

Correlations tune the electronic structure of pentalayer nickelates into the superconducting regime

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Motivated by the recent discovery of superconductivity in the pentalayer nickelate $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ [G. A. Pan *et al.*, *Nat. Mater.* **21**, 160 (2022)], we calculate its electronic structure and superconducting critical temperature. We find that electronic correlations are essential for pushing $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ into the superconducting doping range as they shift the electron pockets above the Fermi energy. As a consequence, $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ can be described with a single $d_{x^2-y^2}$ orbital per Ni. We predict the pentalayer to be overdoped; further improvements in T_c can be expected for a few additional layers. Instead, for the bilayer nickelate $\text{Nd}_3\text{Ni}_2\text{O}_6$ we find correlations to drive the system into a three-orbital regime also involving the Ni $d_{xz,yz}$ states. We suggest, however, that single-orbital physics with optimal doping can be restored by substituting 60% of the trivalent Nd or La by tetravalent Zr.

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Introduction. Even 35 years after the discovery of high temperature (T_c) superconductivity in cuprates [1], understanding the microscopic mechanism and identifying the necessary ingredients for a minimal model remains arguably one of the biggest challenges of solid state theory. The recent synthesis of nickelate superconductors $\text{Sr}(\text{Ca})_x\text{Nd}(\text{La},\text{Pr})_{1-x}\text{NiO}_2$ [2–6] provides a new perspective and has awoken new hope for this quest, not least because nickelates are very similar, but, at the same time, also very distinct from cuprates. In conjuncture with the cuprates they are hence ideally suited to distinguish the essential from the incidental for high- T_c superconductivity. That is, $\text{Nd}(\text{La},\text{Pr})\text{NiO}_2$ shares the same infinite layer structure and formal $3d^9$ valence with the simple cuprate superconductor CaCuO_2 [7–9]. However, besides the $d_{x^2-y^2}$ band, additional electron pockets are present at the $\text{Nd}(\text{La},\text{Pr})\text{NiO}_2$ Fermi surface, as identified in electronic structure calculations [10–21] and evidenced by a negative Hall coefficient [2,3].

Since infinite layer cuprates are not the best superconductors, finding superconductivity in a finite layer nickelate, the undoped pentalayer $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ [22], must be considered a breakthrough for nickelate superconductivity. In contrast to infinite layer nickelates, it has a positive Hall coefficient [22], although density functional theory (DFT) predicts large electron pockets [22,23]. The trilayer nickelate $\text{Nd}_4\text{Ni}_3\text{O}_8$ is, on the other hand, not superconducting [22]. This finding

naturally leads to the question which finite-layer nickelates can be made superconducting and how to dope them.

In this Letter, we analyze the superconducting pentalayer $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ and arguably the simplest finite-layer compound, bilayer $\text{Nd}_3\text{Ni}_2\text{O}_6$ [24], by state-of-the-art DFT plus dynamical mean field theory (DMFT) [25–29]. We show that local, dynamical correlations as included within DMFT push the Nd pockets (that exist in DFT [22]) above the Fermi level, thereby leaving only one correlated $d_{x^2-y^2}$ orbital per layer to form the Fermi surface—reminiscent of cuprate superconductors. The main differences between pentalayer and infinite layer nickelates emerging from our simulations are as follows: (i) When dynamical correlations are accounted for, Nd pockets are present (absent) in the infinite (five-layer) compound. This indicates that the Nd pockets are not important for superconductivity in nickelates. (ii) Our full-orbital DFT+DMFT calculations strongly suggest that the Hubbard model with a single (in-plane) orbital per Ni is the minimal setting for the pentalayer: the fluorite buffer layers mainly suppress the already weak c -axis Ni hopping; the tight-binding parameters and effective masses of the pentalayer’s minimal $d_{x^2-y^2}$ model are very similar to the infinite nickelates. (iii) Further, neglecting the weak interlayer hopping within the pentalayer slabs results in a two-dimensional (2D) one-band Hubbard model, which we study by including nonlocal correlations within the dynamical vertex approximation (DΓA) [30,31]. We find that spin fluctuations are dominant and give rise to superconductivity with a T_c comparable to experiment.

