a_2$) and twice with $J_1$ ($r' = 3a_1, r'' = a_1 + 2a_2$). The same applies for $S_4$ ($S_6$) coupled to $S_3$ ($S_1$).

For the couplings along $[0,0,1]$, we have:

- $S_1$ ($S_2$) couples to $S_3$ ($S_4$) with $J_z$.
- $S_5$ ($S_6$) couples to $S_3$ ($S_4$) with $J_z$.
- $S_3$ ($S_4$) couples with $J_z$ to $S_1$ ($S_2$) and to $S_5$ ($S_6$).

With this information, we can write the torque equations for each of the six spins in the magnetic unit cell, for example:

\[
\begin{align*}
\frac{dS_{0,1}^x}{dt} &= -\frac{1}{\hbar} \left\{ S_{0,1}^y \left( 2J + 2J_1 + J_z \right) \right. \\
&\quad - S \left[ J \left( S_{a_1,2}^y + S_{a_2,2}^y \right) + J_1 \left( S_{-2a_1,2}^y + S_{-2a_2,2}^y \right) + J_z \left( S_{(0,0,-\frac{a}{2})}^y \right) \right] \right\}.
\end{align*}
\]

Substituting Eq. 4, we can rewrite this expression as

\[
\frac{i\hbar \omega}{S} S_{0,1}^x = -S_{0,1}^y \left( 2J + 2J_1 + J_z \right) - S_{3,1}^y J_z e^{-iQ_x} \\
\quad - S_{0,1}^y \left[ J \left( e^{iQ_x} + e^{iQ_y} \right) + J_1 \left( e^{-2iQ_x} + e^{-2iQ_y} \right) \right],
\]

and simplifying

\[
\frac{i\hbar \omega}{S} S_{1}^x = -AS_{1}^y - CS_{1}^y - DS_{1}^y,
\]

where we have defined

\[
\begin{align*}
A &= 2J + 2J_1 + J_z, \\
B &= A + J_z, \\
C &= J \left( e^{iQ_x} + e^{iQ_y} \right) + J_1 \left( e^{-2iQ_x} + e^{-2iQ_y} \right), \\
D &= J_z e^{-iQ_x}.
\end{align*}
\]
Diagonalizing this matrix yields the squared eigenvalues

\[ \frac{i\hbar \omega}{\mathcal{S}} S^y_1 = -A S^y_1 - C S^y_2 D S^y_3, \quad \frac{i\hbar \omega}{\mathcal{S}} S^y_2 = +A S^y_2 + C^* S^y_1 D S^y_3, \]

\[ \frac{i\hbar \omega}{\mathcal{S}} S^x_3 = +B S^y_3 + C S^y_1 D S^y_4 + D^* S^y_1, \quad \frac{i\hbar \omega}{\mathcal{S}} S^x_4 = -B S^y_3 + C^* S^y_1 D S^y_4 - D^* S^y_1, \]

\[ \frac{i\hbar \omega}{\mathcal{S}} S^x_5 = -AS^y_5 - CS^y_5 - DS^y_6 - D^* S^y_5, \quad \frac{i\hbar \omega}{\mathcal{S}} S^x_6 = +AS^y_5 + CS^y_5 + D^* S^y_5, \]

This results in a 12 \times 12 secular matrix (6 spins, 2 components, x, y, per spin)

\[
M = \begin{pmatrix}
0 & -A & 0 & -C & 0 & -D & 0 & 0 & 0 & 0 & 0 & 0 \\
A & 0 & C & 0 & D & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & C^* & 0 & A & 0 & 0 & 0 & D & 0 & 0 & 0 & 0 \\
-C^* & 0 & -A & 0 & 0 & 0 & -D & 0 & 0 & 0 & 0 & 0 \\
0 & D^* & 0 & 0 & 0 & B & 0 & C & 0 & D & 0 & 0 \\
-D^* & 0 & 0 & 0 & -B & 0 & -C & 0 & -D & 0 & 0 & 0 \\
0 & 0 & 0 & -D^* & 0 & -C^* & 0 & -B & 0 & 0 & 0 & -D \\
0 & 0 & 0 & D^* & 0 & C^* & 0 & B & 0 & 0 & 0 & D \\
0 & 0 & 0 & 0 & -D^* & 0 & 0 & 0 & A & 0 & -C & 0 \\
0 & 0 & 0 & D^* & 0 & 0 & 0 & A & 0 & C & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & D^* & 0 & C^* & 0 & A \\
0 & 0 & 0 & 0 & 0 & 0 & D^* & 0 & -C^* & 0 & -A & 0
\end{pmatrix}.
\] (10)

