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Emergence of competing stripe phases in the undoped infinite-layer nickelate

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Recent discovery of superconductivity in the infinite-layer nickelates has ignited renewed theoretical and experimental interest in the role of electronic correlations in their properties. Here, using the first-principles simulations, we show that the parent compound of the nickelate family, $LaNiO_2$, hosts competing low-energy stripe phases, similar to the doped cuprates. The stripe states are shown to be driven by multi-orbital electronic mechanisms and Peierls distortions. Our study indicates that both strong correlations and electron-phonon coupling effects play a key role in the physics of infinite-layer nickelates, and sheds light on the microscopic origin of electronic inhomogeneity and the lack of long-range order in the nickelates.

Introduction. Emergence of symmetry-breaking, competing low-energy phases driven by the complex interplay of spin, charge, orbital, and lattice degrees of freedom is a universal characteristic of correlated systems [1–8]. Examples include the unconventional superconductors, such as the cuprates [4–7, 9–11], iron-based superconductors [12], and twisted bilayer graphene [13], where a wide variety of experiments have revealed the presence of competing inhomogeneous orders and a rich tapestry of phase diagrams. Although the mechanism of high-temperature superconductivity has remained elusive for many decades, it is generally accepted that competing orders play an important role in this phenomenon [1–3].

Recent discovery of superconductivity in the infinite-layer nickelates [14–18] has attracted intense attention [19–22] as an analog of cuprate superconductivity. Much experimental and theoretical effort has been devoted to understanding the relationship between the nickelates and cuprates [23–30]. Experiments have found strong hints of magnetism in the *undoped* infinite-layer nickelates [31–34], spin-glass behaviors [33, 34], and superconductivity [17, 18], similar to the case of the *doped* cuprates. However, these phenomena differ drastically from the *undoped* cuprates, which exhibit a robust antiferromagnetic (AFM) ground state. These similarities and differences between the cuprates and the nickelates have naturally raised many questions [19–21, 30]. For example, why is there no long-range magnetic order in the *undoped* nickelates even at low temperature [31, 34–37]? Do nickelates host competing orders similar to those found in other unconventional superconductors, such as the stripe and nematic phases? Answers to these questions will likely also provide new clues for understanding the mechanism of high-T_c superconductivity.

Here, via first-principles density functional theory (DFT) computations, we demonstrate that, much like the *doped* cuprates but in sharp contrast to the *undoped* cupates [38], the *undoped* infinite-layer nickelate hosts a rich landscape of competing low-energy stripe phases. By analyzing the charge redistribution and electronic structures, we find that the stripe phases are stabilized by a Peierls distortion and the opening of mini-gaps in the Ni $3d_{z^2}$ and $3d_{xz/yz}$ bands in directions perpendicular to the stripes, facilitated by the flat bands dominated by Ni $3d_{z^2}$ orbitals near the Fermi level that induce Van Hove singularities in the density of states (DOS).

Stripe Phases in $LaNiO_2$. Considerable effort has been devoted to studying the connection between inhomogeneous states and superconductivity since the discovery of low-energy stripe phases in cuprates [9–11]. To search for low-energy states in the *undoped* $LaNiO_2$, we have performed systematic calculations on 1D stripe phases as a function of charge periodicity. The computational details are given in Section I of the Supplementary Materials (SM) [39]. An ordered stripe phase results from the interplay between AFM interaction among the magnetic ions and the Coulomb interaction between the charges, where domains of approximately (π,π) AFM order are separated by charged antiphase boundaries. We begin by considering two exemplar stripe phases that are schematized in Fig. 1. We assume the stripe phases have the same ferromagnetic inter-layer exchange couplings as in the C-AFM phase [30]. We first investigate a bond-centered stripe with charge periodicity $P_c = 5$ and magnetic periodicity $P_m = 5$, see Fig. 1 (a). The calculated magnetic moments of Ni atoms are around $1.06\,\mu_B$ with small modulations of $\sim 0.02\,\mu_B$, leading to a slightly enhanced magnetic density around the antiphase boundaries (APBs). This phenomenon is discussed further in

