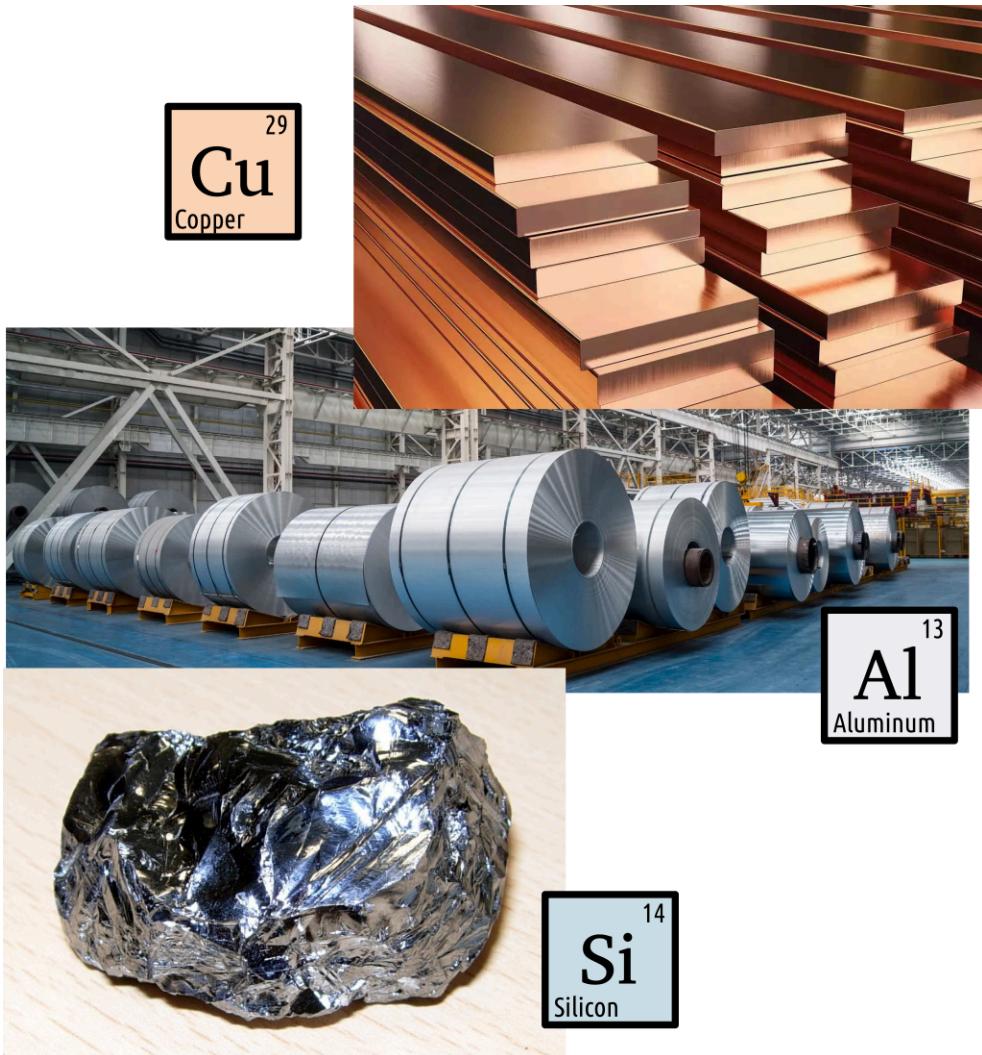


# All I need is U

A gentle introduction to DFT+Dynamical Mean Field Theory (DMFT)

Alberto Carta

# Strongly interacting materials

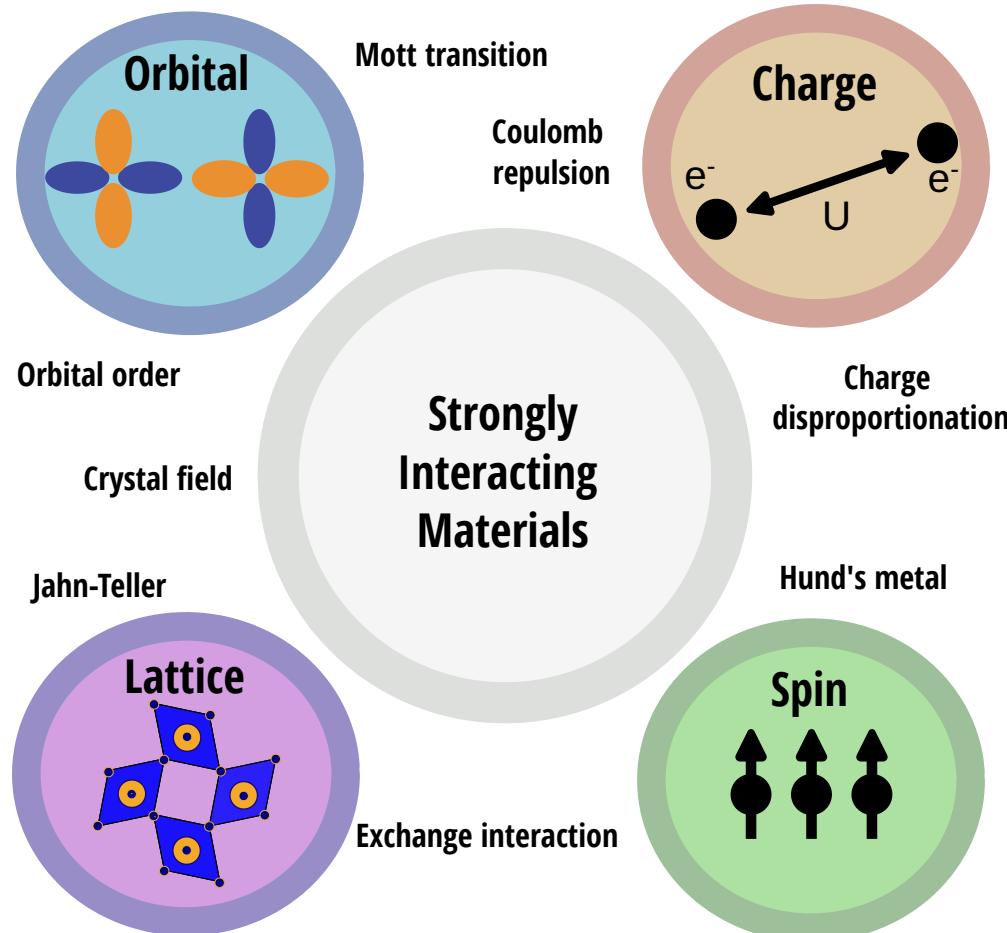


**Periodic Table of the Elements**

Atomic Number	Symbol	Name	Atomic Mass
1	H	Hydrogen	1.008
2	He	Helium	4.003
3	Li	Lithium	6.941
4	Be	Boron	9.012
5	Mg	Magnesium	24.305
6	Ca	Calcium	40.078
7	Sc	Scandium	44.955
8	Ti	Titanium	47.867
9	V	Vanadium	50.942
10	Cr	Chromium	51.996
11	Mn	Manganese	54.938
12	Fe	Iron	55.845
13	Co	Cobalt	58.933
14	Ni	Nickel	63.546
15	Cu	Copper	63.546
16	Zn	Zinc	65.41
17	Ga	Gallium	69.723
18	Ge	Germanium	72.631
19	As	Arsenic	74.922
20	Se	Selenium	78.972
21	P	Phosphorus	30.974
22	S	Sulfur	32.065
23	Cl	Chlorine	35.453
24	Ar	Argon	39.948
25	B	Boron	10.811
26	C	Carbon	12.011
27	N	Nitrogen	14.007
28	O	Oxygen	15.999
29	F	Fluorine	18.998
30	Ne	Neon	20.180
31	Al	Aluminum	26.982
32	Si	Silicon	28.085
33	Pt	Ptadium	196.97
34	Ge	Germanium	102.906
35	Br	Bromine	79.904
36	Kr	Krypton	84.798
37	Rb	Rubidium	85.468
38	Sr	Srtrium	87.62
39	Y	Yttrium	88.906
40	Zr	Zirconium	91.224
41	Nb	Nobium	92.906
42	Mo	Molybdenum	95.95
43	Tc	Technetium	98.907
44	Ru	Ruthenium	101.07
45	Rh	Rhodium	102.906
46	Pd	Palladium	106.42
47	Ag	Argentum	107.868
48	Cd	Cadmium	112.411
49	In	Indium	114.818
50	Sn	Tin	118.711
51	Sb	Antimony	121.760
52	Te	Tellurium	127.6
53	I	Iodine	126.504
54	Xe	Xenon	131.294
55	Cs	Cesium	132.905
56	Ba	Barium	137.328
57	La	Lanthanum	138.905
58	Ce	Cerium	140.116
59	Pr	Praseodymium	140.908
60	Nd	Neodymium	144.242
61	Sm	Proneethym	144.913
62	Eu	Europium	150.36
63	Gd	Gadolinium	151.964
64	Tb	Terbium	157.25
65	Dy	Dysprosium	162.500
66	Ho	Holmium	164.930
67	Er	Erbium	167.259
68	Tm	Thulium	168.934
69	Yb	Ytterbium	173.055
70	Lu	Lutetium	174.967
85	Ac	Actinium	227.028
89	Th	Thorium	227.028
91	Pa	Protactinium	231.036
92	U	Uranium	238.029
93	Np	Neptunium	237.048
94	Pu	Plutonium	244.064
95	Am	Americium	243.061
96	Cm	Berkellium	247.070
97	Bk	Berkeleium	247.070
98	Cf	Californium	251.080
99	Es	Einsteinium	254
100	Fm	Fermium	257.095
101	Md	Mendelevium	258.1
102	No	Nobelium	259.001
103	Lu	Lutetium	262

