Effective Hamiltonian for nickelate oxides \(\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2\)

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We derive the effective single-band Hamiltonian in the flat \(\text{NiO}_2\) planes for nickelate compounds \(\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2\). We first implement the first-principles calculation to study electronic structures of nickelates using the Heyd-Scuseria-Ernzerhof hybrid density functional and derive a three-band Hubbard model for Ni-O \(pd\sigma\) bands of Ni\(^{3+}\) \(3d_{x^2-y^2}\) and O\(^{2-}\) \(2p_{x/\,y}\) orbitals in the \(\text{NiO}_2\) planes. To obtain the effective one-band \(t-t'-J\) model Hamiltonian, we perform the exact diagonalization of the three-band Hubbard model for the \(\text{Ni}_x\text{O}_{16}\) cluster and map the low-energy spectra onto the effective one-band models. We find that the undoped \(\text{NiO}_2\) plane is a Hubbard Mott insulator, and the doped holes primarily locate on Ni sites. The physics of the \(\text{NiO}_2\) plane is a doped Mott insulator, described by the one-band \(t-t'-J\) model with \(t = 265\) meV, \(t' = -21\) meV and \(J = 28.6\) meV. We also discuss the electronic structure for the “self-doping” effect and heavy fermion behavior of electron pockets of \(\text{Nd}^{3+}\) 5d character in \(\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2\).

I. INTRODUCTION

The layered high-temperature superconductors in copper oxides and iron pnictides have motivated the search for new superconductivity compounds with layered structures\(^{1-4}\). Due to the similar crystal and electronic structure, \(\text{LaNiO}_2\) has been theoretical studies as the possible analogs to the cuprates\(^{5,6}\). Recently, the superconductivity with the critical temperature up to \(T_c \approx 15\) K is indeed discovered in \(\text{Nd}_{0.8}\text{Sr}_{0.2}\text{NiO}_2\) thin film\(^7\), albeit the presence of the superconductivity remains debated\(^{8,9}\). Similar to copper oxides, perovskite nickelates (\(\text{RNiO}_3\), where \(R\) is rare earth or heavy metal such as \(\text{Tl}\) or \(\text{Bi}\)) display lots of strongly correlated physics properties, such as the sharp metal-insulator transitions, particular magnetic order, and charge order\(^{10-14}\). The reduced form of \(\text{RNiO}_3\) leads to the infinite layered phase \(\text{RNiO}_2\)^{14-20}, which has a very flat \(\text{NiO}_2\) plane of the square lattice for Ni\(^{3+}\) with one hole in the \(d_{x^2-y^2}\) orbital. The superconductivity likely occurs in the \(\text{NiO}_2\) plane with the charged carrier doping. In the cuprate superconductor compounds, the strong electronic interactions play a significant role in the electronic structure\(^{21}\), and the effective one-band Hamiltonian has been proposed to describe the low energy physics of the correlation effects for \(3d^9\) electrons\(^{21-23}\). To understand the strongly correlated electronic structures of the nickelate oxides \(\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2\), we need to find out the proper effective (one-band) Hamiltonian to explore the similarity and difference from the cuprate compounds.

\(\text{NdNiO}_2\) crystallizes in the \(P4/mnm\) (No. 123) space group, as depicted in Fig. 1 (a). Four oxygens surround the nickel in a planar square environment (Fig. 1 (d)), and the crystal field splits the \(d\) orbitals as shown in Fig. 1 (e). Ni\(^{3+}\) has the \(d^9\) electronic state configuration, and the highest partially occupied \(d\) orbital is \(3d_{x^2-y^2}\). The rare-earth ion \(\text{Nd}^{3+}\) sits in the center of cuboid formed by eight oxygen ions as shown in Fig. 1 (e). As \(\text{Nd}^{3+}\) in \(\text{Nd}_2\text{CuO}_4\)^{24} and \(\text{Ho}^{3+}\) in \(\text{HoNiO}_3\)^{25}, \(\text{Nd}^{3+}\) has the local \(4f\) moment far below the Fermi energy level. \(\text{Nd} \, 5d\) orbitals have the split energy levels, as shown in Fig. 1 (e), and near the Fermi energy, the lowest \(5d\) orbital in \(\text{Nd}^{3+}\) is \(d_{z^2}\). Therefore, in the simple reckoning for the relevant electronic structure, there are 12 bands near Fermi energy level corresponding to mainly Ni \(d\) (5 states) and O \(p\) \((2 \times 3 \) states) and \(\text{Nd} \, d_{z^2}\) (1 state). Previous density functional theory (DFT) studies within the local density approximation (LDA) on \(\text{LaNiO}_2\)^{26} supports the rough impression of the non-magnetic electronic structures.

