RUHR UNIVERSITÄT BOCHUM RUB

A tale of two kinds of superconducting nickelates

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Engineered 2D Quantum Materials

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Collaborations

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- Jannik Gondolf
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Additional support

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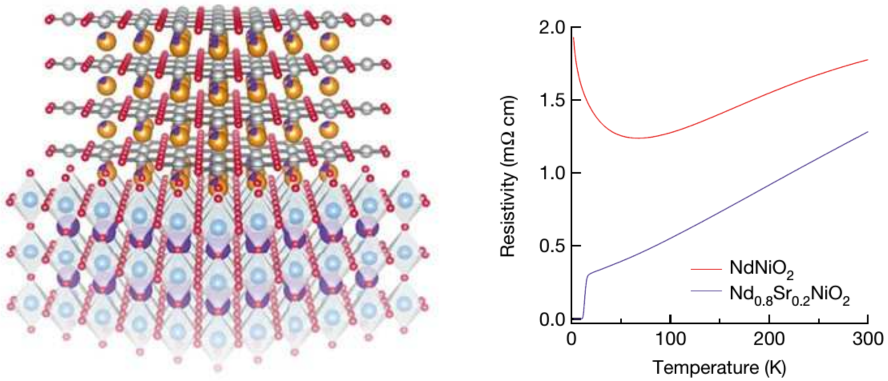
- Andreas Kreisel, Kopenhagen
- Astrid Romer, Kopenhagen
- Evgeny Stepanov, Paris
- Matteo Vandelli, Hamburg
- Alexander I. Lichtenstein, Hamburg

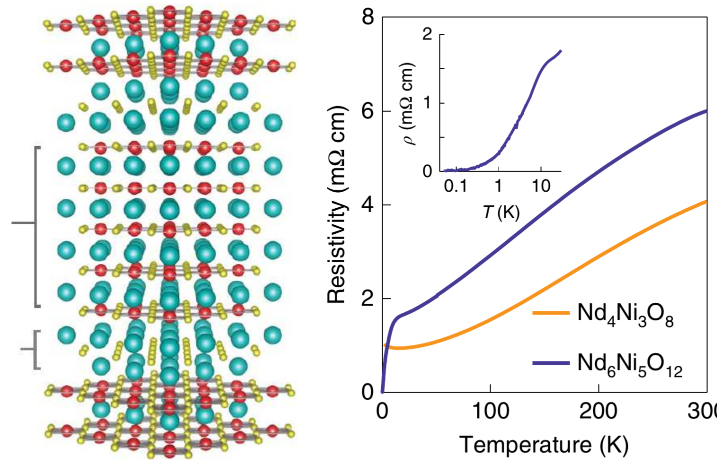
Superconducting nickelates: materials

$9-\delta$



infinite-layer Sr-doped RENiO$_2$, T_c ~10-25 K

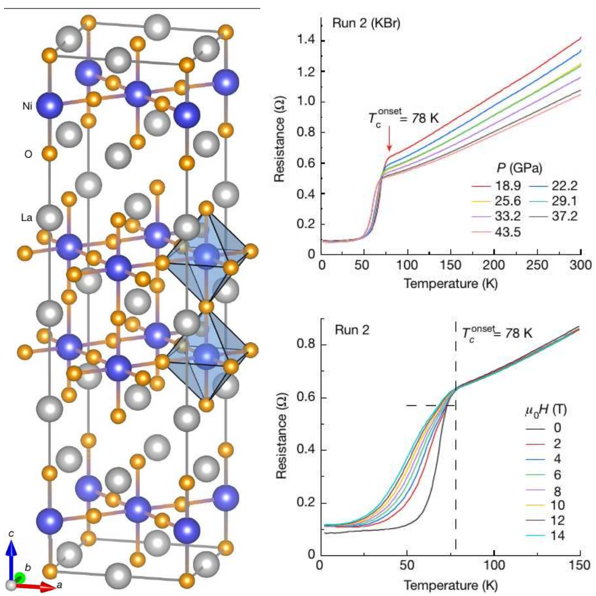
[Li et al., Nature 572, 624 (2019)]



quintuple-layer Nd$_6$Ni$_5$O$_{12}$, T_c ~10 K

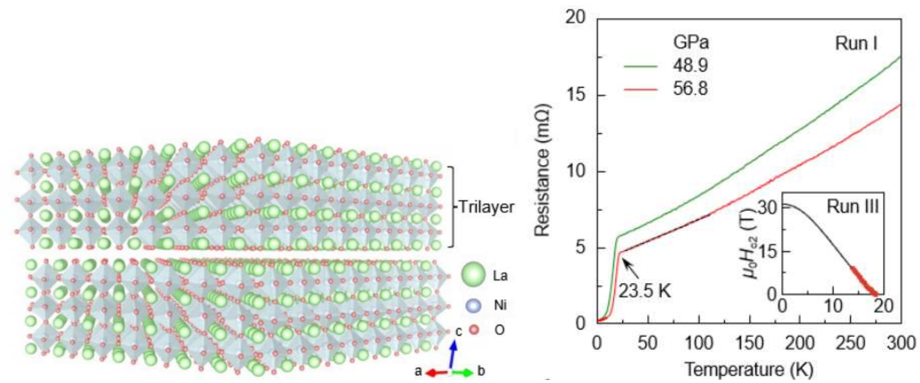
[Pan et al., Nat. Mater. 21, 160 (2022)]

$8\pm\delta$



high-pressure La$_3$Ni$_2$O$_7$, T_c ~65-80 K

[Sun et al., Nature 621, 493 (2023)]

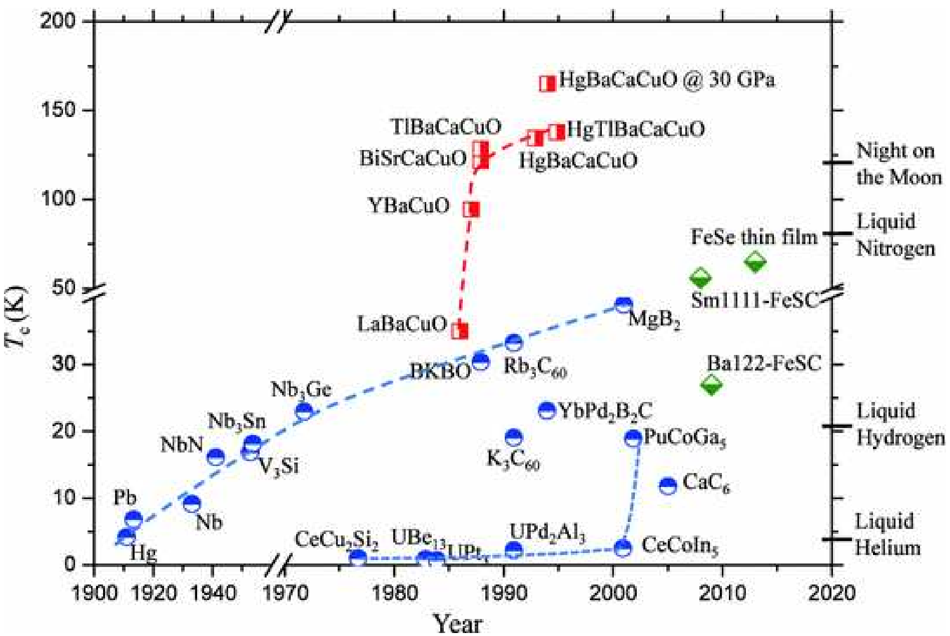


high-pressure La$_4$Ni$_3$O$_{10}$, T_c ~23 K

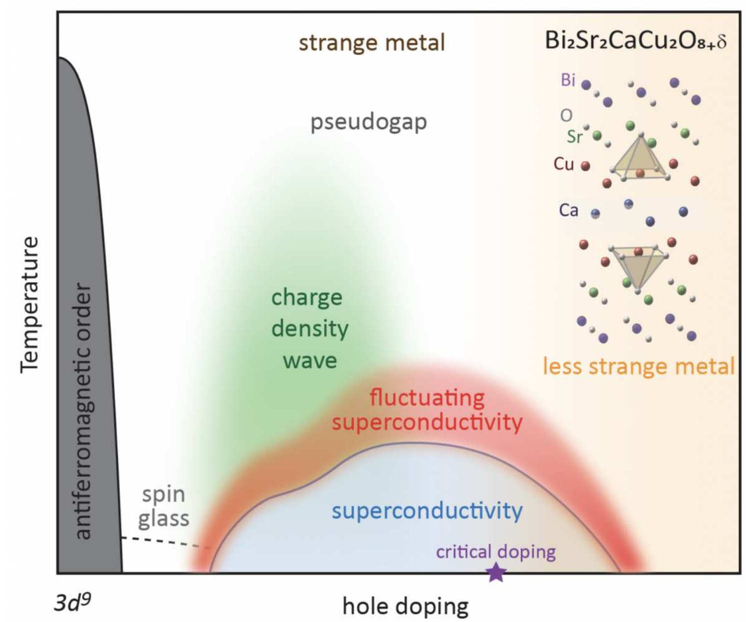
[Zhu et al., Nature 631, 531 (2024)]

[Zhang et al., arXiv:2311.07423 (2023)]

Superconducting nickelates: motivated by high-T_c cuprates



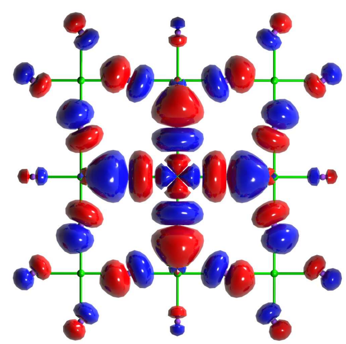
[Rigamonti, Caretta, Structure of Matter]



[Shen group, Stanford]

basic cuprate characteristics

- charge-transfer insulator, Cu^{2+} $(3d^9)$
- square CuO$_2$ planes
- antiferromagnetic (AFM) order
- hole doping: $3d^9\,\underline{L}$
  $\rightarrow$ leads to low-energy Zhang-Rice singlet
- effective one-orbital pic (minimal model):
  Cu-$d_{x^2-y^2}$ hybridized with O$(2p)$



by O. K. Andersen

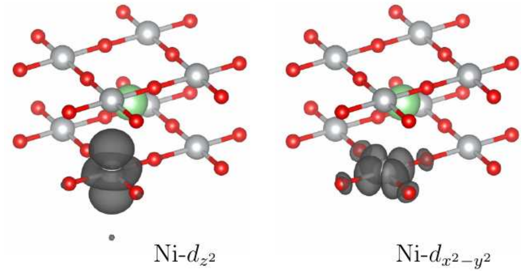
"nickelates to cuprates"?

- prefered in oxides: Ni^{2+} $(3d^8)$
- doping of insulating nickelates often leads to in-gap states
- 'most-natural' Ni^{+} $(3d^9)$ compound: RENiO$_2$ (with RE rare-earth $3+$ ion)

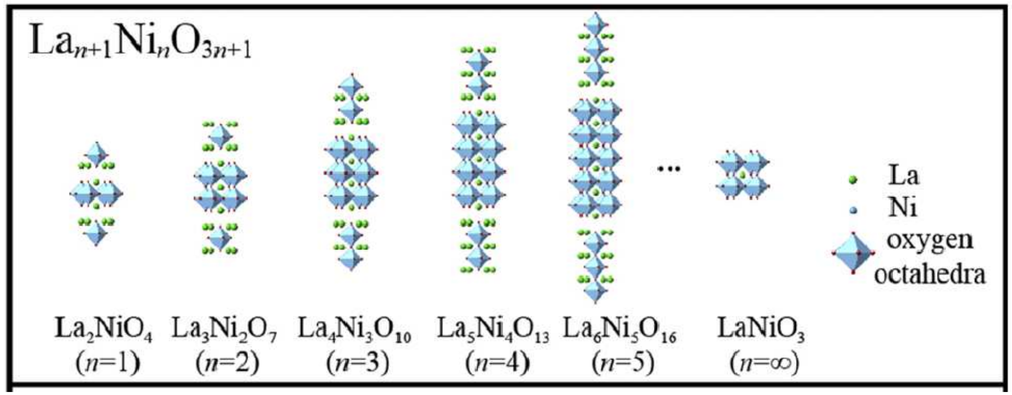
Nickelates: basics

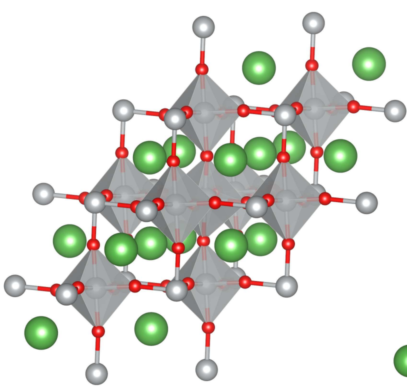
[FL, Elect. Struc. 4, 015005 (2022)]

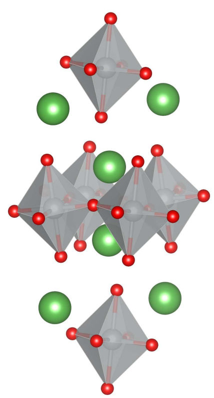
Ni-e_g frontier orbitals

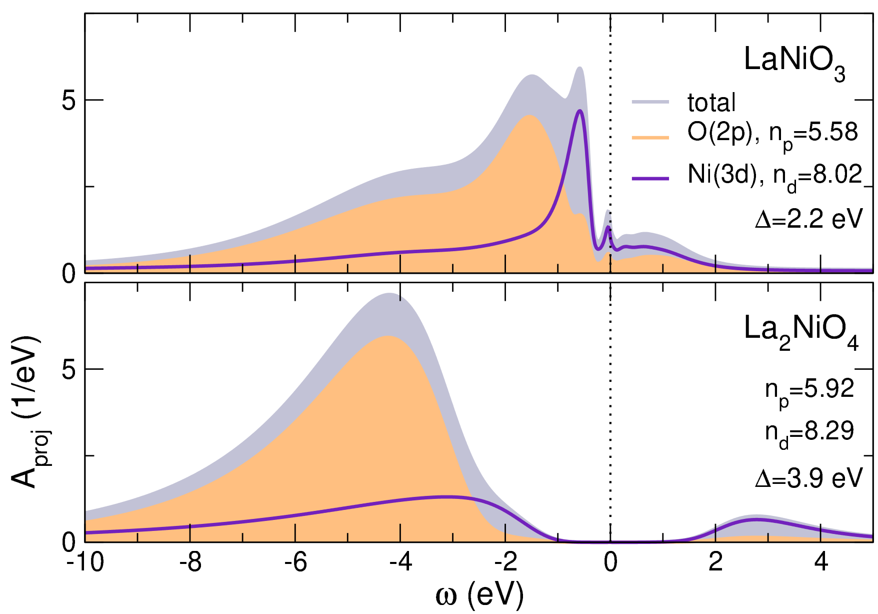


Ruddlesden-Popper compounds









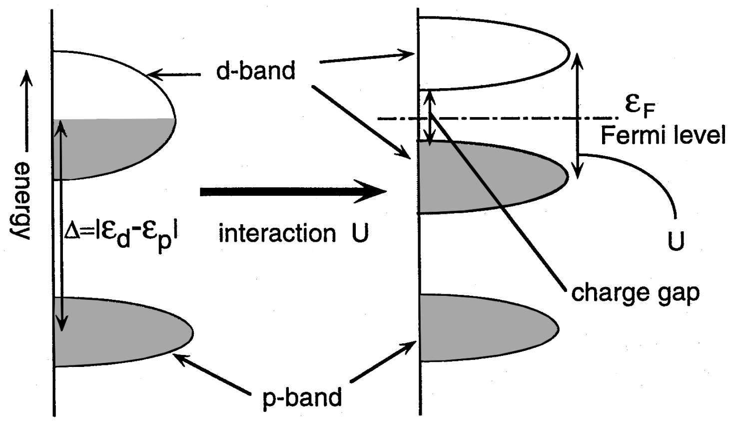
$n \to \infty$
metal

$n = 1$
Mott-charge-transfer insulator

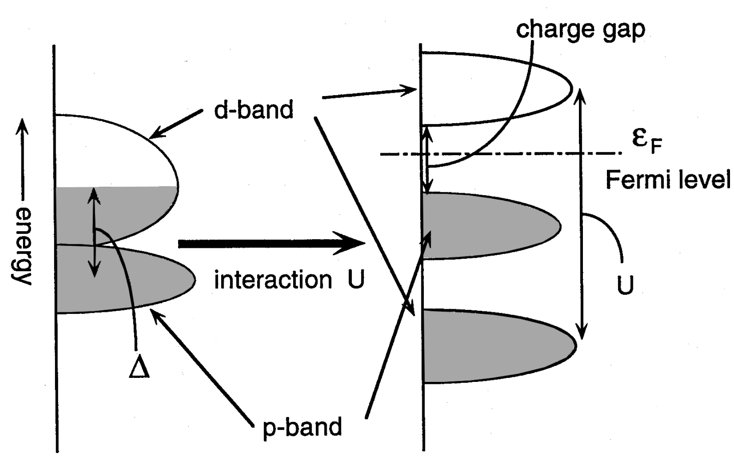
Nickelates: Mott-Hubbard vs. charge-transfer

[Mattheis, PRB 5, (1972)]
[Zaanen, Sawatzky, Allen, PRL 55, (1985)]
[Imada, Fujimori, Tokura, RMP 70, (1998)]

Hubbard U vs. charge-transfer energy $\Delta = \varepsilon_d - \varepsilon_p$



(a) Mott-Hubbard Insulator



(b) Charge Transfer Insulator

formal $3d^n$

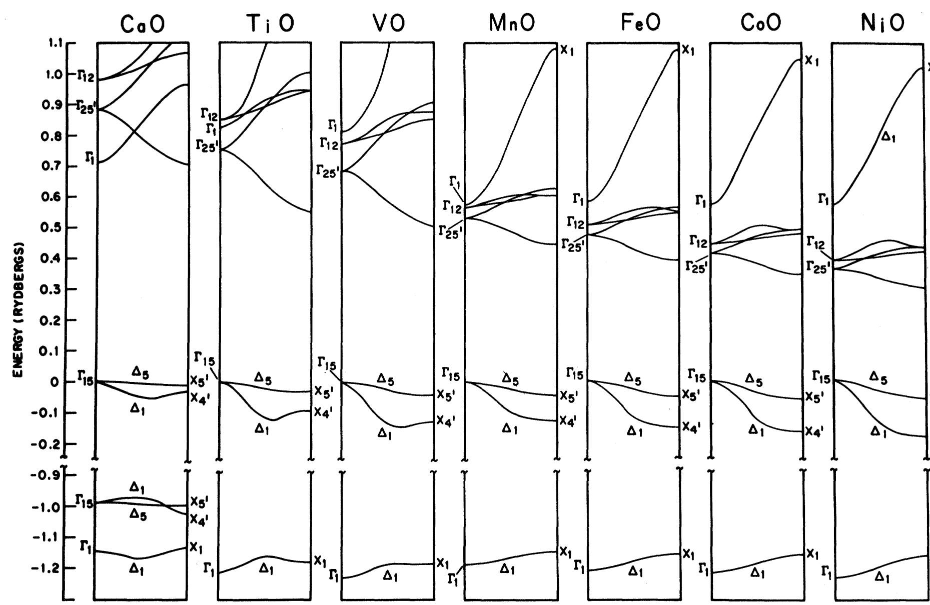


FIG. 2. APW energy-band results for the $3d$ transition-metal oxides plotted along the Δ direction of the Brillouin zone.

