

Material-realistic modeling of Ta_2NiSe_5

– Microscopic Order Parameter –

Malte Rösner

Cavities & Excitonic Insulators Workshop
Flatiron Institute, CCQ, 2019

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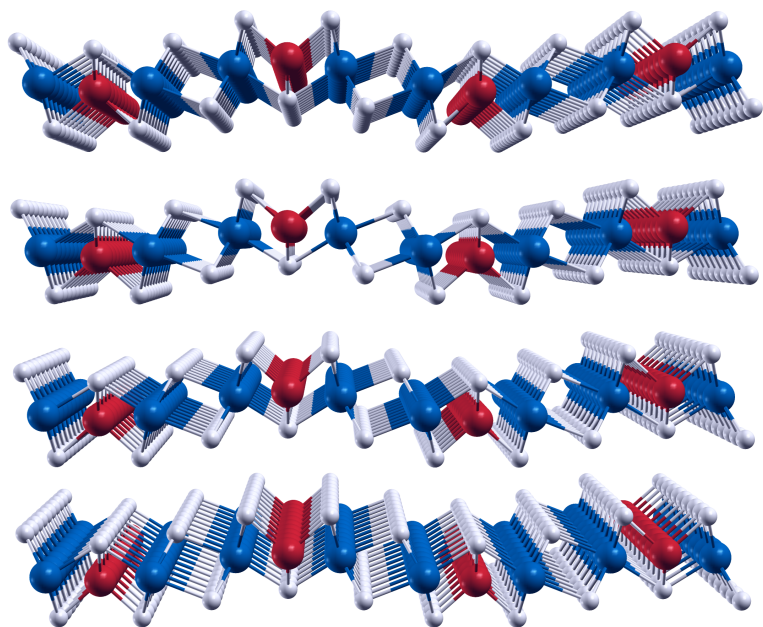
– Microscopic Order Parameter –

Ecole Polytechnique & College de France:
MPSD Hamburg:

Flatiron Institute / CCQ:

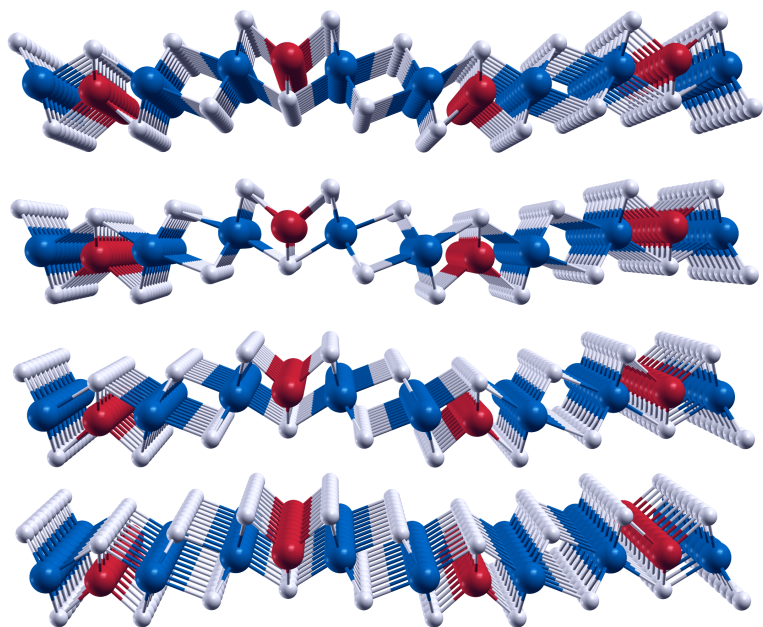
Giacomo Mazza
Lukas Windgaetter, Simone Latini,
Hannes Huebener, Angel Rubio
Jennifer Cano, Andrew Millis,
Antoine Georges

Ta₂NiSe₅ – Motivation



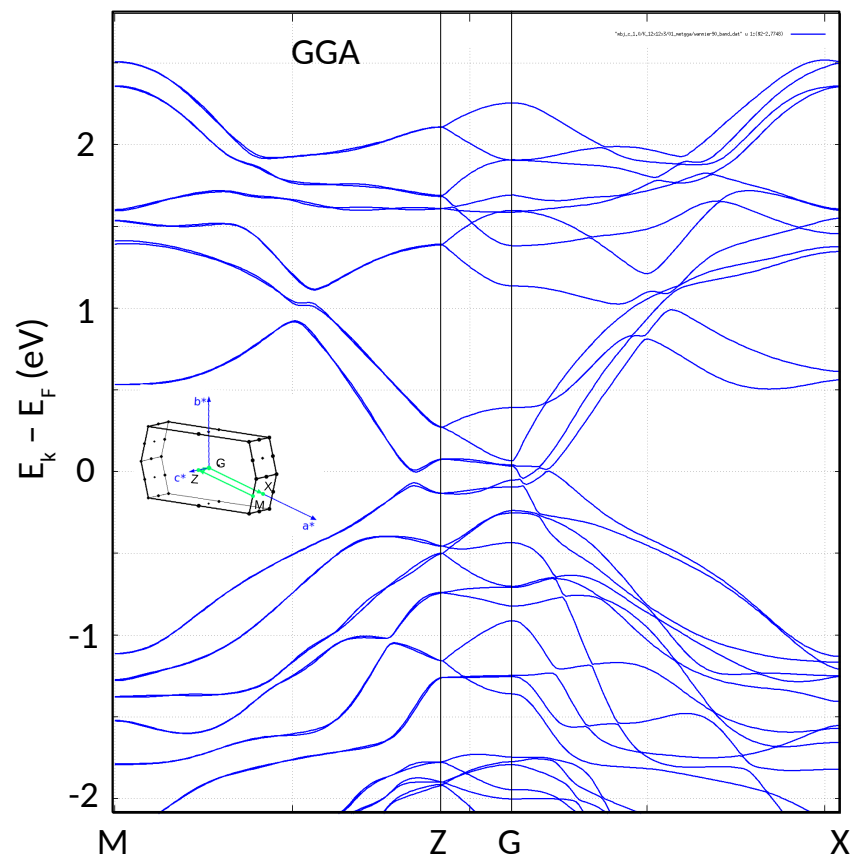
- gain material-realistic understanding of many-body effects
- learn how many-body effects are influenced by
 - number of layers
 - substrate screening
 - cavities
- complicated and involving endeavor due to
 - large number of atoms in the unit cell
 - layered structure
 - strong and long-range Coulomb interactions
 - sizable electron-phonon interactions

Ta₂NiSe₅ – Approach

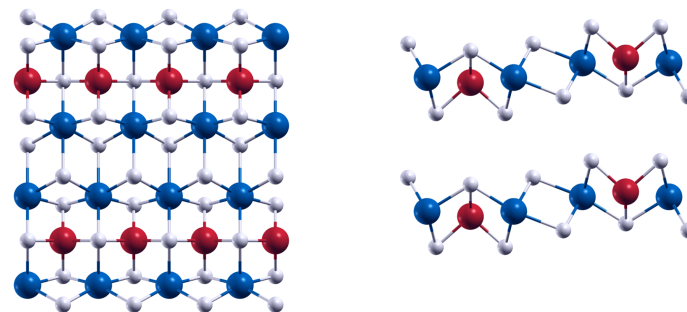


- combine ab initio calculations with higher-level many-body theory
- by down-folding a minimal, yet realistic model including
 - electronic and phononic dispersions
 - electron-electron interactions
 - electron-phonon interactions
- using full RPA or Hartree-Fock as many-body solvers

Ta₂NiSe₅ – Ab Initio Band Structure

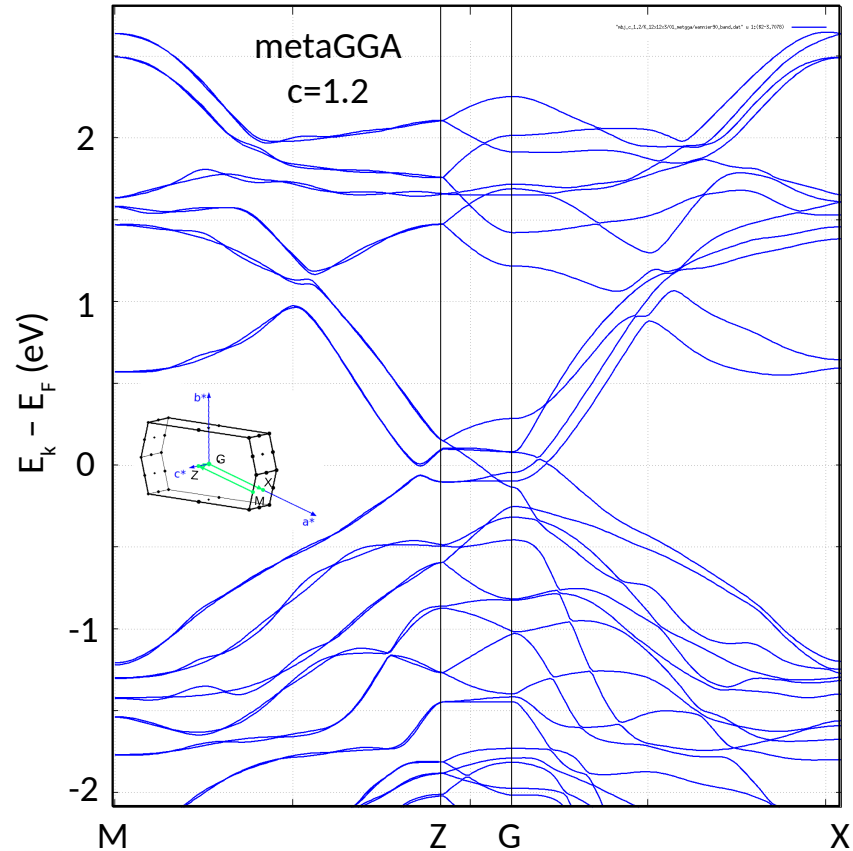


- High-temperature / orthorhombic phase

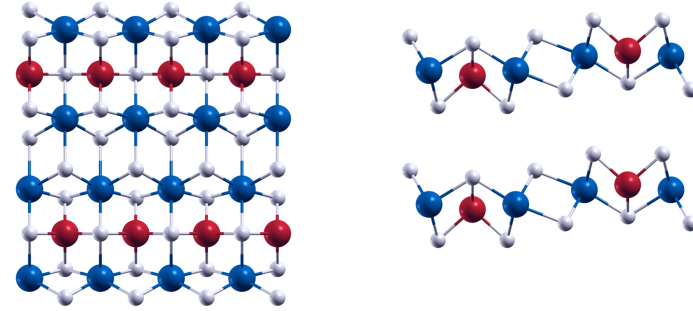


- DFT / GGA:
 - no gap $\leftrightarrow$ 4 to 6 bands crossing Fermi level
 - weak inter-chain dispersion
- long-range Coulomb interactions?
 - GW not feasible (yet!)
 - hybrid / metaGGA (can) mimic the same effects

