

Electronic structure of the parent compound of superconducting infinite-layer nickelates

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The search continues for nickel oxide-based materials with electronic properties similar to cuprate high-temperature superconductors^{1–10}. The recent discovery of superconductivity in the doped infinite-layer nickelate NdNiO₂ (refs. ^{11,12}) has strengthened these efforts. Here, we use X-ray spectroscopy and density functional theory to show that the electronic structure of LaNiO₂ and NdNiO₂, while similar to the cuprates, includes significant distinctions. Unlike cuprates, the rare-earth spacer layer in the infinite-layer nickelate supports a weakly interacting three-dimensional 5d metallic state, which hybridizes with a quasi-two-dimensional, strongly correlated state with 3d_{x₂-y₂} symmetry in the NiO₂ layers. Thus, the infinite-layer nickelate can be regarded as a sibling of the rare-earth intermetallics^{13–15}, which are well known for heavy fermion behaviour, where the NiO₂ correlated layers play an analogous role to the 4f states in rare-earth heavy fermion compounds. This Kondo- or Anderson-lattice-like ‘oxide-intermetallic’ replaces the Mott insulator as the reference state from which superconductivity emerges upon doping.

Although the mechanism of superconductivity in cuprates remains a subject of intense research, early on it was suggested that the conditions required for realizing high-temperature superconductivity are rooted in the physics of a two-dimensional (2D) electron system subject to strong local repulsion^{16,17}. This describes Mott (charge-transfer) insulators in the stoichiometric parent compounds, characterized by spin-½ Heisenberg antiferromagnetism, from which superconductivity emerges upon doping. A long-standing question exists as to whether these ‘cuprate–Mott’ conditions can be realized in other oxides, and extensive efforts to synthesize and engineer nickel oxides (nickelates) promised such a realization^{1–10}. Infinite-layer NdNiO₂ became the first such nickelate superconductor following the recent discovery of superconductivity in Sr-doped samples¹¹. The undoped parent compound, produced by removing the apical oxygen atoms from the perovskite nickelate NdNiO₃ using a metal hydride-based soft chemistry reduction process^{10,18–20}, appears to be a close sibling of the cuprates—it is isostructural to the infinite-layer cuprates with monovalent Ni¹⁺ cations and possesses the same 3d⁹ electron count as Cu²⁺ cations in undoped cuprates. Yet, as we will reveal, the electronic structure of the undoped RNiO₂

(R = La and Nd) remains distinct from the Mott, or charge-transfer, compounds of undoped cuprates and even other nickelates.

As a reference, we first discuss the electronic structure of the canonical nickelates, NiO and LaNiO₃. The rocksalt NiO is a charge-transfer insulator, as characterized in the Zaanen–Sawatzky–Allen scheme²¹, whose charge-transfer energy Δ (promoting charge from oxygen ligands to Ni *d* orbitals) lies below the Coulomb interaction scale *U* on Ni sites. The valence Ni *d* orbitals strongly hybridize with oxygen ligands, yielding wavefunctions with mixed character $\alpha|3d^8\rangle + \beta|3d^9L\rangle$ ($\alpha^2 + \beta^2 = 1$), with $\beta^2 \approx 0.2$ (refs. ^{22,23}) per NiO₆ octahedron, where *L* denotes a ligand hole on the oxygens. Such Ni–O ligand hybridization gives rise to a pre-peak in X-ray absorption spectroscopy (XAS) near the O K-edge (Fig. 1a). In addition, a large bandgap set by Δ appears in the oxygen partial density of states (PDOS) obtained both experimentally (Fig. 1b) and from density functional theory with on-site Coulomb interaction potential (LDA+U) calculations (Fig. 1e). In the perovskite RNiO₃, where formal valence counting would give Ni³⁺ (3d⁷), both theoretical and experimental studies indicate that the perovskite structure leads to a decrease of Δ , such that it effectively becomes negative²⁴. Under such a scenario, electrons from oxygen ligands spontaneously transfer to Ni cations, giving rise to ‘self-doped’ holes on the ligands, and a pre-peak in the O K-edge XAS (Fig. 1a). As expected for a negative charge-transfer metal, no bandgap appears in the oxygen PDOS (Fig. 1c,f)²⁵.

The O K-edge XAS tells a very different story for the infinite-layer nickelates LaNiO₂ and NdNiO₂, as shown in Fig. 1a. The lack of a pre-edge peak suggests that the oxygen ligands carry significantly less weight in the ground-state wavefunction, signalling a weaker effective mixing between oxygen and the Ni¹⁺ cations. Unlike NiO and LaNiO₃, the oxygen PDOS (Fig. 1d) exhibits a diminished weight near the Fermi energy, especially in the unoccupied states, also indicating that O 2*p* orbitals carry less weight in the expected upper Hubbard band by comparison, all of which is consistent with the calculated oxygen PDOS from LDA + *U* (Fig. 1g).

Although the oxygen electronic structure deviates significantly from other nickelates and cuprates²⁶, we examined the electronic structure of the Ni cation in RNiO₂ using both XAS and resonant inelastic X-ray scattering (RIXS) at the Ni L₃-edge (a core-level 2*p* to valence 3*d* transition). As shown in Fig. 2a, while XAS spectra for