formal $3d^{n-1}$

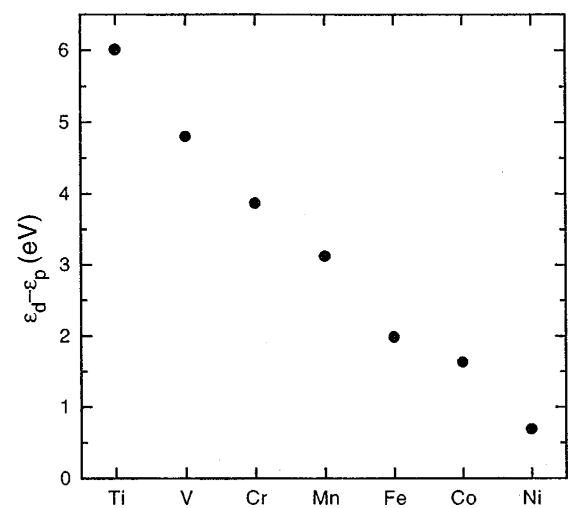
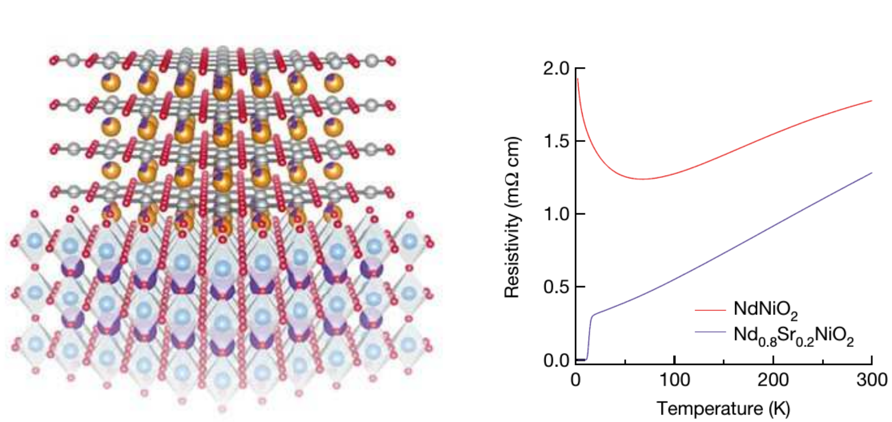


FIG. 59. Variation of $\varepsilon_d - \varepsilon_p$ for the $LaMO_3$ series with the metal M element. From Mahadevan *et al.*, 1996.

Superconducting nickelates: materials

$9-\delta$



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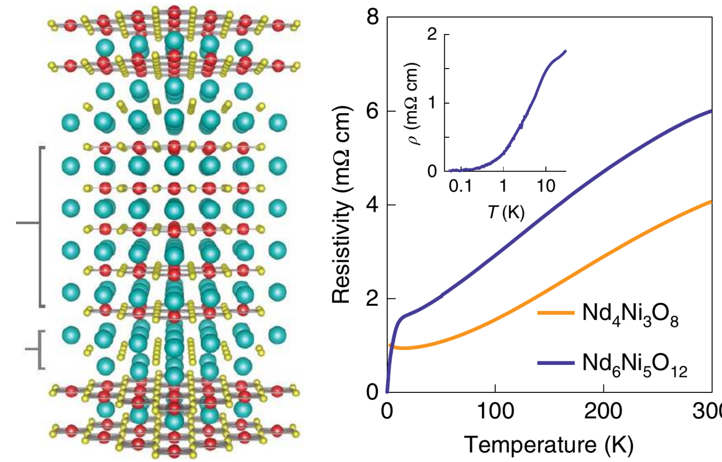
[Li et al., Nature 572, 624 (2019)]



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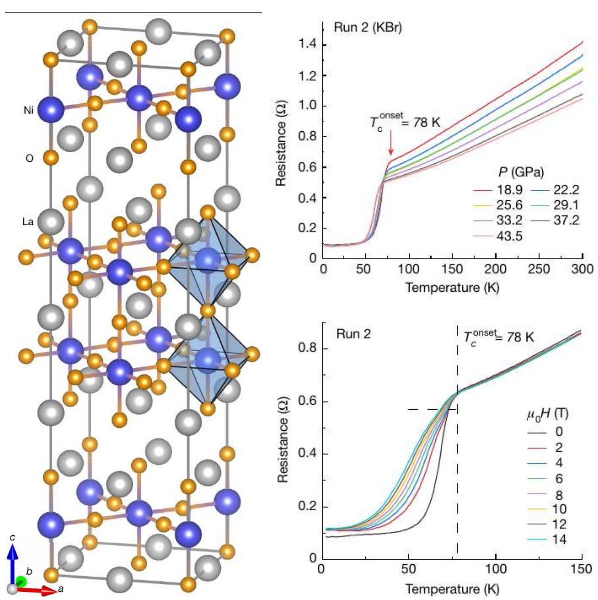

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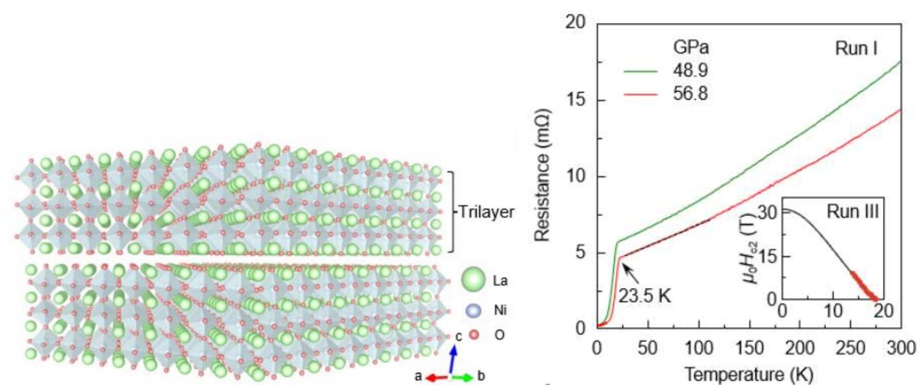
[Sun et al., Nature 621, 493 (2023)]



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[Zhu et al., Nature 631, 531 (2024)]
[Zhang et al., arXiv:2311.07423 (2023)]

$3d^{9-\delta}$ superconductors: initial work on $LaNiO_2$

[Crespin et al,. J. Chem. Soc., Faraday Trans. 2 79, 1181 (1983)]
[Levitz et al., J. Chem. Soc., Faraday Trans. 2 79, 1195 (1983)]
[Hayward et al., J. Am. Chem. Soc. 121, 8843 (1999)]

experiment

$$LaNiO_3 + xH_2 \rightarrow LaNiO_{3-x} + xH_2O.$$

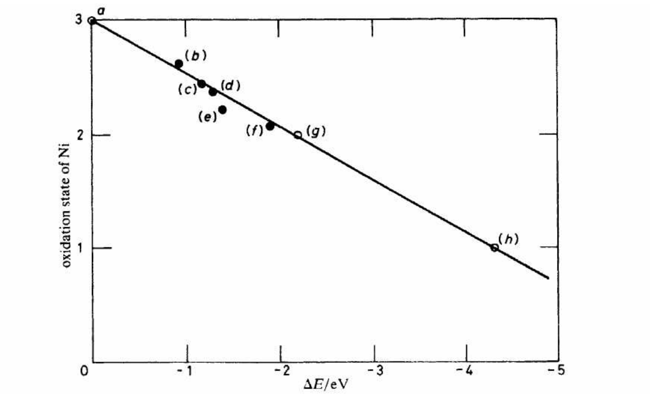


Fig. 6. Oxidation state of nickel in different phases formed during reduction of $LaNiO_3$ plotted against the energy shift of the absorption edge using $LaNiO_3$ as a standard: (*a*)$LaNiO_3$, (*b*) $LaNiO_{2.8}$, (*c*) $LaNiO_{2.7}$, (*d*) $LaNiO_{2.68}$, (*e*) $LaNiO_{2.6}$, (*f*) La_2NiO_4, (*g*) $LaNiO_{2.5}$ and (*h*) $LaNiO_2$.

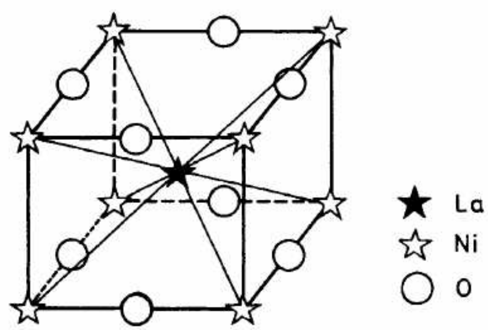
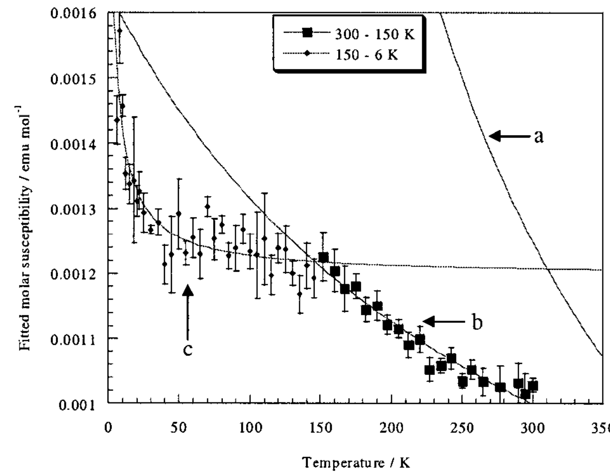
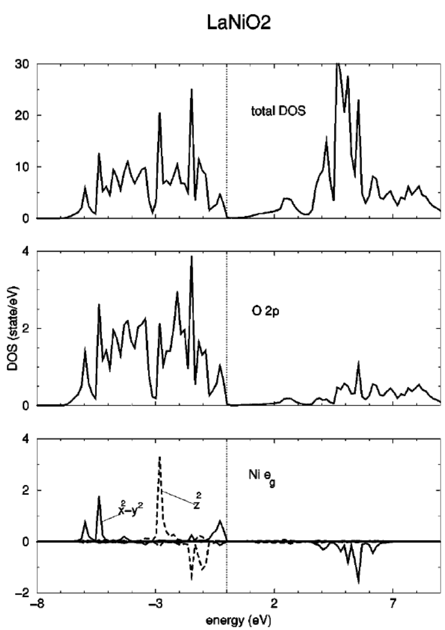


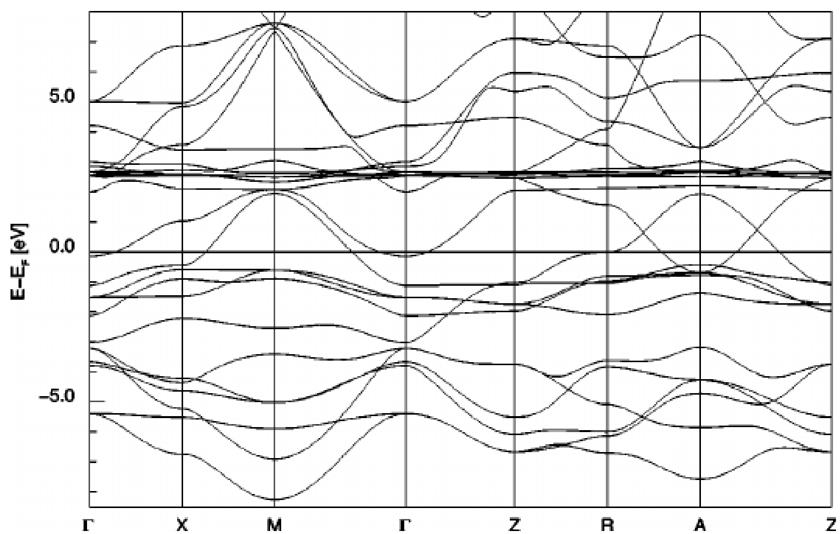
Fig. 1. Proposed structure of $LaNiO_2$.

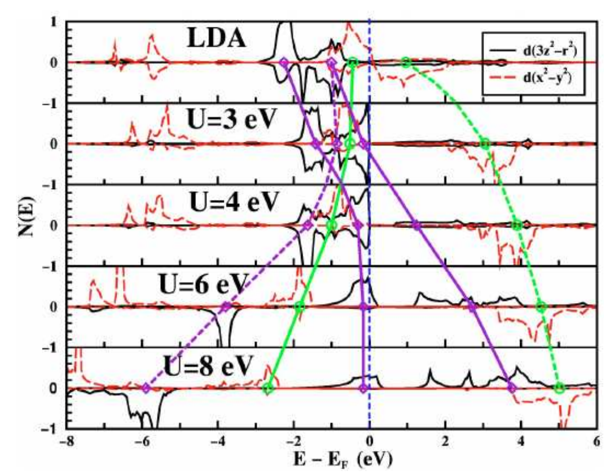


[Choisnet et al., J. Phys. Chem. Solids 57, 1839 (1996)]
[Anisimov, Bukhvalov, Rice, PRB 59, 7901 (1999)]
[Lee and Pickett, PRB 70, 165109 (2004)]

theory







Experimental finding: finally, a superconducting nickelate!

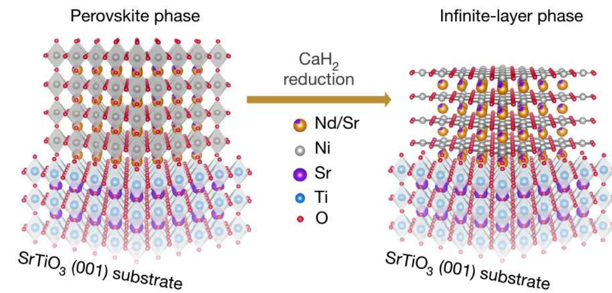
[Li et al., Nature 572, 624 (2019)]
[Lee et al., APL Materials 8, 041107 (2020)]

LETTER

https://doi.org/10.1038/s41586-019-1496-5

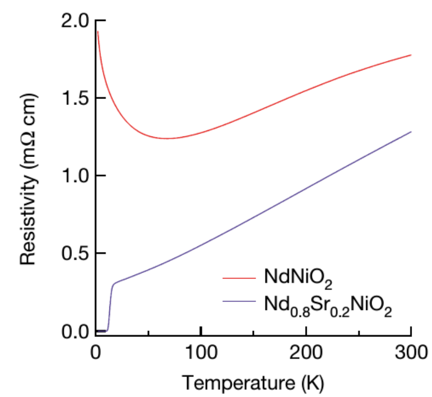
Superconductivity in an infinite-layer nickelate

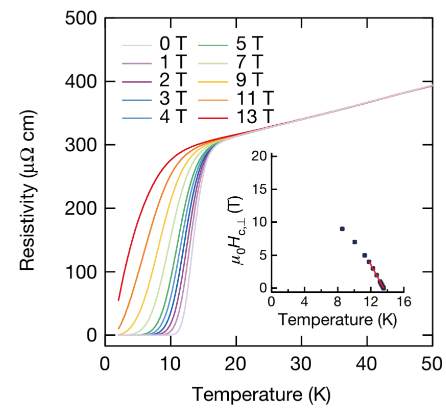
Danfeng Li[1,2]*, Kyuho Lee[1,3], Bai Yang Wang[1,3], Motoki Osada[1,4], Samuel Crossley[1,2], Hye Ryoung Lee[1,4], Yi Cui[1,4], Yasuyuki Hikita[1] & Harold Y. Hwang[1,2]*

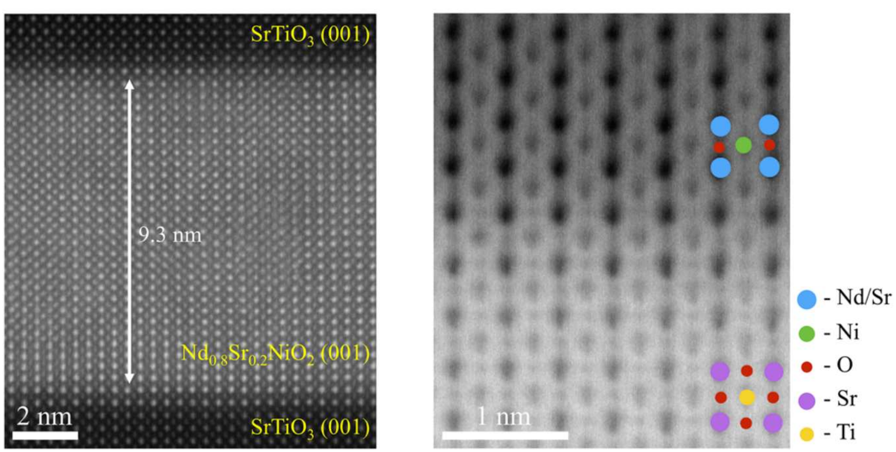


- thin films of infinite-layer $NdNiO_2$ on $SrTiO_3$
- film thickness ~ 10 nm
- resistivity upturn below $T = 70$ K
- 20% Sr (hole) doping: superconducting below $T \sim 10$ K
  - critical current density $J_c \sim 170$ kA cm^{-2}
  - zero-T coherence length ~ 3.25 nm, type-II superconductor
  - recent data: Pauli-limit violation in doped $LaNiO_2$ and $PrNiO_2$?

