Magnetic and *f*-electron effects in LaNiO₂ and NdNiO₂ nickelates with cuprate-like $3d_{x^2-y^2}$ band

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Recent discovery of superconductivity in the doped infinite-layer nickelates has renewed interest in understanding the nature of high-temperature superconductivity more generally. The low-energy electronic structure of the parent compound NdNiO₂, the role of electronic correlations in driving superconductivity, and the possible relationship between the cuprates and the nickelates are still open questions. Here, by comparing LaNiO₂ and NdNiO₂ systematically within a parameter-free, all-electron first-principles density-functional theory framework, we reveal the role of Nd 4*f* electrons in shaping the ground state of pristine NdNiO₂. Strong similarities are found between the electronic structures of LaNiO₂ and NdNiO₂, except for the effects of the 4*f* electrons. Hybridization between the Nd 4*f* and Ni 3*d* orbitals is shown to significantly modify the Fermi surfaces of various magnetic states. In contrast, the competition between the magnetically ordered phases depends mainly on the gaps in the Ni $3d_{x^2-y^2}$ band. Our estimated value of the on-site Hubbard *U* in the nickelates is similar to that in the cuprates, but the value of the Hund's coupling *J_H* is found to be sensitive to the Nd magnetic moment. In contrast with the cuprates, NdNiO₂ presents 3D magnetism with competing antiferromagnetic and (interlayer) ferromagnetic exchange, which may explain why the *T_c* is lower in the nickelates.

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Since the discovery of high- T_c superconductivity (HTSC) in the lanthanum-based cuprates in 1986¹, understanding the mechanism of HTSC has drawn intense interest^{2–6}. Despite vigorous efforts, however, many questions remain unanswered and a clear consensus on the underlying mechanism of HTSC has remained elusive. A promising route in this connection is to find superconducting analogs of the cuprates, which could provide new clues to the origin of HTSC. One such materials family is the perovskite nickel oxides. Here the infinite-layer NdNiO_2 compound holds great promise since it exhibits an intrinsic $3d^9$ filling much like the cuprates, although challenges of crystal growth have presented problems for undertaking systematic investigations.

Superconductivity in the hole-doped infinite-layer nickelate NdNiO_2 at 9–15K has been reported in thin-film samples grown on SrTiO_3 ^{7–10}, although superconductivity in bulk NdNiO_2 has not been observed¹¹. These results⁷ have reinvigorated interest in searching for the microscopic mechanism of HTSC and stimulated many new questions^{10–20}. Notably, superconductivity is present both in the hole-doped NdNiO_2 ⁷ and PrNiO_2 ²¹, but it is absent in LaNiO_2 ⁷. This suggests that the Nd (Pr) f -electrons are not merely spectators, but possibly participate in the emergence of superconductivity. Interestingly, initial reports showed metallic behavior in pristine LaNiO_2 and NdNiO_2 with no sign of long-range magnetic order persisting down to low temperatures^{11,22,23}, calling into question the role of Mott physics in driving superconductivity. However, two recent transport studies^{8,10} report the presence of a weak insulating phase in pristine NdNiO_2 , which could partly be a signature of short-range magnetic fluctuations due to the intrinsic off-stoichiometry produced by the inhomogeneous oxygen deintercalation crystal-growth process⁷.

In this connection, a variety of theoretical studies have been performed to understand the low-energy physics of the nickelates employing density functional theory (DFT)^{14,18,24–26} and “beyond” DFT methods such as DFT+U^{18,27,28}, quasiparticle GW²⁹, dynamical mean-field theory (DMFT)^{27,30–32}, and model Hamiltonians^{15,17,27,33} that have been constructed to understand the low-energy physics. The bulk of these studies focus on the NiO_2 plane and differences in quantities such as the $d-p$ orbital splitting in comparison to the cuprates¹⁸. However, focusing on the NiO_2 plane neglects the effects of the f electrons on the electronic and magnetic structure, despite the presence of superconductivity in the Nd-based and Pr-based compounds but not in the La compounds^{7,21}. An active role of the f electrons is also suggested by a Kondo-like logarithmic temperature dependence of the resistivity and Hall coefficient at low temperatures⁷, and other recent experiments demonstrating strong similarities between the infinite-layer nickelates and the rare-earth intermetallics³⁴, although a recent study attributes this strange behavior to the Nd $5d$ orbitals¹⁵. A few electronic structure studies utilizing the DFT+U¹⁶ scheme or Heyd–Scuseria–Ernzerhof (HSE) hybrid functional²⁵ have considered the f electrons and found significant hybridization between the Nd $4f$ and Ni $3d$ orbitals near the chemical potential along with a possible ferromagnetic order. But, these calculations neglect the effects of spin–orbit coupling (SOC) crucial for capturing the correct f -band splittings, and required the introduction and fine tuning of external ad hoc parameters such as the Hubbard U and the exact-exchange admixture, limiting their predictive power³⁵.