In this paper, we first implement the first-principles simulations to derive a three-band Hubbard model for Ni-O \(pd\sigma\) bands of Ni\(^{3+}\) \(3d_{x^2-y^2}\) and O\(^{2-}\) \(2p_{x/\,y}\) orbitals in the \(\text{NiO}_2\) planes. Based on the three-band Hubbard model, we perform the exact diagonalization for the \(\text{Ni}_x\text{O}_{16}\) cluster and obtain the low-energy one-band effective Hamiltonian for the \(\text{NiO}_2\) planes. We also discuss the electronic structure for the “self-doping” effect and the heavy-fermion behavior of electron pockets of \(\text{Nd}^{3+}\) 5d character in \(\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2\). We present our main results in the two successive stages in Sec. II. We discuss the physics of the one-band \(t - J\) model in the \(\text{NiO}_2\) planes in Sec. III. In the Appendix, we present the results for \(\text{La}_{1-x}\text{Sr}_x\text{NiO}_2\), implying the generic electronic structures of \(\text{NdO}_2\) series. We also include other supplementary results for \(\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2\) in the Appendix.

The main results are summarized as follows. In Sec. II A, we first perform DFT calculations of \(\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2\) within the Heyd-Scuseria-Ernzerhof (HSE) hybrid density functional. We notice that in the previous study of the nickelates \(\text{RNiO}_3\), the HSE hybrid functional method is essential to reproduce the experimentally observed magnetic ground state\(^{27}\). On the generalized gradient approximation (GGA) level, DFT simulations suggest the G-type antiferromagnetic (AFM) within the \(\text{NiO}_2\) plane and AFM between \(\text{NiO}_2\) planes along the \(c\) direction) ground state of the moment on Ni sites in the parent compound \(\text{NdNiO}_2\). The HSE results for \(\text{Nd}_{1-x}\text{Sr}_x\text{NiO}_2\) adopt the G-type spin configuration to mimic strong spin correlations. In Sec. II B, we describe the electronic structures based on the DFT simulations. Three-dimensional electron Fermi pockets of \(\text{Nd} \, 5d\) character are found near Fermi energy and behave as a heavy-fermion system due to its coupling with the local moments of \(\text{Nd} \, 4f\), similar to the electron-doped cuprate \(\text{Nd}_{1-x}\text{Ce}_x\text{CuO}_2\)^{27}. We also
implement PerdewBurkeErnzerhoff (PBE) functional in gen-
Vienna Ab Initio Simulation Package (V A S P) 30
one-band effective Hubbard model is also given.
and the experiment
Ni sites in a good agreement with the HSE results in Sec.II A
in hole-doped nickelates, the doped holes primarily locate on
Mott insulator described by the effective on-band
band
lect and map the low-energy spectra onto the effective one-
cluster within the three-band Hubbard model are used to se-
NdNiO
principles simulations for the non-magnetic ground state for
obtain the three-band Hubbard model for Ni-O p$d$σ bands of
Ni$^+$ 3$d_{x^2}$ and O$^{2-}$ 2$p_x/p_y$ orbitals in the NiO$_2$ planes.
In Sec. II C, we derive the effective one-band Hamiltonian of the NiO$_2$ plane, following the standard procedure in the cuprates 22,23. We derive the parameters for the three-band Hubbard model of the Ni-O p$d$σ bands for Ni$^+$ 3$d_{x^2}$ and O$^{2-}$ 2$p_x/p_y$ orbitals from the LDA results from the first-principles simulations for the non-magnetic ground state for NdNiO$_2$. Exact diagonalization (ED) studies of the Ni$_5$O$_{16}$ cluster within the three-band Hubbard model are used to select and map the low-energy spectra onto the effective one-band t-t$'$.t$''$.J model. According to ED results on finite clusters, in hole-doped nickelates, the doped holes primarily locate on Ni sites in a good agreement with the HSE results in Sec.II A and the experiment 25, while for cuprate the holes mainly locate on oxygen site 22,23. The physics of the NiO$_2$ is a doped Mott insulator described by the effective on-band t-t$'$.t$''$.J model with $t = 265$ meV, $t' = -21$ meV, and $J = 28.6$ meV. The one-band effective Hubbard model is also given.

