The doping-dependent character and possible magnetic ordering of NdNiO$_2$

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The novel nickelate superconductors of infinite-layer type feature challenging electronic peculiarities in the normal-state phase diagram with doping. Distinct many-body behavior and different dispersion regimes of the entangled \{Ni-$d_{x^2-y^2}$, Ni-$d_{z^2-y^2}$\} orbital sector give rise to highly rich physics, which is here studied for the case of the NdNiO$_2$ system. An analysis based on advanced realistic dynamical mean-field theory unveils that the superconducting hole-doped region is the meeting place of a (self-)doped Mott insulator from the underdoped side, and a bad Hund metal from the overdoped side. Fermi-level crossing of the Ni-$d_{x^2}$ flat-band ties both regimes together to form a singular arena for unconventional superconductivity. We furthermore shed light on the intriguing problem of elusive magnetism in infinite-layer nickelates. Antiferromagnetic (AFM) order with small Ni moments is shown to be a vital competitor at low temperature. At stoichiometry, C-AFM order with ferromagnetic spin-alignment along the c-axis benefits from a conceivable coexistence with Kondo(-lattice) screening.

I. INTRODUCTION

Nickelate superconductivity in thin films of Sr-doped NdNiO$_2$ below a critical temperature $T_c \sim 10$ K entered the scene in summer 2019. Recently, the same physics has also been identified in Sr-doped PrNiO$_2$ thin films. Because of the sophisticated sample preparation and the so far lack of bulk single crystals, experimental progress on these Ni($3d^9$)-based infinite-layer systems is challenging.

Several findings still enable further insight into a very rich phenomenology. Ordered magnetism is hard to measure in the thin-film geometry, but neutron scattering on polycrystalline bulk samples provides no clear evidence for long-range antiferromagnetic (AFM) order in NdNiO$_2$. However, recent nuclear-magnetic-resonance (NMR) experiments on similar samples suggest quasistatic AFM ordering for $T < 40$ K. This, together with a resistivity upturn below $T \sim 70$ K, points to intriguing low-energy physics even far from the superconducting region. The latter is sandwiched between weakly-insulating doping regions, in the range $0.125 \lesssim x \lesssim 0.25$ of the phase diagram for hole doping $x$ of Sr kind. Doping-dependent Hall data point to a two-band scenario with $x$. First spectroscopic results reveal hybridization of Ni($3d$) states with other delocalized states and reduced Ni-O intermixing compared to NiO and LaNiO$_2$. Furthermore for finite $x$, substantial Ni($3d^9$) character is unveiled, in contrast to the dominant Cu($3d^9$-$L$), i.e. Cu($3d^9$) with ligand hole on oxygen, weight in high-$T_c$ cuprates. This Ni($3d^9$) signature is either attributed to multi-orbital physics or to Ni($S = 0$) single-orbital physics of $d_{z^2-y^2}$ kind. Concerning the superconducting state, a small anisotropy in the upper critical field and hints to a Pauli-limited coupling, as well as to a two-symmetry gap nature are reported.

From the theoretical side, assessments based on density functional theory (DFT) highlight the self-doping (SD) nature of an otherwise seemingly cuprate-like band structure. The weakly-filled SD band originates from Nd($5d$)-Ni($3d$) hybridization and causes a multi-sheeted Fermi surface at stoichiometry. There seems also agreement that the charge-transfer character of NdNiO$_2$ is reduced compared to estimates in akin cuprates. Further beyond DFT, two main theory concepts are pursued: either (decorated) single-Ni-orbital physics of Ni-$d_{x^2-y^2}$ type, or multiorbital Ni($3d$) processes are assumed key to the correlation phenomenology. In two previous works, we showed that based on calculations utilizing the combination of DFT with dynamical mean-field theory (DMFT) and furthermore including explicit Coulomb interactions on oxygen, the relevance of Ni-multiorbital degrees of freedom is indeed inevitable. Besides the indisputable Ni-$d_{x^2-y^2}$ orbital and its related low-energy band, the Ni-$d_{x^2}$ orbital plays a further crucial role. First, in mediating a Kondo(-lattice) coupling between Ni($3d$) and Nd($5d$) at stoichiometry. And second, for establishing a unique flat-band scenario in the doping region of low-$T$ superconductivity.

In the present work, the goal is to extend our previous studies and shed further light onto the Ni-multiorbital physics in infinite-layer nickelates. In Refs. 18 and 19, paramagnetism has been assumed and doping has been addressed by a supercell description as well as within a minimal three-band model solved for a simplified Fermi-liquid setting. Here, we provide a detailed account of the doping-dependent features of the correlated electronic structure based on the virtual-crystal approximation, and additionally investigate the intriguing problem of magnetic ordering in the NdNiO$_2$ system.

