




















## Magnetic excitations in $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$ Ruddlesden-Popper nickelates observed via resonant inelastic x-ray scattering

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Magnetic interactions are thought to play a key role in the properties of many unconventional superconductors, including cuprates, iron pnictides, and square-planar nickelates. Superconductivity was also recently observed in the bilayer and trilayer Ruddlesden-Popper nickelates, the electronic structure of which is expected to differ from that of cuprates and square-planar nickelates. Here we study how electronic structure and magnetic interactions evolve with the number of layers,  $n$ , in thin film Ruddlesden-Popper nickelates  $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$  with  $n = 1, 3$ , and 5 using resonant inelastic x-ray scattering (RIXS). The RIXS spectra are consistent with a high-spin  $|3d^8\bar{L}\rangle$  electronic configuration, resembling that of  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  and the parent perovskite,  $\text{NdNiO}_3$ . The magnetic excitations soften to lower energy in the structurally self-doped, higher- $n$  films. Our observations confirm that structural tuning is an effective route for altering electronic properties, such as magnetic superexchange, in this prominent family of materials.

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### I. INTRODUCTION

While much remains unknown about unconventional superconductivity, strong magnetic superexchange and reduced dimensionality likely play important roles in achieving high superconducting transition temperatures [1–3]. The square-planar family of nickelates, including the infinite-layer  $R\text{NiO}_2$  ( $R = \text{La}, \text{Pr}, \text{Nd}$ ) and the quintuple-layer  $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ , fits nicely into this picture in many ways, featuring two-dimensional transition-metal oxide planes and a  $d^{9-\delta}$  electronic configuration [4–6]. High energy magnetic excitations are observed throughout the nickelate phase diagram [7–13], although much lower critical temperatures are found in the square-planar nickelates than in cuprates. Comparing these two seemingly similar families of materials can help uncover the origins of superconductivity and identify new strategies to optimize superconductivity.

Recently, superconductivity was also observed in bilayer and trilayer Ruddlesden-Popper nickelates  $R_3\text{Ni}_2\text{O}_7$  ( $n = 2$ )

and  $R_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) for  $R = \text{La}, \text{Pr}$ , under pressure [14–21], or epitaxial strain [22–25]. This discovery has expanded the variety of superconducting nickelates beyond the square-planar geometry [4–6], to octahedrally coordinated nickelates and to substantially higher critical temperatures [14,15]. More generally, the layered Ruddlesden-Popper nickelates,  $R_{n+1}\text{Ni}_n\text{O}_{3n+1}$ , provide a way to explore the nickelate phase diagram through structural tuning. As shown in Fig. 1, the Ruddlesden-Popper structure consists of  $n$  layers of perovskite  $R\text{NiO}_3$  separated by  $(R\text{-O})^+$  charged rock-salt layers. Increasing  $n$  tunes the effective electron count of the  $\text{NiO}_2$  planes by  $1/n$  per nickel, from a  $d^8$  configuration in the  $R_2\text{NiO}_4$  ( $n = 1$ ) compound to a nominal  $d^7$  configuration in the  $R\text{NiO}_3$  ( $n = \infty$ ) compound, tuning the electronic behavior from semiconducting to metallic (see Supplemental Material, Sec. S1 C [26]) while in principle avoiding the disorder associated with chemical doping. This positions the layered Ruddlesden-Popper nickelates as a promising material family for exploring and tuning the superconducting ground state.

Although these materials share the layered perovskite structure common to many cuprates, it is unclear whether they fit into the same “cuprate-like” picture which may be relevant to the square-planar nickelates [27–32]. The square-planar nickelates adopt a  $d^{9-1/n}$  configuration, with dominant in-plane orbital polarization [33]. The key electronic

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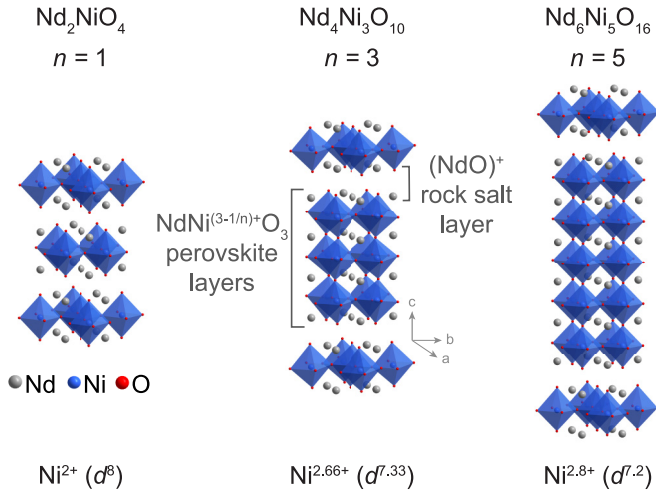


