Electronic Structure

Assessing the correlated electronic structure of lanthanum nickelates

Frank Lechermann

European XFEL, Holzkoppel 4, 22869 Schenefeld, Germany
Center for Computational Quantum Physics, The Flatiron Institute, 162 5th Avenue, New York, NY 10010, United States of America

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Abstract
The series of nickel-oxide compounds LaNiO$_3$ (formal Ni($d^7$)), La$_2$NiO$_4$ (formal Ni($d^8$)) and LaNiO$_2$ (formal Ni($d^9$)) is investigated by first-principles many-body theory, using a combination of density functional theory, self-interaction correction and dynamical mean-field theory. The characteristics of these different nickelates, in good agreement with available experimental data, is revealed by employing a compound-independent choice for the local Coulomb interactions. The dichotomy within the low-energy dominant Ni-$e_g$ sector of \{d$_{z^2}$, d$_{x^2−y^2}$\} kind is rising with growing Ni(3$d$) filling across the series. An intermediate-coupling scheme for spin-polarized calculations is introduced, which leads to very weak Ni ordered moments for the infinite-layer compound LaNiO$_2$ in contrast to the robust-moment system La$_2$NiO$_4$.

1. Introduction

Since the earliest days of studies on strongly correlated condensed matter [1], research on nickel-oxide compounds has ever been an important aspect. But it just recently received a further boost when superconductivity was discovered in thin films of Sr-doped infinite-layer NdNiO$_2$ [2]. Nickelates are key examples of late transition-metal oxides where the t$_{2g}$ manifold of the Ni(3$d$) shell is in most cases filled, and the $e_g$ subshell plays the major active role. While in neighboring (layered) cuprates that latter subshell physics reduces in the most interesting cases to an effective one-band/orbital picture [3], the multiorbital character usually remains intact for nickelates. Rich phase diagrams with various competing orders are the result of this scenario (see e.g. reference [4] and references therein).

Concerning the research over the last 40 years, one may roughly group the respective dominant nickelate studies compound-wise into three areas. First, from traditional work on Mott insulators [1] and the apparent structural affinity to high-$T_c$ cuprates [5], in the 1980s and early 1990s, the emphasis was on formal Ni(3$d^8$) materials, e.g. NiO [6–9] and La$_2$NiO$_4$ [10–15]. Starting from the early 1990s until later 2010s, main attention shifted towards formal Ni(3$d^7$) compounds, mainly to the rare-earth (RE) perovskite(-like) RENiO$_3$ series [16]. Though discussed early on [17], work on formal Ni(3$d^9$) systems heavily increased after the finding of superconducting infinite-layer nickelates in the mid of 2019 [2].

In this work we want to provide an theoretical overview about these different formal Ni(3$d^{m=7,8,9}$) classes, by focussing on one representative material from each class and describing them within an advanced first-principles many-body approach. Goal is to identify the key features of each class, to look for coherent theoretical settings and similarities across the classes, and to weigh the quality of the description for this wider nickelate family. To provide largest coherence from the ligand side, we choose the following series of lanthanum nickelates for the investigation: LaNiO$_3$ (formal $d^7$), La$_2$NiO$_4$ (formal $d^8$) and LaNiO$_2$ (formal $d^9$). While LaNiO$_3$ is well known as a paramagnetic (PM) metal down to lowest temperatures and La$_2$NiO$_4$ as antiferromagnetic (AFM) Mott/charge-transfer insulator over a wide temperature range, the definite electronic and magnetic characterization of LaNiO$_2$ is still a matter of intense debate. The role of an apparent self-doping band [18–20] in the latter compound complicates the correlation problem.
It will be shown that for a rather coherent setting, our theoretical modelling delivers good agreement with the key characterization and long-standing experimental results for LaNiO₃ and La₂NiO₄. For the same setting, we identify in the case of challenging LaNiO₂ an overall weak-metallic state (due to self doping) accompanied by an orbital-selective Mott-insulating Ni-\(e_g\) sector, very similar to recent findings for NdNiO₂.

2. Methodology

2.1. General framework

We utilize the DFT + sicDMFT scheme [21], a charge self-consistent combination [22–24] of density functional theory (DFT), self-interaction correction (SIC) and dynamical mean-field theory (DMFT), to assess the correlated electronic structure of LaNiO₃, La₂NiO₄ and LaNiO₂. A mixed-basis pseudopotential (MBPP) code [25–27] takes care of the DFT part in the local density approximation (LDA). To consider correlation effects beyond LDA driven by the oxygen sites in the given late transition-metal oxides, Coulomb interactions on oxygen are described within SIC, and are incorporated in the O pseudopotential [28–30]. The SIC is applied to the O(2s) and the O(2p) orbitals via weight factors \(w_p\) (see reference [30] for details). While the O(2s) orbital is by default fully corrected with \(w_p = 1.0\), the adequate choice [21, 30] \(w_p = 0.8\) is used for O(2p) orbitals. The La(4f) states are frozen in the pseudopotential core, as they supposed to be empty and appear especially irrelevant for the key physics of the superconducting nickelates [31–33]. The full Ni(3d) shell is used to construct the correlated subspace of the DMFT impurity problem via the projected-local orbitals formalism [34]. The projection is performed on the minimal set of Kohn–Sham (KS) states above the dominant O(2s) bands, associated with O(2p) and Ni(3d). For LaNiO₂, the self-doping band is additionally included in the projection sphere [20, 35, 36]. A five-orbital rotational-invariant Slater Hamiltonian, parametrized by a Hubbard \(U\) and a Hund’s exchange \(J_H\), is active in the correlated subspace. If not otherwise stated, we stick to \(U = 10\) eV and \(J_H = 1\) eV as in previous nickelate studies [20, 21, 35, 36]. One main goal of the present work is to check whether such a compound-independent choice of local Coulomb parameters may satisfyingly describe a wider range of different nickelates.