In this article, we present a systematic in-depth study of the electronic and magnetic structures of both LaNiO_2 and NdNiO_2 using the strongly-constrained-and-appropriately-normed (SCAN) density functional³⁶ with spin–orbit coupling to examine the effects of f electron physics. The SCAN functional has proven accurate for modeling many correlated materials families

including the cuprates^{37–41}, iridates⁴², and the ABO_3 materials⁴³. In particular, SCAN accurately predicts the f -band splitting in SmB_6 in good accord with the corresponding experimental values⁴⁴. We consider a number of magnetic phases, whose energy and ordering are found to be quite similar for LaNiO_2 and NdNiO_2 with sensitivity to the opening of magnetic gaps in the Ni $3d_{x^2-y^2}$ band. The dispersion of this band is quite similar to that of the corresponding band in the cuprates, as is the order of the resulting magnetic phases where the antiferromagnetic state has the lowest energy. In line with this, the estimated values of the Hubbard U are close to those commonly found in the cuprates, while Hund’s coupling J_H varies for different Nd magnetic sublattices. However, unlike the cuprates³⁷, the strength of the interlayer magnetic exchange coupling in the nickelates is comparable to that of the intralayer exchange with a strong antiferromagnetic component. Lastly, the $4f$ electrons play an important role in modifying the Fermi surfaces in the nickelates, where we find the charge transfer energy between the Ni $3d$ and O $2p$ orbitals to be large and do not change much with magnetic order, indicating that the physics of the nickelates is distinct from that of the cuprates.

Results and discussion

Crystal and magnetic structures. Figure 1 shows the crystal structure of LaNiO_2 and NdNiO_2 in the $P4/mmm$ symmetry²², where NiO_2 planes are sandwiched together with La or Nd spacer layers. In the NiO_2 planes the Ni sites are surrounded by four O atoms in square-planar coordination. The primitive cells for the non-magnetic (NM) and ferromagnetic (FM) phases are modeled by the primitive cell (Figs. 1a, b), respectively. Since unconventional superconductivity is often found accompanying an antiferromagnetic (AFM) phase transition, we also consider several AFM phases. In NdNiO_2 , in particular, we find that the Nd and Ni sublattices can both support AFM spin orderings. Specifically, we use a $\sqrt{2} \times \sqrt{2} \times 1$, $\sqrt{2} \times \sqrt{2} \times 2$, and $1 \times 1 \times 2$ supercell for the C-type AFM (C-AFM), G-type AFM (G-AFM), and A-type AFM (A-AFM) orders, respectively, as shown in Figs. 1c–e. In the C-AFM phase, the intralayer coupling in both the Nd and Ni layers is AFM, whereas the interlayer coupling is FM. In the G-AFM phase both the intralayer and interlayer couplings are AFM. In contrast, the A-AFM phase displays an intralayer FM coupling with an AFM interlayer coupling for both the Nd and Ni sublattice.

Table 1 presents our theoretically predicted total energies, lattice constants, and spin magnetic moments for various magnetic phases of LaNiO_2 and NdNiO_2 . For LaNiO_2 , the C-AFM phase is the most stable, with the G-AFM, A-AFM, and NM phases lying at higher energies. The optimized lattice parameters for the magnetic phases are in good accord with the corresponding experimental values, while this is not the case for the lattice parameters obtained for the NM phase. Moreover, the lattice constant and the energy of the FM (C-AFM) phase are almost the same as that of the A-AFM (G-AFM) phase, indicating that the interlayer coupling in LaNiO_2 is very weak. Finally, our predicted local nickel magnetic moment is $\sim 1.0 \mu_B$ irrespective of the magnetic configuration.

Compared to our results, we note that GGA+U^{18,45} ($U = 3$) predicts a much smaller energy separation between the magnetic configurations along with reduced magnetic moments of $\sim 0.7 \mu_B$ and $\sim 0.5 \mu_B$ in the C-AFM and FM phases, respectively. The aforementioned reduced moments could be due to the neglect of SOC effects in the GGA+U calculations, but this is surprising since a significant Hubbard U was introduced on the Ni sites.

We now compare and contrast LaNiO_2 and NdNiO_2 with reference to Table 1. For both compounds, the C-AFM configuration is seen to be the most stable one. Energy of various

Fig. 1 Various crystal structures considered are shown. **a** Non-magnetic (NM), **b** ferromagnetic (FM). Three magnetic exchange couplings in the Ni sublattice are: J_1 (in-plane nearest-neighbor), J_2 (out-of-plane nearest-neighbor) and J_3 (out-of-plane next nearest-neighbor). **c** C-type antiferromagnetic (C-AFM), **d** A-type antiferromagnetic (A-AFM). **e** G-type antiferromagnetic (G-AFM) phase. The light blue, green, and red balls represent La (Nd), Ni, and O atoms, respectively. The light blue and green arrows denote different magnetic moment directions. Note that there is no local magnetic moment on La.

