



Soft X-Ray Spectroscopy of Low-Valence Nickelates

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Low-valence nickelates—including infinite-layer (IL) and trilayer (TL) compounds—are longstanding candidates for mimicking the high-temperature superconductivity of cuprates. A recent breakthrough in the field came with the discovery of superconductivity in hole-doped IL nickelates. Yet, the degree of similarity between low-valence nickelates and cuprates is the subject of a profound debate for which soft x-ray spectroscopy experiments at the Ni *L*- and O *K*-edge provided critical input. In this review, we will discuss the essential elements of the electronic structure of low-valence nickelates revealed by x-ray absorption spectroscopy (XAS) and resonant inelastic x-ray scattering (RIXS). Furthermore, we will review magnetic excitations observed in the RIXS spectra of IL and TL nickelates, which exhibit characteristics that are partly reminiscent of those of cuprates.

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1 INTRODUCTION

Spectroscopy has proven to be a versatile tool for studying the charge, spin, lattice, and orbital degrees of freedom in quantum materials. Arguably, one of the most fascinating families of quantum materials are cuprate high-temperature superconductors [1–3]. Critical insights into cuprates have been provided by electron, neutron, and photon spectroscopy techniques—including photoemission and electron spectroscopy, scanning tunneling microscopy, optical and Raman spectroscopy, as well as inelastic x-ray and neutron scattering [4–10]. More recently, fresh perspectives on the enigmatic ground states of cuprates opened up especially due to resonant soft x-ray spectroscopy studies. For instance, resonant inelastic x-ray scattering (RIXS) [11] revealed that damped spin excitations persist even for high hole-doping levels far away from the parent antiferromagnetically (AFM) ordered phase [12, 13]. Furthermore, RIXS allowed to probe the three-dimensional (3D) dispersion of low-energy plasmons [14–16], which arise due to the characteristic quasi-2D layered crystal structure of cuprates. Moreover, resonant elastic x-ray scattering (REXS) and RIXS studies found that different types of static and dynamic charge orders emerge ubiquitously in cuprates [17–21]. Nevertheless, a comprehensive understanding of the most prominent phases in cuprates—including the pseudogap, strange metal phase, and superconductivity—remains elusive.

One approach to gain a deeper understanding of cuprates involves the targeted design of materials that mimic cuprate-typical properties, such as their layered quasi-2D crystal structure, $3d^9$ electronic configuration, spin $S = 1/2$ magnetic moments with antiferromagnetic (AFM) coupling, strong ligand-oxygen hybridization, and a lifted degeneracy of the active e_g orbitals [22]. In principle, the discovery of superconductivity in a material that emulates at least a subset of

these properties could allow to identify the hallmarks of cuprates that are crucial for invoking their exceptional high-temperature superconductivity.

