Lifshitz transition and frustration of magnetic moments in infinite-layer NdNiO$_2$ upon hole-doping

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Motivated by the recent discovery of superconductivity in the infinite-layer (Sr,Nd)NiO$_2$ films with Sr content $x \approx 0.2$ [Li et al., Nature (London) 572, 624 (2019)], we examine the effects of electron correlations and Sr-doping on the electronic structure, Fermi surface topology, and magnetic correlations in (Nd,Sr)NiO$_2$ using a combination of dynamical mean-field theory of correlated electrons and band-structure methods. Our results reveal a remarkable orbital selective renormalization of the Ni 3d bands, with $m^*/m \sim 3$ and 1.3 for the $d_{x^2-y^2}$ and $d_{x^2+y^2}$ orbitals, respectively, that suggests orbital-dependent localization of the Ni 3d states. We find that upon hole doping (Nd,Sr)NiO$_2$ undergoes a Lifshitz transition of the Fermi surface which is accompanied by a change of magnetic correlations from the three-dimensional (3D) Néel $G$-type (111) to the quasi-2D $C$-type (110). We show that magnetic interactions in (Nd,Sr)NiO$_2$ demonstrate an unanticipated frustration, which suppresses magnetic order, implying the importance of in-plane spin fluctuations to explain its superconductivity. Our results suggest that frustration is maximal for Sr-doping $x \approx 0.1$–0.2, which is in agreement with an experimentally observed doping value Sr $x \approx 0.2$ of superconducting (Nd,Sr)NiO$_2$.

The recent discovery of superconductivity in the infinite-layer Sr-doped NdNiO$_2$ films (Nd$_{0.8}$Sr$_{0.2}$NiO$_2$) with the critical temperature up to $T_c \sim 15$ K has attracted a lot of attention from researchers around the world [1]. NdNiO$_2$ has a similar planar crystal structure to that of the parent “infinite layer” superconductor CaCuO$_2$, which exhibits superconductivity below $T_c \approx 110$ K upon hole doping [2,3]. As Ni is isoelectronic to copper in NdNiO$_2$, it has a nominal $d^9$ configuration. Based on this, it was expected that analogous to cuprates the low energy physics of Sr-doped NdNiO$_2$ is dominated by electrons in the planar Ni $z^2$–$y^2$ states. However, unlike cuprates, in the infinite-layer nickelate the Ni $x^2$–$y^2$ states are found to experience strong hybridization with the Nd 5d orbitals (primarily the 3$d^2$–$s^2$ and $xy$ orbitals), yielding a non-cuprate-like Fermi surface [5,6]. While the electronic structure of NdNiO$_2$ has recently been widely studied using various band structure methods [7,9], model techniques [10,11], and DFT+dynamic mean-field theory (DFT+DMFT) [12,13] methods [14,15], the properties of Sr-doped NdNiO$_2$ are still poorly understood. For NdNiO$_2$, DFT+DMFT calculations reveal significant correlation effects within the Ni 3d orbitals, which are complicated by large hybridization with the Nd 5$d$ states [14,15]. Moreover, based on the experiments two features that are central to copper oxides—the Zhang-Rice singlet and large planar spin fluctuations—were claimed to be absent (or diminished) in (Nd,Sr)NiO$_2$ [11,6].

Here we explore the effects of electronic correlations and Sr-doping on the electronic structure of (Nd,Sr)NiO$_2$ using a fully self-consistent in charge density DFT+DMFT method [12,13] implemented with plane-wave pseudopotentials [20,21]. DFT+DMFT has been proved to be among the most advanced theoretical methods for studying the electronic properties of strongly correlated materials, such as correlated transition metal oxides, heavy-fermions, Fe-based superconductors, e.g., to study the phenomena of a Mott transition, collapse of local moments, large orbital-dependent renormalizations, etc. [22] We use this advanced computational method to study the Fermi surface topology and magnetic correlations, as well as their impact on magnetism of (Nd,Sr)NiO$_2$ upon Sr-doping.