Table 1 Comparison of theoretically predicted properties of various magnetic phases of LaNiO ₂ and NdNiO ₂ .						
Phases	LaNiO ₂			Energy (meV/f.u.)	M _{Ni}	
	Lattice constant (Å)				(μB)	(μB)
	a	b	c			
NM	3.902	3.902	3.392	+400	0	
C-AFM	3.949	3.949	3.346	0	1.05	
G-AFM	3.947	3.947	3.345	+22	0.96	
FM	3.943	3.943	3.366	+62	0.98	
A-AFM	3.941	3.941	3.359	+64	1.00	
Exp. ⁶⁸	3.959	3.959	3.375	-	-	

Phases	NdNiO ₂			Energy (meV/f.u.)	M _{Nd} (μB)	M _{Ni} (μB)
	Lattice constant (Å)					
	a	b	c			
NM	3.836	3.836	3.177	+3792	0	0
C-AFM	3.910	3.910	3.239	0	2.91	1.05
G-AFM	3.901	3.901	3.239	+64	2.89	0.97
FM	3.904	3.904	3.302	+145	3.01	1.01
A-AFM	3.871	3.871	3.312	+155	2.98	0.96
Exp. ²⁰	3.914	3.914	3.239	-	-	-

Lattice constants refer to the primitive unit cell. Energies are given relative to the C-type antiferromagnetic phase. Values of magnetic moments display small variations among different Nd and Ni sites, the results given are the average values. M_{Nd} and M_{Ni} denote the local magnetic moment per Nd and Ni atom, respectively.

magnetic phases are greater in NdNiO₂ compared to the corresponding values in LaNiO₂. The NM phase lies at a high energy of 3840 meV/f.u. in NdNiO₂ and 400 meV/f.u. in LaNiO₂. These results suggest that NdNiO₂ has a stronger magnetic exchange coupling than LaNiO₂, a point to which we will return below. Also, we find the Ni–Nd coupling to be weakly AFM, and the Nd–Nd magnetic couplings to be small with a negligible effect on total energy, so that non-collinear magnetic orders could form in NdNiO₂ [see Supplementary Note 1]. Notably, like LaNiO₂, our calculated lattice constants for NdNiO₂ in the C-AFM and G-AFM phases are very close to the experimental values, while this is not the case for the values obtained in the NM, FM, and A-AFM arrangements.

Our computed local magnetic moments on Nd and Ni are seen from Table 1 to be $\sim 3.0 \mu_B$ and $\sim 1.0 \mu_B$, respectively, suggesting that the corresponding electronic configurations of Nd and Ni are [Xe] 4f³ and 3d⁹. Various orbitals contribute to the total Ni magnetic moment as follows: 0.75 μ_B from $d_{x^2-y^2}$; 0.25 μ_B from d_{z^2} ; and, t_{2g} carries a negligible moment. A number of theoretical studies in the literature have reported magnetic ordering and the associated local moments^{16,28,45,46} with strong spin fluctuations possibly playing a key role in the pairing mechanism^{32,46–48}. Specifically, the GGA yields the local moment on Ni to be significantly smaller with values of ~ 0.52 and $\sim 0.35 \mu_B$ in the FM and AFM phases, respectively¹⁶. [The SCAN values are only recovered when a large Hubbard U of 8(5) eV for Nd(Ni) is used¹⁶.] A previous SCAN-based calculation²⁶ finds a reduced Ni magnetic moment of 0.76 μ_B , but this study neglected Nd 4f electrons and SOC effects. An LDA+ U ($U = 6$ eV) calculation has predicted a Ni moment of 0.97 μ_B ⁴⁹. Lastly, the HSE06 hybrid functional yields moments of $\sim 3.03 \mu_B$ and $\sim 0.89 \mu_B$ on Nd and Ni sites, respectively, for the FM phase²⁵, similar to our SCAN values. Overall, our SCAN-based results are quite close to the expected d and f fillings of NdNiO₂ without invoking any ad hoc parameters such as U .

Regarding the presence of short-range magnetic order in the nickelates, strong spin-fluctuations, and AFM exchange interactions have been reported in NdNiO₂⁵⁰, and a quasi-static AFM order below 40 K has been reported in Nd_{0.85}Sr_{0.15}NiO₂⁵¹. Furthermore, related compounds, such as LaNiO₃⁵² are AFM or are close to an AFM quantum critical point (QCP), suggesting that the nickelates with lower dimensions could also be close to magnetic order. Finally, DMFT calculations indicate the presence of strong spin fluctuations in the nickelates^{32,48} with a fluctuating magnetic moment on Ni^{32,48} of 1.1 μ_B , which is close to our value. Notably, although the DFT is expected to describe the ground state properties well^{53–55}, it is not expected to properly capture the spectral properties of correlated systems such as the self-energies.

Electronic structure of the NM phase. Figures 2a, b show the theoretically obtained band structures and densities of states

Fig. 2 The calculated electronic structures and Fermi surfaces of non-magnetic (NM) phase. Orbital-projected electronic structures of NM phase of **a** LaNiO_2 and **b** NdNiO_2 . **c** The calculated Fermi surface of LaNiO_2 and the projection of Fermi surface on $k_z = 0$ and π/c plane. **d** Same as **c** but for NdNiO_2 . Red and blue colors identify portions of the Fermi surface where the Fermi velocity is maximum and minimum, respectively. The Fermi velocity lies in-between the maxima and minima in the green regions.