[Chow et al., arXiv:2204.12606 (2022)] [B. Y. Wang, et al., arXiv:2205.15355 (2022)]



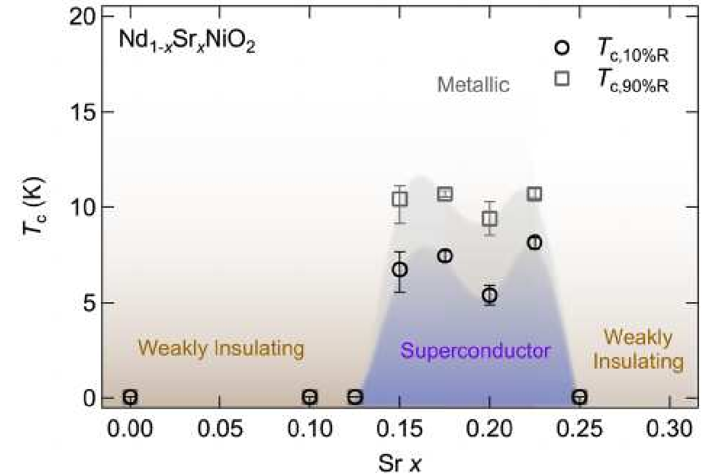


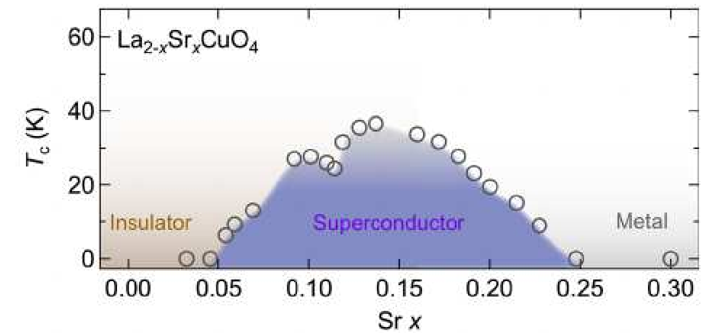


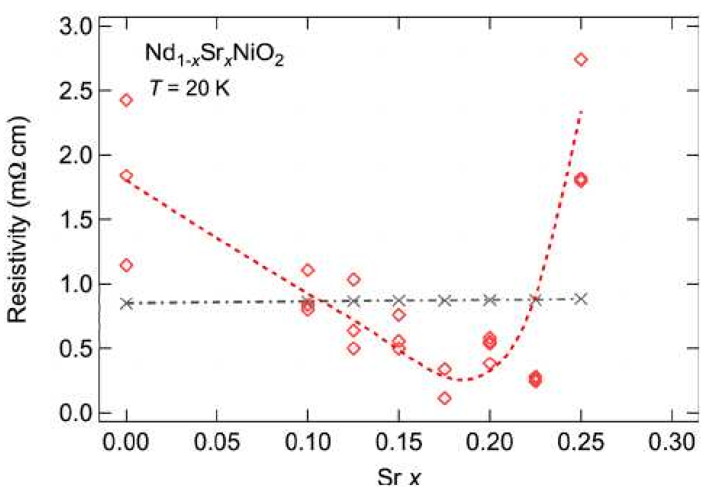
$3d^{9-\delta}$ superconductors: some experimental data

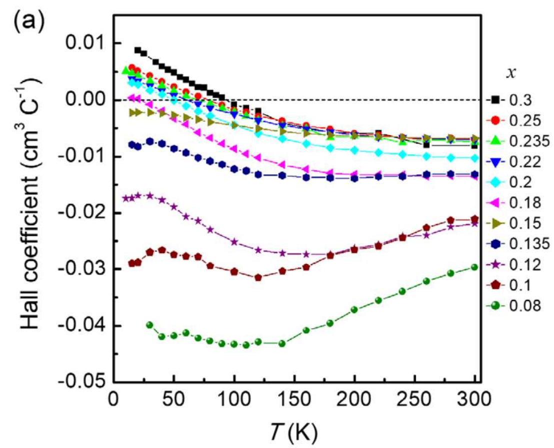
[Li et al., PRL 125, 027001 (2020)]
[Zheng et al., PRL 125, 147003 (2020)]

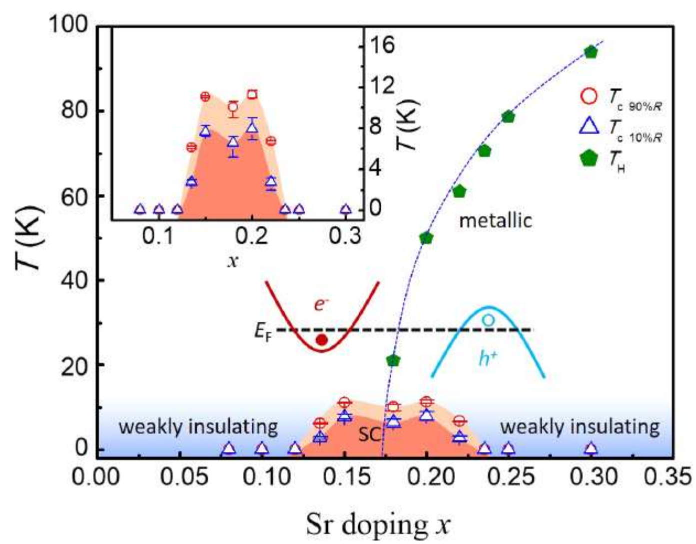
- superconducting “dome” for $0.125 < x < 0.25$
- reminiscent of dome in layered cuprates
- but: weakly-insulating on both sides of superconducting region
- Hall data suggests two-band picture with doping









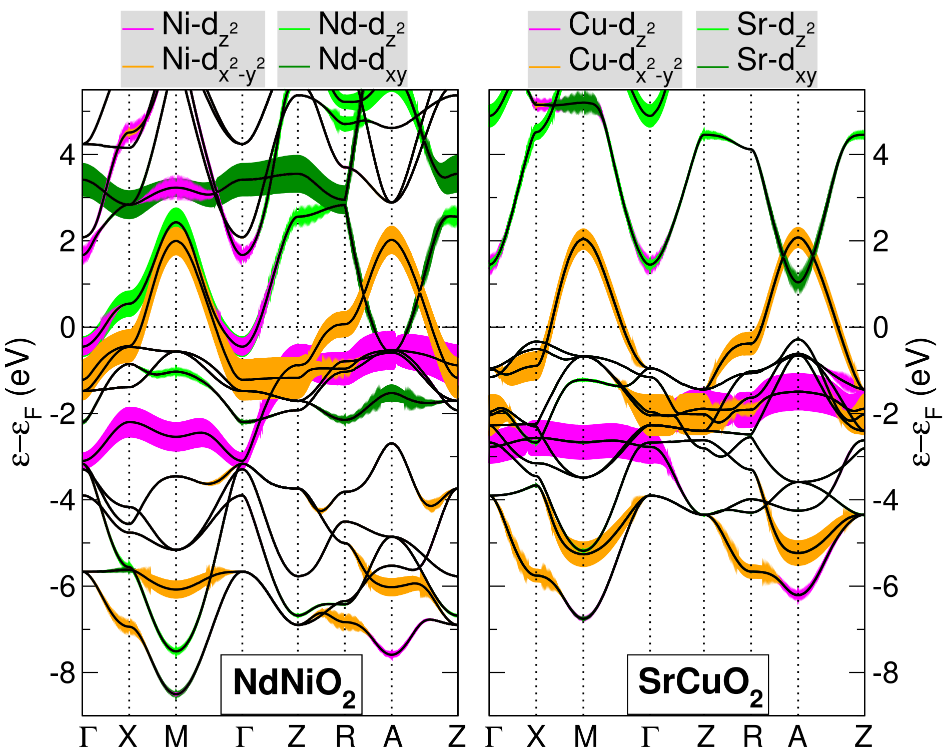


$3d^{9-\delta}$ superconductors: theory concepts

e.g. reviews
[Norman, Physics 13, 85 (2020)]
[Botana, Bernadini, Cano, JETP 132, 618 (2021)]

- indeed cuprate(-like) physics: mainly Ni-$d_{x^2-y^2}$ driven e.g. [Nomura et al. PRB 100, 205138 (2019)] [Wu et al., PRB 101, 060504(R) (2020)][Si et al., PRL 124, 166402 (2020)] [Karp et al. PRX 10, 021061 (2020)][Been et al., PRX 11, 011050 (2021)]
- Kondo-based physics involving Ni-$d_{x^2-y^2}$ and Nd orbitals
  e.g. [Zhang et al. PRBB 101, 020501(R) (2020)]
- dominant interface physics (induced by $SrTiO_3$ substrate)
  e.g. [Geisler and Pentcheva, PRB 102, (020502(R)] [Zhang et al., PRB 102, 195117 (2020)]
- Ni multiorbital physics (Mott vs. Hund (vs. Kondo))
  e.g. [Werner and Hoshino, PRB 101, 041104(R) (2020)] [Wang et al., PRB 102, 161118(R)][Choi et al., PRR 2, 033445 (2020)]

s from
ity functional theory



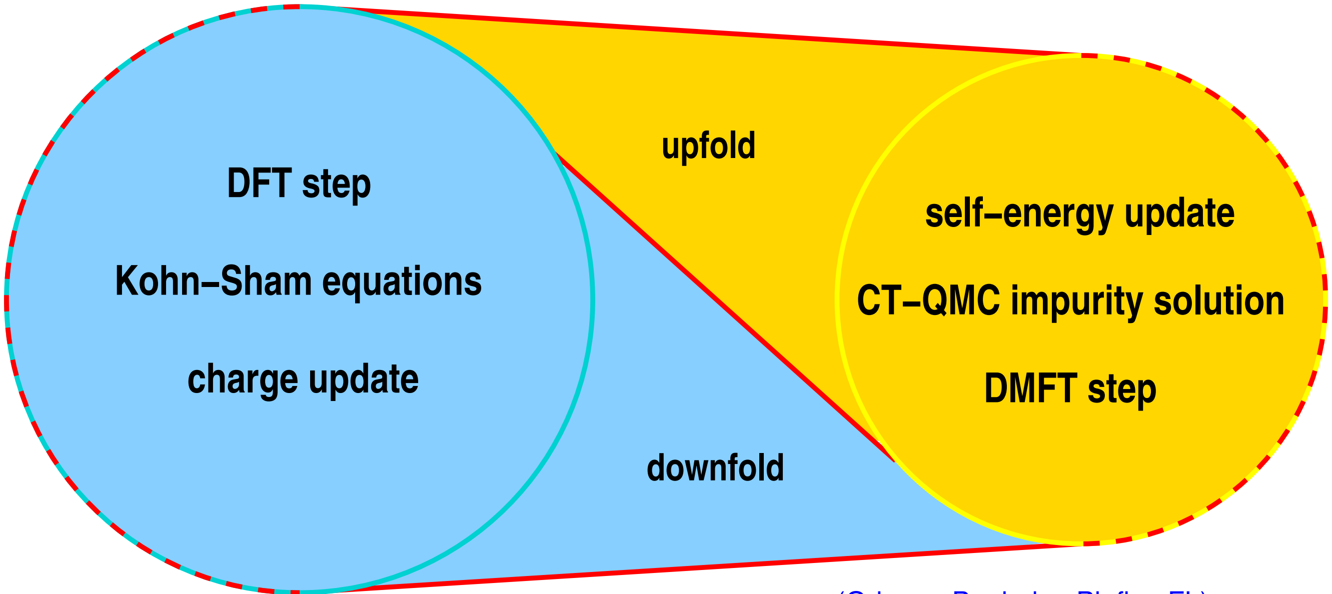
e.g. DFT work
[Botana and Norman, PRX 10, 011024 (2020)]
[FL, PRB 101, 081110(R) (2020)]

harge-transfer energy $\Delta = \varepsilon_d - \varepsilon_p$ $\Delta(\mathrm{NdNiO_2}) \sim 5.0$ eV $\Delta(\mathrm{SrCuO_2}) \sim 1.3$ eV

Theoretical approach: realistic many-body

[Anisimov, Poteryaev, Korotin et al., JPCM 9, 7359 (1997)]
[Held, Keller, Eyert et al., PRL 86, 5345 (2001)]
[Minár, Chioncel, Perlov, at al., PRB 72, 045125 (2005)]
[FL, Georges, Poteryaev et al., PRB 74, 125120 (2006)]
[Lichtenstein and Katsnelson, PRB 57, 6884 (1998)]
[Pavarini, Biermann, Poteryaev et al., PRL 92, 176403 (2004)]
[Kotliar, S. Y. Savrasov, K. Haule et al, RMP 78, 865 (2006)]
[Grieger, Piefke, Peil and FL, PRB 86, 155121 (2012)] . . . , . . . , . . .

Density Functional Theory (DFT) + Dynamical Mean–Field Theory (DMFT)



(Grieger, Boehnke, Piefke, FL)

- DFT part : mixed-basis pseudopotential

[Meyer, Elsässer, Lechermann and Fähnle, MPI for Metals Research]

- DMFT impurity solver : hyb-CT-QMC (TRIQS code)

[Parcollet et al., Comput. Phys. Commun. 196, 398 (2015)]
[Seth et al, Comput. Phys. Commun. 200, 274 (2016)]

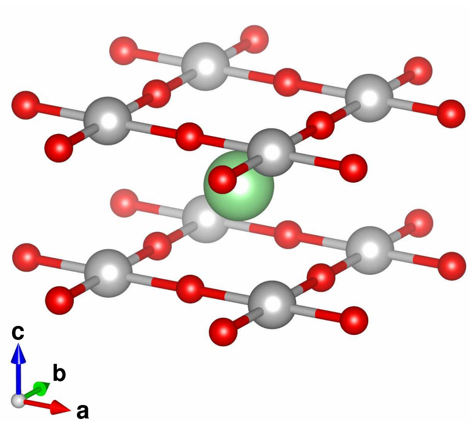
- charge self-consistency
- 1-5 local correlated orbitals
- up to 200-atom unit cells
- inclusion of correlations on oxygen

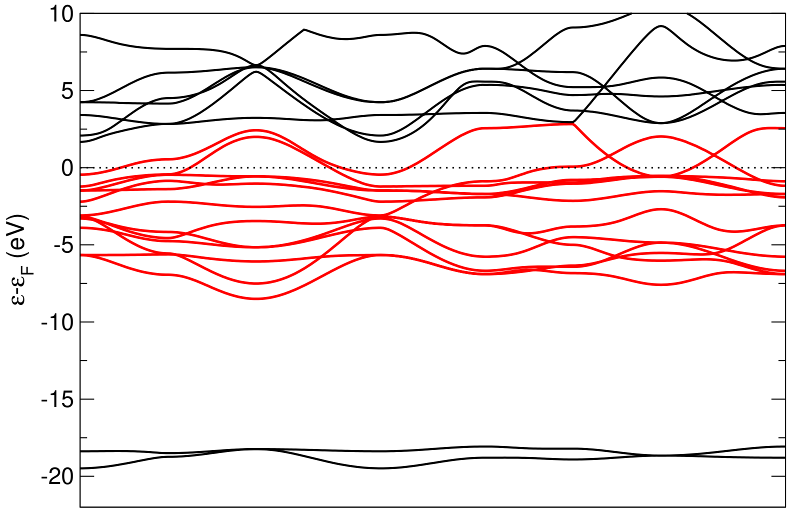
Theoretical approach: settings for nickelates

- correlated subspace: Ni$(3d)$ from projected-local orbitals
- rotational-invariant 5-orbital Slater-Hamiltonian: $U = 10$ eV, $J_{\rm H} = 1$ eV
- self interaction correction (SIC) for oyxgen: $w_p = 0.8$
- fully-localized double counting correction

setting fixed for <u>all</u> nickelates

example: $NdNiO_2$

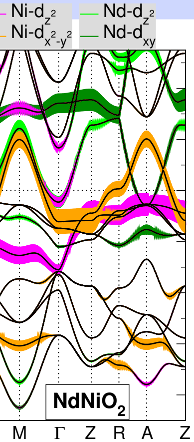


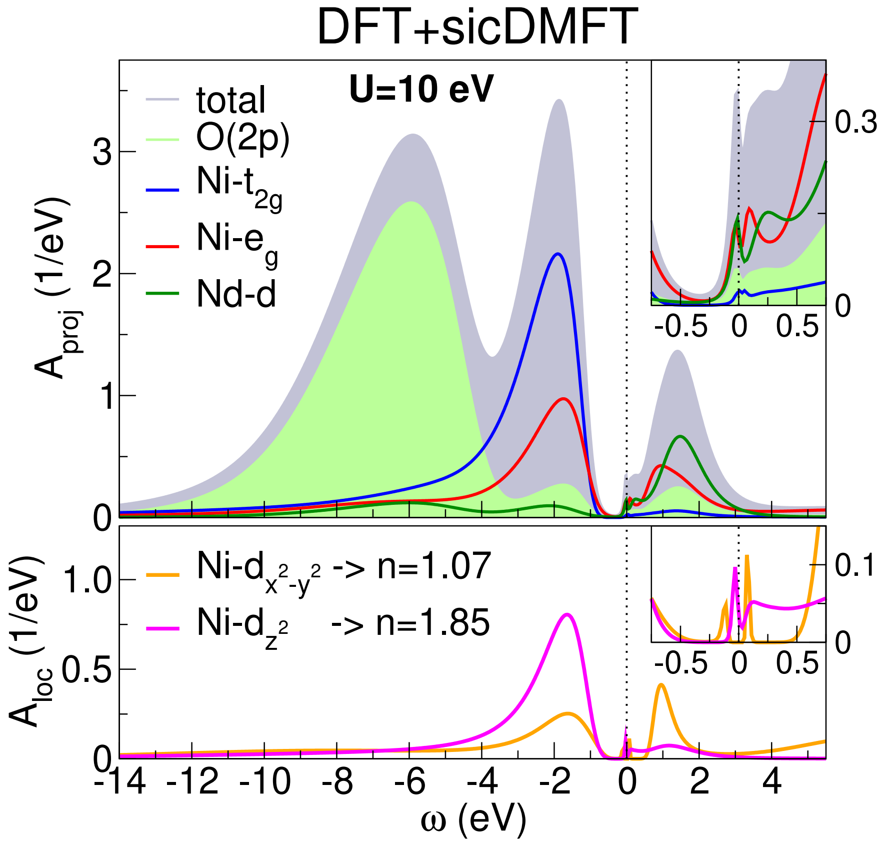


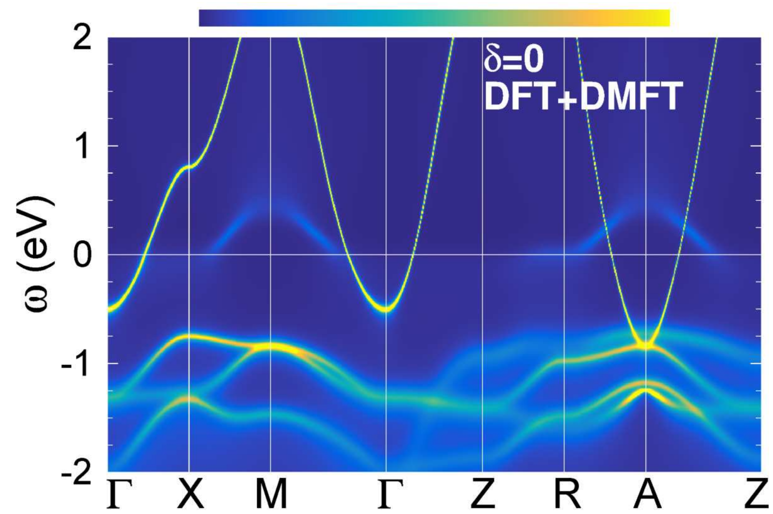
12 Kohn-Sham bands for projection onto Ni$(3d)$

