Building a beyond-DFT database of spectral functions for correlated materials

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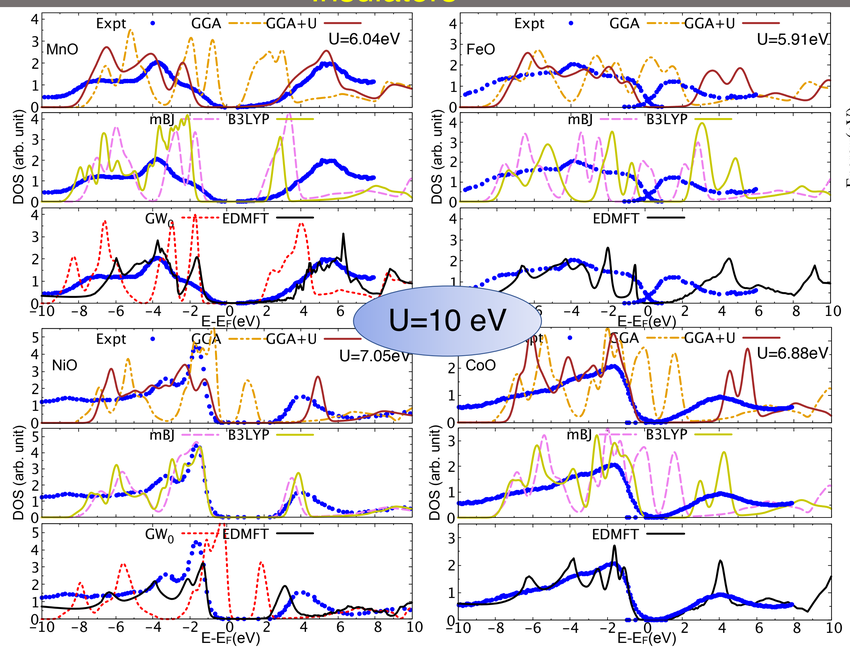
Motivations

- ❑ Most of the materials databases are built with DFT which often predicts incorrect results
- ❑ Many beyond-DFT methods are available: meta-GGA, hybrid-functionals, GW, DFT+DMFT
- ❑ Which one is more appropriate for building up database for correlated materials?
- ❑ Systematic test for strongly correlated materials is required

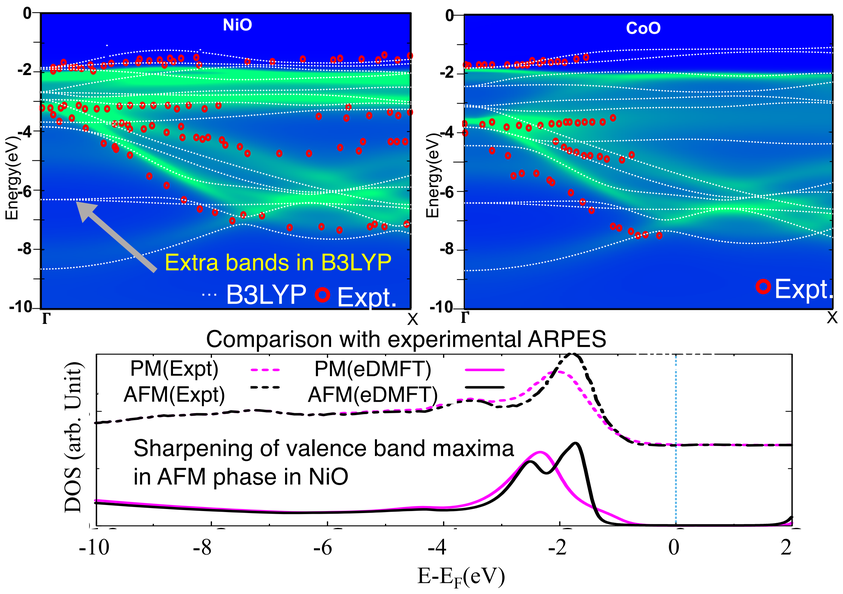
Computational Details

- ❑ All-electron: Wen2k
- ❑ Starting point: LDA
- ❑ Meta-GGA: mBJ
- ❑ Hybrid-DFT: B3LYP
- ❑ DFT+DMFT: eDMFT
- ❑ GW: G_0W_0 software: PyGW
- ❑ Experimental crystal structure: ICSD