Fig. 3 The calculated electronic structures and Fermi surfaces of C-type antiferromagnetic (C-AFM) phase. Orbitally-projected electronic structures of C-type antiferromagnetic phase of **a** LaNiO₂ and **b** NdNiO₂. Band structures are “unfolded” on to the primitive $1 \times 1 \times 1$ Brillouin zone. **c** The calculated Fermi surface of LaNiO₂ and the projection of the Fermi surface on the $k_z = \pi/c$ plane. **d** Same as **c** but for NdNiO₂. Red and blue colors identify portions of the Fermi surface where the Fermi velocity is maximum and minimum, respectively. The Fermi velocity lies in-between the maxima and minima in the green regions.

(DOSs) for LaNiO₂ and NdNiO₂ in the non-magnetic (NM) phase, with the various orbital-resolved and site-resolved projections overlaid. In LaNiO₂, Fig. 2a, two distinct bands are seen crossing the Fermi level: one band is of nearly pure Ni- $3d_{x^2-y^2}$ character, while the other is derived from Ni ($3d_{z^2}$, $3d_{xy/yz}$) and La $5d$ orbitals. The latter band produces a spherical 3D electron-like Fermi surface at Γ and A symmetry points (Fig. 2c), whereas the former band generates a large, slightly warped quasi-2D cylindrical Fermi surface similar to the cuprates. These results are consistent with previous LDA¹⁸, GGA²⁵, DFT+U⁴⁵, and LDA+DMFT³¹ studies. Notably, the $3d_{x^2-y^2}$ band here bears a striking resemblance to the corresponding band in the cuprates³⁷, except

that the position of the VHS here shifts from being below to above the Fermi level along the k_z direction in the Brillouin zone. As a result, the Fermi surface transitions from being open in the Γ plane ($k_z = 0$) to becoming closed in the Z plane ($k_z = \pi/c$) (Fig. 2c).

Figure 2b shows that the f electrons in NdNiO₂ lie close to the Fermi level while this is not the case in LaNiO₂. The Ni $3d$ dispersion in LaNiO₂ and NdNiO₂ is similar and this similarity persists across all the magnetic phases we have investigated (Figs. 3, 4, 5, 6). There is significant hybridization between the Nd $4f$ and Ni $3d$ orbitals which can give rise to self-doping effects and induce Kondo physics^{24,26–28,30}. Regarding the Fermi surface

Fig. 4 The calculated electronic structures and Fermi surfaces of G-type antiferromagnetic (G-AFM) phase. Orbital-projected electronic structures of the G-AFM phase of **a** LaNiO₂ and **b** NdNiO₂. Band structures are unfolded into the primitive $1 \times 1 \times 1$ Brillouin zone. **c** Fermi surface of LaNiO₂ and its projection of on the $k_z = 0$ plane. **d** Same as **c** but for NdNiO₂. Red and blue colors identify portions of the Fermi surface where the Fermi velocity is maximum and minimum, respectively. The Fermi velocity lies in-between the maxima and minima in the green regions.

in Fig. 2d, it is seen to display a double-goblet-like hole pocket along the Γ -Z direction with a narrow stem at Γ . Moreover, a large and complex hole Fermi surface appears surrounding Γ near the $k_z = 0$ plane, along with the formation of electron pockets near the M point. Note that in going from Γ to Z in the BZ, the goblet Fermi surface at the Γ point splits into the Fermi pockets around the Z point as the Fermi surface undergoes a change in topology (Fig. 2d).

Electronic structure of the C-AFM phase. Figures 3a, b present the unfolded electronic structure of LaNiO₂ and NdNiO₂ in the C-AFM phase. Like the cuprates, the C-AFM order is stabilized by the opening of a ~ 2 eV band gap in the $d_{x^2-y^2}$ band. However, unlike the cuprates, the *5d* and *4f* states fill the gap in the

nickelates and maintain their metallic nature. In particular, the states near the Fermi level in LaNiO₂ mainly consist of Ni $3d_{z^2}$ and La $5d$ orbitals (Fig. 3a). Moreover, a flat band is seen pinned at the Fermi level along the Z-R-A-Z line, originating from the Ni $3d_{z^2}$ orbitals. NdNiO₂ hosts a similar flat band, which also stems from Ni $3d_{z^2}$ bands. These flat band features have also been noted by Choi et al.⁴⁶, who used a large *U* to push the Nd *4f* states away from the Fermi level. Flat bands produce highly anisotropic Fermi surfaces near the $k_z = \pi/c$ plane in both LaNiO₂ and NdNiO₂ (Figs. 3c, d). In NdNiO₂, there is one additional (ellipsoidal) electron pocket with good dispersion (Fig. 3d), which result from strong Nd *4f*-Ni $3d_{z^2}$ hybridization.