Ta₂NiSe₅ - Ab Initio Band Structure

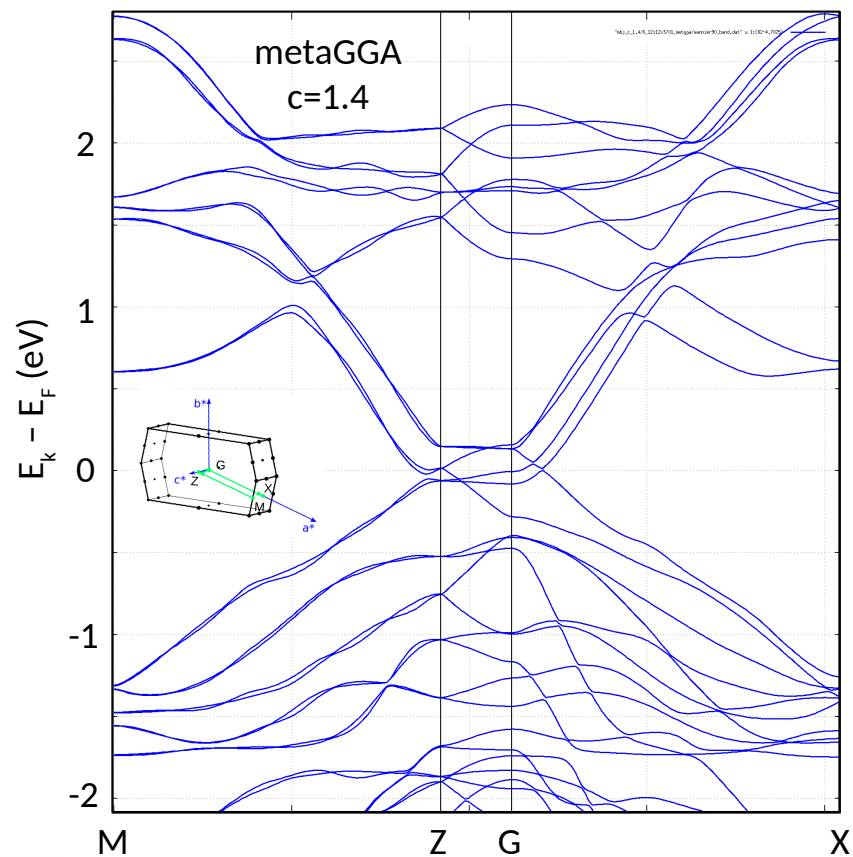


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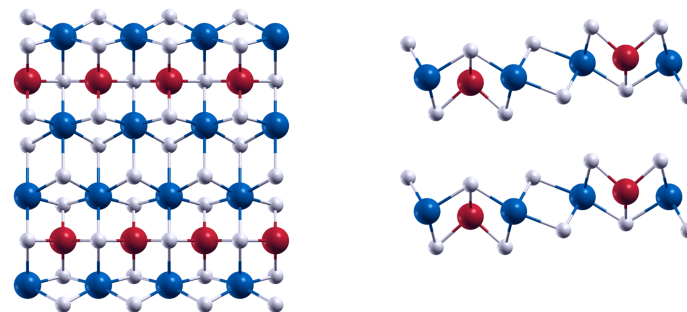


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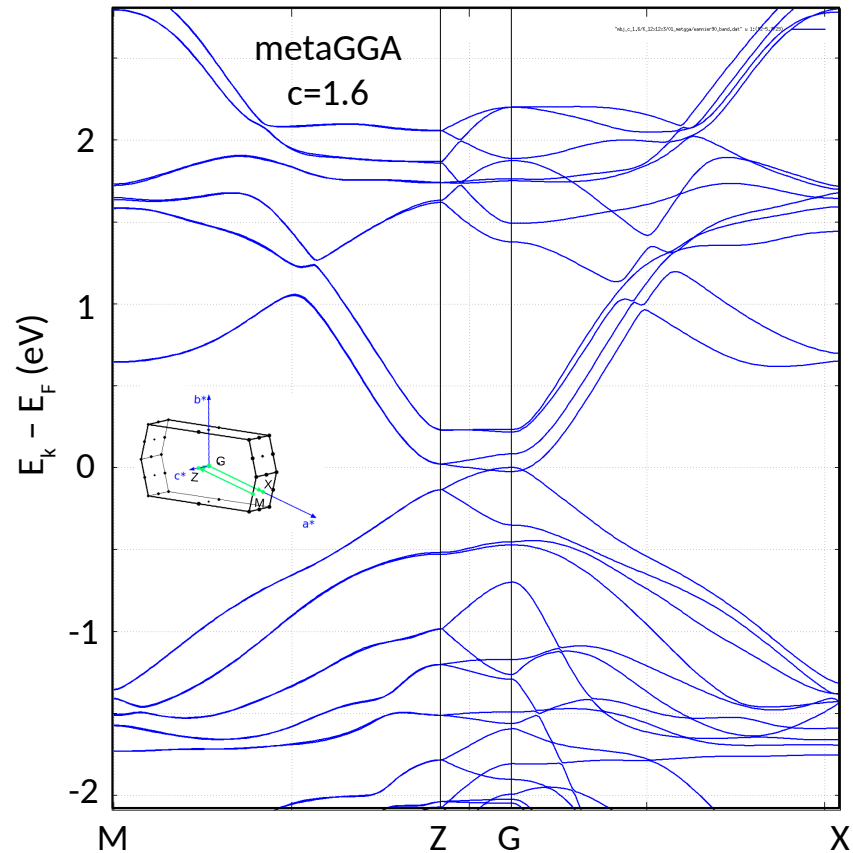


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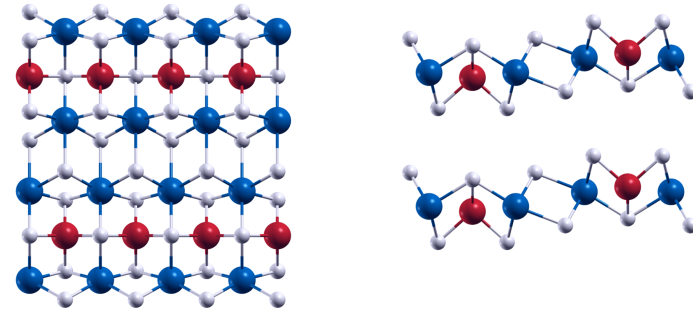


- long-range Coulomb interactions?
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Ta₂NiSe₂ - Ab Initio Band Structure

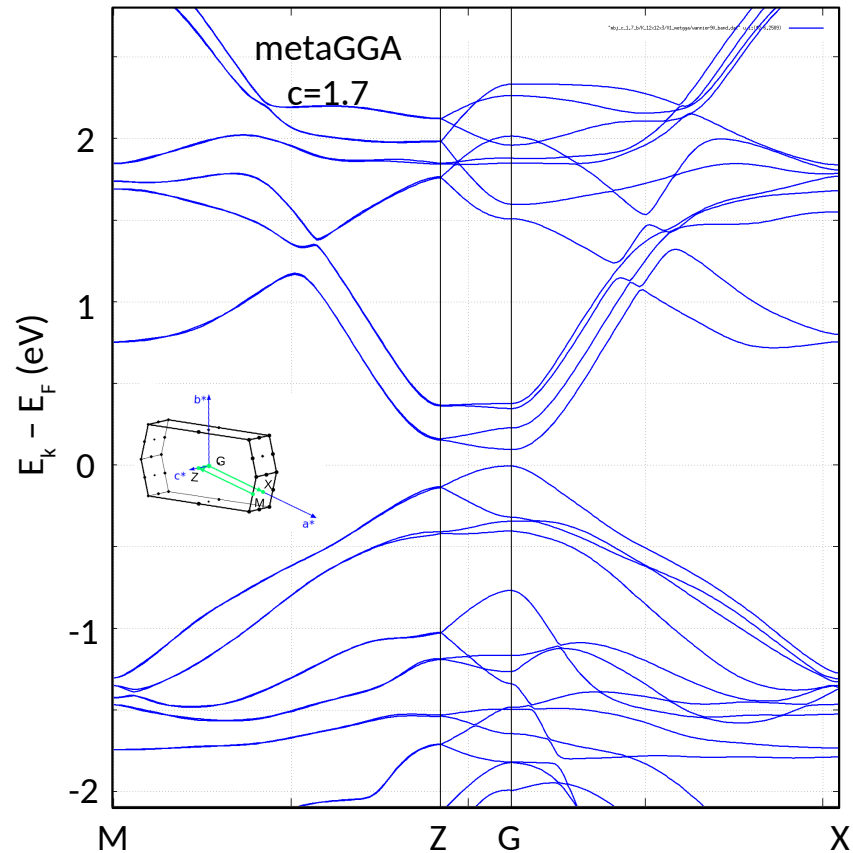


- High-temperature / orthorhombic phase

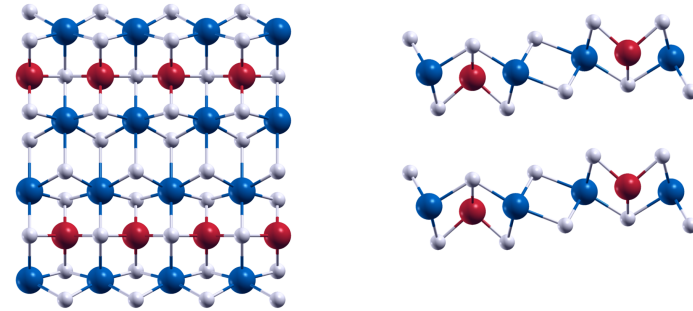


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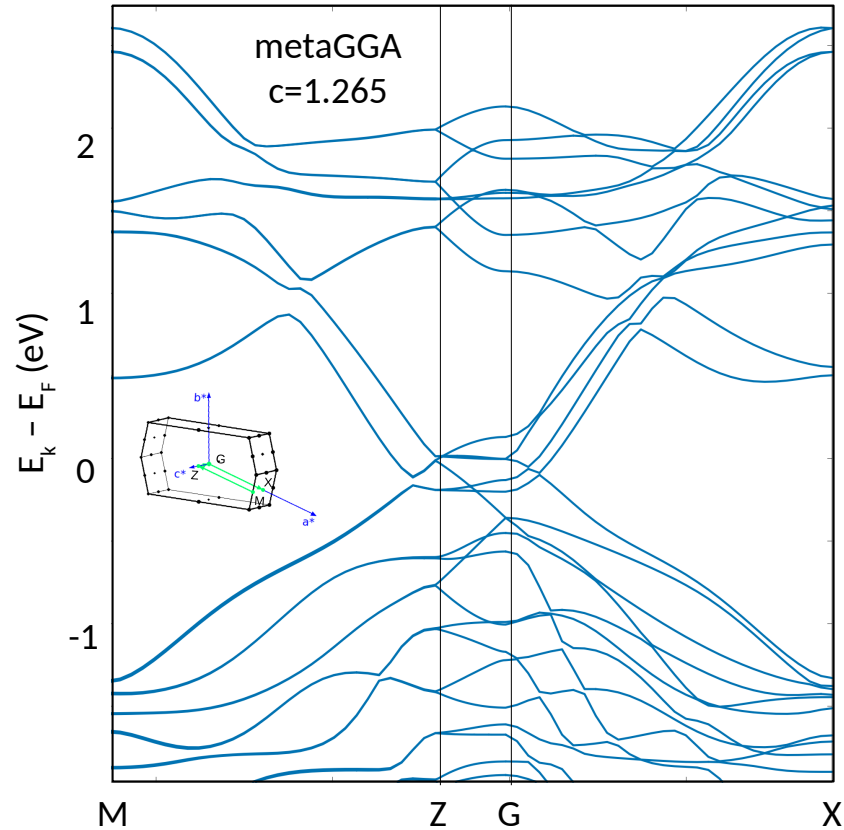


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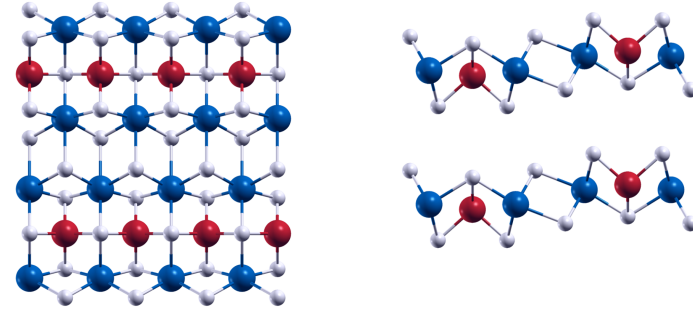


- long-range Coulomb interactions?
 - hybrid / **metaGGA** (can) mimic the same effects
 - band widths widen
 - band around E_F disentangle
 - can even open band gap