2.2. Spin-polarized calculations

The description of ordered magnetism in first-principles many-body calculations of DFT + DMFT type leaves room for methodological interpretation [37]. As a hybrid scheme, the spin polarization can be handled in different ways. Allowing for local ordered spin moments in the DMFT part to investigate possible magnetically ordered phases is definitely adequate (e.g. reference [38]). There is common agreement that explicit correlation effects in partially-filled \(d\)–or \(f\)-shells are a crucial ingredient of most spin-ordering phenomena in condensed matter. An additional treatment also in the DFT part may have pros and cons. On the positive side, a strong itinerant aspect of spin polarization, relevant for some weakly/moderately correlated compounds, might be better included. Also short-range ‘ligand-coupling’ effects could be more realistically covered by a DFT-based spin polarization of the charge density. On the negative side, the double-counting construction is not well suited for the explicit spin-polarization aspect. As a result, an often strong, static and temperature-independent exchange coupling within DFT dominates the physics, and subtle correlation effects in the magnetization are easily outshined. For instance, computation of Curie temperatures is more or less impossible with a full spin-exchange coupling within DFT dominates the physics, and subtle correlation effects in the magnetization are easily outshined. For instance, computation of Curie temperatures is more or less impossible with a full spin-exchange coupling within DFT. Since most DFT + DMFT studies aim at correlations effect at strong coupling (i.e. close to a Mott-critical regime), therefore treating spin polarization only on the DMFT and not on the DFT level, has become the standard method for describing magnetically ordered phases.

In contrast to the both restrictive routes, i.e. DFT part spin-polarized or not, we here want to suggest an alternative way of handling spin polarization in the given context of our nickelate study, which we term intermediate coupling (IC) scheme. Key idea is to neglect only the local DFT part of spin polarization, since that one should for sure be comprehensively covered by the spin-polarized DMFT self-energies. The remaining strictly itinerant DFT charge-density terms may still carry finite spin polarization.

In the MBPP approach [25], the KS wave function for Bloch vector \(\mathbf{k}\), band \(\nu\) and spin \(\sigma = \uparrow, \downarrow\) is expanded into plane waves (pw) and localized functions (HF), reading

\[
\psi_{\mathbf{k}\nu\sigma}(\mathbf{r}) = \frac{1}{\sqrt{4\pi c}} \sum_{\mathbf{G}} \psi_{\mathbf{k}\nu\sigma}^{\mathbf{G}} e^{i(\mathbf{k}+\mathbf{G})\cdot \mathbf{r}} + \sum_{\gamma \lambda \mu m} \beta_{\gamma\lambda\mu}^{\nu\sigma} \phi_{\gamma\lambda\mu}^{\nu\sigma}(\mathbf{r}),
\]  

(1)

where \(\Omega_c\) is the unit-cell volume, \(\mathbf{G}\) a reciprocal-lattice vector, \(\gamma\) labels an atom in the unit cell and \(lm\) are the usual angular-momentum quantum numbers. The localized functions are given by
\[ \phi_{\gamma_{lm}}(\mathbf{r}) = i^l g_{\gamma}(r) K_{\gamma}(\mathbf{r}), \]  
\[ \phi_{\gamma_{lm}}^k(\mathbf{r}) = \sum_{\mathbf{T}} e^{i(\mathbf{T}+\mathbf{R}_i) \cdot \phi_{\gamma_{lm}}(\mathbf{r} - \mathbf{T} - \mathbf{R}_i)}, \]

whereby \( g \) is a radial function, \( K \) a cubic-harmonic function and \( \mathbf{T} \) a lattice vector. Accordingly, the KS electronic charge density \( \rho_\sigma(\mathbf{r}) \) consists of three terms, i.e.

\[ \rho_\sigma(\mathbf{r}) = \sum_{\mathbf{k}} \int_{\mathbf{K}_{\mathbf{r}}} |\psi_{\mathbf{k}_{\gamma\sigma}}(\mathbf{r})|^2 = \rho_{\sigma}^{\text{pw, pw}}(\mathbf{r}) + \rho_{\sigma}^{\text{pw, lf}}(\mathbf{r}) + \rho_{\sigma}^{\text{lf, lf}}(\mathbf{r}). \]

For instance, the purely-local third term \( \rho_{\sigma}^{\text{lf, lf}} \) does not carry any plane-wave part and reads

\[ \rho_{\sigma}^{\text{lf, lf}}(\mathbf{r}) = \sum_{\mathbf{k}} \int_{\mathbf{K}_{\mathbf{r}}} \sum_{\gamma_{lm}} |\phi_{\gamma_{lm}}(\mathbf{r})|^2 = \sum_{\gamma_{lm}} \rho_{\gamma_{lm}}^{\text{lf, lf}}(\mathbf{r}'), \]

with \( \mathbf{r}' = \mathbf{r} - \mathbf{T} - \mathbf{R}_i \) and hence can be understood as an expansion into cubic harmonics.

In the standard DFT + DMFT calculation for magnetically ordered phases of correlated materials, all terms in equation (4) are spin-independent from a nonmagnetic KS treatment in each DFT step. Instead, we here suggest to perform a spin-polarized KS calculation in each DFT step and the following representation of the equation (4) are spin-independent from a nonmagnetic KS treatment in each DFT step. Instead, we here indeed adequate to also average the \( \rho_{\sigma}^{\text{pw, pw}}(\mathbf{r}) \) term in the construction of the correlated subspace [34]. It therefore proves indeed adequate to also average the \( \rho_{\sigma}^{\text{pw, lf}}(\mathbf{r}) \) term in equation (4), since the DMFT-relevant and therefore correlation-relevant intermixing between local and full Hilbert space is already established at a different place in the complete DFT + DMFT framework [24].

Using representation (6) on the pure DFT level results of course in a self-consistent vanishing of a possible ordered moment for correlated materials of interest. However in DFT + (sic)DMFT, the many-body part will reinject spin polarization within the IC scheme, eventually with a more subtle and balanced linkage between the weakly and strongly correlated aspects of magnetic ordering.