II. RESULTS

A. DFT results of Nd$_{1-x}$Sr$_x$NiO$_2$

We performed first-principles calculations based on DFT 29 with the HSE06 hybrid functional 30 as implemented in the Vienna Ab Initio Simulation Package (V A S P) 31-33. We also implement PerdewBurkeErnzerhoff (PBE) functional in gen-
ralized gradient approximation (GGA) 34 and strongly con-
strained and appropriately normed semilocal density func-
tional (SCAN) in meta-GGA 35 for comparisons. We use an energy cutoff of 500 eV and $12 \times 12 \times 12, 4 \times 4 \times 4,$ and $4 \times 4 \times 4$ Monkhorst-Pack grids 36 in the PBE, SCAN and HSE06 calcula-
tions, respectively. The 4$f$ orbitals in Nd$^{3+}$ are expected to
local magnetic moment as Nd$^{3+}$ in Nd$_2$CuO$_4$ 24
Ho$^{3+}$ in HoNiO$_3$ 25, and we treat them as the core-level electrons in the Nd pseudopotential in the HSE06 calculations.
We check the results with 4$f$ valence electrons pseudo-
potential within GGA+U ($U_{Jf} = 10$ eV) scheme 37 in Ap-
pendix A. We also check that the spin-orbit coupling does not
significantly change the DFT results in the Appendix. There-
fore, we only present the simulations by taking 4$f$ the core-
level electrons in Nd pseudopotential and do not include the
spin-orbit coupling in our HSE06 hybrid functional simula-
tions.
We use the bulk lattice constants $a = b = 3.9208$ Å, $c = 3.281$ Å 40 for Nd$_{1-x}$Sr$_x$NiO$_2$ and didn’t optimize the crystal structure during the DFT simulations. In this setup, the NiO$_2$ plane remains flat in the doped case Nd$_{0.75}$Sr$_{0.25}$NiO$_2$. We didn’t consider the lattice distortion effect upon doping in the doped nickelate oxides.

Figure. 1 (b) is the HSE06 band structure for the non-
magnetic state in NdNiO2. The band ordering is differ-
ent from the crystal field theory in Fig. 1 (e) due to the covalent
effect in the hybridizations between $d$ orbitals (Nd and Ni) and
2p orbitals (O). There are 12 electronic bands near the Fermi
energy $E_F$, corresponding to orbitals mainly of Ni d (5 states)
and O p (2×3 states) and Nd d_{z^2} (1 state) for the relevant electronic structure. The O 2p bands extend from about -10 to -5 eV. The Ni 3d bands are distributed from -5 to 2 eV, while the broad Nd 5d states range from -1 to 10 eV. The Ni 3d_{x^2−y^2} and Nd 5d_{z^2} cross the Fermi energy $E_F$ as expected from the crystal field splitting, as shown in Fig. 1 (c). Ni 3d_{x^2−y^2} is very broad along the $Γ$-X-M direction due to the strong d$p_σ$ antibonding interaction with oxygen p$_{x/y}$ states and encloses holes centered at the $M$ point. Ni 3d$_{xy/yz}$ bands localize near -4 eV due to the weak d$p_π$ hybridization with O 2p states. Around $A$ point, the Nd 5d_{z^2} state forms an electron pocket around the $Γ$ point, goes up along $Γ$-Z direction, and lies above the Fermi energy level with $k_z = \pi/c$. The Nd 5d$_{xy}$ lowers down and crosses the Fermi level, forming an electron pocket around $A$ point.

Along $Γ$-Z direction, four O 2p$_{x/y}$ bands and four Ni+ (3dxz/yz, 3d$_{xy}$, and 3d$_{x^2−y^2}$) bands have weak dispersions, indicating the two-dimensional features of these bands. Ni 3d$_{z^2}$ and Nd 5d states are dispersive along with $Γ$-Z directions and three-dimensional extended. The two electron pockets around $Γ$ and $A$ points of the Nd 5d orbital character have the Ni orbitals mixing. However, such mixing is not quickly resolved in Fig. 1 (b) since the Nd 5d characters dominate the electron pockets and cover up the contributions from Ni orbitals. To further clarify the mixing character, we also present the band structure for the simulated SrNiO$_2$ in with the same structure as NdNiO$_2$, in order to eliminate Nd 5d orbitals in Fig. 2. From the comparing between Fig. 1 (b) and Fig. 2, we can see that Nd 5d_{z^2} band crosses $E_F$ around $Γ$ point with Ni 4$s$ mixing, while Nd 5d$_{xy}$ band crosses $E_F$ around $A$ point with Ni 4$p_z$ mixing.