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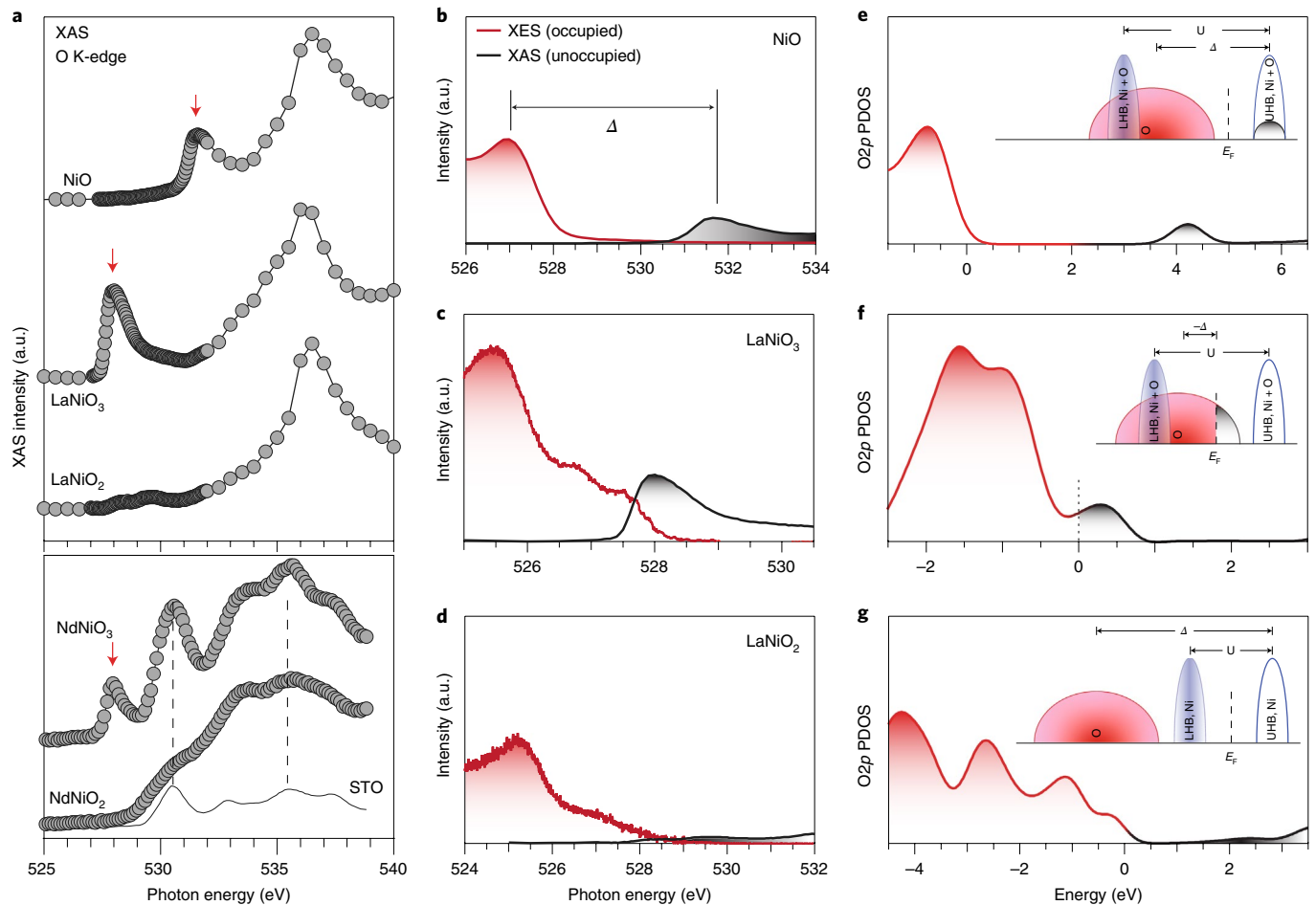


Fig. 1 | X-ray spectroscopy near the O K-edge and LDA + U calculation. **a**, Upper panel: XAS spectra for NiO, LaNiO₃ and LaNiO₂. Red arrows mark the pre-edge peaks indicative of Ni–O hybridization. The lower panel shows the XAS of NdNiO₃ and NdNiO₂ due to the film thickness being thinner than that of the La-based films shown in the upper panel. Spectra are vertically offset for clarity. **b–d**, X-ray emission spectra (XES) and XAS in the pre-edge region, roughly reflecting the occupied (red shading) and unoccupied (black shading) oxygen PDOS, respectively. Vertical lines illustrate the bandgap projected in the oxygen DOS, corresponding to the effective charge-transfer energy Δ in NiO and LaNiO₃. **e–g**, LDA + U calculations for the PDOS with O 2p orbital character ($U = 8$ eV for NiO and LaNiO₃, $U = 6$ eV for LaNiO₂). Red and black shading indicates the occupied and unoccupied oxygen PDOS, respectively. Insets: sketches of the relationship between U and Δ in the Zaanen–Sawatzky–Allen scheme for each compound. UHB and LHB stand for upper and lower Hubbard band, respectively.

both NiO and LaNiO₃ exhibit distinct multi-peak structures originating from $2p^63d^8-2p^53d^9$ and $2p^63d^8L^n-2p^53d^9L^n$ multiplet transitions, respectively^{23,24}, XAS for the infinite-layer nickelates shows a main absorption peak (denoted A) that closely resembles the single peak associated with the $2p^63d^9-2p^53d^{10}$ transition in cuprates²⁷. In particular for LaNiO₂, the XAS exhibits an additional lower-energy shoulder A'. In the RIXS map shown in Fig. 2b, the ~ 1 and ~ 1.8 eV features resemble the dd excitations seen in LaNiO₃ (Fig. 2c) and NdNiO₃ (ref. 24), except they are broader and exhibit a dispersion with incident photon energy. This suggests that the Ni 3d states in LaNiO₂ are mixed with delocalized states, probably the La³⁺ 5d states. Interestingly, at the A' resonance, a 0.6 eV feature appears that is absent in the RNiO₃ compounds (Fig. 2c,e and ref. 24). Using exact diagonalization (see Methods), we reproduce the general features from XAS and RIXS (Fig. 2f–h), including the A' features, highlighting the hybridization between the Ni $3d_{x^2-y^2}$ and La 5d orbitals. Thus, in configuration interaction, the Ni state can be expressed as a combination of $|3d^9\rangle$ and $|3d^8R\rangle$, where R denotes a charge-transfer to the rare-earth cation (see Methods and Supplementary Table 2). RIXS measurements across the La M₄-edge ($3d-4f$ transition) do not show any signatures of 4f orbital excitations, as expected for the

completely empty 4f shell of the La³⁺ cation (Extended Data Fig. 3). Hence, we argue that the La 4f states are frozen in the core and not hybridized with Ni 3d states (Extended Data Fig. 2). In NdNiO₂, the ~ 0.6 eV feature due to the Nd–Ni hybridization also exists in RIXS (Fig. 2d,e), but its resonance energy (A') almost coincides with the main absorption peak A. As a consequence, the A' feature cannot be resolved in XAS (Fig. 2a). We note that the separation between A and A' depends on the energetic balance among microscopic parameters, including site energy, charge-transfer energy and rare-earth Ni hybridization energy, which are expected to vary between infinite-layer nickelates as a function of the rare-earth element.