2.3. Crystal data and further computational settings

All structural data for the La nickelates are taken from experiment. The LaNiO\(_3\) compound crystallizes in a two-formula-unit cell with rhombohedral R\(_{3}\)c space group, and we used the data by Zhang et al [39]. The structure is very similar to, but deviates slightly from a basic perovskite structure (see top of figure 1(a)). The Ni site is in an octahedral coordination and the Ni–O bond length amounts to \( d_{\text{Ni}}^1 = 1.929 \) Å. The same coordination holds also for Ni in La\(_2\)NiO\(_4\), which is the single-layer \( p = 1 \) compound of the Ruddlesden–Popper series La\(_{p+1}\)Ni\(_p\)O\(_{3p+1}\) and basically iso-structural to the famous high-T\(_c\) cuprate La\(_2\)CuO\(_4\) as well as the famous low-T\(_c\) superconductor ruthenate Sr\(_2\)RuO\(_4\). At ambient temperature, La\(_2\)NiO\(_4\) crystallizes in the orthorhombic B\(_{2}\)\(_{2}\)\(_{2}\) structure (see mid of figure 1(a)) with a two-formula-unit cell. The in-plane Ni–O distance amounts to \( d_{\text{Ni}}^{\text{plane}} = 1.948 \) Å, while the out-of-plane distance to the apical oxygen reads \( d_{\text{Ni}}^{\text{out}} = 2.267 \) Å. Above \( T \sim 770 \) K the compound transforms to the simpler tetragonal I\(_4\)/\(_{mm}2\)m space group. Below \( T \sim 80 \) K the system transforms to a new tetragonal phase with P\(_{4}/\text{mmm}\) symmetry, however that phase is not considered in the present work. All lattice data on La\(_2\)NiO\(_4\) is taken from the work by Rodríguez-Carvajal et al [40]. Finally, the LaNiO\(_3\) compound with P\(_4\)/\(_{mmm}\) space group cannot be grown in single-crystal form, but exists as polycrystals [41] and in thin films on SrTiO\(_3\) [32, 33]. This infinite-layer structure consists of NiO\(_2\) layers with \( d_{\text{Ni}}^2 = 1.980 \) Å and lacking apical oxygen (see bottom of figure 1(a)). We use the lattice data for the single-phase model from Hayward et al [41]. The general increase of \( d_{\text{Ni}}^2 \) from LaNiO\(_3\) to La\(_2\)NiO\(_4\) and LaNiO\(_2\) may be associated with the expected lowering of the Ni oxidation state.

For the DFT + (sic)DMFT calculations we utilize a plane-wave cutoff \( E_{\text{cut}} = 16 \) Ry and a \( 13 \times 13 \times 13 \) k-point mesh for LaNiO\(_3\). The respective k-point mesh for the other structure is accordingly tailored to yield the identical k-point density. Localized functions in the MBPP formalism are introduced for La(5\(d\)), Ni(3\(d\)) as
well as O(2s, 2p). Continuous-time quantum Monte Carlo in hybridization expansion [42] as implemented in the TRIQS code [43, 44] is utilized to solve the quantum-impurity problem. The fully-localized-limit double-counting scheme [45] is applied. Maximum-entropy [46] and Padé [47] methods are employed for the analytical continuation from Matsubara space onto the real-frequency axis. PM calculations are performed for LaNiO3. In the case of La2NiO4 and LaNiO2, both, PM and AFM phases are investigated, for details see the corresponding sections 4 and 5.

3. General electronic structure aspects

Before discussing individual aspects of each of the studied nickelate compounds, let us start with an overview of the correlated structure when lowering the Ni ‘oxidation potential’ from LaNiO3 via La2NiO4 to LaNiO2. Figure 1(b) displays from top to bottom the total and site-orbital-projected spectral function $A(\omega)$ for the three oxides. Note again that the local Coulomb parameters $U = 10$ eV and $J_H = 1$ eV are kept identical for each compound.

The slightly-distorted perovskite LaNiO3 is the metallic end member of the RENiO3 series. Formally, nickel is in the 3+ oxidation state, i.e. a local $3d^7(t^6_2g^1e^1)$ configuration would hold. Yet from the present calculations, Ni is closer to a $3d^8(t^6_2g^2e^0)$ configuration with a ligand hole on oxygen, i.e. a $3d^8L^0$ state. In detail, according to the projected spectral function, the average O(2p) filling per oxygen site $n_p = 5.58$ translates into about 1.2 holes per unit cell. A significant oxygen hole density can also easily be observed from substantial O(2p) spectral weight above the Fermi level $\epsilon_F$. This finding is in line with previous works [48–54] that emphasize the preference for oxygen–hole formation in LaNiO3 and presumably in further formal-3d nickelates. Under these circumstances, the (effective) charge-transfer energy $\Delta$ should be small or even negative [4]. We here define $\Delta = \epsilon_d - \epsilon_p$ as obtained from DFT + sic, i.e. nonmagnetic DFT calculations with SIC on oxygen, and $\epsilon_d$, $\epsilon_p$ as the respective centers of the associated Ni(3d) and O(2p) projected-local-orbital bands. The resulting $\Delta_{LaNiO3} = 2.2$ eV is indeed rather small.

The Ni(3d) filling $n_d$ raises in La2NiO4 and the number of oxygen holes is comparatively small. The Ni2+ state with $3d^8$ configuration is indeed closest to reality. The system is an AFM insulator with a charge gap $\Delta_q$ of about 4 eV, in line with experiment [55]. However note that compared to prototypical NiO [21], the valence-band-maximum states are of stronger Ni(3d) character. The charge-transfer energy is computed as

![Figure 1](image-url). Structural and spectral overview for the nickelate compounds LaNiO3, La2NiO4 and LaNiO2. (a) Top to bottom: ambient-temperature crystal structure of the compounds. LaNiO3: rhombohedral $R\bar{3}c$; La2NiO4 orthorhombic $Bmab$; LaNiO2: tetragonal $P4/mmm$. La (green), Ni (grey) and oxygen (red). (b) Top to bottom: DFT + sicDMFT total and projected spectral function for PM-LaNiO3, AFM-La2NiO4 and PM-LaNiO2 at $T = 193$ K, respectively. The values for $n_p$ and $n_d$ refer to the O(2p) and Ni(3d) filling as obtained from integrating $A_{proj}(\omega)$. The respective charge-transfer energy $\Delta$ is computed within DFT + sic (see text).
\[ \Delta_{\text{LaNiO}_3} = 3.9 \text{ eV, significantly larger than for LaNiO}_3 \text{ and also in good accordance with experimental estimates \[15\]. With } \Delta \ll U \text{ and } \Delta \sim \Delta_c \text{ the system may be classified as a charge-transfer insulator.} \]

The LaNiO\(_3\) compound with \(n_{\text{e}}\) closer to 3\(d^\uparrow\) has apparently a more subtle electronic structure than given by an obvious metal or insulator. The Ni(3\(d\)) states are nearly gapped, but minor spectral weight remains at the Fermi level. The oxygen–hole content is non-negligible but not as strong as for LaNiO\(_2\); there are only about 0.5 holes per unit cell. However note that the here provided fillings derived from the projected spectral function may differ somewhat from values extracted from the local spectral function. The values \(n_{\text{loc}}\) associated with the latter function read \{7.95, 8.15, 8.84\} for LaNiO\(_3\), La\(_2\)NiO\(_4\) and LaNiO\(_2\). The infinite-layer compound shows the largest charge-transfer energy \(\Delta_{\text{LaNiO}_2} = 4.8\) eV among the here studied nickelates. In line with the increase of \(\Delta\) with a shrinking formal oxidation state, the O(2\(p\)) levels shift to deeper energies with rising Ni(3\(d\)) filling.

