Optimizing superconductivity: from cuprates via nickelates to palladates

Motoharu Kitatani\textsuperscript{a,b}, Liang Si\textsuperscript{c,d}, Paul Worm\textsuperscript{d}, Jan M. Tomczak\textsuperscript{d,e}, Ryotaro Arita\textsuperscript{b,f} and Karsten Held\textsuperscript{d}

\textsuperscript{a}Department of Material Science, University of Hyogo, Ako, Hyogo 678-1297, Japan
\textsuperscript{b}RIKEN Center for Emergent Matter Sciences (CEMS), Wako, Saitama, 351-0198, Japan
\textsuperscript{c}School of Physics, Northwest University, Xi’an 710127, China
\textsuperscript{d}Institute of Solid State Physics, TU Wien, 1040 Vienna, Austria
\textsuperscript{e}Department of Physics, King’s College London, Strand, London WC2R 2LS, United Kingdom and
\textsuperscript{f}Research Center for Advanced Science and Technology, University of Tokyo 4-6-1, Komaba, Meguro-ku, Tokyo 153-8904, Japan

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Motivated by cuprate and nickelate superconductors, we perform a comprehensive study of the superconducting instability in the single-band Hubbard model. We calculate the spectrum and superconducting transition temperature \(T_c\) as a function of filling and Coulomb interaction for a range of hopping parameters, using the dynamical vertex approximation. We find the sweet spot for high \(T_c\) to be at intermediate coupling, moderate Fermi surface warping, and low hole doping. Combining these results with first principles calculations, neither nickelates nor cuprates are close to this optimum within the single-band description. Instead, we identify some palladates, notably RbSrPdO\(_3\) and A\(_2\)PdO\(_2\)Cl\(_2\) (\(A=\text{Ba}_{0.5}\text{La}_{0.5}\)), to be virtually optimal, while others, such as NdPdO\(_2\), are too weakly correlated.

Introduction—Ever since the discovery of cuprate superconductivity \cite{1}, the material dependence of the transition temperature \(T_c\) and exploring routes toward optimizing \(T_c\) are a central quest of condensed matter physics. The recently discovered nickelate superconductors provide a new perspective to this quest. Similar to cuprates, superconductivity in nickelates emerges from a doped \(3d^{\delta-\delta} (\delta \sim 0.2)\) electronic configuration of the transition metal. Besides the initial infinite-layer superconductor Nd\(_{1-x}\)Sr\(_x\)NiO\(_2\) \cite{2–8}, substituting neodymium with another lanthanoid \cite{9–11} and also the quintuple-layer compound \cite{12} show superconductivity. This indicates that, akin to cuprates, there is a whole family of nickelate superconductors.

As for the theoretical modeling, the one-band Hubbard model is arguably the simplest effective model for cuprates \cite{13, 14}. Its tight-binding parameters can be obtained from \textit{ab initio} calculations, and the relation between model parameters and the experimental \(T_c\) has been analyzed \cite{15–27}. For nickelates, a similar scenario (1) with a one-band Hubbard model plus largely decoupled electron pockets has been put forward \cite{28–35}. Based on the same density functional theory (DFT) and dynamical mean-field theory (DMFT) Fermi surface with Ni \(3d_{x^2−y^2}\) orbital plus vertex corrections around \(A\) and \(\Gamma\) momentum, a second group of scenarios \cite{36–39} emphasizes the role of holes in the Ni \(3d_{x^2−y^2}\) orbital. These originate from an admixture around the \(\Gamma\) pocket that is predominately Nd \(5d_{x^2−y^2}\). In scenario (1) this is argued not to be of primary importance for superconductivity because of the strong doping and rare earth cation dependence of the \(\Gamma\) pocket \cite{3, 40–42}. Finally, scenario (3) proposes an \textit{additional} Ni \(3d_{x^2−y^2}\) Fermi surface based on self-interaction corrected (sic) DFT+DMFT \cite{32, 43–46}. Such an additional Fermi surface is also obtained in antiferromagnetically ordered DFT \cite{47, 48}, \(GW+\text{DMFT}\) \cite{49}, and DFT+DMFT in the overdoped region \cite{28, 50}.

While the relevant low energy model for nickelates is still under debate, a boost for scenario (1) was its successful prediction of the superconducting phase diagram \cite{28} prior to experiments \cite{5, 6} and with high accuracy in the light of new, defect-free films \cite{51}. Also some other experiments including, among others, the Hall coefficient, resonant x-ray spectroscopy \cite{52} and magneto transport \cite{53} point toward this scenario. As for the microscopic origin of high-\(T_c\) superconductivity: while spin fluctuations mediate superconductivity in Ref. \cite{28}, the topic remains highly controversial; many different mechanisms have been proposed \cite{54–57}.

The aim of the present paper is hence two-fold: First, we would like to identify the optimal conditions for superconductivity in the Hubbard model building upon recent progress made with diagrammatic extensions of DMFT \cite{58}. In particular, we will employ the dynamical vertex approximation (DVA) \cite{59}, which accurately describes antiferromagnetic spin fluctuations in the parameter range where numerical quantum Monte-Carlo simulations are still available \cite{60}. Second, from a materials point of view we would like to identify cuprate- or nickelate-like materials that promise even higher \(T_c\)’s. One important factor is the interaction strength \(U\) and its ratio to the hopping \(U/t\). On a qualitative level it has been recently found \cite{28, 61} that the interaction strength is too large in nickelates. Higher \(T_c\)’s should thus be possible using compressive strain or pressure \cite{62}, confirmed experimentally by a record \(T_c > 30\) K for nickelates under a pressure of 12 GPa with no saturation yet discernible \cite{63}. Much more dramatic changes of \(U/t\) are possible when going from \(3d\) to \(4d\) transition metal oxides, which
can be achieved by replacing Ni with Pd [28, 33, 64, 65]. Here, we study this possibility on a quantitative level. We show that a one-band description is justified for palladates, and make a prediction of the superconducting phase diagram which can be tested in experiment.

Let us start by sketching the electronic structures of the above-mentioned materials schematically in Fig. 1. As is well known, the parent cuprate compounds are charge transfer insulators [66]: Cu-3d and O-2p bands exist at a similar energy level and are strongly hybridized. Thus the Hubbard model is justified only as an effective model that mimics the physics of the Zhang-Rice singlet [14]. In contrast, the Ni 3d-orbital is higher up in energy by $\sim 1\text{ eV}$. The increased distance to the oxygen orbitals then makes the single-$d_{x^2-y^2}$-orbital description more suitable than for the CuO$_2$ layers in cuprates. However, density-functional theory (DFT) calculations [61, 65, 67–72] show that the Ni-d bands of NdNiO$_2$ now partially overlap instead with the bands of Nd in-between the NiO$_2$ layers, forming the already mentioned pockets around the A and G momentum.

If we replace Ni (3d) with Pd (4d), the Pd 4d-orbitals are shifted back down by $\sim 1\text{ eV}$ due to the higher ionization energy of Pd compared to Ni. This removes the pockets present in the nickelates and leads to a larger d-p hybridization and minor overlap with the oxygen bands. However, we do not yet have a charge transfer state as in the cuprates. Indeed, our DFT and DMFT computations (shown in Supplementary Materials (SM) [73] Section I–III) of the crystal and electronic structures of the nickelate NdNiO$_2$ and the palladates NdPdO$_2$, RbSr$_2$PdO$_3$ and $A_2'$PdO$_2$Cl$_2$ show that palladate compounds are somewhere in-between cuprates and nickelates – with a single-band $d_{x^2-y^2}$ Fermi surface justifying a modelling by a single-orbital Hubbard model. Tuning the dispersion and interaction strength in palladates thus opens so-far untapped possibilities for finding new superconductors with possibly higher $T_c$'s.