Notably, the appearance of a flat band at the Fermi energy and the associated VHS can produce a landscape of nearly degenerate states⁴⁶. Our calculations on a slightly distorted lattice (see

Fig. 5 The calculated electronic structures and Fermi surfaces of ferromagnetic (FM) phase. Orbital-projected electronic structures of FM phase of **a** LaNiO₂ and **b** NdNiO₂. **c** The calculated Fermi surface of LaNiO₂ and the projection of the Fermi surface on *k_z* = 0 plane. **d** Same as **c** but for NdNiO₂. Red and blue colors identify portions of the Fermi surface where the Fermi velocity is maximum and minimum, respectively. The Fermi velocity lies in-between the maxima and minima in the green regions.

Supplementary Note 2 and Fig. S2) show that the distorted C-AFM phase is lower in energy by only ~3 meV/f.u. compared to the undistorted case. In this connection, we have also carried out computations on possible stripe phases in NdNiO₂ and, as expected, like the cuprates, we have found several low-energy stripe phases lying within 4–6 meV/f.u. of the ground state.

Electronic structure of the G-AFM phase. Figures 4a, b show the electronic structures of LaNiO₂ and NdNiO₂ in the G-AFM magnetic phase. This phase exhibits AFM coupling between the intralayer as well as the interlayer magnetic sites, which is to be contrasted with C-AFM where the interlayer coupling is FM.

Interestingly, here we find no *f* bands near the Fermi level in NdNiO₂, so that the Fermi surfaces in LaNiO₂ and NdNiO₂ become similar. While the G-AFM and C-AFM phases are both dominated by the splitting of the *d_{x²-y²}*-band, and both phases host a region of suppressed DOS within ~0.7 eV of the Fermi level, these two phases differ in that a *3d_{z²}* band lies above the low DOS region in the C-AFM phase, but it lies below this DOS region in the G-AFM phase, resulting in a Fermi energy shift of ~0.7 eV between the two phases.

Electronic structure of the FM phase. Figures 5a, b present the electronic structure of LaNiO₂ and NdNiO₂ in the FM phase. As

Fig. 6 The calculated electronic structures and Fermi surfaces of A-type antiferromagnetic (A-AFM) phase. Figures 6a, b display the unfolded electronic band structure and DOSs of LaNiO₂ and NdNiO₂ in the A-AFM phase. Since the NiO₂ layers are ferromagnetically ordered, the band-splitting here is quite similar to that in the FM phase (Fig. 5). Figure 6a shows that the bands near the Fermi level in LaNiO₂ are mainly of Ni $3d_{x^2-y^2}$ and hybridized Ni $3d_{z^2}$ -La $5d$ characters. However, in NdNiO₂, the main low-lying states near the Fermi level originate from Nd $4f$ states hybridizing with the Ni $3d_{x^2-y^2}$ and $3d_{z^2}$ orbitals. Notably, around -1 eV we see a strong mixing between the Ni $3d_{x^2-y^2}$ and $3d_{z^2}$ orbitals in both LaNiO₂ and NdNiO₂ along the $\Gamma - X$ and $R - A$ directions, which is absent in the NM and FM phases. This strong “orbital-mixing” effect could make the physics of the nickelates quite different from that of the cuprates.

expected, the Ni $3d_{x^2-y^2}$ and $3d_{z^2}$ bands are now spin-split. The Fermi surface is composed of a Ni $3d_{x^2-y^2}$ hole pocket (red) at the M point and an electron pocket from the hybridization between the Ni $3d_{z^2}$ and Nd $4f$ orbitals at the Γ point (blue) (Fig. 5b). Interestingly, the majority spins in the Ni $3d_{x^2-y^2}$ and $3d_{z^2}$ bands point in opposite directions for a given Ni atom. Figures 5c, d show that the Fermi surfaces of LaNiO₂ and NdNiO₂ are quite similar, except that the Γ -point electron pocket in Fig. 5c has grown “propellers” in Fig. 5d, which is due to hybridization between the Ni $3d_{z^2}$ and Nd $4f$ orbitals. A 2D Fermi surface sheet centered at the M point is also seen extending in the k_z direction, produced by the hybridization of the Ni $3d_{xy/yz}$ and Nd $4f$ orbitals. Finally, there is an A-centered hole pocket generated by the Ni $3d_{x^2-y^2}$ band.

Electronic structure of the A-AFM phase. Figures 6a, b display the unfolded electronic band structure and DOSs of LaNiO₂ and NdNiO₂ in the A-AFM phase. Since the NiO₂ layers are ferromagnetically ordered, the band-splitting here is quite similar to that in the FM phase (Fig. 5). Figure 6a shows that the bands near the Fermi level in LaNiO₂ are mainly of Ni $3d_{x^2-y^2}$ and hybridized Ni $3d_{z^2}$ -La $5d$ characters. However, in NdNiO₂, the main low-lying states near the Fermi level originate from Nd $4f$ states hybridizing with the Ni $3d_{x^2-y^2}$ and $3d_{z^2}$ orbitals. Notably, around -1 eV we see a strong mixing between the Ni $3d_{x^2-y^2}$ and $3d_{z^2}$ orbitals in both LaNiO₂ and NdNiO₂ along the $\Gamma - X$ and $R - A$ directions, which is absent in the NM and FM phases. This strong “orbital-mixing” effect could make the physics of the nickelates quite different from that of the cuprates.