Materials containing d and f electrons:  
interplay localized atomic physics +  
delocalized band theory

# The Hubbard model



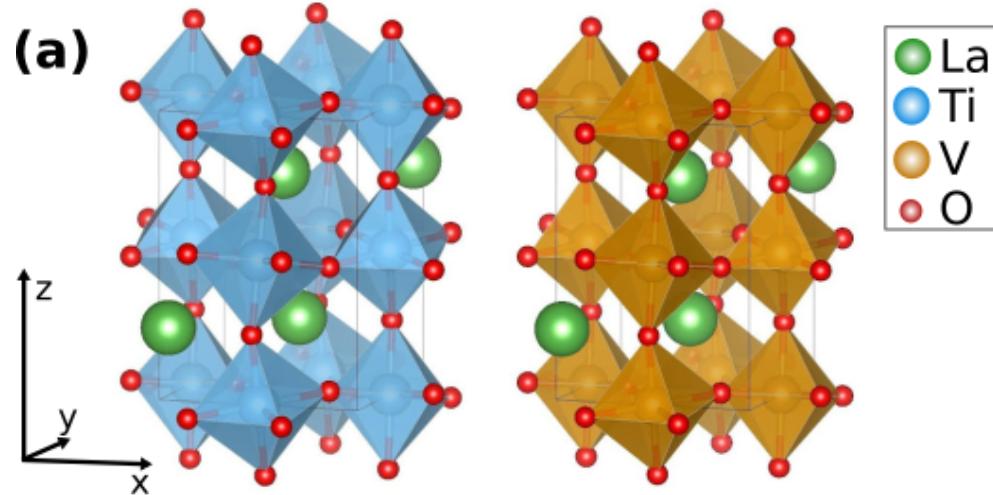
## Technological relevance:

- Transparent conductors:  $\text{SrMoO}_3$ ,  $\text{SrRuO}_3$
- Battery cathode materials:  $\text{LiCoO}_2$

## Promises:

- Motronic transistors: high on-off ratio [1]
- Photovoltaic devices beyond Shockley-Queisser limit

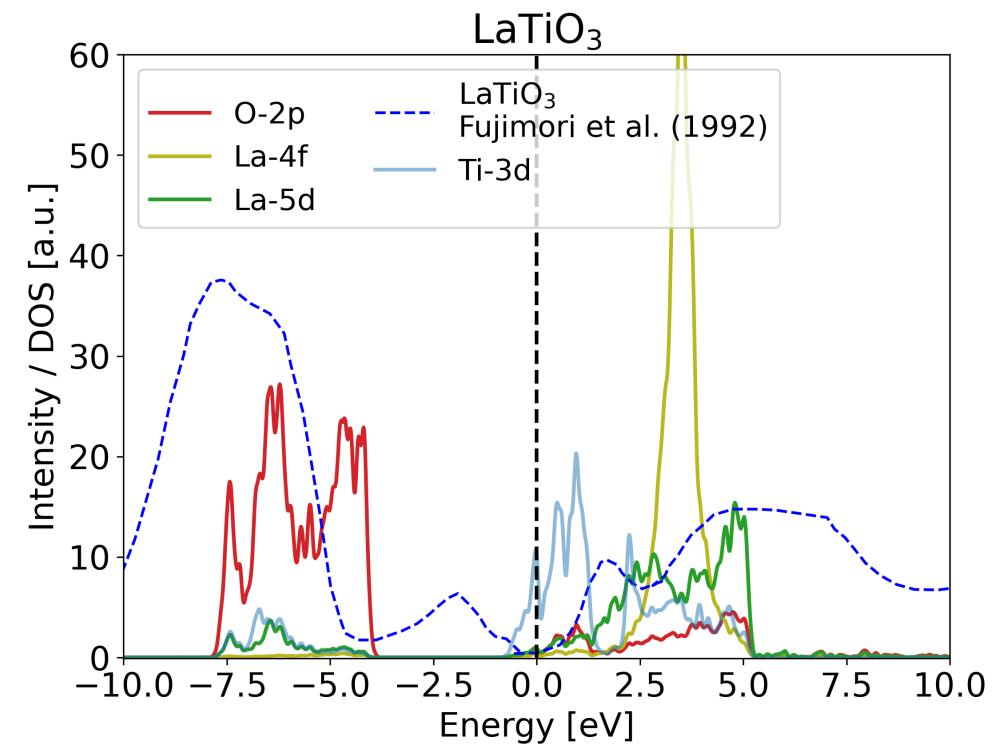
# Some simple examples



Some common examples:  $\text{LaVO}_3$ ,  $\text{LaTiO}_3$  or  $\text{NiO}$

Paramagnetic state (no magnetic order) is experimentally insulating

Nonmagnetic DFT calculation: no gap



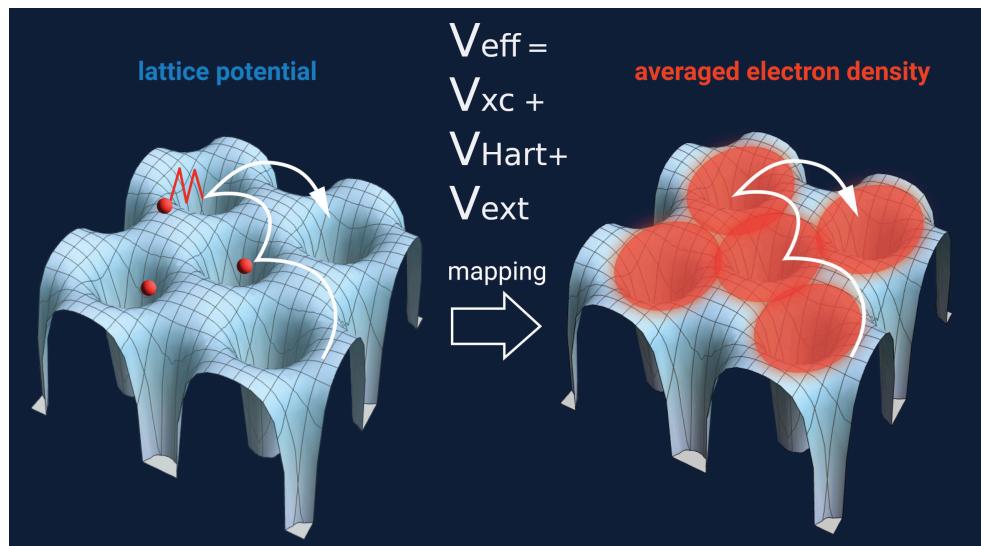
# Computational Techniques

## Density functional Theory (DFT)

Functional of the density

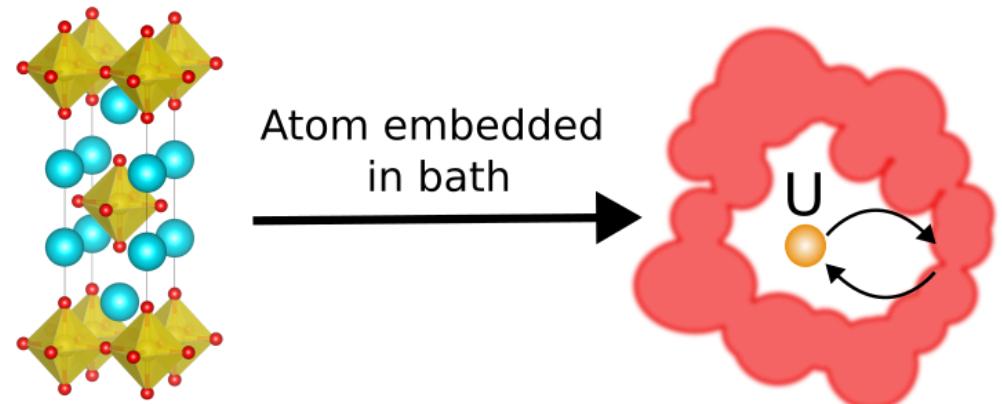


Kohn Sham mapping: effective potential



## Corrections:

- DFT+U (static local treatment)
- DFT+DMFT (dynamic local treatment)