FIG. 1. Structures of Ruddlesden-Popper  $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$  nickelates for  $n = 1, 3,$  and  $5$ . The unit cell hosts  $n$   $\text{NdNiO}_3$  perovskite layers separated by  $\text{NdO}^+$  rock-salt spacer layers. As  $n$  varies, the nominal Ni valence changes as  $d^{7+1/n}$ .

interactions in the octahedrally coordinated Ruddlesden-Popper nickelates may differ substantially from the square-planar nickelates, as Ruddlesden-Popper nickelates have a nominal  $d^{7+1/n}$  configuration, with holes occupying both the in-plane  $3d_{x^2-y^2}$  and out-of-plane  $3d_{3z^2-r^2}$  orbitals [34–36]. The character of the doped holes may also differ: while undoped  $n = 1$  cuprates and nickelates are both antiferromagnetic insulators, lightly hole-doped cuprates quickly become metallic and a superconducting dome emerges [2]. Magnetic excitations are broadened due to increased metallicity, but remain at high energy [37]. In contrast, doped  $n = 1$  nickelates do not exhibit superconductivity, but instead display a wide array of insulating stripe phases [38–42]. Holes and spins are bound via electron-electron and electron-phonon couplings, forming polarons [43]. This localizes spins in the lattice, resulting in a dramatic softening of the magnetic excitations with hole doping [44,45].

Structural tuning across the Ruddlesden-Popper nickelate series offers access to a distinct subspace of material phases and may help identify properties that are inaccessible by doping alone. Here we study how electronic and magnetic interactions evolve with layer number  $n$ , performing high-resolution inelastic x-ray scattering (RIXS) measurements on  $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$  thin films for  $n = 1, 3,$  and  $5$ . Orbital excitations reveal the expected  $|d^8\rangle$  orbital configuration in the  $n = 1$  compound. Upon increasing  $n$ , these features evolve towards a  $|d^8\bar{L}\rangle$  configuration, indicating holes are primarily added to the ligand bands rather than inducing a  $|d^7\rangle$  state. Magnetic excitations with an energy scale of order 50–70 meV are prominent throughout the entire family of materials, with softening at higher  $n$  due to increased doping. The observed softening is smaller than that expected based on effective doping alone, as dimensionality effects from increasing  $n$  partially offset the doping-induced softening. Our observations demonstrate that the Ruddlesden-Popper nickelates share many key features with other perovskite nickelates, with structural tuning providing an effective and unique approach to modify electronic and magnetic properties.

## II. EXPERIMENTAL METHODS

RIXS has proven effective in determining the properties of nickelates [30,46]. We applied this technique to thin films of the  $n = 1, 3,$  and  $5$  layer Ruddlesden-Popper  $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$  compounds, taken at the Ni  $L_3$  edge and with  $\pi$ -polarized light in order to maximize the magnetic signal. RIXS measurements for the  $n = 1$  and  $3$  compounds were performed at the National Synchrotron Light Source II beamline 2-ID [47] at a temperature of 35 K, at a scattering angle of  $2\theta = 150^\circ$ , and with an experimental resolution of 31 meV. Measurements for the  $n = 5$  compound were performed at Diamond Light Source beamline ID21 [48] at a temperature of 20 K, at a scattering angle of  $2\theta = 154^\circ$ , and with an experimental resolution of 36 meV. To probe the dispersion of magnetic excitations, the scattering angle  $2\theta$  was kept fixed while the sample angle  $\theta$  was rotated to control the in-plane momentum transfer to the sample, along the  $[H, 0, L]$  direction of the pseudotetragonal unit cell. In order to compare spectra collected from different samples and at different beamlines, we present spectra normalized to the integrated intensity of the orbital excitations [49].