In the (undoped) bilayer $\text{Nd}_3\text{Ni}_2\text{O}_6$ with formal Ni valence $d^{8.5}$, instead, local correlations drive a charge transfer from the Ni $d_{xz,yz}$ to the $d_{x^2-y^2}$ orbitals that results in a *per se* multi-orbital system with a three-dimensional electronic structure. However, we predict that upon doping to a nominal valence of $d^{8.8}$, the $d_{xz,yz}$ orbitals become fully populated again and one-orbital physics, desirable for superconductivity, is

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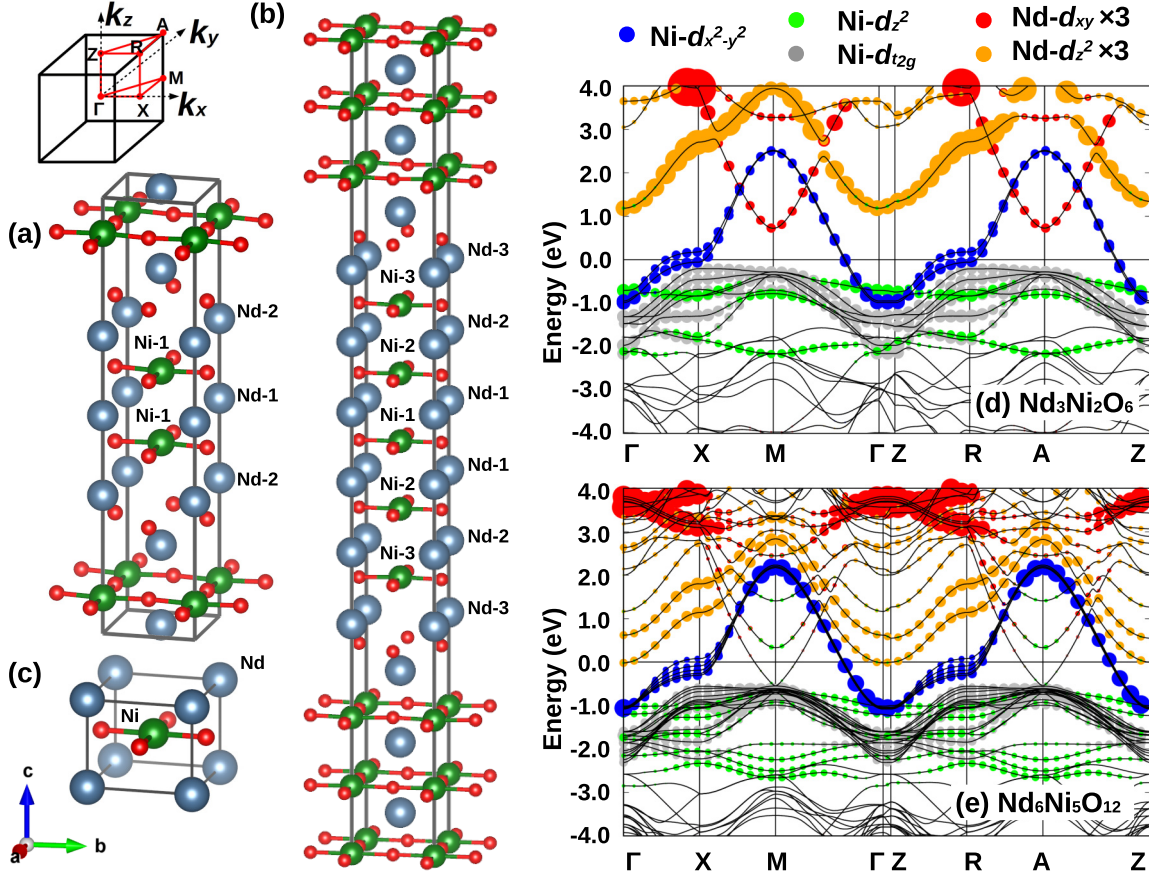