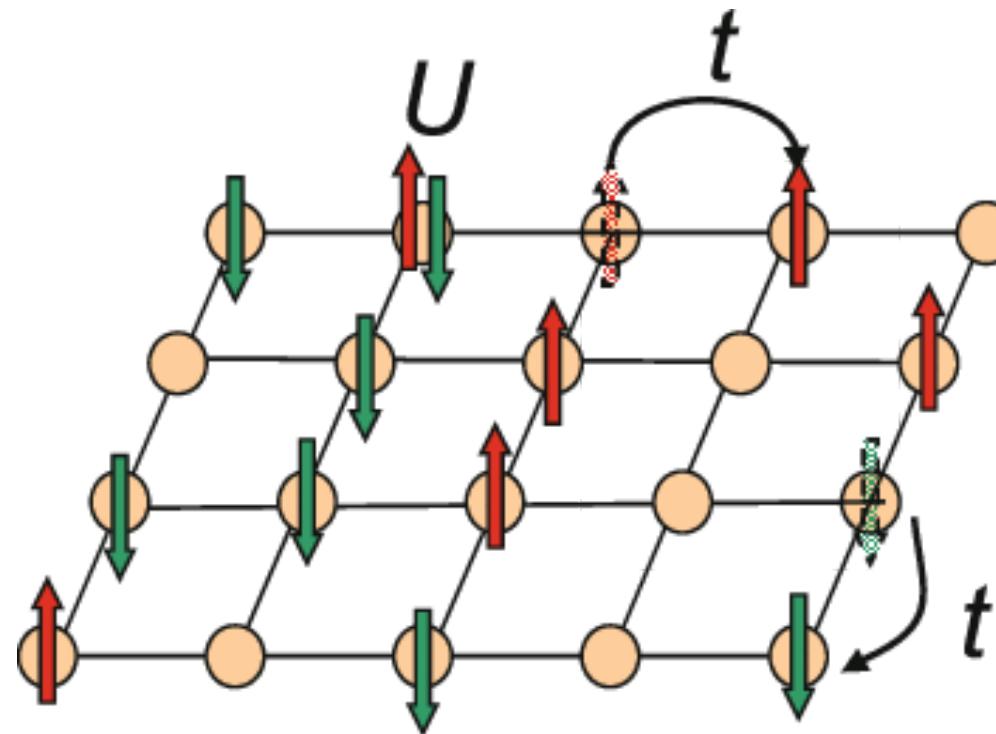
- Interaction condensed in Hubbard  $U$  parameter (Coulomb repulsion) and Hund  $J$  (spin alignment energy)

# In the next half an hour:

- What physics are we trying to describe? When is DFT+U enough?
- How to connect model calculations to real materials
- Practical side: TRIQS and [solid\\_dmft](#)

# What physics are we trying to describe?

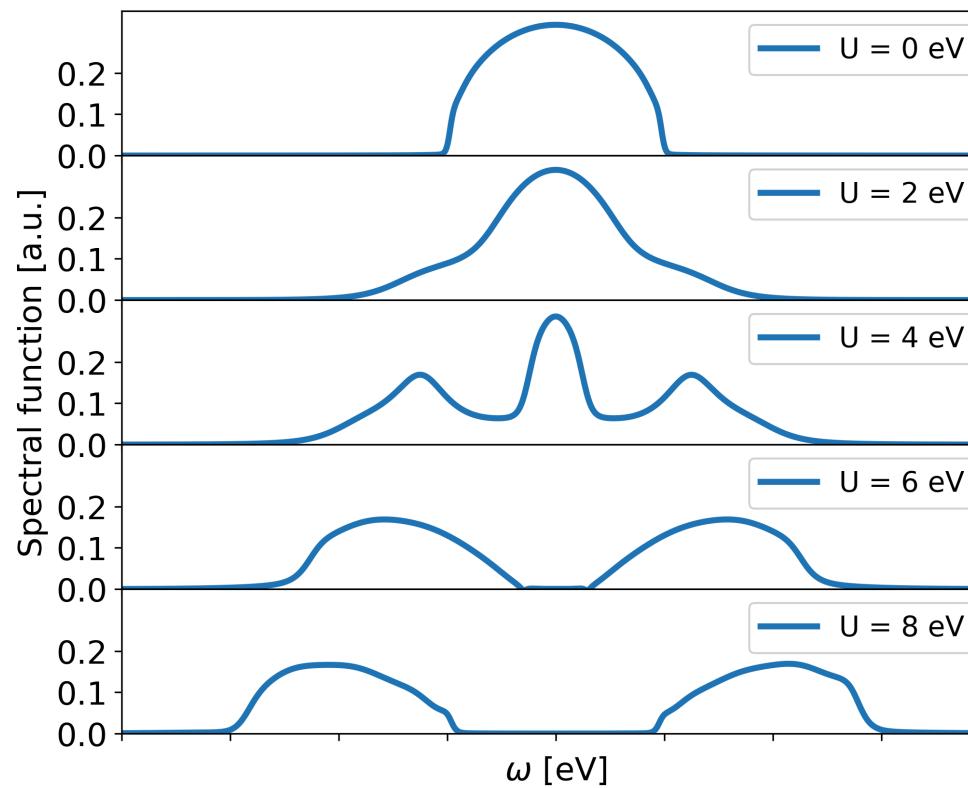
# The Hubbard model



$$H = -t \sum_{\langle i,j \rangle, \sigma} |i\sigma\rangle\langle j\sigma| + U \sum_i n_{i\uparrow} n_{i\downarrow}$$