Ta₂NiSe₅ - Ab Initio Band Structure

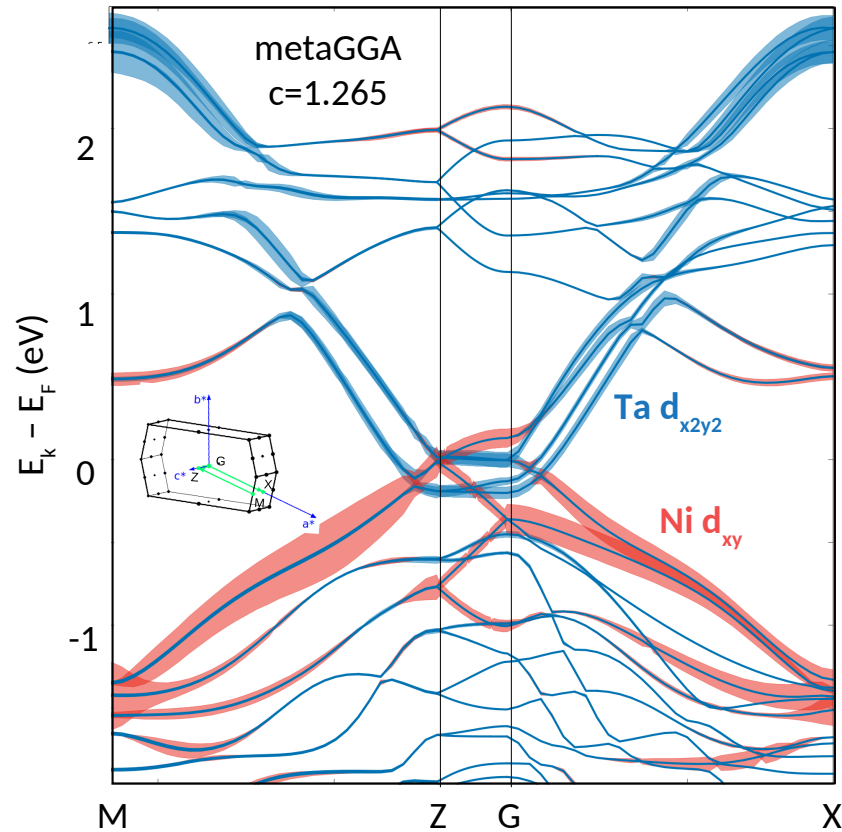


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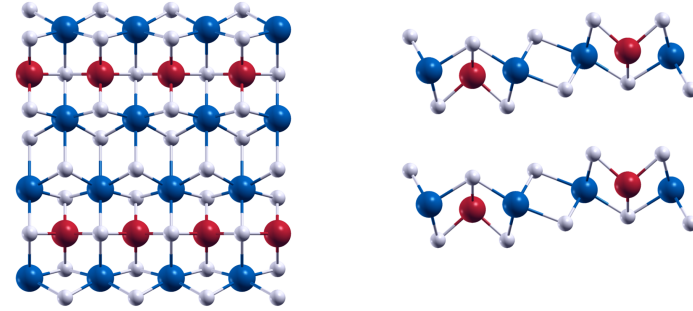


- metaGGA, $c = 1.27$ (automatic) to mimic long-range Coulomb interactions

Ta₂NiSe₅ - Ab Initio Band Structure

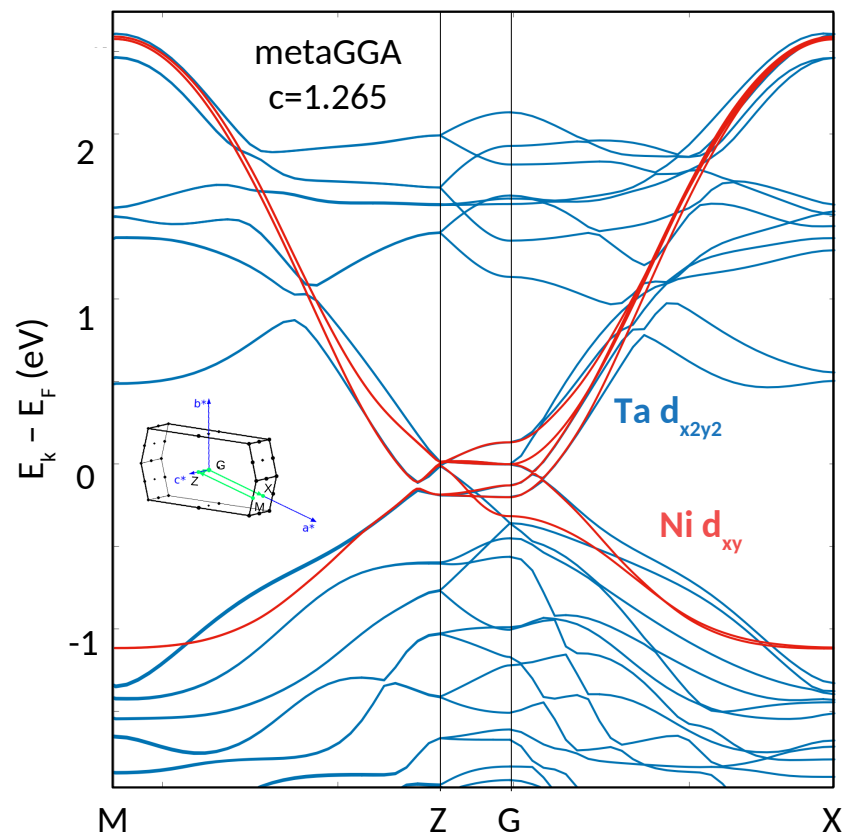


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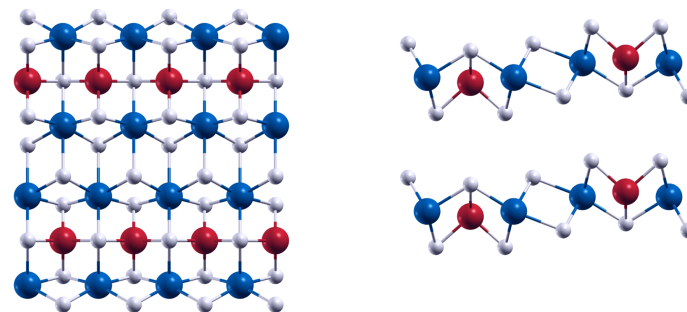


- metaGGA, $c = 1.27$ (automatic) to mimic long-range Coulomb interactions
- Ta $d_{x^2y^2}$ and Ni d_{xy} are main contributions (around Fermi level)

Ta₂NiSe₅ – Material-Realistic Model

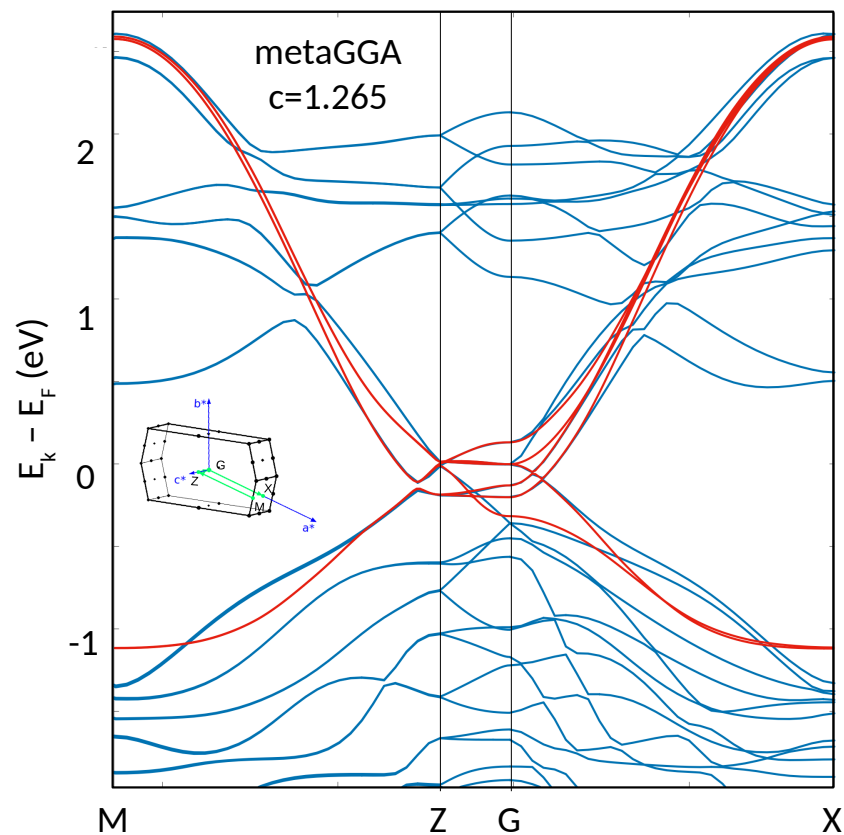


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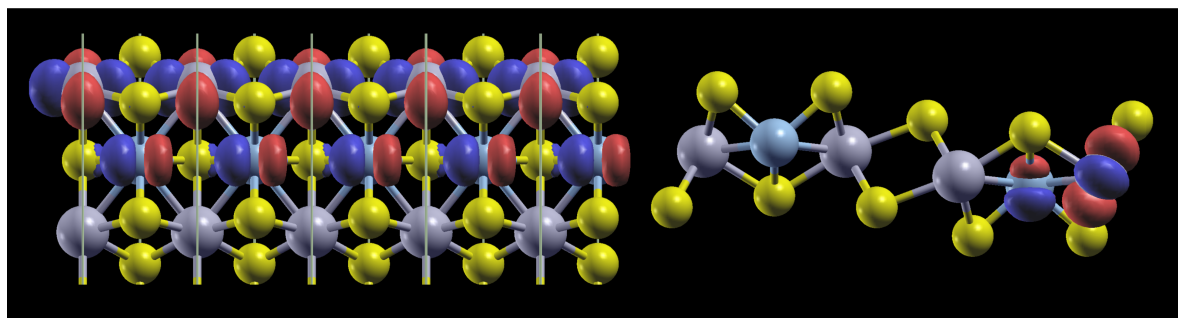
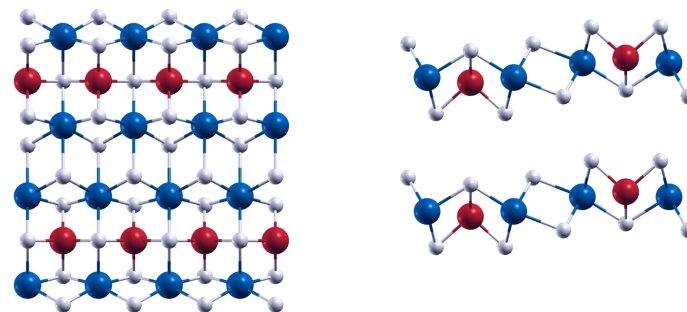


- metaGGA, $c = 1.27$ (automatic) to mimic long-range Coulomb interactions
- Ta $d_{x^2y^2}$ and Ni d_{xy} are main contributions (around Fermi level)
 - serve as good initial projections for Wannierization
 - minimal, material-realistic model involves **6 orbitals**