In this context, long-standing candidates are Ni-based compounds, as Ni is a direct neighbor of Cu in the periodic table. In early works, it was speculated that doped RE_2NiO_4 (RE = rare-earth ion), which is the $n = 1$ member of the Ruddlesden-Popper (RP) homologous series $RE_{n+1}Ni_nO_{3n+1}$ [23], could become superconducting [24], as it is isostructural to the cuprate La_2CuO_4 and possesses similar charge and spin stripe ordered states [25]. The formal electronic configuration of La_2NiO_4 , however, is $3d^8$ (Ni^{2+}) with $S = 1$ [26, 27], providing a possible rationalization for the observed absence of superconductivity. Perovskite nickelates $RENiO_3$ are the $n = \infty$ member of the RP series with a formal $3d^7$ (Ni^{3+}) configuration and $S = 1/2$, although x-ray spectroscopic experiments indicated a $3d^8\bar{L}$ configuration [28], with \bar{L} denoting an O ligand hole. In their seminal work, Chaloupka and Khaliullin proposed a significant modification of the electronic structure of perovskite nickelates via tensile strain and incorporation into thin epitaxial heterostructures [29], breaking of the degeneracy of the e_g orbitals, which in the extreme case could yield a half filled $3d_{x^2-y^2}$ band and a single-sheet, cuprate-like Fermi surface [30]. In fact, spectroscopic studies detected new electronic and magnetic ground states in perovskite nickelate heterostructures, distinct from those of bulk nickelates [31–41]. Nevertheless, superconductivity has not been found to date [42–44], possibly due to the insufficient splitting of the orbital energy levels in the realized heterostructures [45, 46]. More recently, oxygen-reduced variants of the $n = 3$ members of the RP series have attracted intense attention. These $RE_4Ni_3O_8$ trilayer (TL) nickelates are composed of three closely stacked square-planar Ni-O layers with intervening RE ions, which are separated by rocksalt type RE -O blocking structures [47]. The electronic configuration of TL nickelates is $3d^{8.67}$ per Ni on average ($Ni^{1.33+}$). It was found by x-ray spectroscopy that metallic $Pr_4Ni_3O_8$ exhibits low-spin configuration and a significantly lifted orbital degeneracy, suggesting a close analogy to cuprates [48]. Notably, the close parallel between TL nickelates and cuprates was further corroborated by RIXS, revealing the presence of strong AFM exchange coupling J in $La_4Ni_3O_8$ and $Pr_4Ni_3O_8$ [49]. Nonetheless, superconductivity has not been detected in TL nickelates. This could be due to their electronic configuration, which corresponds to 1/3-hole-doping of a Ni^{1+} background and therewith falls into the overdoped regime of cuprates [48, 50, 51]. Experimental efforts to stabilize superconductivity via lowering the $Ni^{1.33+}$ valence by electron-doping are ongoing. Along these lines, also the $n = 5$ oxygen-reduced RP variants $RE_6Ni_5O_{12}$ are promising candidates for superconductivity, as these quintuple layer nickelates exhibit a $3d^{8.8}$ ($Ni^{1.2+}$) configuration—analogue to optimally doped cuprates—already without additional electron-doping.

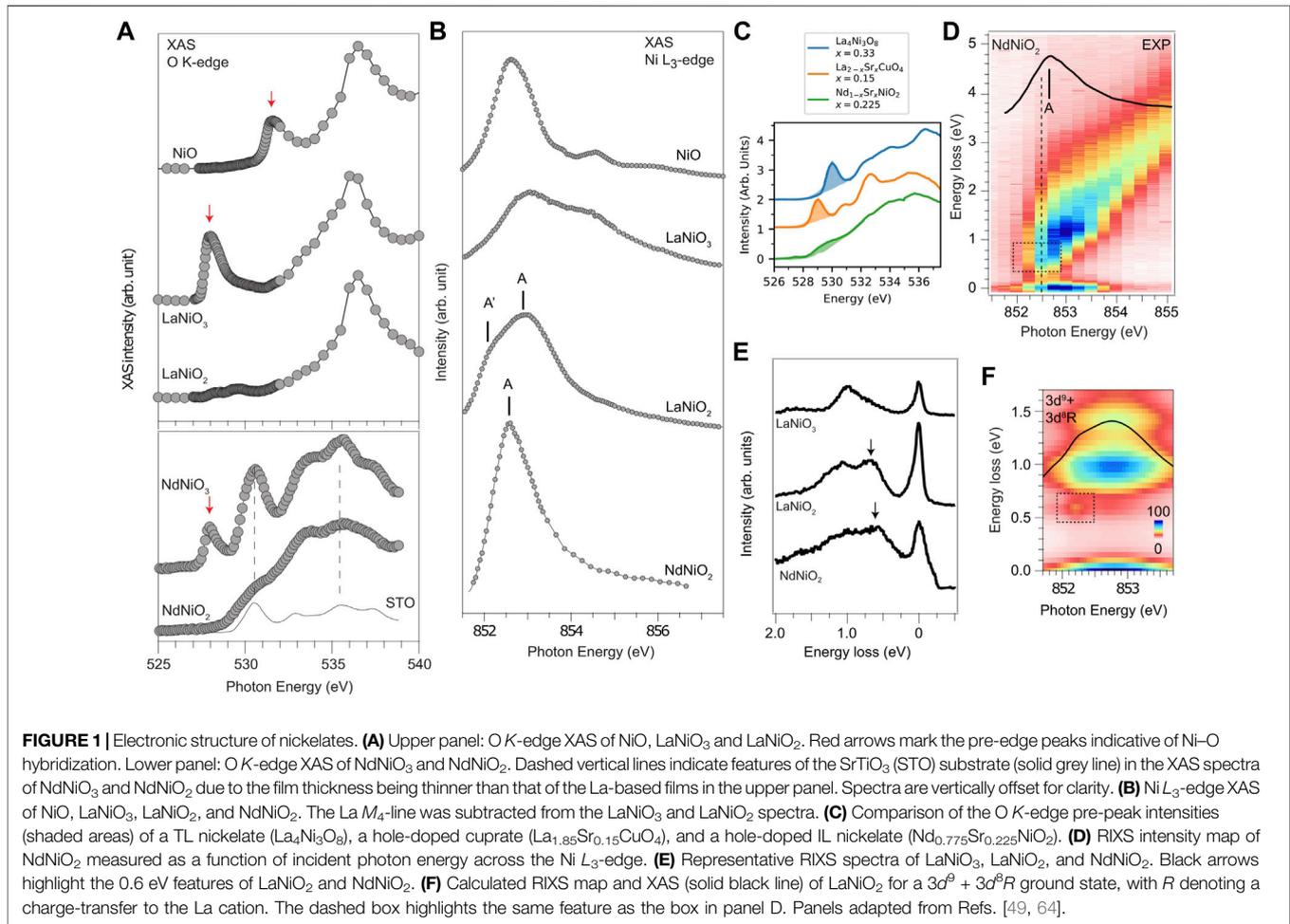
A breakthrough in the field came with the discovery of superconductivity in hole-doped nickelates with the IL crystal structure [52]. In more detail, epitaxial thin films of Sr- or Ca-substituted $RENiO_2$ obtained via topotactic oxygen reduction of

