Nickelate superconductors – a renaissance of the one-band Hubbard model

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Following the discovery of superconductivity in the cuprates1 and the seminal work by Anderson2, the theoretical efforts to understand high-temperature superconductivity have been focusing to a large extent on a simple model: the one-band Hubbard model3–5. However, superconducting cuprates need to be doped, and the doped holes go into the oxygen orbitals5–8. This requires more elaborate multi-band model such as the three-orbital Emery model9,10. The recently discovered nickelate superconductors11 appear, at first glance, to be even more complicated multi-orbital systems. Here, we analyse this multi-orbital system and find that it is instead the nickelates which can be described by a one-band Hubbard model, albeit with an additional electron reservoir and only around the superconducting regime. Our calculations of the critical temperature $T_C$ are in good agreement with experiment, and show that optimal doping is slightly below the 20% Sr-doping of Ref. 11. Even more promising than 3$d$ nickelates are 4$d$ palladates.

The discovery of superconductivity in Sr$_{0.2}$Nd$_{0.8}$NiO$_2$ by Li et al.11 marked the beginning of a new, a nickel age of superconductivity, ensuing a plethora of experimental and theoretical work; see, among others, Refs. 12–33. Similar as for the cuprates, the basic structural elements are NiO$_2$ planes on a square lattice, and Ni has the same formal 3$d^8$ electronic configuration.

But at second glance, there are noteworthy differences, see Fig. 1(a). For the parent compound NdNiO$_2$, density functional theory (DFT) calculations show, besides the Ni $d_{x^2−y^2}$ orbital, additional bands around the Fermi level $E_F$15,20,21,34 that are of predominant Nd-5$d$ character and overlap with the former. Note that the Nd-5$d$ bands in Fig. 1(a) extend below $E_F$ despite their centre of gravity being considerably above $E_F$.

Such bands or electron pockets have the intriguing effect that even for the parent compound NdNiO$_2$, the Ni-$d_{x^2−y^2}$ orbital is hole doped, with the missing electrons in the Nd-5$d$ pockets. In other words, the parent compound NdNiO$_2$ already behaves as the doped cuprates. First calculations16,18,27 for superconductivity in the nickelates that are valid at weak interaction strength hence started from a Fermi surface with both, the Nd-5$d$ pockets and the Ni-$d_{x^2−y^2}$ Fermi surface. Such a multi-orbital nature of superconductivity has also been advocated in Refs. 18 and 29.

There is further the Nd-4$f$ orbital which in density functional theory (DFT) spurious shows up just above the Fermi energy. But these 4$f$ orbitals will be localised which can be mimicked by DFT+$U$ or by putting them into the core, as has been done in Fig. 1(a). Choi et al.32 suggest a ferromagnetic, i.e., anti-Kondo coupling of these Nd-4$f$ with the aforementioned Nd-5$d$ states.

A further striking difference is that the oxygen band is much further away from the Fermi level than for the cuprates, see Fig. 1(a). Vice versa, the other Ni-3$d$ orbitals are closer to the Fermi level and slightly doped because of their hybridisation with the Nd-5$d$ orbitals. Hence, in dynamical mean-field theory (DMFT)35 calculations albeit employing a rather large interaction so that NdNiO$_2$ is a Mott insulator, Lechermann26 finds that the holes in doped Sr$_{0.2}$Nd$_{0.8}$NiO$_2$ go to a larger extent to the Ni-$d_{x^2−y^2}$ orbital, but not the O-$p$ orbitals. In the extreme situation of one extra hole per Ni site, Ni has a 3$d^8$ configuration with only one hole in the $d_{x^2−y^2}$-orbital and the other one in the $d_{z^2}$ orbital forming a local spin-1, as realised e.g. in LaNiO$_2$.28 Other DMFT calculations for the parent compounds NdNiO$_2$33 and LaNiO$_2$, in-

FIG. 1. (a) Energy levels comparing CaCuO$_2$ and NdNiO$_2$, with the DFT bandstructure as a background. (b) Phase diagram $T_C$ vs. Sr-doping as calculated in DΓA together with the hitherto only experimental point: 15 K for 20% Sr-doping11. In the blue-shaded region, a one-band Hubbard model description is possible with its doping given on the upper $x$-axis.
stead report a predominately Ni $3d_{x^2−y^2}$ band plus Nd-5d pockets. This leads to the questions: Which orbitals are depopulated if we dope NdNiO$_2$ with Sr? Is a multi-orbital description necessary for the actual superconducting compound Sr$_x$Nd$_{1−x}$NiO$_2$? In which doping regime is Sr$_x$Nd$_{1−x}$NiO$_2$ superconducting at all? What is the upper limit for the superconducting transition temperature $T_C$?

In this paper, we show that, if we properly include electronic correlation by DMFT, up to a Sr-doping of about 30%, the holes only depopulate the Ni-$3d_{x^2−y^2}$ and Nd-5d bands. Only for larger Sr-dopings, holes are doped into the other Ni-3d orbitals, necessitating a multi-orbital description. The hybridisation between the Ni-$3d_{x^2−y^2}$ and Nd-5d orbitals as calculated from the DFT-derived Wannier Hamiltonian is vanishing. We hence conclude that up to a Sr-doping of around 30% marked as dark blue in Fig. 1 (b), a single Ni-$3d_{x^2−y^2}$ band description as in the one-band Hubbard model is possible. However, because of the Nd-5d pocket(s), which acts like a electron reservoir and otherwise hardly interacts, only part of the Sr-doping [lower $x$-axis of Fig. 1 (b)] goes into the Ni-$3d_{x^2−y^2}$-band [upper $x$-axis], cf. Supplementary Information Section S.2 for the functional dependence. We also take small Sr-doping out of the blue-shaded region since for such small dopings there is, besides the Nd-5d $A$-pocket, the $Γ$-pocket which interacts with the Nd-4f moments ferromagnetically, and might result in additional correlation effects. At larger doping and when including the Nd-interaction in DMFT the $Γ$-pocket is shifted above $E_F$, see Fig. 2 below.