Diagonalizing this matrix yields the squared eigenvalues

\[ \lambda_1^2(Q) = -A^2 + C^2C \] (11)

and

\[ \lambda_{2,3}^2(Q) = -A^2 - \frac{B}{2} + C^2C + 2J_1^2 \pm \frac{1}{2} J_2 \sqrt{(A + B)^2 + 32C^2C - 8J_2^2}, \] (12)

each of which are two-fold degenerate because of the tetragonal symmetry of the ground state. As \( i\omega = \gamma \), these eigenvalues correspond to the magnon branches

\[ \omega_{\text{middle}}(Q) = \sqrt{A^2 - C^2C} \] (13)

and

\[ \omega_{\text{acoustic, optic}}(Q) = \sqrt{\frac{A^2}{2} + \frac{B}{2} - C^2C - 2J_2^2 + \frac{1}{2} J_2 \sqrt{(A + B)^2 + 32C^2C - 8J_2^2}}. \] (14)

To justify these labels, and give some sense of these energies, we consider the \( \Gamma \) point. Substituting \( Q = 0 \) in Eq. 8 gives \( C = 2J + 2J_1 \) and \( D = J_2 \), which shows that our labels were chosen in order of increasing energy:

\[
\omega_{\text{acoustic}}(0) = 0,
\quad \omega_{\text{middle}}(0) = SJ_z \sqrt{1+2C/J_z} \sim SJ_z \sqrt{2J_z C},
\quad \omega_{\text{optic}}(0) = SJ_z \sqrt{1+6C/J_z} \sim SJ_z \sqrt{6J_z C},
\] (15)

where the approximation applies for \( J_z \ll J \). The magnon dispersion for the parameters determined by the fit to the RIXS data, \( J = 69 \text{ meV}, J_1 = 17 \text{ meV} \) (with \( J_z = 13.6 \text{ meV} \)), can be seen in Fig. S5.
FIG. S5. Magnon dispersion with $J=69 \text{ meV}$, $J_1=17 \text{ meV}$ and $J_z=13.6 \text{ meV}$, as in the main text, along the $(Q,Q)$ and $(Q,0)$ directions (with $Q_z=0$). The dashed curves in the left plot are for the twin domain $(Q,-Q)$.

B. Calculation of the RIXS intensities

As explained in the main text and Ref. [7], the RIXS intensity for each mode, labeled $n$, can be written as

$$I_n(Q) = \left| \sum_i k_{in} \cdot M_{n,Q}(r_i) \right|^2,$$

with $i$ summed over the six spins in the magnetic unit cell and

$$M_{n,Q}(r_i) = \left( c_{n,Q,i}^x S_i^x, c_{n,Q,i}^y S_i^y, 0 \right).$$

As the analytic expressions for the eigenvectors are complicated, we chose to determine them by diagonalizing the secular matrix Eq. 10 numerically for each $Q$ using SciPy [8]. As noted above, each distinct magnon branch within our model has two degenerate eigenvalues, with the two members of each pair related by a 90 degree in-plane rotation because of the tetragonal symmetry. That is

$$\left( c_1^x, c_1^y, c_2^x, c_2^y, ... \right)$$

is degenerate with

$$\left( -c_1^y, c_1^x, -c_2^y, c_2^x, ... \right)$$

with $n,Q$ being implicit. It is important to enforce this symmetry when calculating the RIXS intensity.

For the fit shown in the text, we took into account the scattering geometry of the RIXS measurements, with

$$k_{in} = \cos \theta I - \sin \theta Q,$$

and

$$|Q| = \frac{4\pi \sin \theta}{\lambda_E},$$

with $\theta$ the Bragg angle and $\lambda_E$ the photon wavelength. Here,

$$I = \frac{Q \times c \times Q}{|c \times Q|}.$$

In particular, as the in-plane component of $Q$ is swept, the $Q_z$ component changes accordingly.
VI. FIRST-PRINCIPLES DETERMINATION OF EXCHANGE COUPLING CONSTANTS IN La$_4$Ni$_3$O$_8$

We performed DFT calculations for La$_4$Ni$_3$O$_8$ with the all-electron full potential code WIEN2k [9, 10] using the generalized gradient approximation (GGA) exchange-correlation functional [11]. In these calculations, as we did previously [2, 12], we considered the influence of the Coulomb interaction, $U$. The $U$ modification of DFT is usually included to compensate for the under-localization of transition-metal 3$d$-electrons in DFT. Including $U$, conversely, tends to over-localize electrons as it “double counts” the true Coulomb and exchange interactions as explained in, for example, Refs. [13–15]. Because of this, it is not immediately obvious whether including $U$ leads to a more accurate value of $J$. In calculations, we found that the inclusion of $U$ simply increases the size of the gap, and also leads to a modest increase in $J$, as outlined in Table I. Calculations with and without $U$ find the same insulating, charge and spin stripe-ordered ground state, which was predicted before this state was experimentally observed [5]. In our prior work, we examined the role of $U$ at length and found that the low-spin stripe state La$_4$Ni$_3$O$_8$ was appropriately described by GGA calculations even without the inclusion of $U$ [2, 12]. We refer the reader to these papers for the reasoning behind this. Since it allows us to compute $J$ with fewer adjustable parameters, we report values from GGA calculations in the main text, and find that this is in good accord with experiment. As is evident based on how $J$ changes with $U$, the very close match between theory and experiment of 2 meV at the GGA level is likely coincidental.