- $i, j$  are site indices
- $\sigma$  is spin
- $n$  is electron number

# Mott insulators and correlated metals

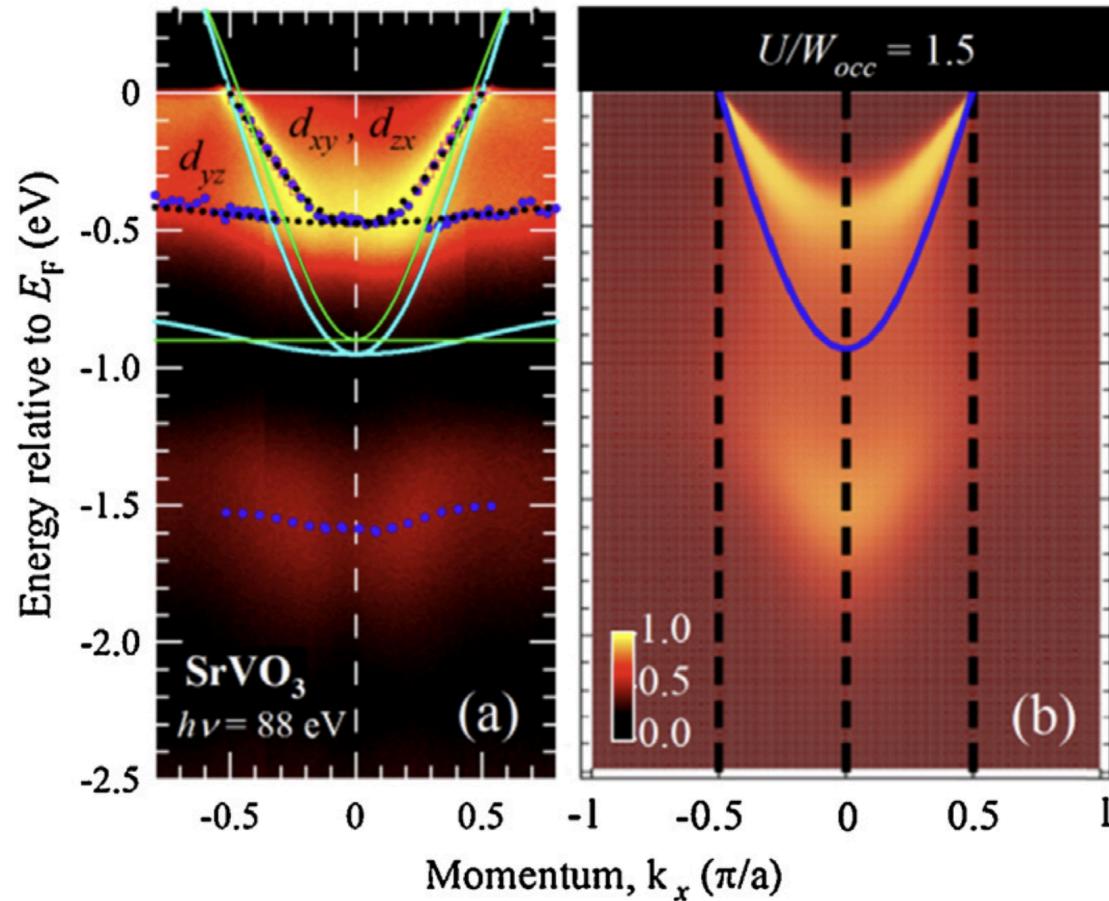


D = bandwidth, proportional to  $t$

Formation of hubbard bands and renormalization (squeezing) of the central peak

Quasiparticle weight:  $Z = \frac{D_{\text{central}}}{D}$

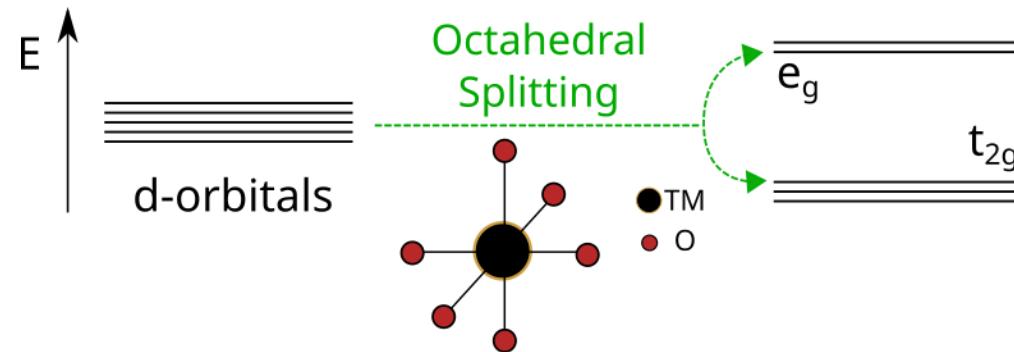
# Example: SrVO<sub>3</sub>



- Left: ARPES photoemission (“real” experimental bandstructure), light blue = LDA calculation
- Right: DMFT angle resolved spectral function

# Multiple orbitals

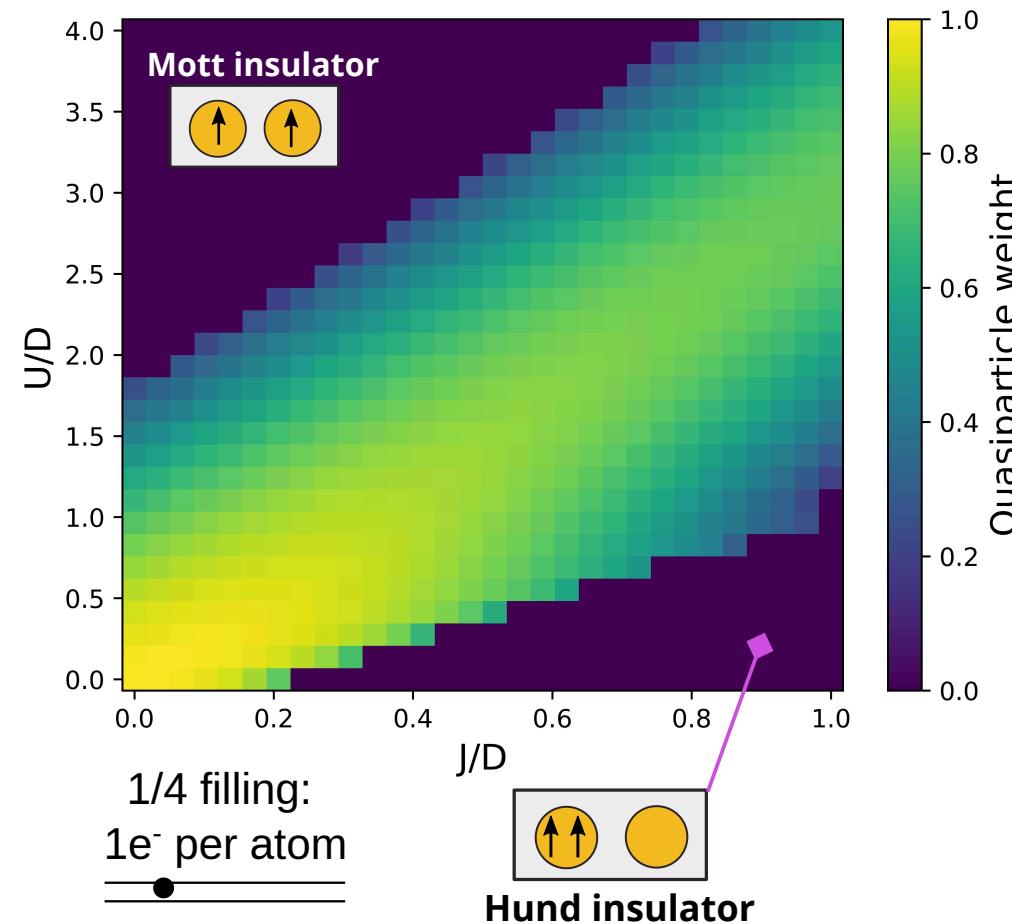
In real materials we have more than one orbitals: 5 d orbitals split by crystal field



Also J (energy of spin alignment) important

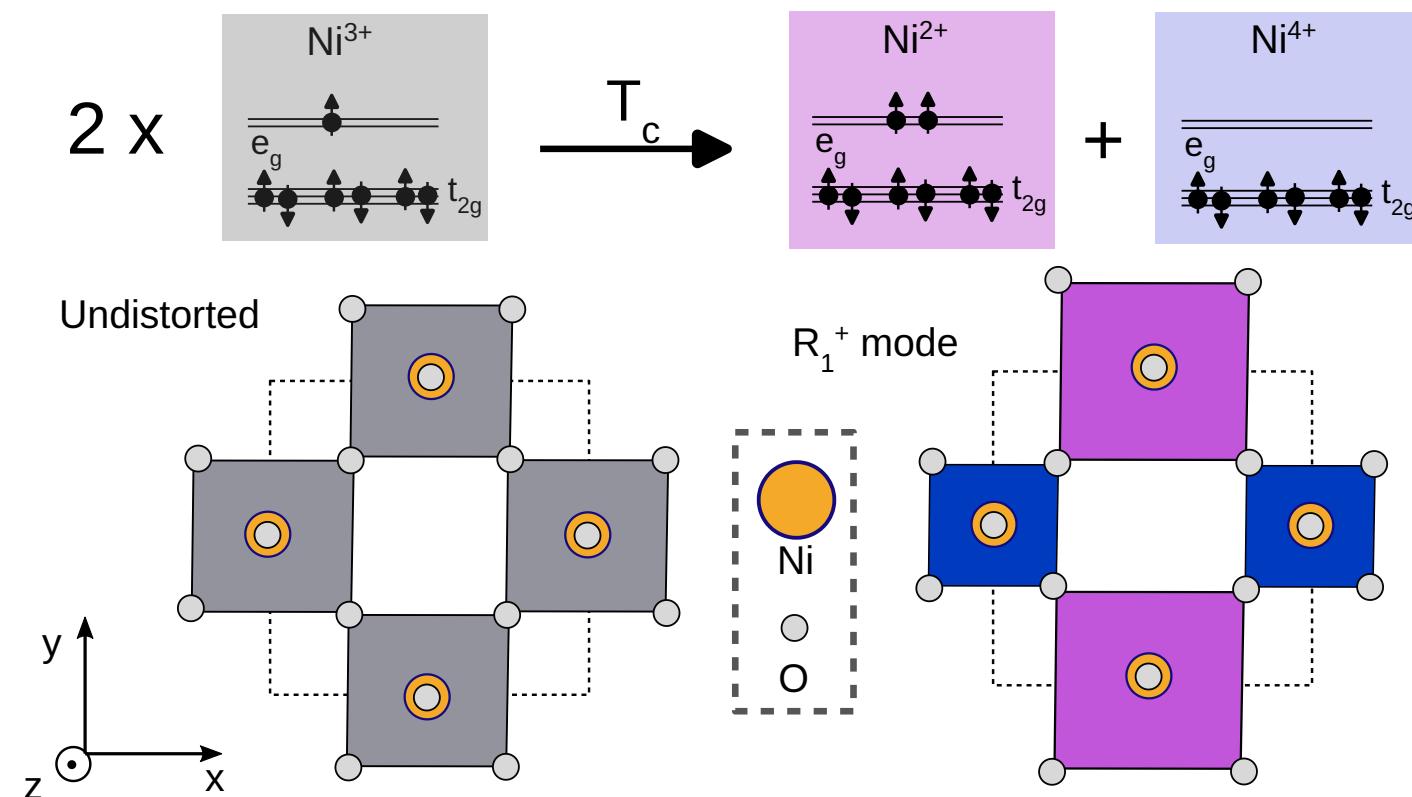
# Interplay of U and J

Two band model:  $e_g$  orbitals



# Practical examples: $\text{LuNiO}_3$ and other nickelates

In real materials electronic order couples to structure



# When is DFT+U good enough?

DFT+U successfully describes magnetically ordered state → antiferromagnets are well described (low temperature phase)



Whenever you can “freeze in” one atomic state, DFT+U is good!