$\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$  films were synthesized using oxide molecular-beam epitaxy as described in Refs. [50,51]. All films are epitaxially strained to the substrate. We focus on samples synthesized on (110)-oriented  $\text{NdGaO}_3$ , which has a pseudoperovskite lattice constant of 3.86 Å and thus provides a small amount of tensile strain,  $\epsilon \approx +1.0\%$ , to the Ruddlesden-Popper nickelate family [52]. Additional data for  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  on (001)-oriented  $\text{LaAlO}_3$  ( $\epsilon \approx -0.9\%$ ) are shown in the Supplemental Material, Sec. S4 [26]. Structural and electrical sample characterization is detailed in the Supplemental Material, Sec. S1. By studying compounds where the rare-earth site is occupied by neodymium rather than the more commonly studied lanthanum series, we avoid contamination on Ni  $L_3$ -edge RIXS from the nearby La  $M_4$  absorption edge. We also emphasize that thin film techniques provide unique access to higher- $n$  Ruddlesden-Popper phases, as only compounds up to  $n = 3$  can be synthesized in bulk [53].

## III. ORBITAL EXCITATIONS AND HOLE CONFIGURATION

To study the electronic structure of  $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$  we examine the orbital excitations observed in RIXS spectra, shown in Fig. 2 for films with  $n = 1, 3,$  and  $5$ . In  $\text{Nd}_2\text{NiO}_4$  ( $n = 1, 3d^8$ ) we observe two sharp orbital excitations at 1.04 and 1.6 eV, in good agreement with published RIXS data on bulk  $\text{La}_2\text{NiO}_4$  [44]. In  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ , nominal  $d^{7.33}$ ) in  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ , nominal  $d^{7.2}$ ), the orbital excitations broaden in energy but remain centered at similar energies.

$\text{La}_2\text{NiO}_4$  is known to adopt a Ni  $3d^8$  high-spin configuration in which the two holes reside in the Ni  $3d e_g$  orbitals and have parallel spin due to the Hund’s exchange interaction (Fig. 2). The orbital excitations were previously shown to be well captured by excitations from this atomic configuration [44] and agree with the excitations we observe in  $\text{Nd}_2\text{NiO}_4$  ( $n = 1$ ). The lower energy orbital excitation at 1.04 eV is attributed to a transition from the  $B_{1g}$  symmetric

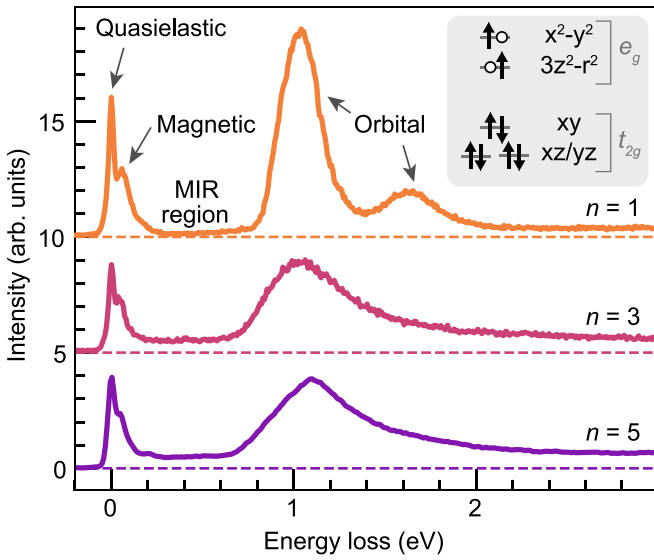


FIG. 2. RIXS spectra of Ruddlesden-Popper nickelates for  $n = 1$  (orange), 3 (dark pink), and 5 (purple). The majority of the spectral weight observed around  $\approx 1$  eV and above corresponds to orbital excitations. Spectra are collected at  $q = (-0.4, 0)$  reciprocal lattice units ( $\theta = 24^\circ$ ) with  $\pi$ -incident polarization, probing primarily out-of-plane orbitals. Data for  $n = 1$  and 3 were collected at 35 K, while data for  $n = 5$  were collected at 20 K. The inset depicts the ground state orbital configuration for  $n = 1$ .

ground state to an  $E_g$  symmetric state with one of the holes moving into the  $t_{2g}$  orbitals. The higher energy peak at 1.6 eV is composed of an  $A_{1g}$  excitation from an  $S = 1$  to 0 configuration, a  $B_{2g}$  excitation with one of the holes transferred to the  $t_{2g}$  manifold, and two  $A_{2g}$  excitations, one with a single hole in the  $t_{2g}$  manifold and one with both holes in the  $t_{2g}$  manifold [44]. Thus the orbital excitations in  $\text{Nd}_2\text{NiO}_4$  ( $n = 1$ ) can be well captured as excitations of the local  $d$  multiple.

