

Softening of dd excitation in the resonant inelastic x-ray scattering spectra as a signature of Hund's coupling in nickelates

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We investigate the effects of Hund's coupling on the resonant x-ray absorption spectra of the recently discovered family of layered nickelate superconductors. We contrast two scenarios depending on the relative strength of the ratio of the effective Hund's coupling (J_H) to the crystal fields (Δ) in these systems. We carry out the cluster and DFT + DMFT simulations of the RIXS signal at the Ni L edge for different values of Hund's coupling. We find the latter dominates for the parent compound while the former becomes important for sufficiently large doping. Our results are consistent with the observations of a softening of a RIXS peak as a function of doping by Rossi *et al.* [Phys. Rev. B **104**, L220505 (2021)], only when the Hund coupling is sizable. To interpret the results, we separate the theoretical RIXS signal into spin conserving and nonspin conserving channels and conclude that the infinite layer nickelates are in a regime where Δ and J compete effectively and suggest further experimental tests of the theory.

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Introduction. A long-standing goal in condensed matter physics has been to discover a superconducting infinite-layer (IL) nickelate, as these compounds are analogous to the high-temperature two-dimensional (2D) superconducting cuprates [1], in which superconductivity was discovered in 1986 [2]. After a long search, superconductivity was discovered in the doped infinite layer nickelate, $\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2$ [3], with the differences in superconducting domes with the 2D cuprates [4,5]. Recently, superconductivity was also reported in $\text{La}_3\text{Ni}_2\text{O}_7$, suggesting it as a generic feature of nickelates [6]. This is a major breakthrough, as the electronic structure of the nickelates is notably different from the cuprates. Most notably, the nickel in a nickelate is understood to exist in electron configurations from d^7 to d^9 , unlike the copper in a cuprate superconductor, which is understood to exist only in the d^9 configuration [7,8]. Nickelates are also known to host metal-insulator transitions where the oxidation state of Ni plays a central role [8].

Superconductivity in doped 2D cuprates is understood to be driven by resonating valence bond states [9] and has d -wave pairing [10,11]. This is usually explored using the Emery model [12], which allows for Zhang-Rice singlets formed with $d_{x^2-y^2}$ orbitals [13] and the oxygens. The parent compound for IL nickelate is weakly insulating in contrast to the Mott insulator realized in cuprates. The possibility of mixed oxidation states in nickelates makes the problem challenging, as multiple d orbitals can play a significant role in the origin of the superconductivity. In particular, $3z^2$ have

been hypothesized to play a role in the ground state [14,15]. The model for nickelates ranges from single band to the multiorbitals of the Ni atom, and even the role of the rare-earth metal has been considered [15–26].

The Fermi surfaces of the rare earth metal have also been postulated to play a role in the IL nickelate NdNiO_2 [27–29], which complicates the understanding of the active degrees of freedom for superconductivity. The relevance of Hund's coupling has also been explored [29–32]. More recently, it has been proposed that since these compounds are created by reduction, the hydrogen atoms in these materials can play a significant role, further complicating the description [33]. Indeed, the origin of superconductivity is a controversial topic, and the proposals are at a very nascent stage.

Recently, resonant inelastic x-ray scattering (RIXS) [34–40] has also been extensively used to explore the relevant degrees of freedom in nickelates. It offers an advantage as it can amplify the response for smaller cross sections, such as small crystals and thin films, due to large light-matter interaction and inherent resonance conditions in the technique. The spectroscopic response allows one to constrain the possible theory for superconductivity in nickelates. For example, in a recent study [39] on low valence nickelates, $\text{La}_4\text{Ni}_3\text{O}_8$ and $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ revealed that the nickelates are of mixed charge-transfer–Mott–Hubbard character with the Coulomb repulsion U , in contrast to the charge-transfer nature of the cuprates in Zaanen–Sawatzky–Allen scheme. Recently, a softening of dd excitations in IL nickelate on doping was reported in Ni L -edge RIXS spectra [34]. We revisit the observation to investigate the relevance of Hund's coupling to the observation.

