Multiorbital processes rule the Nd$_{1-x}$Sr$_x$NiO$_2$ normal state

Frank Lechermann
I. Institut für Theoretische Physik, Universität Hamburg, Jungiusstr. 9, 20355 Hamburg, Germany

The predominant Ni-multiorbital nature of infinite-layer neodymium nickelate at stoichiometry and with doping is revealed. We investigate the correlated electronic structure of NdNiO$_2$ at lower temperatures and show that first-principles many-body theory may account for Kondo(-lattice) features. Yet those are not only based on localized Ni-$d_{x^2-y^2}$ and a Nd-dominated self-doping band, but heavily builds on the participation of Ni-$d_{x^2-y^2}$ in a Hund-assisted manner. In a tailored three-orbital study, the half-filled regime of the former plane Ni orbital remains surprisingly robust even for substantial hole doping $\delta$. Reconstructions of the interacting Fermi surface designate the superconducting region within the experimental phase diagram. They furthermore provide clues to recent Hall measurements as well as to the astounding weakly-insulating behavior at larger experimental $\delta$. Finally, a strong asymmetry between electron and hole doping, with a revival of Ni single-orbital features in the former case, is predicted. Superconductivity in Nd$_{1-x}$Sr$_x$NiO$_2$ is unlike the one in cuprates of distinct multiorbital kind, building up on nearly localized Ni-$d_{x^2-y^2}$ and itinerant Ni-$d_{x^2-y^2}$.

I. INTRODUCTION

The discovery of superconductivity in Sr-doped thin films of infinite-layer (IL) NdNiO$_2$ with a $T_c$ in the 10 K range marks a further milestone in the investigation of transition-metal oxides$^1$. Recent continuing experimental works provided details on the thin-film growth on a SrTiO$_3$ substrate, and moreover yield a phase diagram with doping$^{2,4}$. The latter place the superconducting region in the range $0.125 \lesssim x \lesssim 0.25$ within the Nd$_{1-x}$Sr$_x$NiO$_2$ system. Notably, that area is neighbored by weakly-insulating regions on either side of the doping range. Non-surprisingly, the original findings$^1$ are already covered by many theoretical works$^{30-33}$, with also important earlier studies$^{3,5-29}$, with also important earlier studies$^{30-33}$ on IL(-like) nickelates.

Up to now, three (interlinked) key questions are associated with the challenging physics of superconducting nickelates. The first one deals with the basic comparison to high-$T_c$ layered cuprates. Though seemingly akin, IL nickelate (see Fig. 1a) with the unusual Ni$^2+$ formal oxidation state shows prominent differences at stoichiometry: NdNiO$_2$ is weakly metallic and does not exhibit magnetic ordering down to the lowest measured temperatures$^1$. Theoretical calculations furthermore show that it has an additional self-doping (SD) band across the Fermi level and that the charge-transfer character is weaker than in cuprates$^{11,15}$. Second, strong correlations within the Ni-$d_{x^2-y^2}$ state together with an existing SD band, raises the question about Kondo(-lattice) physics at low temperature. An experimentally found$^1$ resistivity upturn below about $T \sim 70$ K might indeed be indicative for related processes. The third, highly debated, question is most relevant for the superconducting mechanism and deals with the issue of deciding low-energy physics based on single-Ni-orbital processes of Ni-$d_{x^2-y^2}$ kind$^{18-23}$ versus processes of multi-Ni-orbital kind$^{7,9-11,31}$ at stoichiometry and with finite hole doping from replacing Nd by Sr.

In a previous work$^{11}$, we focussed on the one-to-one comparison of IL nickelates to structural-akin cuprates. The present study uncovers crucial Ni-multiorbital mechanisms for Kondo physics at stoichiometry and for the accentuation of the superconducting region with hole doping.

First-principles many-body theory and a realistic three-band Hamiltonian investigation are utilized to establish our current understanding of IL nickelates. Besides the significance of Ni-$d_{x^2-y^2}$ and the Nd-dominated SD band, we unveil the decisive role of Ni-$d_{x^2-y^2}$, both for a Kondo(-lattice) behavior at stoichiometry and for the characterization of the normal-state correlated electronic structure with hole doping $\delta$. While the Ni-$d_{x^2-y^2}$ Fermi-surface sheet in the $k_z = 0$ plane does hardly evolve with increasing $\delta$, an additional Ni-$d_{x^2-y^2}$ sheet becomes available in the $k_z = 1/2$ plane for dopings in the region of the onset of superconductivity. On the other hand, further changes of the Fermi surface at larger hole doping may be connected to the experimentally found weakly-insulating behavior. The given Fermi-surface reconstructions with doping are furthermore in line with the experimentally established Hall-data based multiband picture of an evolution from electron-like to hole-like transport. In a final step, we compare hole doping with theoretical electron doping and detect a stronger single-orbital character of Ni-$d_{x^2-y^2}$ flavor on the electron-doped side.

II. METHODS

In essence, three methodologies are put into practise in this work. First, the charge self-consistent combination$^{34}$ of density functional theory (DFT), self-interaction correction (SIC) and dynamical mean-field theory (DMFT), i.e., the so-called DFT+sicDMFT framework$^{35}$, is used to provide a realistic approach to the temperature-dependent correlated electronic structure of NdNiO$_2$. The complete Ni($3d$) shell enters the correlated subspace of the DMFT impurity problem. Coulomb interactions on oxygen are described within SIC, and are incorporated in the O pseudopotential$^{36}$. A mixed-basis pseudopotential code$^{37-39}$ takes care of the DFT part in the
local density approximation (LDA). The SIC is applied to the O(2s) and the O(2p) orbitals via weight factors $w_p$ (see Ref. 36 for more details). While the O(2s) orbital is by default fully corrected with $w_p = 1.0$, the common choice $w_p = 0.8$ is used for O(2p) orbitals. The Nd(4f) states are put in the pseudopotential core. Continuous-time quantum Monte Carlo in hybridization expansion\(^\text{39}\) as implemented in the TRIQS code\(^\text{41,42}\) is utilized to address the DMFT problem. The DMFT correlated subspace is governed by a five-orbital full Slater-Hamiltonian applied to the Ni projected-local orbitals\(^\text{43}\). A Hubbard $U = 10$ eV and a Hund’s exchange $J_H = 1$ eV prove reasonable for this large-energy window treatment of the given late transition-metal oxide\(^\text{11}\). The fully-localized-limit double-counting scheme\(^\text{44}\) is applied. The DFT+sicDMFT calculations are performed in the paramagnetic regime. Maximum-entropy and Padé methods are employed for the analytical continuation from Matsubara space onto the real-frequency axis. Stoichiometric lattice parameters are overtaken from experiment\(^\text{1}\).

Second, we employ the maximally-localized Wannier-function (MLWF) formalism\(^\text{45,46}\) to construct an effective three-orbital low-energy Hamiltonian for NdNiO\(_2\), to be utilized at stoichiometry and with finite doping. Details of the construction are provided in section IV A.

Third, the derived Hamiltonian is solved by the rotationally invariant slave-boson (RISB) scheme\(^\text{47–52}\) on the mean-field level. The RISB electronic self-energy is local (or extendable via cluster techniques) and consists of a term linear in frequency as well as a static part. It thus lacks the full frequency dependence of the DMFT self-energy, but is still well suited (here at formal $T = 0$) for a large class of correlated materials problems. For details on the computation of quantities such as the quasiparticle weight $Z$ or local spin correlations see e.g. Refs. 48 and 51.