The GGA band structure of NdNiO$_2$ in Appendix A is quite similar to the LDA band structure of LaNiO$_2$\cite{Qiu2001}, indicating a generic electronic structure in the RNiO$_2$ family. Compared with the GGA band structure (Appendix A), for the non-magnetic state, the mixing of the exact exchange in the HSE06 hybrid functional separates the two bands crossing the Fermi energy $E_F$ away from other bands, without significant change of dispersions and relative positions of the bands far from $E_F$. The separation of two bands can also be reproduced in the LDA+U results\cite{Ma2007}. However, in the LDA+U scheme, Ni 3d$_{x^2−y^2}$ is raised by $U$ and crosses $E_F$ when $U$ is large\cite{Ma2007}, different from the HSE06 hybrid functional simulation for the non-magnetic state.

The magnetization measurement and neutron powder diffraction didn’t reveal the long-magnetic order in LaNiO$_2$ and NdNiO$_2$: however, the paramagnetic susceptibilities imply the (at least short) spin correlations\cite{Kanda2001, Koizumi2001}. The absence of long-range magnetic order may be due to poor sample qualities, or due to the “self-doping” effects of the Nd 5d electron pockets. The strong correlation for electrons on Ni$^{+}$ induce the magnetism in the system.

To demonstrate the correlation and magnetism in NdNiO$_2$, we calculate the magnetic moment and compare the ground state with the non-magnetic one within different functionals (GGA, SCAN, and HSE06). To save computation time, we consider the ferromagnetic spin configuration in the primary unit cell. The results are list in TABLE I. Than the non-magnetic state, the magnetic states have increased moments on Ni with increasingly lower ground state energies from GGA, SCAN to HSE06. For GGA, the magnetic state has even higher energy than the non-magnetic state, indicating the non-magnetic ground state within the GGA functional, consistent with the previous study in LaNiO$_2$\cite{Ma2007}. The SCAN functional includes more correlation effects, and the magnetism is significantly enhanced. The magnetic ground state has further

![FIG. 2. GGA band structures of the non-magnetic state in the simulated SrNiO$_2$ in with the same structure as NdNiO$_2$. The projected band structures of d orbitals in Sr, and 4p orbitals in Ni are also shown. The color in (a) has the same meanings as Fig. 1 (b). The color turquoise in (b) indicates the projected Ni 4s orbital (mainly locates at $Γ$ point at around 2.4 eV).](image)

| Functional | Magnetization | $\Delta E$ (meV) |
|------------|---------------|-----------------|
| HSE06      | 0.94          | 744.7           |
| SCAN       | 0.76          | 142.7           |
| GGA        | 0.05          | -0.1            |

TABLE I. Theoretical magnetic moments (in $\mu_B$) on Ni in NdNiO$_2$ calculated with HSE06, SCAN, and PBE functional. $\Delta E$ is the energy difference between the non-magnetic state and the ferromagnetic state.
lower energy than the non-magnetic state in the HSE06 functional. The fact that the correlation achieves the magnetism is a strong indication that NdNiO$_2$ is magnetic if we include the correlations.

Even though there is no long-range magnetic order, we still impose static magnetic configurations on Ni ions in Nd$_{1-x}$Sr$_x$NiO$_2$ to mimic the spin correlations in the DFT simulations for electronic structures. We find the G-type AFM magnetic state has the lowest ground state energy within the GGA DFT simulations. Therefore, in the HSE06 simulations, we adopt the G-type AFM spin configurations on Ni using the $\sqrt{2} \times \sqrt{2} \times 2$ supercell to study the electronic structures of Nd$_{1-x}$Sr$_x$NiO$_2$ ($x = 0, 0.25$). Therefore, the Brillouin zone is folded by the G-type AFM spin configuration. However, without any confusion, we still use notations, $\Gamma$, $X$, $M$, $Z$, $R$, $A$, for high symmetry k-points in the folded Brillouin zone.

Figure 3 displays the HSE06 band structures of Nd$_{1-x}$Sr$_x$NiO$_2$ ($x = 0, 0.25$) with G-type AFM magnetic configuration on Ni. In the HSE06 hybrid functional results, the AFM magnetic configuration not only folds the band structure, but also dramatically change the bands near the Fermi energy $E_F$, very different from the GGA result for the G-type AFM magnetic configuration in the Appendix A. The significant change of band structures in the AFM states between GGA and HSE06 implies strong correlations in Nd$_{1-x}$Sr$_x$NiO$_2$.