FIG. 1. Crystal structure of the reduced Ruddlesden-Popper perovskites $\text{Nd}_{n+1}\text{Ni}_n\text{O}_{2n+2}$: (a) $\text{Nd}_3\text{Ni}_2\text{O}_6$ ($n = 2$), (b) $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ ($n = 5$) and (c) NdNiO_2 ($n = \infty$). From the center to the interface layer, the inequivalent Nd (Ni) atoms are labeled as Nd-1, 2, 3 (Ni-1, 2, 3). Nd, Ni, and O atoms are shown as blue, green, and red balls, respectively. The top-left panel shows the Brillouin zone and the notation for the high-symmetry momenta. DFT band structure and orbital character for (d) $\text{Nd}_3\text{Ni}_2\text{O}_6$ and (e) $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ along a high-symmetry path through the Brillouin zone.

recovered: the *doped* bilayer system shows properties similar to the infinite and pentalayer superconductors. As a dopant we propose Zr, specifically $\text{La}_{2+\delta}\text{Zr}_{1-\delta}\text{Ni}_2\text{O}_6$ with $\delta \lesssim 0.4$.

Methods. Structural relaxations for both lattice constants and all internal atomic positions are performed with the VASP code [32], while the WIEN2K package is used for the DFT electronic structures [33]. For both, the Perdew-Burke-Ernzerhof (PBE) [34] and PBEsol [35,36] versions of the generalized gradient approximation (GGA) are employed on a dense momentum grid with 3000 k points. [See Supplemental Material (SM) [37] Sec. S.1 for detailed information or results with the modified Becke-Johnson (mBJ) [38] and DFT + U functionals.]

For the DMFT calculations, the WIEN2K bands around the Fermi level are projected onto maximally localized Wannier functions [39,40] using WIEN2WANNIER [41,42], visualization by XCrysden [43] and supplemented by local Kanamori interactions for the sites shown in Figs. 1(a)–1(c), taking the fully localized limit [44] as double counting. For $\text{Nd}_3\text{Ni}_2\text{O}_6$ a projection onto only the Ni-3d orbitals is sufficient, since the Nd-5d bands are well separated and above the Fermi level, as shown in Fig. 1(d) (discussed below). Instead, for $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ a projection onto both, the Ni-3d and the Nd-5d shell, is necessary to account for the electron pocket around the M and

A points, that are present in DFT. Expecting only minor variations of the local interaction with the number of nickel layers, we use the same interaction parameters as were obtained for LaNiO_2 [20] by constrained random phase approximation (cRPA): interorbital interaction $U' = 3.10$ eV (2.00 eV) and Hund's exchange $J = 0.65$ eV (0.25 eV) for Ni (Nd). The intraorbital Hubbard interaction follows as $U = U' + 2J$. These interaction parameters are close to those of previous studies [45,46] for 3d oxides. The resulting Hamiltonian is then solved at room temperature (300 K) in DMFT using continuous-time quantum Monte Carlo simulations in the hybridization expansions [47] implemented in w2DYNAMICS [48,49]. The maximum entropy method [50–54] is employed for an analytic continuation of the spectra.

As justified below, a one-band per Ni site description based on the $\text{Ni-}d_{x^2-y^2}$ orbital is possible at low energies. Hence, we also perform a Wannier projection onto this $d_{x^2-y^2}$ orbital only (see SM [37] Sec. S.3 for further information). We supplement this effective tight-binding one-band model with the same on-site interaction $U = 8t$ as in [21], motivated by cRPA. This one-band model for five isolated layers can then be treated by more involved many-body techniques. We use the D Γ A [30,31], employing an additional, admittedly crude, approximation: neglecting the interlayer hopping. We perform

TABLE I. Major tight-binding hopping parameters (in units of eV) for the Ni- $d_{x^2-y^2}$ orbital as obtained from a single-band Wannier projection; t , t' , t'' , and t_z indicate first [hopping along the direction $R = (100)$], second [$R = (010)$], third nearest neighbor [$R = (200)$], and z -direction [$R = (001)$] hopping, respectively. For Nd₆Ni₅O₁₂, Ni-1 is the central layer, Ni-3 denotes the two equivalent interface layers, and Ni-2 the two layers in-between [see Fig. 1(b)].