III. KONDO SIGNATURE IN NdNiO\(_2\) FROM COMPREHENSIVE REALISTIC DMFT

In Ref. 11 we studied the interacting electronic structure of NdNiO\(_2\) using the DFT+sicDMFT method at the system temperature $T = 193$ K. At the reasonable large interaction strength $U = 10$ eV, the half-filled Ni-$d_{x^2-y^2}$ band is Mott insulating and does not participate in the Fermi surface. This is shown in Fig. 2a, where only the electron pockets of the self-doping band cross the Fermi level $\varepsilon_F$ in the $k_z = 0$ plane around $\Gamma$ and in the $k_z = 1/2$ plane around A. The SD band is of mixed Ni-Nd character, namely Nd-$d_{x^2-y^2}$ and especially relevant Ni-$d_{x^2}$ contribution to the $\Gamma$ pocket\(^\text{11,19}\). A more detailed discussion of the orbital hybridizations can be found in Ref. 11.

At lower temperatures, a coupling of the localized Ni-$d_{x^2-y^2}$-based spin-1/2 and the remaining itinerant de-
FIG. 3. (color online) Orbitals weights (i.e. 'fatband' picture) for Ni-$d_{z^2}$ (left) and Ni-$d_{x^2-y^2}$ (right) character in the $k$-resolved DFT+sicDMFT spectrum of NdNiO$_2$. (a) Along high-symmetry lines in the Brillouin zone at $T = 193$ K and $T = 39$ K. (b) On the Fermi surface in the $k_z = 0$ plane at $T = 193$ K. Note that the Ni-$d_{z^2}$ colorscale maximum is 30 times larger than the Ni-$d_{x^2-y^2}$ one.

degrees of freedom is suggested from the strong-coupling situation. An underscreening scenario is then expected because of the low filling of the electron pockets. But importantly, there is zero nearest-neighbor hopping between Ni-$d_{x^2-y^2}$ and Nd-$d_{x^2-y^2}$ (e.g. Ref. 11). Thus an intriguing Kondo(-lattice) picture is expected. Note however that it is computationally challenging to reach very low temperatures within our five-Ni-orbital realistic DMFT framework.

Still, Fig. 2b displays the evolution of the $k$-resolved correlated electronic structure at low energy with decreasing $T$. At $T = 58$ K, a flat-dispersion feature appears in the spectral function $A(k, \omega)$ close to $\varepsilon_F$, which further settles below 40 K. The location of the electron pockets of the SD band remains rather $T$-independent. It is very tempting to associate this flat feature crossing itinerant, seemingly weakly correlated, dispersions at lowest energy with a Kondo scenario. Yet the spectral intensity of the flat feature remains very weak at these reachable temperatures. The orbital-resolved local spectrum depicted in Fig. 2c renders obvious, that the main Ni-derived spectral weight in this region is of Ni-$d_{z^2}$ character. At $T = 193$ K, a single Ni-$d_{z^2}$ quasiparticle peak is located just below $\varepsilon_F$ at $\sim -30$ meV. Lowering temperature first leads to a second peak above the Fermi level that gradually becomes sharper and shifts towards $\varepsilon_F$. Hence the 'Kondo resonance' property, at least in that $T$ range, is carried by the Ni-$d_{z^2}$ character. This points to a relevant participation of Ni-$d_{z^2}$ in the given low-energy physics. The local spectral weight of Ni-$e_g$ kind near $\varepsilon_F$ strongly grows below $T = 58$ K (see inset at bottom in Fig. 2c), in good agreement with the experimental temperature scale for the onset of the resistivity increase. The orbital-resolved electron count is hardly dependent on $T$ and reads $\{n_{d_{x^2}}, n_{d_{x^2-y^2}}\} = \{1.845, 1.065\}$ for Ni.

To investigate the roles of the two Ni-$e_g$ orbitals further, we also extracted their $k$-resolved orbital characters (i.e. 'fatbands') in the interacting regime, as shown in Fig. 3a. It is first once more confirmed that there is no Ni-$e_g$ contribution to the electron pocket around $A$. Second, the Ni-$d_{x^2-y^2}$ orbital weight at low-energy is way smaller than the Ni-$d_{z^2}$ one. The Ni-$d_{x^2-y^2}$ origin of the flat feature at low $T$ is still obvious. Independent of temperature, the Ni-$d_{x^2-y^2}$ orbital displays a low-energy hybridization with Ni-$d_{z^2}$ within the $\Gamma$ pocket, however interestingly, only along $\Gamma-X$ in the chosen Brillouin-zone path. This specific direction-dependent hybridization is already observable on the LDA level by close inspection. Its structure is even better revealed when plotting the Ni-$d_{z^2}$-resolved orbital weight along the $k_z = 0$ Fermi surface (see Fig. 3b). The corresponding weight is strictly zero along $\Gamma-X$, and along $\Gamma-Z$ from Fig. 3a. Since the oxygen positions in real space correspond to the $\Gamma-X$ direction, we attribute this Ni-Ni hybridization to explicit hopping via oxygen. It appears relevant for the present Kondo scenario since it provides the sole explicit Ni-$d_{z^2}$ connection between the single level and the $\Gamma$ pocket at lower temperature (cf. Fig. 3a). As an inspection below $T = 58$ K, a Fermi-surface maximum on the circular sheet appears along $\Gamma-X$ and splits perpendicular to the $\Gamma-X$ direction, respectively (not shown).

In order to eventually also connect to the spin degree of freedom in a simplest manner, we choose a linear-response(-kind) calculation. For $T = 39$ K, we strongly spin split the paramagnetic Ni-$d_{x^2-y^2}$ occupation by hand and perform a single DFT+sicDMFT step allowing for spin-polarized Ni self-energies. This one-step ‘magnetic-order’ calculation leads to a spin contrast in the low-energy spectral function as shown in Fig. 4a. As expected, the single level, mirroring the localized-spin behavior, is split into a larger-occupied spin-up branch and a smaller-occupied spin-down part (due to the per-
formed spin splitting by hand). Important is however the effect of this splitting onto the itinerant pockets, since it may deliver information about the Kondo-(like) exchange coupling. Also here, the A pocket shows only weak spin signature. Yet the Γ pocket displays sizable spin splitting, hence is 'Kondo' affected by the localized Ni-\(d_{x^2-y^2}\) spin. As one can easily see, that pocket's spin splitting is the other way around, i.e., the spin-up band is shifted to somewhat higher energies. This indeed points to an antiferromagnetic (AFM) exchange coupling between Ni-\(d_{x^2-y^2}\) and the SD band.

The size of the SD spin splitting at Γ amounts to 40 meV (see Fig. 4b), which translates in a Kondo antiferromagnetic coupling between Ni-\(d_{x^2-y^2}\) and the SD band. No-.. denoted by the previous more comprehensive DFT+sicDMFT picture. From the results of Ref. 11 and those from the previous section III, the natural conclusion has to be that we definitely need Ni-\(d_{x^2-y^2}\) and SD orbital. That's not unfeasable, but one wonders if that is truly 'minimal', also in a model sense for general IL nickelates. For instance, we learned that the A pocket, where the Nd-orbital contribution is most substantial, is apparently not as important as the Γ pocket in the low-temperature interacting regime. It may therefore be sufficient for a truly minimal setting to merge the Nd(5d) part at low energy with the remaining Ni-\(t_{2g}\), Ni(4s) and O(2p) contributions and ally them altogether in a 'stand-alone bath' degree of freedom coupled to the Ni-\(e_g\) orbitals.