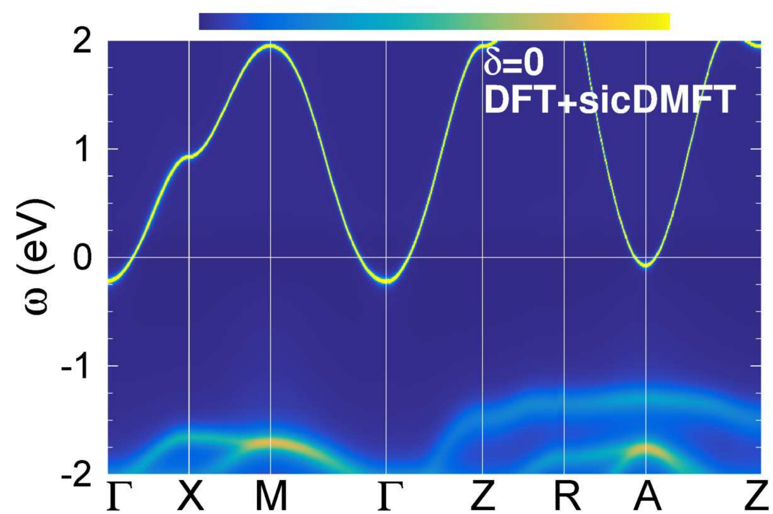
$3d^{9-\delta}$ superconductors: $NdNiO_2$

[FL, PRB 101, 081110(R) (2020)]
[FL, PR Mat. 5, 044803 (2021)]







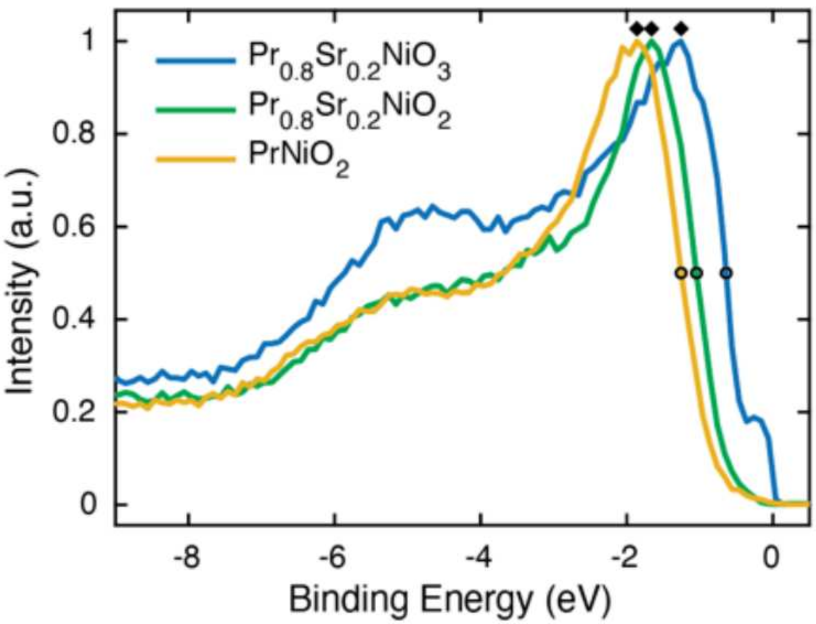


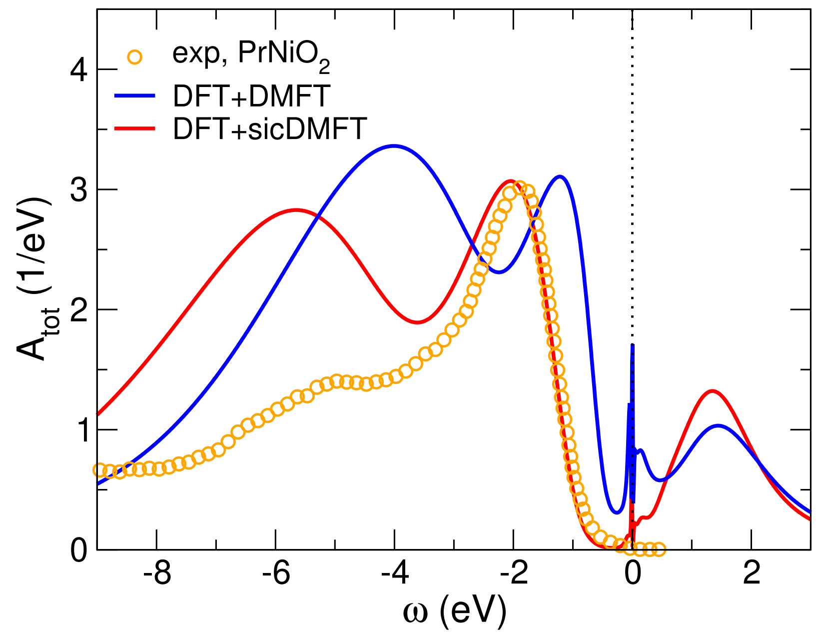
- Ni-$d_{x^2-y^2}$ (nearly) Mott-insulating, Ni-d_{z^2} hybridizes with Nd-d_{z^2} and forms Γ-pocket of self-doping band
- important: oxygen-based correlations (brought in by SIC), render the system highly correlated;
  $NdNiO_2$ is close to Mott criticality!

$3d^{9-\delta}$ superconductors: experimental spectrum

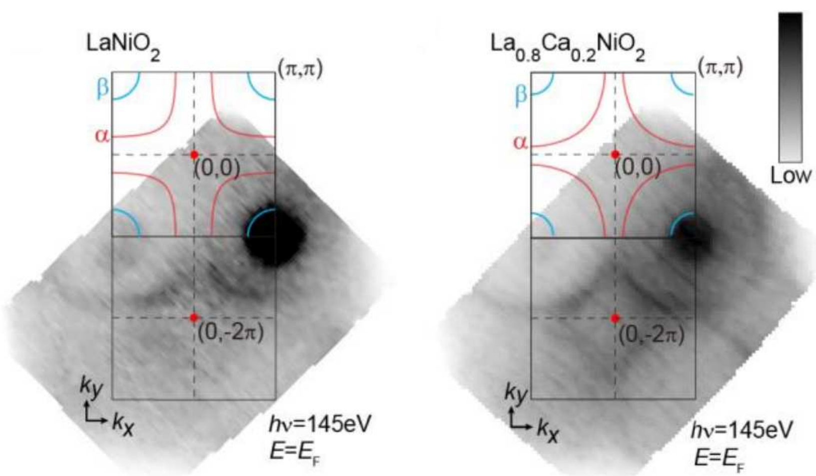
[FL, PRB 101, 081110(R) (2020)]
[FL, PR Mat. 5, 044803 (2021)]
[Chen et al., Matter 5, 1806 (2022)]
[Ding et al., Nat. Sci. Rev. 194 (2024)]
[Sun et al., arxiv:2403.07344 (2024)]

angle-integrated photoemission



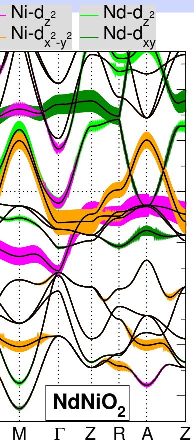


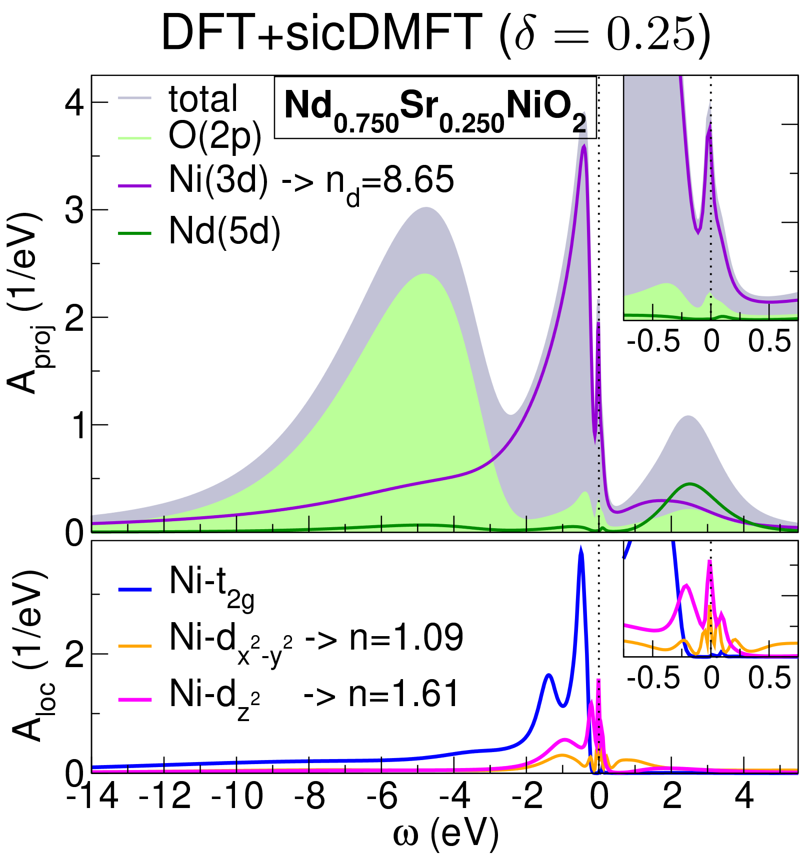
very recent ARPES

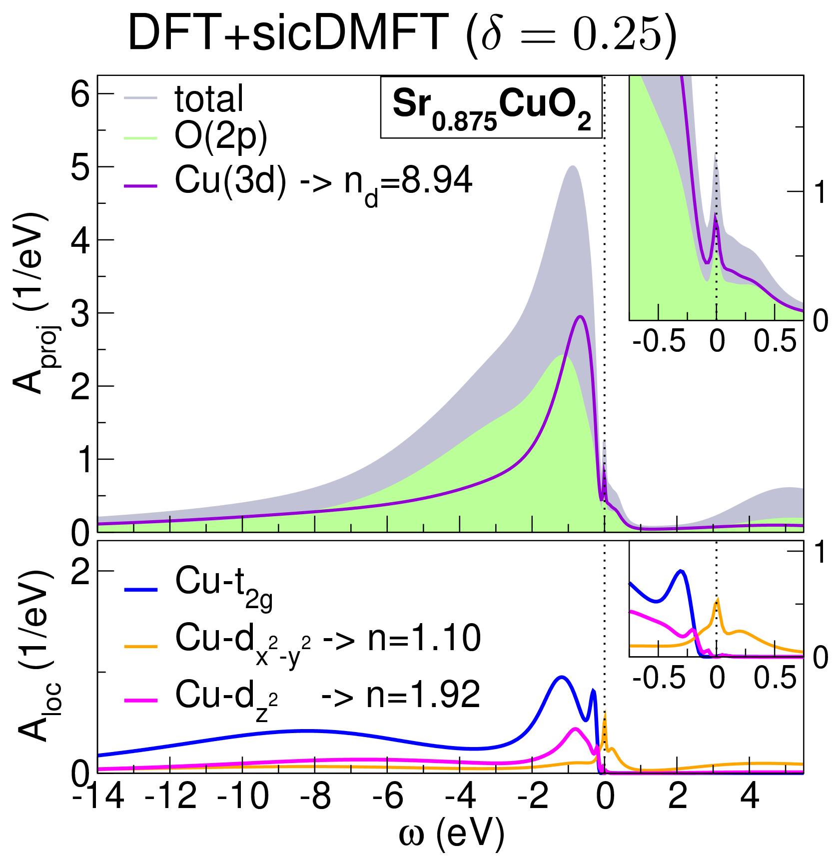


$3d^{9-\delta}$ superconductors: doped $NdNiO_2$

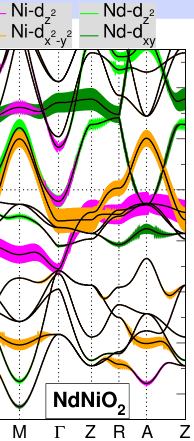
[FL, PRB 101, 081110(R) (2020)]
[FL, PR Mat. 5, 044803 (2021)]





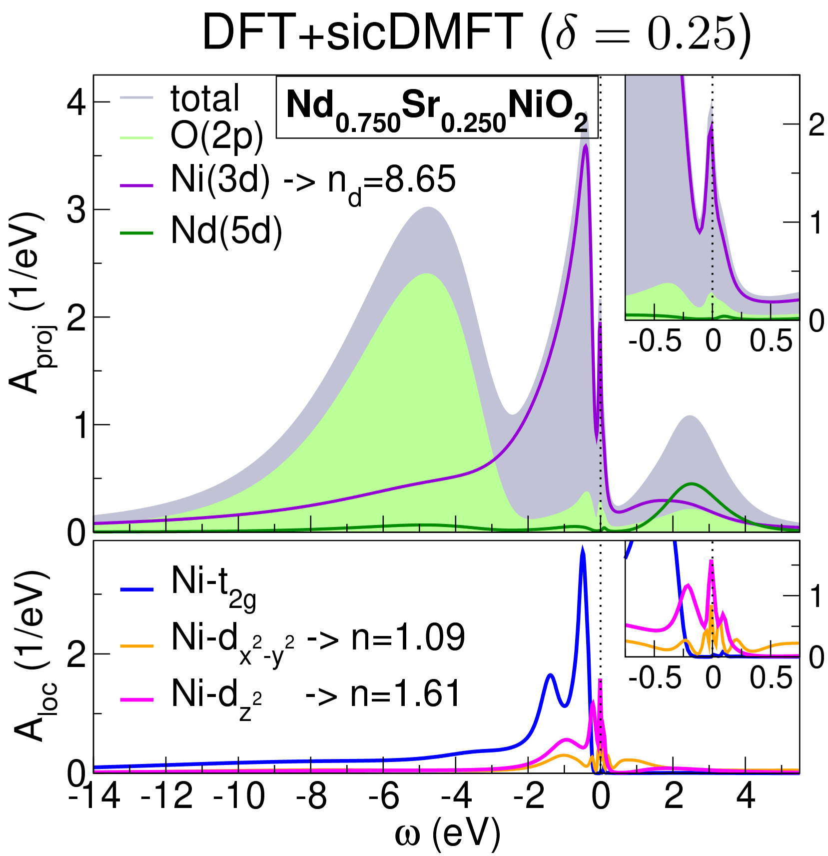


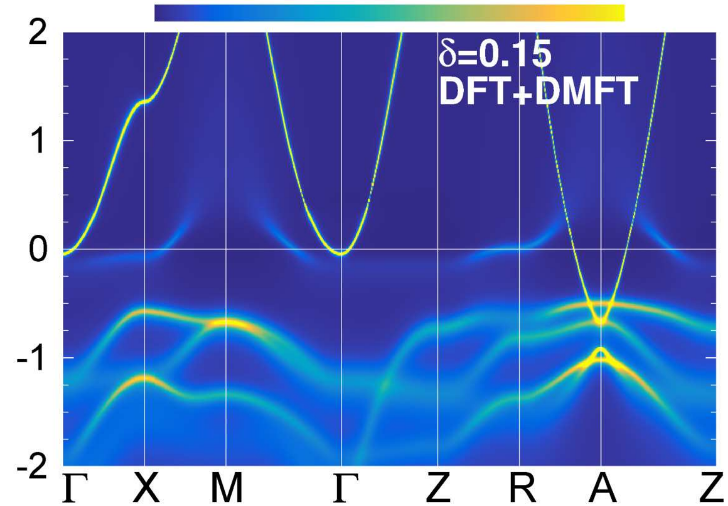
- no strong Zhang-Rice physics

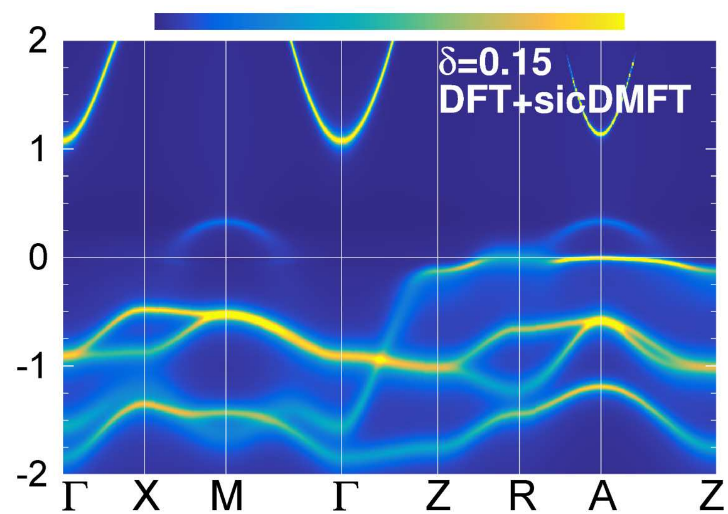


$3d^{9-\delta}$ superconductors: doped NdNiO$_2$

[FL, PRB 101, 081110(R) (2020)]
[FL, PR Mat. 5, 044803 (2021)]