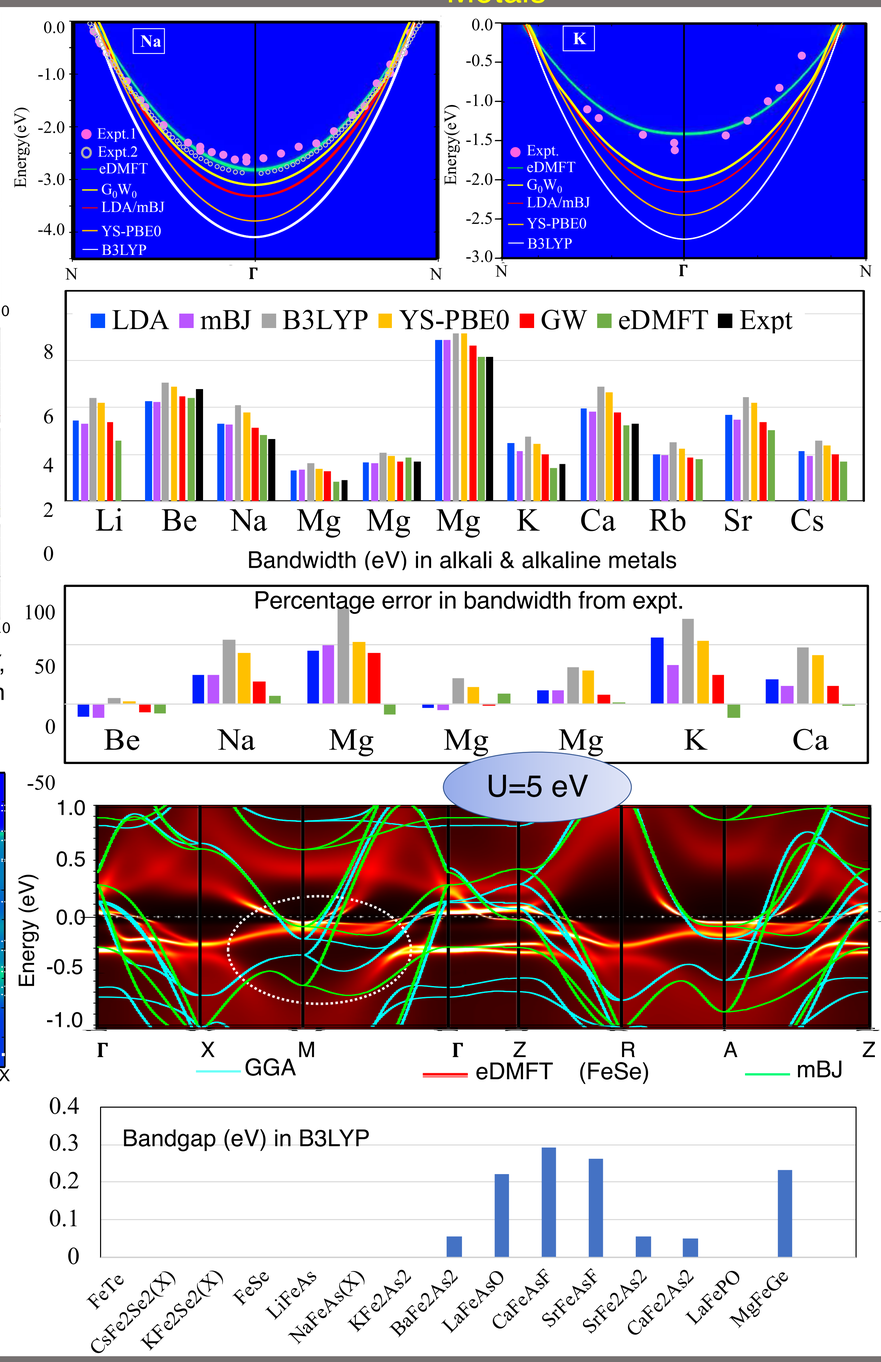
Insulators



Total density of states (states/eV) as computed in GGA, GGA+U, mBJ, B3LPY, and eDMFT. Blue dots indicate photoemission and inverse photoemission (PES/IPES) data in arbitrary unit as obtained from literature.