As  $n$  is increased, additional holes are doped into the perovskite layers, modifying the ground state and the orbital excitations. In  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) the Ni sites have a nominal  $d^{7.33}$  valence and in  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ) the Ni sites have a nominal  $d^{7.2}$  valence. Nonetheless, the orbital excitations remain centered at approximately the same energies, and the polarization dependence indicates that holes remain roughly equally distributed between the in-plane and out-of-plane Ni  $e_g$  orbitals (see Supplemental Material, Fig. S6 [26]). The orbital excitations broaden such that the two features can no longer be separately resolved, but remain centered at similar energies.

The broadenings of the orbital excitations in  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) and  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ) are indicative of a  $d^8\bar{L}$  state rather than a  $d^7$  state, sharing a striking similarity to the orbital excitations observed in hole-doped  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  ( $n = 1$ ) [44] and metallic  $\text{NdNiO}_3$  ( $n = \infty$ ) [54,55], materials both known to adopt a  $d^8\bar{L}$  configuration. In addition to hole doping, possible small variations in the crystal field environment between inequivalent Ni layers in  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) and  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ) could additionally contribute to the observed broadening of the  $d^8$  orbital excitations, with each inequivalent layer contributing excitations at slightly different

energies. In contrast, the observed orbital excitations are inconsistent with a contribution from a  $d^7$  configuration—such a contribution has been calculated in Refs. [44,54] and was shown to yield additional peaks in the RIXS spectrum outside the energy range of these main  $d^8$  excitations, above 2 eV and around 0.25 eV respectively.

We believe the spectral changes with increasing  $n$  are driven primarily by changes in the effective doping rather than changes in hybridization. To leading order, the in-plane environment is expected to remain unchanged, as the Ni-O bond lengths are fixed for all  $n$  by the substrate epitaxy. Changes in the apical Ni-O bonding with increasing  $n$  may play a small role in the observed changes, but would manifest most strongly in changes to the orbital dichroism, which are not observed. We therefore consider doping to be the driving force behind the observed changes, consistent with the strong similarity of these data with prior measurements of hole-doped  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  ( $n = 1$ ) [44].

In addition to hole doping the perovskite layers, structural tuning introduces additional interlayer couplings between adjacent Ni planes within each perovskite block, which might further modify the orbital excitations beyond a single-site picture. In bulk  $\text{La}_3\text{Ni}_2\text{O}_7$  ( $n = 2$ ), an additional, Raman-like orbital excitation was observed at 0.4 eV, which is attributed to transitions between  $d_{x^2-y^2}$  and  $d_{3z^2-r^2}$  orbitals [9]. The energy of this excitation is primarily determined by the interlayer hopping, which results in the formation of molecular subbands [56]. If such features occurred in the higher  $n$  films studied here, they would appear at substantially different energies, with differences of order  $t$  [56], which would be easily resolvable. We do not see evidence of any such additional peaks in the orbital excitations for  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) and  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ), which may be because for higher  $n$  the distinct orbital subbands are strongly concentrated on distinct Ni layers within the perovskite blocks [56], minimizing their cross section for the highly local RIXS process.

As shown in the Supplemental Material, Fig. S5 [26],  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ) and  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) exhibit strong x-ray fluorescence features in which RIXS intensity appears at increasing energy loss as the incident energy increases. This is distinct from regular  $dd$  excitations that appear at fixed energy loss independent of incident energy. These fluorescence features arise from and indicate the presence of hybridization between transition metal states and itinerant ligand states [34,35,57]. Similar hybridization features are commonly observed in cuprates [58] and nickelates [30,32,34,35,44,54,57,59,60], including  $\text{NdNiO}_3$  ( $n = \infty$ ) [54] and hole-doped  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  ( $n = 1$ ) [44], underscoring the role of hybridized itinerant ligand states as a unifying feature across these materials.

