Emergent flat-band physics in $d^{9-\delta}$ multilayer nickelates

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Recent experiments have shown that the reduced multilayer rare-earth (RE) nickel oxides of form RE$_{p+1}$Ni$_3$O$_{2p+2}$ may belong to the novel family of superconducting lanthanide nickelates. Here, the correlated electronic structure of Pr$_4$Ni$_3$O$_8$ and Nd$_6$Ni$_5$O$_{12}$ is studied by means of an advanced realistic many-body framework. It is revealed that the low-energy physics of both systems is dominated by an interplay of Ni-$d_{x^2-y^2}$ and Ni-$d_{z^2}$ degrees of freedom. Whilst the Ni-$d_{x^2-y^2}$ orbitals are always highly correlated near an (orbital-selective) Mott-insulating regime, the Ni-$d_{z^2}$ orbitals give rise to intriguing non-dispersive features. At low temperature, the Pr compound still displays QP-like Ni-$d_{x^2-y^2}$-derived states at the Fermi level, but the interacting fermiology of the Nd compound is outshined by an emergent Ni-$d_{z^2}$-controlling flat band. These findings translate well to the previous characterization of doped infinite-layer nickelates, and hence further make the case for a mechanism of unconventional superconductivity which is distinct from the one in high-$T_c$ cuprates.

I. INTRODUCTION

The physics of layered nickelate compounds has recently (re)gained enormous interest due to the discovery of superconductivity in Sr-doped thin films of infinite-layer NdNiO$_2$\textsuperscript{1,2}. The rise of a novel family of superconducting oxides has been confirmed by subsequent same findings in ams systems of Pr$_4$Ni$_3$O$_8$\textsuperscript{5,6} as well as of LaNiO$_2$ type\textsuperscript{1,2}.

In fact, the quest for superconductivity in $p$-layered nickelates RE$_{p+1}$Ni$_3$O$_{3p+1-(2p+2)}$ of Ruddlesden-Popper(-like) structure with rare-earth ion RE (the oxygen stoichiometry in braces marking the topotactically reduced compounds without apical O) has a rather long history\textsuperscript{7-16}. It started off from investigating single-layer La$_2$NiO$_4$, which is structurally most akin to the Cu-$d_{x^2-y^2}$-driven high-$T_c$ cuprate La$_2$CuO$_4$. But in that setting, the Ni$^{2+}$\textsuperscript{(3d}$^9$) ion turns out orbital-wise rather different from the reference Cu$^+$(3d$^9$) ion. Further layering and topotactic reduction should therefore be most effective in singling out the Ni-$d_{x^2-y^2}$ orbital in a low-energy regime for some (effective) Ni$^+$ setting\textsuperscript{8,13}. However, single crystals for the "straightforward" infinite-layer ($p\rightarrow\infty$) materials were absent until very recently\textsuperscript{17}, and therefore focus consolidated on the multilayer systems with small $p$. And indeed, single crystals of the reduced $p = 3$ system were successfully prepared by Zhang et al.\textsuperscript{15}. Yet superconductivity was still not detected, but then eventually the thin-film realization of hole-doped infinite-layer nickelates made the breakthrough\textsuperscript{1,2}. Interestingly, the optimal hole doping for the infinite-layer compounds can effectively also be realized for systems with $p \sim 5, 6$ layers\textsuperscript{15,18,19}. Actually, in a very recent study Pan et al.\textsuperscript{20} reported superconductivity in the reduced $p = 5$ compound Nd$_6$Ni$_5$O$_{12}$.

This story has a further level of complexity, since the actual electronic structure and the resulting superconducting scenario of the infinite-layer nickelates might be not of straightforward cuprate kind and is still heavily debated (see e.g. Refs. 19, 21–24 for recent reviews).

Main issues are the degree of correlation strength, the role of apparent self-doping bands, and the relevance of a Ni-$d_{x^2-y^2}$ vs. a Ni-multiorbital setting at low energy. Against this challenging background, we here report a comparing first-principles many-body investigation of Pr$_4$Ni$_3$O$_8$ ($p = 3$) and the recently highlighted Nd$_6$Ni$_5$O$_{12}$ ($p = 5$).

In our previous studies of infinite-layer nickelates\textsuperscript{25-28}, we argued that besides local Coulomb interactions on Ni, an effective inclusion of such local interactions on the O sites is indispensable to arrive at a reliable picture of the correlated electronic structure. Based on that viewpoint, it turns out that the infinite-layer nickelate physics is dominated by the intriguing interplay between Ni-$e_g\{d_{x^2}, d_{z^2}\}$ multiorbital degrees of freedom. While Ni-$d_{x^2-y^2}$ is orbital-selective Mott insulating and hardly doped with holes, the Ni-$d_{z^2}$ orbital eagerly collects hole carriers and shifts as a flat band in the $k_z = 1/2$ plane of the Brillouin zone across the Fermi level in the superconducting doping region. Main result of the present work is that this emergent flat band physics in front of a highly correlated Ni-$d_{x^2-y^2}$ state is indeed also relevant for $d^{9-\delta}$ multilayer nickelates, consistent with the comparable effective hole doping for superconducting infinite-layer and multilayer nickelate. Importantly, for the Pr ($p = 3$) compound, the flat band has already passed the Fermi level, dissolving into incoherent (Hund-driven) excitations. However, it is about “in resonance” with the Fermi energy for the Nd ($p = 5$) compound. This underlines the decisive role of the Ni-$d_{z^2}$-dominated flat-band physics for the stabilization of the superconducting phase in infinite- and multilayer nickelates.