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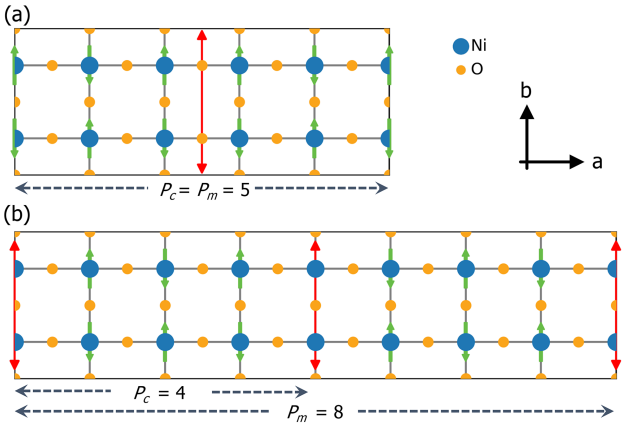


FIG. 1. **Schematics of two 1D stripe phases along the a-axis in $LaNiO_2$.** (a) A bond-centered stripe phase, where the antiphase boundaries (APBs) pass through the Ni–O–Ni bond, i.e., are centered on the bonding oxygen atoms. Here $P_c = P_m = 5$, where P_c and P_m denote the periodicities of the charge and spin orders, respectively. (b) A site-centered stripe phase with $P_m = 8$ and $P_c = 4$, where the APB passes through the Ni atoms. The blue and yellow circles represent Ni and O atoms, respectively. La atoms are omitted in the figure for clarity. The green arrows indicate the relative orientation of magnetic moments on Ni atoms, and the red lines with double arrows point to the APBs.

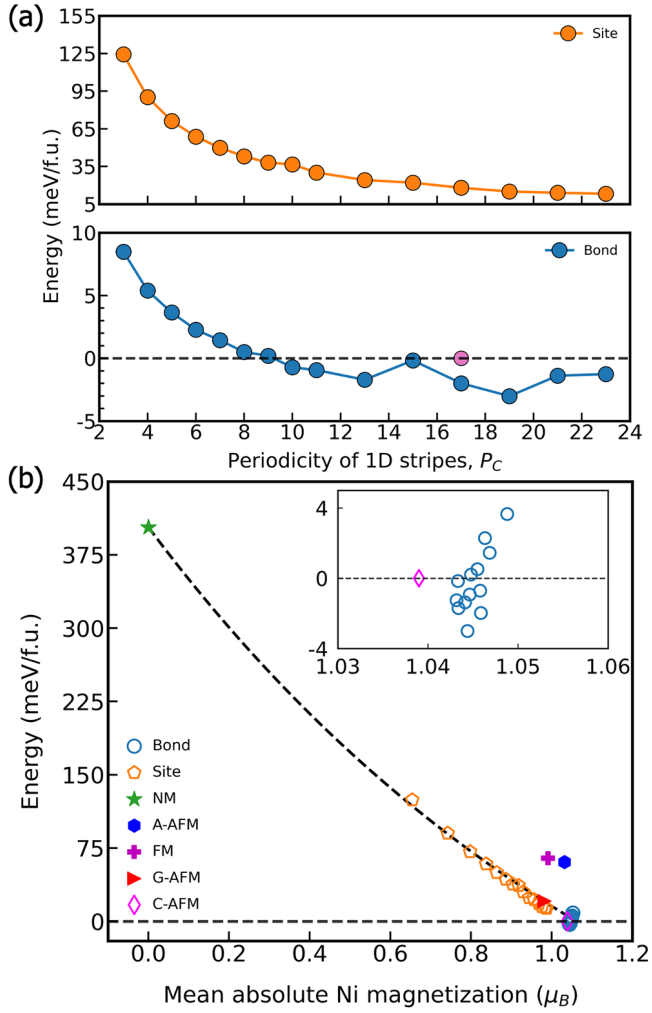


FIG. 2. **Stability of various computed states in $LaNiO_2$.** (a) Relative energies of several stripe phases with respect to the C-AFM state are plotted as a function of their periodicity. The purple circle in the bottom panel gives the total energy of the bond-centered stripe with $P_c = 17$ without atomic relaxation. (b) Energies of the computed states of $LaNiO_2$ relative to C-AFM as a function of the average magnetic moment of Ni atoms. The dashed black line is a guide to the eye. The insert is a blowup of the lower right corner of the plot. A similar plot was also obtained in $YBCO_7$, see Fig. 3 of Ref. [38].

the following section. Next, we study a site-centered stripe with charge and magnetic periodicities of $P_c = 4$ and $P_m = 8$, see Fig. 1 (b). Notably, we find the self-consistent magnetic moment of each Ni atom along the APBs to be $0.00\,\mu_B$, whereas the Ni atoms away from the APB exhibit a $0.99\,\mu_B$ moment in this case. This difference between the bond- and site-centered cases persists more generally among the various stripes we have examined, which go up to P_c values of 23; computations for still larger values of P_c become impractical.