In the following three subsections, we will discuss the specific features of the individual compounds in more detail.

### 4. LaNiO\(_3\)

Down to low temperature, the LaNiO\(_3\) compound remains in a strongly correlated metallic state \[16\], with an experimental mass renormalization \(m'/m\) of about 3–4 \[56, 57\]. There have already been various DFT + DMFT accounts for this nickelate, however most studies focus on a two-orbital Ni-\(e_g\)-correlated subspace, and only few treat the full five-orbital Ni(3\(d\)) shell as correlated within DMFT \[58, 59\].

Based on our calculations utilizing the five-orbital correlated subspace, figure 2(b) displays the \(k\)-resolved spectral function along high-symmetry lines in the Brillouin zone of the R\(3\)c structure (see figure 2(a)). The Ni-\(e_g\) orbitals are degenerate and two corresponding bands cross the Fermi level, showing a DFT bandwidth of ~ 2.5 eV. The Ni-\(t_{2g}\) orbitals are mostly filled and the associated bands remain below \(\varepsilon_F\) \[58\]. Compared to the DFT dispersion, there is a significant band renormalization for the low-energy Ni-\(e_g\) dominated bands. Figure 2(c) documents the \(U\) dependence of the local Ni-\(e_g\) self-energy part \(\Im \Sigma(\iota\omega_n)\) for small Matsubara frequencies \(\omega_n\), with the extracted \(m'/m_{\text{DA}} = (1 - \partial \Im \Sigma / \partial \omega_n|_{\omega_n=0})^{-1}\) in this orbital basis. It is seen that the effective mass is indeed closest to the experimental regime for our general choice \(U = 10\) eV, while it remains too small for a lower on-site Coulomb interaction. Note that the actual effective mass in the band basis, which strictly speaking should be the one for comparison with experiment, is lower and amounts to \(m'/m_{\text{LDAband}} \sim 2.5\). Thus, even a rather large \(U\) value does not quite lead to a perfect-matching magnitude for the experimental Ni-\(e_g\) band renormalization. For instance, non-local spin fluctuations could be a further source for additional renormalization effects.

The explicit local Ni-\(e_g\) occupation amounts exactly to two electrons, i.e. both the Ni-\(d_{\uparrow}2\) and Ni-\(d_{\uparrow}5\) orbital each host one electron. This becomes also clear from the local spectral function shown in the bottom panel of figure 2, where the half-filled \(e_g\) states give rise to a renormalized quasiparticle peak (A) at the Fermi level. The total \(A(\omega)\) exhibits further peaks at \(-0.6\) eV (B), \(-1.5\) eV (C) and \(-4.2\) eV (D) in the occupied part of the spectrum. Peak B is associated with Ni-\(t_{2g}\), C with O(2\(p\)) and \(D\) with O(2\(p\))/Ni(3\(d\))-Hubbard-band. The prominent two-peak O(2\(p\)) structure at higher energies below the dominant Ni(3\(d\)) part, supposedly due to the intricate 3\(d^\uparrow3\)L ground state, can already be drawn from figure 1. Though there is overall agreement with photoemission data \[16\], there are slight differences for the peak positions; in experiment, especially peak C is located at deeper ~ 2.4 eV. Lowering the \(U\) value shifts C to somewhat to higher negative energies, but still not reaching the experimental location. But as discussed before, the quasiparticle \(\varepsilon_F\) renormalization becomes too small for smaller \(U\). Notably, the plain DFT spectrum places the main part of peak C in better accordance with experiment. In summary, DFT + sicDMFT provides a good description of LaNiO\(_3\) for the chosen set of Coulomb parameters, however, some questions remain concerning correlation strength and the general electronic spectrum. Albeit originally-thought a rather unspectacular compound, a very good description/understanding of the LaNiO\(_3\) electronic structure turns out still challenging \[60\].

### 5. La\(_2\)NiO\(_4\)

As the natural nickelate analog to the canonical high-\(T_c\) cuprate La\(_2\)CuO\(_4\), experimentally, the La\(_2\)NiO\(_4\) compound has been studied extensively in the past (see e.g. \[4\] and references therein for the phenomenology with hole doping). However to our surprise, while there are standard-DFT \[13, 61\], DFT + U \[62\] and meta generalized-gradient approximation \[63\] studies, we could not find an in-depth realistic-DMFT investigation for this material in the literature. This section is hence also devoted to fill this void.

From an experimental point of view, ensuring the exact stoichiometry for the undoped compound, especially in view of the oxygen content \[64, 65\], appears the most critical issue in defining the correct electronic
phases. Here, we do not delve into the intriguing question of defect behavior at nominal stoichiometry, but discuss the perfectly-ordered system.

The orthorhombic compound at ambient and lower temperature is described metallic in nonmagnetic DFT (see figure 3(a)). By imposing strong electronic correlations and additional AFM order, La$_2$NiO$_4$ is transferred into a Mott-insulating state with a sizable charge gap of $\sim 4$ eV. That latter value and the associated electronic spectrum (full line in top panel of figure 3(a)) are in very good agreement with data from photoelectron spectroscopy [55]. There is a prominent and broad peak centered at $-4$ eV, mainly constituted by O(2p) (cf mid panel of figure 1(b)). The upper Hubbard-band edges of Ni-$e_g$ character are responsible for a shoulder at $-2$ eV. While the Ni-$t_{2g}$ orbitals are once more completely filled, Ni-$d_{x^2-y^2}$ and Ni-$d_{z^2}$ both host one electron, i.e. $\{n_{d^1}, n_{d^{1-\delta}}\} = \{1.07, 1.09\}$, respectively. The ordered Ni moment in the standard G-type AFM phase turns out $m_{Ni} = 1.65 \mu_B$ at $T = 193$ K from our IC-scheme calculations, with an equal share of spin polarization between both Ni-$e_g$ orbitals. Thus the Ni moment deviates significantly from the value associated with a saturated $S = 1$ spin, but is again in very good agreement with experimental values [40, 66]. Note that from neutron diffraction the Néel temperature is located at $T_N = 330$ K [40]. It is however important to realize that the magnetic order is not decisive for establishing the insulating state. In comparison, the calculated ordered Ni moment turns out too small in the DMFT-only spin-polarization scheme and too large in fully spin-polarized DFT + DMFT. For the same system temperature of $T = 193$ K, a value $m_{Ni} = 0.70 \mu_B$ results for the former case and $m_{Ni} = 1.82 \mu_B$ is obtained for the latter one. The rather small $m_{Ni}$ for the DMFT-only viewpoint may be linked to the strong hybridization between Ni-$e_g$ and O(2p), which renders an inclusion of ligand spin polarization necessary.