System	t	t'	t''	t_z	t'/t	t''/t
NdNiO ₂	-0.395	0.095	-0.047	-0.034	-0.242	0.119
Nd ₃ Ni ₂ O ₆	-0.414	0.092	-0.055	-0.055	-0.223	0.132
Nd ₆ Ni ₅ O ₁₂ : Ni-1	-0.395	0.098	-0.050	-0.031	-0.249	0.127
Nd ₆ Ni ₅ O ₁₂ : Ni-2	-0.392	0.097	-0.050	-0.026	-0.249	0.127
Nd ₆ Ni ₅ O ₁₂ : Ni-3	-0.398	0.097	-0.049		-0.245	0.122

calculations ranging from 300 to 40 K and estimate the critical temperature T_c along the lines of [55,56].

DFT crystal and electronic structure. We consider the series Nd _{$n+1$} Ni _{n} O _{$2n+2$} which can be synthesized from the Ruddlesden-Popper perovskite parent compound by oxygen reduction. Specifically, we concentrate on Nd₃Ni₂O₆ ($n = 2$), Nd₆Ni₅O₁₂ ($n = 5$, experimentally realized in [22]) and NdNiO₂ ($n = \infty$, experimentally realized in [2,3,57]). Their crystal structures are shown in Fig. 1. Note the interface NdO-NdO layers between the n NiO₂ layers are transformed to fluoritelike Nd-O₂-Nd blocking slabs, which we find to be energetically favorable in DFT(+ U) compared to the octahedral rocksalt interface (see SM [37]).

A natural question is: how do the electronic bands in general [23] and the presence of the Nd pocket in particular change with the number of layers n ? DFT studies of NdNiO₂ [11,16,17,58,59] show an electron pocket at the A and Γ point originating from the Nd-5d orbitals. Electronic correlations and Sr doping may push the Γ pocket above E_F . However, the A pocket remains present in the superconducting doping region and acts as an electron reservoir [21] that (self)dopes the Ni 3d bands away from the nominal $3d^9$ configuration. In contrast, Fig. 1(d) for Nd₃Ni₂O₆ shows no electron pockets; the Nd bands are clearly above E_F and well separated from the Ni bands. This indicates a weaker hybridization between Nd and Ni in Nd₃Ni₂O₆, i.e., the fluorite interface makes both NiO₂ and Nd-O₂-Nd layers more 2D. For Nd₆Ni₅O₁₂ in Fig. 1(e), the DFT band structure shows a tubelike pocket (composed of Ni- d_{z^2} and Nd- d_{xy} character) that encompasses the momenta A and M (see SM [37] for a plot). Similar to NdNiO₂ [11,16,59], a band of Nd- d_{z^2} character touches E_F at Γ .

To further investigate prospective similarities between infinite and finite layers, we list the electronic hopping terms from a Wannier projection in Table I: Infinite layer NdNiO₂ [60] and pentalayer Nd₆Ni₅O₁₂ have almost identical in-plane hopping parameters. Also the out-of-plane t_z hopping is almost the same within the five layers of the pentalayer nickelate, while it is practically zero across the fluorite spacing layer. Additionally both systems have very similar ratios of t'/t and t''/t . The bilayer Nd₃Ni₂O₆ has an $\sim 5\%$ larger t which can be attributed to the increased two-dimensionality caused by the fluorite interface, which cuts off z -direction hopping after every second layer, rather than every fifth in the pentalayer.

While differences in hopping parameters are much smaller than between infinite and finite layer cuprates [61], the filling

of the $d_{x^2-y^2}$ band, a key factor for superconductivity, differs considerably: Nd₃Ni₂O₆ hosts a nominal $d^{8.5}$ configuration which is far away from the optimal hole doping $\delta \lesssim 0.2$ ($d^{9-\delta}$). Also the doping of Nd₆Ni₅O₁₂, as predicted by DFT, is not within the superconducting region. Indeed, the A - M pocket takes electrons away, leading to $\delta \sim 0.24$ holes (see SM [37] Sec. S.4 and Table T.I) in the effective Ni- $d_{x^2-y^2}$ band, compared to the formal valency $\delta = 0.2$ ($d^{8.8}$). As we will see next, however, electronic correlations shift the tube-like pocket around A and M above the Fermi energy, and restore the doping to a level that is in line with the observed superconductivity.