We also observe featureless, nondispersing spectral weight in the midinfrared (MIR) region, 0.3–0.7 eV, in the  $n = 3$  and 5 compounds, which is nearly absent in the  $n = 1$  compound (Fig. 2). This energy scale is above that of magnons and multimagnons. This MIR spectral weight appears only at the x-ray-absorption spectroscopy resonance (See Supplemental Material, Fig. S5 [26]), phenomenologically different from the clear Raman peak at 0.4 eV in  $\text{La}_3\text{Ni}_2\text{O}_7$  ( $n = 2$ ) which is attributed to transitions between  $d_{x^2-y^2}$  and  $d_{3z^2-r^2}$  orbitals. Instead, the flat filling in of the

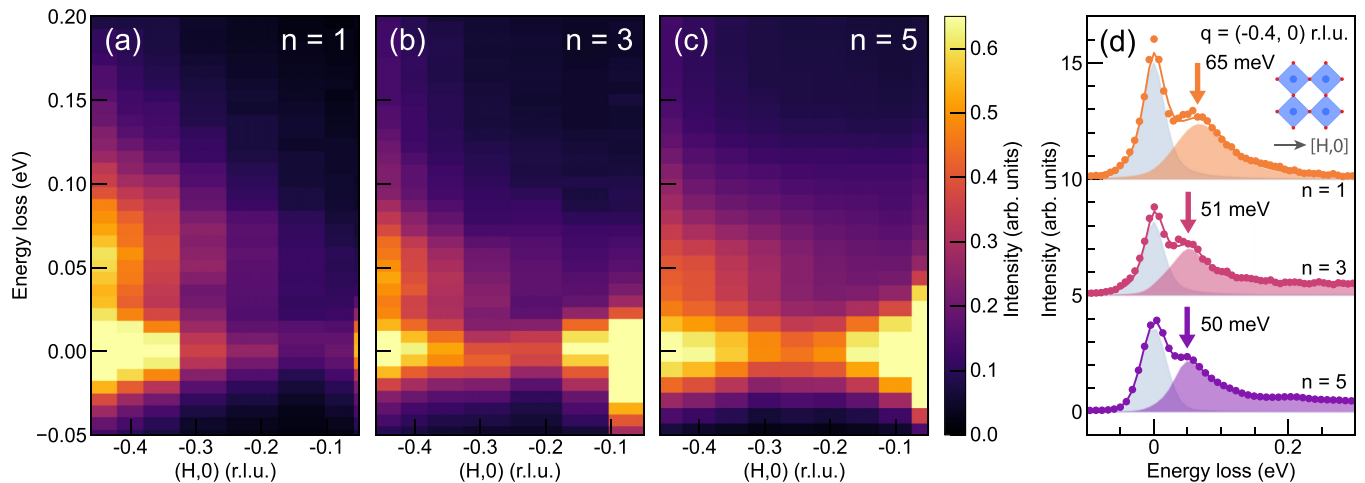


FIG. 3. RIXS spectra along  $[H, 0]$  for  $\pi$ -incident polarization for (a)  $n = 1$ , (b)  $n = 3$ , and (c)  $n = 5$ . The inelastic components of the RIXS spectra are dominated by magnetic excitations. (d) Linecuts at  $q = (0.4, 0)$  for  $n = 1$  (orange),  $n = 3$  (dark pink), and  $n = 5$  (purple) with the quasielastic scattering shown in gray and the inelastic (magnetic) scattering shown in color. Data for  $n = 1$  and 3 were collected at 35 K, while data for  $n = 5$  were collected at 20 K.

MIR spectral weight appears phenomenologically very similar to the behavior observed in  $\text{NdNiO}_3$  ( $n = \infty$ ) upon heating through a metal-insulator transition [54], attributed to charge excitations within the  $|d^8\bar{L}\rangle$  states and associated with partial metallicity. This is consistent with the electronic transport properties of these films: the  $n = 1$  material is semiconducting with an exponentially increasing resistivity in the low-temperature limit, while the  $n = 3$  and 5 materials, on the other hand, are metallic at room temperature with modest resistivity upturns at low temperature (see Supplemental Material, Sec. S1 C [26], as well as Refs. [60–63] therein).

Thus the high energy RIXS features support a  $|d^8\bar{L}\rangle$  configuration for Ruddlesden-Popper nickelates, with mobile holes doped into hybridized ligand states as  $n$  is increased.

#### IV. MAGNETIC EXCITATIONS MODIFIED BY STRUCTURE AND DOPING

To study the magnetic excitations in  $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$ , we examine the momentum dependence of the low energy-loss region of the RIXS spectra along the  $[H, 0]$  direction for films with  $n = 1, 3$ , and 5, shown in Fig. 3. RIXS spectra show an elastic line and a broad inelastic feature extending out to 0.2 eV. In all three compounds these features are most prominent around the zone boundary, near  $(0.4, 0)$ , and disperse to lower intensity and lower energy scales towards  $(0, 0)$ . This behavior is typical for magnetic branches, so, consistent with prior work [44, 64], we assign these features to damped magnetic excitations. With increasing  $n$ , additional inelastic spectral weight appears approaching the zone center,  $q = (0, 0)$ , and extends out to higher energy losses in  $n = 3$  and 5 than in the  $n = 1$  sample.