FIG. 1. (Color online) Schematic picture of the energy levels for copper (Cu$^{2+}$), nickel (Ni$^{+}$), palladium (Pd$^{+}$) superconductors.

| Material            | $|t|(\text{meV})$ | $t'/t$       | $t''/t$ | $U_{\text{eff}}/t$ |
|---------------------|------------------|--------------|---------|------------------|
| NdNiO$_2$           | 395              | -0.25        | 0.12    | 8                |
| NdNiO$_2$(Strained) | 419              | -0.23        | 0.12    | 7.0–7.5          |
| NdPdO$_2$           | 558              | -0.17        | 0.13    | 4.5              |
| RbSr$_2$PdO$_3$     | 495              | -0.24        | 0.16    | 6                |
| $A_2'$PdO$_2$Cl$_2$ | 443              | -0.22        | 0.14    | 7.5              |
| $A_2'$PdO$_2$Cl$_2$ | 470              | -0.22        | 0.14    | 7.0              |
| $A_2'$PdO$_2$Cl$_2$ | 497              | -0.22        | 0.14    | 6.0              |

TABLE I. Summary of the DFT-derived parameters for the single-band Hubbard model, as an effective low-energy model for the nickelate NdNiO$_2$, the palladates NdPdO$_2$, RbSr$_2$PdO$_3$, and $A_2'$PdO$_2$Cl$_2$.

Model and Method—We study the two-dimensional Hubbard model on the square lattice with Hamiltonian

$$\mathcal{H} = \sum_{k,\sigma} \epsilon_k c_{k,\sigma}^\dagger c_{k,\sigma} + U \sum_{i} n_{i,\uparrow} n_{i,\downarrow},$$  \hspace{1cm} (1)

where $c_{k,\sigma}^\dagger$ ($c_{k,\sigma}$) is the creation (annihilation) operator,

$$\epsilon_k = -2t[\cos(k_x) + \cos(k_y)] - 4t'\cos(k_x)\cos(k_y) - 2t''[\cos(2k_x) + \cos(2k_y)],$$  \hspace{1cm} (2)

the energy-momentum dispersion, $U$ the onsite Coulomb repulsion, and $t, t', t''$ are the nearest, second nearest, and third nearest hoppings, respectively. The model parameters are obtained from DFT, using WIEN2K [74] with the PBE [75, 76] exchange correlation functional and WIEN2WANNIER [77] for projecting onto a maximally localized $3d_{x^2-y^2}$ Wannier orbital [78]. SM Sec. VI, VII, VIII, and IX provides details on the Wannier projection, the DFT calculation of $A_2'$PdO$_2$Cl$_2$, the stability against structural distortions, and the antiferromagnetic DFT solution, respectively. Before constructing the single-orbital model for palladates, we performed multi-orbital DMFT calculations which confirm the single orbital nature of the system, see SM [73] Sec. III. The constrained random phase approximation (cRPA) is employed to estimate $U$. Following the previous research [28], we employed slightly enhanced values ($+0.35\text{ eV}$ [79]) from our cRPA calculation for entangled bands [80]: 2.85 eV for NdNiO$_2$, 2.55 eV for RbSr$_2$PdO$_3$, and 2.97 eV for $A_2'$PdO$_2$Cl$_2$ (which are consistent with the preceding study [65]; please note that small changes in the interaction strength $U$ do not change our conclusion). Table I provides a summary of the DFT and cRPA derived parameters [81] (for details see SM [73] Section II), which are used in subsequent DΓA calculations. Besides these material-specific Hubbard models we also include the simplest case with nearest-neighbor hopping only ($t'=t''=0$).

We analyze these single-orbital models by means of the DΓA [59, 82–84], a diagrammatic extension of DMFT [85–87]. Similar techniques have been previously applied
to unconventional superconductivity on the square lattice [88–95]. We mainly use the continuous-time quantum Monte-Carlo solver from w2dynamics [96] as DMFT solver, see SM [73] for details. DΓA can describe dynamical screening effects which are crucial for accurately determining $T_c$ [97]. It has predicted the superconducting dome in nickelates [28] prior to experiments [5, 6, 51] with astonishing accuracy. For a review of DΓA, see [58]; and [98] for how to calculate $T_c$.

Spectrum—We first discuss the electronic spectrum. In Fig. 2, we show the momentum dependence of the imaginary part of the Green’s function $-\Im G/\pi$ for various interactions: $U/t = 4.3, 6, 7, 8$ and fillings $n = 0.80, 0.85, 0.90, 0.95$ for hoppings corresponding to NdPdO$_2$ ($t'/t = -0.17, t''/t = 0.13$) and NdNiO$_2$ ($t' = -0.25t, t'' = 0.12t$). First, we notice in all cases a strong suppression of $\lambda$ around half-filling at strong coupling. This leads to a dome structure of $T_c$ as a function of both interaction strength and filling, which is essential for optimizing superconducting materials.

Even outside the pseudogap region, correlation effects change the Fermi surface structure. Specifically, they decrease the Fermi surface warping, i.e. effectively decrease $t'$. Such change of the spectrum for large $(t', t'')$ region has also been observed in other theoretical studies [99–101]. These Fermi surface changes also affect the superconducting instability. While this effect (coming from the momentum dependence of $\Im G$) is minor compared with the pseudogap physics (stemming from the momentum dependence of $\Sigma$), we find that such a Fermi surface flattening can enhance superconductivity, but only around the optimal conditions in parameter space; see SM [73] for a detailed discussion.

As demonstrated here, DΓA properly captures correlation induced changes of the Fermi surface (e.g., Fermi arc in cuprates [102]) and is consistent with previous results. Superconductivity—Next, we discuss the superconducting instability. To do so, we calculate the eigenvalues of the linearized gap (Eliashberg) equation which is the usual procedure for evaluating the superconducting instability from the paramagnetic solution. The eigenvalue $\lambda$ is a measure of the superconducting instability, and $T_c$ is identified by $\lambda$ reaching unity. While DΓA is unbiased with respect to spin, charge and quantum critical fluctuations, we find that spin fluctuations mediate $d$-wave superconductivity in all cases studied.

In Fig. 3, we plot the superconducting eigenvalues against the interaction $U$ and the filling for three tight-binding parameter sets: the simplest case ($t' = t'' = 0$), parameters for NdPdO$_2$ ($t' = -0.17t, t'' = 0.13t$) and NdNiO$_2$ ($t' = -0.25t, t'' = 0.12t$). First, we notice in all cases a strong suppression of $\lambda$ around half-filling at strong coupling. This leads to a dome structure of $T_c$ as a function of both interaction strength and filling, which is essential for optimizing superconducting materials.

Fig. 3 unequivocally reveals that the origin of this suppression is too strong antiferromagnetic correlations (dark red color scale). These open a pseudogap in Fig. 2 and thus suppress the electron propagator. Even though
the antiferromagnetic pairing glue is huge, this eventually suppresses superconductivity.

Let us note that cluster DMFT [103] shows a similar tendency as in Fig. 3, albeit with still much higher temperatures and weaker superconducting fluctuations. The pseudogap behavior is, on the other hand, consistent with the observation that the insulating regime expands if long-range spatial fluctuations are properly included [60, 104, 105].

Except for the perfectly nested case ($t' = t'' = 0$) where spin-fluctuations are overly strong and open a gap regardless of interaction strength, we can separate the $\lambda$ dependence of the superconducting eigenvalues $\lambda$ at $T = 0.01t$ for three different $t', t''$ from Table I. For each $U$, the linewidth corresponds to the range of $\lambda$ for fillings $0.80 \leq n \leq 0.90$. Materials corresponding to these models and $U$'s are indicated.

In Fig. 4, we also show the points corresponding to each material from Table I. Further we include HgBa$_2$CuO$_4$ as a typical cuprate which has $U/t \sim 7.5t - 8t$ [22, 24, 25] and almost the same $t', t''$ as $A_2'$PdO$_2$Cl$_2$. As mentioned in previous works [28, 61], we can see that NdNiO$_2$ has a too large interaction and thus falls outside the area with highest $T_c$.

Following this insight, we can rationalize now the recent experimental achievements of realizing higher $T_c$'s in NdNiO$_2$: both external pressure [63] and in-plane lattice compressive strains [51, 108] play similar roles at reducing the in-plane lattice constant, shrinking the Ni-Ni distance and weakening the correlation strength $U/t$, while increasing the magnetic exchange $4t^2/U$ [109]. Simply replacing Ni-3$d$ by Pd-4$d$ yields NdPdO$_2$ which is too weakly correlated in Fig. 4. Let us emphasize that this concerns tetragonal NdPdO$_2$ which can be stabilized in thin films. In the bulk and for thick films, there will be a substantial orthorhombic distortion, see SM [73] Section VIII. Due to the tilting of the PdO$_6$ octahedra, $t$ is reduced to 304 meV. This pushes orthorhombic NdPdO$_2$ to $U/t \sim 7$, i.e., close to the optimum in Fig. 4 (average $t'/t \approx 0.25$; $t''/t \approx 1$). However this enhancement of $T_c/t$ is largely compensated by the smaller $t$, altogether yielding a similar $T_c$.