Fig. 1 (b) further shows the superconducting critical temperature $T_C$ of the thus derived and doped Hubbard model, calculated by a method that is appropriate in the strong coupling regime: the dynamical vertex approximation (DVA)\cite{36}. The agreement with experiment is reasonable given that the experimental $T_C$ can be expected to be lower because of e.g. impurity scattering and the theoretical one is somewhat overestimated\cite{37}.

Let us now discuss these results in more detail. We start with a DFT calculation [cf. Supplementary Information Section S.1] which puts the Nd-4f orbitals just above $E_F$. But since their hybridisation with the Ni-$3d_{x^2−y^2}$ orbital is weak\cite{38}, $|V_{x^2−y^2,4f}| = 25$ meV, see Supplementary Information Section S.2, they will localise and not make a Kondo effect. This localisation can be described e.g. by the spin-splitting in DFT+$U$, or by including the Nd-4f states in the core. It leaves us with a well defined window with just five Nd-5d and five Ni-3d around the Fermi energy. For these remaining ten orbitals we do a Wannier function projection (see Supplementary Information Section S.2) and subsequent DMFT calculation with constrained random phase approximation (cRPA) calculated inter-orbital interaction $U' = 3.10$ eV (2.00 eV) and Hund’s exchange $J = 0.65$ eV (0.25 eV) for Ni (Nd)\cite{28}.

Fig. 2 presents the calculated DMFT spectral function for these ten bands. Let us first concentrate on Sr$_{0.2}$Nd$_{0.8}$NiO$_2$ for which also the $k$-resolved spectral function on the right hand side is shown. Clearly in DMFT there is a single, compared to the DFT strongly renormalized Ni-$3d$ band of $d_{x^2−y^2}$ character crossing $E_F = 0$, see the zoom in Fig. 2 (e). Besides, there is also a pocket around the $A$-point of predominately Nd-$5d_{xy}$ character, but the $Γ$ pocket is shifted above $E_F$, cf. Supplementary Information Section S.4 for other dopings.

Hence we have two bands of predominately Ni-$3d_{x^2−y^2}$ and Nd-$5d_{xy}$ character. Their hybridisation is zero, see Supplementary Information Table S.III, which can be inferred already from the lack of any splitting around the DFT crossing points in Fig. 2 (d,e). There is some hybridisation of the Nd-$5d_{xy}$ with the other Ni-bands, which results in minor spectral weight of Nd-$5d_{xy}$ character in the region of the other Ni-3d bands at $−0.5$ to $−2.5$ eV in Fig. 2 (a-c) and vice-versa of the Ni-$3d_{x^2}$ orbital between 1 and $3$ eV. This admixing is however so minor, that it can be described by properly admixed, effective orbitals that are away from the Fermi energy, without multi-orbital physics.

If we study the doping dependence in Fig. 2 (a-c), we see that with Sr-doping all bands move upwards. More involved and beyond a rigid-band picture, also the Ni-$3d_{x^2−y^2}$ band becomes less and less correlated. The effective mass enhancement or inverse quasiparticle weight changes from $m^*/m = 1/Z = 4.4$ for the undoped compound to $m^*/m = 2.8$ at 20% Sr-doping to $m^*/m = 2.5$ at 30% Sr-doping. The Hubbard bands gradually disappear.

At 30% Sr-doping we are in the situation that the other Ni-3d bands are now immediately below the Fermi energy. Hence around this doping it is no longer justified to employ a Ni-$d_{x^2−y^2}$-band plus Nd-$d_{xy}$-pocket around $A$ picture to describe the low-energy physics. However below 30% doping, the other Ni-3d orbitals enter the stage. Because of the weak hybirdization, the Nd-$d_{xy}$-pocket only acts as an electron reservoir, which changes the doping of the Ni-$d_{x^2−y^2}$-band from the lower $x$-axis in Fig. 1 (b) to the upper $x$-axis.

For this $d_{x^2−y^2}$-band we have done a separate Wannier function projection which results in the aforementioned hopping parameters $t = 395$ meV, $t' = −95$ meV, $t'' = 47$ meV between next-nearest and the two next-nearest neighbours; a cRPA calculation for this single orbital yields $U = 3.2$ eV\cite{39}. The hopping parameters in the $z$-direction are negligibly small $t_z = 34$ meV, leaving us with a to a good approximation two-dimensional one-band Hubbard model.

This two-dimensional one-band Hubbard model with the properly translated doping according to Fig. 1 (b) can now be solved using more sophisticated, numerically expensive methods. We employ the DVA\cite{36} in the following, which not only includes all local DMFT correlations, but also non-local correlations that are responsible for spin fluctuations\cite{46} and $d$-wave superconductivity\cite{36,40}. Please note that our DVA calculation also includes charge fluctuations\cite{41} and their effect on superconductivity on
FIG. 2. (a-c) DFT+DMFT k-integrated spectral function \( A(\omega) \) of \( \text{Sr}_x\text{Nd}_{1-x}\text{NiO}_2 \) at 0\%, 20\% and 30\% Sr-doping, orbitally resolved for the Nd-5\(d \) and Ni-3\(d \) orbitals. (d) DFT+DMFT k-resolved spectral function \( A(\omega) \) for \( \text{Sr}_0.2\text{Nd}_{0.8}\text{NiO}_2 \). (e) Zoom-in of (d). The DFT bands are shown as (white) lines for comparison. Around the Fermi level there is a single Ni-3\(d \) orbit which has a hole-like structure for \( \text{Sr}_0 \). If these \( d \) orbitals are not included, studyting a (tetragonal) rotational symmetry broken phase \(^{43}\) requires a full parquet DFA or an eigenvalue analysis like we do for \( d \)-wave superconductivity here.