To further analyse the electronic structure, we turn to density functional theory (DFT). The LDA + U scheme²⁸ has a long track record of correctly reproducing the gross features of correlated electronic structure for transition metal oxides. Although generally first principle, one cannot be certain about the value of the local Coulomb interaction U ; however, we can put bounds on it. The infinite-layer nickelates are undoubtedly less good metals than elemental nickel, characterized by $U \approx 3$ eV, which we can take as a lower bound. From O K-edge XAS, the Coulomb interaction should be smaller than that of the large-bandgap charge-transfer insulator NiO, where

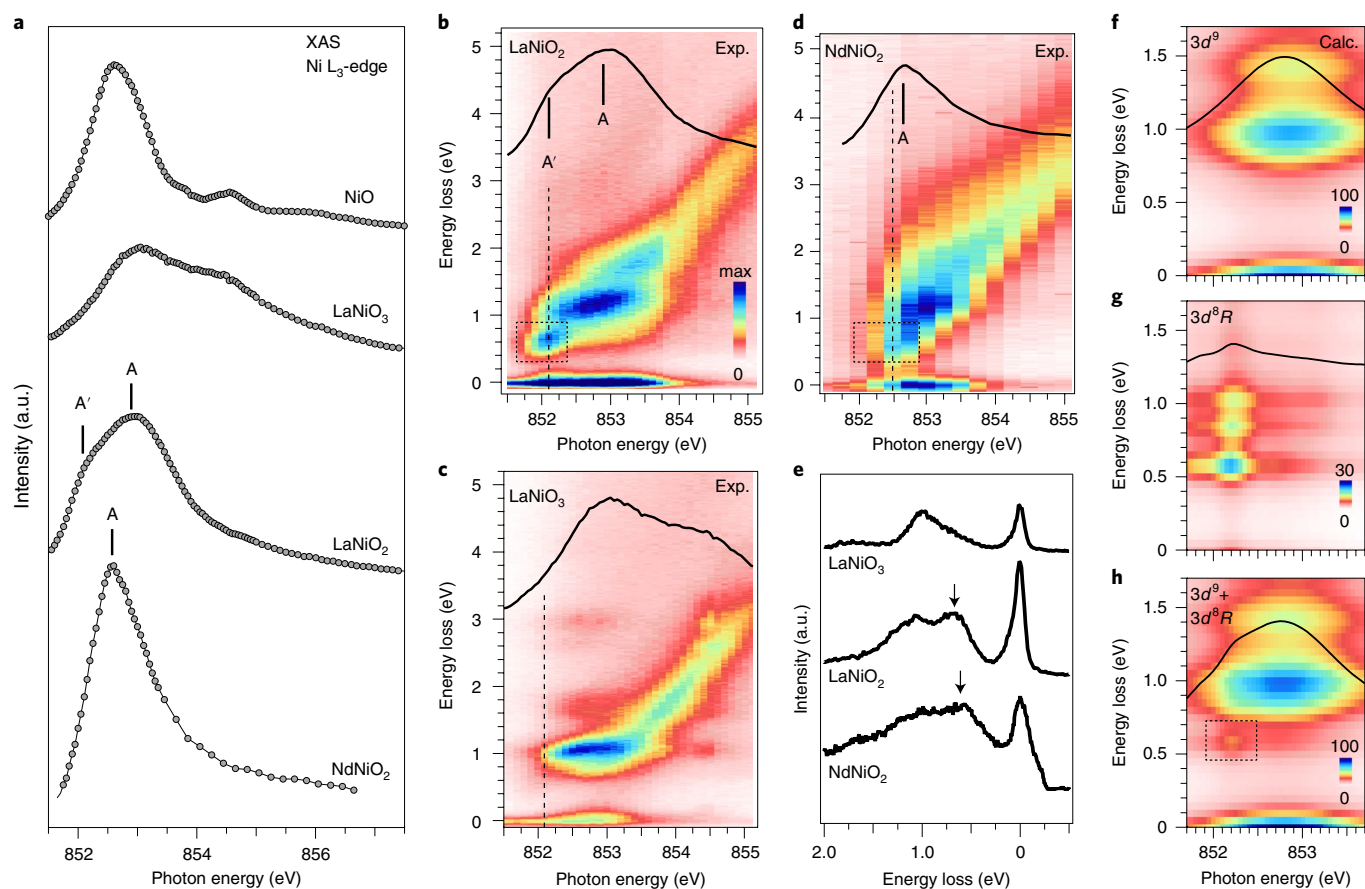


Fig. 2 | XAS and RIXS at the Ni L_3 -edge. **a**, XAS of NiO, LaNiO₃, LaNiO₂ and NdNiO₂. The La M_4 -line was subtracted from the LaNiO₂ and LaNiO₃ spectra (Extended Data Fig. 2). The markers A indicate the main peak of LaNiO₂ and NdNiO₂. A' labels a lower-energy shoulder in the XAS of LaNiO₂. Spectra are vertically offset for clarity. **b–d**, RIXS intensity map of LaNiO₂, LaNiO₃ and NdNiO₂ measured as a function of incident photon energy at $T = 20$ K. The corresponding XAS are superimposed as a solid black line in each map. The dashed boxes in **b** and **d** highlight the -0.6 eV features of LaNiO₃ and NdNiO₂ that are associated with the Ni–La and Ni–Nd hybridizations, respectively. **e**, RIXS energy loss spectra of LaNiO₃, LaNiO₂ and NdNiO₂ at incident energies indicated by vertical dashed lines in **b–d**. Black arrows highlight the 0.6 eV features of LaNiO₂ and NdNiO₂. **f–h**, Calculated RIXS maps and absorption spectra (solid black lines) of LaNiO₂ for $3d^9$ (**f**), $3d^9R$ (**g**) and $3d^9 + 3d^9R$ (**h**) (R denotes a charge-transfer to the rare-earth cation) ground state, respectively. The dashed box in **h** highlights the same feature as the box in **b**.