II. THEORETICAL APPROACH

To reveal the correlated electronic structure of Pr$_4$Ni$_3$O$_8$ and Nd$_6$Ni$_5$O$_{12}$, the charge self-consistent combination\textsuperscript{29,30} of density functional theory (DFT), dynamical mean-field theory (DMFT) and self-interaction correction (SIC) is employed\textsuperscript{31}. The DFT part of this
DFT+sicDMFT scheme builds up on a mixed-basis pseudopotential framework\textsuperscript{32–34} in the local density approximation (LDA). We address the Coulomb interactions on oxygen furthermore beyond DFT within SIC on the pseudopotential level\textsuperscript{35–37}. Whereas the O(2s) orbital is by default fully corrected with a weight factor $w_{2s} = 1.0$, the reasonable choice\textsuperscript{31,37} $w_{2p} = 0.8$ is utilized for the O(2p) orbitals. The further screening parameter $\alpha$ for this SIC pseudopotential is chosen also as $\alpha = 0.8$, such that the SIC inclusion on O asks for one additional parameter in the overall computational scheme. For a further discussion of the relevance of the use of SIC on O in layered nickelates we refer to Ref. 27. Finally, the Ni sites act as quantum impurity problems in multi-site realistic DMFT, with the site-resolved correlated subspace consisting of the full Ni(3d) shell, respectively. The whole scheme is converged until self-consistency in the charge density and the self-energies is reached. Note again that the SIC incorporation enters the given framework as a modified (pseudo)potential, and is not a ‘fixed correction’ or ‘shift’ on the sole DFT level.

In the following the calculational settings are described in more detail. A $11 \times 11 \times 11$ k-point mesh is utilized for Pr$_4$Ni$_3$O$_8$, and a $5 \times 5 \times 5$ one for Nd$_4$Ni$_5$O$_{12}$. The plane-wave cutoff energy is set to $E_{\text{cut}} = 13$ Ry and localized basis orbitals are introduced for Pr$_5$/Nd(5d), Ni(3d) as well as O(2s, 2p). The Pr/Nd(4f) states are put in the pseudopotential frozen core, since they are not decisive for the key physics of infinite-layer nickelates\textsuperscript{38}. The $j$-resolved frozen occupation of the Pr/Nd(4f) shell is chosen with small resulting moment and only scalar-relativistic effects enter the general pseudopotential generation. The role of spin-orbit effects in the overall crystal calculations is neglected. The DMFT correlated subspace on each Ni site is governed by a full Slater Hamiltonian applied to the Ni(3d) projected-local orbitals\textsuperscript{39}. The projection is performed on the $N(O) \times 3 + N(Ni) \times 5 + N(\text{RE})$ Kohn-Sham (KS) states above the dominant O(2s) bands, associated with O(2p), Ni(3d) and possibly relevant RE-based bands. Here, $N(\text{element})$ amounts to the element–specific number of atoms in the unit cell, e.g. four for Pr$_4$Ni$_3$O$_8$ and five for Nd$_4$Ni$_5$O$_{12}$. This choice resembles the use of one additional KS state, i.e. $6+5+1$ in infinite-layer nickelates RENiO$_2$\textsuperscript{25–28}. A Hubbard $U = 10$ eV and a Hund exchange $J_H = 1$ eV prove reasonable for this choice of the energy window\textsuperscript{25,31}. The fully-localized-limit double-counting scheme\textsuperscript{40} is applied. Continuous-time quantum Monte Carlo in hybridization expansion\textsuperscript{41} as implemented in the TRIQS code\textsuperscript{42,43} is used to solve the DMFT problem. Two different system temperatures, namely $T = 193$ K and $T = 50$ K are chosen to reveal relevant coherence effects. Up to $1.5 \cdot 10^9$ Monte-Carlo sweeps are performed to reach convergence. A Matsubara mesh of $1025(2049)$ frequencies is used to account for the higher(lower)-temperature regime. Maximum-entropy\textsuperscript{44} and Padé\textsuperscript{45} methods are employed for the analytical continuation from Matsubara space onto the real-frequency axis. All calculations are conducted for a paramagnetic regime, respectively.