A more promising solution to increase $U/t$ and thus $T_c$ is to enlarge the lattice which is possible by inserting spacing layers between the PdO$_2$ planes. Doing so, our DFT and cRPA calculations indeed place RbSr$_2$PdO$_3$ and $A_2'$PdO$_2$Cl$_2$ close to the optimum.

We finally show in Fig. 5 the phase diagram for these palladate compounds, using the same approach as previously for nickelates [28]. We predict palladates to

$\text{FIG. 4. (Color online) Interaction } U \text{ dependence of the superconducting eigenvalues } \lambda \text{ at } T = 0.01t \text{ for three different } t', t'' \text{ from Table I. For each } U, \text{ the linewidth corresponds to the range of } \lambda \text{ for fillings } 0.80 \leq n \leq 0.90. \text{ Materials corresponding to these models and } U \text{'s are indicated.}$

$\text{FIG. 5. (Color online) Phase diagrams for nickelates (NdNiO}_2\text{) and palladates (RbSr}_2\text{PdO}_3\text{ and } A'_2\text{PdO}_2\text{Cl}_2\text{). In the red region, we expect antiferromagnetism instead of superconductivity for } A'_2\text{PdO}_2\text{Cl}_2 \text{ if there is a weak inter-layer coupling because of the huge } \chi_{sp} \text{ (color code of } \chi_{sp} \text{ in DFT as in Fig. 3).}$
have a \( T_c \gtrsim 60 \) K, which touches the floor level line of cuprates and remarkably exceeds the current upper limit of nickelates, \( T_c = 30 \) K [51, 63]. Indeed, the calculated phase diagram (superconductivity and antiferromagnetism) for \( \mathcal{A}_2 \text{PdO}_2\text{Cl}_2 \) is quite similar to well-known cuprate phase diagrams. Furthermore, weak strain (described by \( U = 6.5t, 7t \) [110], c.f., Table I) would tune the material to an even higher \( T_c \).

Conclusion and outlook—Building on the success of DGA to predict the superconducting dome in nickelates, we have performed a comprehensive survey of the Hubbard model and revealed the optimal phase-space region for unconventional superconductivity. Conditions are optimal at intermediate coupling (\( U/t = 6 - 7 \)), moderate Fermi surface warping (\( |t'| + |t''| \approx 0.3 - 0.4 \)), and low hole doping (\( n \sim 0.90 - 0.95 \)). Combining this insight with first principles calculations, we predict palladates and nickelates grown on compressive substrates to be superconductors with a \( T_c \) comparable to cuprates.

The theoretically proposed palladates have yet to be synthesized. Palladates with a perovskite-like structure [111, 112], however, have already been realized in experiment. Then, provided that a reduction process, similar to that of NdNiO\(_3\) \( \rightarrow \) NdNiO\(_2\), is possible, realizing palladates with PdO\(_2\) layers is a promising route for high-\( T_c \) superconductors. Additionally, the possibility to engineer cuprate analogs based on 4d materials has been discussed based on AgF\(_2\) [113] and silver oxides [114]. Analyzing the relation between those systems and the ones we are proposing in this letter will offer fresh insight into the design of superconductors.

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The rather crude classification [(1) 3d_{x^2} plus decoupled pockets; (2) same Fermi surface as (1) but relevance of 3d_{z^2} holes and coupling to the pockets; (3) additional 3d_{z^2} Fermi surface, possibly no pockets] neglects finer details, e.g., Hund’s vs. Kondo physics in (2) or the conjectured importance of 4f electrons [116, 117].

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Supplementary material for “Optimizing superconductivity: from cuprates via nickelates to palladates”

Motoharu Kitatani\textsuperscript{a,b}, Liang Si\textsuperscript{c,d}, Paul Worm\textsuperscript{d}, Jan M. Tomczak\textsuperscript{d}, Ryotaro Arita\textsuperscript{b,c} and Karsten Held\textsuperscript{d}

\textsuperscript{a}Department of Material Science, University of Hyogo, Ako, Hyogo 678-1297, Japan
\textsuperscript{b}RIKEN Center for Emergent Matter Sciences (CEMS), Wako, Saitama, 351-0198, Japan
\textsuperscript{c}School of Physics, Northwest University, Xi’an 710127, China
\textsuperscript{d}Institute of Solid State Physics, TU Wien, 1040 Vienna, Austria and
\textsuperscript{e}Research Center for Advanced Science and Technology, University of Tokyo 4-6-1, Komaba, Meguro-ku, Tokyo 153-8904, Japan

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This supplementary material contains additional results obtained by density functional theory (DFT), dynamical mean field theory (DMFT) and dynamical vertex approximation (DTA) that further corroborate our conclusions of the main text and is organized in nine sections. In Section I, we perform discussions on the crystal structures, parameters and analyze similarities and differences between the nickelate NdNiO\textsubscript{2} and the palladates NdPdO\textsubscript{2}, RbSr\textsubscript{2}PdO\textsubscript{3} and A′\textsubscript{2}PdO\textsubscript{2}Cl\textsubscript{2} (A′ = La\textsubscript{0.5}Ba\textsubscript{0.5}). Section II provides detailed DFT (and beyond DFT) results of electronic structure computations for all compounds. In Section III we show the DMFT results, including spectral functions and effective mass, for RbSr\textsubscript{2}PdO\textsubscript{3} and A′\textsubscript{2}PdO\textsubscript{2}Cl\textsubscript{2}. Section IV shows the self-energy effect on the spectrum and the superconductivity instability in more detail. In Section V, we show the numerical details on the DTA study of superconductivity. Further, in Section VI we present our Wannier function projection and in Section VII the computational details of the virtual crystal approximation. Section VIII analyzes the phonon dispersion and structural distortions of LaPdO\textsubscript{2}. Finally in Section IX we compute the DFT total energy for different magnetic states of A′\textsubscript{2}PdO\textsubscript{2}Cl\textsubscript{2}.

I. CRYSTAL STRUCTURE OF NICKELATES AND PALLADATES

The crystal structures of nickelate NdNiO\textsubscript{2} and palladates NdPdO\textsubscript{2}, RbSr\textsubscript{2}PdO\textsubscript{3} and A′\textsubscript{2}PdO\textsubscript{2}Cl\textsubscript{2} are shown in Fig. S.1. To guarantee a 3d\textsuperscript{9} configuration of the Ni\textsuperscript{2+} cation in A′\textsubscript{2}PdO\textsubscript{2}Cl\textsubscript{2}, we need a A′\textsuperscript{2.5+} cation which can be achieved by mixing 50%La and 50%Ba. To approach optimal filling/doping in the proposed palladates RbSr\textsubscript{2}PdO\textsubscript{3} and A′\textsubscript{2}PdO\textsubscript{2}Cl\textsubscript{2}, the ration between Rb\textsuperscript{1+}/Sr\textsuperscript{2+} for the former and La\textsuperscript{3+}/Ba\textsuperscript{2+} for the later case can be tuned.

The relaxed parameters for all nickelates and palladates are shown in Table S.1. For the DFT-level calculations, PBESol [1] is employed for structural relaxations, and PBE [2] for electronic structure calculations (e.g., DFT band structures and tight-binding Wannier projections), following the lines of Ref. [3]. Our PBESol structural relaxations yield the lattice constants of NdNiO\textsubscript{2} a=b=3.864 Å and c=3.243 Å. To simulate in-plane compressive strain effects, we tune the in-plane lattices to 3.800 Å [of bulk LaAlO\textsubscript{3} (LAO)], the c-lattice is then simultaneously relaxed to 3.330 Å, assuming that the films are several layers thick as in experiments for nickelate superconductors, this change of the lattice parameters is the dominant effect of the substrate; apart from that a bulk DFT calculation is justified. The corresponding changes of hoppings and U are shown in Table I of the main text.