In the G-type AFM state of NdNiO$_2$ (Fig. 3 (a)), the Nd 5$d_{xz}$ band is raised above the Fermi energy $E_F$ by the correlation, eliminating the electron pocket of the Nd 5$d_{xy}$ character in the non-magnetic state (Fig. 1 (b)). The Nd 5$d_{xy}$ band is also raised, but the electron pocket of this band still exists. We remind here again that the electron pocket of the Nd 5$d_{xy}$ character has the Ni 4$p_z$ orbital mixing. The Nd 5$d_{xy}$ electron pocket locates around $\Gamma$ point in the folded magnetic Brillouin zone. Therefore, the AFM spin correlation has an essential influence on the “self-doping” effect of Nd 5$d$ electron pockets. For the Ni 3$d$ band, the AFM correlation significantly renormalizes the bandwidth. The Ni 3$d_{x^2-z^2}$ bands split into upper and lower Hubbard bands separated by around 5 eV due to the AFM spin configuration, indicating the strong correlation in NdNiO$_2$. The lower Hubbard 3$d_{x^2-z^2}$ band locates lower than the Ni 3$d_{xz}$ band, bringing the doping problem into a complicated situation.

When one Sr$^{2+}$ ion substitutes Nd$^{3+}$ in NdNiO$_2$, we dope an extra hole into the NiO$_2$ planes. To simulate the doped nickelate oxide, we calculate the band structures of Nd$_{0.75}$Sr$_{0.25}$NiO$_2$ (Nd$_3$SrNi$_4$O$_8$) in the $\sqrt{2} \times \sqrt{2} \times 2$ supercell with the G-type AFM spin configuration on Ni ions. Fig. 3 (b) is the HSE06 band structure of Nd$_{0.75}$Sr$_{0.25}$NiO$_2$. The Nd 5$d_{xy}$ band goes up above the Fermi energy level, and the band minimum locates 0.2 eV. We can expect that Nd 5$d_{xy}$ band contributes Hall coefficient at high temperatures at low dopings. The doped hole does not go to the Ni 3$d_{xz}$ band. Instead, it locates on the 3$d_{x^2-y^2}$ band, which goes up above 3$d_{xz}$ in Nd$_{0.75}$Sr$_{0.25}$NiO$_2$. Therefore, the doped hole does not polarize Ni$^{2+}$ into the $S = 1$ local moment, but create an $S = 0$ hole-like doped charge carrier. The Ni 3$d_{x^2-y^2}$ encloses the hole pockets around $X$ point in the folded Brillouin zone ($M$ in the original non-magnetic Brillouin zone). The HSE06 band structures in Fig. 3 suggest that the undoped NiO$_2$ plane is Hubbard-like Mott insulator, not the charge-transfer-like one as in the cuprate. The doped hole goes into Ni 3$d_{x^2-y^2}$ orbital, creating an $S = 0$ hole-like charge carrier, rather than into the O$^{2-}$-2$p_{x/y}$ orbitals.

**B. Electronic structures of Nd$_{1-x}$Sr$_x$NiO$_2$**

The presence of the electron pockets of Nd 5$d$ bands suggests the “self-doping” effect, implying a small charge transfer from these pockets to the Ni-O sheets even without chemical doping. The “self-doping” effect also exists in the cuprate family, e.g., YBa$_2$Cu$_3$O$_7$ and Bi$_2$Sr$_2$CaCu$_2$O$_8$. The three-dimensional electron pockets of Nd 5$d_{xy}$ states have the mixing with Ni 4$p_z$ orbitals. The “self-doping” effect allows some charge transfer and changes the hole count in the NiO$_2$ planes, resulting in the metallic behavior even without chemical dop-
ing. In the Appendix A, we present the GGA band structure with the 4f valence electron pseudopotential within the GGA+U scheme. We can see that Nd $4f$ states have the mixing with Nd $5d$ states. Due to the presence of Nd $4f$ orbitals, the electron pockets of the Nd $5d_{xy}$ character hybridizes with the $4f$ local moments, behaving as a heavy-fermion system

$$H_K = \sum_k \epsilon_k a_k^\dagger a_k + J_K \sum_i s_i \cdot S_i^{\mathrm{HF}},$$

where $a_k^{\sigma}$ creates fermion with momentum $k$ on the electron pockets and $s_i$ and $S_i^{\mathrm{HF}}$ are the spin operators for the $4f$ electrons and $4f$ local moments, respectively.