II. METHODOLOGY

The charge self-consistent combination of DFT and DMFT with the additional incorporation of the self-interaction correction (SIC) is used to investigate the normal-state properties of pristine and doped neodium nickelate in the infinite-layer structure. The realistic part of this DFT+SICDMFT scheme builds up on a mixed-basis pseudopotential representation in the
local density approximation (LDA). Coulomb interactions on oxygen are addressed beyond DFT within SIC applied to the O pseudopotential. While the O(2s) orbital is fully corrected with weight factor $w_p = 1.0$, the reasonable choice $w_p = 0.8$ is used for the O(2p) orbitals.

The Nd(4f) states are put in the pseudopotential frozen core, since they are not decisive for the key physics of infinite-layer nickelates. This also means, that f-electron based magnetism is not considered in this work. As in many other comparable compounds, due to the local nature of the 4f moments and their weak exchange coupling, magnetic ordering of Nd moments is believed to occur at very low temperatures. Note furthermore that the $j$-resolved frozen occupation of the Nd(4f) shell is here chosen with small resulting moment. The role of spin-orbit effects in the overall crystal calculations is neglected. Additional details for the computation of the magnetically ordered phases in this work will be given in section IV.

To facilitate the description at finite doping, we here employ the virtual-crystal approximation (VCA) in the fully charge self-consistent scheme. This amounts to replace the Nd atom by a pseudo atom of nuclear (and electronic) charge $Z = Z_{Nd} - \delta$ in the pristine unit cell. As a choice, the charge content of the Nd(4f) shell is here not modified by this construction. It is then assumed that $\delta = x$ holds, i.e. the VCA doping $\delta$ mimics the true Sr doping $x$ of effectively replacing Nd$^{3+}$ by Sr$^{2+}$. As any theoretical doping description, also the VCA misses certain effects, e.g. local-structural distortions or correct local charge states of the impurity. But it usually provides a faithful description of the overall effects of charge doping. Note that in the previous works Refs. [18] and [19] the other two standard options to describe doping, namely supercell[19] and chemical-potential shift[23] have been put into practice. Thus the present perspective completes the conventional method spectrum in that regard.

The DMFT correlated subspace is governed by a full Slater Hamiltonian applied to the Nd(3d) projected-local orbitals. The projection is performed on the $6 + 5 + 1 = 12$ Kohn-Sham states above the dominant O(2s) bands, associated with O(2p), Ni(3d) and the self-doping band. A Hubbard $U = 10$ eV and a Hund exchange $J_H = 1$ eV prove reasonable for this choice of the energy window in the given nickelate[25][19]. Continuous-time quantum Monte Carlo in hybridization expansion[24] as implemented in the TRIQS code[25][19] is utilized to handle the DMFT problem. The fully-localized-limit double-counting scheme[20] is applied. Maximum-entropy and Padé methods are employed for the analytical continuation from Matsubara space onto the real-frequency axis. Stoichiometric lattice parameters are taken from experiments, reading $a = 3.92$ Å and $c = 3.31$ Å. The system temperature is set to $T = 30$ K in order to properly approach the competition/competition between quasiparticle (QP) formation, Mott criticality, Kondo physics and magnetic order in these challenging nickelates. If not otherwise stated, all shown data is obtained within charge-selfconsistent DFT+sicDMFT.

The inclusion of explicit oxygen-based correlations beyond conventional DFT+DMFT for the NdNiO$_2$ system is believed necessary for mainly two reasons. First generally, in nickelates as late transition-metal (TM) oxides the charge-transfer aspect is stronger as in early TM oxides, and therefore Coulomb interactions from O(2p) will have a larger impact. Second, while Ni-$d_{x^2-y^2}$ is strongly hybridized with O(2p) in the infinite-layer structure, Ni-$d_{z^2}$ is less so since there is no apical oxygen (see Fig. 1). Thus especially the Ni-$e_g \{z^2, x^2 - y^2\}$ orbitals of the DMFT-active Ni(3d) shell are quite differently affected by O(2p) and therefore it should be insufficient to treat the Coulomb interactions from oxygen only implicitly via a chemical-potential or Coulomb-on-Ni shift. In the Appendix we provide a brief comparison between the DFT+sicDMFT and DFT+DMFT picture of pristine and doped NdNiO$_2$.

III. PARAMAGNETIC SYSTEM

A. Stoichiometric compound

A basic characterization of the correlated electronic structure of paramagnetic (PM) NdNiO$_2$ has been given in Ref. [18] for ambient temperatures, and in Ref. [19] for the lower-temperature regime. To set the stage, we here recapitulate the main findings and discuss the different site/orbital contributions within the correlated regime in some more detail.