For NdPdO\textsubscript{2}, an in-plane lattice expansion from 3.864 Å to 4.085 Å results from substituting Ni by Pd. Such an expansion may contribute to smaller orbital overlap and stronger electronic correlations if the B-site atoms are unchanged, however the more delocalized 4d-orbitals of Pd counteract this with their larger orbital spreads (see main text). When the in-plane lattices are fixed to the one of SrTiO\textsubscript{3} to simulate the strain effects from a STO substrate, the out-of-plane is relaxed to 3.255 Å, which boosts its two-dimensional characters.

For the proposed palladates RbSr\textsubscript{2}PdO\textsubscript{3} and A′\textsubscript{2}PdO\textsubscript{2}Cl\textsubscript{2}, the in-plane lattice constants are even larger than those of NdPdO\textsubscript{2}. Another notable difference is the distance between Ni-Ni or Pd-Pd along the

![FIG. S.1. Crystal structures of (a) NdNiO\textsubscript{2}, (b) NdPdO\textsubscript{2}, (c) RbSr\textsubscript{2}PdO\textsubscript{3} and (d) A′\textsubscript{2}PdO\textsubscript{2}Cl\textsubscript{2}.](image-url)
TABLE S.I. DFT-PBESol relaxed lattice parameters of NdNiO\(_2\), NdPdO\(_2\) (with full relaxation or grown on SrTiO\(_3\) substrate), RbSr\(_2\)PdO\(_3\) and \(A'\)\(_2\)PdO\(_2\)\(_{\text{Cl}_2}\) (in units of Å). \(A'\) is La\(_{0.5}\)Ba\(_{0.5}\). In the last column we show the appropriate substrates, in the brackets are the in-plane lattice constants (also in unit of Å).

| Substrate          | \(a/b\) | \(c\) | Ni-Ni/Pd-Pd (in-plane) | out-of-plane | Substrate              |
|--------------------|---------|------|------------------------|-------------|------------------------|
| NdNiO\(_2\)        | 3.864   | 3.243| 3.864                  | 3.243       | LSAT (3.868)           |
| NdNiO\(_2\) (strained: on LaAlO\(_3\)) | 3.800   | 3.330| 3.800                  | 3.330       | LaAlO\(_3\) (3.80)     |
| NdPdO\(_2\)        | 4.085   | 3.114| 4.085                  | 3.143       | PrScO\(_3\) (4.02)     |
| NdPdO\(_2\) (strained: on SrTiO\(_3\)) | 3.905   | 3.255| 3.905                  | 3.255       | SrTiO\(_3\) (3.905)    |
| RbSr\(_2\)PdO\(_3\) | 4.264   | 7.555| 4.264                  | 7.555       | MgO (4.20)             |
| \(A'\)\(_2\)PdO\(_2\)\(_{\text{Cl}_2}\) (unstrained) | 4.366   | 14.639| 4.366                  | 7.944       | Rutile-TiO\(_2\) (4.59)/MgO(4.20) |
| \(A'\)\(_2\)PdO\(_2\)\(_{\text{Cl}_2}\) (-1.5% strain) | 4.301   | 14.740| 4.301                  | 7.973       | MgO (4.20)             |
| \(A'\)\(_2\)PdO\(_2\)\(_{\text{Cl}_2}\) (-3% strain) | 4.235   | 14.870| 4.235                  | 8.016       | MgO (4.20)             |

- direction (out-of-plane); it increases to \(~7.5-8.0\) Å, as shown in Table S.I. In the last column we list some suitable substrates for the proposed materials, e.g., for RbSr\(_2\)PdO\(_3\) and strained (-1.5% and 3%) \(A'\)\(_2\)PdO\(_2\)\(_{\text{Cl}_2}\) that host larger in-plane lattice constants than SrTiO\(_3\) (3.905 Å), MgO is considerable to be employed as substrate as its in-plane lattice is \(~4.2\) Å. For the fully relaxed NdPdO\(_2\) (4.085 Å), PrScO\(_3\) (in-plane lattice 4.02 Å) is possible candidate. For compressive cases (when compared with the in-plane lattice of SrTiO\(_3\): 3.905 Å) such as unstrained NdNiO\(_2\) and strained NdNiO\(_2\), one of the possible substrates (LaAlO\(_3\))\(_{0.3}\)(SrAl\(_{0.5}\)Ta\(_{0.5}\)O\(_3\))\(_{0.7}\) (LSAT: 3.868 Å) had been proved as helpful to enhance its \(T_c\) [4]. For the strained case, LaAlO\(_3\) (3.80 Å) is expected to be more effective at eliminating atomic defects and enhancing \(T_c\). The in-plane lattice constant of unstrained \(A'\)\(_2\)PdO\(_2\)\(_{\text{Cl}_2}\) is predicted as 4.366 Å in DFT structural relaxation, we hence proposed rutile-TiO\(_2\) (4.59 Å) and MgO (4.20 Å) as a possible substrate in realistic experiments.

II. ELECTRONIC STRUCTURE OF NICKELATES AND PALLADATES

To investigate the difference between nickelates and palladates, we first compute their band centroids. The bands centroids in Table S.II are computed by \(E_i = \int g_i(E)E dx/\int g_i(E)\), here \(g_i\) is the partial density of states of the corresponding orbital \(i\) and \(E\) is the energy. The integration ranges covers both the bonding and anti-bonding states for Cu/Ni/Pd-\(d\) orbital and O-\(p\) orbitals (up to 5, 7 and 7 eV for CaCuO\(_2\), NdNiO\(_2\) and NdPdO\(_2\), respectively). As one can see, for the cuprate CaCuO\(_2\), both the centroids of Cu-\(d\) and \(d_{x^2−y^2}\) are just \(~0.4\) eV and \(~0.7\) eV higher than that of O-\(p\). This is consistent with the fact that CaCuO\(_2\) is a charge-transfer insulator.

For NdNiO\(_2\), the \(d\)-bands \((d_{x^2−y^2}\) band) shift up by \(~1\) eV \((~0.9\) eV\) compared with those in CaCuO\(_2\). Moreover, the O-\(p\) bands are shifted down by \(~0.9\) eV. As a consequence, the charge transfer energies \(\Delta(d−p)\) and \(\Delta(d_{y^2−z^2}−p)\) in NdNiO\(_2\) are \(~1.8\) eV larger than in CaCuO\(_2\).

For the palladate NdPdO\(_2\), the band centroid of \(d\)-orbitals \(E(d)\) is comparable with those of CaCuO\(_2\) while \(E(d_{y^2−z^2})\) \(~0.3\) eV higher than in CaCuO\(_2\). Thus, the charge transfer energies \(\Delta(d−p)\) and \(\Delta(d_{y^2−z^2}−p)\) are 1.211 and 1.827 eV, i.e., in between the charge transfer energies of CaCuO\(_2\) and NdNiO\(_2\). The computations strongly hint that NdPdO\(_2\) hosts a similar 2D nature as in both CaCuO\(_2\) and NdNiO\(_2\).

In Fig. S.2 we show the DFT-level band structures of NdPdO\(_2\) and RbSr\(_2\)PdO\(_3\) and \(A'\)\(_2\)PdO\(_2\)\(_{\text{Cl}_2}\). NdPdO\(_2\) has a larger \(d_{x^2−y^2}\) bandwidth than NdNiO\(_2\), which is due to its more delocalized 4d orbitals nature. Its pockets at \(\Gamma\) and \(A\) are energetically deeper than in NdNiO\(_2\). However, as concluded by previous research [5], these pockets are not essential for emergent nickelate superconductivity. Additionally, the band structures of \(A'\)\(_2\)PdO\(_2\)\(_{\text{Cl}_2}\) [Fig. S.2(d)] exhibits a Fermi surface with only the Pd-\(d_{x^2−y^2}\) orbital. For RbSr\(_2\)PdO\(_3\) [Fig. S.2(c)], the pockets merely touch the Fermi energy at the \(A\) and \(\Gamma\) momentum. Indeed, the tiny \(\Gamma\) pocket for RbSr\(_2\)PdO\(_3\) is expected to become unoccupied upon hole doping or with exchange-correlations by using functional beyond standard DFT-PBE: the \(\Gamma\)-pocket is absent, e.g., for Rb\(_{1.2}\)Sr\(_1\)PdO\(_3\) [Fig. S.3(a)] and RbSr\(_2\)PdO\(_3\) computed by employing the improved version of the modified Becke-Johnson (mBJ) exchange potential [Fig. S.3(b)]. On a technical note, the process of hole doping and consequently the noninteger filling at Pd-\(d_{x^2−y^2}\) in RbSr\(_2\)PdO\(_3\) is achieved here by employing virtual crystal approximation [6].