We note that similar values for the superexchange can be obtained from rough estimates using a sum of the Mott and charge transfer contributions, $J = 2t^2/U_d + 2t^2/|\Delta + U_p/2|$, with $t = t_{pd}/\Delta$. Using $U_d = 8.5$ eV and $U_p = 7.3$ eV values from a similar analysis on the cuprates [16], with a $\Delta$ and $t_{pd}$ obtained from our Wannier fit for La$_4$Ni$_3$O$_8$ [17], a comparable $J$ of 99 meV is obtained. But this, obviously, depends on the choice of $U_d$ and $U_p$. Spin-orbit coupling is not expected to have an appreciable effect on the exchange constants for 3d transition metal ions, especially for $e_g$ states where the orbital moment is largely quenched. This has been explicitly verified in our prior calculations [5].

The exchange couplings ($J_1$, $J_2$ and $J_3$) were obtained from total energy calculations for different Ni spin configurations (labeled C1-C4 in Fig. S6) mapped to a Heisenberg model. Configuration C1 is the experimental and theoretical ground state as shown in Fig. 1 of the main text. The magnitudes of the magnetic moments of the Ni$^{2+}$ atoms were between 0.6-0.7 $\mu_B$ and we confirmed that these values were similar within 0.1 $\mu_B$ in the different configurations, an accuracy typical of this type of calculation, justifying the Heisenberg mapping. Different configurations C1-C4 have
differing magnetic bonds:

- C1 – AFM $J$, AFM $J_1$, AFM $J_z$
- C2 – AFM $J$, AFM $J_1$, FM $J_z$
- C3 – FM $J$, FM $J_1$, FM $J_z$
- C4 – FM $J$, AFM $J_1$, FM $J_z$

The energies per trilayer are (with each bond counted once)

$$E_{C1} = E_0 - 4 \times 3S^2 - 3 \times 4J_1S^2 - 2 \times 4J_zS^2$$
$$= E_0 - 3J - 3J_1 - 2J_z$$
$$E_{C2} = E_0 - 3J - 3J_1 + 2J_z$$
$$E_{C3} = E_0 + 3J + 3J_1 + 2J_z$$
$$E_{C4} = E_0 + 3J - 3J_1 + 2J_z$$

where $E_0$ is the non-magnetic energy. Solving this set of linear equations gives

$$J = (E_{C4} - E_{C2})/6$$
$$J_1 = (E_{C3} - E_{C4})/6$$
$$J_z = (E_{C2} - E_{C1})/4.$$

whose values are listed in the main text.

**VII. X-RAY ABSORPTION**

Our X-ray absorption spectrum (XAS) measurements aim to compare the intensity of the O $K$-edge pre-peak feature as a guide to the $3d-2p$ orbital hybridization in nickelates and cuprates. We analyze our data on La$_4$Ni$_3$O$_8$ against literature data for La$_{2-x}$Sr$_x$CuO$_4$ from Ref. [18] and Nd$_{1-x}$Sr$_x$NiO$_2$ from Ref. [19]. As far as we are aware, Nd$_{1-x}$Sr$_x$NiO$_2$ data are only available for in-plane polarization, so we only show this component of the polarization. The intensity of the spectra are scaled to have equivalent intensities for energies above 538 eV past the main O $K$-edge step. Since different measurements can have different absolute energy calibrations, we used measurements of different reference samples to put the spectra on the same energy-scale. We use Ref. [2] as our reference energy calibration for which La$_4$Ni$_3$O$_8$ was measured alongside La$_{2-x}$Sr$_x$CuO$_4$ and SrTiO$_3$. We then used the SrTiO$_3$ reference measurements in [19] to put Nd$_{1-x}$Sr$_x$NiO$_2$ on the same energy scale. While in La$_4$Ni$_3$O$_8$ and La$_{2-x}$Sr$_x$CuO$_4$ the pre-peak is very clear, the pre-peak in Nd$_{1-x}$Sr$_x$NiO$_2$ is broader making isolating the pre-peak less immediately obvious. We took the same ‘background’ intensity as was used in [19], which comes from a measurement of undoped NdNiO$_2$ and use a lorentzian lineshape to fit. The dominant error in our analysis likely comes from inhomogeneity in the doping of the Nd$_{1-x}$Sr$_x$NiO$_2$ results we compare to and some uncertainty in how to isolate all the intensity in the pre-peak. These will likely improve in the future by higher quality sample preparation. Further analysis of the ligand-hole anisotropy will also be important, but also requires polarization-dependent measurements of Nd$_{1-x}$Sr$_x$NiO$_2$ that are not currently available in the literature.


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<th>GGA (meV)</th>
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