the perovskite phase show superconductivity below 9–15 K [52–58]. The parent compounds of these nickelates formally exhibit the $3d^9$ (Ni^{1+}) configuration with $S = 1/2$, which qualifies them as isostructural and isoelectronic to the parent cuprates. Early neutron powder diffraction studies of parent IL nickelates, however, indicated absence of long-range AFM order [59, 60] and electrical transport measurements of films show weakly metallic behavior [61]. This is in stark contrast to parent cuprates, which are AFM Mott (charge-transfer) insulators [1]. Moreover, whereas first theoretical studies proposed that the electronic and magnetic correlations of IL nickelates and cuprates share close similarities [62], other theoretical works suggested significant distinctions, including a multiband character of nickelates [63]. Along these lines, insights from experiments can help to resolve the controversy about similarities and differences between IL nickelates and cuprates. In particular, recent x-ray and electron energy-loss spectroscopic studies [64–66] unveiled a reduced Ni-O hybridization, presence of a weakly interacting RE 5d metallic band, and overdamped spin excitations with a bandwidth as large as 200 meV in IL nickelates.

In the following, we will review recent soft x-ray absorption spectroscopy (XAS) and RIXS studies at the O K -edge and Ni L -edge of IL and TL nickelates. We will discuss the essential elements of their distinct electronic structure. Furthermore, the spin excitation spectra of IL and TL nickelates observed with RIXS will be reviewed.

2 ELECTRONIC STRUCTURE

XAS at the O K -edge measures core-hole excitations from O 1s to unoccupied O 2p states and is also a sensitive probe of the covalent mixing between O 2p and transition-metal d states [67]. In particular the O- K pre-edge fine structure can provide valuable information about Ni-O hybridized states and the associated electronic structure, for instance in the cases of NiO and perovskite $RENiO_3$ [28, 67, 68]. In both materials, Ni 3d-orbitals strongly hybridize with oxygen ligands, giving rise to a pre-peak in the absorption spectra near the O K -edge (Figure 1A). Due to different relative energy scales between the charge-transfer energy Δ and the Coulomb interaction U , according to the Zaanen–Sawatzky–Allen (ZSA) scheme [69], the former material falls into the regime of charge-transfer insulators, whereas the latter is a negative charge-transfer compound. In contrast, the O K -edge absorption spectra of the IL nickelates $LaNiO_2$ and $NdNiO_2$ lack a prominent pre-edge peak (Figure 1A), suggesting a substantially weaker effective mixing between oxygen and the unoccupied 3d states of the upper Hubbard band (UHB) of the Ni^{1+} cations [64]. In the case of cuprates, a prominent pre-peak feature is present in O K -edge absorption spectra [70]. This is known to originate from the charge-transfer nature of these materials, with Δ smaller than U , and O 2p states mixed with both the lower Hubbard band (LHB) and the UHB of the Cu $3d_{x^2-y^2}$ states. A sizable pre-peak has also been observed in TL nickelates (Figure 1C), indicating mixing between O 2p and the Ni UHB [48, 49]. Upon hole-doping of cuprates, spectral weight shifts from the UHB pre-peak to a