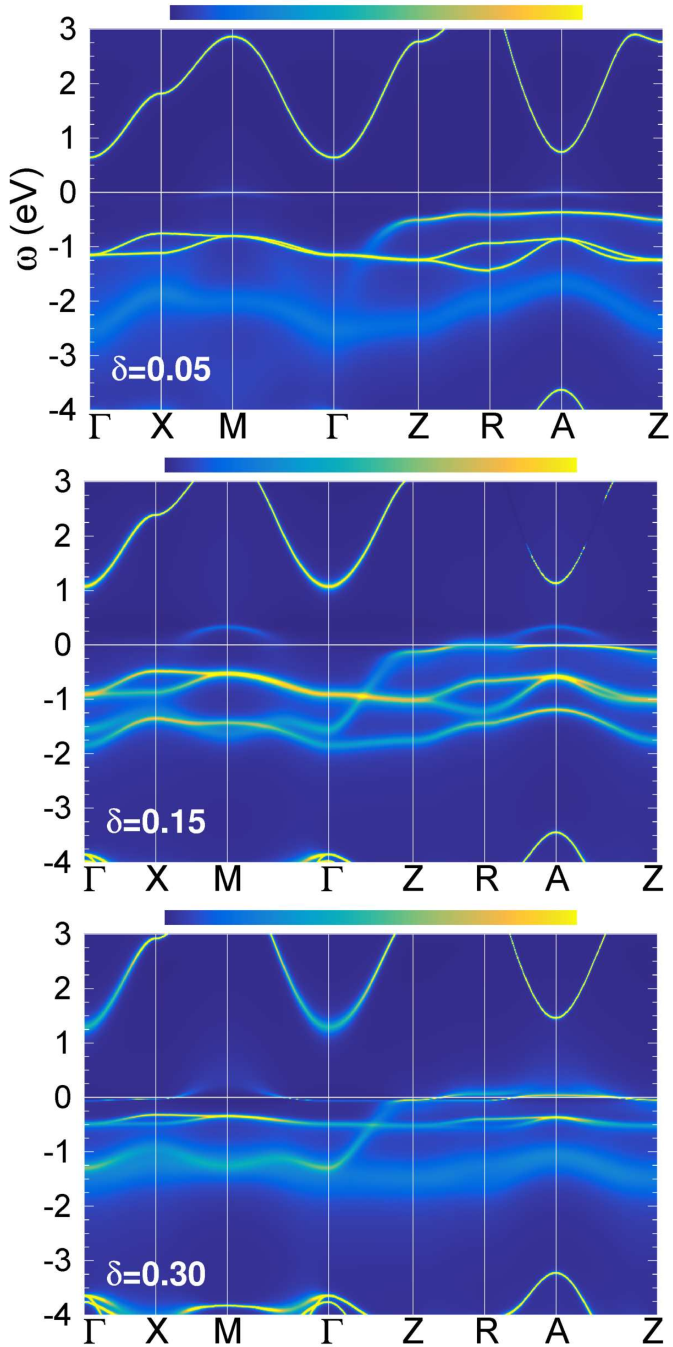


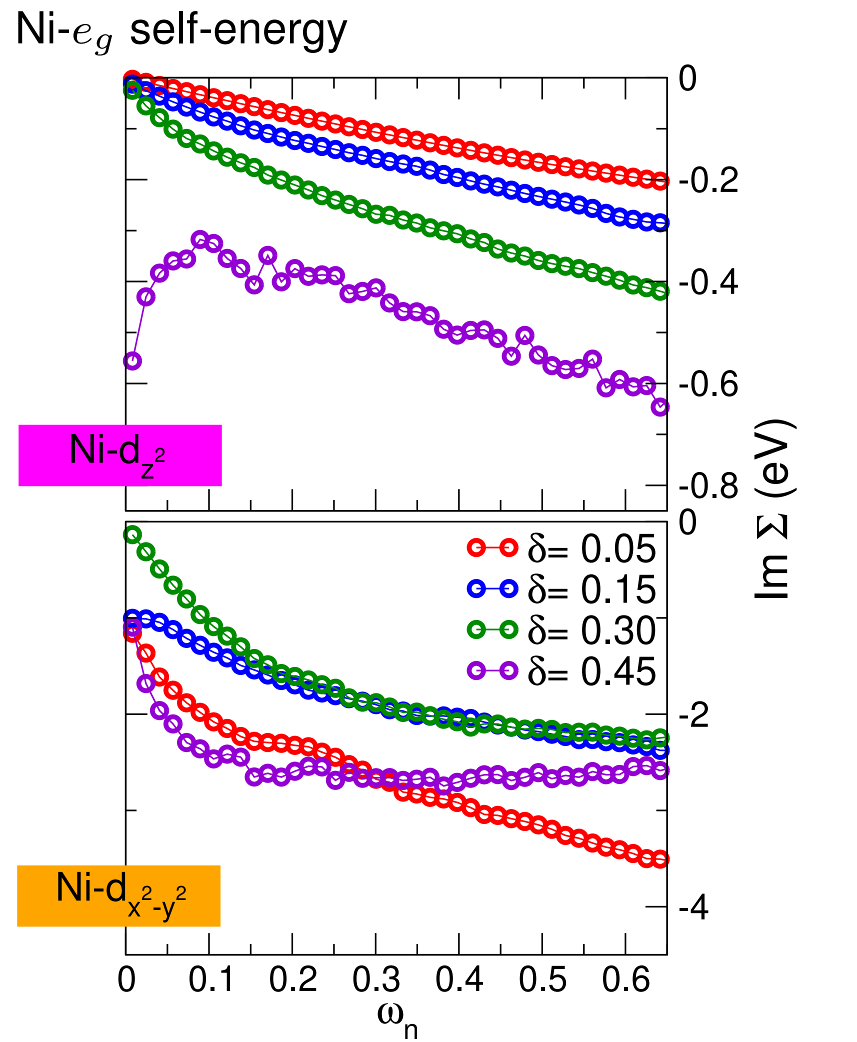


- no strong Zhang-Rice physics
- Ni-d_{z^2} flat-band part in $k_z = 0.5$ plane crosses Fermi level in the superconducting doping region when including oxygen-based correlations (brought in by SIC)

$3d^{9-\delta}$ superconductors: doped $NdNiO_2$

[FL, PR Mat. 5, 044803 (2021)]

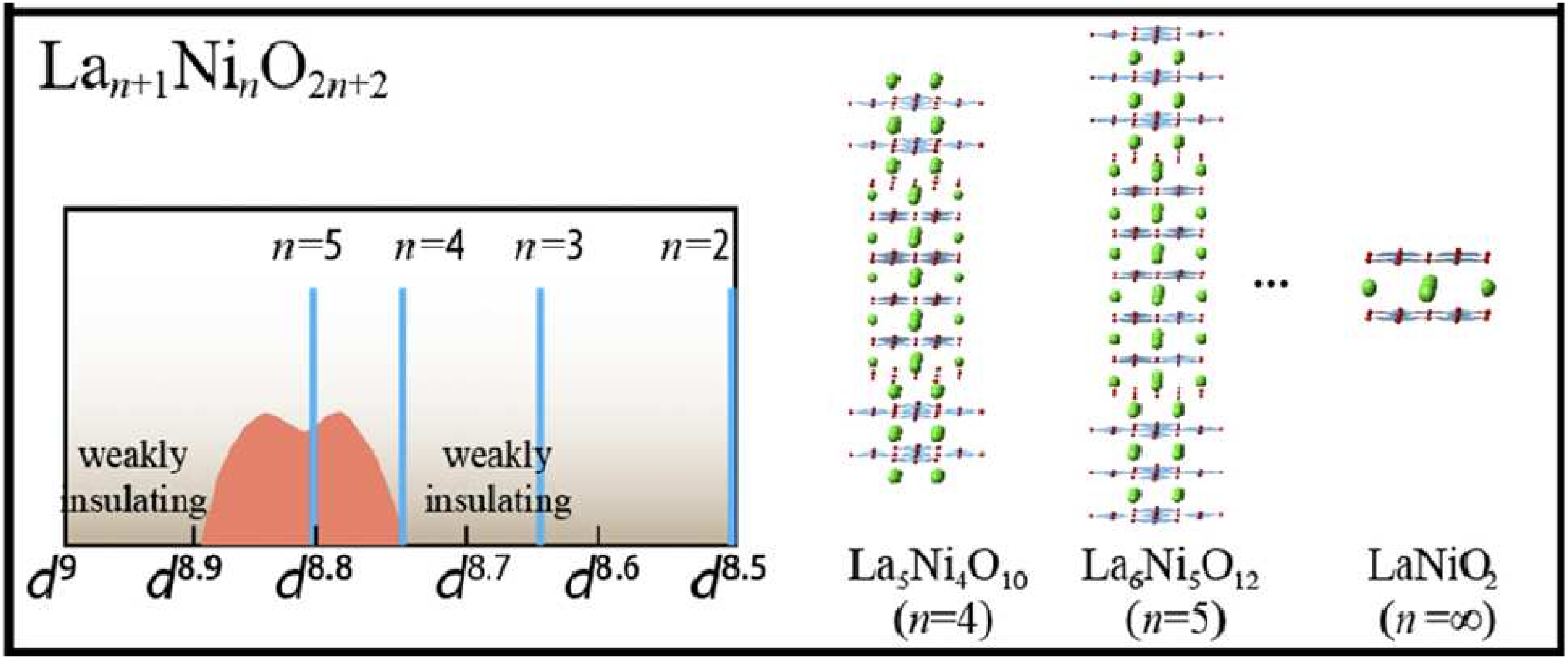




- Ni-d_{z^2} "brings" coherence to Ni-$d_{x^2-y^2}$

Multilayer nickelates: infinite-layer vs. multilayer

e.g. [Li et al., APL Materials 8, 091112 (2020)]



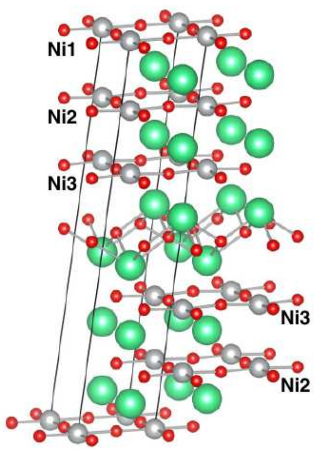
.

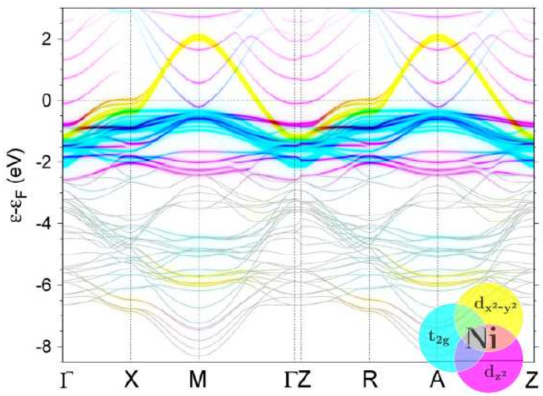
$3d^{9-\delta}$ superconductors: quintuple-layer $Nd_6Ni_5O_{12}$

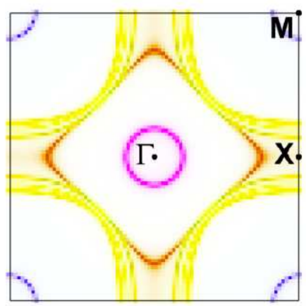
[Pan et al., Nat. Mater. 21, 160 (2022)]
[FL, PRB 105, 155109 (2022)]

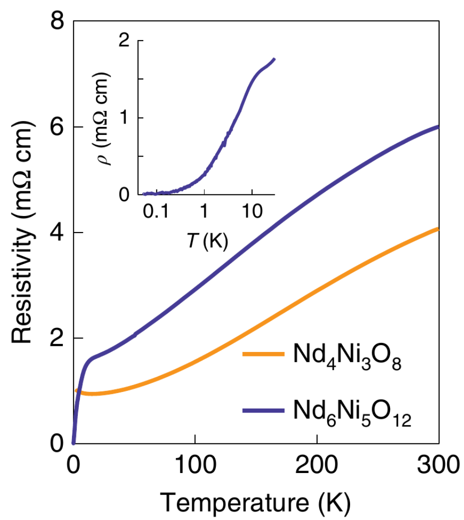
DFT picture

$Nd_6Ni_5O_{12}$

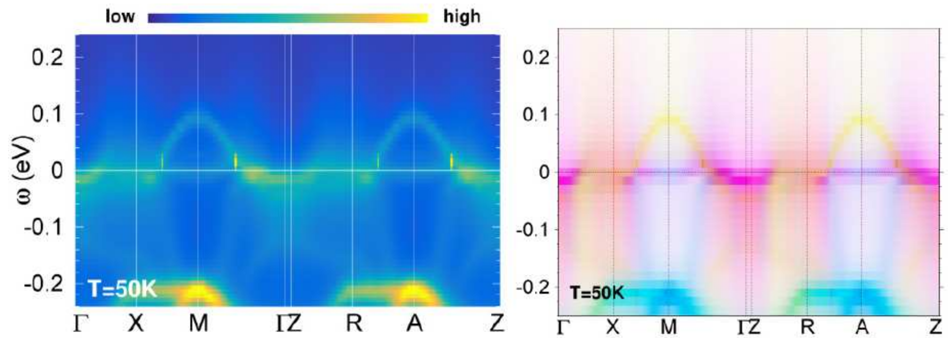


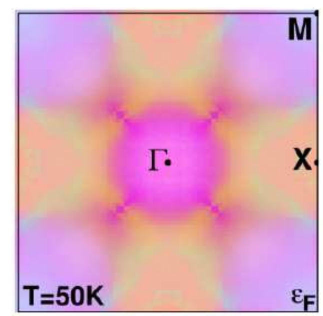






DFT+sicDMFT picture



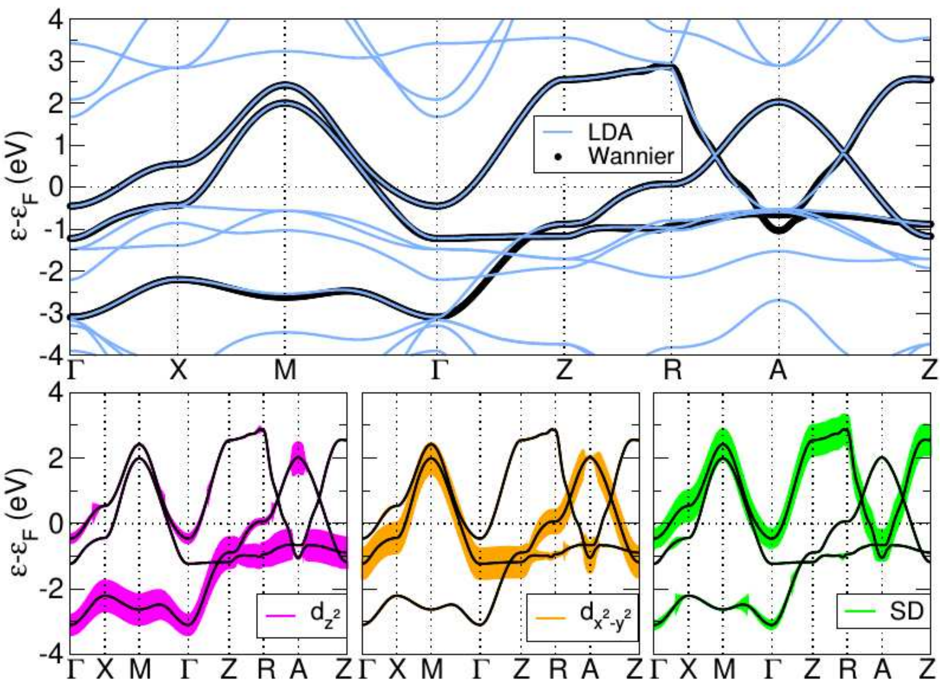


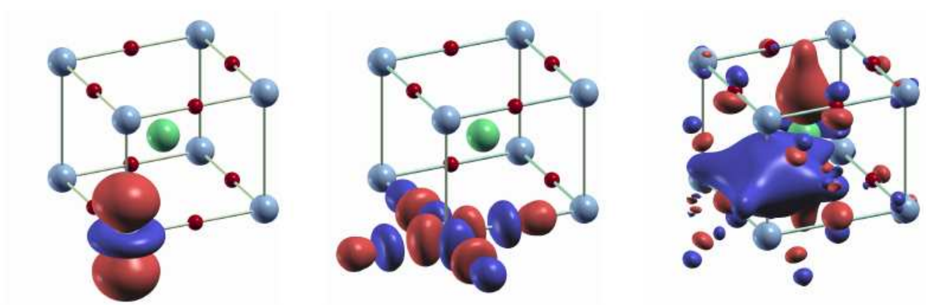
$m^*/m_{\rm DFT} \sim 25$

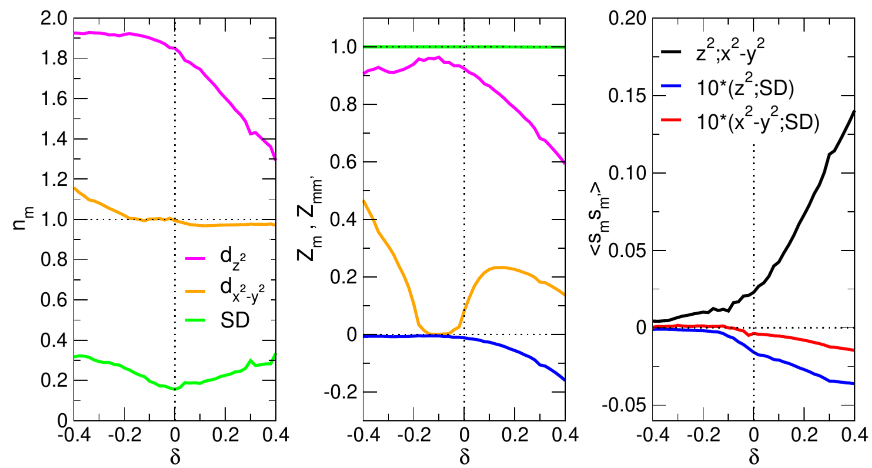
- similar $d_{x^2-y^2}$ vs. d_{z^2} dichotomy from Ni as in infinite-layer compounds
- Ni-d_{z^2} flat band appears essential for superconductivity