We thus arrive at a three-orbital Hamiltonian, based on Ni-\(d_{x^2-y^2}\), Ni-\(d_{x^2-y^2}\) and a third orbital which we still call 'SD', but understandably now carries all the non-Ni-\(e_g\) contributions at low energy. Concretely, the complete Hamiltonian reads

\[
H_{\text{min}} = H_{\text{kin}} + \sum_i \left( H_{\text{int}}^{(i)} + H_{\text{nint, loc}}^{(i)} \right),
\]

using the label \(i\) for the unit cell. It incorporates the kinetic part \(H_{\text{kin}}\), the local-interacting part \(H_{\text{int}}\) and a further local, but non-interacting contribution \(H_{\text{nint loc}}\). The kinetic Hamiltonian, here liberated from all local terms, may be written with hoppings \(t\) as

\[
H_{\text{kin}} = \sum_{i\neq j, mm', \sigma} t_{ij, mm', \sigma} c_{i m \sigma}^\dagger c_{j m' \sigma},
\]

where \(m, m' = \text{Ni-}d_{x^2-y^2}, \text{Ni-}d_{x^2-y^2}, \text{SD}\) and \(\sigma = \uparrow, \downarrow\). For \(H_{\text{int}}\) we choose a canonical Slater-Kanamori form to de-
scribe local interactions among the Ni-\(\epsilon_g\) orbitals, i.e. 
\[
H_{\text{int}}^{(i)} = U \sum_{m \neq z^2, z^2 \sigma} n_m \epsilon_{m \sigma} + \sum \left\{ U' n_{z^2 \sigma} n_{x^2-y^2 \sigma} + U'' n_{z^2 \sigma} n_{x^2-y^2 \sigma} + J_H c_{z^2 \sigma}^\dagger c_{z^2 \sigma} c_{z^2 \sigma} c_{z^2 \sigma} + J_H c_{z^2 \sigma}^\dagger c_{x^2-y^2 \sigma} c_{z^2 \sigma} c_{x^2-y^2 \sigma} \right\}, \tag{3}
\]
and \(U' = U - 2J_H, U'' = U - 3J_H\). Thus notably within the present choice, explicit interactions within the SD orbital, or between Ni-\(\epsilon_g\) and SD, are set to zero in our minimal realistic modeling. The final \(H_{\text{int}}^{\text{loc}}\) does not only deal with the local sum \(\epsilon_{\text{loc}}\) of kinetic energies, but importantly also has to take care of double counting due to a depiction of strongly interacting Ni-\(\epsilon_g\) coupled to a weakly interacting SD state. Furthermore, the SIC description in the DFT+sicDMFT calculations took care of relevant interactions on O(2\(p\)) in this late transition-metal oxide, which now enters also the double-counting term. It proves therefore favorable in this minimal context to split the double-counting (DC) correction into two parts: a negative shift of the SD state through a potential \(\mu_{\text{SD}}\) in the interacting case, and a standard DC correction for explicitly interacting Ni-\(\epsilon_g\). For the latter we choose again the fully-localized-limit form\(^{44}\). The last term in (1) is hence given by 
\[
H_{\text{nint}, \text{loc}}^{(i)} = \sum_{m \neq z^2, z^2 \sigma} \epsilon_{m \sigma} c_{m \sigma}^\dagger c_{m \sigma} - \mu_{\text{SD}} \sum_{\sigma} n_{\text{SD} \sigma} - \sum_{m \neq z^2, z^2 \sigma} \left[ U \left( \hat{n}_{\epsilon_g} - \frac{1}{2} \right) - J_H \left( \hat{n}_{\epsilon_g} - \frac{1}{2} \right) \right] n_{\sigma}. \tag{4}
\]
As we will utilize the minimal Hamiltonian without charge self-consistency, the \(\hat{n}\) occupations in (4) refer to \(\epsilon_g\) fillings at \(U = 0\) (in the following, \(U = 0\) is understood as \(U = J_H = 0\)). The potential \(\mu_{\text{SD}}\) for the SD state is not perfectly straightforward, since therein accumulate various effects, such as electrostatics, interaction with O(2\(p\)), etc. Since NdNiO\(_2\) for the present minimal three-orbital Hamiltonian is located at total half filling \(n = 3\), the most canonical choice is provided by \(\mu_{\text{SD}} = U/2\) (e.g. Ref. 54).

For a concrete representation of the described Hamiltonian form, maximally-localized Wannier functions are derived from the LDA electronic structure. Because of the strong downfolding to three orbitals, the disentangling procedure\(^{46}\) is employed and the dispersions closest to the Fermi surface are fixed in an inner energy window. The result of this Wannier construction is displayed in Fig. 5. The overall Wannier dispersion agrees well with the LDA one, and the fatband analysis clearly identifies the Ni-\(d_{z^2}\), Ni-\(d_{x^2-y^2}\) and SD bands. Solely the dispersions/fatbands around the A point are hard to exactly align within the present orbital setting. This can be easily understood from the fact that the occupied part of this very region has sizable contribution from Ni-\(t_{2g}\). However again, from our more general DFT+sicDMFT description, the very details around the A point are not crucial for the IL nickelate physics.

But importantly note that the present SD orbital does not only describe the electron pockets at \(\Gamma\) and \(A\). It also has nonzero orbital weight on the Ni-\(\epsilon_g\) dominated dispersions, mostly on the Ni-\(d_{z^2}\) band. This fact is generally very relevant to appreciate the role of Ni-\(d_{z^2}\), and to understand the following results with doping.

The real-space Wannier orbitals are shown in the bottom part of Fig. 5. For Ni-\(\epsilon_g\), they resemble the expected appearance. Substantial leaking of Ni-\(d_{x^2-y^2}\) onto in-plane oxygen sites is a natural outcome of the strong downfolding. Of course by construction, here the SD orbital cannot resemble an atomic-like orbitals Since not based on a highly-localized viewpoint, its spread is comparatively large and the Wannier centre is inbetween Ni and Nd. It as contributions from Nd, Ni and O and furthermore breaks the full-cubic symmetry. This orbital is important for its 'stand-alone' features to Ni-\(\epsilon_g\).

Let us conclude this subsection by providing the Wannier values for the local single-particle terms \(\epsilon_{\text{loc}}\) and for near hoppings \(t\) in Tab. I. The nearest-neighbor hopping between Ni-\(d_{x^2-y^2}\) and SD, here notably included in \(\epsilon_{\text{loc}}\), is indeed zero, but there is a sizable one between Ni-\(d_{z^2}\) and SD.

\begin{table}[h]
\centering
\begin{tabular}{|l|c|c|c|c|}
\hline
Orbitals & \(\epsilon_{\text{loc}}\) & \(t_{\text{12}}^{(x)}\) & \(t_{\text{12}}^{(z)}\) & \(t_{\text{12}}^{(e)}\) \\
\hline
Ni-\(d_{z^2}\), Ni-\(d_{x^2}\) & -1479 & -1 & -398 & 0 \\
Ni-\(d_{x^2-y^2}\), Ni-\(d_{x^2-y^2}\) & 232 & -387 & -30 & -50 \\
SD, SD & 1191 & -13 & -229 & 12 \\
Ni-\(d_{z^2}\), SD & 92 & 125 & 77 & -8 \\
Ni-\(d_{x^2}\), Ni-\(d_{x^2-y^2}\) & 0 & 41 & 0 & 9 \\
Ni-\(d_{x^2-y^2}\), SD & 0 & 17 & 0 & 13 \\
\hline
\end{tabular}
\caption{Local single-particle terms and selected hoppings of the derived Wannier Hamiltonian. All energies in meV.}
\end{table}

\section{RISB solution}

The paramagnetic RISB solution of the minimal Hamiltonian \(H_{\text{min}}\) in the stoichiometric half-filled case and with finite doping is discussed. Technically speaking, the present multiorbital Hamiltonian is solved in a ‘cluster-RISB’ fashion, since the onsite Ni-\(\epsilon_g\) effects and the intersite effects between Ni-\(\epsilon_g\) and the SD orbital are treated on equal footing.