In this Letter, we explore the dd excitations in undoped and doped nickelates. In the undoped case, dd excitation is independent of J_H . In the doped case, both the crystal-field

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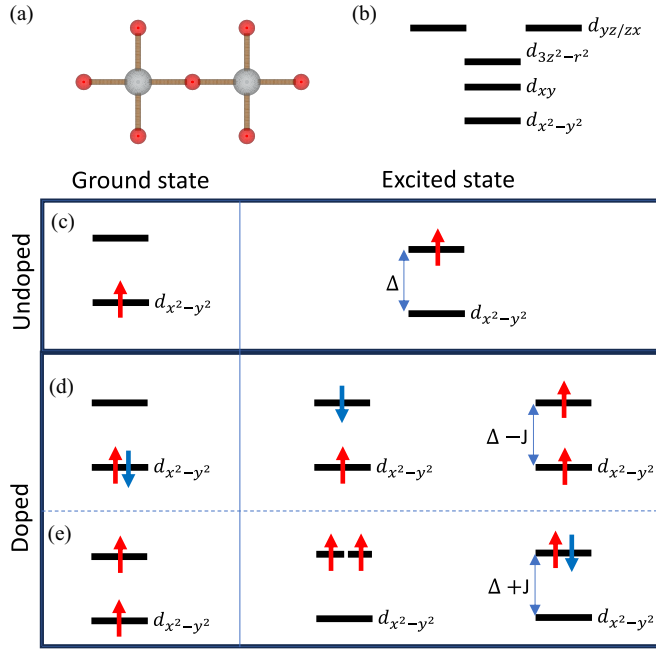


FIG. 1. Schematics of the cluster and the ground state occupancies in the cluster. (a) shows the Ni_2O_7 cluster and (b) shows its Ni 3d energy levels. (c)–(e) shows excitations for single ion configurations. (c) shows schematics for the ground and dd transition for the undoped model. (d) and (e) show the schematics for ground and the dd transitions for the doped model with two ground state configurations; (i) doubly occupied $d_{x^2-y^2}$ orbital for large Δ , and (ii) one hole each in $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ orbitals for large J_H , respectively. In the spin-flip ($\Delta S = 1$) channel, (d) and (e) will soften and harden dd excitations, respectively.

splitting of d orbitals (Δ) and J_H become important and J_H plays a central role in softening of dd excitations. Studying RIXS spectra using a small cluster and DFT + DMFT, we show that J_H plays a central role in softening dd excitations despite Δ dominating the ground state and report results consistent with the experimental softening of dd excitations on doping [34].

Model. We consider the Ni_2O_7 cluster shown in Fig. 1(a) with all the Ni 3d and O 2p orbitals, unless otherwise stated. The full Hamiltonian is given by $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_C^d + \mathcal{H}_C^p + \mathcal{H}_{pd}$ [39]. $\mathcal{H}_0$ is the noninteracting part. $\mathcal{H}_{pd} = V_{pd} \sum_{ij} n_i^d n_j^p$. V_{dp} is nearest neighbor coulomb interaction. The Coulomb interaction on O atoms is given by $\mathcal{H}_C^p = U_p \sum_{i,\mu} n_{i\mu\uparrow}^p n_{i\mu\downarrow}^p$. $\mathcal{H}_C^d$ is the Coulomb repulsion on the 3d of Ni which are accounted for by the multiorbital Hubbard-Kanamori model [41–43] given by

$$\begin{aligned} \mathcal{H}_C^d = & U_d \sum_{i,\alpha} n_{i\alpha\uparrow}^d n_{i\alpha\downarrow}^d + U'_d \sum_{i,\sigma,\sigma',\alpha<\beta} n_{i\alpha\sigma} n_{i\beta\sigma'} \\ & + J_H \sum_{i,\sigma,\sigma',\alpha<\beta} d_{i\alpha\sigma}^\dagger d_{i\beta\sigma'}^\dagger d_{i\alpha\sigma} d_{i\beta\sigma'} + J_P \sum_{i,\sigma,\sigma',\alpha<\beta} d_{i\alpha\sigma}^\dagger d_{i\alpha\sigma'}^\dagger d_{i\beta\sigma} d_{i\beta\sigma'}. \end{aligned}$$

Here, U is the intraband Hubbard repulsion, U' , J_H , and J_P are interband Hubbard repulsion, Hund's exchange interaction, and pair hopping amplitude, respectively.