On the DFT(LDA) level, the $x^2-y^2$ orbital has the lowest crystal-field level within the Ni(3d) sector and the energy splittings to the other levels read \( \{\varepsilon_{x^2}, \varepsilon_{y^2}, \varepsilon_{x^2-y^2}, \varepsilon_{xz}, \varepsilon_{yz}, \varepsilon_{xy}\} = \{356, 0, 286, 286, 536\} \) meV. Then in essence, the compound is a self-doped Mott insulator in DFT+sicDMFT, whereby the effectively Mott-insulating states are of half-filled Ni-$d_{x^2-y^2}$ kind and the self doping originates from a weakly-occupied band resulting from the hybridization between Nd(5d) and

![FIG. 1. (color online) Projected local orbitals of Ni-$d_{z^2}$ (left) and Ni-$d_{x^2-y^2}$ (right) type in the NdNiO$_2$ compound: Ni(grey), Nd(large, green), O(small, red).](image-url)
Ni(3d). Thus there is no Ni-$d_{x^2-y^2}$ quasiparticle dispersion crossing the Fermi level $\varepsilon_F$ in the k-resolved spectral function, as shown in Fig. 2a. At low energy, there are only SD-band based electron pockets at $\Gamma$ and A in the Brillouin zone.

Correspondingly, orbital weights along the given high-symmetry lines are provided in Fig. 2b-f for the most-relevant orbital sectors. The Ni-$d_{x^2}$ contribution is twofold. First, it forms an incoherent part around -3 eV for $k_z = 0$, which becomes more coherent, flat and intense in the $k_z = 1/2$ plane. Second, it has its prominent share in the SD-band formation for the $\Gamma$-pocket part. On the other hand due to Mottness, the Ni-$d_{x^2-y^2}$ weight remains largely incoherent throughout the chosen k-space regions, with the exception of a small part along Z-R in the regime of the overall Ni-$d_{x^2}$-dominated spectrum. The Ni-$t_{2g}$ $\{xz, yz, xy\}$ orbitals cause dispersing features between $[-3, -2]$ eV and have weak contribution to the SD band around A. The Nd($5d$)-based orbitals of $z^2$ and $xy$ character are the main contributors to the SD band from the rare-earth site; Nd-$d_{xy}$ for the A pocket and Nd-$d_{xz}$ pretty much everywhere else. The hybridization between Nd-$d_{xy}$ and Ni-$t_{2g}$ is also evident from the joint weight at deeper energies. The coupling between Ni-$d_{x^2}$ and Nd-$d_{xz}$ is mostly confined to the $\Gamma$-X-M-Z path. Mott-insulating Ni-$d_{x^2-y^2}$ appears disconnected from the Nd($5d$)-Ni(3$d$) hybridization, and indeed nearest-neighbor hopping from both relevant Nd($5d$) orbitals to the in-plane Ni-$e_g$ orbital is zero. The O(2$p$) contribution is surely also vital in the given energy window, but a k-resolved discussion appears not highly instructive (see Ref. 18 for the k-integrated weight).

With high spectral-resolution focus on the low-energy

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**FIG. 2.** (color online) Spectral information for pristine NdNiO$_2$. (a) k-resolved function $A(k, \omega)$ along high-symmetry lines: $\Gamma$-X-M describes a triangle in the $k_z = 0$ plane and Z-R-A marks the same triangle translated to the $k_z = 1/2$ plane. Above the Fermi level, only the single SD dispersion is shown and further dispersions discarded in the plot. (b-f) k-resolved orbital weights (i.e. fatbands) for the given spectral function: (b) Ni-$d_{z^2}$, (c) Ni-$d_{x^2-y^2}$, (d) Ni-$t_{2g}$, (e) Nd-$d_{xz}$ and (f) Nd-$d_{xy}$. Note that with this chosen intensity resolution, the non-dispersing-level feature at $\varepsilon_F$, giving rise to a Kondo(-lattice) mechanism at lower $T$, is not visible in $A(k, \omega)$.

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**FIG. 3.** (color online) Effective Ni(3$d$) occupation in doped NdNiO$_2$ from introducing $\delta$ holes/electrons. (a) Difference $n_{d,0} - n_d$ between the total occupation at $n_{d,0}$ zero doping and at given doping $n_d$, versus $\delta$. The dashed red line marks the regime where the complete doping charge is transferred to Ni(3$d$). (b) Orbital-resolved filling of the Ni-$e_g$ states.
region, a single-level feature at the Fermi level would become additionally visible for the here chosen lower temperature regime. It is stemming from Ni-$d_{x^2-y^2}$ and is part of an intriguing Kondo(-lattice) mechanism, involving furthermore Ni-$d_{z^2}$ and Nd-$d_{z^2}$. Basically, the Ni-$d_{z^2}$ orbital mediates a Hund-assisted Kondo coupling between the $d$ sectors of Ni and Nd, by enabling fluctuations between Ni-$e_g$ and weakly-correlated Nd-$d_{z^2}$.