We adopt the experimental lattice parameters measured for the Nd$_{0.8}$Sr$_{0.2}$NiO$_2$ film grown on the SrTiO$_3$ substrate (space group P4/mmm, lattice parameters $a = 3.91$ Å and $c = 3.37$ Å) [1]. Following the literature, to avoid the numerical instabilities arising from the Nd 4$f$ electrons we focus on La$^{3+}$ instead of Nd$^{3+}$ (4$f^3$) ion [5,11,15]. (Hereafter, we assume La by saying Nd in our calculations.) To explore the effect of Sr-doping on the electronic structure of (Nd,Sr)NiO$_2$ we employ a rigid-band shift of the Fermi level within DFT. In our DFT+DMFT calculations we explicitly include the Ni 3$d$, Nd 5$d$, and O 2$p$ valence states, by constructing a basis set of atomic-centered Wannier functions within the energy window spanned by these bands [22]. This allows us to take into account a charge transfer between the partially occupied Ni 3$d$, Nd 5$d$, and O 2$p$ states, accompanied by the strong on-site Coulomb correlations of the Ni 3$d$ electrons. We use the continuous-time hybridization expansion (segment) quantum Monte Carlo algorithm in order to solve the realistic many-body problem [24]. We take the average Hubbard $U = 6$ eV and Hund’s exchange $J = 0.95$ eV as previously employed for rare-earth nickelates NiO$_2$ [25]. We use the fully localized double-counting correction, evaluated from the self-consistently determined local occupations, to account for the electronic interactions already described by DFT.

In Fig. 1 we display our results for the $k$-resolved spectra of paramagnetic (PM) (Nd,Sr)NiO$_2$ obtained by...
DFT+DMFT as a function of Sr doping $x$. Overall, our results agree well with those published previously [11][15][18]. For $x = 0$ we observe a band formed by the strongly mixed Ni and Nd $3z^2 - r^2$ states crossing the Fermi level near the $\Gamma$ point. Upon Sr doping these states are seen to shift above the Fermi level, resulting in a change of the electronic structure of $(\text{Nd},\text{Sr})\text{NiO}_2$.

Our DFT+DMFT calculations reveal a remarkable orbital-selective renormalization of the partially occupied Ni $x^2 - y^2$ and $3z^2 - r^2$ bands (shown in Fig. 2). In particular, for $x = 0$ the Ni $x^2 - y^2$ states exhibit a large mass renormalization of $m^*/m \sim 3$, while correlation effects in the $3z^2 - r^2$ band are significantly weaker, $m^*/m \sim 1.3$. This behavior is consistent with sufficiently different occupations of the Ni $x^2 - y^2$ and $3z^2 - r^2$ orbitals. In fact, the $x^2 - y^2$ orbital occupancy for $x = 0$ is close to half-filling ($\sim 0.58$ per spin-orbit), while the $3z^2 - r^2$ orbitals are nearly fully occupied ($\sim 0.84$). In addition, our analysis of the local spin susceptibility $\chi(\tau) = \langle \hat{m}_z(\tau)\hat{m}_z(0) \rangle$ (see Fig. S1) suggests the proximity of the Ni $x^2 - y^2$ states to localization, while the Ni $3z^2 - r^2$ electrons are delocalized. Indeed, $\chi(\tau)$ for the Ni $3z^2 - r^2$ states is seen to decay fast to zero with the imaginary time $\tau$, which is typical for itinerant behavior. In contrast to that $\chi(\tau)$ for the $x^2 - y^2$ states is sufficiently larger, $\chi(0) = 0.72 \mu_B^2$, slowly decaying to $\sim 0.07 \mu_B^2$ as $\tau = \beta/2$. Our results therefore suggest that magnetic correlations in NdNiO$_2$ are at the verge of orbital-dependent formation of local magnetic moments [14]. In agreement with this the calculated (instantaneous) magnetic moment of Ni is about $\sqrt{\langle m_z^2 \rangle} \approx 1.1 \mu_B$, which is consistent with nearly a $S = 1/2$ state of nickel.