Metals



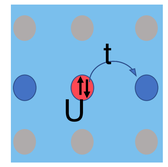
How to find U in DFT+DMFT ?

$$H = -t \sum_{<i,j>,\sigma} (c^{\dagger}_{i,\sigma} c_{j,\sigma} + c^{\dagger}_{j,\sigma} c_{i,\sigma}) + U \sum_{i=1}^{N} N^{I\uparrow} N^{I\downarrow}$$

$$U^{I}_{nl} = IP^{I}_{nl} - EA^{I}_{nl} = E(N^{I}_{nl} + 1) + E(N^{I}_{nl} - 1) - 2E(N^{I}_{nl})$$

For 3d transition metal with configuration $3d^n4s^1$:

$$U = [E(3d^{n-1}4s^2) - E(3d^n4s^1)] + [E(3d^{n+1}4s^0) - (3d^n4s^1)]$$



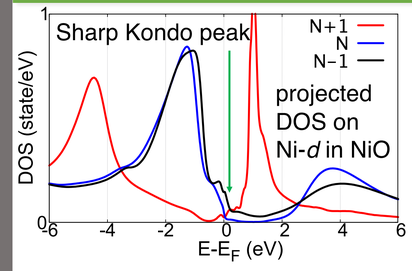
Traditional ways of Computing U

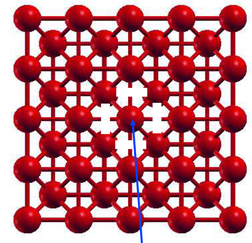
- ❖ Constrained-LDA (c-LDA): $U = \frac{\partial^2 E(n)}{\partial n^2}$
- ❖ Linear response approach: $U = \frac{\partial^2 E(n)}{\partial n^2} - \frac{\partial^2 E^{KS}(n)}{\partial n^2}$
- ❖ Constrained-RPA

Difficulties:
- ❖disentanglement of correlated bands
- ❖Strong dependence of energy window

U is found to be very sensitive for materials
→U is a tuning parameter in LDA+U

U in constrained-eDMFT





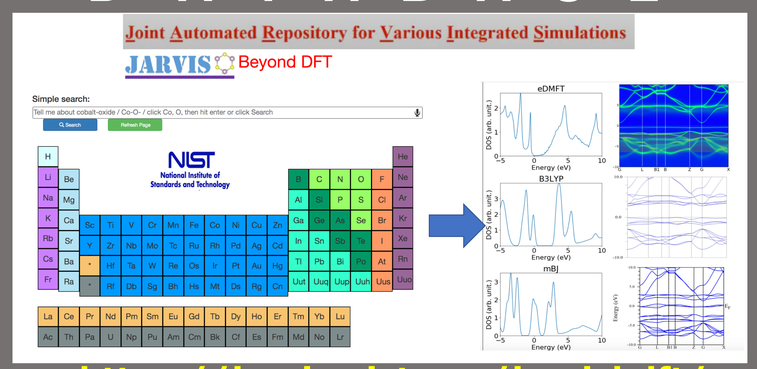
Impurity atom with constrained occupation

Using total energy in eDMFT

Mott insulators: U= 10 eV

3*d*-metals: U=5 eV

D A T A B A S E



https://jarvis.nist.gov/jarvisbdft/

Conclusions

- ❑ A new paradigm with wide range of methods applied to prototypical materials
- ❑ B3LYP and eDMFT perform best for Mott Insulators
- ❑ Hybrid functionals perform worst for metals
- ❑ Computed U in eDMFT is less sensitive for a given class of materials, allowing high-throughput eDMFT
- ❑ PES/IPES with computed U in eDMFT remarkably reproduce experimental spectral properties for pnictides, TMOs, Nickelets, 4d-semimetals, ordinary metals etc.

References

Binary TMOs: **Mandal**, Haule, Rabe, Vanderbilt; npj Comput. Mater. 5, 115 (2019)
Mandal, Haule, Rabe, Vanderbilt; Phys. Rev. B 100, 245109 (2019)
Alkali metals: **Mandal,** Haule, Rabe, Vanderbilt; arXiv:2101.03262 (2021)
PyGW: Haule & Mandal; arXiv:2008.07727
Review of JARVIS: K. Choudhary ..[**Mandal**, Haule, Rabe, Vanderbilt], *et al.* npj. Computer. Mater. 6, 173 (2020)