Fig. 3 shows the thus obtained DFA Fermi surfaces at different dopings and two different temperatures. For the superconducting \( \text{Sr}_0.2\text{Nd}_{0.8}\text{NiO}_2 \), which corresponds to \( n_{d_{x^2-y^2}} = 0.822 \) electrons per site in the Ni-\(d_{x^2-y^2}\)-band, we have a well defined hole-like Fermi surface, whereas for \( n_{d_{x^2-y^2}} = 0.9 \) and, in particular for \( n_{d_{x^2-y^2}} = 0.95 \), we see the development of Fermi arcs induced by strong antiferromagnetic spin fluctuations. The \( A(\pi, \pi, \pi) \)-pocket is not visible in Fig. 3 because it is only included through the effective doping in the DFA calculation. In any case it would be absent in the \( k_z = 0 \) plane and only be visible around \( k_z = \pi \).

Whereas the Ni-3\(d_{x^2-y^2}\)-band is strongly correlated and has a Fermi surface that is prone to high-\( T_C \) superconductivity, the \( A \)-pocket is weakly correlated and hardly hybridises with the former. Nonetheless for some physical quantities, different from superconductivity it will play a role. For example, it will give an electron-like negative contribution to the Hall coefficient. This will be partially compensated by the \( d_{x^2-y^2} \)-contribution which has a hole-like structure for \( \text{Sr}_0.2\text{Nd}_{0.8}\text{NiO}_2 \), corresponding to \( n_{d_{x^2-y^2}} = 0.822 \) which is in-between the two leftmost panels in Fig. 3. Note that also for the cuprates a hole-like Hall coefficient is found\(^{14}\). In contrast, for the undoped compound \( \text{NdNiO}_2 \) (i.e., \( n_{d_{x^2-y^2}} = 0.944 \approx 0.95 \)), the 3\(d_{x^2-y^2}\)-Fermi surface in Fig. 3 has a more electron-like shape but its Hall contribution should be suppressed because of the pseudo gap. This might explain why the Hall coefficient\(^ {11}\) is large and electron-like (negative) for \( \text{NdNiO}_2 \), whereas it is smaller and changes from negative (electron-like) to positive (hole-like) below 50 K. In Fig. 3 we further see that for \( T = 92 K = 0.02t \), we have a strong scattering at the antinodal point \( k = (\pi, 0) \), whereas at the lower temperature \( T = 46 K = 0.01t \), we have a well defined band throughout the Brillouin zone. This indicates that the hole-like contribution of the \( d_{x^2-y^2} \)-orbital becomes more important at lower temperatures, possibly explaining the experimental sign change of the Hall coefficient.

At the temperatures of Fig. 3, \( \text{Sr}_x\text{Nd}_{1-x}\text{NiO}_2 \) is not yet superconducting. But we can determine \( T_C \) from the divergence of the superconducting susceptibility \( \chi \), or alternative the leading superconducting eigenvalue \( \lambda_{SC} \). These are related through, in matrix notation, \( \chi = \chi_0/[1 - \Gamma_{pp}\chi_0] \). Here \( \chi_0 \) is the bare superconducting susceptibility and \( \Gamma_{pp} \) the irreducible vertex in the particle-particle channel calculated by DFA; \( \lambda_{SC} \) is the leading eigenvalue of \( \Gamma_{pp}\chi_0 \). If \( \lambda_{SC} \) approaches 1, the superconducting susceptibility is diverging.

In Fig. 4 we plot this \( \lambda_{SC} \) vs. temperature, and see that it approaches 1 at e.g. \( T = 36 K = 0.008t \) for
\[ n_{d_{x^2-y^2}} = 0.85. \] But outside a narrow doping regime between \( n_{d_{x^2-y^2}} = 0.9 \) and 0.8 it does not approach 1; there is no superconductivity. Let us also note that the phase transition is toward \( d \)-wave superconductivity which can be inferred from the leading eigenvector corresponding to \( \lambda_{SC} \).

Altogether, this leads to the superconducting dome of Fig. 1 (b). Most noteworthy \( \text{Sr}_{0.2}\text{Nd}_{0.8}\text{NiO}_2 \) which was found to be superconducting in experiment\(^{11} \) is close to optimal doping \( n_{d_{x^2-y^2}} = 0.85 \) or \( \text{Sr}_{0.16}\text{Nd}_{0.84}\text{NiO}_2 \). Our results call for a more thorough investigation of superconductivity in nickelates around this doping, which is quite challenging experimentally\(^{12-14} \).

Besides a slight increase of \( T_C \) by optimising the doping, and further room of improvement by adjusting \( t' \) and \( t'' \), our results especially show that a larger bandwidth and a somewhat smaller interaction-to-bandwidth ratio may substantially enhance \( T_C \), see Supplementary Information Fig. S7. One way to achieve this is compressive strain, which enlarges the bandwidth while hardly affecting the interaction. Compressive strain can be realised by e.g. growing thin nickelate films on a \( \text{LaAlO}_3 \) substrate with or without \( \text{SrTiO}_3 \) capping layer, or by \( \text{Ca} \)- instead of \( \text{Sr} \)-doping for the bulk or thick films. Another route is to substitute \( 3d \) \( \text{Ni} \) by \( 4d \) elements, e.g. in \( \text{Nd(La)}\text{PdO}_2 \) which has a similar Coulomb interaction and larger bandwidth\(^{17,45} \).

**Note added:** In an independent DFT+DMFT study Leonov et al.\(^{46} \) also observe the shift of the \( \Gamma \) pocket above \( E_F \) with doping for \( \text{LaNiO}_2 \) and the occupation of further \( 3d \) orbitals besides the \( 3d_{x^2-y^2} \) for large (e.g. 40\%) doping.