**AFM state: “Frozen moment”**

$$|\psi_{local}\rangle = |\uparrow\uparrow\rangle_{site\ 1} + |\downarrow\downarrow\rangle_{site\ 2}$$

The average fock state is similar to the ground state

**Paramagnetic state: fluctuating moment**

$$\begin{aligned} |\psi_{local}\rangle = & \frac{1}{\sqrt{2}} |\uparrow\uparrow\rangle_{site\ 1} + \frac{1}{\sqrt{2}} |\downarrow\downarrow\rangle_{site\ 1} \\ & + \frac{1}{\sqrt{2}} |\uparrow\uparrow\rangle_{site\ 2} + \frac{1}{\sqrt{2}} |\downarrow\downarrow\rangle_{site\ 2} \end{aligned}$$

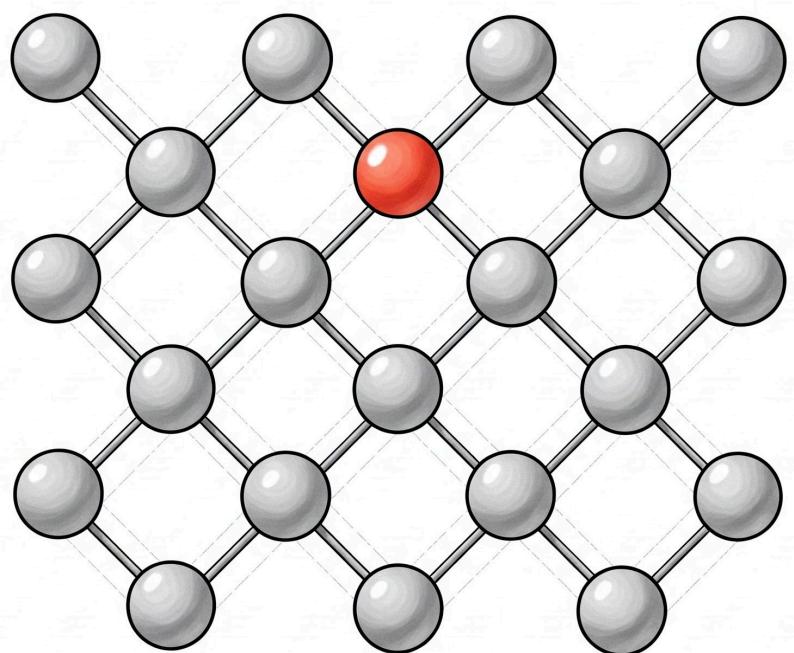
Need multiple determinants to describe

# DFT+DMFT in real materials

# DFT+U

Static contribution to the local potential

$$E_{tot} = E_{DFT}[\rho] + E_{Hubbard}[n_{\text{local}}] - E_{DC}[\rho]$$



Add a local **static** potential (  
 $V_{Hubbard}[n_{\text{local}}]$ ) to Kohn-Sham equations

$$V_{eff} = V_{ext}[\rho] + V_{xc}[\rho] + V_{Hubbard}[n_{\text{local}}] - V_{DC}[\rho]$$

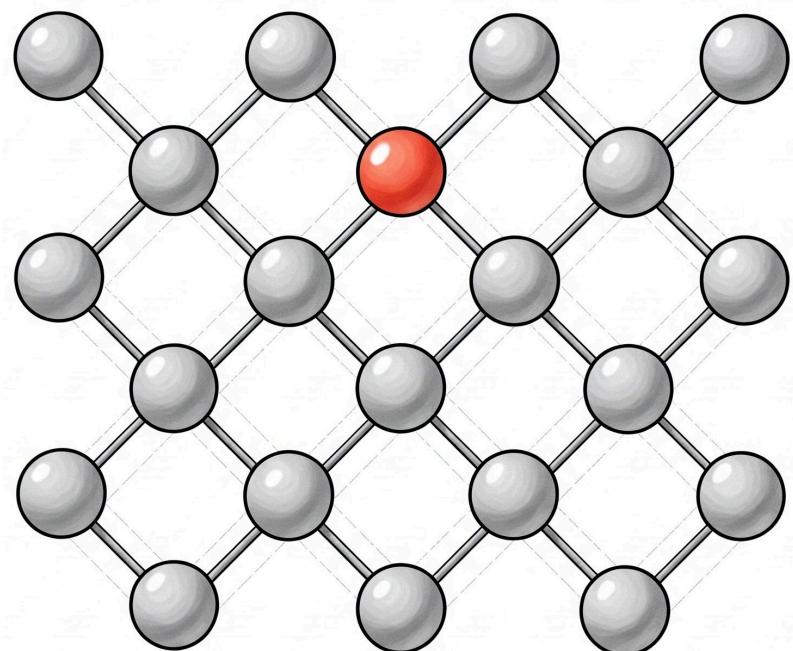
Converge two quantities iteratively:

- electronic density  $\rho$
- local density matrix  $n_{\text{local}}$

# DFT+DMFT

Dynamical contribution to the local potential (derived with free energy F instead of total energy E)

$$F_{tot} = E_{DFT}[\rho] + \Phi_{Hubbard}[G_{\text{local}}(\omega)] - E_{DC}[\rho]$$



Add a local **dynamic** potential (self energy  $\Sigma(\omega)$ ) to Kohn-Sham system

$$V_{eff} = V_{ext}[\rho] + V_{xc}[\rho] + \Sigma(\omega) - V_{DC}[\rho]$$

Converge two quantities iteratively:

- electronic density  $\rho$
- local green function  $G_{\text{local}}(\omega)$

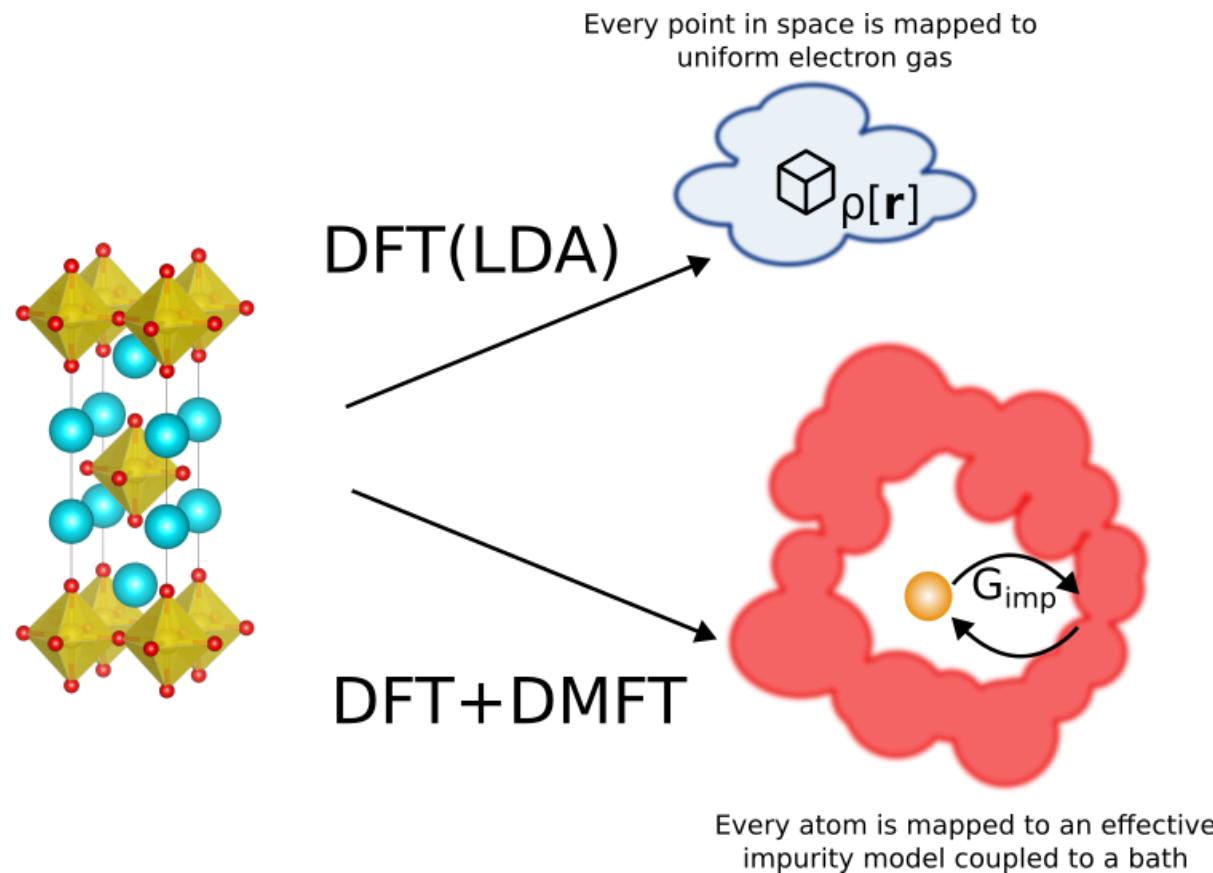
The green function describes the whole **LOCAL** spectral properties of an atom

DFT+DMFT is functional of the **whole LOCAL spectrum**, not just electronic density