$U \approx 8$ eV. Here, we choose $U = 6$ eV in our calculations for LaNiO₂ (with a lowest energy antiferromagnetic solution, see Methods for details), revealing some salient features that correlate with experimental observations. (1) As shown in Fig. 1g (and Fig. 3a), when compared to other nickelates, the oxygen $2p$ bands lie significantly further away from the Fermi energy, signalling reduced oxidation and implying a charge-transfer energy Δ that exceeds U . This places the RNiO₂ infinite-layer nickelates within the Mott–Hubbard regime of the Zaanen–Sawatzky–Allen scheme²¹. (2) The density of states near E_F is dominated by the half-filled Ni $3d_{x^2-y^2}$ states, which appear isolated from the occupied Ni $3d$ bands. The characteristic lower and upper Hubbard bands (Fig. 3a), at least in part, signal a textbook single-band Hubbard model, all but confirming that the Ni cation should be in a very nearly monovalent $3d^9$ state, consistent with the Ni L-edge XAS and RIXS (Fig. 2). (3) The density of states at E_F is actually finite, but small, as shown upon closer inspection of Fig. 3a,b. Near the Γ point, a Fermi surface pocket forms that is mainly of La $5d$ character (Fig. 3b); it is quite extended and 3D (see the wavefunction at a Fermi momentum k_F , Fig. 3c, and the Fermi surface, Fig. 3d). This contrasts with the 2D nature of the correlated $3d_{x^2-y^2}$ Ni states (Fig. 3b). In other words, the electronic structure of the infinite-layer nickelate consists of a low-density 3D metallic rare-earth band coupled to a 2D Mott system.

To theoretically investigate emergent phenomena in infinite-layer nickelates, a low-energy effective model can be derived as a starting point. A minimal model for these materials would look like

$$H = \sum_{k,\sigma} \left(\epsilon_k^R n_{k,\sigma}^R + \epsilon_k^{\text{Ni}} n_{k,\sigma}^{\text{Ni}} \right) + U \sum_i n_{i,\uparrow}^{\text{Ni}} n_{i,\downarrow}^{\text{Ni}} + \sum_{k,i,\sigma} \left(V_{k,i} c_{k,\sigma}^\dagger d_{i,\sigma} + \text{h.c.} \right)$$

where the first term describes the non-interacting rare-earth (R) and Ni bands with energies ϵ_k^R and ϵ_k^{Ni} , respectively, the second term represents the usual on-site Hubbard interaction with strength U in the quasi-two-dimensional Ni layer, and the third term describes the coupling with strength $V_{k,i}$ between the R and Ni subsystems. Here, $n_{k,\sigma}^R$ and $n_{k,\sigma}^{\text{Ni}}$ represent the usual number operators for the R and Ni subsystems, while $c_{k,\sigma}^\dagger$ ($c_{k,\sigma}$) and $d_{k,\sigma}^\dagger$ ($d_{k,\sigma}$) create (annihilate) electrons in the 3D metallic R and 2D Hubbard-like Ni subsystems, respectively. This model resembles the Anderson-lattice (or Kondo-lattice) model for the rare-earth intermetallics^{13–15}, but with the notable addition of a weakly hybridized single-band Hubbard-like model for the Ni layer, rather than strongly interacting $4f$ states

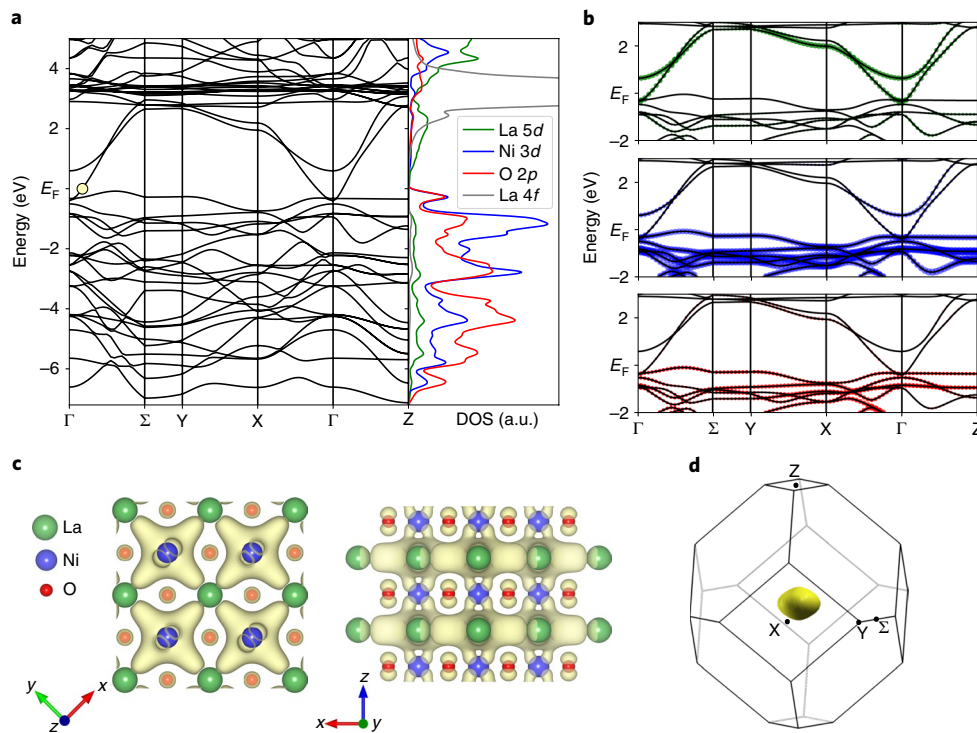


Fig. 3 | Electronic structure of LaNiO₂. Theoretical calculations of the electronic structure in the LDA + U framework with $U=6$ eV (antiferromagnetic solution). **a**, Band structure of LaNiO₂ along high symmetry directions in the body centred tetragonal (bct) Brillouin zone. The Brillouin zone with labelled high symmetry points is also shown in **d**. The right-hand side shows the La 5d (green), Ni 3d (blue), O 2p (red) and La 4f (grey) PDOS with a smaller energy broadening than that used in Fig. 1e–g. **b**, Orbital projected band structure of LaNiO₂ near E_F . The colour code is identical to that used in **a**, representing the projection onto orbitals with different atomic character. **c**, Top and side views of an electron density contour for the single-particle wavefunction at Fermi momentum k_F along Γ – Σ (yellow marker in **a**). **d**, Fermi surface (closed electron pocket) around Γ with dominant La 5d character in the first bct Brillouin zone with labelled high symmetry points.