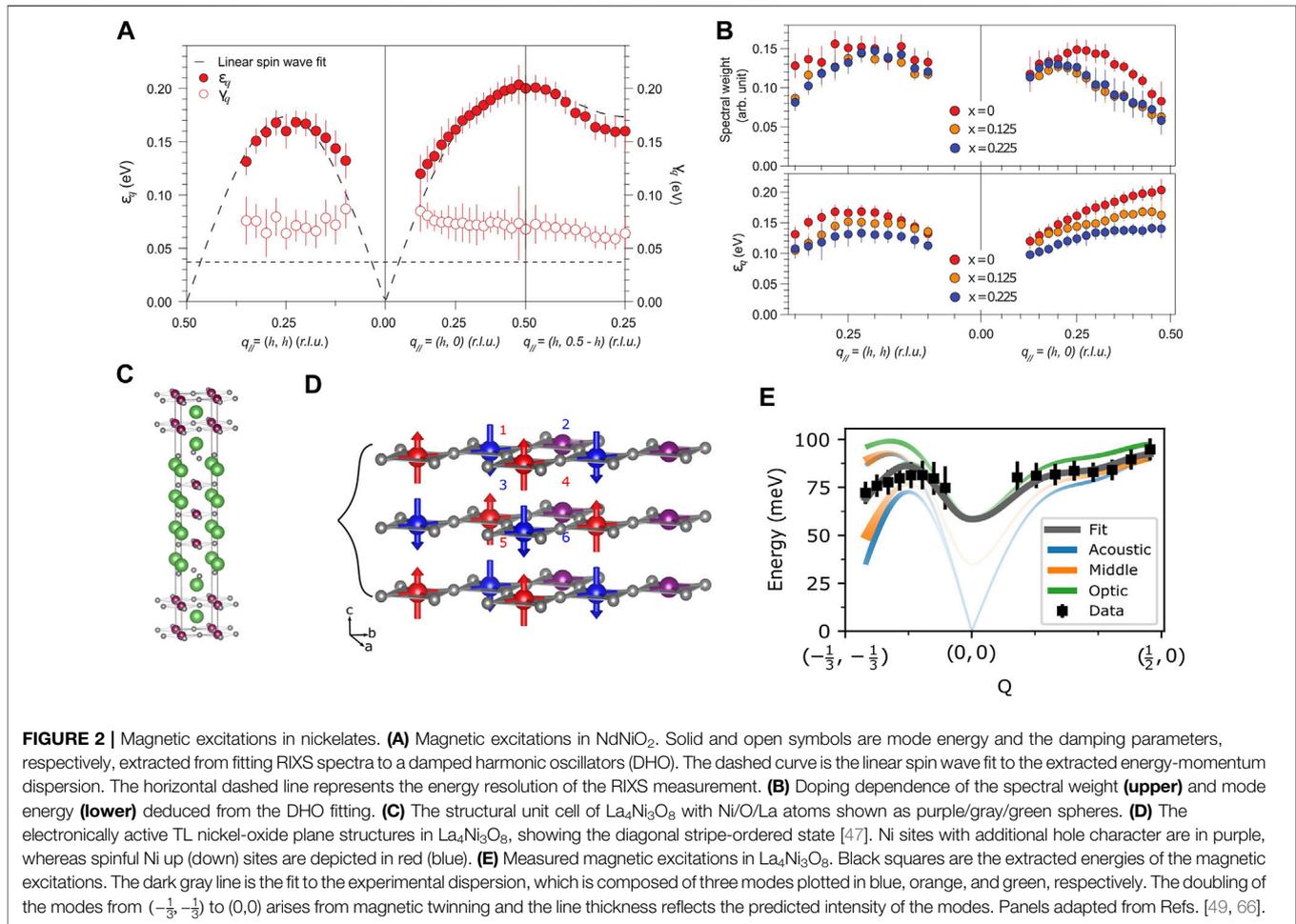


lower-energy peak (**Figure 1C**) that is associated with transitions into the doped hole levels [70, 71]. These hole-states constitute Zhang-Rice singlets (ZRS), which are plaquettes of two doped holes and four O atoms in square planar coordination around a Cu atom, playing the same role as a fully occupied or empty site in an effective single-band Hubbard model [72]. Notably, a recent scanning transmission electron microscopy electron-energy loss spectroscopy (STEM-EELS) study suggested that a similar ZRS peak emerges in IL nickelates upon hole-doping [65], although it carries significantly less spectral weight (**Figure 1C**). Overall, there is good evidence for reduced oxygen hybridization in nickelates compared to cuprates, which likely comes from an enhanced Δ value. Direct and quantitative determination of Δ and U represents an important issue for future soft x-ray studies.

Further insights into the electronic structure can be obtained from Ni *L*-edge XAS, corresponding to $2p$ - $3d$ multiplet transitions, reflecting the valence configuration of the Ni ions. In the cases of NiO and perovskite $RENiO_3$ ($2p^6 3d^8 - 2p^5 3d^9$ and $2p^6 3d^8 \underline{L}^n - 2p^5 3d^9 \underline{L}^n$ transitions, respectively) distinct multi-peak structures emerge across the *L*₃-edge (**Figure 1B**) [28, 64, 73]. Conversely, the line shapes of the IL nickelates LaNiO₂ and especially of NdNiO₂ (**Figure 1B**) resemble rather the single-peak XAS spectrum of IL cuprates with only one possible final

XAS state ($2p^6 3d^9 - 2p^5 3d^{10}$ transition). In more detail, the *L*₃-edge XAS of LaNiO₂ and NdNiO₂ is dominated by a main peak A (**Figure 1B**), while LaNiO₂ shows an additional minor low energy shoulder A' at slightly lower energies.