Ta₂NiSe₅ – Material-Realistic Model

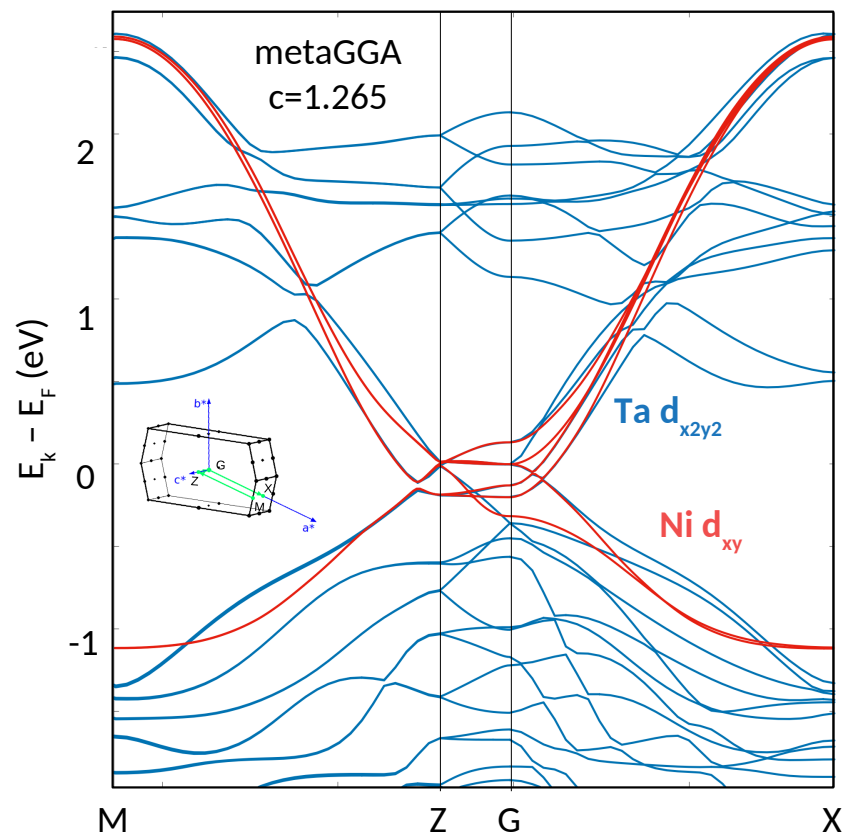


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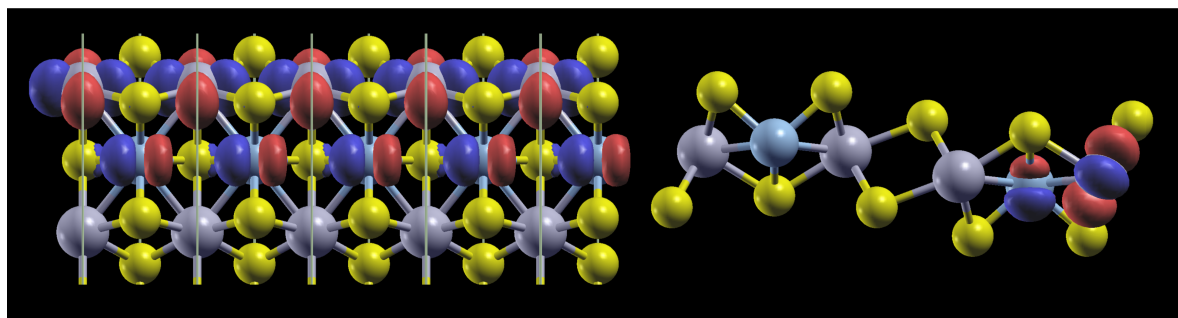
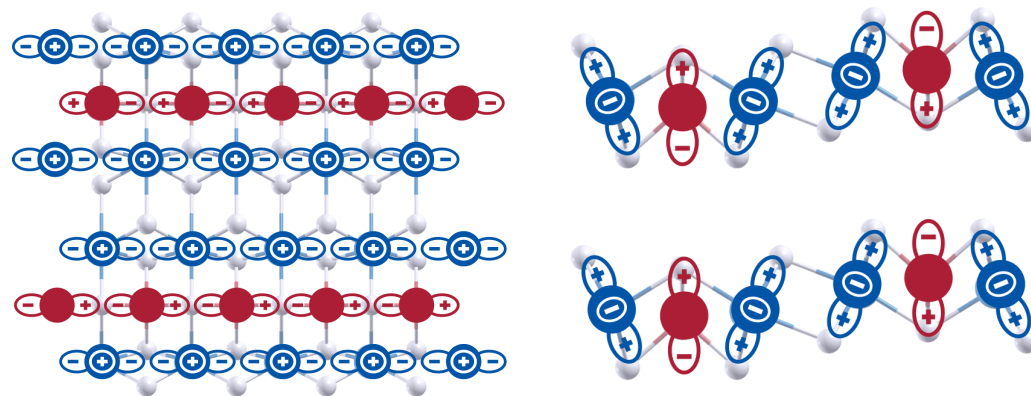


→ tilted Ni & Ta d orbitals take Se contributions into account!

Ta₂NiSe₅ - Ab Initio Model

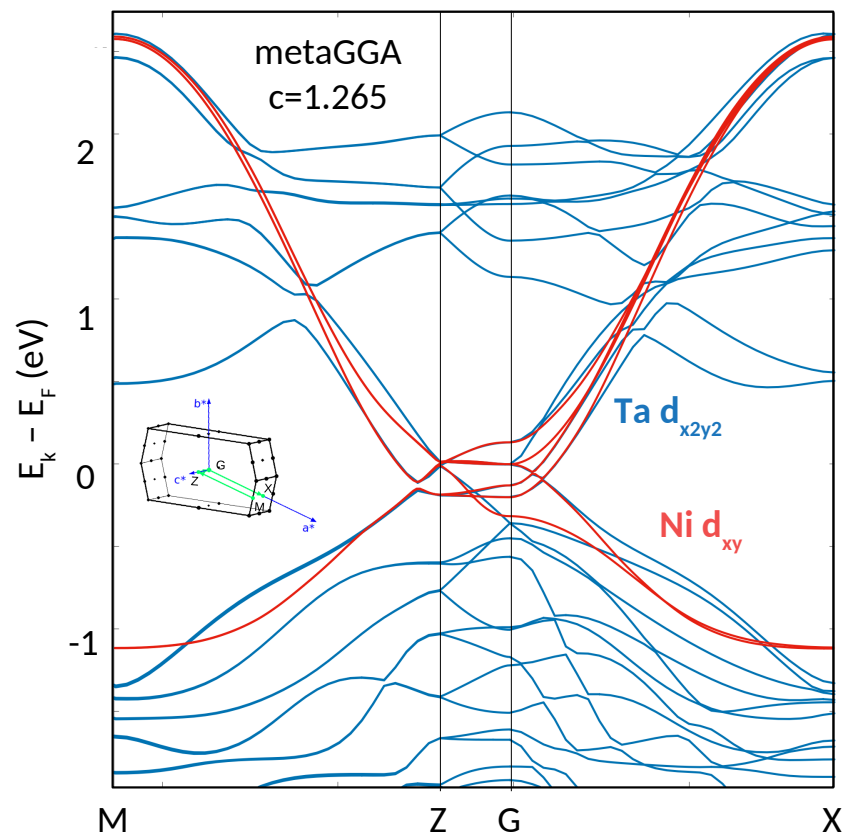


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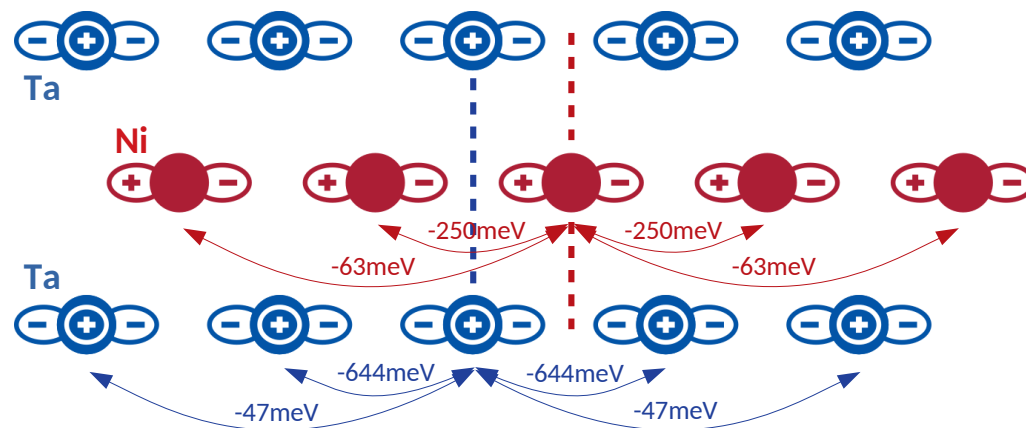


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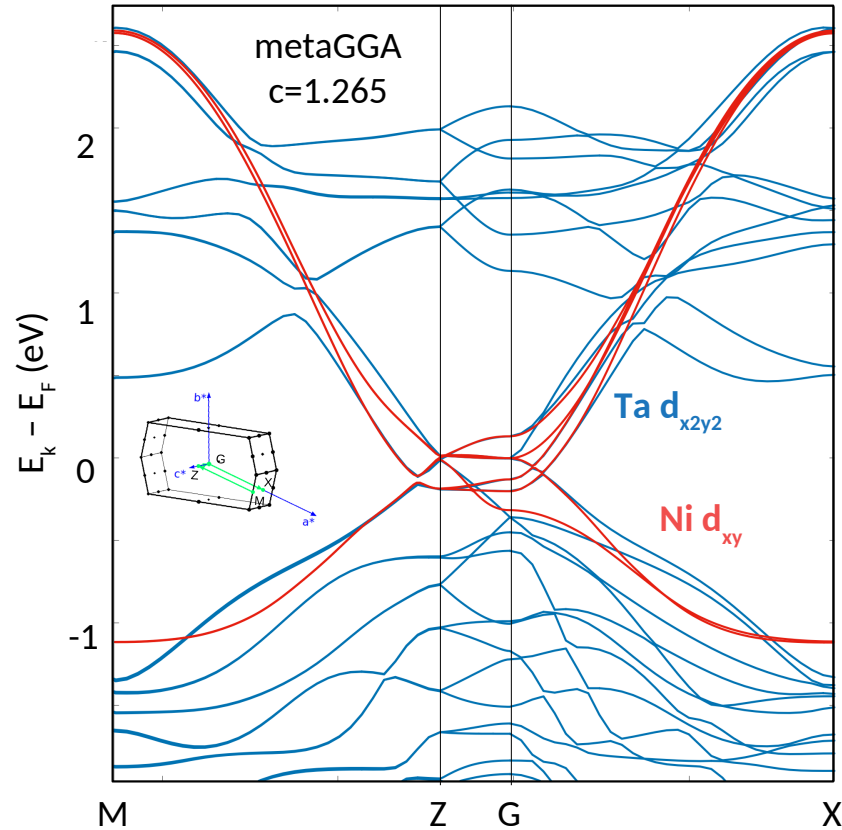
Ta₂NiSe₅ - Material-Realistic Model