\subsection{Stoichiometry}

Pristine NdNiO\(_2\) resembles the half-filled \(n = 3\) case of the three-orbital description. At \(U = 0\), the orbital fillings read \(\{n_{z^2}, n_{x^2-y^2}, n_{\text{SD}}\} = \{1.84, 0.93, 0.23\}\), i.e. the Ni-\(d_{x^2-y^2}\) orbital lies somewhat below true half filling. Figure 6a shows the evolution of key quantities of the
low-energy electronic structure with increasing the Hubbard $U$. Be aware that due to the strong downfolding, the lack of charge self-consistency as well as the RISB treatment, the tailored $U$ value surely differs from $U = 10$ eV of the DFT+xcDMFT study.

As $U$ grows from zero, charge transfers occur such to establish a truly half-filled Ni-$d_{x^2-y^2}$ orbital in an orbital-selective Mott transition scenario. For $U = 7$ eV, the fillings read $\{n_{x^2}, n_{y^2}, n_{SD}\} = \{1.83, 1.0, 0.17\}$ and the quasiparticle (QP) weight $Z$ (i.e. the inverse effective mass) of the in-plane orbital reads $Z_{x^2-y^2} = 0.02$, i.e. is close to zero. In the following, we will understand the electronic states at $U = 7$ eV as the low-energy equivalent to the more comprehensive DFT+xcDMFT picture at $U = 10$ eV. The QP weight $Z_{x^2}$ remains close to unity due to the rather large filling. The localization of the Ni-$d_{x^2-y^2}$ electron is accompanied by the build-up of a corresponding local squared spin moment $\langle s^2 \rangle$ which saturates at the limiting value $s(s+1/2) = 3/4$. The Ni-$e_g$ spin-spin correlations are surely positive because of Hund’s first rule, and increase with the localization degree of Ni-$d_{x^2-y^2}$. On the other hand, the intersite spin-spin correlations of $\{\text{Ni}-e_g, \text{SD}\}$ kind are negative and much smaller in magnitude. The $\{\text{Ni}-d_{x^2}, \text{SD}\}$ spin correlations turn out still larger than the $\{\text{Ni}-d_{x^2-y^2}, \text{SD}\}$ ones. This again proves that a direct spin-spin coupling between the latter orbitals is weak.

In view of the dichotomy between itinerant and localized behavior in strongly correlated materials, let us further focus on Fermi surfaces and local states. The non-interacting Fermi surface in Fig. 6b shows the electron pockets around $\Gamma$ and $\Lambda$, as well as the Ni-$d_{x^2-y^2}$-dominated sheet in the $k_y = 0, 1/2$ planes. Note the closing of the latter sheet along $Z-R$ as a difference to conventional cuprate fermiology (see Ref. 25 for a direct comparison). In the strongly interacting case, the $\Gamma$ pocket is shrunk and since the QP at $U = 7$ eV is not yet exactly zero, the strongly-renormalized Ni-$d_{x^2-y^2}$ sheet

FIG. 6. (color online) Minimal-Hamiltonian properties at half filling for $\mu_{SD} = U/2$. (a) Selected quantities for increasing $U$. The Hund’s exchange is chosen $J_H = U/3$ for $U < 3$ and fixed to $J_H = 1$ eV for $U \geq 3$ eV (dashed gray line separates the regimes). From left to right: orbital occupation $n_m$, orbital-resolved QP weight and local squared-spin moment (top) as well as local spin-spin correlation (bottom). (b) Non-interacting Fermi surface for $k_y = 0$ (left) and $k_y = 1/2$ (right). (c) Interacting Fermi surface for $U = 7$ eV.

FIG. 7. (color online) Local-state behavior of the minimal Hamiltonian via the slave-boson weights $|\phi_{q,q'}|^2$ connecting Fock states $q,q'$. (a,b) Weight matrix in $q,q'$-space covering the $N = 0, \ldots, 6$ particle sectors $N_p$ for weights $> 10^{-5}$: (a) $U = 0$, and (b) $U = 7$ eV. (c) Dominant diagonal weights (along positive axis) for $U = 0$ and for $U = 7$ eV, and dominant off-diagonal weights (along negative axis) within each particle sector for $U = 7$ eV (filled circles).
still contributes to the Fermi surface (yet now slightly enlarged due to the exact half filling). But in the \( k_z = 1/2 \) plane, the near-Mott-insulating sheet apparently now bends electron-like. The warping close to \( k_z = 1/2 \) is therefore further strengthened with strong correlations. We will not further comment on the small-A-pocket features because of possible Wannier-construction artifacts in that region for \( U \neq 0 \).

The slave-boson amplitudes \( \phi_{qq'} \) connecting states \( q, q' \) in the RISB formalism provide useful insight in the local-state behavior for different degrees of electronic correlation. Let us first remind about the corresponding Hilbert space. There are a total of \( 2^6 = 64 \) (Fock) states available for our three-orbital system, with each particle sector \( N = 0, \ldots, 6 \) contributing \( N_q = 6!/(N!(6-N)!)) \) states. Thus, if coupling between all states is allowed, the total number of \( \phi_{qq'} \) amounts to \( 64 \times 64 = 4096 \) for a three-orbital problem. However here, we only study normal-state properties and therefore exclude couplings between different particle sectors. A RISB implementation for pairing problems permitting also such couplings has been put forward by Isidori and Capone. Without pairing, one ends up with \( \sum_N N_q^2 = 924 \) slave-boson amplitudes (without using symmetries) in the RISB calculation. The squared amplitudes of the relevant part of those are depicted in Fig. 7 for the non-interacting case and for \( U = 7 \) eV. The values \( \left| \phi_{qq'} \right|^2 \) may be interpreted as the weight for finding the system in a quantum state characterized by \( q, q' \) to occur, since \( \sum_{qq'} \left| \phi_{qq'} \right|^2 = 1 \) holds. There is of course the option to transfer from a Fock basis to a multiplet basis, but due to the specific itinerant inter-site structure of our Hamiltonian we remain in the Fock basis for a straightforward analysis.

Figures 7a,b first show that for sizable magnitudes the weight matrix in \( q, q'- \) space is mostly diagonal. Especially without interaction, quantum entanglement between unlike states is rather implausible. But also along the diagonal, the weights are much more distributed for \( U = 0 \), just because there is no interaction-driven state selection but only statistics of a Fermi gas. Since describing half filling, for \( U = 7 \) eV the fluctuations to the 0.1 and 5.6 particle sectors is significantly suppressed. The truly relevant diagonal Fock states are easily selected (see Fig. 7c), using the notation

\[
|q\rangle = \mid \uparrow_{z^2} \downarrow_{x^2} \uparrow_{x^2 - y^2} \downarrow_{x^2 - y^2} \uparrow_{SD} \downarrow_{SD} \rangle .
\]

Non-surprisingly the largest weight stems from the \([111000]\) and \([110100]\) states in the 3-particle sector, i.e. two electrons in Ni-\(d_{xz}\) and one in Ni-\(d_{x^2 - y^2}\). Of course, doubly-occupied (sub-)states in Ni-\(d_{x^2 - y^2}\), though appreciated without interactions, are highly suppressed at large \( U \). There are still four relevant diagonal states for \( U = 7 \) eV in the 2- and 4-particle sector, respectively. The ones in the 2-particle sector display Hund’s first rule within Ni-\(e_g\). The ones in the 4-particle sector exhibit fully occupied Ni-\(d_{xz}\) as well as singly-occupied Ni-\(d_{x^2 - y^2}\) and SD, but importantly, without spin-alignment differentiation for the \( \left| \phi_{qq'} \right|^2 \) magnitude. In other words, without charge fluctuation in Ni-\(d_{xz}\) there is no exchange-favored discrimination of \{Ni-\(d_{x^2 - y^2},\)SD\} spin states.