In Fig. S.3, the band structures of NdPdO\(_2\) [Fig. S.3(d)] on a SrTiO\(_3\) substrate, RbSr\(_2\)PdO\(_3\) upon 0.2 hole doping per Pb (i.e., Rb\(_{1.2}\)Sr\(_1\)PdO\(_3\)) [Fig. S.3(a)], and the bands calculated by improved version of the modified Becke-Johnson exchange potential [7] [Fig. S.3(b)] are shown; for a comparison further the band of fully relaxed NdPdO\(_2\) are presented in Fig. S.3(c). As shown in
FIG. S.2. DFT band characters of (a) NdNiO	extsubscript{2}, (b) NdPdO	extsubscript{2}, (c) RbSr	extsubscript{2}PdO	extsubscript{3} and (d) A’	extsubscript{2}PdO	extsubscript{2}Cl	extsubscript{2}. Here, we used the fully relaxed structure for all systems. The top panel shows the symbols for different orbitals and the first Brillouin zone. The size of the symbols for Nd-\textit{d}_{xy} are renormalized by a factor of 2 for clearness.

Table S.II. DFT calculated band centroids (in units of eV) of CaCuO	extsubscript{2}, NdNiO	extsubscript{2} and NdPdO	extsubscript{2} and charge transfer energy \(\Delta\).

| Material         | \(E(d)\) | \(E(d_{x^2-y^2})\) | \(E(O-p)\) | \(\Delta(d-p)\) | \(\Delta(d_{x^2-y^2}-p)\) |
|------------------|----------|-------------------|-------------|----------------|-----------------------------|
| CaCuO	extsubscript{2} | -2.367   | -2.047            | -2.772      | 0.405          | 0.725                       |
| NdNiO	extsubscript{2} | -1.384   | -1.148            | -3.674      | 2.289          | 2.525                       |
| NdPdO	extsubscript{2} | -2.358   | -1.742            | -3.570      | 1.211          | 1.827                       |

As we discussed in the main text, the fully relaxed in-plane lattice constants of NdPdO	extsubscript{2} are \(a=b=4.085\) Å, 4.6% more than a SrTiO\textsubscript{3} substrate: 3.905 Å. A suitable substrate for an almost unstrained film is PrScO\textsubscript{3} whose lattice is 4.02 Å (as shown in Table S.I). If NdPdO\textsubscript{2} is grown on SrTiO\textsubscript{3}, the in-plane lattice is fixed as 3.905 Å. As a consequence, the out-of-plane lattice is relaxed to larger value of 3.25 Å, and thus a better 2D character of the \(d_{x^2-y^2}\) orbital. As shown in [Fig. S.3(c-d)], such a strain does not change the main character of the Pd-\(d_{x^2-y^2}\) orbital.

Finally, as Pd is heavier than Ni, one may consider the spin-orbital coupling (SOC) effects in palladates. We hence perform DFT+SOC band computations for NdPdO\textsubscript{2} (fully relaxed case) with and without SOC, as...
FIG. S.3. Comparison between the DFT band characters of (a) hole doped Rb$_{1.2}$Sr$_{1.8}$PdO$_3$, (b) RbSr$_2$PdO$_3$ with mBJ exchange-correlation functional, (c) fully relaxed NdPdO$_2$ and (d) NdPdO$_2$ on SrTiO$_3$ (STO) substrate. The top panel shows the symbols for contribution of the different orbitals and the first Brillouin zone. The size of the symbols for Nd-$d_{xy}$ are renormalized by a factor of 2 for clearness.

shown in Fig. S.4. Fig. S.4(a) and (b) show the DFT band of NdPdO$_2$ without and with SOC, respectively. For better visibility, we also plot these bands in a smaller energy region from -2 to 4 eV in Fig. S.4 (c,d). We find three band-crossing regions in which hybridization gaps open after switching-on SOC effect, see S.4(c). The first region is at $E \sim$-1 to -2 eV and $k=X-M$. Here, the SOC opens a hybridization gap between the Pd-$t_{2g}$ bands. The second region is at $E \sim$-0.5 eV and $k=A$, where there is a band-crossing between Nd-$d_{xy}$ orbitals, which mostly contribute to the $A$-pocket, and Pd-$t_{2g}$ bands. Since these two regions are irrelevant to our target Pd-$d_{x^2-y^2}$ band, the effect on superconductivity will be minor. The third region is at $E \sim$2 eV and $k=A$. In Fig. S.4(c), as one see that there is a double-band degeneracy composed of Nd-$d_{yz}$ and $d_{xz}$ bands, labeled by the arrow in (c). This double degeneracy exchanges its band characters with Pd-$d_{x^2-y^2}$ band at A-point. And the gap opens after including SOC effect because the double degeneracy between Nd-$d_{yz}$ and Nd-$d_{xz}$ is lost, as shown in the orange region in Fig. S.4(d). However, we can exclude this small band-opening as a possible factor suppressing superconductivity in palladates because the energy of this gap is high ($\sim$2 eV) compared to $E_f$ at the very top of the energy region of Pd-$d_{x^2-y^2}$ band (-2 to 3 eV). Additionally, hole-doping will further reduce $E_f$ and weaken this hybridization.

All in all, we conclude that for the low energy physics around the Fermi level, the SOC does not play an important role. This is because there is only a single Pd-$d_{x^2-y^2}$ band crossing the Fermi level. While SOC could be disadvantageous for superconductivity, in principle,
for superconductivity in the studied palladates it does not seem to be relevant. Hence, we do not consider it in the subsequent DMFT and DΓA calculations.

### III. FULL $d$-SHELL PROJECTION IN DMFT

The DFT bands are projected onto maximally localized Wannier functions [8–11] of (i) only the Pd $d_{x^2−y^2}$ band (for the single-band DMFT and DΓA calculations presented in the main text) and of (ii) all Pd $d$ bands (for the full-$d$ set calculations in this section), using the WANNIER90 [12] and WIEN2WANNIER [13] codes. These corresponding low-energy effective tight-binding Hamiltonians are generated and subsequently supplemented by a local Kanamori interaction, using the fully localized limit as double counting [14]. DMFT calculations in the present research are carried out at 300 K with the W2DYNAMICS code [15], which solves the corresponding impurity problem using the continuous time quantum Monte Carlo (CTQMC) approach in the hybridization expansion (CT-HYB) [16]. Real-frequency spectra are obtained with the ANACONT code [17] via analytic continuation using the maximum entropy method (MaxEnt) [18, 19].

The corresponding DMFT results of RdSr$_2$PdO$_3$ and $A_2$PdO$_4$Cl$_2$ are shown in Fig. S.5, Fig. S.6 and Fig. S.7. Fig. S.5 shows the DMFT $k$-resolved ($A(k,\omega)$) and $k$-integrated spectral function ($A(\omega)$) spectral functions of the parent compounds RdSr$_2$PdO$_3$ [Fig. S.5(a)] and $A_2$PdO$_4$Cl$_2$ [Fig. S.5(b)], i.e., at 4$d^9$ Pd electronic configuration which corresponds to a half-filled Pd-$d_{x^2−y^2}$ orbital. Including DMFT electronic correlations, the other $d$-bands ($t_{2g}+d_{z^2}$) of Pd are well separated from

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**FIG. S.4.** Comparison between the DFT band characters of NdPdO$_2$ with (a,c) and without (b,d) SOC. The arrow in (c) indicates the double degeneracy of the Nd-$d_{yz}$ and Nd-$d_{xz}$ orbital at the $A$ point. The dashed orange regions in (d) indicate the SOC induced hybridization gaps.
FIG. S.5. Spectral function, \( k \)-resolved (\( A(k,\omega) \)) and \( k \)-integrated (\( A(\omega) \)), for a full \( d \)-shell projection at a nominal filling of \( n = 9.0 \). (Left) \( \text{RdSr}_2\text{PdO}_3 \) and (right) \( \text{A}'_2\text{PdO}_2\text{Cl}_2 \).

FIG. S.6. Same as Fig. S.5 but now at a nominal filling of \( n = 8.8 \), which corresponds to the superconducting region.

the \( d_{x^2-y^2} \) band. This makes both \( \text{RdSr}_2\text{PdO}_3 \) and \( \text{A}'_2\text{PdO}_2\text{Cl}_2 \) electronically similar to cuprates superconductors.