¹ **Figure 1D** displays the RIXS intensity map of NdNiO₂ as a function of the incident photon energy and **Figure 1E** shows RIXS spectra for selected incident energies. Importantly, the RIXS spectra of LaNiO₂ and NdNiO₂ exhibit a distinct feature around 0.6 eV energy loss (**Figures 1D,E**), which is visible in the RIXS spectra with incident energies coinciding with the XAS peak A (A'). Furthermore, this feature emerges exclusively in the IL compounds and not the perovskite nickelate LaNiO₃ (**Figure 1E**). Using exact diagonalization, the general XAS and RIXS features can be reproduced (**Figure 1F**) and the 0.6 eV feature can be assigned to the hybridization between the Ni

¹The A' feature in XAS is only visible in LaNiO₂ films without a SrTiO₃ (STO) capping layer. Based on our recent measurements on LaNiO₂ films with a STO capping layer, the A' feature, which arises from the resonance of the ~ 0.6 eV feature in the RIXS map, coincides with the main XAS peak and becomes invisible. In other words, the XAS of La- and Nd-based infinite layer nickelates (with a STO capping layer) are essentially the same



$3d_{x^2-y^2}$ and La $5d$ orbitals, involving a charge-transfer from Ni to the RE cation [64]. From LDA + U it is found that a Fermi pocket of mainly La $5d$ character forms near the Γ point, which is quite extended and three-dimensional. On the other hand, the Ni $3d_{x^2-y^2}$ states in the NiO₂ planes are quasi-2D and strongly correlated [64, 74]. Nevertheless, the relevance of the rare-earth $5d$ bands for the low-energy physics of IL nickelates is still under debate and proposals range from effective single-band to multi-orbital models, including various Ni $3d$ and rare-earth $5d$ as well as interstitial orbitals [62, 63, 75–89]. Hence, future experiments probing the Fermi surface topology, such as angle resolved photoemission (ARPES) and quantum oscillation measurements, are highly desirable.