Let us finally turn to the off-diagonal slave-boson weights, of which there are a few in the strongly interacting regime with sizable magnitude (cf. Figs. 7b,c), but still much smaller than the diagonal ones. There is a prominent \( \phi_{qq'} \) in the 2-particle sector, connecting the two Ni-\(e_g\) states \([101000]\) and \([011000]\). This clearly marks the relevant singlet \( S = 0, S_z = 0 \) and triplet \( S = 1, S_z = 0 \) entanglement in the \( e_g \) manifold. The respective twofold off-diagonal weights \( \alpha^{(s)} \), \( \beta^{(s)} \) and \( \gamma^{(s)} \) in the 3-particle sector are more complicated, but most interesting. For clarity, let us enumerate them in words:

- \( \alpha^{(s)} \): scattering between doubly Ni-\(d_{xz}\), spin-up(down)
- Ni-\(d_{x^2 - y^2}\) and spin-up(down) Ni-\(d_{xz}\), spin-down(up) Ni-\(d_{x^2 - y^2}\), spin-down(up) SD
- \( \beta^{(s)} \): scattering between singlet/triplet Ni-\(d_{xz}\), Ni-\(d_{x^2 - y^2}\) and spin-up/down SD
- \( \gamma^{(s)} \): scattering between doubly Ni-\(d_{xz}\), spin-up(down)}
Ni-d_{z^{2}-y^{2}} and spin-up(down) Ni-d_{z^2}, spin-up(down) Ni-d_{x^{2}-y^{2}}, spin-down(up) SD.

Thus these weights provide the desired spin differentiation in the scattering processes between the three orbitals, relevant for paving the way toward establishing a proper Kondo-(lattice) picture. The weights accordingly scale as $\gamma^{(i)} > \alpha^{(i)} > \beta^{(i)}$, which importantly provides further quantitative evidence for a Hund-assisted Kondo scenario via Ni-d_{z^2}, leading to AFM exchange between localized Ni-d_{z^{2}-y^{2}} and the SD band.

To conclude this discussion at stoichiometry, we want to comment on the role of the potential shift $\mu_{SD}$, which was set so far to $\mu_{SD} = U/2$. Figure 8 depicts the modifications in the electronic states with varying $\mu_{SD}$ to smaller and larger values. Generally, the effects from changing $\mu_{SD}$ by reasonable amounts are comparatively weak. The given potential shift defines the energy location of the SD band, thus a smaller(larger) value shifts it upwards(downwards) with respect to $\mu_{SD} = U/2$ (see Fig. 8a). Understandably, a larger $\mu_{SD}$ leads a stronger filling of the SD band and a decrease of correlation strength in Ni-d_{z^{2}-y^{2}} (see Fig. 8b). The growth of the $\{\text{Ni-d}_{z^2}, \text{Ni-d}_{x^{2}-y^{2}}\}$ spin-spin correlation with increasing $\mu_{SD}$ in Fig. 8c may be attributed to the less-filled Ni-d_{z^2} in this situation, strengthening the Ni-d_{z^2} moment. The $\mu_{SD}$ variation renders furthermore the coupling between itinerant and local degrees of freedom obvious. Figure 8c shows for instance the growth of an off-diagonal $|\phi_{qq'}|^2$ near the diagonal at the upper end of the 4-particle sector. This amounts to the coupling of states $\{|011011\}$, $\{|001111\}\}$, describing scattering between singlet/triplet Ni-$\epsilon_g$ and doubly-occupied SD. This means, that the growth of SD occupation from a sinking SD band uplifts also $\phi_{qq'}$ amplitudes with a completely-filled SD state.

The observed $\mu_{SD}$ variations provide confidence that at stoichiometry, the setting $\mu_{SD} = U/2$ indeed proves reasonable. For instance, the existence of the $\Gamma$ electron-pocket Fermi surface is a robust feature of the DFT+sicDMFT study (and also reported from many other theoretical works).

2. Hole doping

The experimental doping with Sr and accordingly the replacement of Nd^{3+} by Sr^{2+} leads to the doping of holes into the nickelate compound. In Ref. 11 we studied this doping process by a DFT+sicDMFT treatment of realistic supercells for Nd_{1-x}Sr_{x}NiO_{2} at $T = 193$ K. It was found that for $x = 0.125$, while both Ni-$\epsilon_g$ orbital gain holes (though Ni-d_{z^2} slightly more), the lowest-energy response comes mainly from Ni-d_{x^{2}-y^{2}}. Whereas for $x = 0.25$, the number of holes in Ni-d_{x^2} is quite large and the lowest-energy response is twofold but dominated by Ni-d_{z^2}. This is apparently not in line with a cuprate-like, single-orbital hole doping of Ni-d_{x^{2}-y^{2}}.

In the following, hole doping is investigated in more detail from the low-energy perspective within the minimal-Hamiltonian approach. It amounts to change the total filling to $n = 3 - \delta$ with $\delta > 0$. This approach surely misses some effects of the realistic Nd substitution by Sr. But it should be geared to unveil the key qualitative features with doping. For instance, one (also electrostatic-supported) effect of true Sr doping is the shifting of the Fermi electron pockets into the unoccupied region\(^{11,20,21,27}\). In the minimal-Hamiltonian scheme, part of the corresponding underlying mechanism surely goes into the value of $\mu_{SD}$. We hence used the two values $\mu_{SD} = 3/7U,U/2$ for comparison and to truly include that upward shifting qualitatively correct.

Figure 9 summarizes the key changes with hole doping from the minimal-Hamiltonian perspective for $U = 7$ eV. Notably, the differences between $\mu_{SD} = 3/7U,U/2$ are small and mostly quantitative. The first surprising (though already DFT+sicDMFT suggested) observation is the very weak change of the Ni-d_{x^{2}-y^{2}} orbital filling with increasing $\delta$ (cf. Fig 9a). It remains very near half filling up to the investigated limiting value $\delta = 0.4$. In fact starting from stoichiometry, $n_{z^2}$ first decreases slightly until $\delta \sim 0.1$, and increases afterwards progressively toward half filling. On the other hand, $n_{x^2}$ decreases steadily with $\delta$. The second surprise is the increase of the SD filling with hole doping. As one associates with SD the electron pockets, one naively ex-
The Fermi sheets also vary. Reason is that the original bands of the SD state increases its weight on the deep-lying Ni-electron pocket. This off-diagonal \( Z \) monitors the outlined inter-site hybridization modification between Ni-\( d_{z^2} \) and SD state. Thus, the strong doping of Ni-\( d_{z^2} \) is triggered by the direct hole creation from \( \delta \), as well as additional charge transfer to the SD state. All this apparently being energetically more favorable than putting more holes into Ni-\( d_{z^2} \).