- hopping matrix elements in minimal Wannier basis

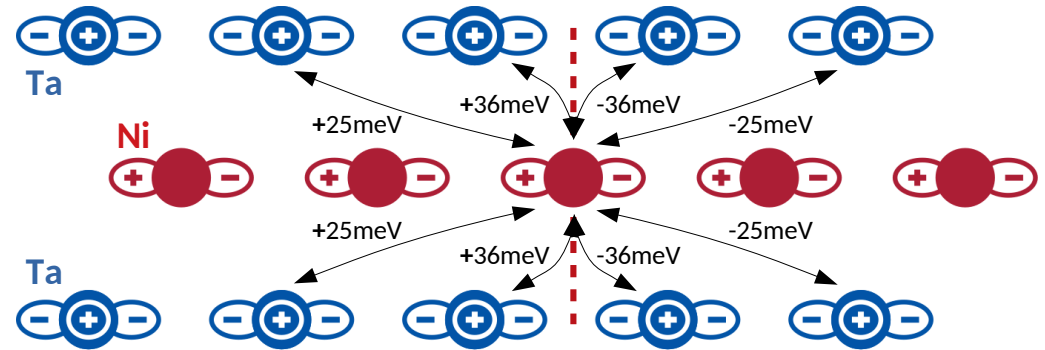


Ta₂NiSe₅ - Material-Realistic Model

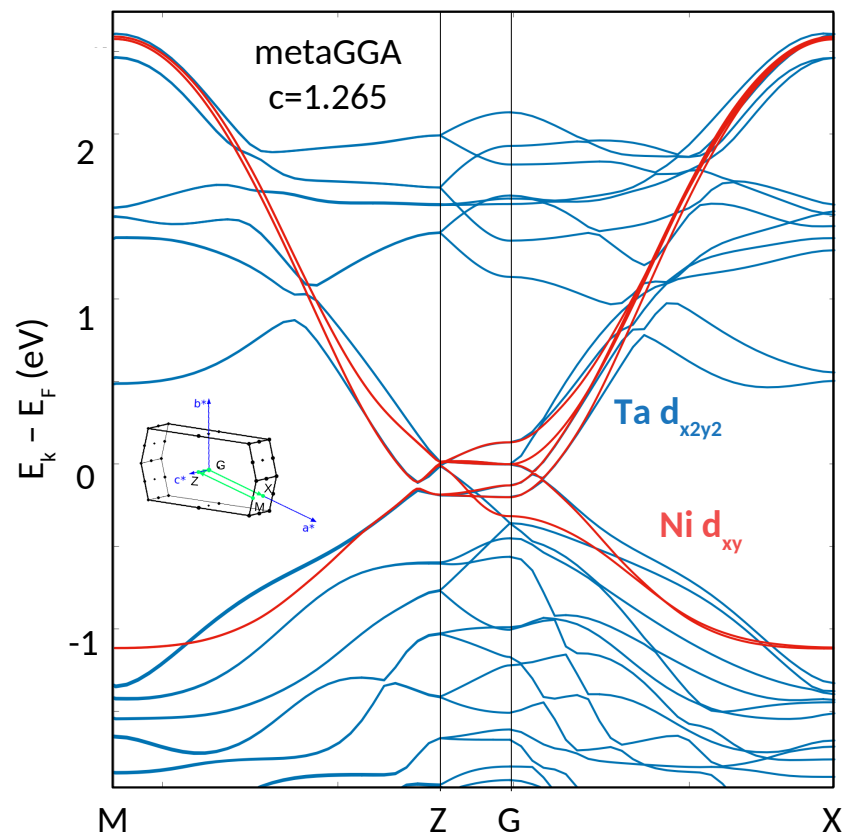


- hopping matrix elements in minimal Wannier basis

→ non-zero Ta-Ni hoppings with inverted signs

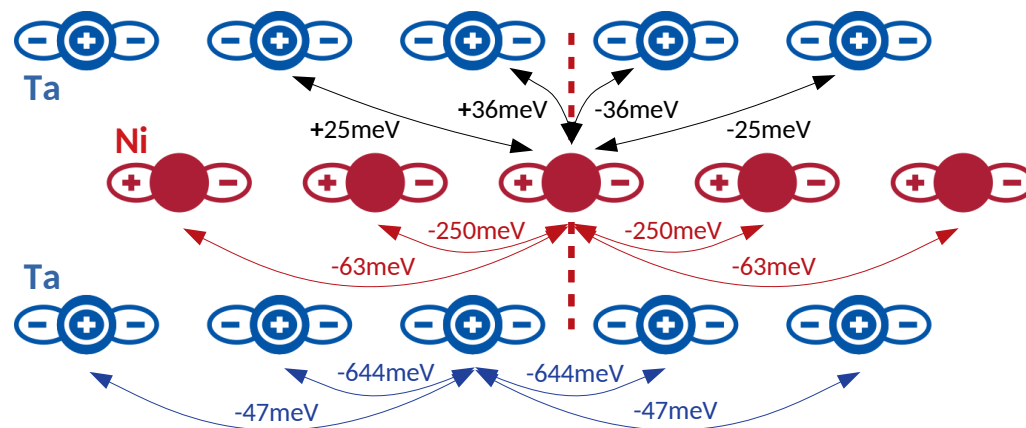


Ta₂NiSe₅ - Material-Realistic Model

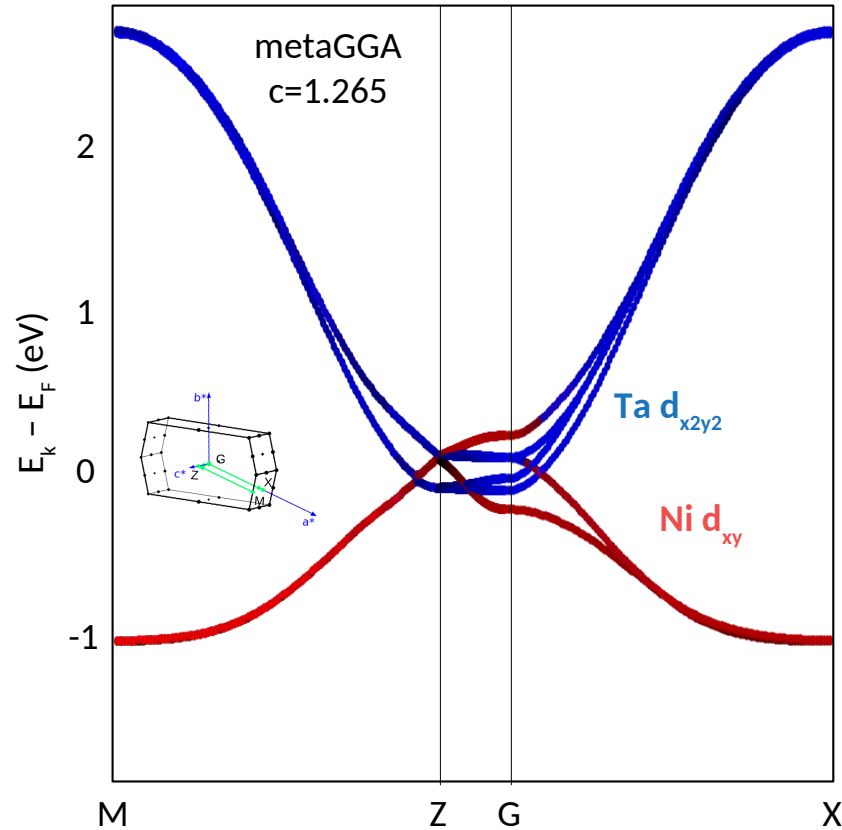


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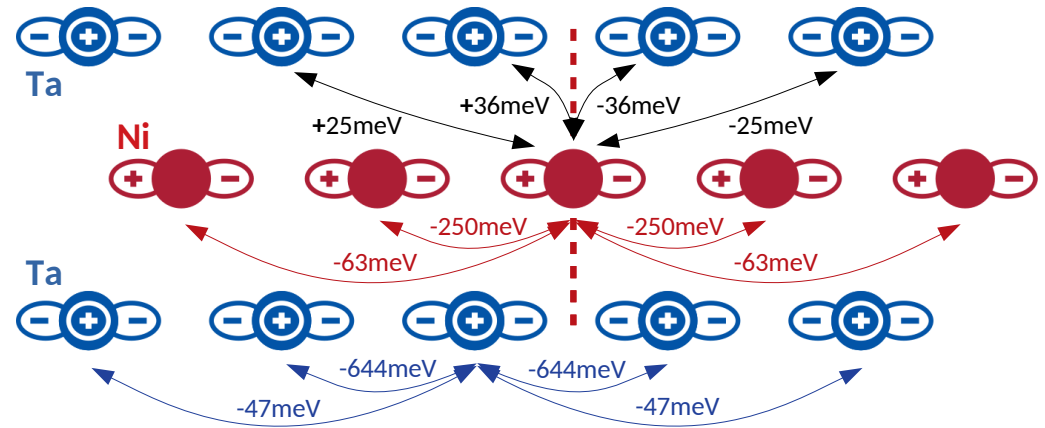
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Ta₂NiSe₅ – Material-Realistic Model



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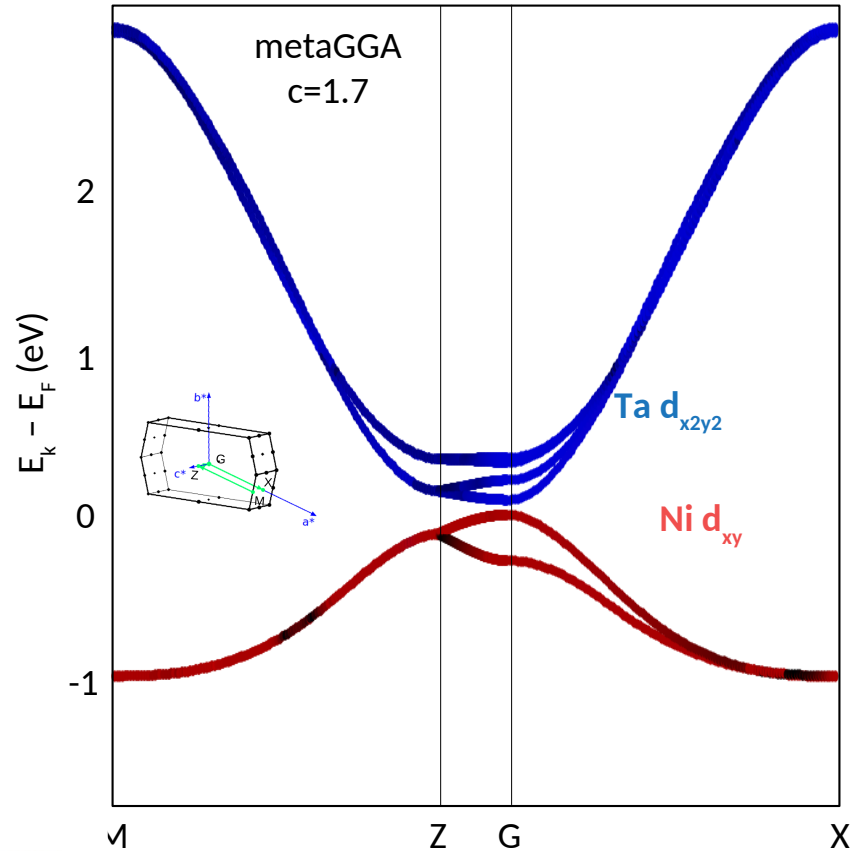


→ yields Ta / Ni hybridization throughout the Brillouin zone

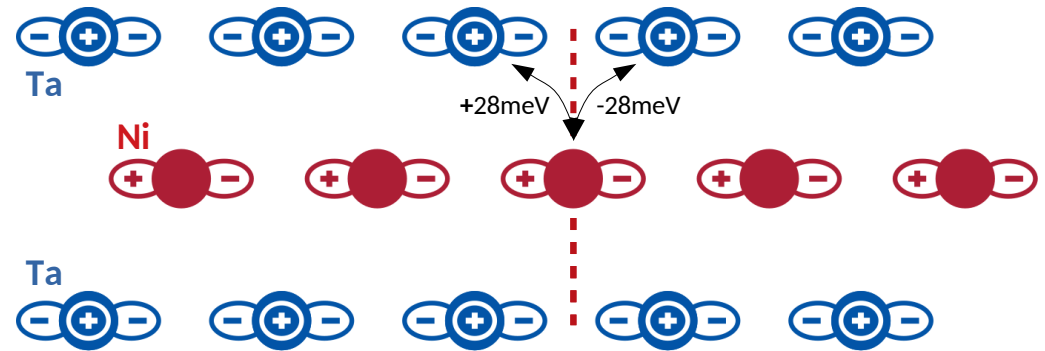
→ but not at Gamma!

$$H_{\alpha\beta}(\mathbf{k}') = \sum_{\mathbf{R}} e^{i\mathbf{k}'\cdot\mathbf{R}} t_{\alpha\beta}(\mathbf{R})$$

Ta₂NiSe₅ – Material-Realistic Model



- hopping matrix elements in minimal Wannier basis
- → non-zero Ta-Ni hoppings with inverted signs