3 MAGNETIC CORRELATIONS

3.1 Magnetic Excitations in Infinite-Layer Nickelates

Despite the involvement of rare-earth $5d$ states, the fact that the electronic structures of the Ni $3d$ states resemble a cuprate-like $3d^9$ system raises a curious question: whether the Mott-physics, a key ingredient in the cuprate phenomenology [90], also play an

important role in sculpting the electronic structures in IL nickelates. Since a strong AFM interaction is a consequence of Mott physics due to strong onsite Coulomb interaction, information about the magnetic structures in IL nickelate is imperative to gain further insight into this issue. Early investigations of bulk polycrystalline LaNiO₂ and NdNiO₂ found no evidence of AFM order [59, 60], which appeared to suggest a significantly weaker magnetic interaction than in cuprates. On a different ground, theories have been debating the energy scale of magnetic interactions in the IL nickelates. Some theories predict a small AFM interaction (\sim an order of magnitude smaller than that of cuprates) because of the larger charge transfer energy Δ [91–94]. Conversely, other theories argue that the magnetic interactions are comparable to those in cuprates [74, 92, 95]. Experimental information about magnetic excitations is crucial to clarify this important issue.

Recently, magnetic excitations in Nd_{1-x}Sr_xNiO₂ have been revealed using RIXS at the Ni L_3 -edge [66]. As shown in **Figure 2A**, a branch of dispersive magnetic excitations has been observed in NdNiO₂, whose energy-momentum dispersion resembles the spin wave excitations of AFM coupled spins in a square lattice. Importantly, the bandwidth of the magnetic excitations is approximately 200 meV,

corresponding to a nearest neighbor spin interaction $J_1 \sim 65$ meV. This is about half of the J_1 in cuprate superconductors and similar to that in the TL nickelates [49], but is notably higher than in the stripe-ordered single-layer ($n = 1$) nickelates [25–27], perovskite nickelates ($n = \infty$) [96], and cubic NiO [97, 98]. Therefore, the observation of the high energy scale of J_1 in IL nickelates confirms the presence of a strong onsite Coulomb interaction, indicating that the strong correlation effect associated with the Mott-physics is likely also at play in the nickelate superconductors. Notably, distinct from the sharp magnetic modes observed in undoped cuprates, the magnetic excitations in the undoped parent compound of IL nickelates are damped, which is likely due to the coupling to the metallic Nd $5d$ states.

Upon hole doping, the magnetic excitations become less dispersive as a function of momentum and significantly damped. By fitting the spectrum to a damped harmonic oscillator function, it is found that mode energies soften accompanied by slightly reduced spectral weight (Figure 2B). The observed doping dependence is consistent with spin dilution in a Mott insulator. This is in fact different from those observed in cuprates, in which the mode energy and spectral weight do not decrease with increasing doping [99]. The doping dependence of the magnetic excitations in cuprates has been attributed to the longer-range charge dynamics emergent with increasing hole doping, for example, the three site terms in a Hubbard model [100, 101]. Such dynamics appear to be less prominent in the doped IL nickelates, likely due to the larger charge transfer energy Δ and the presence of the rare-earth $5d$ metallic state, calling for further investigation.

We note the next nearest-neighbour exchange interaction J_2 extracted from the magnetic excitations dispersion possesses an opposite sign to the nearest neighbor J_1 , which should favor the formation of AFM ordering at (0.5, 0.5). Unfortunately, RIXS at the Ni L_3 -edge cannot reach (0.5, 0.5) due to insufficient momentum transfer of the photons, preventing a direct scrutinization on the putative AFM order. Notably, recent susceptibility measurement on bulk powder samples indicated spin glass behaviors, but signatures of an AFM phase transition were still not observed. Thus, it would be interesting to investigate why IL nickelates are a failed AFM. However, one should be cautious about the difference between bulk and thin film samples, as well as the disorders in both types of materials, which were significantly reduced over time along with the optimization of material synthesis protocols.

Interestingly, the IL nickelates add one more case in which the magnetic correlations are in proximity to superconductivity in the phase diagram, similarly to a number of unconventional superconductors, such as cuprates, iron-based superconductors, and heavy fermion superconductors [102]. It might be tempting to attribute magnetic fluctuations as a candidate mechanism of superconductivity. However, among these superconducting compounds, including the nickelate superconductors, there appears no clear correlation between the energy scale of the magnetic excitation and the superconducting transition temperature, casting doubt on this notion. In any case, the relationship between magnetic fluctuations and the

superconductivity remains an important issue in nickelate superconductors.