As shown in Fig. 1, the unit cells of  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) and  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ) include sets of coupled nickel-oxide trilayers and quintuple layers, so spin wave theory predicts a large number of distinct spin wave modes (Supplemental Material, Sec. S3 [26]). These modes are split by the interlayer magnetic exchange coupling,  $J_z$ , as has

been observed in the bilayer systems  $\text{La}_3\text{Ni}_2\text{O}_7$  [9] and  $\text{Sr}_3\text{Ir}_2\text{O}_7$  [65, 66]. Due to the large number of modes and mode-broadening effects arising from finite effective doping, resolving all the predicted modes individually is unlikely to be feasible. In fact, previous measurements of the trilayer square-planar nickelate  $\text{La}_4\text{Ni}_3\text{O}_8$  observed only the average of the three modes expected based on spin wave theory [8].

We attribute the enhanced spectral weight around 0.1 eV near  $q = (0, 0)$  in the higher  $n$  compounds to a magnetic branch with partially optical character. The optical spectral weight in  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) appears weaker than in  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ), which may be due to a reduced importance of interplane interactions in  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$ . Neutron scattering measurements on bulk  $\text{La}_4\text{Ni}_3\text{O}_{10}$  indeed reveal a concentration of spin density in the outer layers of the trilayer block [67], reducing the magnetic coupling between planes.

Since the individual spin wave modes in the  $n = 3$  and 5 materials cannot be resolved individually, we use a phenomenological model to separate the inelastic features from the elastic line and compare results for  $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$  compounds with  $n = 1, 3$ , and 5. The phenomenological model includes a pseudo-Voigt peak for the quasielastic scattering, and an error function for the flat midinfrared background in the  $n = 3$  and 5 compounds. For  $n = 1$  and 3 compounds we use a single damped harmonic oscillator function to capture the remaining inelastic spectral weight. For  $n = 5$  we were better able to capture the inelastic features by instead using four pseudo-Voigt peaks, though our results are insensitive to the exact fitting model used. An example of the total fit is shown for  $q = (0.4, 0)$  in Fig. 3(d). The quasielastic scattering is indicated in light gray, while the sum of the inelastic components is indicated by the shaded colored region. The position of maximum intensity of the inelastic components is taken as the magnon bandwidth and is indicated by the arrows. At  $q = (0.4, 0)$  the magnetic excitations are peaked at 65 meV in  $\text{Nd}_2\text{NiO}_4$  ( $n = 1$ ) and soften to 51 and 50 meV in  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) and  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ), respectively.

The RIXS spectra for the  $\text{Nd}_2\text{NiO}_4$  ( $n = 1$ ) sample qualitatively agree with published magnetic dispersions for  $\text{La}_2\text{NiO}_4$  ( $n = 1$ ) [44,64], but the magnon bandwidth is reduced to about 75% of that observed in the La-based material, consistent with reported differences between bulk  $\text{Nd}_2\text{NiO}_4$  and  $\text{La}_2\text{NiO}_4$  [68]. Larger perovskite distortions and rotations in Nd-based compounds can play a key role in driving this reduction in magnetic energy scales by bending the Ni-O-Ni bond angles [63,69,70], weakening antiferromagnetic superexchange interactions. These rotations can also increase spin canting, introducing a ferromagnetic out-of-plane magnetization component which can cause an effective reduction in the magnon energy [68,71–73]. Lastly, the large magnetic moment on the neodymium atoms can further interact with magnetic moments on Ni sites, competing with in-plane magnetic exchange interactions and introducing a large magnetic anisotropy [68].