The diagonal QP weights \( Z_{m} \) behave then as expected with \( \delta \), i.e. \( Z_{z^2} \) decreasing and \( Z_{x^2-y^2} \) exhibiting a non-monotonic behavior with a maximum around \( \delta \approx 0.1 \). Note again that the Ni-\( d_{z^2} \) correlation strength at the large doping of \( \delta \approx 0.4 \) becomes comparable to the one at half filling. Expectedly, the spin-spin correlation between the Ni-\( e_g \) orbitals increases with \( \delta \) due to the enhancement of the Ni-\( d_{z^2} \) localization.

The described evolution in fillings and many-body observables are accompanied by obvious changes of the interacting Fermi surface, shown in Fig. 9b. For \( \delta < 0.11 \), the fermiology in the \( k_z = 1/2 \) plane is basically equivalent to the stoichiometric scenario, i.e. showing an electron-like pocket around \( Z \) (see right part of Fig. 6c). Yet for \( \delta > 0.11 \), there is a Lifshitz transition with an emerging second pocket around \( A \). This pocket should not be confused with the original small SD-based electron pocket.

Hence the Fermi surface reconstructs at \( \delta \approx 0.11 \), giving rise to the topology shown in Fig. 10 for \( \delta = 0.16 \). Going along \( k_z \), the original Ni-\( d_{z^2} \) hole pocket bends over toward an electron pocket, shown for \( k_z = 0.41 \). At \( k_z = 0.45 \) a new heart-shaped hole pocket starts out along the nodal line. It opens up to a large hole sheet around \( A \) at \( k_z = 1/2 \). The nature of the novel larger pocket around \( A \) is of mixed Ni-\( e_g \) kind. There is still another Lifshitz transition at larger \( \delta \). For \( \delta < 0.26 \) the van-Hove singularity at \( Z \) lies below the Fermi level, and above for \( \delta > 0.26 \). Therefore, the \( k_z = 1/2 \)-pocket around \( Z \) vanishes for hole doping beyond \( \delta = 0.26 \). It is surely tempting to relate these reconstructions with the experimental phase boundaries of superconductivity\(^{41}\).

The \( k \)-integrated orbital-resolved QP spectral functions shown in Fig. 11 for selected dopings, underline the scenario we just discussed. Also from there, it seems that the intermediate region has the strongest susceptibility for Ni-\( d_{z^2} \)-driven instabilities in a highly-localized Ni-\( d_{z^2} \) background: below that region the Ni-\( d_{z^2} \) weight at \( \varepsilon_F \) is still small, and above the Fermi level is located in Ni-\( d_{z^2} \) pseudogap.

Yet a further complexity issue has to be discussed. Upon increasing \( \delta \) and running through the described Fermi surface reconstructions, the character partition between Ni-\( d_{z^2} \) and Ni-\( d_{x^2-y^2} \) on the (near-)\( k_z = 1/2 \) Fermi sheets also varies. Reason is that the original bands of both of these orbital characters sweep through each other in that region of the Brillouin zone with doping (see Fig. 6b). Hence a nontrivial mixing of characters due to correlation-modified hybridizations takes place. Table II summarizes the basic information in simple terms for selected dopings in the three distinct regions. It is seen that the intermediate doping region \( 0.11 < \delta < 0.26 \) displays the most intriguing case with the strongest multiorbital character of the Fermi surface. Surely note that the issue of mixed Fermi-surface character does not arise in the \( k_z = 0 \) plane: The \( \Gamma \)–\( \text{M} \) pocket is of perfect Ni-

![FIG. 10. (color online) The interacting Fermi surface along \( k_z \) at \( \delta = 0.16 \), for \( U = 7 \text{eV} \) and \( \mu_{SD} = 3/7U \).](image)

| orbital | Fermi sheets | \( \delta = 0.06 \) | \( \delta = 0.16 \) | \( \delta = 0.30 \) |
|---------|-------------|----------------|----------------|----------------|
| Ni-\( d_{z^2} \) | around \( Z \) | moderate | strong | weak |
| Ni-\( d_{z^2} \) | around \( A \) | moderate | weak |
| Ni-\( d_{z^2} \) | around \( Z \) | strong | moderate |
| Ni-\( d_{z^2} \) | around \( A \) | strong | strong |

TABLE II. Qualitative orbital-character strength on the interacting Fermi-surface sheets in the \( k_z = 1/2 \) plane for selected hole dopings. See bottom right of Fig. 10 for a visualization of both Fermi sheets.

- \( \delta \)-induced depopulation of that orbital. However, one has to recall the important fact that the SD state hybridizes substantially with Ni-\( d_{z^2} \) over a large energy region (see right bottom of Fig. 5). With large \( U \) on Ni, the \( e_g \) electrons try hard to escape to the SD state, if they cannot achieve complete localization. Hence there is a correlation-induced change of the hybridization, here apparently activated for hole doping, such that the SD state increases its weight on the deep-lying Ni-\( d_{z^2} \)-dominated band and more electrons can benefit from the favorable SD-orbital location. This mechanism is proven by \( Z_{SD} \) being the only sizable off-diagonal QP weight, and its absolute value steadily increasing with \( \delta \) (see bottom of Fig 9b). This off-diagonal \( Z \) monitors the outlined inter-site hybridization modification between Ni-\( d_{z^2} \) and SD state. Thus, the strong doping of Ni-\( d_{z^2} \) is triggered by the direct hole creation from \( \delta \), as well as additional charge transfer to the SD state.
In fact, recent experimental Hall data in the normal state is interpreted as going from an electron- to a hole-like transport scenario with doping. This is in line with our findings, since for small $\delta$, transport should mainly arise from the vanishing SD electron pockets and the electron-like bended region around $Z$ which has mixed Ni-$e_g$ character (cf. Tab. II). But for $\delta > 0.11$, the hole pocket around $A$ appears, showing strong Ni-$d_{x^2-y^2}$ weight for intermediate doping.

Last but not least, the present results allow us moreover to also provide an explanation for the puzzling experimental region for superconductivity, is designated with the most pronounced multiorbital character.

3. Comparison between electron and hole doping

The dichotomy between electron and hole doping is an essential part of the high-$T_c$ cuprates physics. Therefore in the final result section, the opportunity is used to compare in basic terms the electron-doped region within the minimal-Hamiltonian approach with the hole-doped one. Experimentally, electron doping of NdNiO$_2$ has not been achieved yet. In analogy to cuprates, it could formally being realized by e.g. replacing Nd$^{3+}$ with Ce$^{4+}$. The theoretical calculations are straightforwardly extended to electron doping by fixing the total particle number at $n = 3 - \delta$ with $\delta < 0$.

Figure 13 displays the comparison of both doping regimes. We lowered the Hubbard interaction to $U = 6$ eV, since the RISB numerics of the electron-doped region turns out more challenging. Nonetheless, the hole-doped side behaves qualitatively identical to the results for $U = 7$ eV.

Let us thus focus on the electron doping, which shows singular differences to hole doping. Starting with the QP weights, it is seen that for small $\delta$ the correlation strength in Ni-$d_{x^2-y^2}$ apparently increases compared to half filling. After a minimum at $\delta \sim -0.1$, the QP weight $Z_{x^2-y^2}$ then strongly recovers and grows up to the studied limiting value $\delta = -0.4$. The doping $\delta = -0.1$ apparently marks the true orbital-selective Mott-transition point in the IL nickelate. The Ni-$d_{x^2-y^2}$ filling remains fixed at half filling for small $\delta$, but then also grows beyond the latter transition point. Expectedly, the Ni-$d_{x^2}$ orbital has to accommodate even more electrons with electron doping. Yet because of the large associated Coulomb-repulsion cost, it succeeds in not becoming completely filled, but can delegate some charge to the remaining orbitals. Also without surprise, the SD orbital becomes further filled, too. With electron doping, no sophisticated correlation-induced change of hybridization has to be invoked in or-



der to place additional electrons in the SD state. Correspondingly, the off-diagonal QP weight $Z_{\delta,SD}$ is small for electron doping. Thus, albeit original electron pockets of the SD band shift deeper into the occupied region, the coupling at least to Ni-$d_{z^2}$ appears weaker. The Ni-$e_g$ spin-spin correlations are much smaller for electron doping, because of the even stronger Ni-$d_{z^2}$ filling. Interestingly when adding electrons, the spin-spin correlations between Ni-$d_{x^2-y^2}$ and SD change sign from negative to positive at the critical point $\delta = -0.1$.