Upon hole doping the Ni 3$d$ occupations slightly decrease to 0.52 and 0.80 (per spin-orbital) for the Ni $x^2 - y^2$ and $3z^2 - r^2$ orbitals, respectively, for $x = 0.5$. This corresponds to a $\sim 0.17$ decrease of the total Wannier Ni 3$d$ occupation, whereas the Nd 5$d$ and O 2$p$ states occupancies drop by $\sim 0.21$ and 0.06. In addition, we observe a gradual decrease of mass renormalization of the $x^2 - y^2$ states to $m^*/m \sim 2.3$ at $x = 0.5$. In contrast to that for the $3z^2 - r^2$ orbital $m^*/m$ slightly increases to $\sim 1.5$. We notice no qualitative change in the self-energy upon changing of the Sr content $x$. The Ni 3$d$ states obey a Fermi-liquid-like behavior with a weak damping at the Fermi energy. Moreover, doping with Sr does not affect much magnetic moments in the paramagnetic phase of $(\text{Nd},\text{Sr})\text{NiO}_2$. Thus, the instantaneous magnetic moments $\sqrt{\langle m_z^2 \rangle}$ tend to increase only by about 5%. Interestingly in our model calculations (with absent self-consistency over the charge density, i.e., for the fixed tight-binding parameters of the DFT Wannier Hamiltonian) this increase is more significant, about 63%, suggesting the proximity to spin freezing, in accordance to recent model DMFT calculations [13].

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FIG. 2: Orbitally resolved quasiparticle mass enhancement $m^*/m$ together with the instantaneous $\sqrt{\langle m_z^2 \rangle}$ and fluctuating magnetic moments $M_{\text{loc}} = [T \int_0^{\beta/T} \langle \hat{m}_z(\tau)\hat{m}_z(0) \rangle]^{1/2}$ of Sr-doped NdNiO$_2$ calculated by DFT+DMFT for the paramagnetic state, at $T = 290$ K. $M_z$: DFT+DMFT results for magnetization per Ni site for the N´eel (111), C-type (110), and ($1\frac{1}{2}$) AFM states at $T = 290$ K.

FIG. 3: Quasiparticle Fermi surface of Sr-doped NdNiO$_2$ for Sr $x = 0$, 0.2, and 0.4 calculated by DFT+DMFT for the paramagnetic state at $T = 290$ K.
the dependence of the calculated FS’s as a function of Sr $x$. We note that our results for $x = 0$ are in qualitative agreement with previous band-structure studies \[5, 7\]. In particular, we obtain that the FS consists of three FS sheets, with the elliptical FS centered at the Brillouin zone (BZ) center (Γ point), originating from the mixed Ni 3$d$ and Nd 3$z^2 - r^2$ states. The electron FS pockets centered at the A-point are mainly of the Ni $xz/yz$ character. Similarly to the cuprates, the FS of NdNiO$_2$ is dominated by the quasi-two-dimensional (quasi-2D) holelike FS sheet with a predominant Ni $x^2 - y^2$ character, centered at the A-M BZ edge. In close similarity to the cuprates, our results for the FS topology imply an in-plane nesting with magnetic vector $q_m = (110)$ ($M$-point).

Upon increase of the Sr content, we observe a remarkable change of the electronic structure of (Nd,Sr)NiO$_2$ which is associated with an entire reconstruction of the FS topology, i.e., a Lifshitz transition. Thus, at $x = 0.2$ the elliptical FS centered at the Γ point vanishes. In addition, the holelike quasi-2D FS sheets at the top and the bottom of the BZ merge near the $R$ point to a quasi-3D electron-like FS that forms a neck at the top and the bottom of the BZ. Overall, this suggests that the Lifshitz transition is accompanied by a reconstruction of magnetic correlations in infinite-layer (Nd,Sr)NiO$_2$ that appears near to the experimentally observed doping Sr $x \simeq 0.2$.