The parameters for this model for modeling nickelate and cuprates have been extensively reported in the literature and notably varied [see Table I in the SM] [14,27,44–46]. The literature has a notably varied set of parameters, albeit all these studies agree that nickelates have large TM-ligand splitting and larger hopping overlap (t_{pd}) compared to cuprates. The relevant noninteracting parameters for this work are also shown in Table I of the SM [44]. For the interacting part, we use $U_d = 6.5$, $U_p = 4.1$, $V_{pd} = 1.0$, and $J_H = 0.8$ (unless otherwise stated) for the nickelate model.

For evaluating the RIXS spectra, we consider the core-hole Hamiltonian given by $H_{\text{ch}} = \mathcal{H} + U_{\text{ch}} \sum_i n_i^d n_i^p$ for the L edge [47,48]. The RIXS cross section is evaluated using the Kramers-Heisenberg formula [35,49,50] given by

$$\mathcal{I} = \sum_f \left| \sum_{i,n,\sigma} \frac{\langle f | \mathcal{D}^\dagger | n \rangle \langle n | \mathcal{D} | g \rangle}{E_n - E_g - \hbar\omega_{\text{in}} + i\Gamma_n} \right|^2 \delta(E_f - E_g - \Omega). \quad (1)$$

Here, $|g\rangle$, $|n\rangle$, and $|f\rangle$ are the ground, intermediate, and final states with energies E_g , E_n , and E_f , respectively and $\Omega (= \hbar\omega_{\text{in}} - \hbar\omega_{\text{out}})$ is the energy loss of the incoming photon. Γ is the inverse core-hole lifetime. $\mathcal{D}$ is the dipole operator given by $\mathcal{D} = \sum_{i,\alpha,\sigma,J} e^{i\mathbf{k}_i \cdot \mathbf{R}_i} p_{i,J}^\dagger d_{i,\alpha,\sigma}$ for the Ni L edge and involve core 2p (with spin-orbit coupling) and valence 3d shells of the nickel site. We evaluate spin-resolved Ni L -edge RIXS, where the angular dependence is integrated out, and the spectra are resolved into nonspin conserving (NSC, $\Delta S = 1$) and spin conserving (SC, $\Delta S = 0$) channels, depending on odd or even numbers of spin-flips in the excitations [37,48,51–53].

Results. We now present results for the nickelates using a combination of techniques.

Figure 1 shows the cluster considered in our work and schematics for the dd excitations. Panel (a) shows the Ni_2O_7 cluster considered later in our work, and panel (b) shows the energy Ni 3d onsite levels considered in our cluster. The Ni cluster with ligand can be mapped to energy levels for the single Ni site, integrating out the ligand (see Sec. S2 of the SM [44]). We start by considering the different scenarios of dd excitation for a single Ni ion (undoped d^9 , and doped d^8) case. We consider that the lowest energy orbital is $d_{x^2-y^2}$, and other d orbitals are roughly separated by a Δ energy gap in the hole language.

Panel (c) presents the schematics for dd excitations realized in undoped single-site nickelates, where dd excitations can be understood as simple excitations between $d_{x^2-y^2}$ to other d orbitals (Δ) as elucidated in the panel. In the doped case, there are two possible ground state configurations depending on the strength of J_H and Δ : (i) $\Delta > J_H$, doubly occupied $d_{x^2-y^2}$; or (ii) large $J_H > \Delta$, one hole each in $d_{x^2-y^2}$ and $d_{3z^2-r^2}$ (the preferential occupancy on this orbital is discussed later) as shown in panels (d) and (e), respectively. The NSC channel in (d) leads to the softening of dd excitations. This configuration is also consistent with the ground state occupancy estimates. On the other hand, (e) ground state will lead to hardening of the dd excitations. We use these schematic pictures as a guiding tool to understand RIXS spectra in the models below.

RIXS spectra. We present the simulated RIXS spectra for a set of models to decipher the effect of J_H . We use Ni_2O_7