Large hole doping had been proved being able to destroy the single-band picture and suppress superconductivity states in both cuprates and nickelates. Hence it is worth to investigate the electronic structures of \( \text{RdSr}_2\text{PdO}_3 \) and \( \text{A}'_2\text{PdO}_2\text{Cl}_2 \) with a certain amount of hole doping, in particular, in the hole doping region were superconductivity may be expected. Here, we show thee DMFT spectra of \( \text{Rd}_{1.2}\text{Sr}_{1.8}\text{PdO}_3 \) (\( 4d^{8.8} \)) and
A$_2'$PdO$_2$Cl$_2$ (with $A'^2.4+ = (La_{0.4}Ba_{0.6})^{2.4+}$ and consequently 4$d^{8.8}$). For both compounds, the single-band $d_{x^2-y^2}$ Fermi surface is conserved.

Finally, we investigate the effective mass ($m^*$) of both compounds. Fig. S.7 displays the mass renormalization of the $d_{x^2-y^2}$ band as obtained in DMFT (including all Pd $d$ orbitals). For the A$_2'$PdO$_2$Cl$_2$ compound we show two values of the interaction strength: $U=3.3$ eV for RdSr$_2$PdO$_3$, 3.3 eV and 4 eV for A$_2'$PdO$_2$Cl$_2$. As antiferromagnetic correlations become large towards half-filling so does the effective mass, in particular for the larger interaction. This indicates the strong tendency toward an anti-ferromagnetic Mott insulator at strong coupling. The mass renormalization for both RdSr$_2$PdO$_3$ and A$_2'$PdO$_2$Cl$_2$ at optimal filling for superconductivity, i.e. $n \sim 1 - 0.8$, is around $m^*/m \sim 2$, which is consistent with the values of NdNiO$_2$ as $n$ approaches $\sim 0.82$ per Ni-$d_{x^2-y^2}$: $m^*/m = 2.81$. This comparison indicates that these palladates are quite strongly correlated but less so than NdNiO$_2$. This is advantageous for superconductivity, as discussed in the main text.

IV. EFFECT OF FERMI SURFACE SHAPE ON $T_c$

As shown in the main text, the self-energy weakens the spectral intensity and flattens the Fermi surface shape. Here, we discuss these effects on the Fermi surface structure in more detail, by artificially separating the different effects.

In Fig. S.8, we show the spectrum at two interaction strength [(a) $U = 7.5t$ and (b) $U = 6.5t$] comparing the full DΓA and DMFT self-energy [top left (DΓA) and top right subpanels (DMFT)] in (a) and (b)] to the following artificially modified DΓA self-energies: only the real part $[\text{Im} \Sigma = 0$; middle left subpanels], only the imaginary part $[\text{Re} \Sigma = 0$; bottom left subpanels], momentum-averaged DΓA self-energy $[\Sigma_{\text{ave}}$; middle right subpanels] and momentum average for the real part only, keeping the momentum-dependence of the imaginary part $[\text{Re} \Sigma_{\text{ave}}$, bottom right subpanels]. We fix the filling to the original value (i.e., recalculate the chemical potential) for all cases. We also show the corresponding superconductivity eigenvalues $\lambda$ calculated with these spectra, while keeping the pairing vertex the same for solving the linearized gap equation.

We can first see that the imaginary part of the self-energy has the most relevant effect; it leads to the pseudogap physics, as expected. From DΓA to $\text{Im} \Sigma = 0$ in Fig. S.8, the shape of the Fermi surface remains the same, but its intensity becomes stronger, in particular in the antinodal region around $(\pi, 0)$. The corresponding superconducting eigenvalues is a factor 6-7 higher. This clearly shows that the pseudogap is counterproductive for superconductivity.

Comparing DΓA to $\text{Im} \Sigma = 0$ and the non-interacting Fermi surface (white dots in the top left subpanels) further shows that the flattening of the Fermi surface is still captured for $\text{Im} \Sigma = 0$. That is it is caused by the real part of the DΓA self-energy, hence there is also no flattening in the $\text{Re} \Sigma = 0$ Fermi surface.

Next, if we compare the DMFT self-energy and the momentum averaged DΓA self-energy, they give similar results. That is, the Fermi surface structure is similar to the non-interaction line (white dots in top left subpanels). Some self-energy damping exists in both cases, but as a matter of course, there is no momentum differentiation and hence no pseudogap. In this case, the superconductivity eigenvalues are in between those for $\text{Im} \Sigma = 0$ and the DΓA self-energy.

When ignoring the momentum dependence of the real part of the self-energy, $\text{Re} \Sigma = 0$ and $\text{Re} \Sigma_{\text{ave}}$ give similar results in Fig. S.8. The Fermi surface is the same as the non-interacting case, but strong damping exists around the anti-nodal $(\pi, 0)$ region (and symmetrically related momenta). This is a consequence of the momentum differentiation of the imaginary part of the DΓA self-energy.

When we focus on the effect of the Fermi surface flattening on superconductivity, we can compare the superconducting eigenvalue $\lambda$ for $\text{Re} \Sigma = 0$ and $\text{Re} \Sigma_{\text{ave}}$ with the original DΓA $\lambda$. We surprisingly find the flattening of the Fermi surface suppresses superconductivity at $U = 7.5t$, $t' = -0.22t$, $t'' = 0.14t$, $n = 0.85$ but enhances it at $U = 6.5t$, $t' = -0.25t$, $t'' = 0.12t$, $n = 0.95$.

To study this dichotomy further, We analyze the parameter dependence in Fig. S.9. We can see that there is almost no effect of $\text{Re} \Sigma$ in the weak coupling regime. In contrast, at strong coupling, the eigenvalues are almost everywhere suppressed if the flattening of the Fermi surface is properly included in Fig. S.9 (a). This Fermi
surface flattening, given by the real part of the DΓA self-energy, enhances $T_c$ only around the optimum in the original phase diagram in Fig. S.9(a). That is, for intermediate $U$ and fillings closer to half filling.

This result can be understood as follows: If the strength of the spectrum is similar, i.e., if there is no pseudogap, spectral weight closer to $(\pi,0)$ is favorable because the $(d$-wave) gap function has a peak there and because the van Hove singularity results in a particular strong spectral contribution. On the other hand, if the pseudogap opens around $(\pi,0)$, this trend will eventually reverse. For weak coupling, the Fermi surface flattening does not occur nor does the pseudogap open. There is hence no effect. For larger interactions, the flattening of the Fermi surface moves the Fermi surface toward $(\pi,0)$. At intermediate coupling, there is no pseudogap yet [see Fig. S.8 (b)] and superconductivity becomes enhanced. This leads to the optimum (or hot spot) for superconductivity in Fig. S.9 (a) at $U \approx 6t$ and $n \approx 0.95$. In contrast for larger interactions a pseudogap opens [see Fig. S.8 (a)]. Thus despite the flattened Fermi surface, superconductivity is suppressed. If it was possible to switch of this pseudogap [as in the artificial $\text{Im} \Sigma = 0$ panel of Fig. S.8 (a)] we would get the strongest tendency toward superconductivity in this parameter regime.

V. DETAILS OF DΓA CALCULATION

In this section, we explain computational details for DΓA results. For further details and the general procedure how to calculate spin (and charge) fluctuations and from these superconductivity in ($\lambda$-corrected), we refer the reader to [5, 20].

First, for solving the impurity problem and calculating local vertices, which act as a starting point, we use the continuous-time quantum Monte-Carlo (CT-QMC) on w2dynamics [15] as an impurity solver [except for $U = 9t$ results, which are taken from the previous results [5] using exact diagonalization (ED)]. We have also...
checked that both solvers give quantitatively consistent results at $U = 8t$, $t' = -0.25t$, $t'' = 0.12t$.

In the DΓA calculations, we separate the Matsubara frequency range into two parts following the previous works [5, 20]. We use 120 (80) Matsubara grids for the case $\beta t = 100$, $U \geq 5t$ (others) for which the vertices are treated directly. For an extended grid of 2048 Matsubara frequencies on the positive side we use the bare-$U$ contribution instead of the local vertex. This is sufficient to obtain well-converged results even at low temperatures [21]. For momentum grids, we take a $120 \times 120$ momentum mesh, which may not be enough to get quantitatively converged results in the critically fluctuating regime (dark magenta region in Fig.3), but elsewhere is sufficient [22]. Nevertheless, this mesh is enough to study the superconductivity dominating region, which is the main scope of this paper.