B. Doped compound

Superconductivity in the nickelate thin films is reached by Sr doping, expected to introduce holes to the system. For completeness we here discuss both, electron and hole doping within the VCA framework. From a preparation perspective, electron doping, e.g., by substituting Nd$^{3+}$ by Ce$^{4+}$, should be more challenging, the already delicate Ni$^+$ ionic state is reduced even further and the charge imbalance within the NiO$_2$ plane becomes additionally stretched. Indeed, the DFT+sicDMFT calculations show that the progress of the doping level $\delta$ toward the Ni(3$d$) shell is strongly hindered on the electron-doped side (see Fig. 5a). On the other hand on the hole-doped side, the doping originating on the rare-earth site is nearly exclusively transfered to the Ni site. This furthermore underlines the weaker charge-transfer character compared to cuprates, where most holes reside on oxygen. The orbital-resolved filling of the Ni-$e_g$ states with doping $\delta$ in Fig. 5b exhibits the strong hole attraction of Ni-$d_{z^2}$ and its weaker additional electron filling on the electron-doped side. On the contrary, the Ni-$d_{x^2-y^2}$ filling is only weakly affected by the charge doping of the system and remains nearly inert. All these observations are in line with previous findings in a minimal-Hamiltonian description. Note that the Ni-$t_{2g}$ states are always close to complete filling throughout the doping landscape and seemingly do not play a key role for the phase diagram with $\delta$.

We now turn to the $k$-resolved spectral evolution with doping, displayed in Fig. 4. Electron doping mainly leads to an expected downshift of the overall spectrum, resulting in an increased filling of the SD band. The Mott-insulating character of the Ni-$d_{x^2-y^2}$ orbital remains apparently robust, though the corresponding self-energy is considerably weakened with electron doping (cf. lower-left part of Fig. 5). Note however that this weakening proceeds even stronger with hole doping (cf. lower-right part of Fig. 5), as observed already in the minimal-Hamiltonian picture. It leads to a partial melting of

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**FIG. 4.** (color online) Spectral function $A(k, \omega)$ with doping $\delta$. Top row, from left to right: $\delta = -0.30, -0.15, -0.05, 0$. Bottom row, from left to right: $\delta = 0.05, 0.15, 0.30, 0.45$. Same setting as in Fig. 2a.

**FIG. 5.** (color online) Imaginary part $\text{Im} \Sigma(i\omega_n)$ of the Ni-$e_g$ self energy on the Matsubara axis $\omega_n = (2n + 1)\pi T$ for different dopings $\delta$. Top: Ni-$d_{x^2}$ and bottom: Ni-$d_{x^2-y^2}$. Left part: $\delta = -0.30, -0.15, -0.05, 0$ and right part: $\delta = 0.05, 0.15, 0.30, 0.45$. 
FIG. 6. (color online) Spectral information of hole-doped NdNiO$_2$ with $\delta = 0.15$. (a-f) As in Figs. 2a-f. (g) Constant-energy surfaces in the $k_z = 1/2$ plane. Note that the maximum intensity for the surfaces in (g) is reduced by a factor of three compared to the one for $A(k, \omega)$ in (a).

The actual Fermi surface with obvious Ni-$d_{z^2}$ weight at the A point is shown on the left of Fig. 6g. Because of the flat-band character, the topology of the constant-energy surface changes quite significantly within a $\sim 10$ meV range below $\varepsilon_F$. Going down in energy from $\omega = 0$, a hole surface around A transforms into an electron surface around Z. The substantial hole character just below the Fermi level is in line with experimental findings of hole-like transport in that doping region$^{7,8}$. But note that the “actual” Fermi surface for a given $0.125 \lesssim \delta \lesssim 0.25$ in the region with low-T superconductivity is still very delicate, and perfect agreement between experiment and theory will be hard to reach. Nonetheless, we predict that the experimental $\delta = 0.15$ Fermi surface has the appearance of one of the three $\omega = [0, -0.045]$ eV plots of Fig. 6g.

The Ni-$e_g$ self-energies have a rather intriguing development with hole doping (cf. right part of Fig. 5). For Ni-$d_{z^2}$, the self-energy part $\text{Im} \Sigma(i\omega_n)$ grows with $\delta$, and the low-frequency curvature increasingly departs
The filling scenario and k-space topology behind the Ni-d$_{z^2}$ pseudogap structure for hole dopings beyond the superconducting region is one reason for weak metallicity, especially for in-plane transport, as already pointed out in Ref. 13. However, in addition, the non-Fermi-liquid and heavily bad-metallic character of the Ni-d$_{z^2}$-dominated states, associated with the pseudogap structure and a specific Hund-metal scenario of Ni-e$_g$, is a second vital reason.