Figure 2. Data for LaNiO$_3$. (a) Brillouin zone with high-symmetry lines for the $R3c$ structure. (b)–(d) DFT + sicDMFT results for $T = 193$ K: (b) $k$-resolved spectral function with comparison to DFT dispersion; (c) imaginary part of the Ni-$e_g$ self-energy for different $U$ values, yielding varying effective masses; (d) $k$-integrated total spectrum (top) and local Ni(3$d$) spectrum (bottom).
The high-temperature phase of La$_2$NiO$_4$ has been a matter of debate in the past. Transport studies [67] motivated a (bad-)metallic scenario for the tetragonal phase of La$_2$NiO$_4$, i.e. some kind of metal-to-insulator transition with rising temperature. Goodenough et al [10, 68] suggested an orbital-selective Mott transition upon heating, whereby the Ni-$d_{z^2}$ states remain insulating but the Ni-$d_{x^2−y^2}$ states become metallic. From a structural point of view, the absence of the further extension and rotation of the NiO$_6$ octahedron known within the $Bmab$ phase, leads to a weaker splitting between both $e_g$ states [61]. The DFT crystal-field splitting $\Delta_{cf} = \varepsilon_{z^2} − \varepsilon_{x^2−y^2}$ amounts to 66 meV in the orthorhombic phase and to 38 meV in the tetragonal phase. Additionally, while the total Ni-$e_g$ bandwidth in the tetragonal phase amounts to $\sim 2.8$ eV, the individual orbital contributions differ (cf figure 3(b)). The $d_{x^2−y^2}$ dominated DFT band crossing the Fermi level along $\Gamma$–X is indeed significantly wider than the $d_{z^2}$ dominated band crossing along $X$–$M$. However, the orbital $e_g$ mixing within these bands is still about 2:1 (and vice versa, respectively), due to the reduced energy splitting. As a further interesting aspect, the DFT band structure of tetragonal La$_2$NiO$_4$ shows self-doping behavior through a largely La($5d$) dominated band creating a small electron pocket around the $Z$ point. This highlights the stronger three-dimensional character compared to e.g. La$_2$CuO$_4$.

The plotted spectral parts and self-energies in figure 3 render obvious that within DFT + sicDMFT there is no orbital-selective physics in the tetragonal phase at higher temperature. While indeed the Ni-$d_{x^2−y^2}$ orbital turns out somewhat weaker correlated than the Ni-$d_{z^2}$ one, the gap opening takes place in both orbital sectors also for $U < 10$ eV. Still, the $U = 10$ eV regime appears again best fitting to La$_2$NiO$_4$, as the charge gap results too small for reduced $U$ values. Thus the possible (bad-)metallic behavior at higher temperatures in this compound should be associated with the intriguing defect characteristics at stoichiometry [64, 65].

6. LaNiO$_2$

The infinite-layer LaNiO$_2$ belongs to the growing family of superconducting nickelates upon thin-film growth and Sr doping [2, 32, 33]. This finding has stimulated a vast theory interest in these type of compounds...
(see e.g. [18–20, 35, 36, 69–89] among others), with notable earlier work [90, 91]. For a detailed discussion within the DFT + sicDMFT approach for the case of NdNiO₂, we refer to our previous works [20, 35, 36].

Non-surprisingly, the essential physics of LaNiO₂ far away from the low-temperature regime turns out very similar to NdNiO₂ on the present level of theory. Figures 4(a)–(c) display the key features of the PM electronic spectrum. The DFT density of states describes a good metallic system, yet strong electronic correlations transfer most of the low-energy spectral weight to higher energies. Note that the DFT + sicDMFT predicted spectrum agrees well with data from recent photoemission spectroscopy performed on thin films of PrNiO₃ [92], where the two main peaks at $-2$ eV and $-5.5$ eV are well confirmed by experiment. The Ni$^\dagger$(3$d^P$) configuration leads to an intriguing situation where the Ni-$d_{x^2−y^2}$ orbital is close to half filling and residual hole character in the Ni-$d_z$ orbital remains (cf figure 4(a), lower panel). That latter character is achieved via a self-doping band of dominant La(5$d$) nature, causing electron pockets around the $\Gamma$ and A point in reciprocal space, which still carry minor Ni-$d_z$ hybridization weight. Contrary to the self-doping band in tetragonal La₅Ni₄O₁₃, the electron pockets at the Fermi level survive the impact of strong electronic correlations. Reason is that those pockets do not carry Ni-$d_{x^2−y^2}$ weight and are therefore not shifted to higher energies. Thus the effective Ni-$d_{x^2−y^2}$ band is most-proximate to a (orbital-selective) Mott transition and the largely-filled and therefore weakly-to-moderately correlated Ni-$d_z$ orbital remains in a joint metallic state with the La(5$d$) orbitals (see figure 4(b)). For illustration, we also show in figure 4(c) the imaginary diagonal part of the Ni-$e_g$ hybridization function $\Delta_{hyb}$ in the DFT limit [89] as well as in the interacting regime with $\Delta_{hyb}(\omega) = \omega + \mu - G_{loc}^{-1}(\omega) - \Sigma_{imp}(\omega)$. In the DFT limit, both Ni-$e_g$ have sizable weight at low-energy, whereas the hybridization function for the Ni-$d_{x^2−y^2}$ vanishes in DFT + sicDMFT close to the Fermi level because of its Mott-insulating nature. The hybridization structures with O(2$p$) are dominant at higher energy around $-6$ eV especially for Ni-$d_{x^2−y^2}$. Note the energy shift towards deeper energies due to the SIC inclusion compared to standard DFT. Comparing LaNiO₂ and NdNiO₂ does not reveal striking differences in $\Delta_{hyb}$. But it is revealed that the frequency-integrated hybridization (dashed lines in lower panel of figure 4(c)), is somewhat stronger for the Nd compound. This could affect the low-temperature physics in some regard, and different features seem indeed revealed experimentally [32] (see also text below).

Loosely speaking, infinite-layer nickelate with the missing apical oxygen can be viewed as a coupling of two dichotomic subsystems: strongly hybridized/correlated Ni-$d_{x^2−y^2}$–O(2$p$) and weakly-correlated Ni-$d_z$–RE(5$d$). Hole doping leads to a further rise of complexity in the entanglement of these two subsystems, namely a change from (orbital-selective) Mottness to Hundness via the enclosed superconducting region [20, 35, 36]. But we will not pursue this physics in the present context.