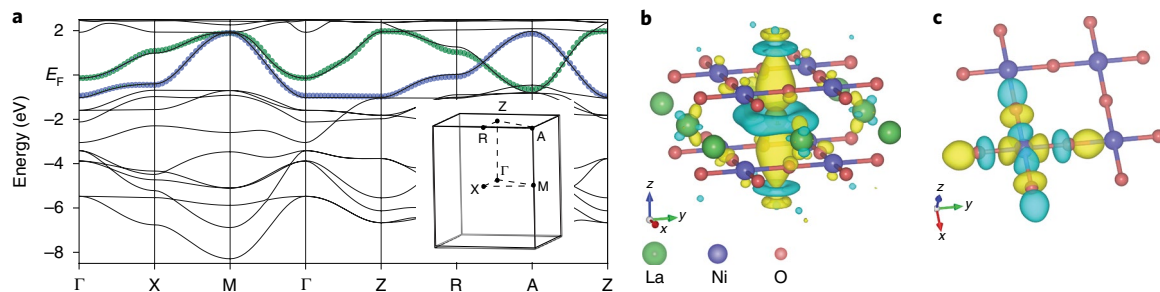


Fig. 4 | Deriving a minimal model for the rare-earth infinite-layer nickelates. **a**, Band dispersion of LaNiO₂, highlighting two bands that cross E_F in the paramagnetic LDA calculation. The inset shows the high symmetry points in the tetragonal Brillouin zone. **b, c**, Isosurface plots for an extended La-centred $d_{3z^2-r^2}$ -like (**b**) and essentially planar Ni-centred $d_{x^2-y^2}$ -like (**c**) Wannier orbital for the minimal low-energy model of LaNiO₂. These two orbitals produce the 3D band (La, green) and quasi-2D band (Ni, blue) highlighted in **a**. The derived parameters of the minimal model are listed in Supplementary Table 3.

(or localized spin moments). We can take this a step further and derive the parameter of this model Hamiltonian by performing Wannier ‘downfolding’ on the band structure in the one-Ni unit cell with $U=0$ eV. Figure 4a shows the band structure for LaNiO₂ obtained from LDA without a Hubbard U (see Supplementary Information for details). Here, consistent with previous calculations², two bands cross the Fermi level: a fully 3D band with predominantly La 5d character and a quasi-2D band with Ni 3d character. Wannier downfolding²⁹ produces one extended orbital with $d_{3z^2-r^2}$ symmetry centred on La (Fig. 4b) and another orbital confined primarily to the NiO₂ planes with $d_{x^2-y^2}$ symmetry centred

on Ni (Fig. 4c), which are fully consistent with the expected orbital arrangements given the crystal and ligand field symmetries for this material and the LDA + U results shown in Fig. 3. Full details about the downfolded model, including effective model parameters, are provided in Supplementary Table 3.

This downfolded model is, to the best of our knowledge, unique to this particular system. Viewed theoretically, this ‘Hubbard–Kondo’ model is uncharted territory and it is a natural question to ask what happens to the basic single-band Hubbard model when its states weakly hybridize with a metallic band. For example, do the spins in the NiO₂ layers order antiferromagnetically or will the Kondo effect

strongly screen the local moments and give rise to electronic band hybridizations^{13–15} in analogy to the case of heavy fermions? Note that, unlike the rare-earth intermetallics, here Ni spins interact via the strong short-range super-exchange interaction, which replaces the Ruderman–Kittel–Kasuya–Yosida interactions in the heavy fermion compounds. More importantly, can superconductivity emerge in this model by introducing doped charge carriers? Apparently, experimental information, particularly about the Fermi surface and magnetic susceptibility, and information about other elementary excitations such as spin, charge and phonon excitations will be required to gain further insights. Nevertheless, our results have provided a glimpse into the remarkable electronic structure of the parent compounds of superconducting infinite-layer nickelates, which appear to serve as a birthplace of superconductivity upon doping.

Online content

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Methods

Materials. LaNiO₃ films with 12 and 50 nm thicknesses were grown on top of 5 × 5 mm² TiO₂-terminated SrTiO₃ (001) substrates by pulsed laser deposition using a 248 nm KrF excimer laser. Before growth, SrTiO₃ substrates were pre-annealed at an oxygen partial pressure (p_{O_2}) of 5 × 10⁻⁶ torr for 30 min at 950 °C to achieve sharp step-and-terrace surfaces. The films were subsequently grown at a substrate temperature T_g of 575 °C and $p_{O_2} = 34$ mtorr, using 1.4 J cm⁻² laser fluence and 4 mm² laser spot size on the target. The growth was monitored by reflection high-energy electron diffraction intensity oscillations. After the growth, the samples were cooled to room temperature in the same oxygen environment. Characterization by X-ray diffraction (XRD) scans with Cu K α radiation indicated the presence of the perovskite phase of (001)-oriented LaNiO₃ and high epitaxial quality for all as-grown films. AFM topographic scans showed atomically flat surfaces. Reducing conditions³⁰ were adapted to remove apical oxygen to produce both the (001)-oriented LaNiO_{2.5} and LaNiO₂ phases. For reduction experiments, each LaNiO₃ sample was cut into two pieces with dimensions of 2.5 × 5 mm². The 2.5 × 5 mm² sample was then vacuum-sealed together with blocks of CaH₂ powder in a Pyrex glass tube (pressure <0.1 mtorr). The tube was heated to 240 °C at a rate of 10 °C min⁻¹ and kept at this temperature for 30–120 min, before being cooled to room temperature at a rate of 10 °C min⁻¹. After the annealing process, remnant CaH₂ powder on the sample surface was rinsed off with 2-butanone. The XRD scans in Extended Data Fig. 1a show the characteristic Bragg peaks of the 12 nm LaNiO₃ film and the ~50 nm LaNiO₂ film used in the XAS and RIXS measurements in the main text. Additionally, a ~50 nm LaNiO_{2.5} film was characterized as a reference sample. The 2 θ peak positions of these three films coincide with that of similar films on SrTiO₃ (ref. 30). The c -axis lattice constants extracted from the XRD scans were 3.809, 3.771 and 3.407 Å for the LaNiO₃, LaNiO_{2.5} and LaNiO₂ films, respectively. In comparison to LaNiO₂ powder^{17,30}, the c -axis lattice constant of the film is slightly expanded due to the compressive strain induced by the SrTiO₃ substrate.