Let us finally note, that the present behavior with hole doping does not seem perfectly commensurable with the experimental phase diagram. There, the superconducting region stretches from 0.125 $\lesssim \delta \lesssim$ 0.25, whereas the prominent role of Ni-d$_{z^2}$ seemingly peaks here at $\delta$ = 0.30. There may be several reasons for this difference. First, due to our sparse doping sampling, a ‘true’ peaking Ni-d$_{z^2}$ behavior within the experimental superconducting doping range is still possible, as for $\delta$ = 0.30 there is already the pseudogap onset. Second, we only assume $\delta$ = x to hold, but since the present VCA approach has its limitations, a small shift of behavior would not be surprising. Moreover throughout the study, the stoichiometric lattice parameters are employed, and e.g. the c-axis increase with hole doping is not included in the calculations. And third, albeit the Ni-d$_{z^2}$ flat band neighborhood to the Fermi level is believed to be the key driving force for superconductivity, its $\varepsilon_F$ entry and departure points do not necessarily have to agree exactly with the phase boundaries for superconductivity.

**IV. ANTFERROMAGNETIC SYSTEM**

A key feature of structurally akin high-$T_c$ cuprates is the prominent AFM phase at and near stoichiometry. So far, static magnetic order in infinite-layer nickelates has been elusive in experiment, but the investigations point to a low-temperature behavior that deviates from straightforward paramagnetism.

From theory, there are various studies that address ordered magnetism in these systems. Besides more unlikely ferromagnetic (FM) order, the canonical orderings examined are G-type and C-type anti-

| magnetic order | $m_{Ni} (\mu B)$ | $E^{AFM}_{tot} - E^{FM}_{tot}$ (meV/atom) |
|---------------|------------------|------------------------------------------|
| $\delta = 0$  |                  |                                          |
| G-AFM         | 0.076, 105       | 0.059, 126                               |
| C-AFM         | 0.081, 20        | 0.000, –                                 |

**TABLE I.** Ordered Ni magnetic moment in the G-AFM and C-AFM phase at $T = 30$ K and energy difference to the PM phase.
ferromagnetism. While the G-AFM order displays antiparallel alignment between in-plane and c-axis nearest-neighbor (NN) Ni spins, the C-AFM order deviates therefrom in showing ferro-alignment along the c-axis (see Figs. 8a,b). Most results from DFT(+)U and model-Hamiltonian DMFT calculations reveal a large ordered magnetic moment of $m_{Ni} \sim 1 \mu_B$ on the Ni site for these patterns. At stoichiometry, the C-AFM state appears energetically favorable against the nonmagnetic state and against FM order within DFT(+)U and additionally against G-AFM when utilizing the SCAN (i.e. meta generalized-gradient approximation) functional in DFT\cite{SCAN}. However note that electronic correlations are only treated on a static level in these approaches (e.g. there is no true PM state straightforwardly reachable), and furthermore they formally describe the system at $T = 0$. Nearest-neighbor in-plane exchange constants between Ni spins are estimated 65 meV\cite{DFT+U} and 82 meV\cite{DFT(+)U}, i.e. roughly half the value usually revealed for high-$T_c$ cuprates.

Here we report DFT+sicDMFT calculations for possible G-AFM and C-AFM ordering at $T = 30\,K$. Two technical aspects have to be mentioned. First, the possible additional ordering of the Nd spins is not considered in this investigation. This is not unreasonable, as in most rare-earth compounds the corresponding 4$f$-based spins order at much lower temperature. Second, in a hybrid scheme of DFT and DMFT, there are in principle two basic formalisms to handle spin polarization: either it is treated both in the DFT and the DMFT part, or only on the DMFT level. The former approach includes ligand-based exchange mechanisms more directly, but the $T$-independent exchange splitting from the DFT part usually ruins a reliable assessment of temperature effects. In other words, especially for transition-metal oxides the robustness of the ordered magnetic moments with temperature is most often severely overestimated. Therefore, spin polarization is here treated only on the DMFT level, i.e. the Ni(3$d$) self-energies are solely exchanged with temperature.

With this in mind, let us discuss the results by first stating that the calculations are quite demanding. For instance, we did so far not succeed to stabilize a magnetically ordered phase in electron-doped NdNiO$_2$. At stoichiometry and with hole doping where this proves possible, the Ni magnetic moments turn out generally very small, with values $m_{Ni} \lesssim 0.1 \mu_B$ (see Tab. I). But importantly, the differentiation between an actual ‘zero moment’ and such a small one is always very manage-

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**FIG. 8.** (color online) Comparing PM and AFM-ordered NdNiO$_2$. (a) G-AFM ordering and (b) C-AFM ordering. (c) Total (top) and local Ni-$\epsilon_p$ (bottom) spectral function for PM, G-AFM and C-AFM phase, respectively. (d-f) k-resolved spectral function along high symmetry lines for G-AFM (d), C-AFM (e) and PM (f) phase. The $\sqrt{2} \times \sqrt{2}$ enlarged in-plane unit cell translates into X$\leftrightarrow$M and R$\leftrightarrow$A for the (G,C)-AFM structure. And the upper Ni-$d_{x^2}$ flat-band part is flipped with respect to $k_z$ for the G-AFM structure, due to the doubled unit cell along c.
Fig. 9. (color online) Comparison of PM and G-AFM low-energy spectral functions at doping level $\delta = 0.15$. (Top) Total spectrum and (bottom) local Ni-\(e_g\) spectrum.