### III. INITIAL CHARACTERIZATION OF THE $p = 3, 5$-LAYER NICKELATES

The reduced layered nickelates of type RE$_{p+1}$Ni$_p$O$_{2p+2}$ (see Figs. 1a,c) crystallize in a tetragonal $I4/mmm$ space group and host blocks of $p$ NiO$_2$ square-lattice layers separated by RE layers. These blocks of NiO$_2$ and RE layers are again separated by fluorite-like REO slabs, increasing the two-dimensional character of the compounds. The here studied systems Pr$_4$Ni$_3$O$_8$ and Nd$_4$Ni$_5$O$_{12}$ are chosen for the following reasons. First, the nominal average Ni(3d) occupation amounts to\textsuperscript{46} $\delta^8.67$ for $p = 3$ and to $\delta^8.80$ for $p = 5$. Since the superconducting region of the infinite-layer nickelates nominally spans between $\sim \delta^8.75$–$8.85$, the $p = 5$ compound should effectively be prone to superconductivity, while the $p = 3$ compound should reside in the hole-overdoped regime. Thin films of Nd$_4$Ni$_5$O$_{12}$ have indeed been shown to exhibit superconducting properties\textsuperscript{20}. For the $p = 3$ case we select Pr$_3$Ni$_3$O$_8$ since it was prepared in single-crystal form\textsuperscript{47,48} with metallic behavior down to lowest temperatures.

The trilayer Pr compound has two symmetry-inequivalent Ni classes: the Ni1 class corresponding with the inner NiO$_2$ layer and the Ni2 class associated with the outer Ni2 layer. The quintuple-layer Nd compound has three such different classes: Ni1 for inner, Ni2 for intermediate and Ni3 for outer NiO$_2$ layer. Lattice parameters $a = 3.935$ Å and $c = 25.485$ Å, as well as atomic positions are here overtake from experiment\textsuperscript{47} for Pr$_3$Ni$_3$O$_8$. In the case of Nd$_5$Ni$_5$O$_{12}$, the experimental $c$-axis parameter of the thin-film study\textsuperscript{20}, i.e. $c = 38.8$ Å, and the inplane $a = 3.92$ Å from NdNiO$_2$\textsuperscript{1} are used. The atomic positions for the Nd compound are obtained via DFT structural relaxation using the generalized-gradient approximation. This results in a somewhat stronger buckling of the non-NiO$_2$ layers compared to the one in the Pr compound (compare Figs. 1a,c).

The DFT characterization (to be compared with previous studies\textsuperscript{20,46,49–52}) of both multilayer compounds is summarized in Figs. 1b,d. We choose a fatspec representation for the band structures. It colors the spectrum according to the orbital weight in the spectral function $A(k, \omega)$ at given $(k, \omega)$ point. Of course, in the DFT limit, $A(k, \omega)$ reduces to the KS band structure. Note that this fatspec representation differs slightly from the usual ‘fatband’ picture, since overlayed/crossing bands and thus non-hybridized energy areas may also appear as ‘mixed-orbital’ character. For instance, a Ni-$d_{z^2}$-dominated dispersion (magenta-colored) starts in the top panel of Fig. 1b at $\sim -1$ eV and closes at $X$ with energy $\sim -0.75$ eV. It appears blue-colored inbetween, because it overlays with Ni-$t_{2g}$ (cyan-colored) in that energy range. Surely, the truly mixed hybridization on an individual dispersion is also correctly signalled. Here, the fatspec representation focuses on the Ni(3d) nature of
FIG. 1. (color online) Crystal structure and DFT spectrum of Pr$_4$Ni$_3$O$_8$ (a,b) and Nd$_6$Ni$_5$O$_{12}$ (c,d). (a) Structure of the p = 3 Pr compound with Pr (blue), Ni(grey) and oxygen (red); Ni1 and Ni2 class with associated NiO$_2$ layers are indicated. (b) Band structure along high-symmetry lines (top) and Fermi surface (bottom) of Pr$_4$Ni$_3$O$_8$ in Ni(3d)-fat-spec (see text) representation. Note that mixed colors represent joint contributions from the involved orbitals, e.g. red color corresponds to a mixing of Ni-$d_{x^2-y^2}$ (yellow) and Ni-$d_z$ (magenta). The O(2p)-based block of bands is drawn with full grey lines. (c) Structure of the $p = 5$ Nd compound with Nd (green), Ni(grey) and oxygen (red); Ni1, Ni2 and Ni3 class with associated NiO$_2$ layers are again indicated. (d) Same as (b), but for Nd$_6$Ni$_5$O$_{12}$.

the electronic structure.

Starting with Pr$_4$Ni$_3$O$_8$, it is seen that the Ni-$t_{2g}$-dominated bands are mostly full and only the Ni-$e_g$-derived bands are partially occupied. Note that there is no self-doping band as in infinite-layer nickelates$^{25}$, the corresponding bands with sizable Ni-$d_{x^2-y^2}$, Ni-$t_{2g}$ and Pr(5d) character are well above the Fermi level $\varepsilon_F$. Along $\Gamma$-$X$, one band of mixed Ni-$e_g$ type crosses $\varepsilon_F$, while along $X$-$M$ there are two dominant Ni-$d_{x^2-y^2}$ dispersions at low-energy. In other words, there are $p$ Ni-$e_g$ bands crossing the Fermi level This results in a three-sheeted Fermi surface (FS) for the Pr compound, with an electron-like mixed Ni-$e_g$ inner sheet centred around $\Gamma$ and two close-running Ni-$d_{x^2-y^2}$-based hole-like sheets. The latter are reminiscent of the hallmark hole sheet in high-$T_c$ cuprates.