Figure 2 (a) shows the total energies of various site- and bond-centered stripes for *undoped* $LaNiO_2$ relative to the C-AFM phase [30]. The bond-centered stripe is seen to be more stable than the site-centered stripe. Moreover, the bond-centered stripes become more stable as the charge periodicity increases. Figure 2 (b) compares the total energies of various magnetic phases with net Ni magnetization. Here, we include all the 1D stripe phases up to $P_c = 23$, along with a variety of uniform magnetic phases that have been discussed previously [30]. The more stable magnetic phases generally carry a larger average Ni magnetic moment. Several bond-centered stripes with $P_c \geq 10$ are found to be more stable than C-AFM. Many phases with strong local magnetic moments are found to be nearly degenerate in energy, with energy differences of less than 4 meV. The bond-centered stripe with $P_c = 19$ yields the ground state. These results are in line with our recent study of the yttrium-based cuprates [38], which revealed that "intertwined orders" with larger magnetic moments are key for understanding the physics of correlated materials. Notably, the total energy of the $P_c = 17$ stripe without distortions is higher by 2.19 meV/Ni compared to the case where distortions are included, suggesting that *e-ph* coupling plays an important role in the nickelates: see SM for further discussion.

We emphasize that, like the doped cuprates, Fig. 2 clearly provides evidence for the existence of competing low-energy states in $LaNiO_2$. It should be noted that when energy differences are very small, the true ground state would fluctuate between these stripe states. This is consistent with the absence of long-range order in neu-

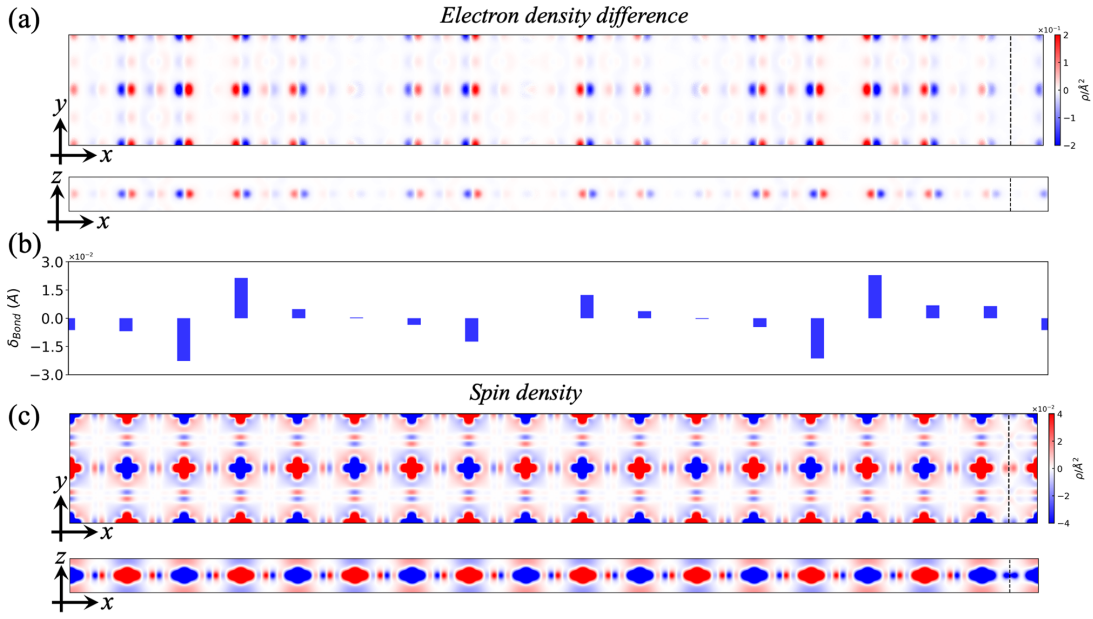


FIG. 3. **Analysis of charge and spin redistribution and lattice distortions.** (a) Top and side views of the projection of electron density difference between the stripe with $P_m = P_c = 17$ and the C-AFM phase on the xy-plane. Red (blue) colored regions indicate electron accumulation (depletion). (b) Bar plot of bond length differences for the right and left side Ni-O bonds for various Ni atoms. (c) Top and side views of spin density of the bond-centered stripe with $P_m = P_c = 17$; red (blue) color denotes spin-up (spin-down). The antiphase boundary is denoted by the black dashed line.