We proceed with analysis of the symmetry and strength of magnetic correlations in (Nd,Sr)NiO$_2$. For this purpose we compute the momentum-dependent static magnetic susceptibility $\chi(q)$ within DFT+DMFT using the particle-hole bubble approximation. Orbital contributions of $\chi(q)$ along the BZ path and their dependence on the Sr content $x$ are shown in Fig. 4. Our results for the total $\chi(q)$ as a function of Sr doping $x$ are summarized in Fig. 5. Interestingly for $x = 0$ our results for $\chi(q)$ exhibit two well defined maxima at the $M$ and $A$ points of the tetragonal BZ. This suggests the existence of (at least) two leading magnetic instabilities due to the Ni $x^2 - y^2$ states (for $x = 0$) with a wave vector near to $q_m = (110)$ and (111), that corresponds to the $C$-type and the Néel AFM ordering, respectively. In the same time $\chi(q)$ for the $3z^2 - r^2$ states is seen to be small and nearly $q$-independent. We notice that $\chi(q)$ appears to be somewhat larger in the $A$ than that in the $M$ point. We therefore expect that the three-dimensional Néel AFM state is more energetically favorable than the quasi-2D $C$-type (for Sr $x = 0$). In fact, this qualitative analysis agrees well with our total-energy calculations within the spin-polarized DFT and DFT+DMFT methods (see Fig. S3). Both reveal that for Sr $x = 0$ the Néel AFM ordering is more energetically favorable by about 3-4 meV/f.u. with respect to the $C$-type AFM and the PM state within DFT+DMFT, at $T = 290$ K. We note that within DFT the Néel and the staggered dimer ($11\frac{1}{2}$) and $C$-type (110) states are differ by about 5-7 meV/f.u., while the non-magnetic state appears much above, by about 85 meV/f.u.

Our results for $\chi(q)$ and total energies suggest that various types of spin order are competing (nearly energetically degenerate) in (Nd,Sr)NiO$_2$. Indeed, for Sr $x = 0.2$, $\chi(q)$ is seen to be nearly flat and degenerate at around the $M$ and $A$ points (see Fig. 4), implying possible frustration of the Ni 3$d$ moments. Upon further increase of Sr $x$, our results provide a clear evidence of an entire reconstruction of magnetic correlations, with the $3z^2 - r^2$ states now playing a major role. While for Sr $x = 0.4$ $\chi(q)$ for the $x^2 - y^2$ orbital is seen to be nearly flat (degenerate for different $q_i$), suggesting in-plane frustration of the Ni 3$d$ moments. The out-of-plane $3z^2 - r^2$ orbital contribution reveals a flat maximum near the $M$ point.

![Graph](https://example.com/graph.png)

**FIG. 4:** Orbitally resolved static spin susceptibility $\chi(q)$ of Sr-doped NdNiO$_2$ calculated by DFT+DMFT at $T = 290$ K.

![Graph](https://example.com/graph.png)

**FIG. 5:** Long-range ordered magnetic moments of Ni as a function of hole doping calculated for NdNiO$_2$ by DFT (empty symbols), DFT+DMFT results for the Néel, $C$-type (110), single stripe (100), and staggered dimer (11$\frac{1}{2}$) AFM states at $T = 290$ K are shown by filled symbols.