The DFT spectrum of the Nd compound displays further-increased complexity (see Fig. 1d). Now five Ni-$e_g$-based bands cross $\varepsilon_F$; a strongly-mixed one again along $\Gamma$-$X$ and four along $X$-$M$. Three out of these latter four bands are dominantly of Ni-$d_{x^2-y^2}$ kind and the highest-lying one with some Ni-$d_{z^2}$ mixed in. Note that the mixed Ni-$e_g$ dispersion crossing in $\Gamma$-$X$ direction has its van-Hove point at X closer to the Fermi level than in the Pr compound. Furthermore, there are two additional self-doping bands, one close to $\Gamma$ and the second close to M. The first one has sizable Ni-$d_{z^2}$ flavor, whereas the one next to M has also Ni-$t_{2g}$ mixed in. Accordingly, besides the five-sheeted dominant Ni-$e_g$ fermiology, the FS exhibits two additional electron-like pockets around $\Gamma$ and M. As a further difference to Pr$_4$Ni$_3$O$_8$, the most-mixed Ni-$e_g$ sheet (i.e. red-colored in bottom panel of Figs.1b,d) is larger and has more square shape in the Nd compound. Interestingly, the self-doping bands are seemingly missing in the $p = 5$ compound of La type$^{46,52}$, but are also observed in the DFT result for Nd$_6$Ni$_5$O$_{12}$ from Ref. 51.

Finally, we want to comment on the charge-transfer energy $\Delta = \varepsilon_d - \varepsilon_p$, whereby $\varepsilon_d$,$\varepsilon_p$ are the respective band centre of Ni(3d) and O(2p). Using the DFT+$+$sc approach, a value $\Delta_{\text{NdNiO}_2} = 5.0$ eV was obtained for infinite-layer NdNiO$_2$.$^{25}$ For the present multilayer systems, calculations from the same perspective result in $\Delta_{\text{Nd}_6\text{Ni}_5\text{O}_{12}} = 4.2$ eV and $\Delta_{\text{Pr}_4\text{Ni}_3\text{O}_8} = 3.9$ eV. This drop of the charge-transfer energy with an increase of the Ni effective oxidation state is in line with previous findings.$^{28,53}$

### IV. DFT+$+$sicDMFT RESULTS

#### A. Pr$_4$Ni$_3$O$_8$

Let us turn to the interacting problem beyond DFT by first focussing on the trilayer Pr compound. Intuitively, from the anticipated similarity to the overdoped infinite-layer system$^{27}$, one may expects the correlated electronic structure of Pr$_4$Ni$_3$O$_8$ to be in a Hund(-like) regime where coherence effects and possible non-Fermi-liquid (NFL) characteristics due to an interplay of Ni-$d_{z^2}$
and Ni-$d_{x^2-y^2}$ may be important. Figure 2a shows the k-resolved spectrum at $T = 193$ K $(\beta = 1/T = 60$ eV$^{-1})$, with an indeed absence of well-defined quasiparticle (QP) dispersions at the Fermi level. The Ni-$d_{x^2-y^2}$-dominated states are very strongly correlated with large scattering rate for Ni1 and an even diverging imaginary Matsubara self-energy for Ni2 (cf. orange circle-dashed lines in right panel of Fig. 3a). The fatspec representation reveals that the Ni-$t_{2g}$-dominated bands remain well below $\varepsilon_F$. Furthermore, it displays that the near-dispersionless spectral weight at $\sim 0.15$ eV above the Fermi level is of Ni-$d_{z^2}$ character. Further such character with somewhat more (incoherent) dispersion may be observed in a similar energy range below $\varepsilon_F$.

Lowering the temperature to $T = 50$ K leads to a significant increase of the Ni-$d_{x^2-y^2}$ coherence, resulting in highly-renormalized QP dispersions as shown in Fig. 2b,c. This Fermi-liquid (FL) regime is also observable from the associated Ni-$d_{x^2-y^2}$ self-energies for both Ni sites (cf. orange square-full lines in right panel of Fig. 3a). The effective mass $m^*/m_{\text{DFT}} \approx 14$ turns out very large for this orbital sector. In addition, notably, the impact of temperature, leading also to energy shifts for the Ni-$t_{2g}$-dominated bands, is stronger than in stoichiometric infinite-layer NdNiO$_2$, where Ni-$d_{x^2-y^2}$ resides in an effective Mott-insulating regime throughout the accessible temperature range$^{25,26}$. However as seen in Fig. 2c, there is still strong incoherent Ni-$d_{z^2}$ spectral weight at and close to the Fermi level, which adds up to an appreciable total low-energy weight upon k-integration for both symmetry-in equivalent Ni sites (cf. Fig. 3b). The incoherent nature of this Ni-$d_{z^2}$ weight is also drawn from the displayed imaginary part of the corresponding self-energies in Fig. 3a (cf. magenta square-full lines in left panel): for Ni1 it shows a clear NFL upturn at small Matsubara frequencies, and for Ni2 the bending in the same frequency range is too strong to account for the linear-frequency behavior of a FL. In other words, the electronic regime that one encounters in Pr$_4$Ni$_3$O$_8$ is that of an partly incoherent metal with still highly-renormalized Ni-$d_{x^2-y^2}$ QP(-like) dispersions. Concerning the aforementioned connection of effective Ni charge states, this finding matches with a principal Hund-metal identification of the overdoped infinite-layer regime$^{27}$.