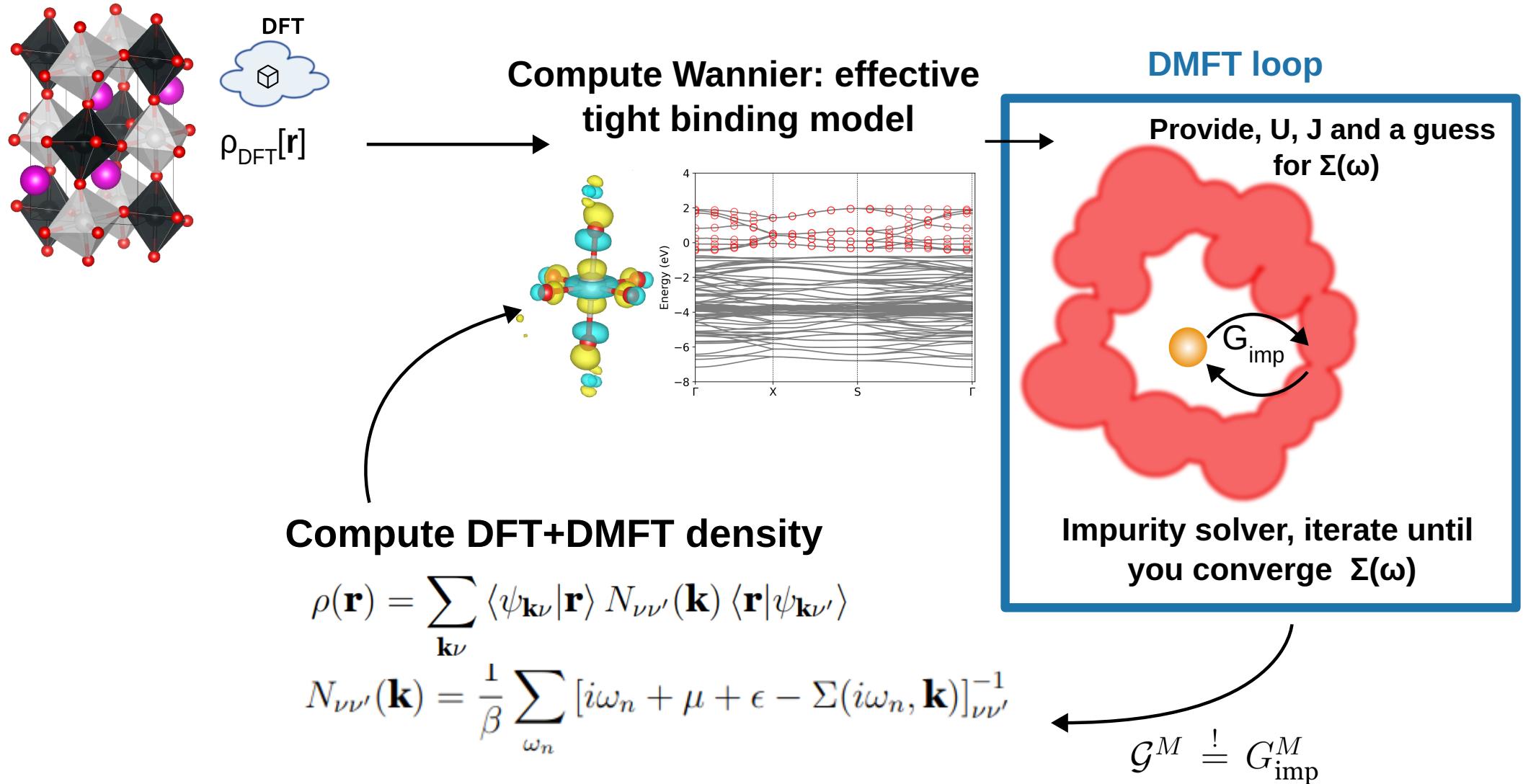
# DFT+DMFT

The local problem: we extract one atom from the lattice and treat the interaction only here

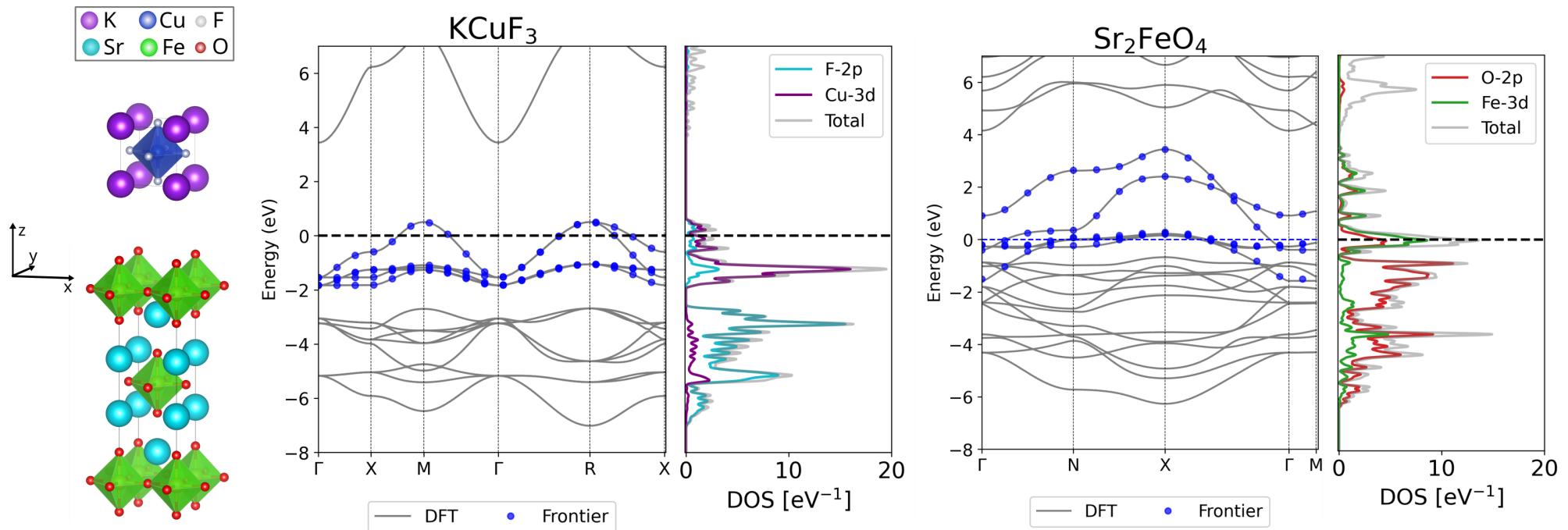
Atom + bath (rest of the crystal): Anderson impurity model



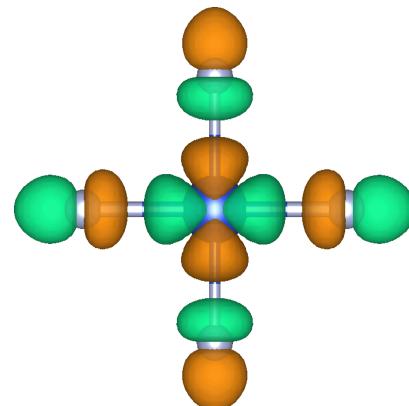
# DFT+DMFT complete cycle



# Local basis: Wannier functions

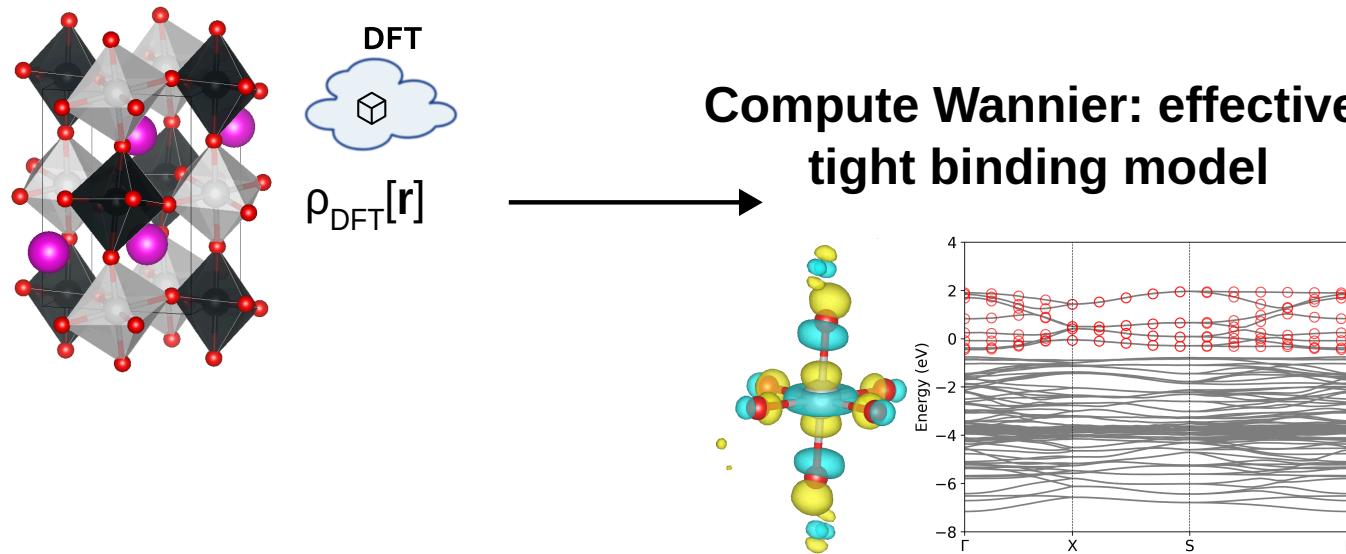


Wannierization gives you hopping between orbitals.