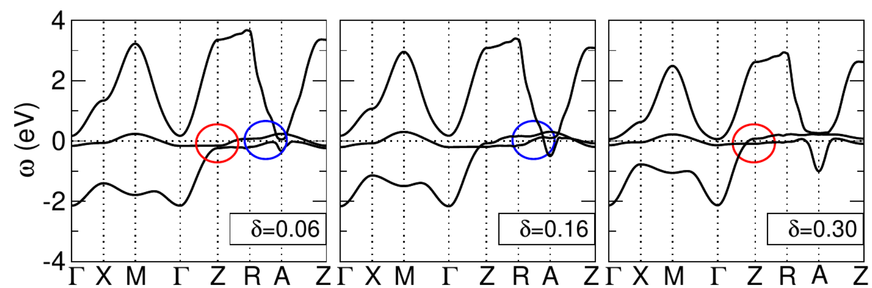
Theoretical picture: three-band model for $Nd_{1-x}Sr_xNiO_2$

[FL, PRX 10, 041002 (2020)]







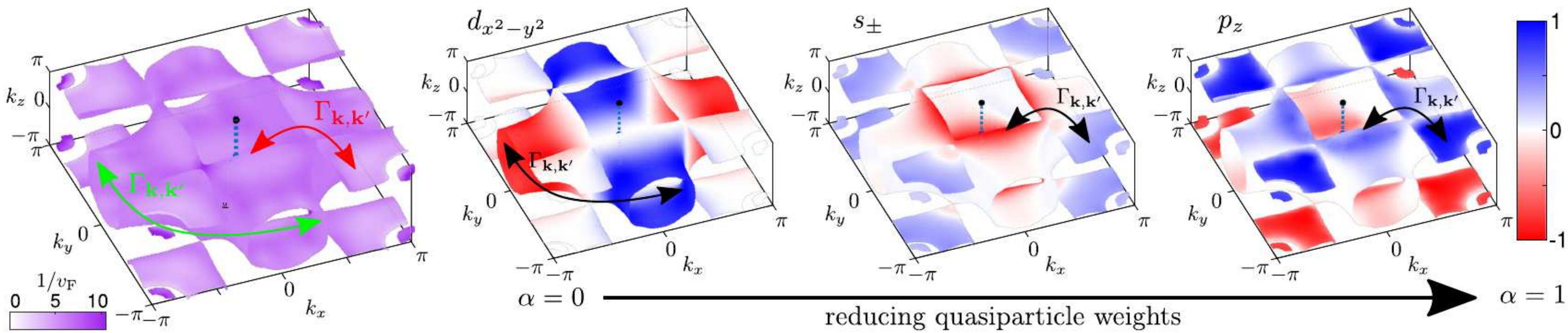


solved with rotational-invariant slave bosons (RISB)

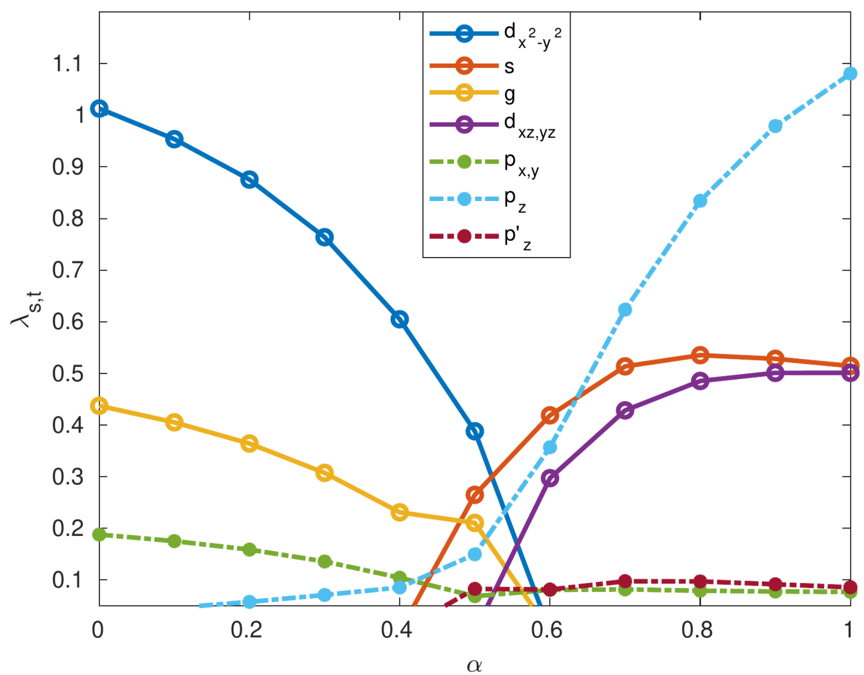
$3d^{9-\delta}$ superconductors: model superconductivity

[Kreisel et al., PRL 129, 077002 (2022)]

Leading SC instabilities from RPA spin fluctuations

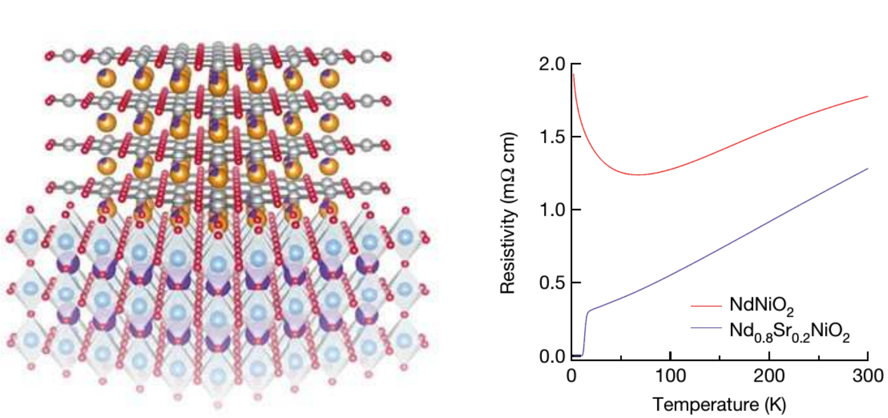


⇒ various competing instabilities, nodal $s_\pm$ a promising candidate



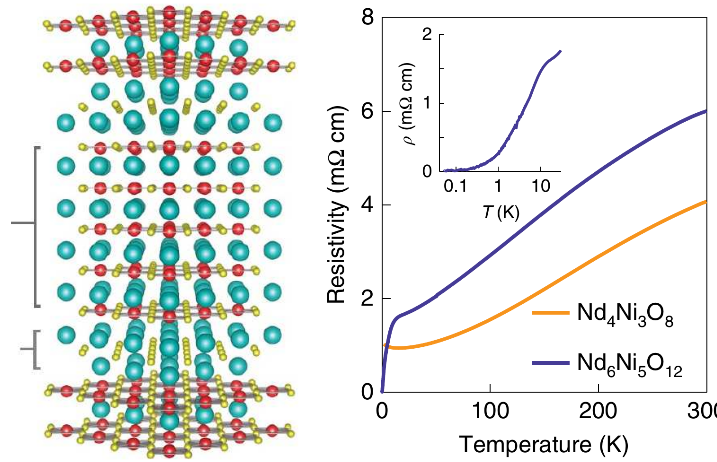
Superconducting nickelates: materials

$9-\delta$



infinite-layer Sr-doped RENiO$_2$, T_c ~10-25 K

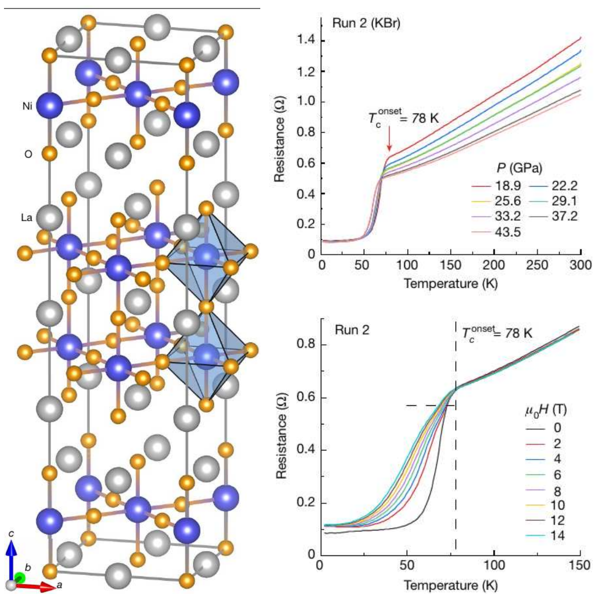
[Li et al., Nature 572, 624 (2019)]



quintuple-layer Nd$_6$Ni$_5$O$_{12}$, T_c ~10 K

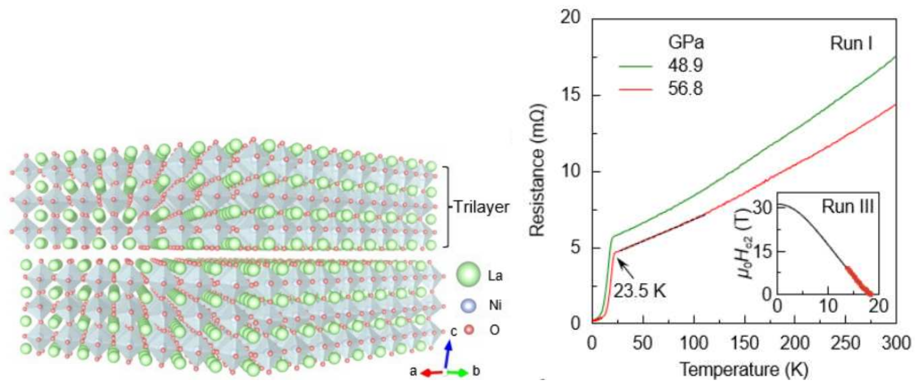
[Pan et al., Nat. Mater. 21, 160 (2022)]

$8\pm\delta$



high-pressure La$_3$Ni$_2$O$_7$, T_c ~65-80 K

[Sun et al., Nature 621, 493 (2023)]



high-pressure La$_4$Ni$_3$O$_{10}$, T_c ~23 K

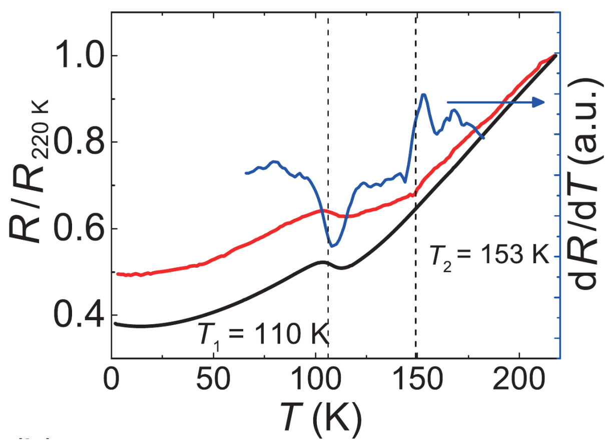
[Zhu et al., Nature 631, 531 (2024)]

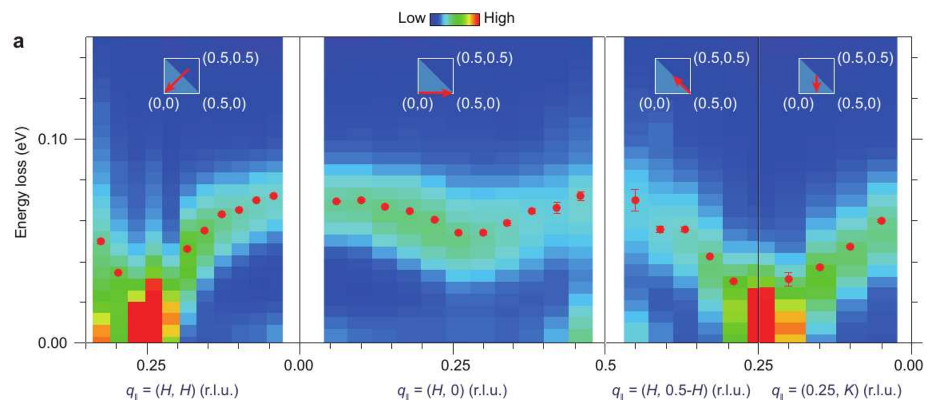
[Zhang et al., arXiv:2311.07423 (2023)]

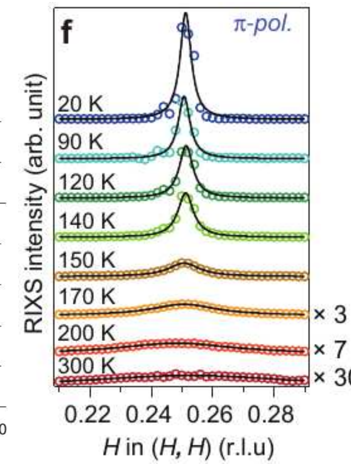
$3d^{8\pm\delta}$ superconductors: ambient-pressure $La_3Ni_2O_7$

[Liu et al., Sci. China Phys. Mech. Astron. 66, 21741 (2023)]
[Liu et al., arXiv:2307.02950 (2023)][Yang et al., arXiv:2309.01148 (2023)]
[Chen at al., arXiv:2401.12657 (2024)]

temperature anomalies

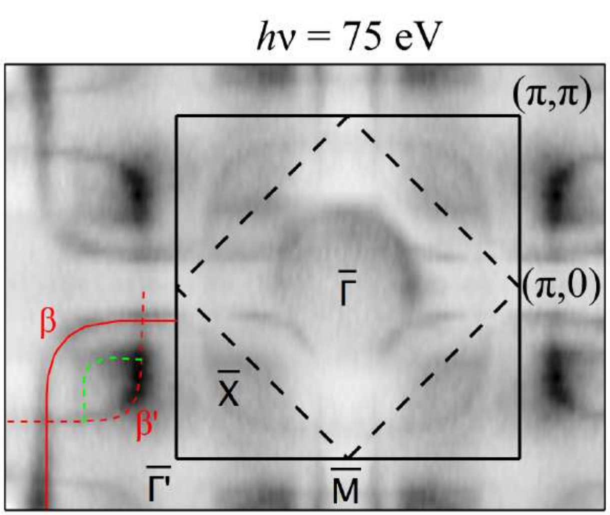


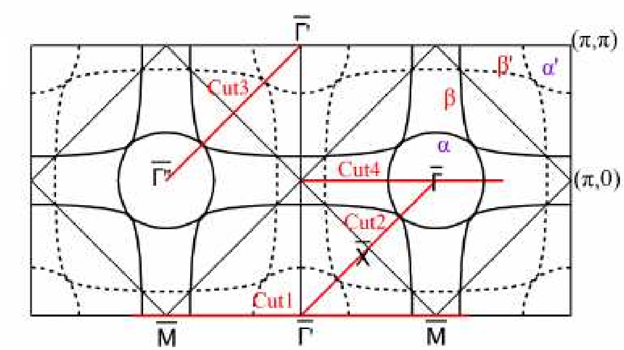




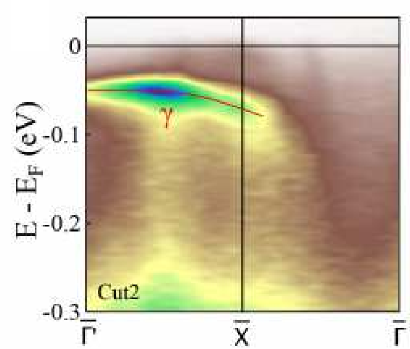
$T \sim 150\,$K anomaly is S(C)DW transition with $\mathbf{q} = (0.25, 0.25) = X$

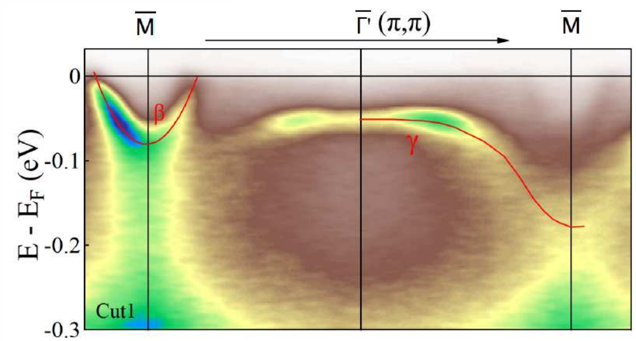
Fermi surface and flat band





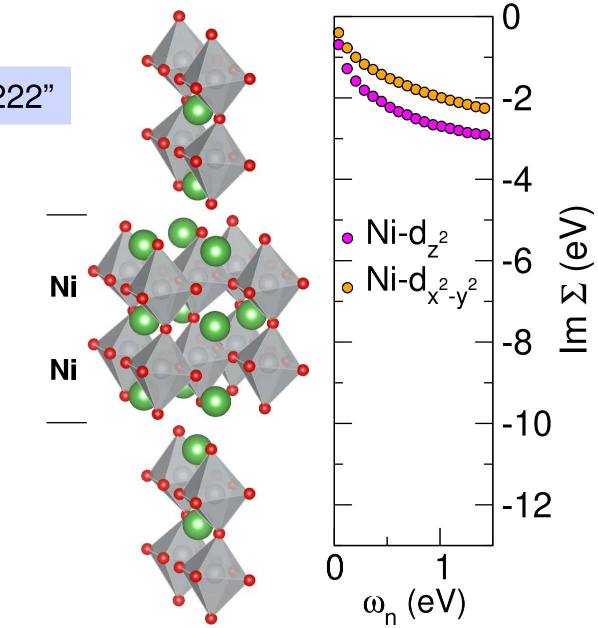
- flat band 50 meV
  below Fermi level
  at $\sim 10\,$K
  in ordered state

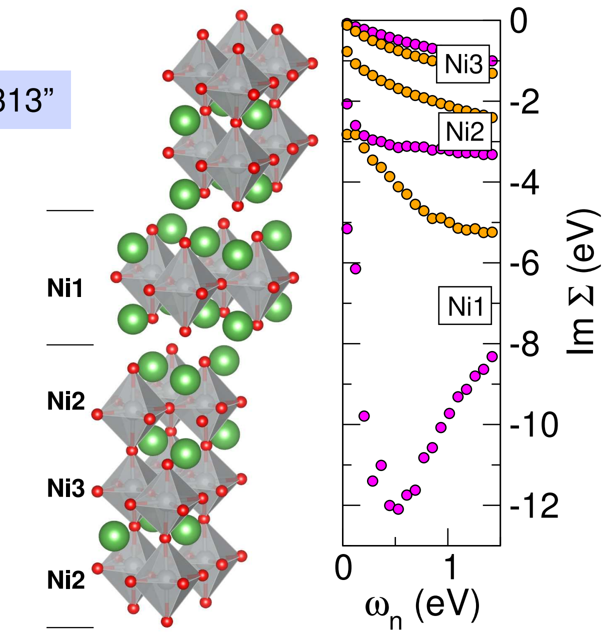




$3d^{8\pm\delta}$ superconductors: ambient-pressure $La_3Ni_2O_7$, DFT+sicDMFT

[Ling et al., J. Sol. Stat. Chem. 152, 517 (1999)]
[Chen et al., JACS 146, 3640 (2024)]
[Puphal et al., arXiv:2312.07341 (2023)]
[FL, Bötzel, Eremin, arXiv:2403.12831 (2024)]