The total spectral function $A(\omega)$ (see top panel of Fig. 3b), besides the metallic weight at $\varepsilon_F$, shows a dominant Ni($3d$) peak at $\sim -0.5$ eV and a dominant broader O($2p$) peak at $\sim -4.5$ eV. Those dominant $d$ and $p$ peaks are shifted towards the Fermi level compared to stoichiometric NdNiO$_2$, again understandable from the doping connection. Note that there is also some non-minor Pr($5d$) weight located within the broader O($2p$)-dominated peak, associated with the Pr sites in the fluorite-like REO slab. The Zhang-Rice character$^{54}$ of the low-energy spectral weight, i.e. a spectral sharing between Ni($3d$) and O($2p$), is small; the latter weight is dominantly of Ni($3d$) type. In view of the Ni1,Ni2 differentiation, the inner Ni1 site exhibits a somewhat stronger Ni-$d_{x^2-y^2}$ low-energy peak structure than the outer Ni2 site. But this weight is of pseudogap structure for both sites, possibly due to the coupling to the NFL-behaving Ni-$d_{z^2}$ states.

Table I provides the Ni($3d$) fillings, and while the values for Ni-$d_{x^2-y^2}$ are always close to half filling, the Ni-$d_{z^2}$ orbitals are mainly controlling the hole content. For Pr$_4$Ni$_3$O$_8$, the filling $n(d_{z^2})$ is lowest with an average value $\sim 1.45$ and some larger filling for the outer Ni2 site (translating to a slight overall larger $n(3d)$ filling for Ni2).
Fig. 3. (color online) Ni-ε_{0} self-energies (a) and k-integrated DFT+sicDMFT spectrum (b) of Pr_{2}Ni_{3}O_{8}. (a) Imaginary part of the Ni-d_{2} (left) and the Ni-d_{x^2-y^2} (right) Matsubara self-energy Σ(ω_{n}) for Ni1 (top) and Ni2 (bottom) at T = 193 K (circle-dashed) and T = 50 K (square-full). (b) Top: total as well as element- and site-resolved projected spectral function; middle and bottom: local Ni1(3d) and Ni2(3d) spectral function at T = 193 K (dashed) and T = 50 K (full), respectively. Inset: low-energy window.

Albeit far off from a formal Ni^{2+}, Ni^{2+} dichotomy, the larger Ni(3d) charge on Ni2 is qualitatively in line with this simple picturing. Note that the theoretical filling values from DFT+sicDMFT, as usual depending on e.g. the given definition of the correlated subspace, are generally somewhat smaller than from the simplistic aforementioned oxidation-state analysis for p = 3, 5. From a direct theory comparison to calculations for hole-overdoped Nd_{1-x}Sr_{x}NiO_{2} with x = 0.3, the present n(d_{z^2}) for the trilayer compound is also significantly lower. Thus the comparison between multilayer and doped-infinite-layer physics has still to be performed with caution.

Let us finally compare to experiment. Zhang et al.\cite{27} reported metallic behavior down to lowest T, with however a rather high room-temperature resistivity of 67.2 Ω cm. Additionally, the specific-heat measurements from that work give rise to a large γ ∼ 75 mJ mol^{-1} K^{-2}. In comparison, for La_{2}NiO_{3} a value γ ∼ 15 mJ mol^{-1} K^{-2} is obtained\cite{56} and a mass enhancement of about 3-4 inferred. Disregarding possible Pr(4f) physics, an enormous formal mass enhancement of ∼15 for Pr_{2}Ni_{3}O_{8} is thus not completely out of range. Magnetic properties have been measured by Huangfu et al.\cite{57} showing complex magnetic behavior without ordering down to lowest T. But the data hints to a coexistence of localized spins and itinerant degrees of freedom.

### B. Nd_{6}Ni_{5}O_{12}

Very much in parallel to the discussion of the correlated electronic structure of Pr_{2}Ni_{3}O_{8} in the last section, the focus is henceforth on the p = 5 Nd compound. Figure 4 displays the k-resolved data, with obvious low-energy differences to the trilayer Pr system. The seemingly non-dispersive spectral weight just above the Fermi level for the latter compound, appears here more or less right at ε_{F} for both investigated temperatures (cf. Fig. 4a,b). At T = 193 K, this Fermi-level spectral weight is again very incoherent, and the Ni-d_{x^2-y^2} self-energies are (about to) diverging at low frequency (see Fig. 5a). In the low-energy window of Fig. 4c for T = 50 K, there is some strongly renormalized Ni-d_{x^2-y^2} dispersion identifiable, but its intensity seems reduced compared to the one in Pr_{2}Ni_{3}O_{8}. Instead, the non-dispersive Ni-d_{z^2} weight right at ε_{F} turns out stronger for the present quintuple-layer Nd compound.