3.2 Magnetic Excitations in Trilayer Nickelates

In parallel with the measurement of magnetic excitations in the IL material $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$, magnetic excitations were also measured in the TL materials $\text{La}_4\text{Ni}_3\text{O}_8$ and $\text{Pr}_4\text{Ni}_3\text{O}_8$ [49]. The crystal structure of $\text{La}_4\text{Ni}_3\text{O}_8$ is shown in Figure 2C. This material has some features that indicate that it might be especially promising as a cuprate analog. The rock salt RE-O layers present in its structure make it more two-dimensional than IL compounds and the rare-earth orbitals that are populated in IL are predicted to have a less significant role in TL systems [48, 103, 104]. As explained in the introduction, this compound is naturally self-doped and has a nominal hole concentration of 1/3. A disadvantage of the $\text{La}_4\text{Ni}_3\text{O}_8$ series is that they have, to date, proven difficult to chemically dope. Like cuprates, and some other complex oxides, $\text{La}_4\text{Ni}_3\text{O}_8$ has charge and spin order [47, 105]. This structure, illustrated in Figure 2D, features diagonal rows of Ni sites with enhanced hole character and neighboring diagonal stripes of up and down spin-ordered sites with reduced hole character. The overall magnetic dispersion, measured with Ni L_3 edge RIXS, is plotted in Figure 2E and features a bandwidth of ~ 80 meV with a downturn near $(-\frac{1}{3}, -\frac{1}{3})$, which is the charge and spin stripe-ordering wavevector. $\text{La}_4\text{Ni}_3\text{O}_8$ was modeled by solving a Heisenberg Hamiltonian which accounts for stripes, and assumes complete charge disproportionation into d^9 and d^8 sites, similar to prior studies of other stripe-ordered cuprates and nickelates [18, 106]. This includes J_1 , which connects sites 1 and 2 in Figure 2D, J_3 , which couples atom 2, through the purple doped site, to atom 1 in the next unit cell and J_z , which reflects interactions along the c -axis, for example, site 1 to site 3. The diagonal J_2 interaction that was included in the analysis of NdNiO_2 is not expected to be important here, as it couples to the sites with an enhanced hole character and which would be spinless when hosting an extra hole ($S = 0$) [48]. Solving this Hamiltonian in the spin-wave approximation yields three modes. Since these modes could not be resolved separately, the RIXS intensity of these modes was computed and summed to predict the intensity of the magnetic feature in RIXS. Values of $J_1 = 69(4)$ meV and $J_3 = 17(4)$ meV were obtained by fitting a Hamiltonian of this type.² Similar to the $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ case, the effects of J_z were too small to be constrained by the experiment, so the theoretical value of $J_z = 13.6$ meV was used. Very similar dispersions were found in $\text{Pr}_4\text{Ni}_3\text{O}_8$, which may imply that dynamical stripes exist in this compound even though long range stripe order has not been detected, as has been suggested independently in muon spin rotation studies [107].

The leading value of $J_1 = 69(4)$ meV in $\text{La}_4\text{Ni}_3\text{O}_8$ is strikingly close the 65(1) meV value obtained for NdNiO_2 . It should be noted that this similar value arises from a much smaller magnetic

²The notation used here was been modified from the original work of Ref. [49] to facilitate comparison with Ref. [66].

bandwidth, as within the stripe-ordered state, each Ni will have only two magnetic neighbors. An approximate extrapolation of the magnetic dispersion of $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ to a doping of $x \sim 1/3$ implies that it would have a bandwidth comparable to $\text{La}_4\text{Ni}_3\text{O}_8$ at this doping; yet, whether stripe order or fluctuations exist in the IL nickelates, like those found in the TL nickelates, remains an important open question. Overall, this suggests that the local correlated physics in these reduced RP cousins is very similar provided they are compared at the same effective doping, although their precise low-energy ground states might be more different.

4 CONCLUSION

In summary, soft x-ray spectroscopic studies have provided valuable insights into the physics of RP-phase and RP-derived nickelates. Nevertheless, for low-valence nickelates there is still limited consensus on the essential ingredients of their electronic structure. Along these lines, we anticipate that advances in sample synthesis and the application of complementary experimental techniques, including ARPES and quantum oscillation measurements, will be helpful. Moreover, the role of disorder and capping layers, as well as the apparent differences between film and bulk samples need further clarification. These insights could point the way towards improved low-valence nickelate superconductors, including multilayer systems [108]. Finally, a pertinent question is whether suitable sample preparation allows to realize other cuprate-typical ground states in nickelates, such as

antiferromagnetism, pseudogap, as well as nematic, charge, and spin orders.

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