Instead, we here want to briefly comment on the issue of ordered magnetism, since the spin physics of RENiO₂ compounds is highly puzzling in experiment. Absence of long-range order [41, 93], quasi-static ordering [94], spin-glass physics [95], pseudogap vs Curie–Weiss [96] and (para)magnon dispersions [97] are discussed. Various theoretical accounts [84, 85, 98] find a nearest-neighbor superexchange coupling $J \sim 60–70$ meV, i.e. about half the size of the value in high-Τc cuprates, and AFM order should thus be favorable. However one must also remember that all available calculations find a metallic state for RENiO₂ at stoichiometry, either due to a robust Ni-$d_{x^2−y^2}$ quasiparticle weight and/or, as here, because of the existent self-doping band. This is in stark contrast to layered cuprates. But metallicity, strong electronic correlations and AFM order do not go along very well in nature, reason why there are only few reported materials cases (see e.g. discussion in [99]). Furthermore, the stronger three-dimensional character of RENiO₂ compared to layered cuprates does not help either to stabilize AFM order under these circumstances. Therefore given the electronic structure of the infinite-layer nickelates, the absence of robust AFM order in experiment is in fact not totally surprising. To underline this viewpoint, we show in figure 4(d) the diagonal part of the LaNiO₂ static weak-coupling spin susceptibility $\chi(q,\omega) = -\frac{1}{\pi} \sum_{k,\alpha\beta} G_{\alpha}(k,\omega)G_{\beta}(k+q,\omega)$, where $\omega$ are fermionic Matsubara frequencies and $G_{\alpha}$ denotes the converged DFT + sicDMFT Green’s function in Bloch space for band indices $\nu\nu'$. This simplified Lindhard-like susceptibility neglects vertex contributions and makes only sense in a system with existing low-energy spectral weight. Because of the self-doping electron pockets around $\Gamma$ and A (plus some very weak leftovers from the effectively Mott-insulating Ni-$d_{x^2−y^2}$ band), $\chi_\nu$ is nonzero and obviously has highest intensity around $\Gamma$. Note that the absolute values are still low and far off any instability regime. Still, the maximum close to the zone center puts the system in favor of ferromagnetic ordering tendencies from this perspective. These tendencies oppose the strong-coupling AFM tendencies from the superexchange perspective. Again to be clear, we do not at all claim that the given $\chi_\nu$ describes the full range of spin fluctuations in LaNiO₂ adequately, it only shines a light on the implications coming from the metallic side of the problem. Let us also mention that additionally, a unique Kondo scenario involving the Ni-$d_{x^2−y^2}$ spin and the self-doping band has been detected below $T \sim 60$ K for the case of NdNiO₂ [35]. Note that again the Ni-$d_z$ orbital, hybridized onto the self-doping band, plays a decisive role in arranging for link between Ni-$d_{x^2−y^2}$ spin and itinerant electrons. It was shown [36], that this Kondo coupling may interfere/coexist with AFM ordering tendencies at low temperatures.
Equipped with our IC scheme to handle finite spin polarization in the interacting regime, the DFT + sicDMFT calculations for most promising C-type AFM order [35, 82, 84], i.e. AFM in-plane and FM out-of-plane, results in ordered Ni moments of size $m_{\text{Ni}} \lesssim 0.05 \mu_B$. This result holds for $T = 190$ K as well as for the lower-temperature regime of $T = 30$ K. Thus in qualitative agreement with our previous work on NdNiO$_2$, the tendency to establish sizable-moment antiferromagnetism in RENiO$_2$ is weak, in agreement with experimental findings. Note that we employed identical initialization protocols with starting moments to eventually stabilize magnetic order, and while the $d^9$ compound La$_2$NiO$_4$ develops substantial Ni moments (see previous section), the $d^8$ compound LaNiO$_2$ does not. However, the residual magnetic order apparently helps to establish a stable electronic LaNiO$_2$ phase at lower temperature $T = 30$ K. In contrast to NdNiO$_2$ [35, 36], the Ni-$d_{z^2}$ orbital in the PM phase seemingly becomes in some way critical at lower $T$ as the associated self-energy develops unphysical features at small Matsubara frequencies. This might be a numerical artifact, but we still want to report it here, since several careful attempts to cure these features were unsuccessful. One then surely enters highly speculative territory, but the basic message could be that the Kondo-decorated PM phase (and its underlying unconventional Ni-$e_g$–RE(5$d$) coupling structure) detected for NdNiO$_2$, is in some way ‘disturbed’ in LaNiO$_2$ at comparable temperatures.

7. Conclusions

In this work we presented a comparison of the correlated electronic structure of three different 3$d^9$ nickelates, i.e., formal-$d^7$ LaNiO$_3$, $d^8$ La$_2$NiO$_4$ and $d^9$ LaNiO$_2$, based on calculations within the DFT + sicDMFT scheme. One goal was to evaluate the performance of the latter scheme for the wider class of nickel-oxide compounds. Moreover, we chose to fix the local Coulomb parameters $U$ and $J_{\text{H}}$ to examine if key features of the different compounds can still be well described without fine tuning (or direct calculation) of the interaction integrals. Concerning both aspects, positive results are reported here. The DFT + sicDMFT approach is very capable of...
describing the correlated-metal nature of LaNiO₃ with its hole on oxygen as well as the Mott-insulating characteristics of La₂NiO₄. And importantly, the constant-interaction strength approach shows that while a detailed knowledge/calculation of the Hubbard parameters is surely a relevant goal, main electronic-structure features are well describable without strong changes of the interaction strength from one compound to another. For the case of LaNiO₂, comparison to experiment remains still difficult due to the scarce availability of measured data. But the present results for the other two compounds and recent photoemission data [92] provide some confidence that the here (and elsewhere [20, 35, 36]) established picture for infinite-layer nickelates will also prove reliable.

In the course of this materials study, a refined scheme to perform spin-polarized DFT + (sic)DMFT studies was introduced. This intermediate-coupling scheme spin averages the DFT charge density terms that connect to the local part of the problem, but still allows for finite spin polarization in the plane-wave-derived charge density within DFT. Hence most of the spin polarization should be generated in the DMFT part, yet a minor feedback into the DFT part is restored. This new scheme provides ordered Ni moments for La₂NiO₄ in excellent agreement with experiment. It furthermore confirms our recent results [36] of a (nearly) vanishing AFM-ordered Ni moment in infinite-layer nickelates, also in line with experiment. The intriguing metallic(-like) nature at stoichiometry may be blamed for prohibiting a strongly correlated AFM state.