Note that the self-doping bands forming electron pockets around Γ and M in the previous DFT picture are shifted well-above the Fermi level and do not play a role in the fermiology. Compared to infinite-layer NdNiO_{2},

| compound       | Ni site | n(d_{z^2}) | n(d_{x^2-y^2}) | n(3d) |
|----------------|--------|------------|---------------|-------|
| Pr_{2}Ni_{3}O_{8} | Ni1    | 1.40       | 1.13          | 8.47  |
|                 | Ni2    | 1.48       | 1.11          | 8.53  |
| Nd_{6}Ni_{5}O_{12} | Ni1   | 1.65       | 1.08          | 8.67  |
|                 | Ni2    | 1.54       | 1.09          | 8.57  |
|                 | Ni3    | 1.56       | 1.08          | 8.58  |
| NdNiO_{2}       | Ni     | 1.85       | 1.07          | 8.86  |
| Nd_{5.5}Sr_{0.5}NiO_{2} | Ni     | 1.76       | 1.04          | 8.74  |
| Nd_{5.7}Sr_{0.3}NiO_{2} | Ni     | 1.60       | 1.08          | 8.58  |

TABLE I. Site and orbital-resolved Ni(3d) fillings n in the p = 3, 5-layer compounds from DFT+sicDMFT at T = 50 K. The Ni-d_{z^2} filling varies only marginally and is always close to n(t_{2g}) = 5.94. The comparing data\cite{27} for stoichiometric and hole-doped infinite-layer NdNiO_{2} was obtained for T = 30 K.
where the self-doping band ‘survives’ correlation effects and remains at the stoichiometric Fermi level, this is understandable for two main reasons. First the charge-transfer from Pr(5d) to Ni(3d) in order to realize a favorable (near) half-filled Ni-$d_{x^2-y^2}$ scenario in a strong-coupling limit is apparently larger for $p = 5$. Second, the self-doping bands carry quite some Ni-$d_{z^2}$ weight in both cases, however importantly, in the quintuple-layer compound the filling $n(d_{z^2})$ is way smaller than in NdNiO$_2$ (cf. Tab. I). Therefore, correlations in Ni-$d_{z^2}$ are stronger, enabling a larger correlation-induced shifting of the corresponding spectral weight. As for the trilayer system, the Ni-$t_{2g}$-dominated states are mostly full. Yet there is a residual Ni-$d_{z^2}/t_{2g}$ hybridization visible in the fatspec-representation panel of Fig. 4c, extending towards the Fermi level.

Finally, the fermiology at $T = 50$ K looks quite different from the one of Pr$_x$Ni$_3$O$_8$ and rather intriguing (see Fig. 4d). The role of the previously dominant Ni-$d_{x^2-y^2}$ states is hard to decipher, but much weaker (to say the least). The original hole-like topology from this orbital sector becomes only well tractable somewhat above the Fermi level close to the M point. Apparently, the coherence scale for (possibly) robust QP-like Ni-$d_{x^2-y^2}$ excitations has not yet reached at $T = 50$ K for Nd$_6$Ni$_5$O$_{12}$. Also therefore, mass-enhancement estimates are not too well-defined in this case. Yet from a crude examination of the low-frequency Ni self-energy, the $m^*$ is about the twice the value than for the trilayer Pr compound. The interacting Fermi surface appears to be dominated by a correlated flat-band Ni-$d_{z^2}$ sheet around $\Gamma$. Further conclusive FS details are hard to draw from the generally low-coherence level at this temperature. Lowering the temperature much further is however numerically tough for the given theoretical approach to these multilayer systems. But still, the present results for the two $T$ scales render the fact robust, that emergent flat-band physics of Ni-$d_{z^2}$ type rules the low-energy response. Form a site-averaged view onto the Ni-$d_{z^2}$ selfenergies shown in the left panel of Fig. 5a, the correlation effects on this flat band seem somewhat weaker than for the similarly non-dispersive parts in the Pr compound. There are no very strong irregular features, however the low-frequency bending for the Ni3 site is still NFL-like.

The $k$-integrated spectra plotted in Fig. 5b are for the total content similar to the Pr$_x$Ni$_3$O$_8$ case, with a somewhat stronger low-energy peak at $\varepsilon_F$ and slightly deeper-energy location of the Ni(3d) and O(2p) peaks. The Zhang-Rice nature of the low-energy weight remains minor. The site-resolved Ni spectra exhibit seemingly a monotonic Ni-$\varepsilon_g$ evolution at low energy, in the sense that the Ni-$d_{z^2}$ character grows from inner-to OUTER-layer and vice versa for Ni-$d_{x^2-y^2}$ (with similar pseudogap structure as for the trilayer compound). The emergent flat-band physics appears to be dominantly driven from the outer Ni3 site. Interestingly, the Ni(3d) charge hierarchy between inner and outer layers turn out reversed in Nd$_6$Ni$_5$O$_{12}$; the inner Ni1 site has a slightly higher $d$ count than Ni2,Ni3 by about 0.1 electrons (see Tab. I).