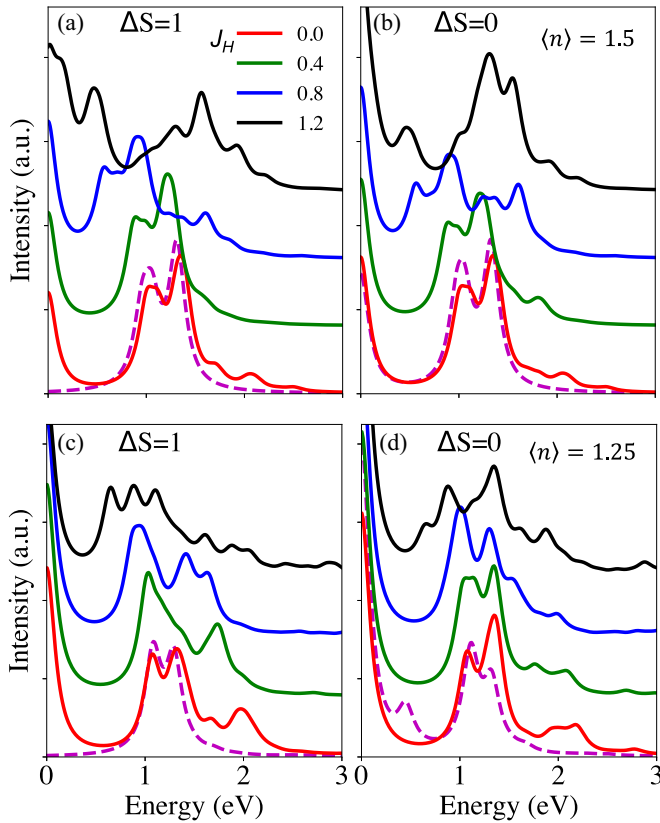


FIG. 2. Spin-resolved Ni L -edge RIXS spectra. (a) and (b) show results for Ni_2O_7 clusters in the NSC ($\Delta S = 1$) and SC ($\Delta S = 0$) channels. The dashed magenta line shows spectra for the undoped ($\langle n \rangle = 1$) model. Solid lines show spectra for the doped ($\langle n \rangle = 1.5$) model for the values of J_H mentioned in the legend. Similarly, (c) and (d) show results for Ni_4O_8 clusters in the NSC and SC channels, respectively, for undoped and doped ($\langle n \rangle = 1.25$) models.

clusters to explore the undoped and hole-doped (50%) models. We additionally present results for a smaller doping (25%) model using Ni_4O_8 clusters and an Anderson impurity model.

We start by examining the Ni L -edge RIXS spectra using Ni_2O_7 clusters with (1 $\uparrow$, 1 $\downarrow$) and three (2 $\uparrow$, 1 $\downarrow$) hole fillings to simulate the undoped and doped (50%) models in both the non-spin-conserving (NSC, $\Delta S = 1$) and spin-conserving (SC, $\Delta S = 0$) channels using KH formalism.

Panels (a) and (b) in Fig. 2 show NSC and SC channels of the Ni L edge for the Ni_2O_7 cluster. The dashed magenta line shows the RIXS spectra for the undoped model and is independent of J_H . The doped (50%) model results are also shown in both panels using solid lines for a set of the J_H . The prominent peaks for the doped model $J_H = 0$ occur at the same energies as in the undoped case. Additionally, we have new peaks extending up to 2 eV in the doped case. We notice a softening of the major peaks of dd excitation in both channels with the increase in J_H in both the NSC and SC channels. The new peaks in the doped case also soften with the increase in J_H for $J_H \leq 0.8$. We will further revisit these results from this cluster later to examine the nature of the various peaks in both the channels.

To examine results for the lower doping (25%), we consider a Ni_4O_8 cluster with (3 $\uparrow$, 2 $\downarrow$) filling and evaluate the

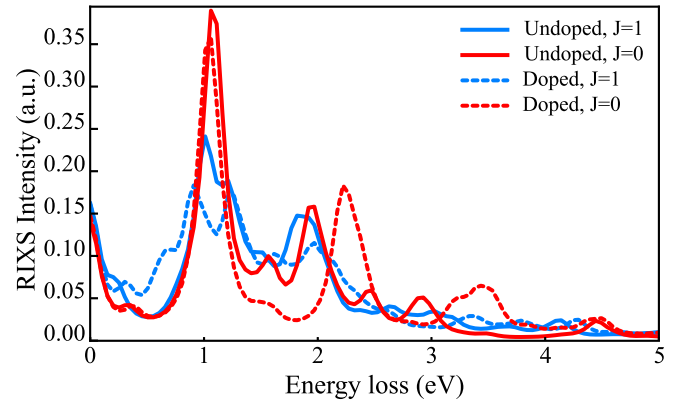


FIG. 3. RIXS spectra from the AIM for both the undoped and doped (0.25 hole doping) model. The results with and without J_H are shown in blue and red, respectively.