Figures 6c, d show that the Fermi surfaces of LaNiO_2 and NdNiO_2 in the A-AFM phase fall into two categories: (1) The $M - A$ direction in both materials is similar to the corresponding FM Fermi surfaces, except for the splitting along the k_z direction due to the AFM stacking of the adjacent FM layers, which leads to the appearance of two pockets near the M point in the Γ -plane. And (2), the $\Gamma - Z$ direction is reminiscent of the NM case, with a $\text{Ni } 3d_{z^2}$ electron pocket at the Γ point in LaNiO_2 , while in NdNiO_2 , the goblet Fermi surface of the NM phase is seen split

into the Z -centered pockets. The blue color of these features (low Fermi velocity) suggests the presence of strong f -electron mixing.

f -electron dispersion. Based on the preceding discussion of the electronic structures, we adduce that the various magnetic phases of NdNiO_2 are mainly stabilized by the $\text{Ni } d$ -electrons, with the f electrons playing a minor role. The Fermi surfaces, however, are strongly affected by the $\text{Nd } 4f$ -electrons through their strong mixing with the $\text{Ni } 3d$ orbitals. The cluster of f -derived bands in the NM phase splits into three subbands once the Nd atoms become polarized, where the detailed structure of the bands depends on the magnetic order, much as is the case in our recent study of SmB_6 ⁴⁴.

Table 2 Comparison of properties of various phases of the nickelates and cuprates. Δ_{dp} and Δ_{eg} denote the splitting of the metallic (Ni and Cu) $3d$ and $O 2p$ bands, and the splitting of the transition-metal ions (Ni and Cu) e_g bands, respectively.

Phases	Δ_{dp} (eV)	Δ_{eg} (eV)	U (eV)	J_H (eV)
<i>LaNiO</i> ₂				
NM	3.21	1.93	–	–
FM	2.70	2.23	4.88	1.01
A-AFM	2.68	1.93	4.99	1.14
C-AFM	2.73	2.02	5.21	1.11
G-AFM	2.72	2.09	5.70	1.45
<i>NdNiO</i> ₂				
NM	3.17	1.94	–	–
FM	2.19	2.04	5.08	1.23
A-AFM	2.31	2.00	4.99	2.14
C-AFM	2.39	2.11	5.27	1.00
G-AFM	2.69	2.13	5.78	1.94
<i>CaCuO</i> ₂				
C-AFM	0.21	2.65	5.35	1.29
<i>La</i> ₂ <i>CuO</i> ₄				
G-AFM	0.30	1.23	4.85 ³⁷	1.25 ³⁷

U and *J_H* denote effective on-site Hubbard potential and the Hund's coupling, respectively

Comparison with the cuprates. Table 2 gives the calculated $d - p$ splitting Δ_{dp} , $\text{Ni } d_{x^2-y^2} - d_{z^2}$ splitting Δ_{eg} , Hubbard U , and Hund's coupling J_H values for the investigated magnetic phases of LaNiO_2 and NdNiO_2 , along with the corresponding values for the cuprates. These quantities are defined precisely in the “Methods” section. Δ_{dp} for LaNiO_2 ranges from 2.68 (A-AFM) to 3.21 eV (NM), while in NdNiO_2 , it spans 2.19–3.17 eV. The partial-DOSs for the Ni and $\text{Cu } 3d$ and $O 2p$ orbitals are shown in Figs. 7a, c, which refer to the C-AFM phase of NdNiO_2 and CaCuO_2 , respectively. The $O-2p$ band-center is seen to be lower than that of the $\text{Ni-}3d$ bands by ~ 2 eV in NdNiO_2 , but the $O-2p$ levels are strongly hybridized with $\text{Cu } 3d_{x^2-y^2}$ orbitals near the Fermi level in CaCuO_2 . We estimate Δ_{dp} in CaCuO_2 in the C-AFM phase to be 0.19 eV, which is significantly smaller than its value in the nickelates. This is also the case for the single-layer La_2CuO_4 in the G-AFM phase, where we estimate Δ_{dp} to be 0.30 eV. These results suggest that LaNiO_2 and NdNiO_2 are closer to the Mott–Hubbard limit rather than being the charge-transfer case in the Zaanen–Sawatzky–Allen classification scheme⁵⁶.

For the NM state, our values of Δ_{dp} for LaNiO_2 and NdNiO_2 are 3.21 eV and 3.17 eV, respectively, which are smaller than the previously reported values of 4.4¹⁸ and 3.7 eV⁵⁷. Note, however, that we obtain Δ_{dp} directly from our first-principles results,

Fig. 7 Comparison the partial density of states (DOS) between C-type antiferromagnetic (C-AFM) phase of NdNiO_2 and CaCuO_2 . a, b Partial DOSs for various sites and orbitals (see legend) in the C-AFM phase of NdNiO_2 . c, d Same as a, b, except that these panels refer to the C-AFM phase of CaCuO_2 .

Table 3 Estimated exchange interactions for the Ni sub-lattice in LaNiO₂ and NdNiO₂.