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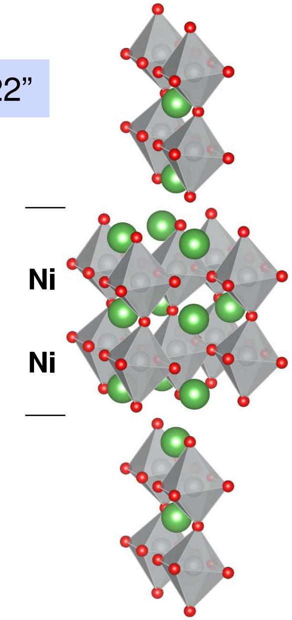
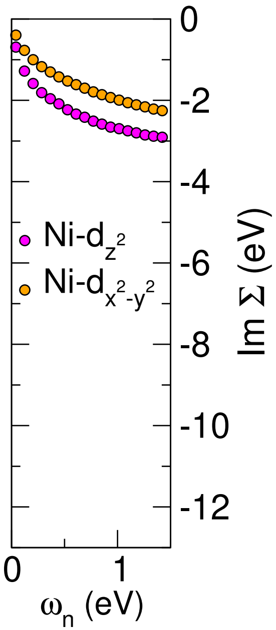
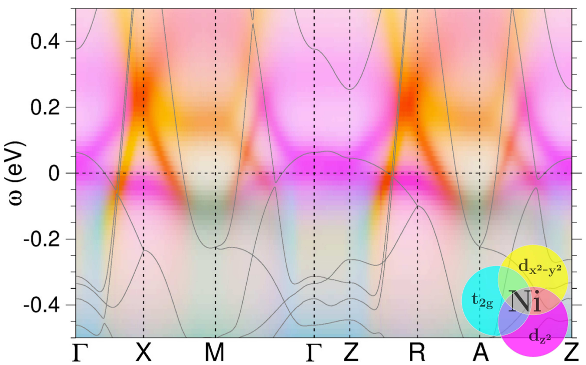
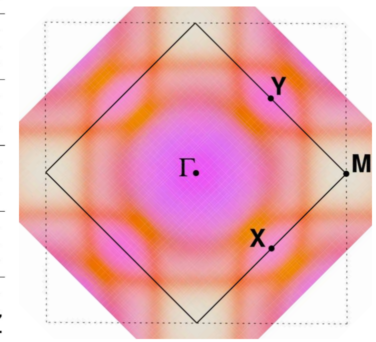
[Puphal et al., arXiv:2312.07341 (2023)]
[FL, Bötzel, Eremin, arXiv:2403.12831 (2024)]

222”



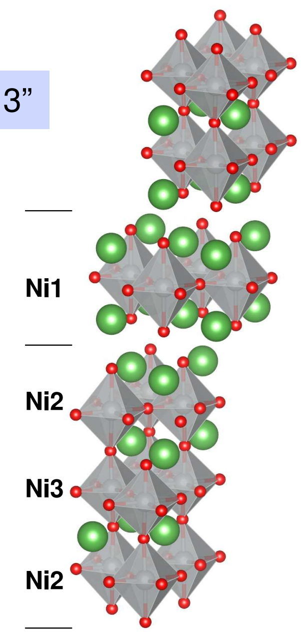


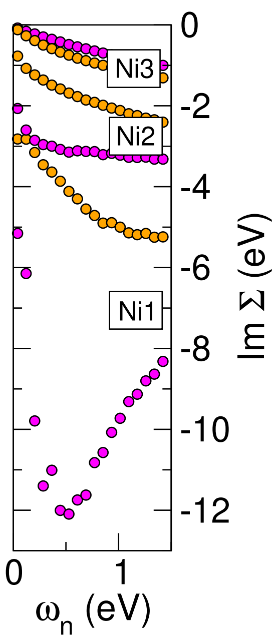


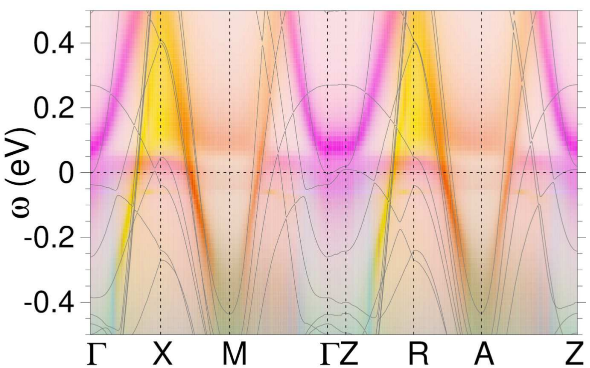


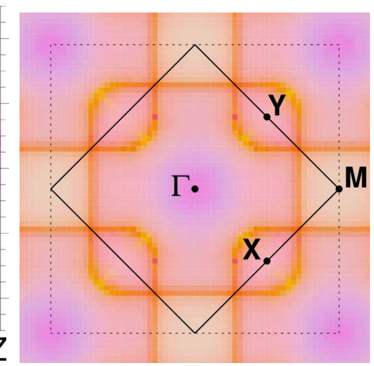
- strong shift of Ni-d_{z^2} spectral weight to low energy
- flat band still very close to Fermi level

313”





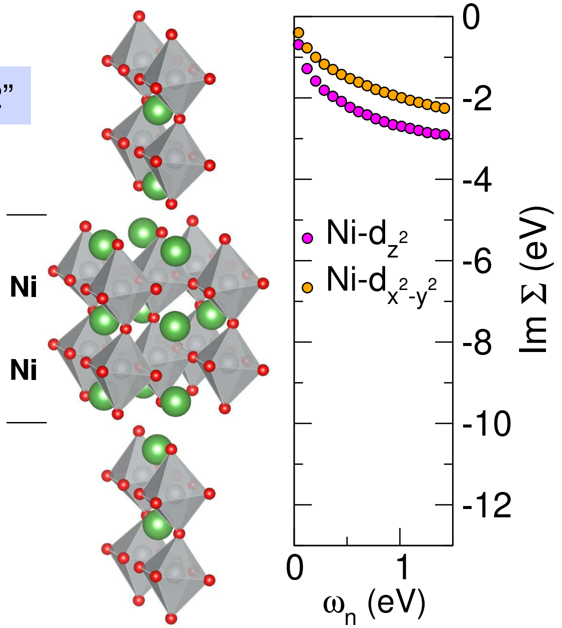


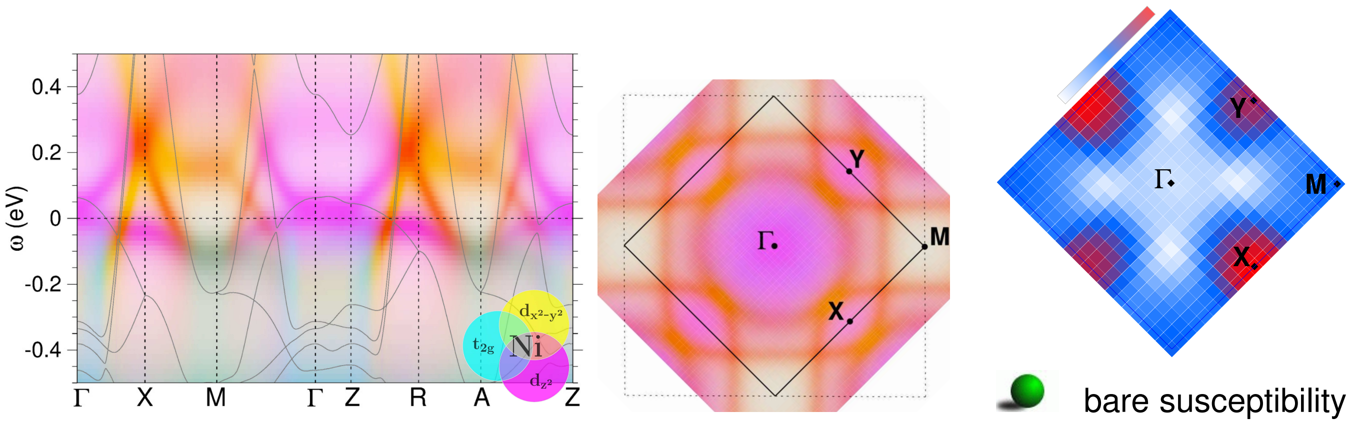


$3d^{8\pm\delta}$ superconductors: ambient-pressure $La_3Ni_2O_7$, DFT+sicDMFT

[Ling et al., J. Sol. Stat. Chem. 152, 517 (1999)]
[Chen et al., JACS 146, 3640 (2024)]
[Puphal et al., arXiv:2312.07341 (2023)]
[FL, Bötzel, Eremin, arXiv:2403.12831 (2024)]

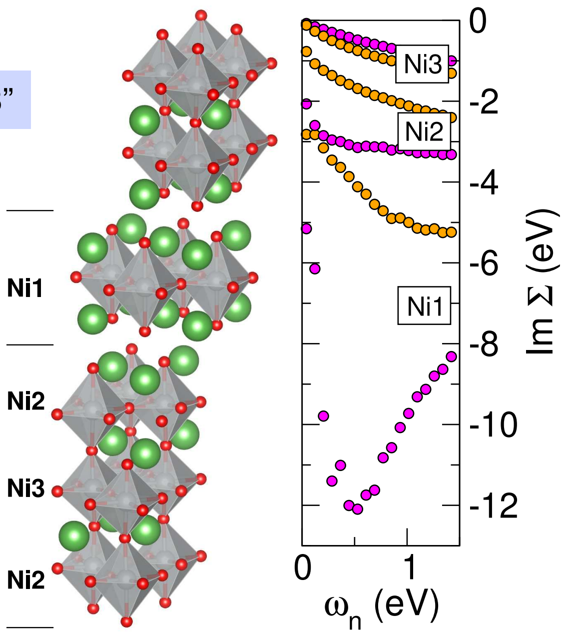
"2222"

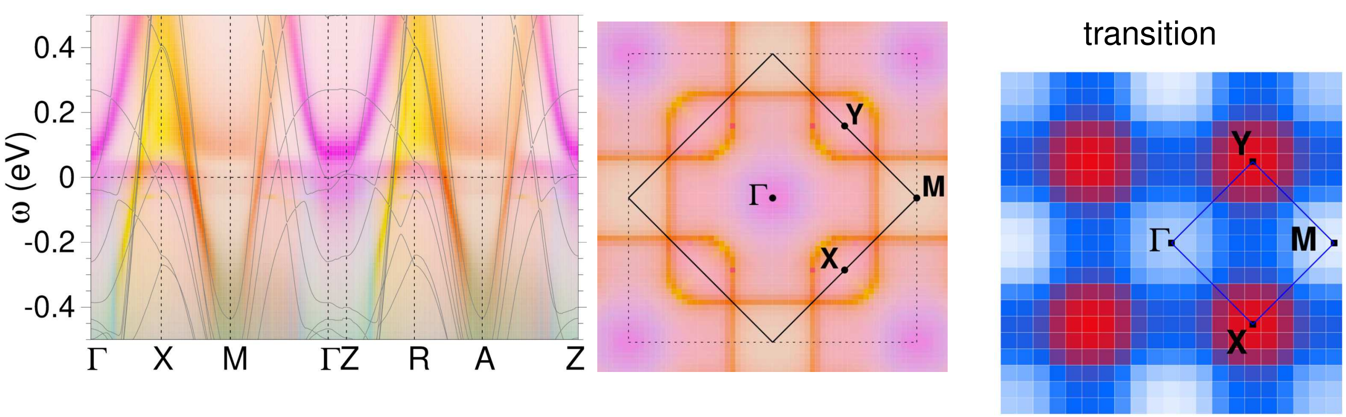




- bare susceptibility $\chi_0(\mathbf{q}) \sim \mathrm{Tr}\, GG$ shows maximum near $\mathbf{q}_X = (0.25, 0.25)$ ✓
- strong shift of Ni-d_{z^2} spectral weight to low energy
- flat band still very close to Fermi level
- flat band will be shifted below Fermi level due to ordering transition

"1313"

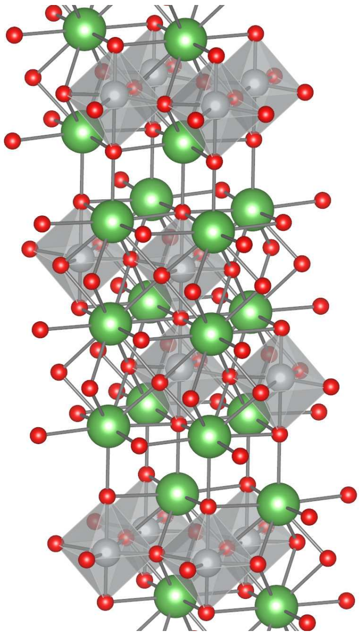


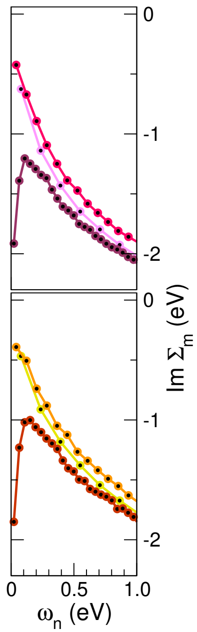


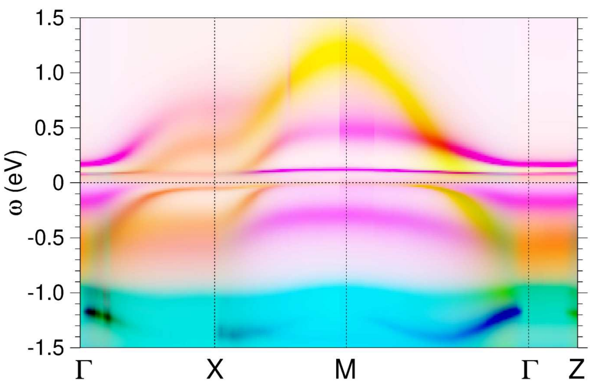
$3d^{8\pm\delta}$ superconductors: high-pressure $La_3Ni_2O_7$, DFT+sicDMFT

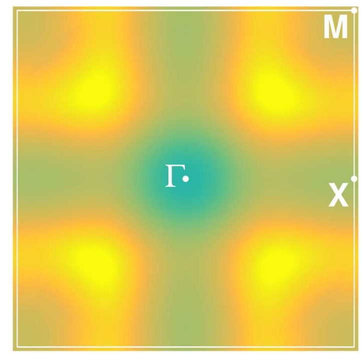
[FL, Gondolf, Bötzel, Eremin, PRB 108, L201121 (2023)]
[FL, Bötzel, Eremin, arXiv:2403.12831 (2024)]

222



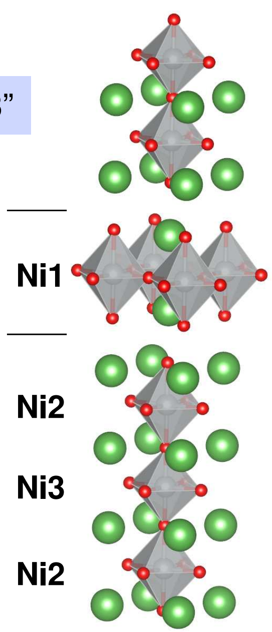


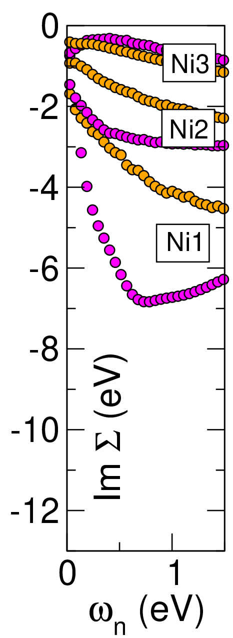


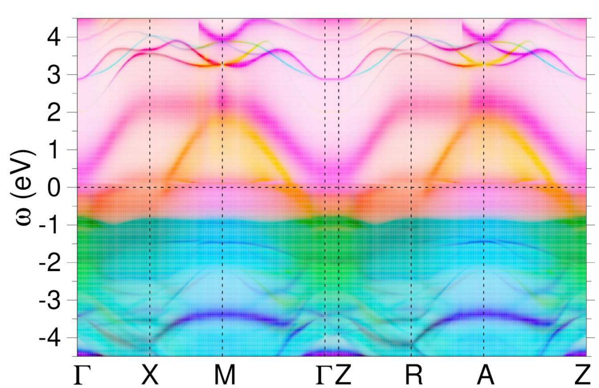


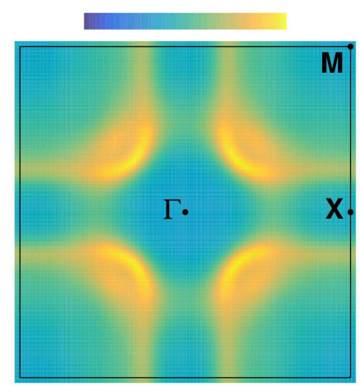
- flat band remains at the Fermi level, triggers non-Fermi-liquid physics
- Fermi arcs in 1313: multiorbital/layer coherence vs. flat-band-induced fluctuations

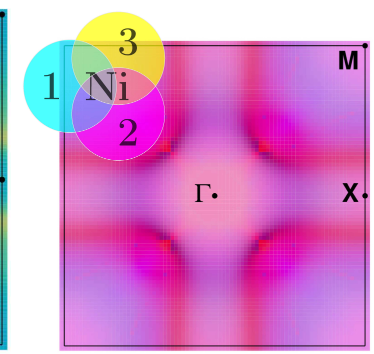
313”