As an exotic phase with coexistence of antiferromagnetic ordering and Kondo-(lattice) screening\textsuperscript{63–66}. Because of the weak filling of the the SD band, the latter Kondo physics takes place in an underscreening scenario, leaving room for magnetic ordering. Additionally, since the Kondo screening is mainly mediated by the \(d_{x^2-y^2}\) orbitals of Ni and Nd\textsuperscript{19}, the ferro-alignment of spins along the c-axis in the C-AFM phase enables a screening action without additional spin flip. This may explain as to why the G-AFM phase cannot easily coexist with Kondo screening.

For finite hole doping $\delta = 0.15$, the C-AFM phase has transformed into the PM phase with zero ordered Ni moment. On the contrary, the G-AFM phase still remains (meta)stable with somewhat reduced $m_N$ compared to $\delta = 0$ (see Tab. I). Notably, as the SD pockets are shifted above the Fermi level for that hole-doping level, partial Kondo screening of the pristine-scenario kind has vanished. Figure 8 shows that close to the Fermi level, the Ni-\(d_{x^2}\) character is enhanced and the Ni-\(d_{x^2-y^2}\) one suppressed in the doped G-AFM phase compared to the PM phase. Though energetically less favorable (see Tab. I), if NN-AFM ordering is indeed stable at low temperature and with doping, there would be a $\delta$-induced transition from C-AFM to G-AFM ordering. Note that a qualitative change of magnetic order with doping has also been deduced by Leonov and Savrasov\textsuperscript{23} from DFT+DMFT calculations for the system at ambient temperature, albeit differently with G-AFM order at stoichiometry and C-AFM order at finite hole doping.

V. SUMMARY AND DISCUSSION

The present DFT+sicDMFT study of pristine and doped NdNiO$_2$ uncovers two key results. First, the character of the paramagnetic electronic structure changes distinctively with doping. On the electron-doped side, which may be challenging to explore experimentally, the nickelate is a (Ni,Nd)-\(d_{x^2}\) based one-band system in a Mott-critical background of Ni-\(d_{x^2-y^2}\) kind. At stoichiometry, the latter background is of robust Mott-insulating nature and the solid one-band part at the Fermi energy has evolved into a weakly-filled self-doping entity. Since the SD dispersion carries relevant Ni-\(d_{x^2}\) itinerancy, one may also picture this phase as of orbital-selective type, eventually decorated with partial Kondo screening at low temperature\textsuperscript{19}. Already small hole doping leads to a sizable upward shift of the SD band into the unoccupied region, and the stabilization of a Ni-\(d_{x^2-y^2}\)-based weakly-doped/pre-melted Mott-insulator in which Ni-\(d_{x^2}\) degrees of freedom are frozen. For hole doping $\delta \gtrsim 0.15$, the Ni-\(d_{x^2}\) flat-band part in the $k_z = 1/2$ region crosses the Fermi level and the system displays intriguing \{Ni-\(d_{x^2}\), Ni-\(d_{x^2-y^2}\)\} physics at low energy. As a result thereof, unconventional superconductivity sets in at low temperature. For even larger hole doping $\delta > 0.30$, the nickelate does not attain a conventional Fermi-liquid state, but enters a pseudogap and a bad Hund-metal
regime. In other words, the two-orbital Ni-$e_g$ subspace with filling $n_{e_g} \sim 2.5$ becomes strongly correlated with vanishing coherence scale due to the interplay of the Hund’s $J_H$ with the remaining local Coulomb interactions in the Ni(3$d$) and O(2$p$) shells. Take notice that other recent theoretical works mark general Nd$_1-x$Sr$_x$NiO$_2$ exclusively as a strong Hund metal, and suggest overall similarities with iron-pnictide physics. However from our point of view, the (near) Mott-insulating state of Ni-$d_{x^2-y^2}$ and large filling of Ni-$d_{z^2}$ close to stoichiometry render this somewhat questionable. As described above, strong Hund-metal physics occurs here only for larger hole dopings, and therefore loosely speaking, superconductivity in doped NdNiO$_2$ appears to take place when 'cuprates' (from the underdoped side) and 'pnictides' (from the overdoped side) meet and bind together through a flat-band scenario.