**Second note added:** Given that our phase diagram Fig. 1 (b) has been a prediction with only a single experimental data point given, the most recently experimentally determined phase diagram\(^{47} \) turned out to be in very good agreement. In particular if one considers, as noted already above, that the theoretical calculation should overestimate \( T_C \), while the experimentally observed \( T_C \) is likely suppressed by extrinsic contributions such as disorder etc. Supplemental Information Section S. 6 shows a comparison, which corroborates the modeling and theoretical understanding achieved in the present paper.

**METHODS**

1. **Density functional theory (DFT)**

   We mainly employ the \textsc{wien2k} program package\(^{48} \) using the PBE version of the generalised gradient approximation (GGA), a \( 13 \times 13 \times 15 \) momentum grid, and \( R_{MTK_{max}}=7.0 \) with a muffin-tin radius \( R_{MT} = 2.50, 1.95, \) and 1.68 a.u. for \( \text{Nd}, \text{Ni}, \) and \( \text{O} \), respectively. But we also double checked against \textsc{vasp}\(^{49} \), which was also used for structural relaxation (\( a = b = 3.86 \) Å, \( c = 3.24 \) Å\(^{38} \), and \textsc{fplo}\(^{50} \), which was used for Fig. 1 (a) and Supplementary Information Fig. S1. The \( 4f \) orbitals are treated as open core states if not stated otherwise.

2. **Wannier function projection**

   The \textsc{wien2k} bandstructure around the Fermi energy is projected onto maximally localized Wannier functions\(^{51} \) using \textsc{wien2wannier}\(^{52} \). For the DMFT we employ a projection onto five \( \text{Ni-3d} \) and five \( \text{Nd-5d} \) bands; for the parameterization of the one-band Hubbard model we project onto the \( \text{Ni-3d}_{x^2-y^2} \) orbital. For calculating the hybridisation with the \( \text{Nd-4f} \), we further Wannier projected onto a 17 bands, with the \( \text{Nd-4f} \) states now treated as valence bands in GGA.
3. Dynamical mean-field theory (DMFT)

We supplement the five Ni-3d plus five Nd-5d orbital Wannier Hamiltonian by a cRPA calculated Coulomb repulsions\textsuperscript{28} $U' = 3.10$ eV (2.00 eV) and Hund’s exchange $J = 0.65$ eV (0.25 eV) for Ni (Nd or La). The resulting Kanamori Hamiltonian is solved in DMFT\textsuperscript{45} at room temperature (300 K) using continuous-time quantum Monte Carlo simulations in the hybridisation expansions\textsuperscript{53} implemented in w2DYNAMICS.\textsuperscript{54} The maximum entropy method\textsuperscript{55} is employed for the analytic continuation of the spectra.

4. Dynamical vertex approximation

For calculating the superconducting $T_C$, we employ the dynamical vertex approximation (DΓA), for a review see Ref.\textsuperscript{36}. We first calculate the particle-particle vertex with spin-fluctuations in the particle-hole and transversal-particle hole channel, and then the leading eigenvalues in the particle-particle channel, as done before in Ref.\textsuperscript{40}. This is like the first iteration for the particle-particle channel in a more complete parquet DΓA\textsuperscript{36}.

\textbf{ADDENDUM}

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This value is slightly larger than the cRPA zero frequency value $U = 2.6 \, eV$ to mimic the frequency dependence as well as beyond cRPA contributions. In the Supplementary Information Section S.5 we present further calculations within a realistic range of $U$ values.

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Supplementary Information “Nickelate superconductors – a renaissance of the one-band Hubbard model”

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Here, we provide information supplementary to the main manuscript. Specifically, Section S.1 presents the orbital character of the three bands crossing the Fermi level for undoped LaNiO2 and NdNiO2 in density functional theory. Section S.2 discusses details of the Wannier function projections onto 1, 10 and 17 bands and tabulates the most important hopping parameters. Section S.3 shows the dependence of the doping of the Ni-3d_{x^2-y^2} band and the quasiparticle mass on Sr-doping as calculated in dynamical mean-field theory (DMFT). Section S.4 supplements the main text by additional DMFT spectra at other doping levels for NdNiO2 as well as for LaNiO2. Section S.5 provides additional dynamical vertex approximation spectra and phase diagrams obtained for a somewhat smaller and larger interaction U than expected and employed in the main text. This hints that Nd(La)PdO2 with a larger bandwidth and slightly smaller interaction might host even larger critical temperatures. Section S.6 compares with the recently obtained experimental phase diagram.

S.1. ORBITAL CHARACTER OF THE IMPORTANT DFT BANDS

Starting point of our analysis is the density functional theory (DFT) bandstructure which we have calculated by wien2k1,2, VASP3, and FPLO4 using the PBE5 version of the generalized gradient approximation (GGA). There are three relevant orbitals that cross the Fermi energy \( E_F = 0 \) in DFT. We determine their orbital character in FPLO4 version 18-00-55, using a \( k \)-mesh of \( 19 \times 19 \times 22 \) points for the scalar relativistic calculations. For NdNiO3, the Nd-4f states were put into the core (“open core”), their occupation is fixed to three electrons. At high-symmetry points, some orbitals may belong to the same two-dimensional irreducible representation. As a result, the respective orbital characters can show a seeming discontinuity in the band structure plot.

The results are presented in Fig. S.1 for both, LaNiO2 and NdNiO2. The band in the first row is predominately of Ni-3d_{x^2-y^2} character, with some oxygen character mixed in. This oxygen admixture is however much less than for the cuprates, where the oxygen orbitals are much closer, see Fig. 1 of the main paper. This band is close to half-filling and can be described, as we show in our paper, by a one-band Hubbard model, if the doping is adjusted properly. We casually refer to it as the “Ni-3d_{x^2-y^2}” band in the main text.