Finally, let us reiterate on the fact that nickelates, in general, are manifest Ni-\(e_g\) systems. While for LaNiO₃, the \(d_{z^2}\) and \(d_{x^2-y^2}\) orbital act quite coherently, some dichotomy sets in for La₂NiO₄. Crystal-field splitting and different bandwidths cause orbital differentiation for this layered \(d^9\) compound, but in the end both orbital sectors give rise to similar qualitative behavior. Then for LaNiO₂ this dichotomy is forced to an utmost limit, where half-filled \(d_{z^2}\) is (nearly) Mott-insulating and largely-filled \(d_{z^2}\) remains metallic. It is interesting to note that, as shown, orbital selectivity and self-doping character are already a matter of debate for La₂NiO₄, namely in its high-temperature tetragonal phase. However, these issues apparently only become severe as a ‘game changer’ for the \(d^9\) LaNiO₂. In this context it may also be worth to state that the mechanism of getting rid of the DFT self-doping band via rotation/distortion in the orthorhombic phase of La₂NiO₄, has just been shown to may also be a proper mechanism for later RENiO₂ systems [100, 101]. It still has to be explored if the even more extreme dichotomic limit of being able to abandon one of the Ni-\(e_g\) orbitals completely from a low-energy discussion can eventually be realized. In any case, the plethora of puzzling and demanding physics that emerges from the Ni-\(e_g\) manifold in nickel oxides will remain an exciting research area in condensed matter physics.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