We now turn to the physics of the NiO$_2$ plane with the hole doping, following the process for the CuO$_2$ planes in cuprates $^{22,23}$. The HSE06 band structures in Fig. 3 suggest that the undoped NiO$_2$ plane is Hubbard-like Mott insulator, and the doped hole goes into Ni $3d_{x^2-y^2}$ orbital, creating an $S = 0$ hole-like charge carrier. Therefore, the physics of the NiO$_2$ plane with the charge doping is described by the three-band Hubbard model for the $d\sigma\tau$ bands of Ni $3d_{x^2-y^2}$ and O $2p_{x/y}$ orbitals. Fig. 4 schematically presents the orbitals of Ni $3d_{x^2-y^2}$ and O $2p_{x/y}$ with the green and brown colors corresponding to the positive and negative signs of wave functions, respectively.

We assume a vacuum state $a^\dagger_0 \phi_0^\dagger$ and introduce the operators $d^\dagger_{\sigma}$ and $p^\dagger_{\sigma}$ creating the Ni $3d_{x^2-y^2}$ hole and the O $2p_{x/y}$ hole at the $i$-the Ni site and the $l$-th O site, respectively, with the spin $\sigma = \uparrow / \downarrow$. The holes hop between Ni $3d_{x^2-y^2}$ and O $2p_{x/y}$ orbitals with the amplitude $t_{dp}$, and among O $2p_{x/y}$ orbitals with the amplitude $t_{pp}$. We set the on-site potential of Ni $3d_{x^2-y^2}$ as $\epsilon_d = 0$, and the chemical potential difference between Ni $3d_{x^2-y^2}$ and O $2p_{x/y}$ orbitals as $\epsilon = \epsilon_p - \epsilon_d$. The strong correlations involve the on-site interactions $U_p$ and $U_d$, and inter-site interactions $U_{dp}$ and $U_{pp}$ for holes on O $2p_{x/y}$ and Ni $3d_{x^2-y^2}$ orbitals. The three-band Hubbard model of the dp$\sigma\tau$ bands reads out

$$H_{dp} = \sum_{\langle il \rangle} t_{dp}(d^\dagger_{i\sigma}p_{l\sigma} + \text{h.c.}) + \sum_{\langle ll' \rangle} t_{pp}(p^\dagger_{l\sigma}p_{l'\sigma} + \text{h.c.})$$

$$+ \sum_{l\sigma} \epsilon_p^l_{\sigma} p_{l\sigma} + U_d \sum_i n_{d\uparrow} n_{d\downarrow} + U_p \sum_l n_{p\uparrow} n_{p\downarrow} + U_{dp} n_{d\uparrow} n_{p\downarrow} + U_{pp} n_{p\uparrow} n_{p\downarrow}.$$  

Here $\langle \cdots \rangle$ denotes the nearest neighbor bonds.

### C. Effective $t'$-J model Hamiltonian of NiO$_2$ planes

The tight-binding parameters $t_{dp}$, $t_{pp}$, and $\epsilon$ can be obtained from the Wannier fitting $^{39,40}$ of the 12 bands in the LDA simulations for the non-magnetic band structure of NdNiO$_2$. It is noteworthy that the HSE06 band structure in Fig. 1 (b) already contains the renormalization effect due to the strong correlations. There is double-counting for the correlations if we obtain the tight-binding parameters from the HSE06 band structure. The parameters are given as $\epsilon = 4.2$ eV, $t_{pd} = 1.3$ eV, $t_{pp} = 0.6$ eV. We choose a proper basis of the $3d_{x^2-y^2}$ and $2p_{x/y}$ for all positive $t_{dp}$ and $t_{pp}$. To match the similar sizable band splitting of the lower and upper Hubbard bands of the Ni $3d_{x^2-y^2}$ orbitals in the HSE06 band structure in Fig 3, we set the interaction terms as $U_d = 7.5$ eV, $U_p = 5.0$ eV, $U_{pd} = 4.0$ eV, and $U_{pp} = 2.0$ eV in the ED calculation for the derivation of the effective one-band Hamiltonian.