The band in the second row forms a Fermi surface pocket around the \( \Gamma \)-pocket. It is mainly of Nd(La)-5d_{3z^2-r^2} character, but with much more admixture from the other orbitals, in particular, the Ni-3d_{3z^2-r^2} orbital. In case of LaNiO2 it is difficult to disentangle this band from the the La-4f orbitals above 2 eV, but this only concerns the less relevant upper band edge. In case of NdNiO2, these Nd-4f have been removed by treating them as part of the open core.

An important observation is that the \( \Gamma \)-pocket is only slightly below \( E_F \) for LaNiO2 but extends noticeable below \( E_F \) for NdNiO2. This is however, possibly an artifact of the open core treatment. In a plain GGA calculation with Nd-4f states as valence states (not shown), the distance between the bottom of the Nd-5d_{3z^2-r^2} pocket and \( E_F \) is more similar to that in LaNiO2. In a DFT+U treatment this pocket is spin-split due to a ferromagnetic interaction with the Nd-4f magnetic moment6. The \( \Gamma \)-pocket disappears, i.e., is shifted above \( E_F \), when including the La-5d interaction in DMFT for LaNiO27. Even if we treat the Nd-4f as open core states, which stabilizes the \( \Gamma \)-pocket the most, the \( \Gamma \) pocket disappears for Sr-doping above 10-15\%8.

Finally, there is the pocket around the \( A \)-point in Fig. S.1, which extends much further below \( E_F \) and hence remains present even up to a Sr-doping of 30\% in DMFT, see Fig. S.4 below. Hence it is this \( A \)-pocket which serves as an electron reservoir, with the important consequence that the doping of the Ni-3d_{x^2-y^2} orbital is different from the Sr-doping. This \( A \)-pocket can be associated in part of the Brillouin zone with the Nd-5d_{xy} orbital. However it is quite intermixed with other orbitals and at its bottom (around the \( A \)-point) it crosses the Nd-3d orbitals which makes it difficult to trace.
FIG. S.1. Band weights for three bands that cross the Fermi energy $E_F = 0$ for LaNiO$_2$ (left panels) and NdNiO$_2$ (right panels), as calculated by DFT. In each panel, the top part shows the selected band (thick blue curve) along a path in the Brillouin zone, while the bottom part depicts the orbital composition (band characters) of the respective band along the same path. Here and in the following, the Fermi level is at $E_F = 0$.

In the next Section, we will see that the hybridization with the Ni-3$d_{x^2-y^2}$ orbital is vanishing so that the $A$-pocket can be considered as an electron reservoir which otherwise does not affect the strongly correlated Ni-3$d_{3z^2-r^2}$ orbital (or the one-band Hubbard model description). The proper translations between the Sr-doping and that of the Ni-3$d_{x^2-y^2}$ orbital is given in Fig. 1 (right) of the main text and in Fig. S.3 below.
Next, we present details of the Wannier function projection, which has been done using *wien2wannier*\textsuperscript{9,10} for the projection of the *wien2k* bandstructure onto maximally localized Wannier functions\textsuperscript{11–13}. Fig. S.2 provides for an overview, showing the DFT bandstructure together with the Wannier function projection on 10-bands (La/Nd-5\textit{d}+Ni-3\textit{d}; blue dots) and on the Ni-\textit{d}_{\text{x}^{2}-\text{y}^{2}} band only (red dots). The former projection is used for the subsequent dynamical mean-field theory (DMFT) multi-orbital calculations. These calculations however show that a properly doped Ni-\textit{d}_{\text{x}^{2}-\text{y}^{2}}-band (one-band Hubbard model) description is sufficient. Hence, we have also performed a projection onto this one-band only. We have further done Wannier projections onto 17 orbitals (Nd-4\textit{f}+Nd-5\textit{d}+Ni-3\textit{d}) which is not included Fig. S.2, as it starts from the plain GGA calculation which puts the Nd-4\textit{f} states just above \( E_F \). For this GGA calculation, the Wannier projections onto 1- and 10-band have been done as well.

![Diagram](image)

**FIG. S.2.** Bandstructure of LaNiO\textsubscript{2} (a) and NdNiO\textsubscript{2} (b). For NdNiO\textsubscript{2}, we treat the Nd-4\textit{f} orbitals as open core states. Single-band (Ni-\textit{d}_{\text{x}^{2}-\text{y}^{2}}, red dots) and 10-bands (La/Nd-5\textit{d}+Ni-3\textit{d}, blue dots) Wannier bands are superimposed on the DFT bandstructure (black lines).

In Table S.1, we present the hopping parameters of the Ni-3\textit{d}_{\text{x}^{2}-\text{y}^{2}} orbital which forms the one-band Hubbard model, as obtained for LaNiO\textsubscript{2}, NdNiO\textsubscript{2} as well as for LaPdO\textsubscript{2}, treating the Nd-4\textit{f} either as valence bands in GGA or as open core in GGA. Here \( t_{R_i,R_j} \) denotes the hopping by \( R_i \) unit cells in the \( i \) direction. That is, \( t_{000} \) is the on-site potential, \( t = -t_{100} \) the nearest neighbor hopping, \( t' = -t_{110} \) and \( t'' = -t_{200} \) the next nearest neighbor hopping, and
LaNiO$_2$ (GGA)  
1-band (Ni-$d_{x^2-y^2}$) 0.2689 -0.3894 -0.0362 0.0977 -0.0465 -0.0037  
10-bands (La-$d$+Ni-$d$) 0.2955 -0.3975 -0.0458 0.0985 -0.0491 0.0000  
17-bands (La-$f$+La-$d$+Ni-$d$) 0.3514 -0.3943 -0.0239 0.0792 -0.0422 -0.0008  

NdNiO$_2$ (GGA)  
1-band (Ni-$d_{x^2-y^2}$) 0.2502 -0.3974 -0.0287 0.0933 -0.0474 -0.0027  
10-bands (Nd-$d$+Ni-$d$) 0.1998 -0.4068 -0.0763 0.1007 -0.0428 0.0015  
17-bands (Nd-$f$+Nd-$d$+Ni-$d$) 0.2892 -0.4065 -0.0289 0.0773 -0.0429 0.0026  