The electron-doped region might be interesting because of obviously stronger Ni-$d_{z^2}$ correlation at low doping and possibly intriguing competition between Kondo correlations and magnetic order. Superconducting tendencies of similar kind as on the hole-doped side are not expected, since electron-doping should not lead to comparable intricate Fermi-surface reconstructions as Ni-$d_{z^2}$ becomes too strongly filled. However apparently, the electron-doped side supports a stronger Ni single-orbital picture of Ni-$d_{x^2-y^2}$ kind. Thus despite the coexistence with the remaining SD band, similarities to cuprates might be more pronounced with electron doping.

V. SUMMARY AND DISCUSSION

Using comprehensive DFT+sicDMFT as well as an aligned minimal-Hamiltonian representation solved within RISB, we were able obtain important insight into the very rich physics of NdNiO$_2$ in pristine condition and with finite doping.

Relevant features of a stoichiometric Kondo-(lattice) scenario at lower temperature are revealed, showing that a calculated sizable AFM Kondo coupling $J_K \sim 120$ meV builds up onto the cooperation of Ni-$d_{x^2-y^2}$, Ni-$d_{z^2}$ and the self-doping band. The Ni-$d_{z^2}$ and the SD band both take part in the screening of the Ni-$d_{x^2-y^2}$ spin, whereby the former orbital has a mediating role through a Hund-assisting mechanism. The onset of significant Kondo correlations around $T \sim 60$ K matches well with the experimental $T = 70$ K for the beginning resistivity upturn$^3$. An interesting k-selective Ni-$e_g$ hybridization via oxygen around $\Gamma$ deserves further investigation.

A realistic minimal three-orbital Hamiltonian is advocated, that describes the low-energy competition of both Ni-$e_g$ orbitals linked to a SD state that carries the effect of remaining Nd(5d), O(2p) and Ni(4s). This Hamiltonian serves the goal of providing a faithful representation of the key degrees of freedom of IL nickelate. Its canonical structure, i.e. two strongly interacting orbitals coupled to a 'stand-alone bath' state, may prove useful for other nickelates or related problems.

The system with hole doping $\delta$ is remarkably different to hole-doped cuprates. The Ni-$d_{x^2-y^2}$ occupation is only very weakly departing from half filling and the orbital even regains correlation strength at larger $\delta$. On the other hand, the Ni-$d_{z^2}$ orbital eagerly collects holes. Also on the expense of the SD orbital, which counterintuitively gains electrons by a correlation-induced change of hybridization to Ni-$d_{z^2}$. While the QP weight for the latter orbital decreases with $\delta$, it remains in a weak-to-moderate correlation regime of $Z_{\delta,SD} \sim 0.7$ at intermediate doping. Further key aspect at hole doping are two apparent Fermi-surface constructions that designate $0.11 < \delta < 0.26$ as the region with strongest multiorbital nature and entanglement. It is an orbital-selective(-kind) situation with highly-correlated, hardly-doped Ni-$d_{x^2-y^2}$ and still substantially-filled Ni-$d_{z^2}$ sharing two Fermi sheets in the $k_z = 1/2$ plane of the Brillouin zone. This specific doping region agrees well with the two experimentally-determined ranges, i.e. $0.125 < x < 0.25$ by Li et al.$^2$ and $0.12 < x < 0.235$ by Zeng et al.$^4$, for the appearance of superconductivity in Nd$_{1-x}$Sr$_x$NiO$_2$.

Moreover, our realistic description may not only explain the experimentally observed change from electron-like to hole-like transport in Hall measurements, but may also provide reason for the weakly-insulating behavior found on both sides of the superconducting region. In essence, Ni-$d_{x^2-y^2}$ remains nearly localized for any reasonable hole doping, and the itinerant contribution of Ni-$d_{z^2}$ remains generally small for $\delta < 0.11$ while contributing mostly to $c$-axis transport for $\delta > 0.26$. The latter should be masked in the so far available thin-film studies of Nd$_{1-x}$Sr$_x$NiO$_2$.

Finally, theoretical electron doping leads to quite different characteristics than hole doping. Most notably, even further correlation-strength enhancement within Ni-$d_{z^2}$ is found for small negative $\delta$, and the spin exchange between the latter and the SD orbital switches to ferro-like behavior. The predominant Ni-$d_{x^2-y^2}$ characteristics on the electron-doped side deserve future experimental investigations.

The present theoretical results raise several further questions. For instance, one wonders about the role of Ni-$d_{z^2}$ states with hole doping, since those are also located in close distance to the Fermi level. However from
our defect-supercell DFT+sicDMFT study\textsuperscript{11}, which included all Ni(3d) on equal footing, they become only weakly doped and their contribution to relevant low-energy physics appears minor up to described $\sim 0.25$. An intuitive reason may be given by the fact that already on the LDA level, Ni-$e_g$ is stronger dispersing than Ni-$t_{2g}$. At strong coupling, with Ni-$d_{z^2}$ becoming half filled, the only appreciable itinerant Ni(3d) degree of freedom to balance the electronic energy cost is thus provided by Ni-$d_{z^2}$. Second, if the IL nickelate superconductivity is based on multi-orbital Ni-$e_g$ processes and not on the so far singular ‘cuprate mechanism’, the question arises why stable pairing is not much more common in nickel-based transition-metal oxides. Yet one has to be aware that the orbital-selective(-like) scenario allied with intriguing fermiology in the intermediate doping region is quite specific, and to our knowledge has not yet been reported for other nickelates. This brings us surely to the most pressing question concerning the pairing mechanism that emerges from the present normal-state scenario. A straightforward simple answer is not available, due to the complex entanglement between localized and itinerant multi-orbital degrees of freedom. From the renormalized-band perspective, a nesting mechanism involving the flattened larger hole-pocket sheets around $\Gamma$ might be conceivable. From a more localized perspective, recent suggestions based on Ni-$e_g$ couplings have been put forward\textsuperscript{9,10}. However, those interesting proposals miss out on the details of the intricate low-energy dispersions that evidently mark the intermediate doping region.

Beyond speculations, as the main conclusion, the superconductivity in thin films of Nd$_{1-x}$Sr$_x$NiO$_2$ is most definitely not of single-orbital cuprate kind. This should not be seen as a disappointment, but quite on the contrary, appreciated as the opening of a new fascinating chapter in the research on superconducting correlated materials.