Comparison to experiment is even more difficult in the quintuple-layer case. First, there is so far only the thin-film work by Pan et al. on this system. Second, even for $T = 50$ K, the theoretical spectrum/fermiology has not yet completely settled in terms of coherence. The measured room-temperature resistivity of $\sim 6 \, \text{m\Omega cm}$ yet is way smaller than for single-crystal Pr$_4$Ni$_3$Os, which could be in favor of a high density of states at $\varepsilon_F$. The experimental Hall coefficient is positive over the whole accessible $T$ range. A negative sign of this coefficient for stoichiometric NdNiO$_2$ is understandable from the self-doping electron pockets, and indeed, such pockets are missing in the interacting regime of the present multilayer

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**FIG. 4.** (color online) Same as Fig. 2 but for Nd$_6$Ni$_5$O$_{12}$. 
V. DISCUSSION

The revealed data for the paramagnetic correlated electronic structure of Pr$_4$Ni$_5$O$_8$ and Nd$_6$Ni$_5$O$_{12}$ shows that the two compounds share certain similarities, but possess a rather different low-energy nature. Similar to both systems is the way higher susceptibility to temperature effects than for the stoichiometric NdNiO$_2$ compound. Akin to the infinite-layer nickelate is the orbital-selective scenario of a highly correlated, (near) Mott-insulating Ni-$d_{x^2-y^2}$ orbital sector and an itinerant Ni-$d_z^2$ sector. However while the latter is close to complete filling and only weakly-correlated in stoichiometric NdNiO$_2$, the correlation effects are here more severe due to a lower filling with stronger overall Ni-$e_g$ inter-orbital processes. These processes are apparently based on a sophisticated interplay between orbital-selectivity, Hund-driven mechanisms and flat-band physics within the two-orbital Ni-$e_g$ manifold. The very details of this interplay have to be addressed in a tailored model-Hamiltonian study and are not subject of the present investigation. But as result thereof, due to a different doping scenario, the trilayer Pr compound still displays Ni-$d_{x^2-y^2}$ QP(-like) excitations within a background of non-dispersive incoherent Ni-$d_z^2$ states, whereas Nd$_6$Ni$_5$O$_{12}$ exhibits a more incoherent Ni-$d_{x^2-y^2}$ dispersion coexisting with Ni-$d_z^2$ flat-band features.

Trying to rationalize this behavior, let us first focus on the Ni-$d_{x^2-y^2}$ part. From DFT+stcDMFT calculations for NdNiO$_2$, we learned that Ni-$d_{x^2-y^2}$ is effectively Mott-localized at stoichiometry and remains incoherent upon hole doping until a specific regime within the superconducting doping region. In the infinite-layer overdoped region it becomes again incoherent. Thus the Ni-$d_{x^2-y^2}$ electrons behave very differently compared to the Zhang-Rice-bound carriers in high-$T_c$ cuprates, where coherent QPs (modulo the notorious pseudogap regime) are robust over a wide doping ranging starting just beyond stoichiometry. The strong charge-transfer character of compounds. But the very intriguing (incoherent) fermiology shown in Fig. 4d does not really allow for more serious conclusions on this issue. At least, many-body calculations of the Hall coefficient for optimally-doped infinite-layer NdNiO$_2$ results also in a positive sign when having a previously-predicted Ni-$d_z^2$ flat-band at the Fermi level.

FIG. 5. (color online) Same as Fig. 3 but for Nd$_6$Ni$_5$O$_{12}$. The Ni site-resolved data spans here over Ni1, Ni2 and Ni3.

FIG. 6. (color online) Emergent flat-band character from the band-resolved correlated spectral function $A_{\nu}(\omega)$ (see text). Left: Pr$_4$Ni$_5$O$_8$ and right: Nd$_6$Ni$_5$O$_{12}$. Inset: focus around the Fermi level. The non-black colored functions denote the $\nu$-resolved $z\nu$-crossing spectral weight with strongest near-$\omega = 0$ contribution. Note that the coloring in terms of Ni-$d_z^2$ and Ni-$d_{x^2-y^2}$ marks only dominance and is for illustration, since this is not a fatspec/band plot.
these cuprates, lacking in present nickelates, appears to be main origin for this. But then it is still surprising that Pr$_4$Ni$_3$O$_8$ shows QP(-like) physics in this nominally overdoped regime from the infinite-layer perspective. Reason could be that a strong similarity between trilayer and infinite layer just breaks down in the overdoped region. For instance, the additional degree of freedom of Ni-site differentiation might allow for electronic relaxation such as to render QP propagation more robust. Another aspect may be the fact that the lanthanide neighboring compounds La$_4$Ni$_3$O$_8$ and Nd$_4$Ni$_3$O$_8$ are experimentally identified insulating at low temperature$^{15,50}$ (though the trilayer Nd compound may be metallic in thin-film geometry$^{20}$). Thus the competition between metal and insulator is seemingly tight, and a QP-based metallicity of the Pr compound may also be based on subtleties that are outside the general rule.