DMFT results. Some correlations, defined by the difference to a one-particle Hartree-Fock solution, are already taken into account on the level of DFT (spin-polarized or not) and further static ones in DFT+ U (shown in the SM [37]). However, for the pentalayer we find prevalent dynamical correlations including a strong quasiparticle renormalization [37], which requires a method such as DMFT. For both, Nd₃Ni₂O₆ and Nd₆Ni₅O₁₂, electronic correlations dramatically change the electronic structure (see Figs. 2 and 3). In bilayer Nd₃Ni₂O₆, Hund's exchange favors a more equal occupation of orbitals since this way a spin-1 can be formed and also the Coulomb repulsion is lower ($U' = U - 2J$ instead of U). As a result, there is an electron transfer from the $d_{xz/yz}$ into the $d_{x^2-y^2}$ orbital (see SM [37] Table T.I). The now partially depopulated $d_{xz/yz}$ orbitals are pushed up in energy, and both $d_{x^2-y^2}$ and

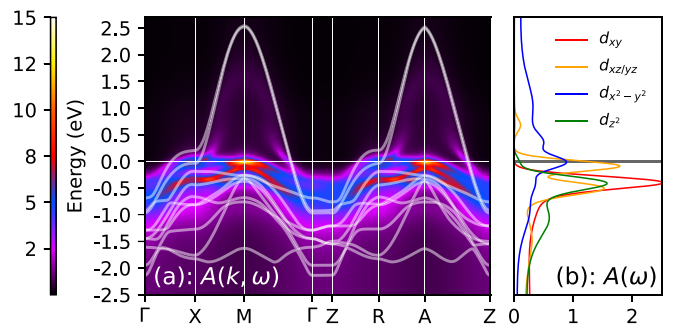


FIG. 2. (a) DMFT k -resolved spectral function $A(k, \omega)$ for Nd₃Ni₂O₆. White lines are the DFT Wannier bands. (b) Corresponding k -integrated spectral function $A(\omega)$. Local correlations push the $d_{xz/yz}$ orbitals up in energy, which now contribute to the Fermi surface at the M point (cf. SM Fig. S7).