Ni L -edge RIXS spectra. Panels (c) and (d) case show results for both the undoped and doped models for this cluster. The dashed magenta line shows the RIXS spectra for the undoped model and is independent of J_H . In the SC channel, an additional bimagnon peak at around 0.3 eV [50,54,55] is observed for the undoped case. The large value of bimagnon can be attributed to the finite size of the cluster. The doped (25%) model results are shown using solid lines. The $J_H = 0$ case has the same characteristics as the undoped case. We observe softening of dd excitation in both cases with an increase in J_H . We, therefore, observed softening here for realistic doping.

These cluster results reveal the softening of dd excitations on doping consistent with reported RIXS experiments on NdNiO_2 [34] and more recently in trilayer nickelates where the dd excitations were reported at even lower energies [40]. This shows that Hund's coupling is important for understanding the softening of dd excitations observed in nickelates.

We now present the RIXS spectra using the Anderson impurity model (AIM) obtained from a charge self consistent LDA + DMFT calculation following Ref. [56]. We use tools from COMSUIE [57]: EDRIXS [36] for the RIXS calculations, LQSGW [58,59], and Portobello [60] for the LDA + DMFT calculations (also see S7 in the SM [44]). The DFT + DMFT calculations are conducted with $U = 5$ eV and $J_H = 1$ and 0 eV using the GGA functional [61] and a fully localized limit double counting with a nominal Ni- d^8 valence. Figure 3 shows the Ni L -edge RIXS spectra for the AIM. The undoped results are shown in solid lines using red and blue colors for $J_H = 0$ and $J_H = 1$, respectively. The dd excitations exist at 1–2.5 eV and are minimally affected by J_H for the undoped case. We also notice a feature at 0.5 eV, also reported in Ref. [27], but absent in the cluster calculation. The RIXS spectra for the doped model are shown in the dashed line in the respective colors. We find a significant softening of dd excitations for the $J_H = 1$ case compared to the undoped case and the $J_H = 0$ of the doped case. We overall find these results consistent with our small cluster calculations and, therefore, further confirms the crucial role of Hund's coupling in the softening of dd excitations observed in the experiment [34].

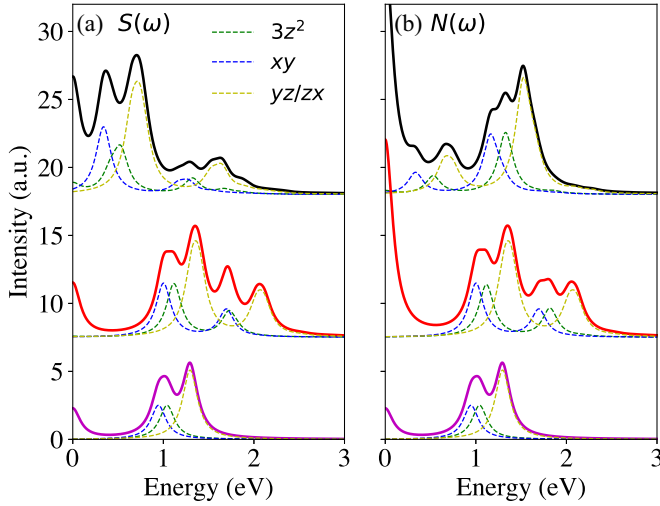


FIG. 4. $S(\omega)$ and $N(\omega)$ for the Ni_2O_7 cluster are shown in (a) and (b). Solid magenta shows the response for the undoped model. Dashed lines show the orbital-resolved excitations in the spectra. Solid red and black lines show the response for the doped model with $J_H = 0$ and 1, respectively.

The RIXS response given by Eq. (1) is complex; we, therefore, examine the model in the $\Gamma_n \rightarrow \infty$ limit, which eliminates the core-hole effects. We evaluate the dynamical spin [$S(\omega)$, $\Delta S = 1$] and charge [$N(\omega)$, $\Delta S = 0$] response on the Ni_2O_7 cluster. Figure 4 shows the $S(\omega)$ and $N(\omega)$ response in panels (a) and (b), respectively. The undoped model results are shown in magenta solid lines for $J_H = 1.2$ and the doped model in red and black lines for $J_H = 0$ and 1, respectively. We also plot the orbital resolved spectra where the outgoing dipole operator in Eq. (1) is restricted to a single orbital, which allows us to identify the different dd excitations. In both spectra, the ordering of orbital excitations with energy loss is as follows for the undoped model: d_{xy} , d_{3z^2} , and then $d_{yz/zx}$. This hierarchy of dd excitations differs from Ref. [34] which reports that $3z^2$ is the highest excitation in NdNiO_2 using the polarization dependence of the excitations. We re-examine the data (see S6 in SM [44]) and assign d_{3z^2} as lower energy compared to $d_{yz/zx}$ orbitals due to their similar angular dependence.