# DFT+DMFT: step 1

## Get the basis



# Computing the $U$ parameter

Both DFT+U and DFT+DMFT rely on the screened Coulomb interaction  $U$  between electrons on the same atomic site.

Often,  $U$  is treated as a fitting parameter:

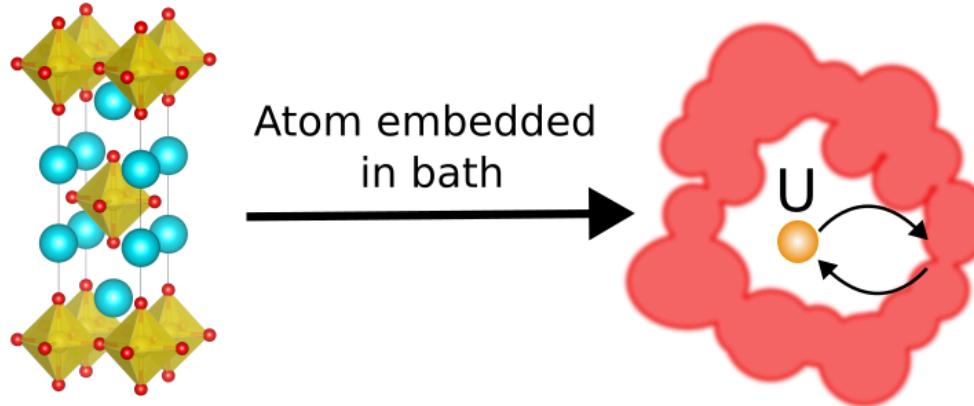
- Reproduce experimental bandgap, lattice constant, etc.
- This produces a scatter of possible  $U$  values in literature, especially if different DFT codes are used

Not satisfactory, we want to compute  $U$  ab-initio.

Two established methods used by different communities:

- Linear-response theory (LRT): **DFT+U community**
- Constrained random-phase approximation (cRPA): **DFT+DMFT community**

# Mapping to the impurity model



1. Compute total green function

$$G(\omega, \mathbf{k}) = [\omega - \epsilon_{KS}(\mathbf{k}) - \Sigma(\omega)]^{-1}$$

2. Get  $G_{\text{local}}$ : average G over BZ

$$G_{\text{local}}(\omega) = \frac{1}{N_k} \sum_{\mathbf{k}} G(\omega, \mathbf{k})$$

3. Get effective local noninteractive model  $G_0(\omega)$  (Weiss field)

$$G_{\text{local}}(\omega) = G_0(\omega) - \Sigma_n(\omega)$$

4. Using U, J and impurity solver, solve the problem and get  $G_{\text{imp}}(\omega)$ . Extract new guess for  $\Sigma$

$$G_{\text{imp}}(\omega) = G_0(\omega) - \Sigma_{n+1}(\omega)$$

5. Iterate from (1) until  $G_{\text{imp}} = G_{\text{local}}$

# Impurity solvers

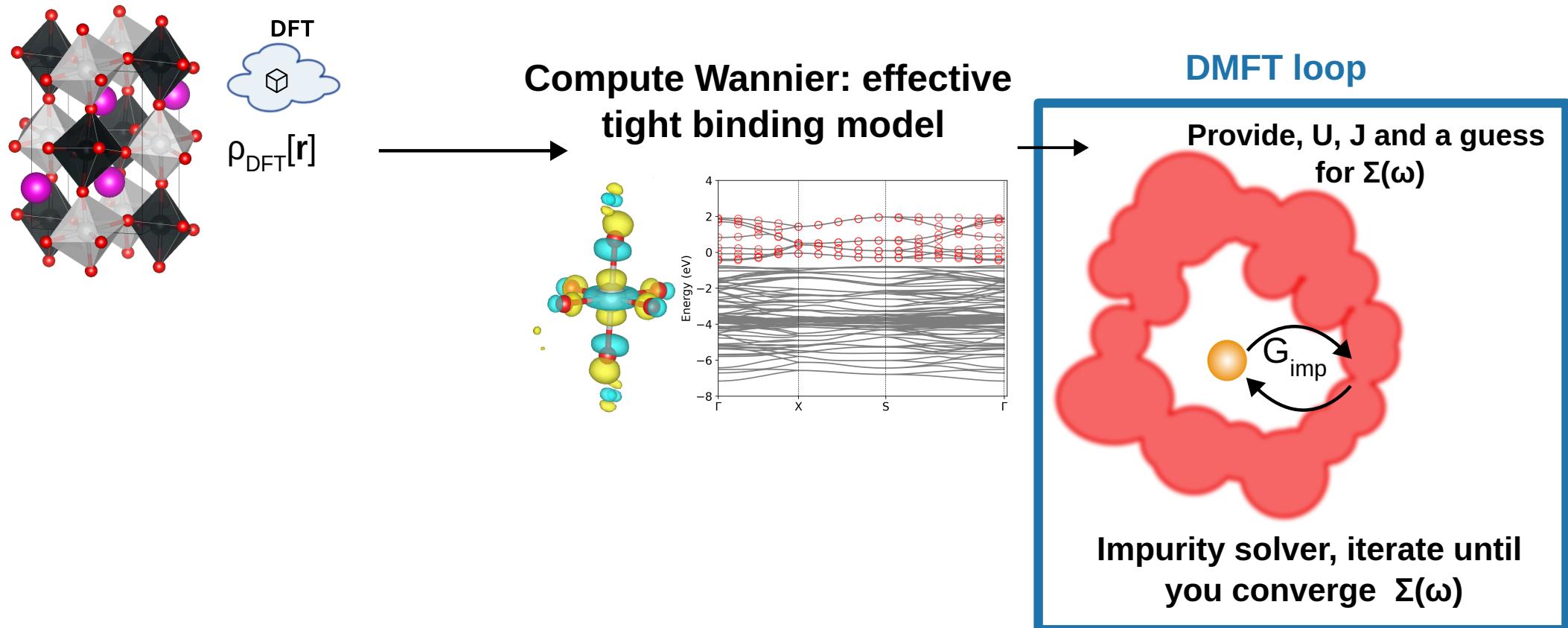
Computational bottleneck: exact solution can be very heavy to compute:

Types:

- Quantum Monte Carlo in imaginary time: sample Feynman diagrams
- Exact diagonalization with discrete bath
- Tensor network solvers
- Approximate solvers:
  - Hartree-Fock = DFT+U
  - Hubbard I: ignore hybridization
  - Finite order perturbation: WIP