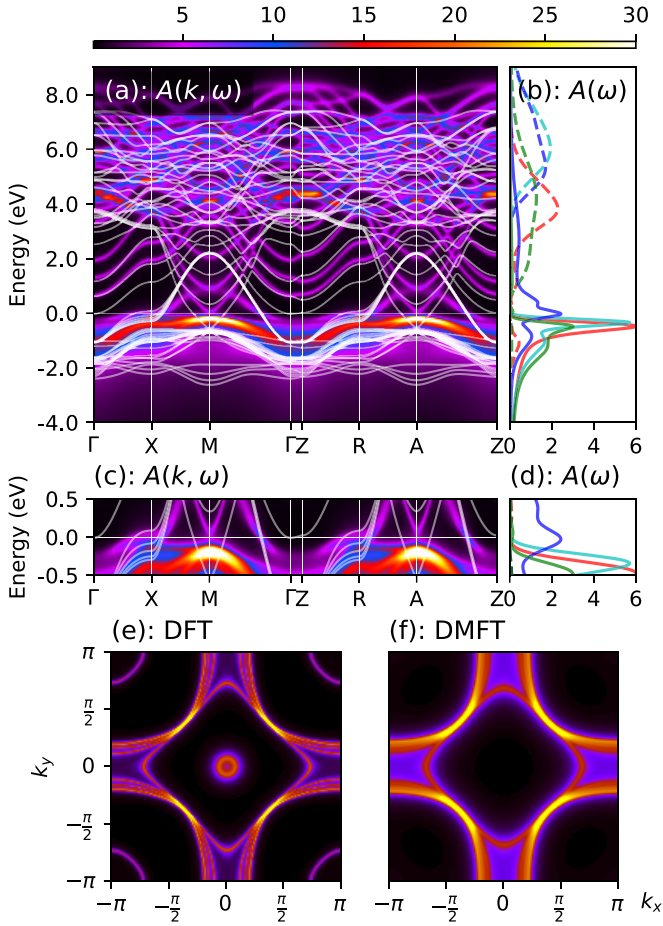


FIG. 3. DMFT k -revolved spectral functions $A(k, \omega)$ (a) and k -integrated $A(\omega)$ (b) of $\text{Nd}_6\text{Ni}_5\text{O}_{12}$; (c) and (d) are a zoom-in around $E_F = 0$; solid lines in (a) and (c) are the DFT Wannier bands. Fermi surface for (e) DFT and (f) DMFT. The colorbar at the top is for all plots. The DMFT Fermi surface has been multiplied by two for better visibility. Color code for the k -integrated spectral function is the same as in Fig. 2, with solid lines for Ni and dashed for Nd.

$d_{xz/yz}$ form quasiparticle peaks in Fig. 2 and contribute to the Fermi surface (see SM [37] Secs. S.6 and S.7). This multi-orbital, three-dimensional electronic structure is, according to common wisdom, not favorable for superconductivity.

For the pentalayer, $\text{Nd}_6\text{Ni}_5\text{O}_{12}$, dynamical correlations have a different game-changing effect (see Fig. 3): The Nd pocket around A and M momentum is pushed above the Fermi level, which only leaves the Ni $d_{x^2-y^2}$ contributing to the Fermi surface (cf. SM [37] Secs. S.6 and S.7). We thus have a one-orbital system that is essentially two-dimensional and prone to strong antiferromagnetic fluctuations. The predominant Ni $d_{x^2-y^2}$ orbitals now host $\delta = 0.19, 0.21$, and 0.20 holes in the three inequivalent layers, compared to $\delta = 0.24, 0.27$, and 0.26 in DFT. As seen below, this pushes $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ into the superconducting doping regime. Further, electronic correlations enhance the effective mass to $m^*/m_b \sim 2.5$ (see SM [37] Sec. S.5). Altogether, our results hence show a clear similarity of the pentalayer and the infinite layer system, with the noteworthy difference being the presence (absence) of the Nd A pocket in NdNiO_2 ($\text{Nd}_6\text{Ni}_5\text{O}_{12}$). This evidences/supports that

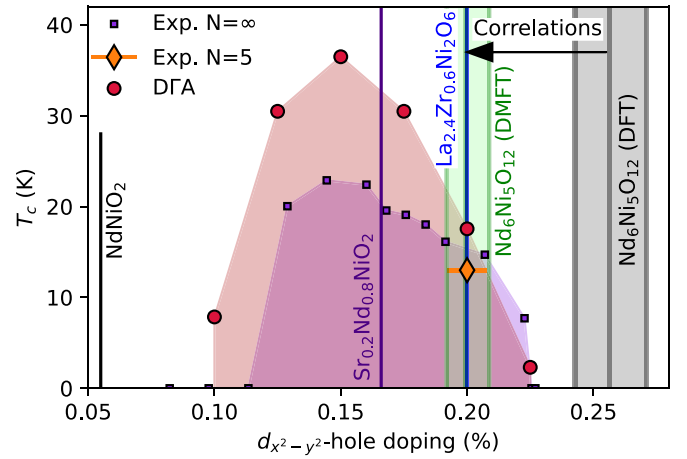


FIG. 4. Phase diagram T_c vs hole doping of the Ni $d_{x^2-y^2}$ band as calculated by DFT. For $\text{Nd}_6\text{Ni}_5\text{O}_{12}$, the doping of the inequivalent layers is marked by three green (DMFT) and gray lines (DFT) and the experimental T_c (diamond) is from [22]. For Sr-doped NdNiO_2 , experimental data are from [62].

the pocket is not essential for superconductivity in nickelates as argued in [21].

This brings us back to the bilayer compound $\text{Nd}_3\text{Ni}_2\text{O}_6$ and the question: can we tune its electronic structure to one that is favorable for superconductivity? We believe one can do so through chemical doping/substitution: Here, we consider $\text{La}_{2+x}\text{Zr}_{1-x}\text{Ni}_2\text{O}_6$ which has a nominal valency $3d^{9-x/2}$. Indeed, at $x = 0.4$ no electron pockets are present within DFT+DMFT anymore, and the Fermi surface is composed only of a single Ni $d_{x^2-y^2}$ orbital (see SM [37] Sec. S.5), hosting $\delta = \frac{x}{2} = 0.2$ holes, akin to the pentalayer and doped infinite layer nickelates.