tron scattering experiments [35], the observation of local dispersing magnon excitations in RIXS [31], and with the large magnetic volume fraction seen at the lowest temperatures in muon spin-relaxation measurements [32], all suggestive of an intrinsically magnetic sample. Therefore, it is important to examine the properties of an array of low-energy stripes rather than focusing on just a single phase. In addition, we can rule out any significant role of the non-magnetic (NM) phase, which lies ∼400 meV above the C-AFM phase, since it is not energetically competitive.

Computational cost of first-principles DFT-based modeling becomes prohibitive for P_c values greater than 23. With this in mind, to explore the full manifold of possible low-energy stripe phases, we have carried out a random phase approximation (RPA) based analysis of the NM state, see Section 1 of SM. These results are in line with our DFT-based findings and show that the total energy can likely be further lowered considering stripes with large periodicities.

Charge Redistribution. To understand the stabilization of the stripe phases, we plot charge differences between selected bond-centered stripes and the C-AFM phase in Figs. 3 (a) and S2 (Section 2 of SM). For $P_c \leq 9$, strong charge redistribution is found around the APBs, which minimizes the effect of parallel neighboring magnetic configurations on the boundaries. However, for $P_c \geq 11$, the charges are deconfined from the APBs, redistributing largely in the magnetic domains, which leads to stripes with $P_c > 11$ (except $P_c = 15$) showing lower energies than the C-AFM phase. Taking $P_c = 17$ as an example [40], in Fig. 3 (a) we plot the electron charge density difference between the $P_c = 17$ stripe and the C-AFM phase which is stretched to have the same lattice constant. A strong charge polarization is seen along the x but not the z direction. The electron clouds around Ni and O atoms are polarized to point away from a depletion region centered halfway between the two neighboring antiphase boundaries. The accompanying Peierls distortion leads to a more stable stripe phase than C-AFM. Due to unequal atomic displacements, the Ni atoms are shifted away from the midpoint between the two neighboring O atoms, Fig. 3 (b), resulting in additional polarization effects.

Stripe related charge order has been experimentally observed in cuprates [41], and may be enhanced in a magnetic field [42]. However, the lattice distortion in Fig. 3 (b) and charge disproportionation in Fig. S3 are much smaller than in the case of the cuprates, suggesting that observing the charge orders in experiments in the nickelates will be challenging. While incommensurate charge density waves (CDWs) were reported in *undoped* nickelates [17, 43–45], recent experiments have suggested that these CDWs may not be an essential component of the nickelate ground state [46] but rather an effect associated with apical oxygen ordering [47], see SM for a discussion of the competing CDW orders.

Fig. 3 (c) shows the spin density of the bond-centered stripe with $P_c = 17$. From the shape of the Ni orbitals, one can see that the local magnetic moments on Ni are mainly composed of Ni $3d_{x^2-y^2}$ and $3d_{z^2}$ orbitals. O atoms in the magnetic domains show small spin polariza-