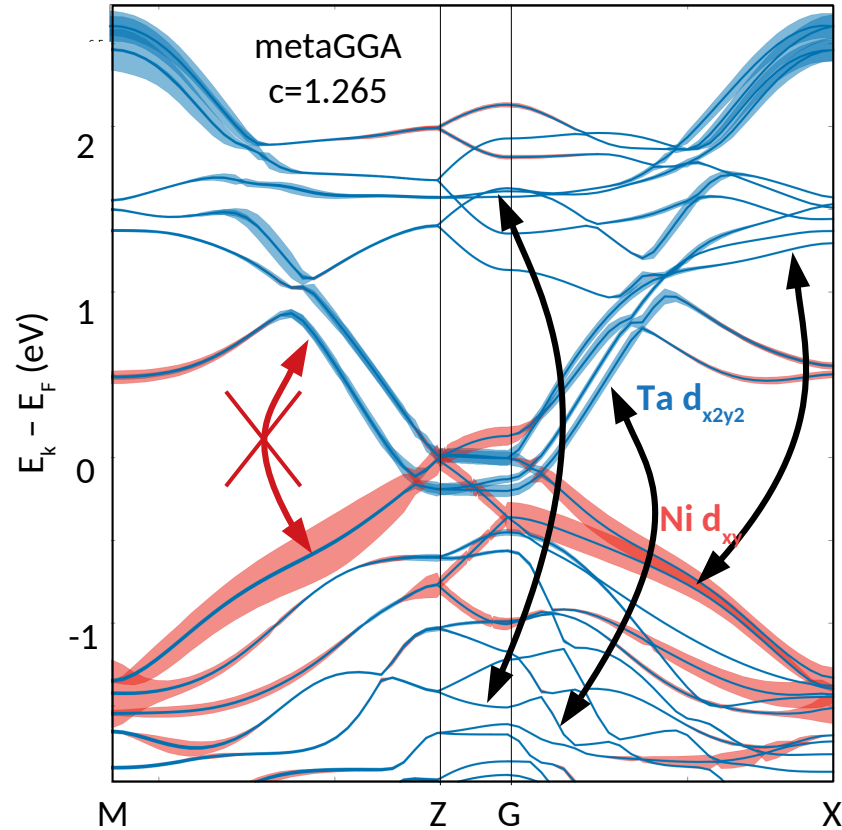


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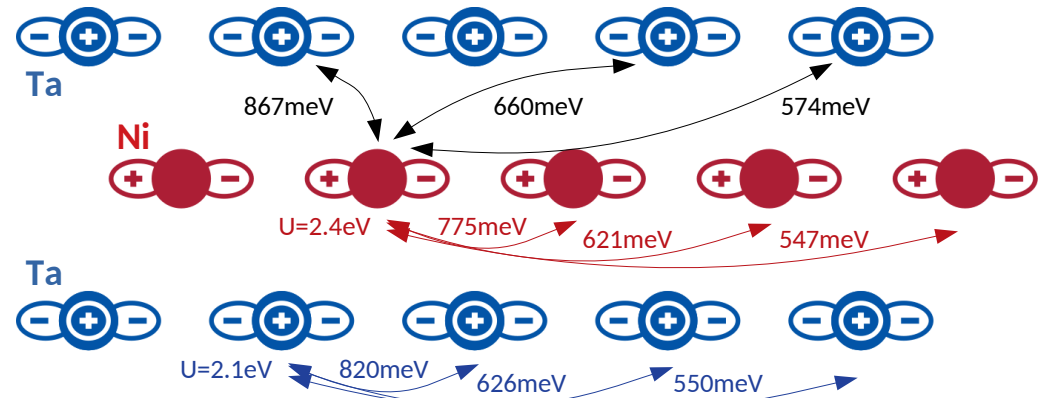
→ but not at Gamma! $H_{\alpha\beta}(k') = \sum_{\mathbf{R}} e^{i\mathbf{k}'\cdot\mathbf{R}} t_{\alpha\beta}(\mathbf{R})$

→ independently on gap / functional:

Ta₂NiSe₅ - Material-Realistic Model

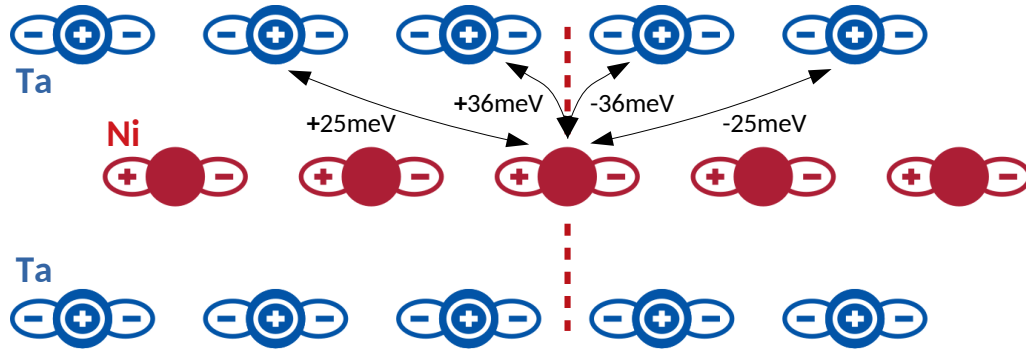


- Coulomb matrix elements
 - Careful: include “background” screening from neglected bands
→ cRPA values are needed!



- slowly decaying / long-range Ta-Ta and Ni-Ni interactions
- and long-range Ta-Ni interactions
- exchange elements negligible

Ta₂NiSe₅ – Microscopic Order Parameter



- Falicov-Kimball model:

$$H = H_0 + H',$$

$$H_0 = \sum_{\mathbf{k}} \varepsilon_v(\mathbf{k}) d_{\mathbf{k}}^\dagger d_{\mathbf{k}} + \sum_{\mathbf{k}} \varepsilon_c(\mathbf{k}) c_{\mathbf{k}}^\dagger c_{\mathbf{k}},$$

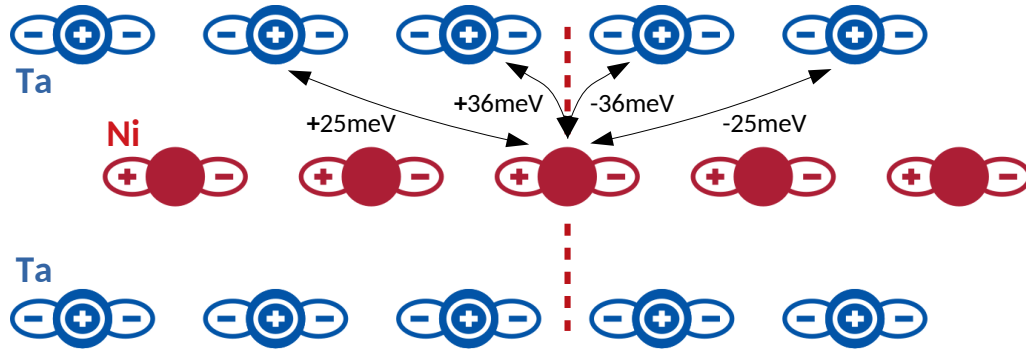
$$H' = \sum_{\mathbf{q}} \sum_{\mathbf{k} \mathbf{k}'} V_c(\mathbf{q}) d_{\mathbf{k}+\mathbf{q}}^\dagger d_{\mathbf{k}} c_{\mathbf{k}'}^\dagger c_{\mathbf{k}'+\mathbf{q}'},$$

→ not enough anymore

→ additional terms are needed to describe Ta ↔ Ni
(c ↔ d) hybridization

→ which affects the definition of the order parameter

Ta₂NiSe₅ – Microscopic Order Parameter



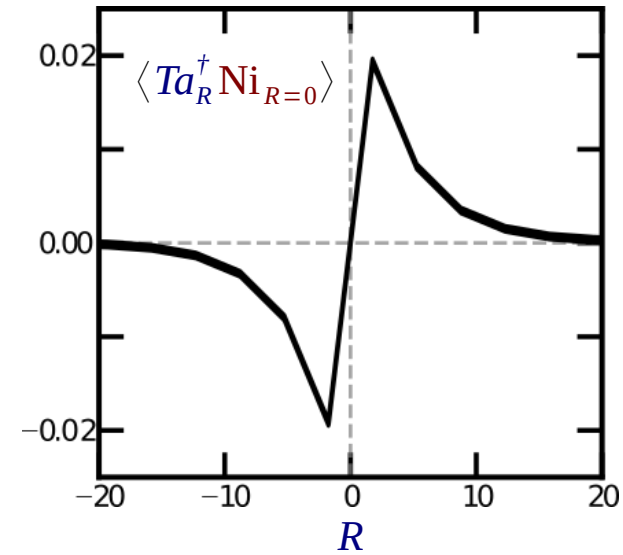
- by mirror symmetry:

$$\langle Ta_R^\dagger Ni_R \rangle = -\langle Ta_{R+1}^\dagger Ni_R \rangle$$

- new order parameter:

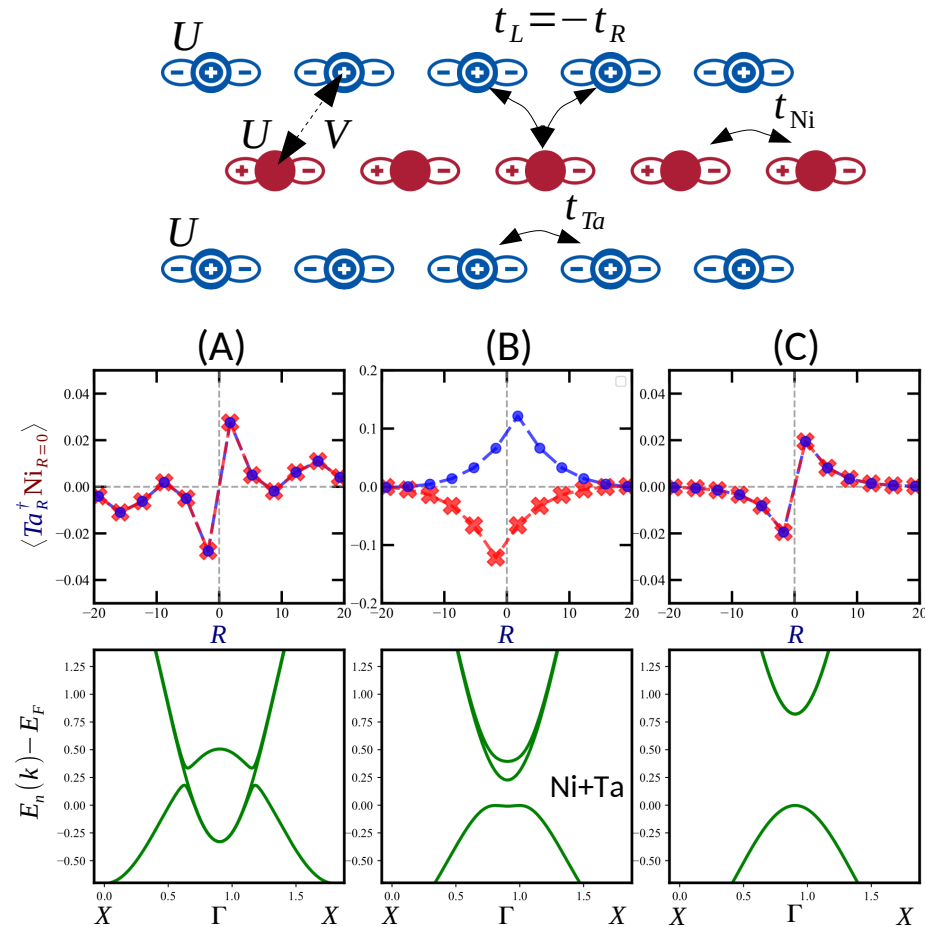
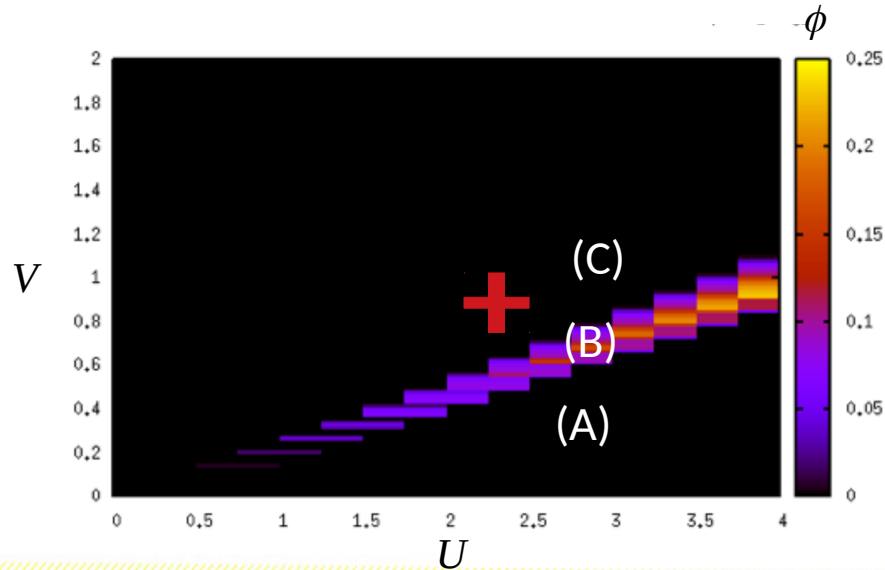
$$\phi = \langle Ta_R^\dagger Ni_R \rangle + \langle Ta_{R+1}^\dagger Ni_R \rangle$$

→ Test this definition for a the most simple toy model!