NdNiO$_2$ (GGA open core)  
1-band (Ni-$d_{x^2-y^2}$) 0.3058 -0.3945 -0.0336 0.0953 -0.0471 -0.0031  
10-bands (Nd-$d$+Ni-$d$) 0.3168 -0.3976 -0.0389 0.0949 -0.0480 -0.0008  
1-band (Ni-$d_{x^2-y^2}$) 0.4094 -0.5373 -0.0448 0.0975 -0.0708 -0.0058  

| TABLE S.I. Major hopping elements (in units of eV) of the Ni-$3d_{x^2-y^2}$ orbital from 1-band (Ni-$3d_{x^2-y^2}$), 10-bands (La/Nd-$d$+Ni-$d$) and 17-bands (La/Nd-$f$+La/Nd-$d$+Ni-$d$) model projections. In the last two lines, we also show the hopping parameters for LaPdO$_2$. The DFT-relaxed lattice parameters are: LaNiO$_2$ ($a = b = 5.88 \text{ Å}, c = 3.35 \text{ Å}$), NdNiO$_2$ ($a = b = 3.86 \text{ Å}, c = 3.24 \text{ Å}$), LaPdO$_2$ ($a = b = 4.13 \text{ Å}, c = 3.27 \text{ Å}$). |
|---|---|---|---|---|---|---|
| LaNiO$_2$ (GGA) | $t_{100}$ | $t_{100}$ | $t_{100}$ | $t_{110}$ | $t_{200}$ | $t_{210}$ |
| Ni-$d_{x^2-y^2}$ | 0.0000 | 0.0030 | -0.0000 | -0.0851 | -0.0851 | -0.0203 |
| NdNiO$_2$ (GGA) | $t_{100}$ | $t_{100}$ | $t_{100}$ | $t_{100}$ | $t_{200}$ | $t_{210}$ |
| Ni-$d_{x^2-y^2}$ | -0.0215 | -0.0215 | -0.0000 | -0.0612 | -0.0612 | -0.0160 |

| TABLE S.II. Hybridization (hopping amplitude in eV) between the Ni-$3d_{x^2-y^2}$ and the Nd(La)-$4f$ orbitals, as obtained from Wannier projections onto 17-bands (La/Nd-$4f$+La/Nd-$d$+Ni-$d$) including the $4f$ as valence states in DFT(GGA). |
|---|---|---|---|---|---|---|
| LaNiO$_2$ | Ni-$d_{x^2-y^2}$ (10-bands model, GGA) | 0.0000 | 0.0085 | -0.0085 | -0.0168 | 0.0000 |
| NdNiO$_2$ (17-bands model, GGA) | 0.0000 | 0.0084 | -0.0084 | -0.0037 | 0.0000 |
| NdNiO$_2$ (GGA with open core) | 0.0000 | 0.0070 | -0.0070 | -0.0088 | 0.0000 |
| Nd-$d_{x^2-y^2}$ (10-bands model, GGA) | 0.0000 | 0.0077 | -0.0077 | -0.0066 | 0.0000 |
| Nd-$d_{x^2-y^2}$ (17-bands model, GGA) | 0.0000 | 0.0081 | -0.0081 | -0.0023 | 0.0000 |

| TABLE S.III. Hybridization (hopping amplitude in eV) between the Ni-$d_{x^2-y^2}$ and the La/Nd-$5d$ orbitals. The results are obtained from Wannier projections onto 17-bands (La/Nd-$4f$+La/Nd-$5d$+Ni-$d$) and 10-bands (La/Nd-$5d$+Ni-$d$). The Nd-$4f$ bands are treated as core states in “GGA with open core” and as valence states in “GGA”. |
|---|---|---|---|---|---|---|
| LaNiO$_2$ | Ni-$d_{x^2-y^2}$ (10-bands model, GGA) | 0.0000 | 0.0085 | -0.0085 | -0.0168 | 0.0000 |
| NdNiO$_2$ | Nd-$d_{x^2-y^2}$ (10-bands model, GGA) | 0.0000 | 0.0084 | -0.0084 | -0.0037 | 0.0000 |
| Nd-$d_{x^2-y^2}$ (17-bands model, GGA) | 0.0000 | 0.0077 | -0.0077 | -0.0066 | 0.0000 |
| Ni-$d_{x^2-y^2}$ (10-bands model, GGA) | 0.0000 | 0.0081 | -0.0081 | -0.0023 | 0.0000 |

$ t_z = -t_{001} $ the hopping in the $z$-direction perpendicular to the NiO$_2$ plane. The hopping parameters are surprisingly similar for LaNiO$_2$ and NdNiO$_2$ and the different Wannier projections, considering the fact that e.g. the $4f$ orbitals are at very different energies for the three DFT calculations in Table S.I. LaPdO$_2$, on the other hand, with $4d_{x^2-y^2}$ instead of $3d_{x^2-y^2}$ has a much larger bandwidth. In the main text, we give the values for the open core GGA 1-band Wannier projection for NdNiO$_2$.

A further relevant result of the Wannier projection is the hybridization with the $4f$ orbitals which is shown in Table S.II. These hybridizations are rather small, maximally $V = 60 \text{ meV}$ for NdNiO$_2$. Such a hybridization is by far too small to give rise to a Kondo effect. Even if we take this maximal hybridization and a typical $4f$-Coulomb interaction of $U = 5 \text{ eV}$, the Kondo coupling is only $J = 4U^2/U = 3 \text{ meV}$. Which even, yields an exponential factor of $e^{-1/(N \rho_0 \cdot J)} \approx 10^{-54}$ for the Kondo temperature (taking a typical $\rho_0 = 0.2 \text{ eV}^{-1}$ from Fig. 2 of the main text and $N = 2 \times 7$ as a maximal upper bound). The prefactor is of the order of $1 \text{ eV}$ or smaller, so that we can conclude that there is no Kondo effect between the localized Nd-$4f$ moments and the Ni $3d_{x^2-y^2}$ orbital.