Concerning the Ni-$d_{x^2}$ part, the difference between the 'non-dispersive' dispersion just above the Fermi level in the trilayer Pr compound and the 'flat band' right at $\varepsilon_F$ in the quintuple-layer Nd compound has to be understood. Key is the observation that as in doped NdNiO$_2$ a flat band (there in the $k_z = 1/2$ plane because of a missing layer differentiation) shifts through the Fermi surface with growing hole content. This is illustrated in Fig. 6, where the band-resolved correlated spectral function $A_\nu(\omega)$ is plotted. The function $A_\nu(\omega)$ results from upfolding the local DMFT Green's function to the original crystal Hilbert space, i.e. displays how strong each original KS band is dressed with correlations and contributes to the total interacting spectrum. The red/magenta-highlighted $A_\nu$ mark the strongest non-Mott-critical contributions to the Fermi spectral weight. In the case of Pr$_4$Ni$_3$O$_8$ (left pane of Fig. 6), there are two such contributions, whereby especially the magenta-colored one still shows sizable sign of correlation (i.e. sideband features). The non-dispersive part at $\sim 0.15$ eV above the Fermi level in Fig. 2c may be identified with the upper sideband of that latter $A_\nu$, peaking just a bit below $\varepsilon_F$. The sideband energy scale is too small for an ordinary upper Hubbard band, so a Hund-mediated origin is likely. Note that hence only two of the three original KS bands that cross the Fermi level are nearly Mott-insulating (cf. inset, orange-colored lines in Fig. 6). To make contact to the original DFT band structure (see Fig 1b): the here red-colored $A_\nu$ connects to the highest Fermi-level KS band with mixed Ni-$d_{z^2}$/Ni-$d_{x^2}-y^2$ character; the here magenta-colored $A_\nu$ connects to the highest-occupied KS Ni-$d_{x^2}$-dominated dispersion, starting at $\sim -1$ eV at $\Gamma$ and closing at $\sim -0.75$ eV at $X$. Hence intrinsically, a KS-occupied band is partly depleted in the interacting regime and causes the non-dispersive sideband feature in DFT+sicDMFT.

On the other hand for the Nd$_4$Ni$_3$O$_{12}$ compound, it is easily observed in the right panel of Fig. 6 that there is one certain $A_\nu$ of strong Ni-$d_{x^2}$ nature dominating right at the Fermi level. It is again connected to the original-KS Ni-$d_{x^2}$-dominated dispersion just below the Ni-$d_{x^2}-y^2$-dominated bands (cf. Fig 1d), and describes the emergent flat band at $\varepsilon_F$. Hence in both multilayer cases, the Ni-$d_{x^2}$ dispersions below the Ni-$d_{x^2}-y^2$-dominated ones are an important key to the problem. However it only evolves into an emergent flat band in the quintuple-layer case. In the trilayer case, since at a nominal higher hole-doping level, the formerly flat-band part of the Ni-$d_{x^2}$ dispersion has already crossed the Fermi level and dissolves into stronger (Hund-)correlated states with incoherent spectral weight. Note that as a further difference, the topmost mixed Ni-$d_{x^2}$/Ni-$d_{x^2}-y^2$ dispersion (red color in Fig. 6) is stronger correlated in the Nd compound.

Let us finally note, that recent DFT+DMFT studies on multilayer nickelates$^{50-52}$ differ from the here established low-energy picture. From those works, Ni-$d_{x^2}$-influenced physics is mainly negligible and moderately-to-strongly correlated Ni-$d_{x^2}-y^2$ physics (yet distant from an obvious Mott-critical regime) is dominating. Key difference lies in the neglect of oxygen-based correlations in Refs. 50–52, which renders the layered nickelates (much) weaker correlated than described here. Future experiments have to decide which correlation regime and low-energy picture is more fitting. Note in that respect, that a recent photoemission study$^{60}$ for stoichiometric thin-films of infinite-layer PrNiO$_2$ reports an electronic spectrum in very good accordance with the DFT+sicDMFT predicted$^{25,27}$ spectrum for NdNiO$_2$.

VI. CONCLUSION

The present DFT+sicDMFT study predicts emergent flat-band character of Ni-$d_{x^2}$ type in the quintuple-layer nickelate Nd$_4$Ni$_3$O$_{12}$. It is due to the formation of near Mott-insulating Ni-$d_{x^2}-y^2$ states in an orbital-selective manner within the Ni-$e_g$ subshell, in conjunction with the effective (optimal) hole doping compared to stoichiometric infinite-layer NdNiO$_2$. Multiorbital correlations of seemingly Hund-driven kind are also observable, but those are stronger for the trilayer nickelate Pr$_4$Ni$_3$O$_8$. The latter compound is in a nominal hole-overdoped regime compared to NdNiO$_2$, however still displays as QP-like Ni-$d_{x^2}-y^2$-dominated fermiology. Whilst the principal mapping between doped infinite-layer and multilayer nickelates exists as envisioned, some caution has to be taken concerning the details. Still in the end, the present results for the $p = 3, 5$ multilayers are consistent with our previous results on infinite-layer nickelates$^{25–28}$: the eventually arising superconductivity should be unconventional in a non-cuprate manner, with the Ni-$e_g$ interplay, and especially the Ni-$d_{x^2}$ flat-band physics at the Fermi level, as a key driving force.
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