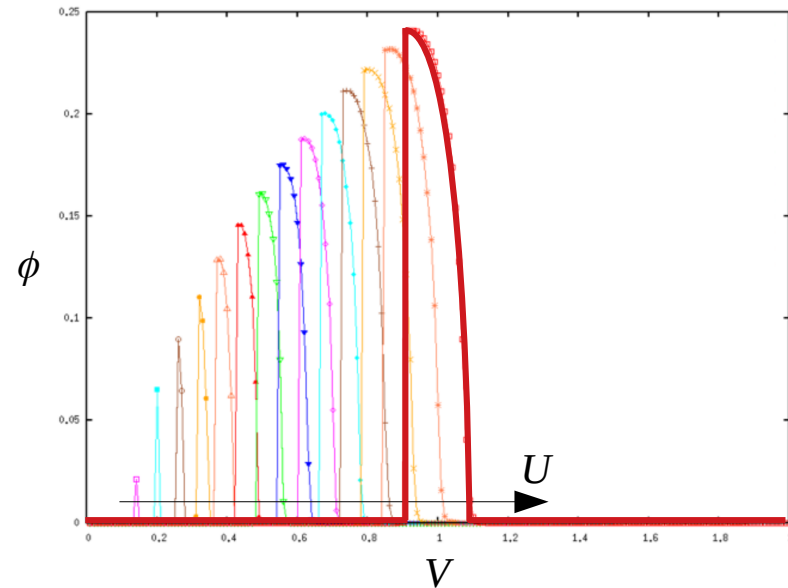
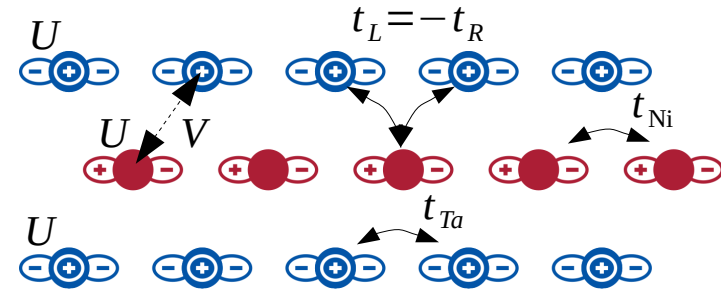
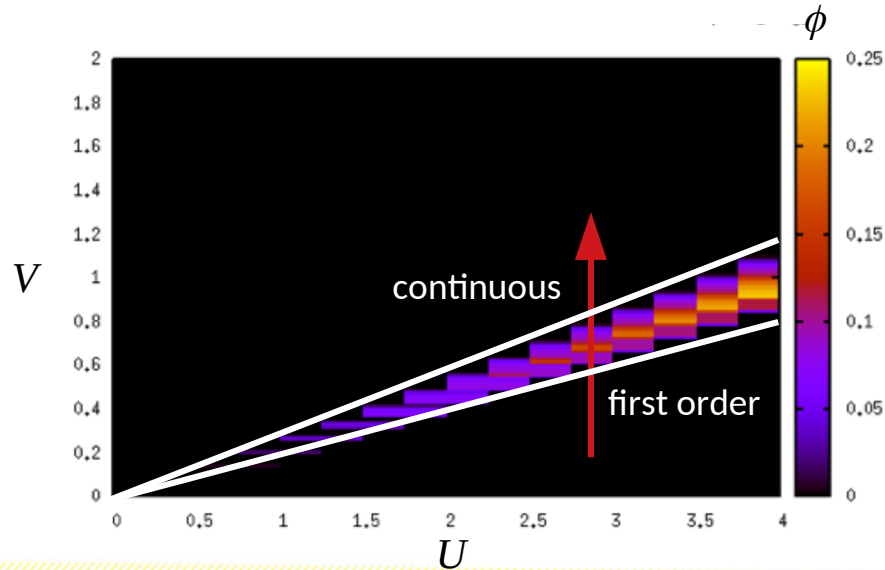
Ta₂NiSe₅ – Microscopic Order Parameter

- Use minimal 1D model
- with Ta / Ni hoppings similar to full model
- onsite Coulomb interaction U
- and Ta-Ni Coulomb interaction V
- calculate: $\phi = \langle Ta_R^\dagger Ni_R \rangle + \langle Ta_{R+1}^\dagger Ni_R \rangle$



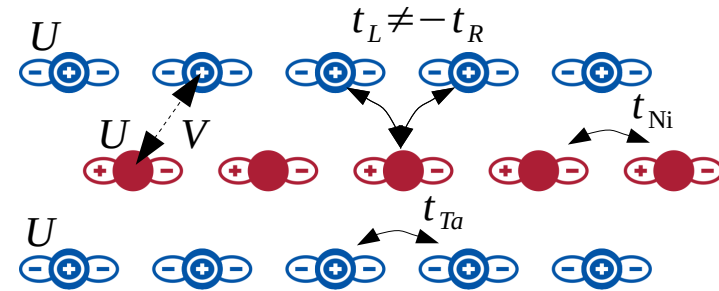
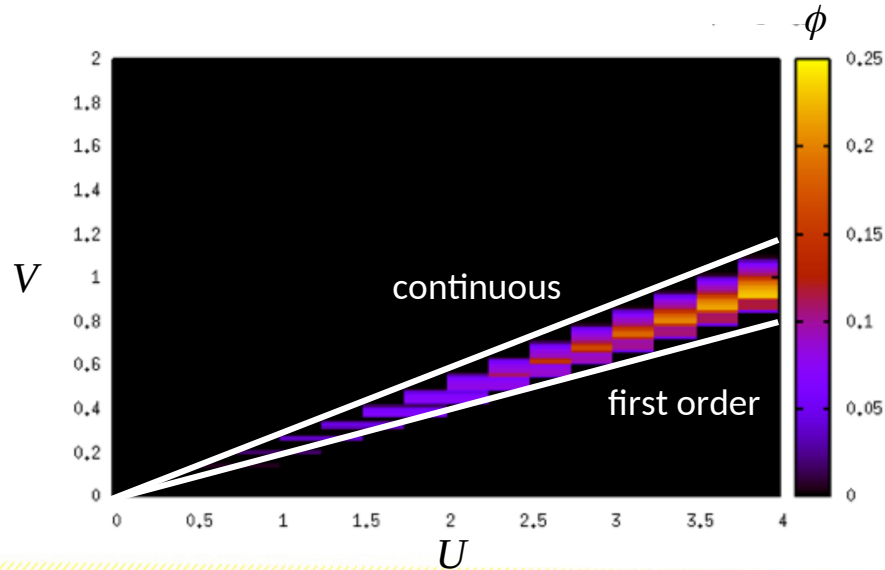
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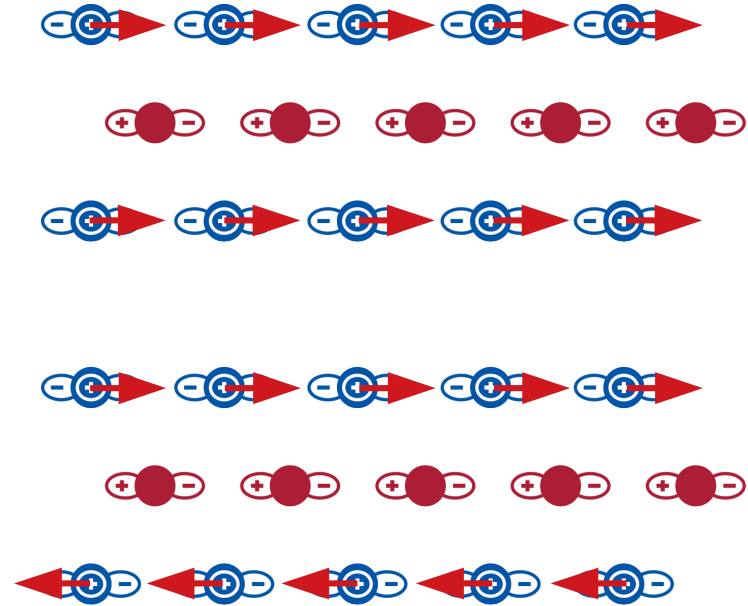
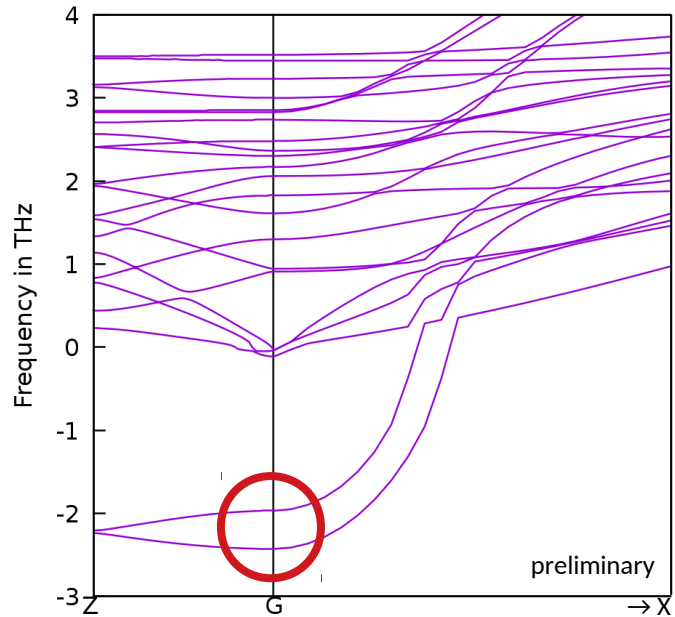


→ Purely electronically-driven symmetry breaking!

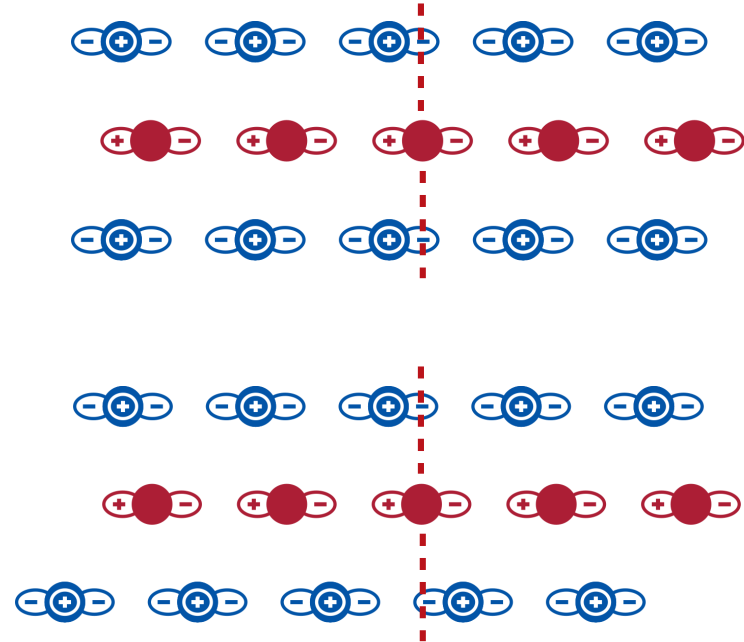
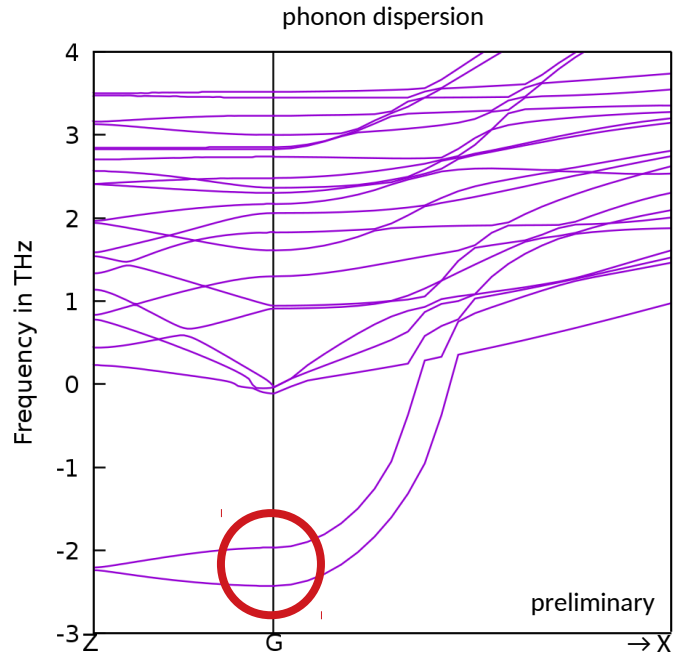
$$\langle Ta_R^\dagger Ni_R \rangle \neq -\langle Ta_{R+1}^\dagger Ni_R \rangle$$

→ What about coupling to the lattice?