Next we turn to the hybridization between the relevant Ni-$3d_{x^2-y^2}$ band and the La/Nd-$5d$ orbitals. The important observation is that the hybridization between the Ni-$3d_{x^2-y^2}$ band and the Nd(La)-$5d_{xy}$ and the Nd(La)-$5d_{z^2}$ is zero. These are the most important hybridizations since these two Nd(La) orbitals form the basis of the $A$- and $\Gamma$-pocket,
respectively. Hence we can, to a very good approximation, indeed consider these pockets to be independent of the Ni-3d_{x^2-y^2} band, except for that they may serve as an electron reservoir. Part of the holes, induced by e.g. Sr-doping will go into the Ni-3d_{x^2-y^2} band, and part into the A- and Γ-pocket. The latter is completely depopulated in the superconducting regime.

### S.3. DOPING AND MASS ENHANCEMENT OF THE Ni-d_{x^2-y^2} ORBITAL IN DMFT

In Fig. 1 of the main text, the scales of the lower and upper x-axis already provide a translation between the Sr-doping (in the virtual crystal approximation) and the doping of the Ni-d_{x^2-y^2} orbital (or the one-band Hubbard model). In Fig. S.3 we additionally show the functional dependence explicitly, and also compare the NdNiO with the LaNiO compound. For both compounds we see that a similar amount of ~ 60% of the holes go to the Ni-d_{x^2-y^2} orbital. For (very) small dopings a little bit less so in case for NdNiO than for LaNiO because we also have to depopulate the Γ-pocket, whereas this is already shifted above E_F for LaNiO if we include the La-5d interaction in DMFT. The mass enhancement gradually decreases with the doping of the d_{x^2-y^2} orbital, as close to half-filling electronic correlations are strongest. For the parent compound LaNiO the mass enhancement is slightly larger than for NdNiO, which agrees with the observation that it is closer to half-filling.

![Graph showing doping and mass enhancement](image)

**FIG. S.3.** Occupation of the Ni-d_{x^2-y^2} orbital [blue; right y-axis], its effective mass enhancement m*/m [black ; left y-axis in panels (a,c)] and quasiparticle renormalization Z [red ; left y-axis in panels (b,d)] vs. Sr-doping for LaNiO (left) and NdNiO (right).

### S.4. ADDITIONAL DMFT SPECTRA

In the main text, we have already shown the k-dependent DMFT spectral function of Sr_{0.2} Nd_{0.8}NiO in Fig. 2. In Fig. S.4 we supplement this with the spectral functions at 0 and 30% Sr-doping. For those Sr-dopings we have already presented the k-integrated spectra in Fig. 2 of the main text. One sees that for the parent compound, NdNiO, there is a Γ pocket whereas it is shifted above E_F for Sr_{0.3}Nd_{0.7}NiO, as well as for Sr_{0.2}Nd_{0.8}NiO in the main text. For Nd_{0.7}Sr_{0.3}NiO the other Ni-3d bands almost touch E_F at the A-point. Hence around this doping a one-band Hubbard model description is not possible any longer, all Ni orbitals and the interaction among these needs to be taken into account.
With Fig. S.5, we provide for exactly the same overview of the DMFT results as in Fig. 2 of the main text but now for LaNiO$_2$ instead of NdNiO$_2$. An important difference is that for LaNiO$_2$ the Ni-$d_{xy}$ is immediately below $E_F$ already at 20% Sr-doping. At 30% Sr-doping it accommodates already many holes and crosses the Fermi energy. Hence, in case of Sr-doped LaNiO$_2$ the one-band Hubbard model description is only valid up to about Sr-20% doping.

Another difference is that the Γ pocket, which was present in DFT, is shifted considerably higher up in energy in Fig. S.5 (d,e). This agrees with the aforementioned observation that it is absent already for the parent compound LaNiO$_2$.

**FIG. S.4.** DMFT $k$-resolved spectral function $A(k,\omega)$ of undoped NdNiO$_2$ (a-b) and 30% Sr-doped NdNiO$_2$, i.e., Sr$_{0.3}$Nd$_{0.7}$NiO$_2$ (c-d). Panels (b,d) are zoom-ins of (a,c). White lines are the corresponding Wannier bands.

### S.5. $U$-DEPENDENCE OF THE DΓA RESULTS

In this section, we show the dependence of our DΓA results within a reasonable range of $U$ values above the cRPA value of $U = 2.6\,eV = 6.7t$. Specifically, in addition to $U = 8t$ (in the main text), we consider $U = 7t$ and $9t$. As already pointed out in the main manuscript, the adequate value should be a bit larger than the cRPA value if we disregard the frequency dependence of $U$. These $U = 7t$ and $U = 9t$ values are the smallest and largest $U$ value, respectively, which we consider still conceivable given the cRPA calculated value. As for the hopping parameters we have employed the rounded ratios $t'/t = -0.25$ and $t''/t = 0.12$ in DΓA.

In Fig. S.6, we show the momentum dependence of the $k$-resolved spectrum and the superconducting eigenvalue ($\lambda_{SC}$) vs. temperature ($T$), which are the same plot as the Fig. 3 in the main text, but now at $U = 7t$ and $U = 9t$ instead of $U = 8t$. For $U = 7t$, the self-energy damping effect becomes smaller and we can still see the Fermi surface (peak of the spectrum) around $k = (0, \pi)$ even for low dopings. The superconducting eigenvalue $\lambda_{SC}$ and hence the superconducting susceptibility is slightly increasing towards low doping: $n_{d_{x^2-y^2}} = 0.90$. For $U = 9t$, we see that the damping effect becomes stronger instead and there is a strong momentum dependence even for large doping ($n_{d_{x^2-y^2}} = 0.80$). A consequence of this increased damping is that $\lambda_{SC}$ becomes smaller than for $U = 7$ and $U = 8t$.