NdNiO₂ films grown on a SrTiO₃ substrate with a thickness of ~10 nm were prepared using the conditions described in ref. 11. NdNiO₂ films with and without a capping layer of 5 unit cells of SrTiO₃ were measured and show the same spectral properties. As a reference, we also measured a NdNiO₃ film grown on a SrTiO₃ substrate and capped with 5 unit cells of SrTiO₃.

Extended Data Fig. 1b displays the resistivity as a function of temperature of the LaNiO₃ film in a four-probe geometry, which shows metallic behaviour down to 2 K. The LaNiO₂ film exhibits higher resistivity than LaNiO₃ at 300 K, which increases further with decreasing temperature (Extended Data Fig. 1b). Similar transport properties are reported in refs. 20,30,31.

Commercially available NiO powder (>99.995% purity, Sigma-Aldrich) was used for the measurements.

XAS and RIXS measurements. The XAS and RIXS spectra of the La-based nickelate samples were measured at the ADDRESS beamline with the SAXES spectrometer at the Swiss Light Source (SLS) of the Paul Scherrer Institut³². For RIXS measurements the scattering angle was fixed to 130° and the combined instrument resolution was ~100 meV at the Ni L₃-edge. The scattering plane coincided with the crystallographic a - c (b - c) plane with a grazing incident angle of $\theta = 15^\circ$. The XES and RIXS spectra shown in Fig. 1 were measured with π -polarized incident photons. Due to the strong fluorescence signal from the STO substrate, the XES of LaNiO₃ and LaNiO₂ shown in Fig. 1 were obtained from the fluorescence signal identified in RIXS incident-photon-energy and emission-energy map across the oxygen pre-edge (incident photon energy from ~525 eV to ~530 eV). The elastic line and weak Raman-like excitations (in LaNiO₂) were removed for clarity.

The XAS and XES spectra of NiO shown in Fig. 1a were measured at beamline BL8.0 using the q-RIXS endstation of the Advanced Light Source (ALS) of the Lawrence Berkeley National Laboratory. For the RIXS/XES measurements the scattering angle was fixed at 130° and the combined instrument resolution was ~300 meV at the Ni L₃-edge and ~200 meV at the O K-edge. The XAS spectra at the O K-edge for NdNiO₃ and NdNiO₂ (Fig. 1a) were taken at 41A BlueMagpie beamline at Taiwan Photon Source. The XAS and RIXS maps at the Ni L-edge of NdNiO₂ were taken at beamline I21 at the Diamond Light Source. The RIXS spectrometer was set at 146°, with a resolution of ~50 meV. The scattering plane coincided with the crystallographic a - c (b - c) plane with a grazing incident angle of ~10°. π -polarized incident photons were used for this measurement.

All XAS spectra at the O K-edge (Fig. 1) were taken in fluorescence yield mode with a grazing incident angle of 10 and 20° for the La-based and Nd-based nickelates, respectively. The grazing incident geometry was used to reduce the signal arising from the STO substrate. The spectra were normalized such that the intensity at the pre-edge and the post-edge were 0 and 1, respectively.

All XAS spectra at the Ni L-edge (Fig. 2) were taken in fluorescence yield mode with normal incident geometry. These XAS were normalized such that the intensity at the pre-edge and the post-edge were 0 and 1, respectively. For the XAS spectra for the La-based nickelates, the intense La M₄-line centred around 850.5 eV (Extended Data Fig. 2a) was fitted by a Lorentzian peak profile and subtracted from the LaNiO₃ and LaNiO₂ XAS to correct for the overlap between the tail of the La M₄-line and the Ni L₃-edge. The resulting spectra are shown in Fig. 2.

Theory calculations. For the oxygen PDOS shown in Fig. 1e–g and the electronic structure of LaNiO₂ shown in Fig. 3, LDA + U calculations were performed using the GGA method and the simplified version from Cococcioni and de Gironcoli³³, as implemented in QUANTUM ESPRESSO³⁴. We find that an antiferromagnetic solution, with wavevector (π, π, π) , leads to the lowest energy, with a two-Ni bct unit cell and corresponding Brillouin zone.

The Ni L₃-edge RIXS calculations (Fig. 2) were performed using an exact diagonalization technique^{35,36}, which accounts for the full overlap of the many-body wavefunctions. The microscopic Hamiltonian used for these calculations includes both material-specific on-site energies and hybridizations as encoded in a Wannier downfolding of the band structure³⁹ and the full set of Coulomb interactions as expressed in terms of Slater integrals. The relevant parameters used for Wannier downfolding paramagnetic LaNiO₂ (a one-Ni tetragonal unit cell), as shown in Supplementary Table 1, were obtained from Wannier90³⁷ for 12-orbital (O $p_x/p_y/p_z$, Ni $d_x^2-y^2/d_{xy}/d_{xz}/d_{yz}$, La d_z^2). The Slater integrals for Ni 3d in the LaNiO₂ calculations were $F^0 = 0.5719$ eV, $F^2 = 11.142$ eV and $F^4 = 6.874$ eV. The Slater integrals for Ni 3d–2p interactions were $F^0_{pd} = 0.148$ eV, $F^2_{pd} = 6.667$ eV, $G^1_{pd} = 4.922$ eV and $G^3_{pd} = 2.796$ eV. The values of F^2 , F^4 , F^2_{pd} , G^1_{pd} and G^3_{pd} are taken from ref. 29. We take 0.7 as a screening factor for the non-monopole terms. A core-level spin-orbit coupling of 12.5 eV was used for the Ni 2p core electrons. The resulting weight of the Ni wavefunction is shown in Supplementary Table 2.

The two-orbital, low-energy model for the physics of LaNiO₂ is shown in Fig. 4. This model, obtained once again by Wannier downfolding the DFT paramagnetic solution for LaNiO₂ in the one-Ni tetragonal unit cell (the same method as that used to obtain the non-interacting part of the Hamiltonian for the LaNiO₂ RIXS calculation, but only for the two bands that cross E_F), yields the independent hopping parameters listed in Supplementary Table 3, cut off for absolute values smaller than 0.008 eV. The two Wannier orbitals are shown in Fig. 4b: (1) a very extended orbital centred on La with $d_{3z^2-r^2}$ character, which makes up the majority character of the 3D band and (2) a more localized orbital, centred on Ni and primarily confined to the NiO₂ plane, with $d_{x^2-y^2}$ character, which makes up the majority character of the quasi-2D band. Note that this paramagnetic solution in the tetragonal Brillouin zone has one large quasi-2D hole-like Fermi surface from the Ni-centred orbital and two smaller 3D electron-like Fermi surfaces centred at the Γ - and A-points from the La-centred orbital. The low-energy, antiferromagnetic band structure from LDA + U (Fig. 3) would result from a (π, π, π) band-folding of the La-centred band, which moves the A-point to the Γ -point, formation of upper and lower Hubbard Ni-centred bands, gapping-out of the large hole Fermi surface, and a shift in chemical potential to compensate for the loss of carriers, which leaves a single electron pocket at the Γ -point.