ORCID iDs

Frank Lechermann https://orcid.org/0000-0002-3756-9722

References

[1] Mott N F and Peierls R 1937 Proc. Phys. Soc. 49 72
[2] Li D, Lee K, Wang B Y, Osada M, Crossley S, Lee H R, Cui Y, Hikita Y and Hwang H Y 2019 Nature 572 624
[3] Zhang F C and Rice T M 1988 Phys. Rev. B 37 3759
[4] Imada M, Fujimori A and Tokura Y 1998 Rev. Mod. Phys. 70 1039
[5] Bednorz J G and Müller K A 1986 Z. Phys. B 64 189
[6] Sawatzky G A and Allen J W 1984 Phys. Rev. Lett. 53 2339
[7] Hüfner S, Osterwalder J, Riesterer T and Hulliger F 1984 Solid State Commun. 52 793
[8] van der Laan G, Zaanan J, Sawatzky G A, Karnatak R and Esteva J-M 1986 Phys. Rev. B 33 4253
[9] van Elp J, Eskes H, Kuiper P and Sawatzky G A 1992 Phys. Rev. B 45 1612
[10] Goodenough J B and Ramasesha S 1982 Mater. Res. Bull. 17 383
[11] Sayer M and Odier P 1987 J. Solid State Chem. 67 26
[12] Aeppi G and Buttrey D J 1988 Phys. Rev. Lett. 61 203
[13] Guo G Y and Temmerman W M 1988 J. Phys. C. 21 L917
[14] Batile X et al 1989 Physica C 162–164 1273
[15] Kuiper P, van Elp J, Sawatzky G A, Fujiwara A, Bosoya S and de Leeuw D M 1991 Phys. Rev. B 44 4570–5
[16] Horiike K, Eguchi R, Taguchi M, Chianani A, Kikkawa A, Senba Y, Ohashi H and Shin S 2007 Phys. Rev. B 76 155104
[17] Crespin M, Levita P and Gaitzne L 1983 J. Chem. Soc. Faraday Trans. II 79 1181–94
[18] Takeuchi N, Hon K M, Chan C T, Braun P and Fahnle M 1990 J. Phys.: Condens. Matter. 2 4371
[19] Lechermann F, Welsch F, Elsasser C, Ederer C, Fahnle M, Sanchez J M and Meyer B 2002 Phys. Rev. B 65 132104
[20] Meyer B, Elsasser C, Lechermann F and Fahnle M 1998 FORTRAN 90 Program for Mixed-Basis-Pseudopotential Calculations for Crystals (Stuttgart: Max-Planck-Institut für Metallforschung)
[21] Vogel D, Kräger P and Pollmann J 1996 Phys. Rev. B 54 5495–511
[22] Filippetti A and Spaldin N A 2003 Phys. Rev. B 67 125109
[23] Körner W and Elsasser C 2010 Phys. Rev. B 81 085324
[24] Zhang G M, Yang Y F and Zhang F C 2020 Phys. Rev. B 101 020501
[25] Osada M, Wang B Y, Goodge B H, Harvey S P, Lee K, Li D, Kourkoutis L F and Hwang H Y 2021 arXiv:2105.13494
[26] Amadon B, Lechermann F, Georges A, Jollet F, Wehling T O and Lichtenstein A I 2008 Phys. Rev. B 77 205112
[27] Lechermann F 2020 Phys. Rev. X 10 041002
[28] Lechermann F 2021 Phys. Rev. Mater. 5 044803
[29] Keshavarz S, Schött J, Millis A J and Kvaushnin Y O 2018 Phys. Rev. B 97 184404
[30] Körner W and Elsasser C 2010 Phys. Rev. B 81 085324
[31] Zhang G M, Yang Y F and Zhang F C 2020 Phys. Rev. B 101 020501
[32] Osada M, Wang B Y, Goodge B H, Harvey S P, Lee K, Li D, Kourkoutis L F and Hwang H Y 2021 arXiv:2105.13494
[33] Zeng S W et al 2021 arXiv:2105.13492
[34] Amadon B, Lechermann F, Georges A, Jollet F, Wehling T O and Lichtenstein A I 2008 Phys. Rev. B 77 205112
[35] Lechermann F 2020 Phys. Rev. X 10 041002
[36] Lechermann F 2021 Phys. Rev. Mater. 5 044803
[37] Keshavarz S, Schött J, Millis A J and Kvaushnin Y O 2018 Phys. Rev. B 97 184404
[38] Flesch A, Gorelov E, Koch E and Pavarin E 2013 Phys. Rev. B 87 195141
[39] Zhang J, Zheng H, Ren Y and Mitchell J F 2017 Cryst. Growth Des. 17 2730
[40] Rodrigo-Carvajal J, Fernandez-Diaz M T and Martinez J L 1991 J. Phys.: Condens. Matter. 3 3215
[41] Hayward M A, Green M A, Rosseinsky M J and Sloan J 1999 J. Am. Chem. Soc. 121 8843
[42] Werner P, Comanac A, de Medici L, Troyer M and Millis A J 2006 Phys. Rev. Lett. 97 076405
[43] Parcollet O, Ferrero M, Ayrat T, Hafemeister H, Krivenko I, Messio L and Seth P 2015 Comput. Phys. Commun. 196 398
[44] Seth P, Krivenko I, Ferrero M and Parcollet O 2016 Comput. Phys. Commun. 196 398
[45] Anisimov V I, Solovyev I V, Korotin M A, Czyzyk M T and Sawatzky G A 1993 Phys. Rev. B 48 16929
[46] Jarrell M and Gubernatis J E 1996 Phys. Rev. B 54 16929
[47] Vidberg H J and Serene J W 1977 J. Low Temp. Phys. 29 179
[48] Demourgues A, Weiß F, Darriet B, Wattiaux A, Grenier J C, Gravereau P and Pouchard M 1993 J. Solid State Chem. 106 330
[49] Mizokawa T, Khomskii D I and Sawatzky G A 2000 Phys. Rev. B 61 11263–6
[50] Park H, Millis A J and Marianetti C A 2012 Phys. Rev. Lett. 109 136402
[51] Lau B and Millis A J 2013 Phys. Rev. Lett. 110 126401
[52] Johnston S, Mukherjee A, Eltsov I, Bercić M and Sawatzky G A 2014 Phys. Rev. Lett. 112 106404
[53] Subedi A, Peil O E and Georges A 2015 Phys. Rev. B 91 075128
[54] Bisogni V et al 2016 Nat. Commun. 7 13017
[55] Eisaki H et al 1992 Phys. Rev. B 45 12513–21
[56] Ouellette D G, Lee S, Kim J, Stemmer S, Balents L, Millis A J and Allen J S 2010 Phys. Rev. B 82 165112
[57] Nowadnick E A, Ruf J P, Park H, Koh C D C, Schlom D G, Shen K M and Millis A J 2015 Phys. Rev. B 92 245109
[58] Deng X, Ferrero M, Mavrou J, Aichhorn M and Georges A 2012 Phys. Rev. B 85 125137
[59] Liao X, Singh V and Park H 2021 Phys. Rev. B 103 085110
[60] Karner V J et al 2019 Phys. Rev. B 100 165109
[61] Zhou N, Chen G, Zhang H J and Zhou C 2009 Physica B 404 4150–4
[62] Pardo V, Botana A S and Baldomir D 2012 Phys. Rev. B 86 165114
[63] Lane C and Zhu J-X 2020 Phys. Rev. B 101 155135
[64] Jorgensen J D, Dabrowski B, Pei S, Richards D R and Hinks D G 1989 Phys. Rev. B 40 2187–99
[65] Bassat J M, Odier P and Loup J P 1994 J. Solid State Chem. 110 124–35
[66] Lander G H, Brown P J, Spalek J and Honig J M 1989 Phys. Rev. B 40 4463–71
[67] Ganguly P and Rao C N R 1973 Mater. Res. Bull. 8 405
[68] Fontcuberta J and Goodenough J B 1985 J. Solid State Chem. 56 116–21
[69] Hu Y, Liu G, Hao W, Liu Y and Liu Z 2019 J. Phys.: Cond. Mat. 31 083204
[70] Jiang M, Berciu M and Sawatzky G A 2020 Phys. Rev. Lett. 124 207004
[71] Jiang P, Si L, Liao Z and Zhong Z 2019 Phys. Rev. B 100 201106
[72] Werner P and Hoshino S 2020 Phys. Rev. B 101 041104
[73] Li D, Wang B Y, Lee K, Harvey S P, Osada M, Goodge B H, Kourkoutis L F and Hwang H Y 2020 Phys. Rev. Lett. 125 027001
[74] Gu Y, Zhu S, Wang X, Hu J and Chen H 2020 Comput. Phys. 3 84
[75] Leonard J, Skornykov S L and Savrasov S Y 2020 Phys. Rev. B 101 241108
[76] Wang Y, Kang C-J, Miao H and Kotляр G 2020 Phys. Rev. B 102 161118
[77] Hepting M et al 2020 Nat. Mater. 19 381
[78] Wang Y and Vishwanath A 2020 Phys. Rev. Res. 2 023112
[79] Botana A S and Norman M R 2020 Phys. Rev. X 10 011024
[80] Si L, Xiao W, Kaufmann J, Tomczak J M, Lu Y, Zhong Z and Held K 2020 Phys. Rev. Lett. 124 166402
[81] Olevano V, Bernardini F, Blase X and Cano A 2020 Phys. Rev. B 101 161102
[82] Choi M Y, Pickett W E and Lee K W 2020 Phys. Rev. Res. 2 035445
[83] Karp J, Botana A S, Norman M R, Park H, Zingl M and Millis A 2020 Phys. Rev. X 10 021061
[84] Zhang R, Lane C, Singh B, Nakelainen J, Barbiellini B, Markiewicz R S, Bansil A and Sun J 2021 Commun. Phys. 4 118
[85] Katukuri V M, Bogdanov N A, Weser O, van den Brink J and Alavi A 2020 Phys. Rev. B 102 241112
[86] Been E, Lee W S, Hwang H Y, Cui Y, Zaanan J, Devereaux T, Moritz B and Jia C 2021 Phys. Rev. X 11 011050
[87] Plienbumbrew T, Dughofer M and Olesi A M 2021 Phys. Rev. B 103 104515
[88] Choubey P and Eremin I M 2021 Phys. Rev. B 104 144504
[89] Karp J, Hampel A and Millis A J 2021 Phys. Rev. B 103 195101
[90] Anisimov V I, Bukhvalov D and Rice T M 1999 Phys. Rev. B 59 7901–6
[91] Lee K-W and Pickett W E 2004 Phys. Rev. B 70 165109
[92] Chen Z et al 2021 arXiv:2106.03963
[93] Wang B X et al 2020 Phys. Rev. Mater. 4 084409
[94] Cui Y et al 2020 arXiv:2011.09610
[95] Lin H, Gawryluk D J, Klein Y M, Huangfu S, Pomjakushina E, von Rohr F and Schilling A 2021 arXiv:2104.14324
[96] Zhao D et al 2021 Phys. Rev. Lett. 126 197001
[97] Lu H et al 2021 Science 373 213
[98] Wan X, Ivanov V, Resta G, Leonov I and Savrasov S Y 2021 Phys. Rev. B 103 075123
[99] Komarek A C et al 2011 Phys. Rev. B 84 125114
[100] Xia C, Wu J, Chen Y and Chen H 2021 arXiv:2110.12405
[101] Bernardini F, Bosin A and Cano A 2021 arXiv:2110.15580