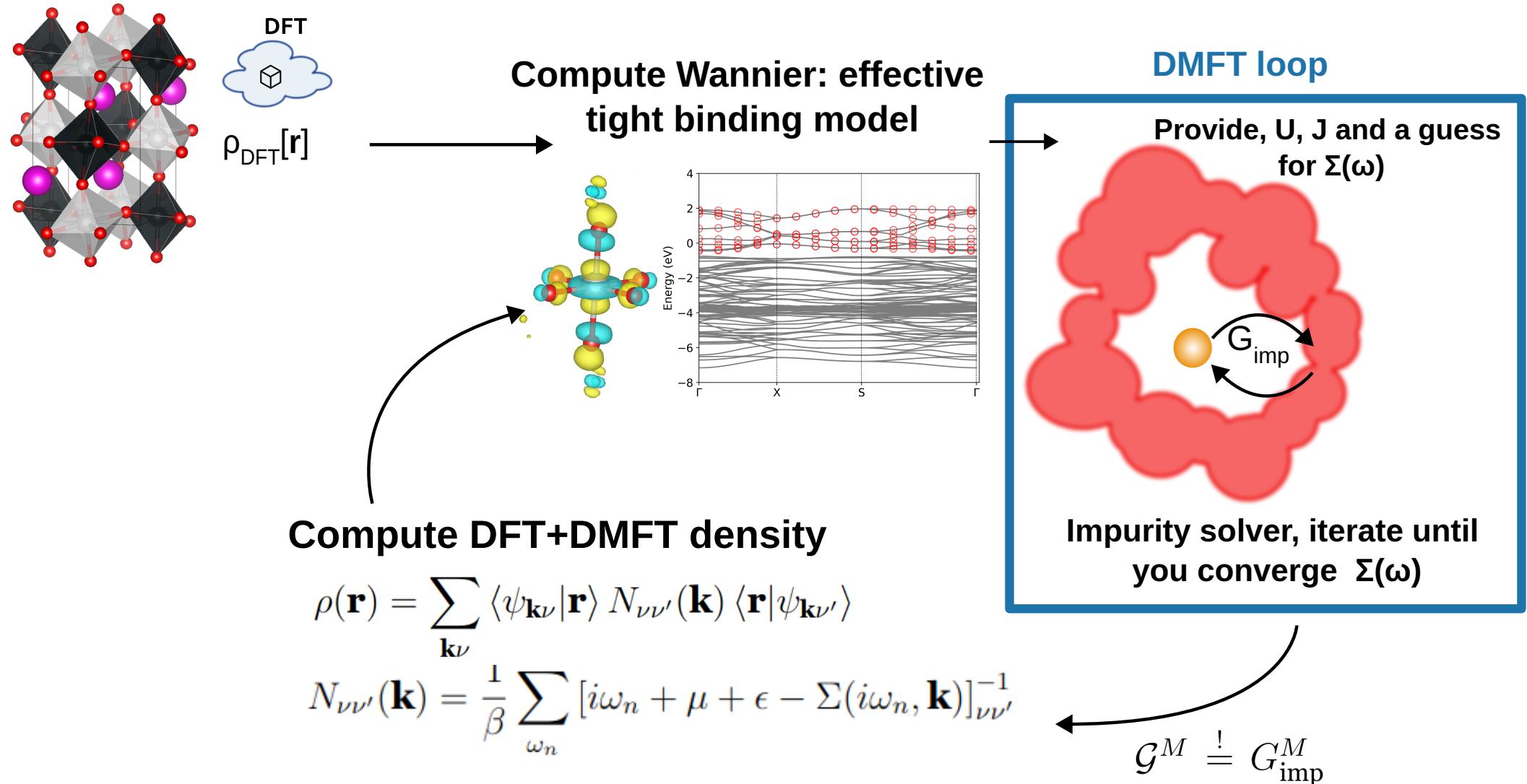
# DFT+DMFT: step 2

## Solve the impurity problem



# DFT+DMFT: closing the cycle

## Add DMFT charge correction



# Practical calculations

# solid\_dmft:

Developed by: Alex Hampel, Sophie Beck,  
me, Francesco Martinelli, Claude Ederer



Based on TRIQS we have developed a package that implements the DMFT cycle

Available on github:  
[https://github.com/flatironinstitute/solid\\_dmft](https://github.com/flatironinstitute/solid_dmft)

/ solid\_dmft

## solid\_dmft

This program allows to perform DFT+DMFT “one-shot” and charge self-consistent (CSC) calculations from h5 archives or VASP/Quantum Espresso input files for multiband systems using the [TRIQS software library](#), and the DFT code interface [TRIQS/DFTTools](#). Works with triqs >3.x.x. solid\_dmft takes advantage of various [impurity solvers available](#) in triqs: cthyb, HubbardI, ForkTPS, ctint, and ctseg. Postprocessing scripts are available to perform analytic continuation and calculate spectral functions.

### solid\_dmft 3.3.2

This is the homepage of solid\_dmft 3.3.2. For changes see the [changelog page](#). visit us on:

[GitHub](#)



# solid\_dmft

## Quantum espresso style input documentation

The aim of this section is to provide a comprehensive listing of all the input flags available for the `dmft_config.ini` input file. We begin by listing the possible sections and follow with the input parameters.

- [\[general\]](#): General parameters
- [\[solver\]](#): solver specific parameters
- [\[dft\]](#): DFT related inputs
- [\[GW\]](#): GW embedding inputs
- [\[advanced\]](#): Advanced inputs

Below an exhaustive list containing all the parameters marked by section.

**[general]**

```
afm_order; beta; block_threshold; broy_max_it; calc_energies; calc_mu_method; csc; dc; dc_dmft; dc_type; dlr_eps; dlr_wmax; enforce_off_diag;
eta; fixed_mu_value; g0_conv_crit; g0_mix; g0_mix_type; gimp_conv_crit; gw_embedding; h_field; h_field_it; ;;;;;;-1; h_int_basis; h_int_type;
h5_save_freq; J; jobname; load_sigma; load_sigma_iter; magmom; magnetic; mu_gap_gb2_threshold; mu_gap_occ_deviation; mu_initial_guess;
mu_mix_const; mu_mix_per_occupation_offset; mu_update_freq; n_iter_dmft; n_iter_dmft_first; n_iter_dmft_per; n_iw; n_tau; n_w;
noise_level_initial_sigma; occ_conv_crit; path_to_sigma; prec_mu; ratio_F4_F2; sampling_h5_save_freq; sampling_iterations; seedname; set_rot;
sigma_conv_crit; sigma_mix; store_solver; U; U_crpa_threshold; U_prime; w_range;
```

**[solver]**

```
type; idx_impurities; crm_dyson_solver; crm_dlr_wmax; crm_dlr_eps; delta_interface; diag_delta; fit_max_moment; fit_max_n; fit_max_w;
fit_min_n; fit_min_w; imag_threshold; legendre_fit; length_cycle; loc_n_max; loc_n_min; max_time; measure_chi_insertions; measure_chi;
measure_density_matrix; measure_G_l; measure_pert_order; move_double; move_shift; n_cycles_tot; n_l; n_warmup_cycles; off_diag_threshold;
perform_tail_fit; random_seed; length_cycle; max_time; measure_pert_order; move_double; n_cycles_tot; n_warmup_cycles; random_seed;
crm_dyson_solver; crm_dlr_wmax; crm_dlr_eps; diag_delta; fit_max_moment; fit_max_n; fit_max_w; fit_min_n; fit_min_w; improved_estimator;
```

# solid\_dmft

## Quantum espresso style input documentation

[View page source](#)

### [general]: General parameters

Frequently used parameters that apply to the whole simulation.

**afm\_order**

**type** = bool; **default** = False

copy self energies instead of solving explicitly for afm order

**beta**

**type** = float; **default** = None

inverse temperature. If set, solid\_dmft stores all Greens functions etc on an imaginary-frequency grid and also n\_iw and n\_tau have to be specified. If not set, it uses a real-frequency grid and eta, n\_w and w\_range have to be set

**block\_threshold**

**type** = float; **default** = 1e-5

threshold for finding block structures in the input data (off-diag yes or no)

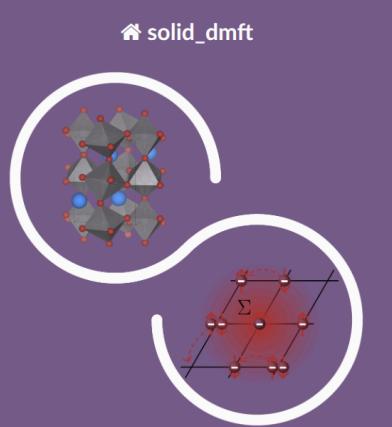
**broy\_max\_it**

**type** = int; **default** = -1

maximum number of iteration to be considered for broyden mixing. Only used if general.g0\_mix\_type='broyden'. 1 corresponds to simple linear mixing

# solid\_dmft

Extensive tutorials covering several use cases



solid\_dmft

3.3.2

[Search docs](#)

Installation Documentation

Tutorials

- 1. OS with QE/W90 and cthyb: SrVO<sub>3</sub> MIT
- 2. CSC with VASP PLOs: charge order in PrNiO<sub>3</sub>
- 3. CSC with QE/W90 and HubbardI: total energy in Ce<sub>2</sub>O<sub>3</sub>
- 4. OS with VASP/PLOs and cthyb: AFM state of NdNiO<sub>2</sub>
- 5. Plotting the spectral function

Support & contribute

Changelog

[/ Tutorials](#)

[View page source](#)

## Tutorials

These tutorials provide an overview about typical workflows to perform DFT+DMFT calculations with solid\_dmft. The tutorials are sorted by complexity and introduce one after another more available features.

**Note**

The tutorials are run with the 3.3.x branch of triqs. Please use the 3.3.x branch for triqs and all applications to reproduce the results shown here.

Short description of the tutorials linked below:

1. Typical one-shot (OS) DMFT calculation based on prepared hdf5 archive for SrVO<sub>3</sub>
2. Full charge self-consistent (CSC) DFT+DMFT calculation using the PLO formalism with Vasp for PrNiO<sub>3</sub>
3. Full CSC DFT+DMFT calculation using w90 in combination with Quantum Espresso utilizing the lighter HubbardI solver
4. OS magnetic DMFT calculation for NdNiO<sub>2</sub> in a large energy window for 5 d orbitals
5. Postprocessing: plot the spectral function after a DFT+DMFT calculation

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- 1. OS with QE/W90 and cthyb: SrVO<sub>3</sub> MIT
  - 1. Starting out with DMFT
  - 2. Looking at the Metal-Insulator Transition
  - 3. Refining the diagram
  - 4. Plotting the spectral function
  - 5 Visualizing the MIT
- 2. CSC with VASP PLOs: charge order in PrNiO<sub>3</sub>
  - 1. Running the initial scf DFT calculation
  - 2. Running the CSC DMFT calculations

# Thank you!

Questions?