In the doped model, the results are sensitive to J_H . For $J_H = 0$, the orbital resolved dd excitations has primarily two peaks, with the bright lower energy peaks coinciding with the undoped case. For $J_H = 1$, we find that the prominent excitations soften in the $S(\omega)$ channel, consistent with the RIXS results. This contrasts with the $N(\omega)$, where mild hardening is observed, and the softening in the SC channel of RIXS can be described as a consequence of finite core-hole lifetime.

Finally, we compare our findings for Ni_4O_8 clusters with the RIXS experimental data from Ref. [34] in Fig. 5. Panel (a) shows the experimental RIXS data for $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ for undoped ($x = 0$) and with the Sr doped ($x = 0.225$) crystals, which is equivalent to hole doping in the material. The hole doping leads to a striking softening in the dd excitations. Panel (b) shows the results evaluated using ED for undoped ($n = 1.0$) and doped ($n = 1.25$) models using Ni_4O_8 clusters. In the case of undoped, the RIXS spectra is unaffected by

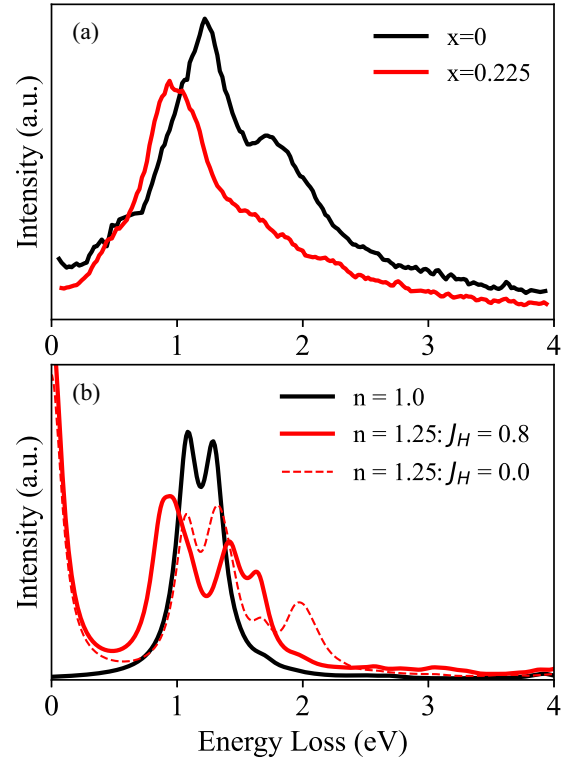


FIG. 5. Comparison of cluster calculation with the experiment data. (a) shows the experimental data extracted from the Ref. [34] for $\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2$ with doping (x) indicated in the panel. (b) shows the spectra evaluated using Ni_4O_8 cluster for the undoped ($n = 1.00$) and doped ($n = 1.25$) cases for $J_H = \{0.8, 0.0\}$.

J_H as we have only one hole per nickel site. In the case of the doped model, we see an interesting behavior, where the spectra hardens on doping for $J_H = 0$. In contrast, the spectra softens for $J_H = 0.8$ which is consistent with what is observed in the experiment, showcasing the crucial role of Hund's coupling in the system. There are some discrepancies though, such as the doped case having some additional peak above 1.5 eV but are significantly weaker than the other peak. The peak at zero energy loss is an elastic peak in the case of the doped model. Considering the simplicity of our model due to the limitation of computational limitation, the differences are nevertheless not unexpected. Nevertheless, the overall softening of dd excitations is quite clear. This is further supported by the schematic picture given in Fig. 1 and the softening in the AIM model in Fig. 3, pointing to a qualitative signature of Hund's coupling in softening the dd excitations, explaining the origin of their softening in the experiment on doping.