Second, it is shown that magnetic order of AFM type may be a vital competitor with and without doping at low-enough temperatures, albeit the associated features are far from those of conventional antiferromagnetism in e.g. stoichiometric high-$T_c$ cuprates. At $T = 30$ K, the ordered Ni moments are already very small for all encountered cases, which either points to a critical temperature in the same range or to strong quantum fluctuations/screening. The size of the local moment surely also depends on calculational settings, e.g. on the method to handle spin-symmetry breaking in realistic DMFT and the choice of the correlated subspace. But the present choices are believed physically sound, and therefore the general outcome of significantly reduced Ni moments around $T = 30$ K should be qualitatively robust. The C-AFM phase is energetically more favorable than the G-AFM phase at stoichiometry, obviously benefiting from a coexistence with partial Kondo screening. This result sounds rather exotic for a TM(3$d$) oxide, however the experimental searches for magnetic order in infinite-layer nickelates also point to singular low-$T$ behavior. Furthermore, we here detect an apparent transition from C-AFM to G-AFM order/correlations with hole doping. This would be coherent with the fact that the Kondo-screening tendencies at stoichiometry vanish with hole doping, because of the shift of the SD band above the Fermi level.

This work complements our previous investigation for infinite-layer nickelates. It is worthwhile to note that the obtained essential picture of relevant Ni multiorbital physics, and especially the importance of the Ni-$d_{z^2}$ flat-band part for hole dopings in the superconducting region, is very robust across these studies. For instance, three different frameworks to describe the doped NdNiO$_2$ compound, i.e. supercell with impurities, minimal Hamiltonian with chemical-potential shift and eventually the present VCA approach, all lead to the same qualitative result. Thereby, the role of explicit Coulomb interactions on oxygen is not of 'decorative' type, but truly decisive for the revealed physics (see Appendix for further details). When abandoning the SIC inclusion into the theoretical framework and employing conventional DFT+DMFT calculations, the Ni-$d_{z^2}$ flat-band part remains well below the Fermi level for the relevant dopings, in agreement with comparable studies of this type, and also the recent GW+EDMFT investigation of Petocchi et al., which includes beyond-Ni(3$d$)-based correlations, supports our picture.

Let us finally take the opportunity to selectively base with further theoretical studies from other research groups. In DFT+U studies by Choi et al., the C-AFM order also turns out to be a proper candidate for magnetism in NdNiO$_2$. Furthermore, notably at stoichiometry, the Ni-$d_{z^2}$ flat-band part in the $k_z = 1/2$ region is placed right at the Fermi level for this order within the given static-correlation scheme. Though at a first glance looking quite different to our result for the C-AFM phase (compare with Fig. 3), this outcome could carry similar physics. The DFT+U scheme may mimic Kondo(-like) physics by placing a spin-resolved flat band right at the Fermi level. In this way, the (corresponding (local) spin becomes highly susceptible, and thats more or less the best a static-correlation scheme may achieve for Kondo-based spin fluctuations. The fact that this band is of dominant Ni-$d_{z^2}$ character underlines our previous finding of the crucial Ni-$d_{z^2}$ role in mediating Kondo-screening in NdNiO$_2$. However, this DFT+U result for Ni-$d_{z^2}$ at stoichiometry should not be directly related to the DFT+sicDMFT result of the Ni-$d_{z^2}$ flat-band part crossing the Fermi level with sizable hole doping.

In a different DFT+U assessment by Liu et al., the G-AFM order is found stable at stoichiometry, but the Ni magnetic moment and the general AFM ordering is also easily weakened by the impact of the SB band. The resistivity upturn below $T \sim 70$ K is associated with the transition into this bad-AFM metal when lowering temperature from the PM phase. Albeit details differ, that bad-AFM metal regime could resemble our AFM plus Kondo-screening phase.

Finally, all our theoretical studies on the thin-film NdNiO$_2$ system, i.e. Refs. 18 and 19 and this one, are conducted in a 'bulk-like' fashion by using the thin-film lattice parameter for a bulk-crystal calculation. Already several works addressed the explicit thin-film geometry on a SrTiO$_3$ substrate via supercell DFT(+U) calculations. However, while nickelate/SrTiO$_3$ interface physics may have some impact, from the comparison of the currently available experimental data and the results of a theoretical bulk-like approach there is no pressing evidence that key effects are missing in the latter. Furthermore, the experimental thin films with ~10 nm thickness are rather bulky, accounting for more than thirty vertical unit cells. The measured critical current densities of more than 300 kA/cm$^2$ would also be quite large for sole two-dimensional superconductivity in the interface. Nonetheless, investigating NdNiO$_2$ in an ultrathin limit of only a few unit cells would surely be interesting.