Ta₂NiSe₅ - Ab Initio Lattice Structure

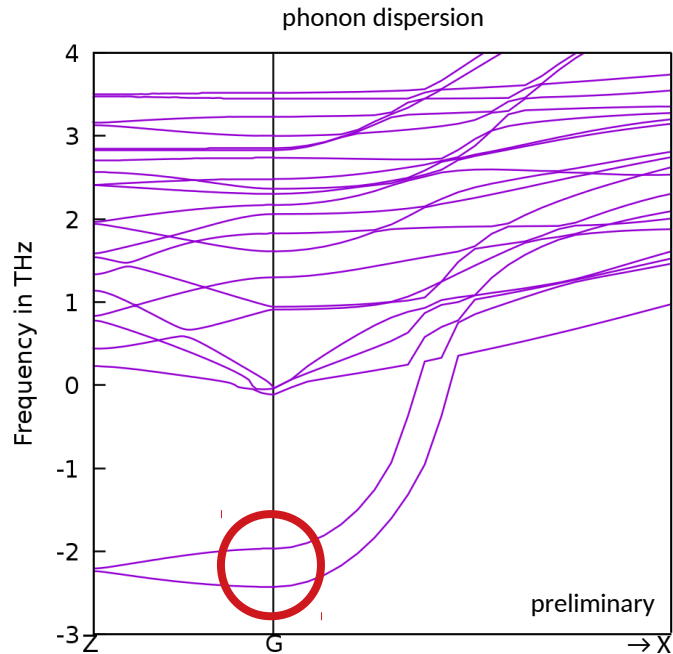


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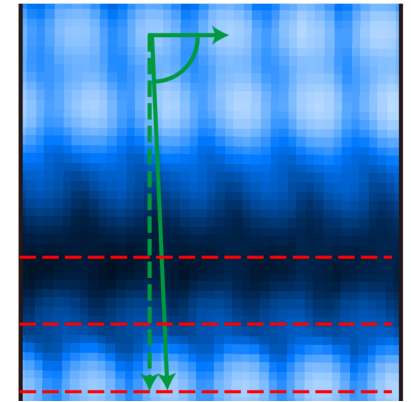
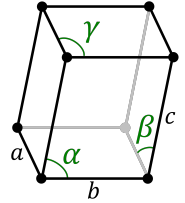
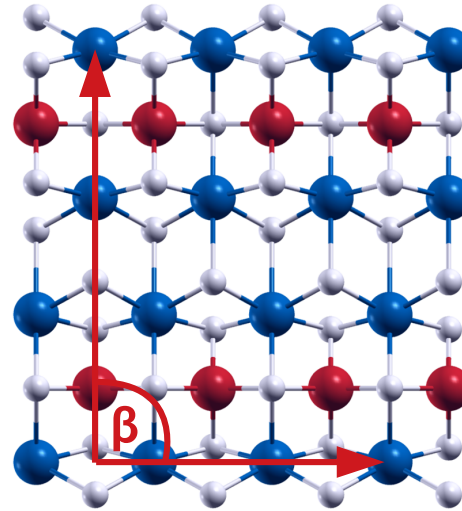
→ Both unstable phonon modes break the mirror symmetry!

Ta₂NiSe₅ – Ab Initio Lattice Structure



- full relaxation yields triclinic structure with

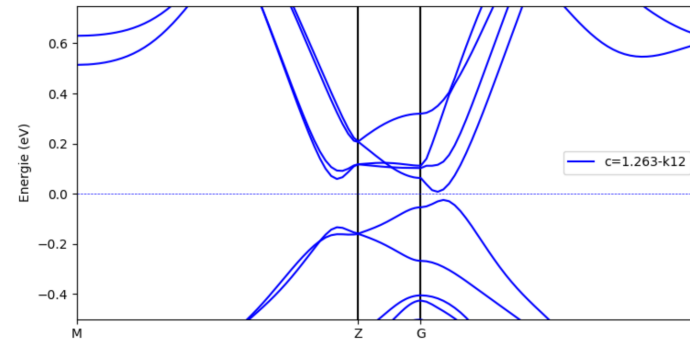
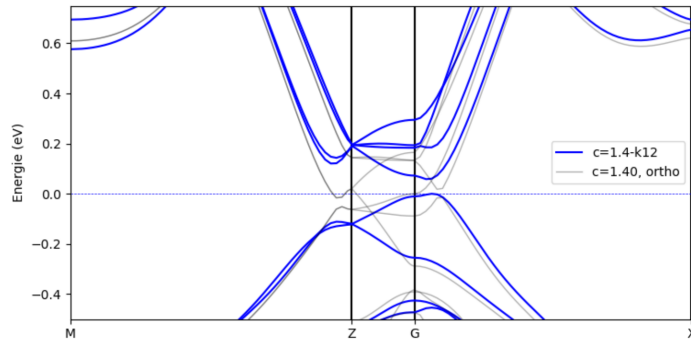
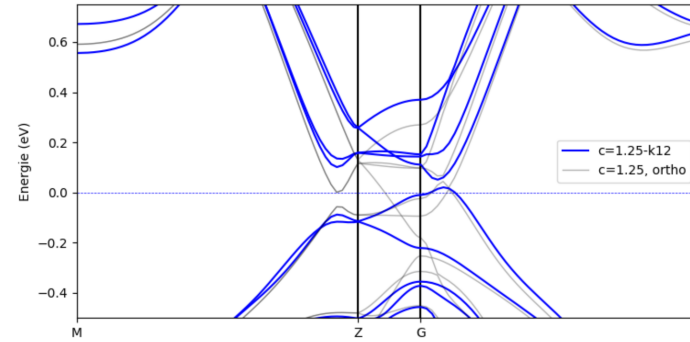
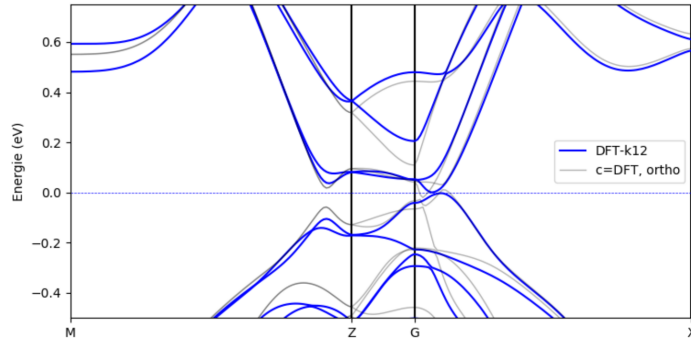
$$\alpha = 90.15^\circ \quad \beta = 90.57^\circ \quad \gamma = 76.0^\circ$$



Lee et al., PRB 99, 075408 (2019)

- lateral effects are the same as in monoclinic distortion
- too small in comparison to experimental values?

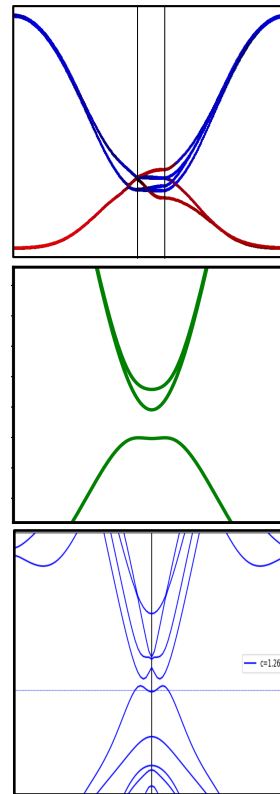
Ta₂NiSe₅ – Ab Initio Lattice Structure



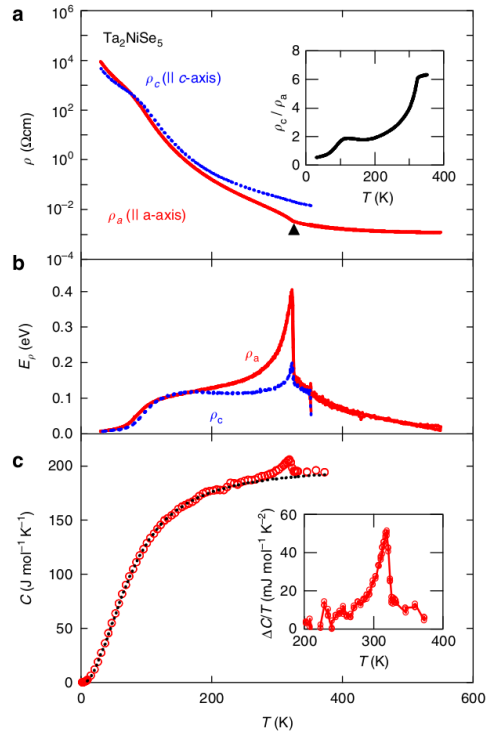
→ triclinic structure & Coulomb interaction open gap!

Ta₂NiSe₅ – Summary

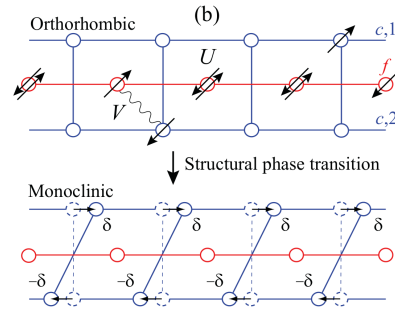
- Derived a minimal, material-realistic 6 band model from first principles
 - persistent finite Ta ↔ Ni hybridization
 - sizable and long-range Coulomb interactions
- Introduced a new order parameter
 - HF calculations show purely electronically driven symmetry breaking
 - opening gap & introduces flat band
- Lattice instability on its own not strong enough to open sizable gap



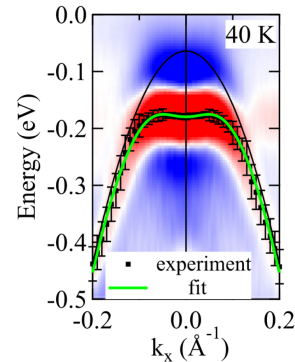
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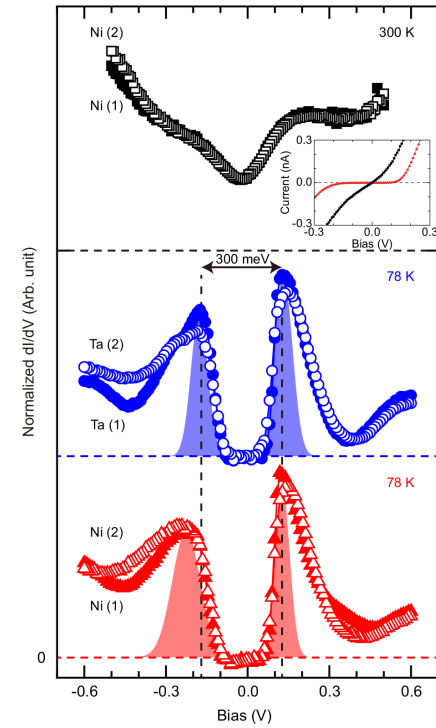
Lu et al., Nat. Comm. 8, 14408 (2017)



Kaneko et al., PRB 87, 035121 (2013)



Seki et al., PRB 90, 155116 (2014)



Lee et al., PRB 99, 075408 (2019)