Magnetic energy scales are reduced by about 25% in  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) and  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ) relative to  $\text{Nd}_2\text{NiO}_4$  ( $n = 1$ ) (Fig. 3). We are unable to resolve a notable difference between  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) and  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ). In fact, the magnon bandwidth in both  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) and  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ) agrees well with that observed in  $\text{NdNiO}_3$  ( $n = \infty$ ) [55,74], suggesting that as  $n$  increases, magnetic interactions quickly approach the behavior seen in the end member of the series. For all  $n$  studied, the magnetic bandwidth is similar to that of the Ruddlesden-Popper bilayer  $\text{La}_3\text{Ni}_2\text{O}_7$  [9] and the square-planar trilayer  $\text{La}_4\text{Ni}_3\text{O}_8$  [8], indicating common magnetic interaction strengths across these diverse layered nickelate materials.

We interpret the softening of magnetic interactions with increasing  $n$  to be dominantly driven by the increase in effective doping, which can disrupt the antiferromagnetic exchange by introducing states that act as spinless impurities. This suggests analogies between Ruddlesden-Popper nickelates and other materials such as square-planar nickelates [7] and  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  ( $n = 1$ ), where magnon energies are reduced by a factor of 2 or more upon the addition of 0.5 holes/site [44,45,75,76]. Compared to these chemically doped nickelates, however, the 25% magnon softening in  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) and  $\text{Nd}_6\text{Ni}_5\text{O}_{16}$  ( $n = 5$ ) (Fig. 3) is less than that predicted solely on the basis of the effective doping of an additional 0.67 and 0.8 holes/site, respectively. The effective doping might be overestimated by simple electron counting arguments, as structurally driven doping is predicted to introduce additional correlations that renormalize the effective doping amount in the layered square-planar nickelates [77–79]. Holes may also be unevenly distributed between planes within  $n$ -layer blocks, leading to contributions from inequivalent  $\text{NiO}_2$  layers, further complicating the magnon mode structure probed by RIXS. However, both these effects would likely impart only small changes to the effective doping.

Magnetic energy scales may be further modified by the increase of dimensionality with increasing  $n$ , in addition to changes due to the effective doping of  $1/n$  holes per  $\text{Ni}^{2+}$  ion. Increasing  $n$  introduces an additional out-of-plane exchange pathway between neighboring  $\text{NiO}_2$  layers which is absent in  $\text{Nd}_2\text{NiO}_4$  ( $n = 1$ ). In  $\text{Nd}_2\text{NiO}_4$  ( $n = 1$ ), the magnetic Ni sites can interact with four neighboring Ni sites. In higher  $n$

compounds, Ni atoms in the inner layers can interact with six neighboring Ni sites, while Ni atoms in the two outer layers can interact with five neighboring Ni sites. Because each Ni atom now has more magnetic exchange pathways, we expect an overall increase in the magnetic bandwidth with increasing  $n$  (Supplemental Material, Sec. S3 [26]), competing with the overall decrease due to increased doping. The out-of-plane exchange interaction  $J_z$  can be quite strong due to the coupling of partially filled  $d_{3z^2-r^2}$  orbitals by apical oxygen atoms. In fact, we see already for  $\text{Nd}_4\text{Ni}_3\text{O}_{10}$  ( $n = 3$ ) that the magnetic bandwidth closely approaches the  $\approx 50$ -meV bandwidth seen in  $\text{NdNiO}_3$  ( $n = \infty$ ) [55,74]. Thus the introduction of these out-of-plane exchange interactions may play a key role in setting the magnetic energy scales in higher order  $n$  Ruddlesden-Popper nickelates, partially offsetting the magnetic softening due to increased doping.

## V. CONCLUSION

We used Ni  $L_3$ -edge RIXS to determine the evolution of the electronic and magnetic structure of layered Ruddlesden-Popper nickelates  $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{3n+1}$  with layer number  $n$ . We show that as  $n$  is increased, holes are doped into itinerant ligand bands, moving the system from a  $|d^8\rangle$  configuration for  $n = 1$  to a  $|d^8\bar{L}\rangle$  configuration. This finding agrees well with the behavior of the end members of the series,  $\text{NdNiO}_3$  ( $n = \infty$ ) and Sr-doped  $\text{La}_{2-x}\text{Sr}_x\text{NiO}_4$  ( $n = 1$ ), confirming systematic changes across this family of materials with doping and structural tuning. These doped holes cause a softening of the magnetic excitations in the higher  $n$  compounds; however, the softening is partially mitigated due to the introduction of additional out-of-plane magnetic interactions with increasing  $n$ , which increase the energy scale of magnetic interactions. Thus, structural and dimensional control can tune both electronic and magnetic properties of materials in a complex and interdependent fashion, allowing access to a richer material space than that accessible by chemical doping alone.

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#### DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

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