We follow the process in the cuprates $^{22,23}$ to obtain the effective one-band Hamiltonian of the NiO$_2$ planes. We perform direct ED studies of the three-band Hubbard model $H_{dp}$ in Eq. 2 and find the effective one-band Hamiltonian by the

![FIG. 4. Schematic representation of the orbitals (Ni $3d_{x^2-y^2}$ and O $p_{x/y}$) included in the three-band Hubbard model in the Ni$_2$O$_{16}$ cluster. The effective one-band model has the degree of freedom only on the five Ni $3d_{x^2-y^2}$ orbitals connected by the solid ($t$ and $J$) and dashed ($t'$ and $J'$) lines.](image)

![FIG. 5. The low-energy spectrum for Ni$_2$O$_{16}$ cluster calculated in the three-band Hubbard model in comparison to the mappings onto the Heisenberg Hamiltonian for the five-hole case.](image)
low-energy spectrum mapping. We carry out the ED calculation for the Ni$_5$O$_{16}$ cluster as shown in Fig. 4 with five and six holes for the undoped and hole-doped NiO$_2$ planes, respectively. The Ni$_5$O$_{16}$ cluster is embedded in an array of Ni 3$d^9$ sites which shift the effective on-site energy of the outer O orbitals due to the inter-site Coulomb energy.

Figure 5 is the low-energy spectrum mapping for the Ni$_5$O$_{16}$ cluster with five holes in the insulating ground state of the undoped NiO$_2$ planes. The spin-1/2 Heisenberg model

$$H_J = \sum_{ij} J_{ij} S_i \cdot S_j,$$  \hspace{1cm} (3)

with the nearest neighbor and next nearest neighbor exchange interactions $J = 28.6$ meV and $J' = 0.4$ meV well reproduces the low-energy spectrum for the three-band Hubbard model as shown in Fig. 5. Therefore, we obtain the effective Heisenberg model for the undoped NiO$_2$ planes.

For the hole-doped phase, the calculated low-energy spectra for six holes in the Ni$_5$O$_{16}$ cluster is shown in Fig 6. The spin-1/2 Heisenberg model for Ni-O based compounds describes the low-energy physics of the nickelate oxides with $t$-$t'$-$J$ model Hamiltonian, we find that the physics of NiO$_2$ planes in NdNiO$_2$ is a doped Mott insulator, even further away from a conventional Fermi liquid than the superconducting cuprates. The undoped CuO$_2$ plane in the cuprate is a charge-transfer type Mott insulator, but the undoped NiO$_2$ plane in the nickelate is a Hubbard Mott insulator. Both the NiO$_2$ and CuO$_2$ planes have the same effective one-band $t$-$t'$-$J$ model Hamiltonians, and they have the similar physics of the superconductivity.

The detecting of the superconductivity in the nickelate oxide certainly motivates the studies of the effective Hamiltonian in this paper. However, two recent experimental papers reported non-superconductivity results in Nd$_{1-x}$Sr$_x$NiO$_2$ getting the presence of superconductivity into controversy. The existence of the superconductivity in the doped nickelate oxides is a crucial issue and needs further exploring in the experiments. In this paper, our study is the consequence of a quantum-chemical description of Nd$_{1-x}$Sr$_x$NiO$_2$; however, we cannot provide direct theoretical implication of the existence of the superconductivity. The effective Hamiltonian in this work provides essential support for, and constraint on, models to describe the low-energy physics of the nickelate oxides, regardless of the presence of the superconductivity.
and RPA analysis of the pairing symmetry is done in Refs.\textsuperscript{44,47}. The hopping parameters for the three-band Hubbard model are (implicitly or explicitly) given from the Wannier fitting in Refs.\textsuperscript{33,44,47–50}, with similar values to our work. The hopping parameters (\(t\) and \(t'\)) of the effective Ni \(3d_{z^2−r^2}\) band are also given in Refs.\textsuperscript{43,47–49} with \(t \sim 370\) meV and \(t' \sim −100\) meV, larger than the values \(t = 265\) meV and \(t' = −21\) meV renormalized by correlations in our work. The exchange interaction is estimated as \(J = 100\) meV in Ref.\textsuperscript{47}, larger than our values \(J = 28.6\) meV. The Hubbard-Mott scenario is also proposed in Ref.\textsuperscript{46} and the charge-transfer gap is estimated \(\epsilon = 7 ∼ 9\) eV larger than our value \(\epsilon = 4.2\) eV. In the Ref.\textsuperscript{46}, the \(S = 1\) Ni\(^{2+}\) state is proposed when the hope is doped into the NiO\(_2\) planes, different from the \(S = 0\) Ni\(^{2+}\) state in our work. We notice that if the charge-transfer gap is taken as the value we use in the work, \(\epsilon = 4.2\) eV, the \(S = 0\) Ni\(^{2+}\) state is favored according to the calculation in Ref.\textsuperscript{46}.