In Fig. 5 we show our results for the long-range ordered magnetic moments of nickel calculated within the spin-polarized DFT and DFT+DMFT. The latter are about 0.67 $\mu_B$/Ni as obtained by DFT+DMFT for the Néel (111), $C$-type (110), and staggered dimer (11$\frac{1}{2}$) AFM states for Sr $x = 0$, at $T = 290$ K. Notably, we observe a sharp suppression of the calculated magnetization $M_z$ and hence of the Néel temperature evaluated from the spin-polarized DFT+DMFT calculations with Sr $x$. In particular, for Sr $x = 0.2$ we find no evidence for a mag-
netically ordered state at $T \geq 290$ K. Thus, all magnetic configurations discussed here, namely, the (100), (110), (111) and (11 $\frac{1}{2}$) AFM and FM configurations collapse in the PM state. That is, for Sr $x = 0.2$ the Néel (Curie) temperature is much below the room temperature that suggests rising of quantum spin fluctuations with $x$.

We note, however, that analysis of the finite-temperature DFT+DMFT results may often be problematic. For example, the single stripe (100) AFM and ferromagnetic orderings are found to be unstable at $T = 290$ K, i.e., both collapse to the PM state. We therefore first perform the spin-polarized DFT calculations of the ground state energy differences between different magnetic states (see Fig. [S3]). In fact, the DFT calculations give qualitatively similar results to those obtained by DFT+DMFT with significantly larger values of the total energy difference (with respect to the non-magnetic state) of $\sim 85$ meV/f.u., and $8$ meV/f.u. for the $C$-type, Néel, and staggered dimer (11 $\frac{1}{2}$), and single stripe (100) magnetic states, respectively. Both spin-polarized DFT and DFT+DMFT calculations reveal a near degeneracy of various types of spin orders, implying frustration of magnetic correlations in (Nd,Sr)NiO$_2$. The latter is most notable for the Sr content of about $x \approx 0.2-0.3$, which is close to the experimental Sr doping $x \approx 0.2$. Moreover, the calculated magnetization for the various AFM states tends to decrease in both the DFT and DFT+DMFT calculations upon increase of Sr $x$ (see Fig. [5]). In addition, within DFT+DMFT magnetization is found to sharply collapse to the PM state for Sr $x > 0.1$ at $T = 290$ K, suggesting a sharp increase of spin fluctuations with $x$.

Our results point out an anomalous sensitivity of the electronic structure and magnetic correlations of (Nd,Sr)NiO$_2$ with respect to the Sr $x$ doping. In particular, we found a remarkable frustration of (orbital-dependent) magnetic moments of Ni sites near to the optimal Sr doping $x \approx 0.2$. To help check these results, we computed magnetic exchange couplings within the spin-polarized DFT and DFT+DMFT using the magnetic force theorem [26]. Our findings for the Néel AFM state are summarized in Fig. [6]. We observe that for Sr $x = 0$ the interlayer couplings $J_{1}$ is small and weakly antiferromagnetic, $J_{\perp} \sim -23$ K [27]. The in-plane couplings $J_{1}$ (nearest-neighbor) and $J_{2}$ (next-nearest-neighbor) are both antiferromagnetic and are sufficiently higher by modulus, $\sim 198$ K and $-45$ K, respectively. Interestingly that for pure NdNiO$_2$, $|J_{1}| = 198$ K is comparable to that found experimentally in infinite-layer CaCuO$_2$. [3]. Most importantly, our results reveal a remarkable change of the $J_{2}/J_{1}$ ratio with respect to Sr $x$, which is increasing from $\sim 0.36$ to 0.56 for $x = 0.1-0.3$, i.e., near to the experimental doping Sr $x \approx 0.2$. While in DFT+DMFT magnetization is found to quickly collapse to the PM state for Sr $x > 0.1$, the exchange couplings evaluated from the spin-polarized DFT+DMFT calculations do follow the same trend, with $J_{2}/J_{1} \approx 0.26$ for Sr $x = 0$, which is found to increase to 0.87 for Sr $x = 0.1$.