The non-interacting bands of the effective low-energy model can be written in tight-binding form as

$$\begin{aligned} \epsilon_k^R = & \epsilon_0^R + 2t_{[0,0,1]}^R \cos(k_z) + 2t_{[0,0,2]}^R \cos(2k_z) + 2t_{[0,0,3]}^R \cos(3k_z) \\ & + 2t_{[1,0,0]}^R [\cos(k_x) + \cos(k_y)] \\ & + 4t_{[1,0,1]}^R [\cos(k_x) + \cos(k_y)] \cos(k_z) \\ & + 4t_{[1,0,2]}^R [\cos(k_x) + \cos(k_y)] \cos(2k_z) \\ & + 4t_{[1,1,0]}^R \cos(k_x) \cos(k_y) \\ & + 8t_{[1,1,1]}^R \cos(k_x) \cos(k_y) \cos(k_z) \\ & + 8t_{[1,1,2]}^R \cos(k_x) \cos(k_y) \cos(2k_z) \\ & + 8t_{[1,1,3]}^R \cos(k_x) \cos(k_y) \cos(3k_z) \\ & + 4t_{[2,0,1]}^R [\cos(2k_x) + \cos(2k_y)] \cos(k_z) \\ & + 8t_{[2,1,1]}^R [\cos(2k_x) \cos(k_y) + \cos(k_x) \cos(2k_y)] \cos(k_z) \end{aligned}$$

$$\begin{aligned} \epsilon_k^{Ni} = & \epsilon_0^{Ni} + 2t_{[1,0,0]}^{Ni} [\cos(k_x) + \cos(k_y)] \\ & + 4t_{[1,1,0]}^{Ni} \cos(k_x) \cos(k_y) \\ & + 2t_{[2,0,0]}^{Ni} [\cos(2k_x) + \cos(2k_y)] \\ & + 2t_{[0,0,1]}^{Ni} \cos(k_z) \\ & + 8t_{[1,1,1]}^{Ni} \cos(k_x) \cos(k_y) \cos(k_z) \end{aligned}$$

where the appropriate matrix elements are provided in Supplementary Table 3.

Data availability

Raw data are shown in Figs. 1a–d and 2a–e, Extended Data Fig. 1 and Extended Data Fig. 2. The data that support the plots within this paper and other findings of this study are available from the corresponding authors upon reasonable request.

Code availability

QUANTUM ESPRESSO and Wannier90 are freely available at <https://www.quantum-espresso.org> and <http://www.wannier.org>, respectively. Access to RIXS exact diagonalization and Python analysis codes will be accommodated upon reasonable request to the corresponding authors.

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Author contributions

W.S.L., M.H. and H.Y. Hwang conceived the experiment. M.H., H.L., E.P., Y.T., T.S. and W.S.L. conducted the experiment at SLS. H.L., W.S.L., Z.H. and Y.-D.C. conducted the experiment at ALS. H.L., W.S.L., A.N. and K.J.Z. conducted XAS measurement at Diamond Light Source. M.R., W.S.L., H.Y. Huang and D.J.H. conducted XAS measurements at NSRRC. J.S.L. contributed to XAS characterization of samples at an early stage of the work. M.H., H.L. and W.S.L. analysed the data. C.J.J., B.M., J.Z. and T.P.D. performed the theoretical calculations. D.L., X.F., Y.H., M.O. and H.Y. Hwang synthesized and characterized the nickelate samples using transport and XRD. M.H., Z.X.S. and W.S.L. prepared and aligned samples for X-ray spectroscopy measurements. M.H., B.M., J.Z. and W.S.L. wrote the manuscript with input from all authors.

Competing interests

The authors declare no competing interests.

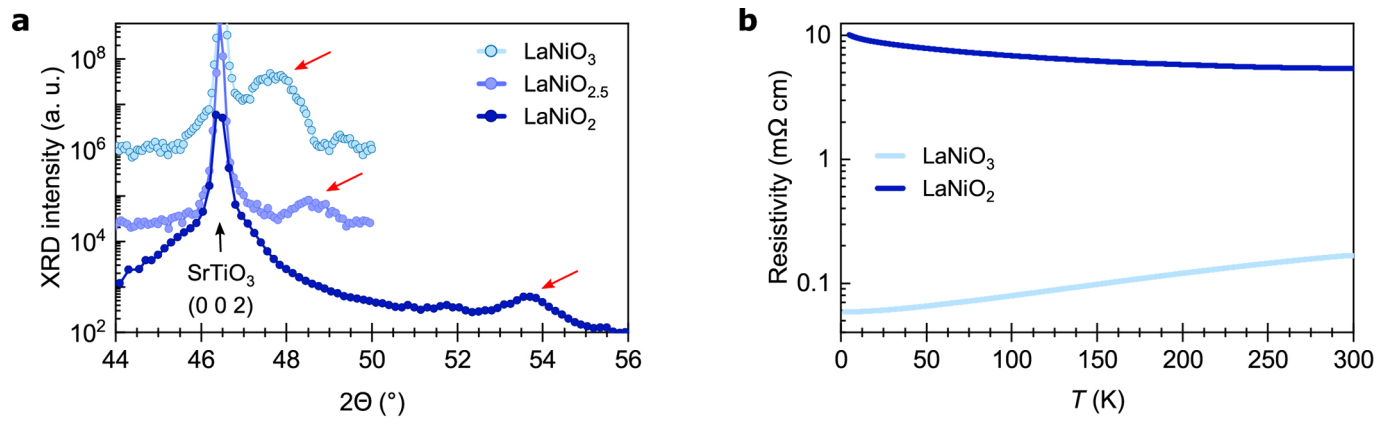
Additional information

Extended data is available for this paper at <https://doi.org/10.1038/s41563-019-0585-z>.