We also show the phase diagram for these three $U$ values in Fig. S.7. As discussed in the main paper the phase diagrams are obtained from $\lambda_{SC} \rightarrow 1$, which for higher $T_C$’s is interpolated and for lower $T_C$’s extrapolated with a fit function of the form $a - b \ln(T)$. This fit function is plotted as a dashed line (also in Fig. 4 of the main text). As a matter of course the extrapolation over a large temperature interval, i.e. for $n_{d_{x^2-y^2}} = 0.775$ in Fig. 4 of the main text and $n_{d_{x^2-y^2}} = 0.875$ in S.7 (d), leads to a large uncertainty. We hence cannot say whether $T_C$ is still finite for $n_{d_{x^2-y^2}} = 0.775$ or whether we are already outside the superconducting regime at this doping.
FIG. S.5. DMFT k-integrated (a-c) and k-revolved (d-e) spectral functions $A(\omega)$ and $A(k, \omega)$ of undoped LaNiO$_2$ (a), 20%Sr-doped LaNiO$_2$ (La$_{0.8}$Sr$_{0.2}$NiO$_2$) (b) and 30%Sr-doped LaNiO$_2$ (La$_{0.7}$Sr$_{0.3}$NiO$_2$) (c). The k-revolved spectral function $A(k, \omega)$ of La$_{0.8}$Sr$_{0.2}$NiO$_2$ is shown in (d); (e) is a zoom-in of (d).

The physical conclusions from Fig. S.7 are: $T_C$ goes down and the optimal doping level moves to the larger doping side as we go toward stronger interaction. At 20% Sr-doping, all results indicate a bit higher $T_C$ than the experimental $T_C$. The result for $U = 9t$ would be very close to the experimental result, but we think that the theoretical $T_C$ has to be larger than the experimental one (for the reasons stated in the main manuscript). This also agrees with our expectation that $U = 8t$ is probably the best estimate for a static interaction parameter. Our results indicate that nickelates are in the strong-coupling, larger $U$ regime with a dome-shaped $T_C$ vs. $U$ phase diagram, similar as is considered to be the situation for the cuprates.

An important conclusion from Fig. S.7 is that larger critical $T_C$’s can be obtained by enhancing the bandwidth and reducing the ratio of Coulomb interaction to bandwidth. This can be achieved by strain or by replacing Nd(La)NiO$_2$ with Nd(La)PdO$_2$ which has $t = -t_{100} = 537$ meV instead of $t = 395$ meV [see Table S.1]. The cRPA $U$ value does not change that strongly$^{18}$, so that we have $U/t \approx 6$ for Nd(La)PdO$_2$ instead of $U/t \approx 8$ for Nd(La)NiO$_2$.

S.6. COMPARISON WITH EXPERIMENTAL PHASE DIAGRAM

*Added:* Here, we plot on top of Fig. 1(b) of the main text also the recently determined experimental phase diagram of Sr$_2$Nd$_{1-x}$NdO$_2$ by Li *et al.*$^{19}$ Since we calculate the onset of the second order phase transition to the superconducting phase, we compare to the experimental onset of the phase transition ($T_{c,90\%}$ in Ref.19) and the upper limit in Ref. 20. As already mentioned in the last section, the extrapolation for $n_{d_{x^2-y^2}} = 0.775$ (22.5% doping of the Hubbard model, $\approx 30\%$ Sr doping) covers a too large temperature range to say for sure whether $T_C = 0$ or small but finite.

Please consider that our calculation is a prediction of a most difficult quantity to calculate, namely the superconducting $T_C$ and its doping dependence, and that we discussed already before that theory should overestimate $T_C$, while the experimentally observed $T_C$ is likely suppressed by extrinsic contributions such as disorder etc. One may of course most easily compensate this over- and underestimation by changing the $U$ value. Indeed, $U = 9t$ of Fig. S.7 would be in almost perfect agreement with the experimental phase diagram, it also was already in excellent with the previously only available data point (Ref.20; “Experiment” in Fig. S.8).

While $U = 9t$ would be essentially on top of the experimental phase diagram, we raised some factors that theory overestimates $T_C$ on the other hand, and we still think that the true $U$ is $8t$ or in-between $U = 8t$ and $U = 9t$, and that further improving the calculation and purifying the crystals will eventually converge experimental and theoretical phase diagrams.
FIG. S.6. (a) Imaginary part of the Green function at the lowest Matsubara frequency $A(k, \omega_0 \equiv \pi/\beta) \equiv -\Im G(k, \omega_0)/\beta$ and (b) temperature dependence of the superconducting eigenvalue ($\lambda_{SC}$) for $U = 7t$. (c,d) Same figures but for $U = 9t$.

We conclude that given this reasonable over- and underestimation we have a very good agreement of the absolute value of $T_C$ and its doping dependence.
FIG. S.7. Superconducting $T_C$ vs. Sr-doping ($d_{x^2−y^2}$ filling on the upper x-axis) comparing $U = 7t$, $8t$ and $9t$.

FIG. S.8. Comparison of the theoretical phase diagram at $U = 8t$ with the experimental phase diagram of Ref. 19 (“arXiv: 2003.08506”); “Experiment” refers, as before, to Ref. 20. While $U = 9t$ in Fig. S.7 would be in almost perfect agreement with experiment, we believe the $T_C$ of the present DΓA calculation should be slightly larger than the experimental one.

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than NdNiO$_2$. Note that the bandwidth of NdPdO$_2$ is even larger than for the compounds considered in Ref. 21.

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