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1. D. Li, K. Lee, B. Y. Wang, M. Osada, S. Crossley, H. R. Lee, Y. Cui, Y. Hikita, and H. Hwang, Nature **572**, 624 (2019).
2. D. Li, B. Y. Wang, K. Lee, S. P. Harvey, M. Osada, B. H. Goodge, L. F. Kourkoutis, and H. Y. Hwang, arXiv:2003.08506 (2020).
3. K. Lee, B. H. Goodge, D. Li, M. Osada, B. Y. Wang, Y. Cui, L. F. Kourkoutis, and H. Y. Hwang, APL Mater. **8**, 041107 (2020).
4. S. Zeng, C. S. Tang, X. Yin, C. Li, Z. Huang, J. Hu, W. Liu, G. J. Omar, H. Jani, Z. S. Lim, et al., arXiv:2004.11281 (2020).
5. Y. Gu, S. Zhu, X. Wang, J. Hu, and H. Chen, arXiv:1911.00814 (2019).
6. M. Hepting, D. Li, C. J. Jia, H. Lu, E. Paris, Y. Tseng, X. Feng, M. Osada, E. Been, Y. Hikita, et al., Nat. Mater. doi:10.1038/s41563-019-0585-z (2020).
7. L.-H. Hu and C. Wu, Phys. Rev. Research **1**, 032046 (2019).
8. H. Zhang, L. Jin, S. Wang, B. Xi, X. Shi, F. Ye, and J.-W. Mei, Phys. Rev. Research **2**, 013214 (2020).
9. Y.-H. Zhang and A. Vishwanath, Phys. Rev. Research **2**, 023112 (2020).
10. P. Werner and S. Hoshino, Phys. Rev. B **101**, 041104(R) (2020).
11. F. Lechermann, Phys. Rev. B **101**, 081110(R) (2020).
12. T. Zhou, Y. Gao, and Z. D. Wang, arXiv:1910.05757 (2019).
13. J. Hirsch and F. Marsiglio, Physica C: Superconductivity and its Applications **506**, 135534 (2019).
14. Z. Liu, Z. Ren, W. Zhu, Z. F. Wang, and J. Yang, arXiv:1912.01332 (2019).
15. M. Jiang, M. Berciu, and G. A. Sawatzky, arXiv:1909.02557 (2019).
16. P. Jiang, L. Si, Z. Liao, and Z. Zhong, Phys. Rev. B **100**, 201106 (2019).
17. S. Ryee, H. Yoon, T. J. Kim, M. Y. Jeong, and M. J. Han, Phys. Rev. B **101**, 064513 (2020).
18. X. Wu, D. D. Sante, T. Schwemmer, W. Hanke, H. Y. Hwang, S. Raghu, and R. Thomale, Phys. Rev. B **101**, 060504(R) (2020).
19. Y. Nomura, M. Hirayama, T. Tadano, Y. Yoshimoto, K. Nakamura, and R. Arita, Phys. Rev. B **100**, 205138 (2019).
20. A. S. Botana and M. R. Norman, Phys. Rev. X **10**, 011024 (2020).
21. G.-M. Zhang, Y.-F. Yang, and F.-C. Zhang, Phys. Rev. B **101**, 020501(R) (2020).
22. L. Si, W. Xiao, J. Kaufmann, J. M. Tomczak, Y. Lu, Z. Zhong, and K. Held, Phys. Rev. Lett. **124**, 166402 (2020).
23. M. Kitatani, L. Si, O. Janson, R. Arita, Z. Zhong, and K. Held, arXiv:2002.12230 (2020).
24. M.-Y. Choi, K.-W. Lee, and W. E. Pickett, Phys. Rev. B **101**, 020503 (2020).
25. J. Karp, A. S. Botana, M. R. Norman, H. Park, M. Zingl, and A. Millis, arXiv:2001.06441 (2020).
26. V. Olevano, F. Bernardini, X. Blase, and A. Cano, Phys. Rev. B **101**, 161102 (2020).
27. I. Leonov, S. L. Skornyakov, and S. Y. Savrasov,
arXiv:2003.04368 (2020).
E. Been, W.-S. Lee, H. Y. Hwang, Y. Cui, J. Zaanen, T. Devereaux, B. Moritz, and C. Jia, arXiv:2002.12300 (2020).
E. M. Nica, J. Krishna, R. Yu, Q. Si, A. S. Botana, and O. Erten, arXiv:2002.12300 (2020).
V. I. Anisimov, D. Bukhvalov, and T. M. Rice, Phys. Rev. B 59, 7901 (1999).
K.-W. Lee and W. E. Pickett, Phys. Rev. B 70, 165109 (2004).
J. Chaloupka and G. Khaliullin, Phys. Rev. Lett. 100, 016404 (2008).
P. Hansmann, X. Yang, A. Toschi, G. Khaliullin, O. K. Andersen, and K. Held, Phys. Rev. Lett. 103, 016401 (2009).
D. Grieger, C. Piefke, O. E. Peil, and F. Lechermann, Phys. Rev. B 86, 155121 (2012).
F. Lechermann, W. Körner, D. F. Urban, and C. Elsässer, Phys. Rev. B 100, 115125 (2019).
W. Körner and C. Elsässer, Phys. Rev. B 81, 085324 (2010).
C. Elsässer, N. Takeuchi, K. M. Ho, C. T. Chan, P. Braun, and M. Fähnle, J. Phys.: Condens. Matter 2, 4371 (1990).
F. Lechermann, F. Welsch, C. Elsässer, C. Ederer, M. Fähnle, J. M. Sanchez, and B. Meyer, Phys. Rev. B 65, 132104 (2002).
B. Meyer, C. Elsässer, F. Lechermann, and M. Fähnle, FORTRAN 90 Program for Mixed-Basis-Pseudopotential Calculations for Crystals, Max-Planck-Institut für Metallforschung, Stuttgart (1998).
P. Werner, A. Comanac, L. de’Medici, M. Troyer, and A. J. Millis, Phys. Rev. Lett. 97, 076405 (2006).
O. Parcollet, M. Ferrero, T. Ayral, H. Hafermann, I. Krivenko, L. Messio, and P. Seth, Comput. Phys. Commun. 196, 398 (2015).
P. Seth, I. Krivenko, M. Ferrero, and O. Parcollet, Comput. Phys. Commun. 200, 274 (2016).
B. Amadon, F. Lechermann, A. Georges, F. Jollet, T. O. Wehling, and A. I. Lichtenstein, Phys. Rev. B 77, 205112 (2008).
V. I. Anisimov, I. V. Solovyev, M. A. Korotin, M. T. Czyżyk, and G. A. Sawatzky, Phys. Rev. B 48, 16929 (1993).
N. Marzari and D. Vanderbilt, Phys. Rev. B 56, 12847 (1997).
I. Souza, N. Marzari, and D. Vanderbilt, Phys. Rev. B 65, 035109 (2001).
T. Li, P. Wölfle, and P. J. Hirschfeld, Phys. Rev. B 40, 6817 (1989).
F. Lechermann, A. Georges, G. Kotliar, and O. Parcollet, Phys. Rev. B 76, 155154 (2007).
A. Isidori and M. Capone, Phys. Rev. B 80, 115120 (2009).
N. Lanatà, Y. Yao, X. Deng, V. Dobrosaljević, and G. Kotliar, Phys. Rev. Lett. 118, 126401 (2017).
C. Piefke and F. Lechermann, Phys. Rev. B 97, 125154 (2018).
J. I. Facio, J. Mravlje, L. Poutrovskii, P. S. Cornaglia, and V. Vildosola, Phys. Rev. B 98, 085121 (2018).
Y.-F. Yang, Z. Fisk, H. O. Lee, J. D. Thompson, and D. Pines, Nature 454, 611 (2008).
S. Schwalow, C. Piefke, and F. Lechermann, Phys. Rev. B 85, 205132 (2012).
N. P. Armitage, P. Fournier, and R. L. Greene, Rev. Mod. Phys. 82, 2421 (2010).