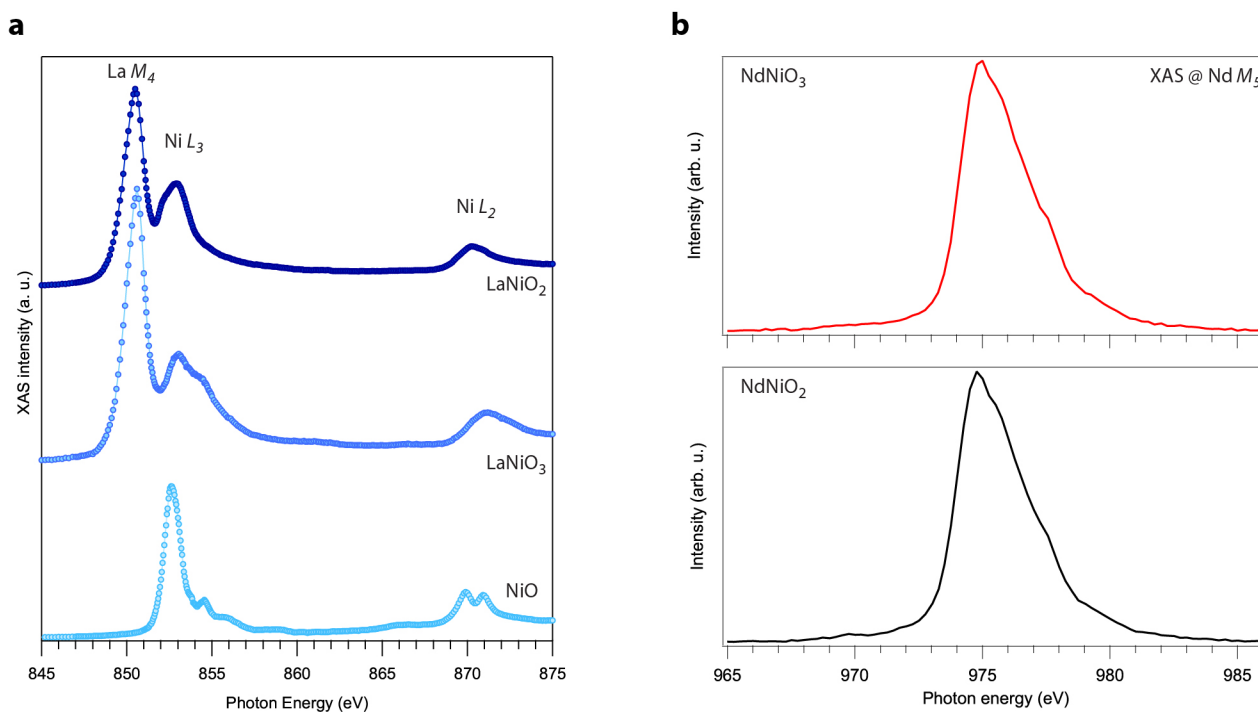
Supplementary information is available for this paper at <https://doi.org/10.1038/s41563-019-0585-z>.

Correspondence and requests for materials should be addressed to C.J.J. or W.S.L.

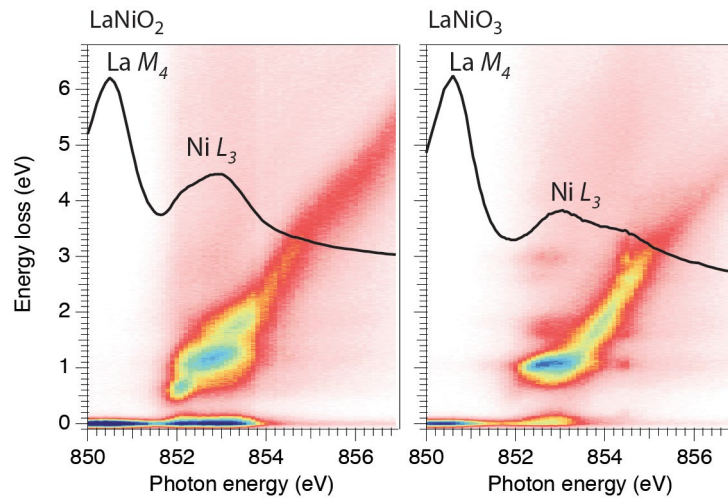
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Extended Data Fig. 1 | XRD characterization and electrical transport measurements. a, XRD pattern of the LaNiO₃, LaNiO_{2.5}, and LaNiO₂ films grown on SrTiO₃ (001) substrates, measured with Cu K α radiation. The red arrows indicate the nickelate film peaks and the black arrow the (002) SrTiO₃ substrate peak. The film peak shifts to higher 2θ values as a function of apical oxygen reduction. The curves are offset in vertical direction for clarity. **b**, Resistivity vs. temperature of the LaNiO₃ and LaNiO₂ film.



Extended Data Fig. 2 | Ni L-edge, La M_4 -edge and Nd M_5 -edge x-ray absorption spectra (XAS). **a**, XAS of LaNiO_3 and LaNiO_2 across the Ni $L_{3,2}$ -edge before subtraction of the La M_4 absorption peak. The XAS of NiO powder is also shown. Spectra are offset in vertical direction for clarity. **b**, XAS of NdNiO_3 and NdNiO_2 across the Nd M_5 -edge. The XAS were taken using total fluorescence yield. The XAS spectra across the rare-earth M -edges of the La- and Nd-based infinite-layer nickelates are essentially identical to those of their perovskite counterparts (LaNiO_3 and NdNiO_3). This implies a similar configuration of $4f$ states for both types of nickelates and that the $4f$ states of La^{3+} and Nd^{3+} do not hybridize in any significant way with the Ni $3d$ states.



Extended Data Fig. 3 | RIXS maps across the La M_4 -edge. RIXS maps for (left) LaNiO_2 and (right) LaNiO_3 with incident photon energies including the La M_4 edge. No orbital excitations exist in the inelastic channel of the RIXS spectra. This is consistent with a nominally empty $4f$ shell of the La^{3+} ion, implying that the $4f$ states are far from near- E_F states and not hybridized with the Ni $3d$ states.