### IV. ACKNOWLEDGEMENTS

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Appendix A: Supplementary DFT results for Nd\(_{1−x}\)Sr\(_{x}\)NiO\(_2\)

In this part, we provide the supplementary DFT results for Nd\(_{1−x}\)Sr\(_{x}\)NiO\(_2\). The main purpose of the supplementary results is two-folded: (a) check the validity of the Nd pseudopotential with the core-level \(4f\) electrons; (b) compare the GGA and SCAN band structures to the HSE06 results in the main text. The spin-orbital coupling is also checked in this section.

1. GGA+U calculations for the Nd \(4f\) valence electron pseudopotential

As Nd\(^{3+}\) in Nd\(_2\)CuO\(_4\)\textsuperscript{24} and Ho\(^{3+}\) in HoNiO\(_3\)\textsuperscript{25}, Nd\(^{3+}\) has the local \(4f\) moment far below the Fermi energy level \(E_F\). In the main text, we treat the \(4f\) electrons in Nd\(^{3+}\) as the core-level electrons in the Nd pseudopotential. In this subsection, we verify the validity of this treatment.

Figure 7 are the band structure of G-type AFM states for NdNiO\(_2\) and Nd\(_{0.75}\)Sr\(_{0.25}\)NiO\(_2\) calculated with \(4f\) valence electron pseudopotential within GGA+U scheme. Without the U term, the \(4f\) electrons form very localized bands near the Fermi level. In the G-type AFM states, the local moments of \(4f\) electrons are also AFM within the same Nd plane and between Nd planes along the \(c\) direction. We take the on-site interaction for \(4f\) electrons \(U_{4f} = 10\) eV in the GGA+U calculation which splits the \(4f\) bands above and below the Fermi energy level \(E_F\). During the calculations, we also include the spin-orbital couplings.

According to Fig. 7, we can see that the \(4f\) electron states couple to Nd \(5d\) bands, however, doesn’t significantly change the band structures near the Fermi level. Thus we can treat \(4f\) orbitals as the core-level electrons in the Nd pseudopotential.

2. GGA and SCAN band structures

In this subsection, we present the band structures for Nd\(_{1−x}\)Sr\(_{x}\)NiO\(_2\) within GGA and SCAN functionals without/with spin-orbital couplings in Fig. 8 and Fig. 9, respectively. Again, once soc is taken into account, there is no significant change in the band structures. The main features of SCAN band structures are very close to those in HSE06 band structures.

![Fig. 7. GGA band structures of G-type AFM states for a supercell in (a) NdNiO2 and (b) Nd0.75Sr0.25NiO2 calculated with 4f valence electrons pseudopotential within GGA+U (U4f = 10 eV) scheme with SOC. The notations of Γ, X, M, Z, R and A are in the folded magnetic Brillouin zone. The projected band structures of f orbitals (red) in Nd are also shown. The Fermi level is set at 0 eV.](image-url)
FIG. 8. GGA band structures of Nd$_{1-x}$Sr$_x$NiO$_2$. (a), (b), and (c) correspond to Fig. 1 (b) (the non-magnetic state for NdNiO$_2$), Fig. 3 (a) (the G-type AFM state for NdNiO$_2$) and (b) (the G-type AFM state for Nd$_{0.75}$Sr$_{0.25}$NiO$_2$), respectively, without SOC. (d), (e), and (f) are the results with turning on SOC.

Appendix B: HSE06 results of LaNiO$_2$

The LDA result for LaNiO$_2$ has been studied previously. In this subsection, we implement the HSE06 hybrid functional to calculated band structures of the non-magnetic state in LaNiO$_2$ and G-type AFM states for a supercell in LaNiO$_2$ and La$_{0.75}$Sr$_{0.25}$NiO$_2$ in Fig. 10. The band structures are very similar to the results of NdNiO$_2$, implying the generic electronic structures in the RNiO$_2$ family.
FIG. 9. SCAN band structures of Nd$_{1-x}$Sr$_x$NiO$_2$, similar to Fig. 8.

FIG. 10. HSE06 band structures of La$_{1-x}$Sr$_x$NiO$_2$ without SOC. (a) non-magnetic state for LaNiO$_2$, (b) G-type AFM state for LaNiO$_2$, and (c) G-type AFM state for La$_{0.75}$Sr$_{0.25}$NiO$_2$, corresponding to Fig. 1(b), Fig. 3(a) and (b), respectively.
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