Estimating T_c with DFT. The DMFT calculation with all Ni and Nd d orbitals for $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ suggests single-orbital physics. Indeed, one-band DMFT (SM [37] Secs. S.6 and S.7) agrees well with the full-orbital calculation, including effective masses (cf. SM [37] Tables T.I and T.III), justifying a projection onto only the $d_{x^2-y^2}$ orbital. Since interlayer hoppings are as small as in the infinite layer system, for what follows, we treat the layers as independent as done previously for NdNiO_2 [21].

This allows us to calculate T_c as a function of the $d_{x^2-y^2}$ orbital filling in an effective two-dimensional Hubbard model. To this end, we include nonlocal spin and superconducting fluctuations with DFT: The thus calculated superconducting dome is displayed as the red-shaded region in Fig. 4, and experiment for $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ [22] as a diamond. Also shown is the experimental phase diagram for NdNiO_2 (violet-shaded region, translated to the $d_{x^2-y^2}$ filling according to [21]) which thanks to higher quality films with less defects [62] now agrees much better with DFT than previously [2–4]. As we can see, the doping of the pentalayer parent compound $\text{Nd}_6\text{Ni}_5\text{O}_{12}$ is indeed already in the doping range for superconductivity. This was not the case without correlations: DFT (gray lines in Fig. 4) would predict a doping outside of the superconducting dome, due to the presence of the Fermi surface tube around A and M . Also the proposed $\text{La}_{2.4}\text{Zr}_{0.6}\text{Ni}_2\text{O}_6$ has a very similar doping (blue line), but it has slightly

different hopping parameters, in particular a larger t which should result in a slightly larger T_c as (i) t sets the unit of energy and (ii) according to D Γ A, a smaller U/t is favorable for T_c .

Conclusion. We have seen that electronic correlations decisively alter the electronic structure of finite-layer nickelates, beyond a mere effective mass renormalization of the bands. In the case of the pentalayer nickelate Nd₆Ni₅O₁₂, DFT predicts an electron pocket around the A and M momenta and thus a doping which is not in line with the observed superconductivity. However, electronic correlations push this electron pocket above the Fermi energy so that there are 0.2 holes per Ni (with only a tiny variation of ~ 0.02 holes between the layers). This way the Ni $d_{x^2-y^2}$ orbital is placed in the optimal superconducting doping range; it also has virtually the same hopping terms as NdNiO₂. Hence, electronic correlations are key to explaining the experimental observation of superconductivity in Nd₆Ni₅O₁₂ [22]. These predictions also call for a careful angular photoemission spectroscopy study of this pentalayer system to experimentally confirm the absence of the Nd-derived pockets.

While the A pocket in NdNiO₂ is possibly only a passive bystander for superconductivity [21], it will certainly contribute to the Hall coefficient. We therefore suggest the

very different Hall coefficient for $n = \infty$ [22] and $n = 5$ [2,3] to be explained by the presence (NdNiO₂) or absence (Nd₆Ni₅O₁₂) of these electron pockets. Further, it is tempting to attribute (at least part of) the unusually broad superconducting transition to the layer-dependent doping of Nd₆Ni₅O₁₂.

According to our phase diagram in Fig. 4, pentalayer nickelate is overdoped. Without electron pockets, Nd _{$n+1$} Ni _{n} O _{$2n+2$} has a hole doping $\delta = 1/n$, suggesting that T_c will increase with a few more Ni layers and the parent compounds will likely remain inside the superconducting dome up to about $n = 10$. For $n < 5$, we have instead too many holes. Then, electron doping with a tetravalent cation as, e.g., in La _{$2+\delta$} Zr _{$1-\delta$} Ni₂O₆ with $\delta \lesssim 0.4$ is needed.

Note added. Recently, [63] also reported DMFT calculations for Nd₆Ni₅O₁₂.

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