	LaNiO ₂	NdNiO ₂	La ₂ CuO ₄
J_1 (meV)	52	118	138 ³⁷
J_2 (meV)	−24	−74	−
J_3 (meV)	5	13.5	−

J_1 (intralayer nearest neighbor), J_2 (interlayer nearest neighbor), and J_3 (interlayer next-nearest neighbor) for LaNiO₂ and NdNiO₂. The in-plane J_1 for La₂CuO₄ is also listed for comparison.

which, as ref. ¹⁸ has pointed out, yield smaller values than those based on Wannier fitting of the band structure used by refs. ^{18,57}. For the magnetic phases, the values of Δ_{dp} are smaller than for the NM phase due to the wider d and p orbital spread.

Interestingly, values of Δ_{eg} for the various magnetic phases of LaNiO₂ and NdNiO₂ are all about 2 eV. For example, the Δ_{eg} for LaNiO₂ in the NM phase is 1.93 eV, which is close to the value of 1.95 eV reported in ref. ¹⁸. The similarity of Δ_{eg} values across the infinite-layer nickelates suggests that the Nd $4f$ electrons play a limited role in splitting the Ni $3d$ levels. Values of Δ_{eg} for C-AFM CaCuO₂ and G-AFM La₂CuO₄ read from Figs. 7b, d are comparable to the values for the nickelates given in Table 2.

In order to ascertain the strength of electronic correlations on the nickel site, we have estimated the value of effective Hubbard U and Hund's coupling J_{H} in LaNiO₂ and NdNiO₂ for various magnetic arrangements using Eqs. (2, 3, 4), see “Methods” section below. Values of U so obtained for LaNiO₂, NdNiO₂, CaCuO₂, and La₂CuO₄ are similar, suggesting that the electronic correlations in all these materials are of similar strength. [Our estimated U values are consistent with other recent studies^{24,57}.] The behavior of J_{H} is more subtle. J_{H} values for the nickelates and cuprates are similar, although J_{H} for NdNiO₂ is larger in the A-AFM and G-AFM phases, indicating that J_{H} is sensitive to the effects of interlayer coupling.

Although the mechanism of superconductivity in the HTSCs remains controversial, the view that spin-fluctuations play a central role in determining the physical properties of the cuprates has been gaining increasing acceptance. In this scenario, the exchange-coupling strength would be a good descriptor for the robustness of superconductivity. Accordingly, we consider three different exchange interactions in the Ni sub-lattice: J_1 (intralayer nearest-neighbor), J_2 (interlayer nearest-neighbor), and J_3 (interlayer next-nearest-neighbor), which are shown in Fig. 1b. We estimated strengths of these exchange couplings by computing the total energies of various magnetic phases using the following Heisenberg Hamiltonian.

$$H = \frac{1}{2} \sum_{ij} J_{ij} \mathbf{S}_i \mathbf{S}_j, \quad (1)$$

where $S = \frac{1}{2}$. Here, $J > 0$ and $J < 0$ will represent the AFM and FM spin exchange interaction, respectively. Notably, Nomura et al.⁵⁸ have recently used an approach very similar to ours, where they obtain J in the nickelates by estimating its value in closely related insulating compounds. We have also used our technique in YBCO₇³⁸ and found the resulting J value to be in good agreement with RIXS experiments. For these reasons, we expect our approach for estimating J in the infinite-layer nickelates to be reasonable.

The calculated exchange parameters for LaNiO₂ and NdNiO₂ are listed in Table 3. The in-plane exchange coupling is seen to be antiferromagnetic where J_1 is the largest with a value of 52 meV in LaNiO₂ and 118 meV in NdNiO₂, which are comparable to the values based on a recent LDA+ U calculation⁴⁹. Interestingly,

Fig. 8 Comparison the $3d_{x^2-y^2}$ band between G-type antiferromagnetic (G-AFM) phase of La₂CuO₄ and C-type antiferromagnetic (C-AFM) of NdNiO₂. **a** Cu $3d_{x^2-y^2}$ band for G-AFM La₂CuO₄. **b** Ni $3d_{x^2-y^2}$ band for C-AFM NdNiO₂. Band structures have been unfolded into the $1 \times 1 \times 1$ primitive Brillouin zone.

the out-of-plane couplings show a competition between the ferromagnetic and antiferromagnetic orders. The larger J values in NdNiO₂ (compared to LaNiO₂) are due to its smaller lattice constant.

Superconductivity in the cuprates evolves out of a Mott insulator³, whereas in the iron pnictides it emerges from a metallic state⁵⁹ with strong local magnetic fluctuations. Is magnetic order necessary for d -electron high- T_c superconductivity? The Ni-based superconductors appear to present a counter-example, although both Ni and Nd generally display significant magnetic moments with evidence of magnetic fluctuations or short-range order^{32,46}. Notably, the undoped nickelates are not ordinary metals, but weak insulators^{8,10}. In our previous SCAN-based studies of various correlated materials, we found many competing low-energy magnetic phases indicative of the presence of prominent magnetic fluctuations^{37,38,44}. In the nickelates, our study of the various AFM orders finds a ~ 0.7 eV “pseudogap” (region of low DOS) near the Fermi level, which could explain their weak insulating behavior. Figure 8 compares the AFM gaps in the ground states of the cuprates (a) and the nickelates (b): The gap is seen to be about twice as large in the nickelates compared to the cuprates³⁷.