In Fig. S.10, we display the actually calculated data points. From these points, we obtain the contour plots of Fig. 3 in the main text. For the $t' = t'' = 0$ case, we linearly interpolated some irrelevant (i.e., $\lambda \leq 0.6$) points: $(U/t, n) =$ (4.75, 0.775), (5.25, 0.775), (5.5, 0.75), (5.5, 0.775), (5.5, 0.8675) before making the contour plot.

As for the $T_c$ calculation, we calculate the superconducting $\lambda$ down to $T = t/100$, and extrapolated the $\lambda$ vs. $T$ curve (using the form $\lambda \approx a - b \ln(T)$) as in previous publications [5, 20]. As examples, we show in Fig. S.11 DΓA results, fit function, and $T_c$ for (a) $U = 7.5t$, $t' = -0.22t$, $t'' = 0.14t$ and (b) $U = 6t$, $t' = -0.24t$, $t'' = 0.16t$ results (corresponding the RbSr$_2$PdO$_3$ and $\mathcal{M}_2$PdO$_2$Cl$_2$, respectively).

Besides the superconducting eigenvalue, we also calculate the antiferromagnetic (AFM) susceptibility $\chi_{\text{sp}}(Q_{\text{max}}, \omega = 0)$. DΓA respects constraints of the two-dimensionality (i.e., Mermin-Wagner theorem [23, 24]) and does not exhibit an antiferromagnetically ordered phase at finite temperatures ($\chi_{\text{sp}}$ remains finite). On the other hand, we can naturally expect that AFM will stabilize for a strongly fluctuating regime (dark red region in Fig. 3 of the main text) once allowing for a weak threedimensionality, that are present in the actual materials.

VI. WANNIER PROJECTIONS AND CONSISTENCY BETWEEN DFT AND WANNIER BANDS.

Essential for interfacing DFT and DMFT is the Wannier projections. It not only provides the electron hopping parameters for the construction of the minimal low-energy model, but also allows to investigate the degree of localization of the orbitals. An indication for the quality of Wannier projection is its consistency with the target DFT bands. As in previous theoretical studies in the case of nickelates [5, 25], we performed projections for both the single $d_{x^2-y^2}$ band, as well as, all Pd-4d orbitals. In the latter case also additional orbitals that are in the vicinity of, or overlapping with, the Pd bands are included. The single-band $d_{x^2-y^2}$ projection is here used to construct the single-band Hamiltonian for the subse-
FIG. S.12. DFT band structure of (a,b) NdNiO$_2$ and (c,d) NdPdO$_2$. The blue and red dots represent the Wannier bands of projections onto only the Pd-$d_{x^2-y^2}$ orbital and the full Pd-$4d$ plus Nd $5d$ orbitals, respectively. In the Wannier projection for NdPdO$_2$ in (d), further the O-$p$ orbitals are included in the projection because of the stronger Pd-$d$ - O-$p$ hybridization, compared with that of NdNiO$_2$.

The corresponding hopping parameters of the single-orbital projection and the comparison with the original DFT bands is shown in Fig. S.12(a,c), Fig. S.13(a,c) and Table S.III. All these single-band projections for the Ni-$d_{x^2-y^2}$ and the Pd-$d_{x^2-y^2}$ orbital exhibit excellent agreement between the DFT and Wannier band, indicating the quality of these projections and reliability of the corresponding parameters.

For obtaining orbital occupation and spectra, including the positions of pockets, we have done multi-band DMFT calculations, based on the multi-band projections. For the previous calculations of NdNiO$_2$, a Nd(La)-$d$+Ni-$d$ basis with 10-bands was adopted. [26] For NdPdO$_2$, a projection onto 16 bands, Nd(La)-$d$+Ni-$d$+O-$p$, was necessary because of the stronger Pd-$d$-O-$p$ hybridization than in NdNiO$_2$, see Fig. S.12(b,d). For $A_2$PdO$_2$Cl$_2$ and RbSr$_2$PdO$_3$, on the other hand, a projection onto only the Pd-$d$ 5 bands is sufficient, because the other occupied bands are well separated from these Pd-$d$ bands, see Fig. S.13(b,d). All Wannier bands reproduce the DFT bands well, especially the low energy parts near Fermi energy.

VII. COMPUTATIONAL DETAILS OF THE VIRTUAL CRYSTAL APPROXIMATION FOR $A_2$PdO$_2$Cl$_2$

The virtual crystal approximation (VCA) [6] has been implemented in the VASP code and applied to the study of Sn$_x$Ge$_{1-x}$ alloys [27]. Such application of VCA in VASP is justified because both VCA and VASP are based on (Vanderbilt) pseudopotentials. However, because WIEN2K uses the full-potential augmented plane-wave and local-orbitals basis set to solve the Kohn-Sham equations, this full VCA is not applicable in WIEN2K. An alternative way to realize atomic substitution and charge doping effect in WIEN2K is to modify the core and valence electron states in the input configuration files [28].

We perform VCA calculations for $A_2$PdO$_2$Cl$_2$ using WIEN2K and VASP as outlined in the last paragraph. The corresponding bands are shown in Fig. S.14(a) and (b), respectively. As the comparison shows, both codes give exactly the same bands, especially the parts below
the optimized lattice parameters are $a=b=4.126$ Å, $c=3.265$ Å. Compared with La(Nd)NiO$_2$, the $a(c)$ lattice

equation (e) yields essentially the same results as VASP.

In Fig. S.14(a), we additionally show the Wannier band of the Pd $d_{x^2−y^2}$ orbital. The excellent agreement between DFT and Wannier band indicates the high quality of our Wannier projection and localization of the Pd-$d_{x^2−y^2}$ orbital. In Fig. S.14(c,d), we compute and show the DFT band of (c) undoped Ba$_2$PdO$_2$Cl$_2$ and (d) La$_2$PdO$_2$Cl$_2$. In the former and later cases the bivalent Ba$^{2+}$ and trivalent La$^{3+}$ lead to nominal Pd$^{2+}$ ($4d^8$) and Pd$^0$ ($4d^{10}$), respectively. Please note that in La$_2$PdO$_2$Cl$_2$ due to the hybridization between La-$d$ and Pd-$d$, the oxidation state of Pd is in fact between Pd$^{1+}$ and Pd$^0$, as in A$'_2$PdO$_2$Cl$_2$.

In Fig. S.14(c,d), we show again the Pd-$d_{x^2−y^2}$ Wannier band taken from Fig. S.14(a), and add (subtract) a constant, leading to the red dots in Fig. S.14(c,d). The shifted Wannier bands exhibit excellent consistency between the Pd-$d_{x^2−y^2}$ bands in Fig. S.14(c,d), indicating the atomic substitution and charge doping effects can, in fact, be well described by a rigid band shift. This explains why the simplified VCA method in WIEN2K yields essentially the same results as VASP.

**VIII. PHONON CALCULATIONS AND STRUCTURAL DISTORTIONS OF LaPdO$_2$**

To investigate the dynamical stability of NdPdO$_2$, we compute its phonon spectra with the frozen phonon method (finite displacement method) using the PHONONY [29] code interfaced with VASP [30, 31]. For the phonon spectra of the other two palladates, A$'_2$PdO$_2$Cl$_2$ and RbSr$_2$Pd$_2$O$_6$, similar calculations can be found in Fig. 11(b,c) of Ref. [3]. Here, to increase computational efficiency and to avoid the issue of bad convergence arising from Nd-$4f$ bands, we replace Nd by La. Previous theoretical research [32] and the experimentally confirmation of superconductivity in both Ca-doped LaNiO$_2$ [33] and Sr-doped NdNiO$_2$ [34, 35] indicate that the replacement of Nd by La does not significantly change the physics. $T_c$ is reduced a bit in the La compound, which likely originates mainly from different levels of disorder. The corresponding phonon spectra of the ideal $P4/mmm$ phase of LaPdO$_2$ is shown in Fig. S.15 and the structural evolution is shown in Fig. S.16(a-c).