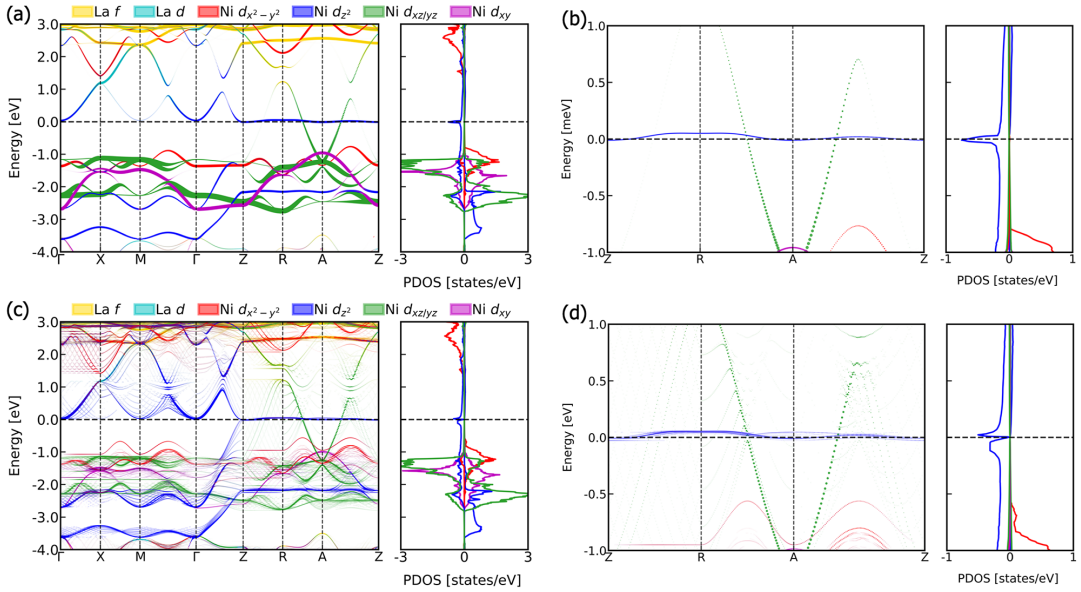


FIG. 4. **Comparison of electronic structures of the C-AFM and bond-centered stripe phases.** (a) Orbitally projected electronic structures of the C-AFM phase. The site-projected, orbitally resolved partial densities of states are for Ni sites with positive magnetic moments. Positive (negative) values indicate spin-up (spin-down) DOS. (b) Blowup of (a) over the energy window of -1 eV to 1 eV. (c) Same as (a), except that this figure refers to the bond-centered stripe with $P_c = P_m = 17$. (d) Blowup of (c) over an energy window of -1 eV to 1 eV. Band structures in (a) and (c) are unfolded onto the primitive $1 \times 1 \times 1$ Brillouin zone, where the width of the bands is proportional to their spectral weight.

tion, while O atoms around the APBs show a dumbbell-shaped spin density with a small magnetic moment of $0.037\mu_B$. These results strongly indicate that nickelates are not a charge-transfer system like the cuprates [38].

Electronic Structures. To gain insight into the electronic properties of the ground state, we compare the electronic band structures of the $P_c = P_m = 17$ bond-centered stripe and the uniform C-AFM phase. Figure 4 presents the unfolded electronic band structures of $LaNiO_2$ in the C-AFM and $P_c = P_m = 17$ bond-centered stripe phases. As pointed out in Ref. [30], the magnetic state appears as a Hund's phase, where the magnetic moments primarily consist of $d_{x^2-y^2}$ character ($0.75\mu_B$), with a smaller admixture of d_{z^2} ($0.25\mu_B$). This is also reflected in the band structure. The Ni-$d_{x^2-y^2}$ band (red lines) is fully gapped out, consistent with recent experiments [44], while the d_{z^2} is spin-split (blue lines), similar to the cuprates [48]. Due to its fractional filling, the upper Ni-d_{z^2} band is pinned to the Fermi level producing a flat band in the $k_z = \pi$ plane, consistent with previous studies [30, 49]. Flat bands dominated by Ni-d_{z^2} have also been predicted in multilayer nickelates using DFT+DMFT calculations [50]. We find similar results for the $P_c = P_m = 17$ stripe (0.78 μ_B from $d_{x^2-y^2}$, $0.26\mu_B$ from d_{z^2}). Remarkably, the pinning which arises from a strong reorganization of the d_{z^2} dispersion when a magnetic gap opens in the $d_{x^2-y^2}$ band, is absent in the NM phase. This flat band gives rise to a Van Hove singularity (VHS) in the DOS, which can enhance many-body interactions and drive more exotic correlated phenomena [51]. It has been suggested that this flat band could also couple electronic and lattice instabilities [30, 49]. Note that the VHS in $LaNiO_2$, which involves the Ni $3d_{z^2}$ orbital, is stronger than the $3d_{x^2-y^2}$-derived VHS in the cuprates. However, it is extremely rare to find a VHS exactly at the Fermi level, without driving a transition to a new phase.