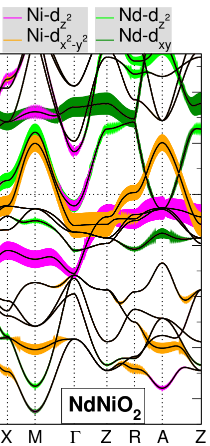


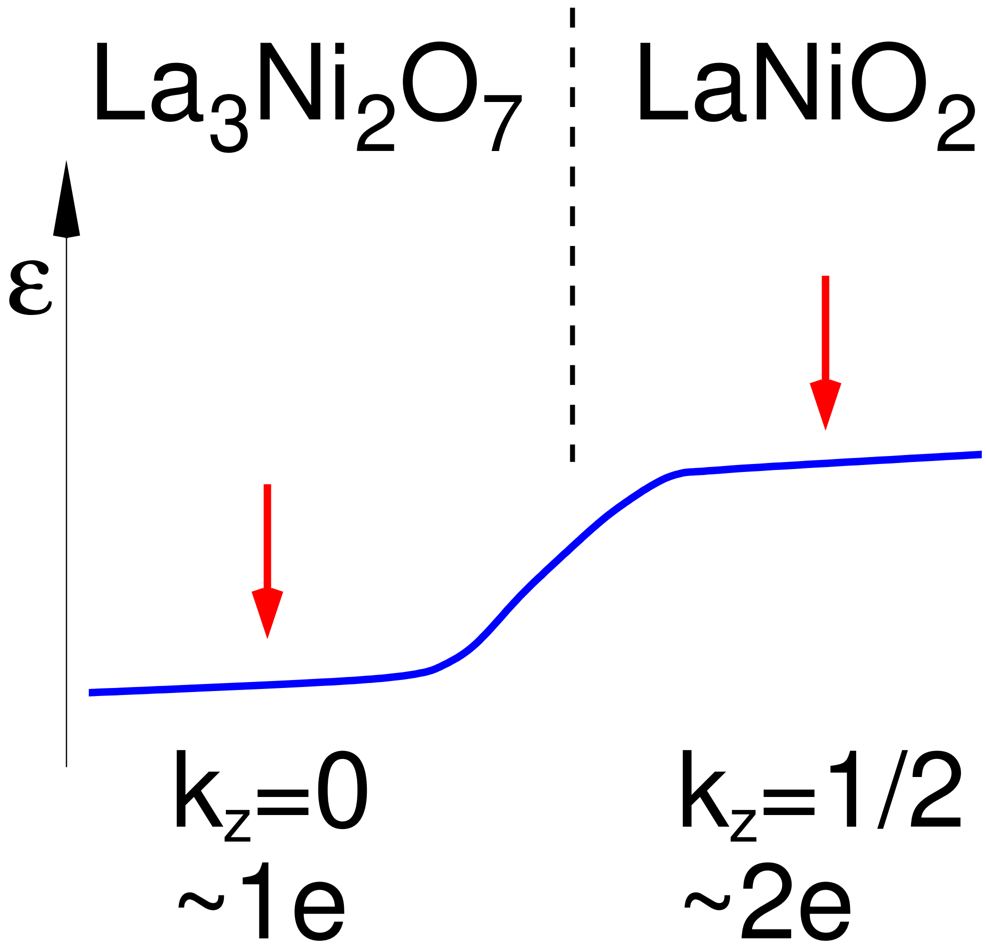




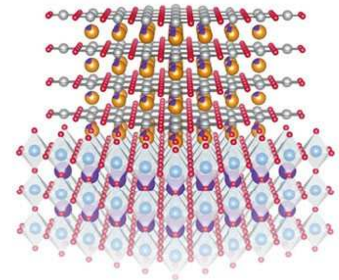


connection between d^9-like and d^8-like nickelate superconductivity?

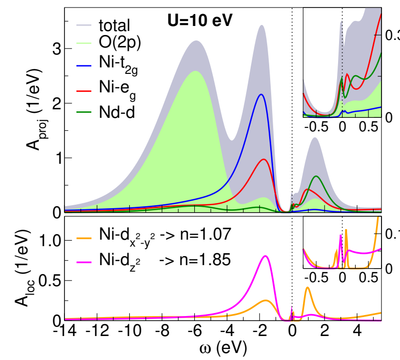


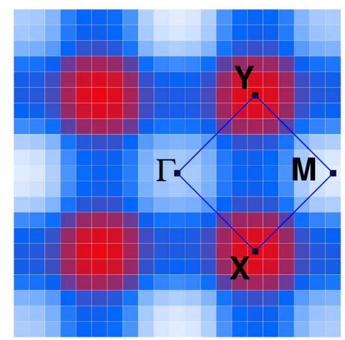


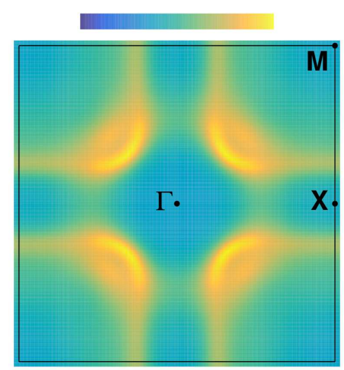
Summary



- superconducting nickelates split into d^8-like and d^9-like systems
- DFT+sicDMFT provides an adequate tool for investigations
- d^9-like systems
  - way larger charge-transfer energy than cuprates
  - orbital-selective Mottness at stoichiometry, flat-band physics with hole doping
- d^8-like systems
  - again Ni-d_{z^2} flat-band characteristics
  - different structural motifs: layer selectivity in 1313
  - ambient pressure: (S)DW transition at $T \sim 150$ K driven by flat band
  - high pressure: strong non-Fermi-liquid physics and Fermi arcs
- common features (?)
  - Ni-d_{z^2} flat-band physics
  - coherence-incoherence dichotomy

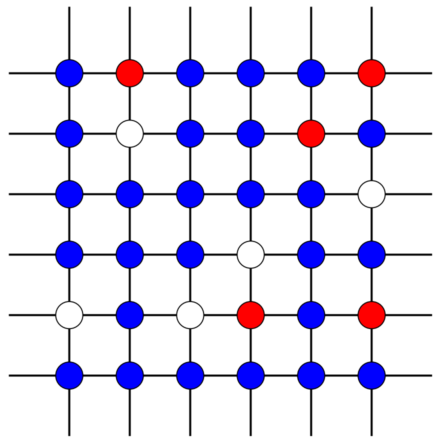




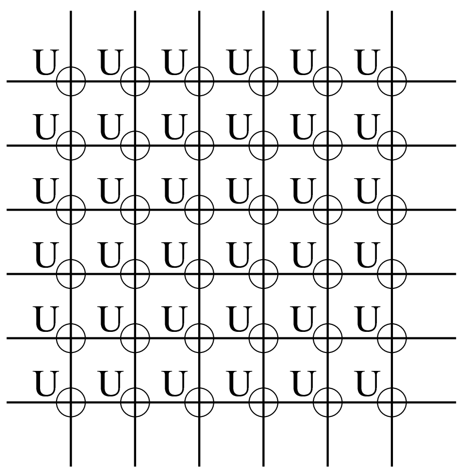


Theoretical approach: dynamical mean-field theory

[Metzner, Vollhardt, PRL (1989)] [Georges, Kotliar, PRB (1992)]

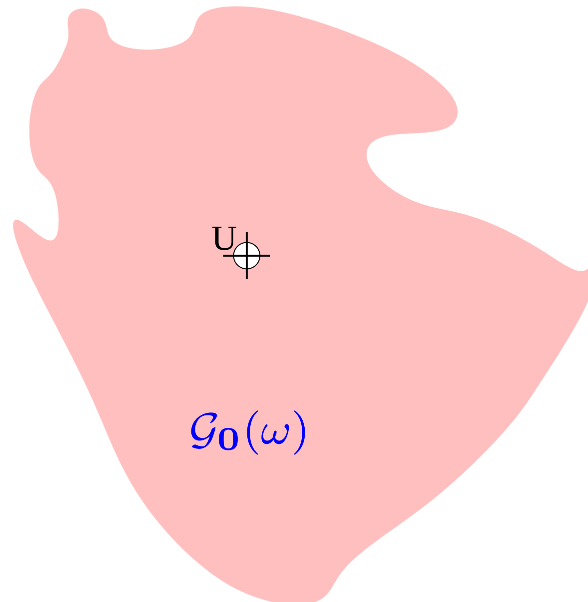


$$H = -t \sum_{\langle ij \rangle \sigma} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_i n_{i\uparrow} n_{i\downarrow}$$



$$G_{\rm loc}(\omega) = \sum_{\mathbf{k}} \left[\omega + \mu - \varepsilon_{\mathbf{k}} - \Sigma(\mathbf{k}, \omega)\right]^{-1}$$

$\Rightarrow$



$$G_{\rm imp}(\omega) = \left[\mathcal{G}_0^{-1}(\omega) - \Sigma_{\rm imp}(\omega)\right]^{-1}$$

DMFT approximation: $G_{\rm loc}(\omega) \stackrel{!}{=} G_{\rm imp}(\omega)$

$$G^{\rm DMFT}_{\rm loc}(\omega) = \sum_{\mathbf{k}} \left[\omega + \mu - \varepsilon_{\mathbf{k}} - \Sigma_{\rm imp}(\omega)\right]^{-1}$$

$\rightarrow$ local correlations are fully taken into account non-pertubatively, explicit non-local correlations are neglected

Theoretical approach: efficient inclusion of ligand-Coulomb in DFT+DMFT

[FL, Körner, Urban, Elsässer, PRB 100, 115125 (2019)]

Issue with late transition-metal oxides

The explicit role of O$(2p)$ and concomitat charge-transfer physics becomes more and more relevant for later transition-metal oxides ($\rightarrow$ Zaanen-Sawatzky-Allen). Therefore, conventional DFT+DMFT, with treating electronic correlations as only emerging from the transition-metal site, has to be extended to include explicit Coulomb interactions on oxygen.

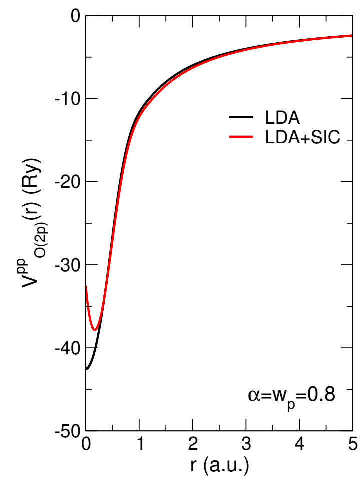
self-interaction correction (SIC) for ligand pseudopotential

[Vogel et al., PRB 54, 5495 (1996)]
[Filippetti and Spaldin, PRB 67, 125109 (2003)]
[Pemmaraju et al., PRB 75, 045101 (2007)]
[Körner and Elsässer, PRB 81, 085324 (2010)]

$$(-\nabla^2 + V_l + V_{\rm H}[n_v] + V_{\rm xc}[n_v] + V_{\rm cor}[n_l])\Psi_l^{pp} = \epsilon_l^{pp}\Psi_l^{pp}\ , \qquad (1)$$

$$V_{\rm cor}[n_l] = -w_l[V_{\rm H}[n_l] + V_{\rm xc}[n_l]]\ , \qquad (2)$$

$$V_l^{\rm SIC}(r) := V_l(r) - \alpha\langle\Psi_l^{pp}, V_{\rm cor}[n_l]\Psi_l^{pp}\rangle\Psi_l^{pp}(r)\ , \qquad (3)$$



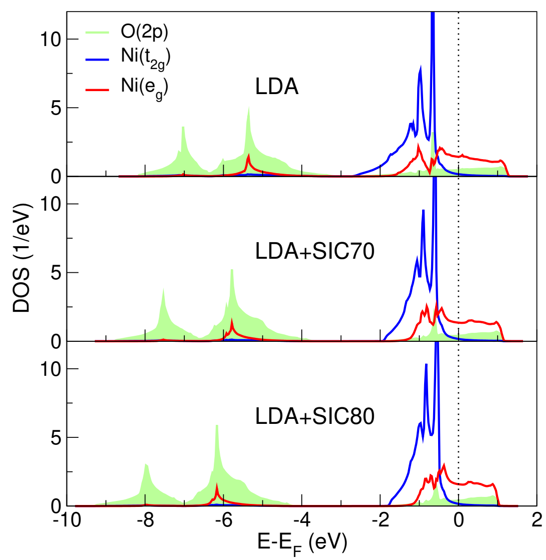
DFT+sicDMFT

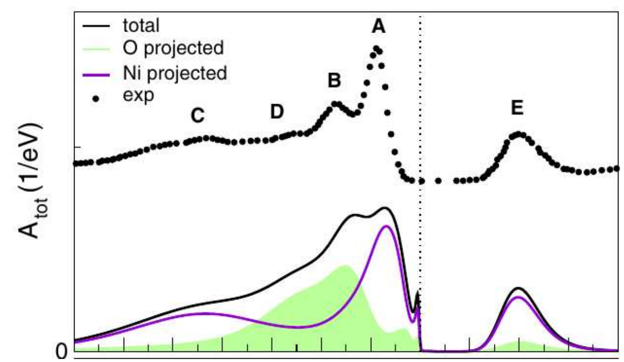
utilizing ligand $V^{\rm SIC}$ in established DFT+DMFT scheme

key effects of SIC inclusion

- enhancing $p-d$ splitting
- t_{pd} renormalization

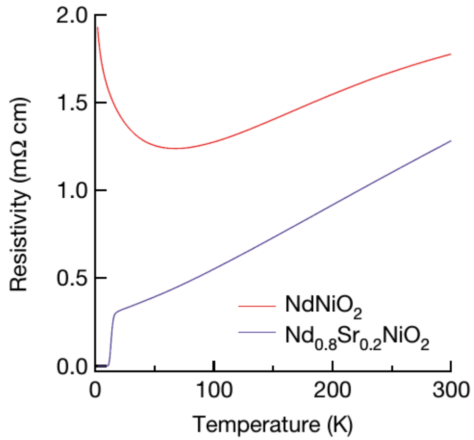
NiO



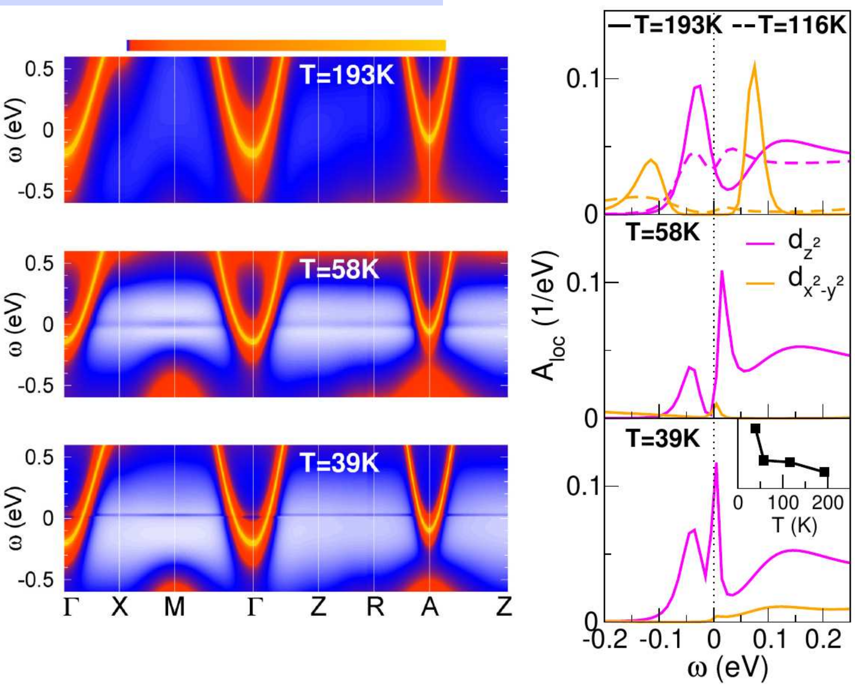


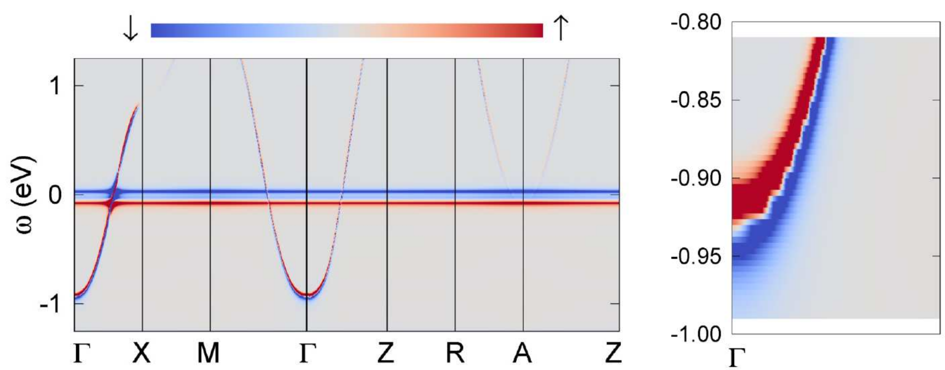
$3d^{9-\delta}$ superconductors: $NdNiO_2$

[Li et al., Nature 572, 624 (2019)]
[FL, PRX 10, 041002 (2020)]



Kondo physics at lower T(?)



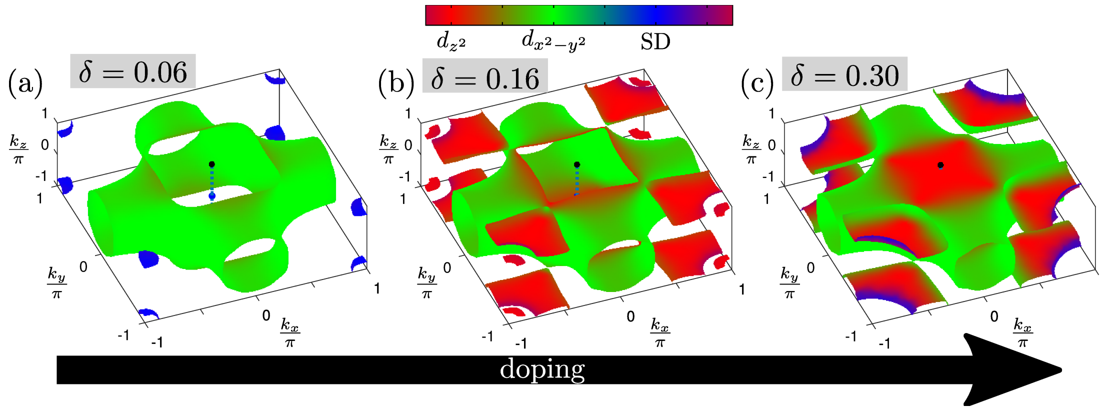


- AFM Kondo coupling due to Hund assistance from Ni-d_{z^2}
- $J_K \sim 120\,$meV

Theoretical picture: model spin excitations

[Kreisel et al., PRL 129, 077002 (2022)]
[Lu et al., Science 373, 213 (2021)]

Interacting Fermi surface with hole doping



Paramagnetic RIXS response