It is clear that there is a good deal of similarity between the cuprates and nickelates, both in the dispersion of the NM $d_{x^2-y^2}$ band and in the resulting magnetic orders, with the f electrons playing little role in the magnetic transitions even though f electrons modify the Fermi surfaces substantially. Although our normal-state DFT calculation cannot directly address superconductivity, our observation of competing AFM and FM exchange couplings in the nickelates may provide insight into why the T_c dome is smaller in the nickelates compared to the cuprates. Note that, Gu et al.⁹ recently reported mixed s -gaps and d -gaps in scanning tunneling microscopy, while a theory paper by Wu et al.⁶⁰ suggested that the s -wave gap here is a surface effect due to a distorted top NiO₂ layer, which would of course not be captured in our bulk calculations. On the other hand, superconductivity has been found in two rare-earth substituted nickelates^{7,21}, but not in the parent La-based compound, suggesting a more significant role of f electrons. An interesting possibility is that the f electrons lead to heavy-fermion physics (flat bands), which is not present in the cuprates.

Conclusions. We present an in-depth examination of the role of f electrons and magnetic ordering effects in LaNiO₂ and NdNiO₂ within a parameter-free, all-electron first-principles framework.

The magnetic orders in the nickelates are found to be very similar to those in the cuprates in that the transitions are driven by the gapping of the $d_{x^2-y^2}$ band. Compared to the cuprates, the nickelates have a reduced J value with competing AFM and FM spin orders. While the $4f$ electrons play little role in the magnetism of the nickelates, they substantially modify Fermi surfaces in various magnetic states. Our analysis indicates that competing low-energy states could be driven in the nickelates via lattice distortions and stripe formation like the cuprates. Our study thus further reveals the importance of fluctuating magnetic order in correlated materials³⁸.

Methods

All calculations were performed by using the pseudopotential projector-augmented wave method⁶¹ as implemented in the Vienna ab initio simulation package (VASP)^{62,63}. A high-energy cutoff of 520 eV was used to truncate the plane-wave basis set. The exchange–correlation effects were treated using the SCAN³⁶ meta-GGA functional. Spin–orbit coupling effects were included self-consistently. The crystal structures and ionic positions were fully optimized using a force convergence criterion of 0.01 eV/Å for each atom along with a total energy tolerance of 10^{-5} eV. The Fermi surfaces were obtained with the FermiSurfer code⁶⁴. The unfolded band structures including orbital characters were extracted from the supercell pseudo-wavefunction calculations⁶⁵ using the VaspBandUnfolding code⁶⁶.

In order to facilitate comparison with the cuprates, we calculated two quantities: (1) the charge-transfer energy between the Ni $3d$ and O $2p$ orbitals, $\Delta_{dp} = \varepsilon_d - \varepsilon_p$; and, (2) the energy-splitting between the two Ni e_g orbitals, $\Delta_{eg} = \varepsilon_{x^2-y^2} - \varepsilon_{z^2}$. Here, ε_i refers to the band center of the i th orbital. Following previous work,^{18,67} we defined band centers as, $\varepsilon_i = \frac{\int g_i(\varepsilon) \varepsilon d\varepsilon}{\int g_i(\varepsilon) d\varepsilon}$, where $g_i(\varepsilon)$ refers to the partial-density-of-states (PDOS) associated with orbital i . The integration range for Δ_{dp} is set to cover the full bonding and antibonding bands⁶⁷, whereas Δ_{eg} is obtained from an integral over the antibonding bands alone, using an energy window of -3.5 to 2 eV and -4 to 4 eV for the NM and magnetic phases, respectively.

In order to estimate the effective on-site Hubbard potential U and the Hund's coupling J_H , we follow the method of Lane et al.³⁷. Using the site-projected orbital-resolved PDOS $g_{\mu\sigma}$, we determine the average spin-splitting of the μ levels as follows:

$$\bar{E}_{\mu\sigma} = \int_W E g_{\mu\sigma}(E) dE, \quad (2)$$

$$\bar{E}_{d_{x^2-y^2}\uparrow} - \bar{E}_{d_{x^2-y^2}\downarrow} = U(N_{\uparrow} - N_{\downarrow}), \quad (3)$$

$$\bar{E}_{\mu\neq d_{x^2-y^2}\uparrow} - \bar{E}_{\mu\neq d_{x^2-y^2}\downarrow} = J_H(N_{\uparrow} - N_{\downarrow}), \quad (4)$$

where $N_{\uparrow}$ ($N_{\downarrow}$) is the occupation of the spin-up (down) $d_{x^2-y^2}$ orbital and the integration is over the full bandwidth W .

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

R.Z., C.L., B.S., and J.N. performed computations and analyzed the data. B.B., R.S.M., A.B., and J.S. led the investigations, designed the computational approaches, provided computational infrastructure and analyzed the results. All authors contributed to the writing of the manuscript.

Competing interests

The authors declare no competing interests.

Additional information

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