Upon unfolding the unit cell for the $P_c = P_m = 17$ stripe, we find that the resulting band dispersions [Figs. 4(c)] are only slightly modified compared to the C-AFM phase. The degeneracy of bands in the stripe phases gets lowered due to the reduced symmetry. Despite this symmetry-lowering, the $3d_{z^2}$ flat-band remains pinned to the Fermi level, which is also the case in other stripes, see Section 5 of SM. Away from the Fermi level, minigaps form in the Ni-$3d_{z^2}$/La-d hybridized band due to the supermodulation of the crystal potential, typical of stripe-like structures [52]. This is also consistent with the case of the cuprates, where the stripes can be viewed as topological defects in an underlying AFM order. Figure 4(d) makes it clear that the stripe stabilization energy is associated with the minigaps induced by lattice distortions in the $3d_{z^2}$ and $3d_{xz/yz}$ bands, which open minigaps in the $3d_{z^2}$ and a small gap in the $3d_{xz/yz}$ band. These results suggest that nickelates are bad metals, like the cuprates. Several experimental studies [37] also report weakly insulating behavior in the nickelates.

$LaNiO_2$ differs from the insulating cuprate parent compounds in that $LaNiO_2$ is a metal with localized Ni $3d_{z^2}$, itinerant $3d_{xz/yz}$, and La $5d$ hybridized bands, so that

a multiorbital model is necessary to properly describe valence fluctuations between Ni^{2+} (d^8) and Ni^{1+} (d^9). We also note that a variety of multi-bands models have been proposed in this connection [23, 53–57]. Behavior of the magnetic state is similar to the cuprates, except for a somewhat larger energy scale in that the magnetic gap in the Ni-$d_{x^2-y^2}$ band is approximately double that in the cuprates owing to the larger magnetic moments. Concomitantly, the energy scale of magnetic and stripe states in Fig. 2(b) is a factor of 5-7 larger than in $YBCO_7$ [38].

Discussion and Conclusion. Our study supports the experimental observation of magnetic fluctuations in nickelates [17, 31, 34–37, 43, 44, 58, 59]. We have identified nearly-degenerate orders of bond-centered stripes and C-AFM phases, along with the presence of low-energy flat bands around the Fermi energy. This underscores the importance of treating orbitals, spins, charges, and lattice interactions equally when studying complex materials [60]. Competing spin and charge orders are suggested by our RPA calculations, which could yield intertwined orders in pristine $LaNiO_2$. Spin fluctuations associated with such nearly degenerate states could play a significant role in driving superconductivity [1–3]. A doping-dependent study is needed to fully elucidate this connection.

Despite significant differences between the cuprates and nickelates, there is a great deal of similarity between the results of Fig. 2 and those of Figs. 2 and 3 in Ref. [38] for $YBCO_7$. We suggest that this similarity is a signature of strong correlations being at play in correlated materials that are realized as a form of short-range order in which topological defects–here domain walls–play a significant role. This suggests that the nickelates may be closer to the (weak coupling) Slater regime [61], where Fermi surface nesting can lower the stripe energy. The enhanced stripe-AFM competition may explain why $LaNiO_2$, like $YBCO_7$, has no long-range magnetic order [14, 34]. Indeed, the similarity of Fig. 2(b) here and Fig. 3 of Ref. [38], with both figures displaying an accumulation point of phases near the ground state is striking, and it provides a plausible basis for the existence of intertwined orders found in many correlated materials.

Our study shows that the mechanism driving the formation of competing stripe phases in the pristine infinite-layer nickelates is the presence of strong correlations and Peierls instabilities involving several Ni d-orbitals, a situation that is quite distinct from the case of the cuprates. Our analysis points to the key role of electron-phonon coupling effects in stabilizing the stripe phases, in addition to the electronic mechanisms [17]. However, the lattice distortions and charge disproportionation in the stripe phases of nickelates are much smaller than those in the cuprates [38], which would make experimental observation of CDWs in nickelates quite challenging [17, 43–47]. The Ni $3d_{z^2}$ orbital is found to lie close to the Fermi energy, which could play a role in superconductivity. Overall, a multi-orbital minimal model is needed to capture the rich physics of the nickelates.

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