Our findings resemble us the behavior of the spin-1/2 frustrated $J_{1}$-$J_{2}$ Heisenberg model on the two-dimensional (2D) square lattice, with an unusual quantum spin liquid ground state to appear in the highly frustrated region $J_{2}/J_{1} \approx 0.4-0.5$, sandwiched between the Néel and stripe type (or valence-bond solid) ordered states [28]. This analogy is very striking, taking into account our results for the change of the electronic structure and magnetic couplings $J_{2}/J_{1}$ ratio in (Nd,Sr)NiO$_2$ with Sr $x$. Thus, the frustration region is sandwiched between the two different (long- or short-range ordered) antiferromagnets [28]. We find that magnetic couplings in (Nd,Sr)NiO$_2$ near to the optimal doping demonstrate an unanticipated frustration, which suppresses a long-range magnetic order (resulting in a drastic drop of the Néel temperature), and can lead to formation of unusual quantum spin liquid ground states. Moreover, our results suggest that frustration is maximal for Sr-doping $x = 0.1-0.2$ that corresponds to the highly frustrated region of the spin-1/2 frustrated $J_{1}$-$J_{2}$ Heisenberg model. Overall, our results suggest the importance of in-plane spin fluctuations to explain superconductivity in (Nd,Sr)NiO$_2$, in contrast to the previous claims [1]. We point out that strong frustration of magnetic interactions in (Nd,Sr)NiO$_2$ suggests that superconductivity in infinite-layer (Nd,Sr)NiO$_2$ appears to be similar to that observed in iron chalcogenides and pnictides [29].

In conclusion, we employed the DFT+DMFT computational approach to study the effects of electronic correlations and Sr-doping on the electronic structure and magnetic properties of (Nd,Sr)NiO$_2$. We show that upon hole doping it undergoes a Lifshitz transition of the Fermi surface which is accompanied by a reconstruction of magnetic correlations. Most importantly, magnetic interactions in (Nd,Sr)NiO$_2$ are found to demonstrate an unanticipated frustration. We find that frustration is maximal for Sr-doping $x = 0.1-0.2$ that nearly corresponds to the experimentally observed doping value of (Nd,Sr)NiO$_2$. Our results for (Nd,Sr)NiO$_2$ reveal a fea-

![FIG. 6: Exchange interaction parameters (in-plane nearest-neighbor (NN) $J_1$, next-nearest-neighbor $J_2$, 3-rd NN $J_3$, 4-th NN $J_4$ and interlayer coupling $J_{1\perp}$) of Sr-doped NdNiO$_2$ calculated within spin-polarized DFT (empty symbols) and DFT+DMFT (filled symbols).]
ture that is central to copper oxides as well as to iron chalcogenides and pnictides – large in-plane spin fluctuations. We propose that superconductivity in nickelates is strongly influenced, or even induced, by in-plane spin fluctuations.

Acknowledgments

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[27] Here we adopt the following notation for the Heisenberg model
\[ H = -\sum_{ij} J_{ij} \mathbf{e}_i \cdot \mathbf{e}_j \]
where \( \mathbf{e}_{i,j} \) are the unit vectors.

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**Supplementary Material**

![Supplementary Material](image_url)

**FIG. S1:** Orbitally resolved local spin correlation functions \( \chi(\tau) = \langle \hat{m}_z(\tau) \hat{m}_z(0) \rangle \) of Sr-doped NdNiO\(_2\) as a function of hole doping Sr \( x \) calculated by DFT+DMFT at \( T = 290 \) K.

**FIG. S2:** Static spin susceptibility \( \chi(q) \) of Sr-doped NdNiO\(_2\) as a function of hole doping Sr \( x \) calculated by DFT+DMFT at \( T = 290 \) K.
FIG. S3: Total energy difference $\Delta E = E_{\text{mag}} - E_{\text{NM}}$ between the long-range magnetically ordered and non-magnetic states of NdNiO$_2$ as a function of hole doping calculated by DFT (empty symbols). Ferromagnetic (FM), Néel (111), C-type (110), (101) and single stripe (100), and staggered dimer (11½) states are shown. DFT+DMFT results for the total energy difference between the Néel and C-type AFM states and the paramagnetic state at $T = 290$ K are depicted by filled symbols.