We first check the phonon spectra of the ideal $P4/mmm$ phase LaPdO$_2$. Using the PBESol functional, the optimized lattice parameters are $a=b=4.126$ Å, $c=3.265$ Å. Compared with La(Nd)NiO$_2$, the $a$ ($c$) lattice

![DFT band structure](image-url)
parameter is increased (reduced) by 6.1% (2.1%). The instability of $P4/mmm$ LaPdO$_2$ is evidenced by negative frequencies in its phonon spectra [Fig. S.15(a,b)]. The phonon spectra and atomic-resolved DOS are computed by employing a $2\times2\times2$ supercell of LaPdO$_2$, which is shown in Fig. S.16(a). As shown in Fig. S.15, the phonon spectrum of $P4/mmm$ LaPdO$_2$ reveals a dynamical instability at both the $X$ [$q=(1/2,0,0)$] and $A$ [$q=1/2,1/2,1/2$] points of the $q$-path. These unstable phonons are in fact consistent with those in SmNiO$_2$, YNiO$_2$ [36], LuNiO$_2$ [37, 38], and EuNiO$_2$ [38]. We further predict the structures with lower symmetry and total energies on the basis of the eigenvectors (atomic vibrations) induced by the $X^{-}_2$, $A^{-}_3$ mode and the combination between both of them, using group theory and the workflow of [37]. The eigenvector of the unstable phonon at $X$-point is $X^{-}_2$ [37] and is related to a ferroelectric distortion perpendicular to the PdO$_2$ layers. The phonon mode at the $A$-point is $A^{-}_3$ and is related to the in-plane rotation of PdO$_4$ planes, leading to a $I4/mcm$ symmetry [38] [Fig. S.16(b)] phase. Further, the combination of $X^{-}_2$ and $A^{-}_3$ corresponds to an orthorhombic distortion, i.e., the $Pbcn$ phase [Fig. S.16(c)]. After (non-spin-polarized) DFT-PBESol structural relaxations, the $I4/mcm$ and $Pbcn$ phase are energetically more stable than the ideal $P4/mmm$ phase by 102 meV and 225 meV per LaPdO$_2$ formula unit, respectively. We hence conclude that the $P4/mmm$ ($Pbcn$) structures are corresponding to high (low) temperature phases.

We additionally compute the Pd-$d_{x^2-y^2}$ hopping of the ideal $P4/mmm$, tetragonal $I4/mcm$ and the low symmetry $Pbcn$ phase of LaPdO$_2$. As shown in Table S.III, the 1st nearest hopping $t$ is reduced from -537 meV in ideal $P4/mmm$ phase to -357 meV in $I4/mcm$ phase, finally to -304 meV in $Pbcn$ phase. Due to the both, the reduced $t$ and $t'$, the DFT bandwidth is reduced is reduced from ~4.4 eV in the $P4/mmm$ phase to ~2.4 eV in the $Pbcn$ phase. The $t$ in $I4/mcm$ and $Pbcn$ phase is even smaller than that in NdNiO$_2$ (~397 meV), indicating stronger correlations than in the ideal $P4/mmm$ phase.

For the comparison between DFT and Wannier bands of distorted $Pbcn$ phase of LaPdO$_2$, see Fig. S.17. Compared with the bands of idea $P4/mmm$ phase, the bandwidth of $Pbcn$ phase of LaPdO$_2$ is effectively reduced.
FIG. S.15. (a,c) DFT phonon spectra and (b,d) phonon density of states of the (fully relaxed) ideal $P4/mmm$ (a,b) and distorted $Pbcn$ (c,d) phase of LaPdO$_2$.

FIG. S.16. DFT-relaxed structures of LaPdO$_2$: (a,d) ideal tetragonal $P4/mmm$ phase; (b,e) tetragonal $I4/mcm$ phase; (c,f) orthorhombic $Pbcn$ phase. The panels (d,e,f) are the top-view along the $z$-direction (001) of panels (a,b,c), respectively.
from 4.60 eV in $P4/mmm$ phase to 2.45 eV in $Pbn$ phase, these are consistent with the reduced major hoppings shown in Table S.III. Additionally, in both Table S.III and Fig. S.17(b,c) we demonstrate the emergence of anisotropic hoppings terms of $t'$ and $t''$ along (110) and (110), as well as along the (200) and (020) direction, respectively. Such anisotropic hopping is a consequence of the orthorhombic distortion is $Pbcn$ phase.

IX. DFT+$U$ MAGNETIC TOTAL ENERGIES OF $A'_{2}PdO_{2}Cl_{2}$

To investigate the magnetic ground state of $A'_{2}PdO_{2}Cl_{2}$ ($A$=La$_{0.5}$Ba$_{0.5}$), we perform DFT+$U$ ($U$=3 eV, 3.3 eV and 4 eV) calculations with a $\sqrt{2}\times\sqrt{2}\times1$ supercell, as shown in Fig. S.18(a). In each layer of (PdO$_2$)$_2$, there are two Pd sites, and in total two such (PdO$_2$)$_2$ layers in the supercell. We define the following three types of magnetic order: FM (inter-layer FM and intra-layer FM), A-AFM (inter-layer AFM and intra-layer FM), A-AFM (inter-layer AFM and intra-layer FM), A-AFM (inter-layer AFM and intra-layer FM).
intra-layer FM), G-AFM (inter-layer AFM and intra-layer AFM) [39]. Our DFT+U calculations confirm G-AFM to be the magnetic ground state for all $U$ values. Specifically, the G-AFM order is for the aforementioned three $U$ values, more stable than FM order by 84 meV, 103 meV and 162 meV per $A'_2$PdO$_2$Cl$_2$ chemical formula. The magnetic moments in FM state for all three considered $U$ values are $\sim 0.2 \mu_B, 0.25 \mu_B$ and $0.40 \mu_B$ per Pd, such small moments are not very effective in $\sim$ between PdO$_2$ layers along the z-direction ($\sim 3.3$ eV) and 0.63 $\mu_B$ per Pd, residual magnetic moments are located at the O-p orbital and interstitially. This demonstrates the d-p hybridization between Pd and O is stronger than that in nickelates. This puts palladates in-between cuprates and nickelates, as already indicated from the energy levels in Fig. 1 of the main text.

Table S.III. Wannier hoppings of the Pd $d_{x^2-y^2}$ orbital for the ideal $P4/mmm$, tetragonal $I4/mcm$ and orthorhombic $Pbcn$ phase of LaPdO$_2$ (in unit of meV). The $t$, $t'$ and $t''$ indicate the 1st, 2nd and 3rd nearest hopping along the real-space vectors of (100), (110) and (200), respectively. The ratios between $t'/t$ and $t''/t$ are also given. The emergence of the second $Pbcn$ values is due to the orthorhombic distortion in $Pbcn$ phase, leading to nonequivalent $t$ and $t''$ along (110) and (110) as well as between the (200) and (020) direction.

| Phase   | $t$ | $t'$ | $t''$ | $t'/t$ | $t''/t$ |
|---------|-----|------|-------|--------|---------|
| $P4/mmm$ | -537 | 97 | -71 | -0.181 | 0.131 |
| $I4/mcm$ | -357 | 77 | -15 | -0.214 | 0.043 |
| $Pbcn$   | -304 | 56 | 3  | -0.184 | 0.014 |
|          | -304 | 95 | -4  | -0.313 | 0.012 |

Fig. S.18(a) show the density-of-states (DOS) of $G$-type antiferromagnetic order in $A'_2$PdO$_2$Cl$_2$, resolved for the two inequivalent sites and spin-up and spin-down channel ($U=3.3$ eV as in our multi-orbital DMFT calculations). With such a degree of correlations a gap of $\sim 0.6$ eV is opened in the presence of $G$-AFM order. The highest occupied orbital is Pd-$d_{x^2-y^2}$, indicating a single-band AFM order that is similar to cuprates. The magnetic moment projected onto the Pd site is $0.55 \mu_B$ ($U=3$ eV), $0.57 \mu_B$ ($U=3.3$ eV) and $0.63 \mu_B$ ($U=4$ eV) per Pd, residual magnetic moments are located at the O-p orbital and interstitially. This demonstrates the $d$-$p$ hybridization between Pd and O is stronger than that in nickelates. This puts palladates in-between cuprates and nickelates, as already indicated from the energy levels in Fig. 1 of the main text.

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