In the end, different theoretical concepts for infinite-layer nickelates are presently put to the test. Ultimately,
only experiment can tell which of these concepts is closest to the truth. The here presented evolution of the doping-dependent phenomenology is already in line with various experimental findings, most notably concerning the identification of weakly-insulating regions on either side of the superconducting doping region as well as the dominance of hole-like transport for \( \delta > 0.15 \). In a next step, angle-resolved photoemission (ARPES) measurements appear deciding, since therefrom the correlation strength and the role of the Ni-\(d_{z^2}\) flat-band part with doping is most clearly revealed. But ARPES on these thin film architectures, together with the necessary resolution for possible flat-band physics out of the \( k_z = 0 \) plane, is surely demanding. By any means, the infinite-layer nickelate physics is highly puzzling and appears to integrate various condensed-matter regimes into one materials class, providing room for intense research in the future.

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Appendix: Role of explicit Coulomb interactions on oxygen

As discussed in more detail in Ref. 38, the SIC treatment for most relevant O(2\(p\)) in the present form effectively introduces a \( U_{pp} \) and a \( U_{pd} \) interaction term. As a result, the \( pd\)-splitting between O(2\(p\)) and Ni(3\(d\)) is enhanced compared to LDA, and the hoppings \( t_{pd} \) between those site-orbitals are renormalized. In order to assess the effect of including explicit Coulomb interactions on oxygen via a SIC-modified O pseudopotential, we performed also calculations within the conventional DFT+DMFT scheme for the paramagnetic system. There, the standard O pseudopotential based on LDA is employed.

In Fig. 10 the total spectral function \( A_{tot}(\omega) \) and the local Ni-\(e_g\) part are shown in comparison for both computational schemes. One may infer that though the Hubbard \( U \) on the Ni site is identical, the overall correlation strength in the standard DFT+DMFT approach is much weaker than with including additionally Coulomb interactions on oxygen. At stoichiometry, the system

![Figure 10](image_url)

**FIG. 10.** (color online) Comparison of DFT+DMFT and DFT+sicDMFT spectral functions for pristine NdNiO\(_2\) (left) and at doping level \( \delta = 0.15 \) (right). (Top) Total spectrum and (bottom) local Ni-\(e_g\) spectrum.

![Figure 11](image_url)

**FIG. 11.** (color online) Comparison between DFT+DMFT and DFT+sicDMFT for pristine (left) and \( \delta = 0.15 \) hole-doped (right) NdNiO\(_2\). (a) \( k \)-resolved spectral function \( A(k,\omega) \) and (b) imaginary part of the Ni-\(e_g\) self-energy \( \text{Im } \Sigma(i\omega_n) \).

(c)
is a strongly renormalized metal, i.e. with Ni-$d_{x^2-y^2}$ not reaching a Mott-insulating state, in line with other works.[24,14] This can be explained by the fact that a sizable renormalization of $t_{pd}$ is missing in DFT+DMFT. Thus a sole focus on the Hubbard $U$ on Ni in order to assess the correlation strength in infinite-layer nickelates may be misleading, since a vital contribution is carried by the Coulomb interactions on oxygen because of the late-TM-oxide context. Furthermore, the distance between oxygen and Ni(3d) states, associated with both main peaks in the respective total spectrum, is somewhat larger from the DFT+sicDMFT calculations, which reflects the increase of $pd$ splitting.

For the hole-doped compound, it is additionally seen that the Ni-$d_{x^2}$ character does not show a peak close to the Fermi level for DFT+DMFT. This is understood from Fig. 11, which displays the comparison between DFT+sicDMFT and DFT+DMFT on the level of the $k$-resolved spectral function $A(k, \omega)$ as well as the imaginary part of the Ni-$e_g$ self-energy $\text{Im} \Sigma(\omega_n)$. It becomes obvious that the previously discussed key effect of hole doping, namely the shift of the Ni-$d_{x^2}$ flat-band part in the $k_z = 1/2$ plane is missing in DFT+DMFT. Moreover, the SD electron pockets are much wider shifted to higher (unoccupied) energies, also in line with previous DFT+DMFT studies.[24,16,18] This difference in doping evolution of those specific dispersions has to be related to the fact, that both individual Ni-$e_g$ orbitals hybridize very differently with O(2p) (see last paragraph in section II). Consequently, the Ni-$d_{x^2-y^2}$ self-energy at $\delta = 0.15$ also does not show a clean low-frequency anomaly in DFT+DMFT, since that one should be linked to the Ni-$d_{x^2}$ flat-band part residing at $\varepsilon_F$. However, note that the Ni-$e_g$ occupations up to $\delta = 0.15$ do not seem to differ strongly between both correlation schemes.

In summary, there are strong qualitative differences between the DFT+DMFT and the DFT+sicDMFT description of pristine and doped NdNiO$_2$, which ask for a rethinking of the sole TM-driven correlation picture of late transition-metal oxides in many numerical approaches.

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