Figure 6 shows results for the orbital-resolved ground state occupancies dependence on J_H for Ni_2O_7 clusters. Panel (a) shows results for the undoped model. Here, the occupancies in Ni 3d orbitals (n_α) given by $n_\alpha = \langle g | \sum_\sigma n_{i,\alpha,\sigma} | g \rangle$ dependence on J_H is presented. We find that the holes are restricted to $d_{x^2-y^2}$ orbitals and the oxygen p orbitals. Panels (b)–(d) show results for the doped model. Panel (b) shows n_α dependence on J_H . For small J_H , the holes are restricted to the $d_{x^2-y^2}$ orbital and O 2p orbitals, the occupancy on d_{3z^2} increases with J_H till 1.25 eV, where it reaches around 20% of $d_{x^2-y^2}$. This is despite d_{xy} having a lower onsite energy. The dominant weight

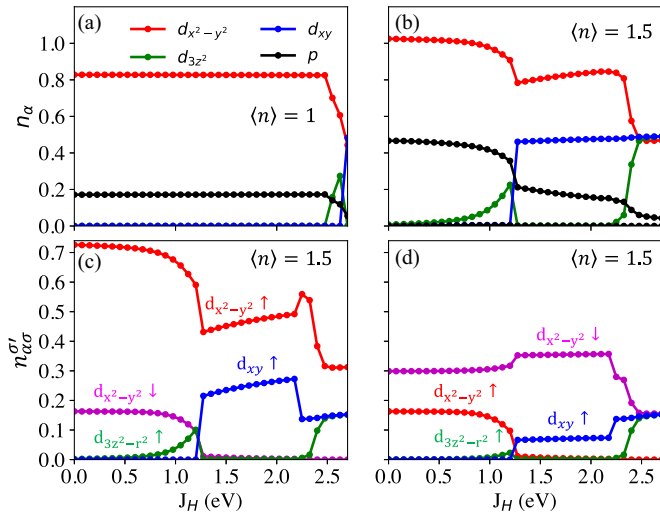


FIG. 6. Ground state analysis of the Ni_2O_7 cluster. (a) and (b) show the orbital resolved n_α dependence on J_H for undoped ($\langle n \rangle = 1$) and doped ($\langle n \rangle = 1.5$), respectively. (c) and (d) show the spin-resolved orbital double occupancies ($n_{\alpha\sigma}[n_{d_{x^2-y^2}\sigma'}]$) on the Ni site in the doped case for $\sigma' = \uparrow$ and $\downarrow$, respectively.

on d_{3z^2} can be attributed to the large hopping and the oxygen p orbitals shared between the two d orbitals (see Fig. S1 in the SM [44]). To investigate where the extra hole resides, we plot the constrained occupancies, $n_{\alpha\sigma}' = \langle g | n_{d_{\alpha\sigma}} [n_{d_{x^2-y^2}\sigma'}] | g \rangle$. The occupancy on $d_{x^2-y^2}$ with spin $\sigma' = \uparrow$ and $\downarrow$ is constrained

and are shown in panels (c) and (d), respectively. For small J_H , the $d_{x^2-y^2}$ has double occupancy, which is expected due to the penalty of the CF splitting. We notice an increase in occupancy on d_{3z^2} for $\sigma = \sigma'$ mediated by Hund's coupling till $J_H = 1.25$ eV. This makes the scenario discussed in Fig. 1(d) relevant for the nickelates, which leads to the softening of dd excitations.

Conclusions. In this work, we have explored the effects of Hund's coupling in undoped and doped nickelates using small cluster models and DMFT. Using these models, we find that Hund's coupling plays a central role in softening the dd excitations and softening is enhanced for larger doping. This softening of dd excitations with the increase in doping is consistent with the recent reported experimental study of IL nickelates [34]. Our small cluster analysis shows that doubly occupied $d_{x^2-y^2}$ is the preferred state for the doped nickelates, but the dd excitations are softened due to lowering of the dd excitations in the RIXS spectra. This contrasts with the cuprate, where the extra hole typically resides on the ligands [13].

Future experiments using photon polarization dependent RIXS allow for spin resolution [62], which can test the relative contributions of spin and charge fluctuations to the RIXS signal. Also, a comparative study of dd excitations of IL nickelates and cuprates can be a useful tool to highlight the difference between the two.

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