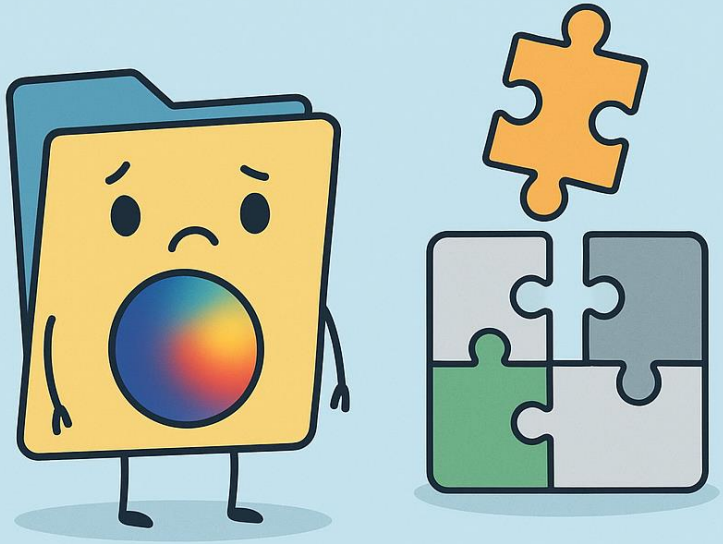
Towards Fully Functioning DMFT in SIESTA



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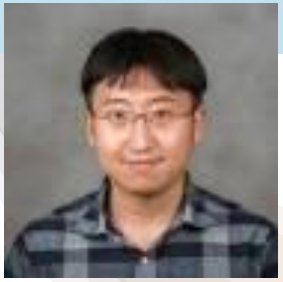
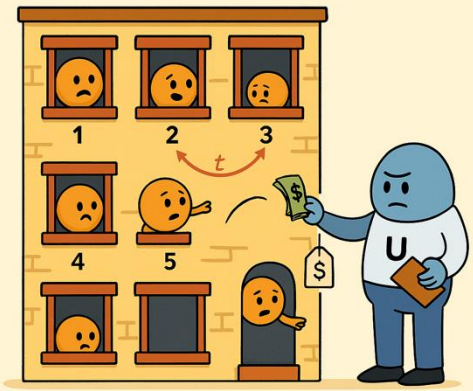
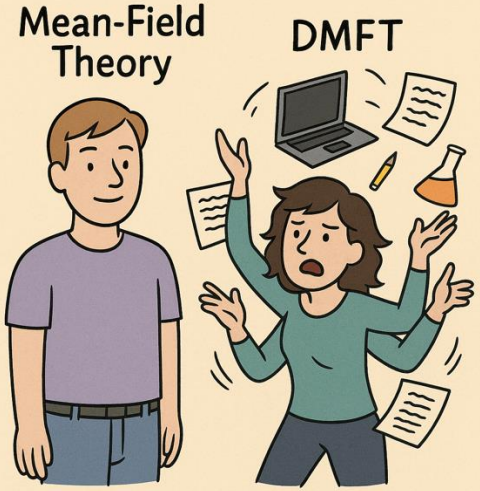


In collaboration:

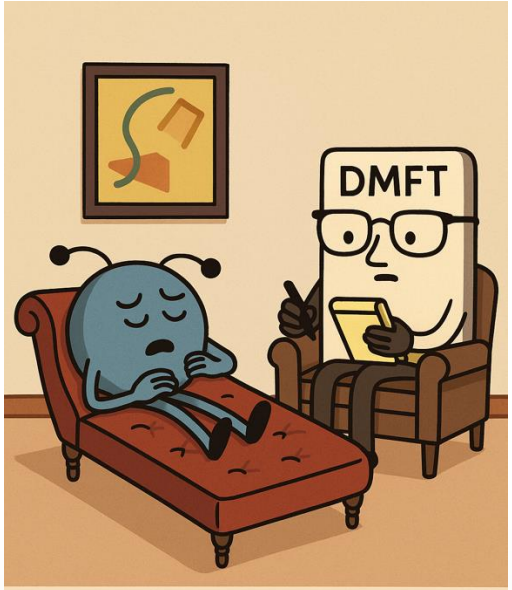
Hyowon Park, UIC, USA
Uthpalah Herath, Duke University, USA



In the Hubbard model, hopping comes at a cost.



Because somettimes, you need a dynamic approach to handle the drama.



Helping electrons deal with their complex relationships.

Our goal:

Having a fully self-consistent DMFT implementation within Siesta. This will offer a better description for strongly correlated materials.

Where are we?

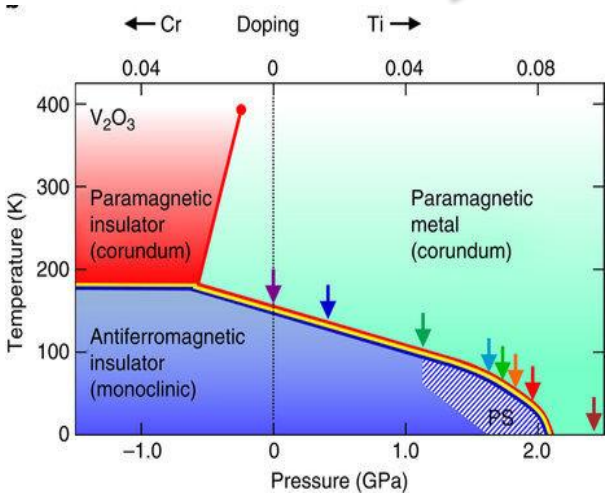
Single shot is working

Self consistent works sometimes and sometimes it does not converge a all.. New actions coming from new Wannier interface

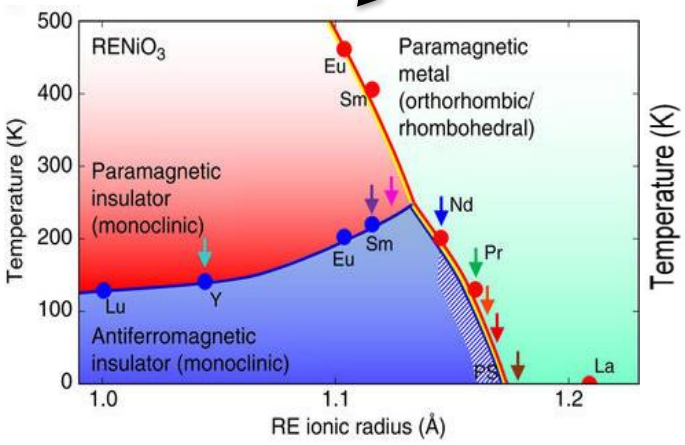
Strong correlations in materials

3d
4d
5d

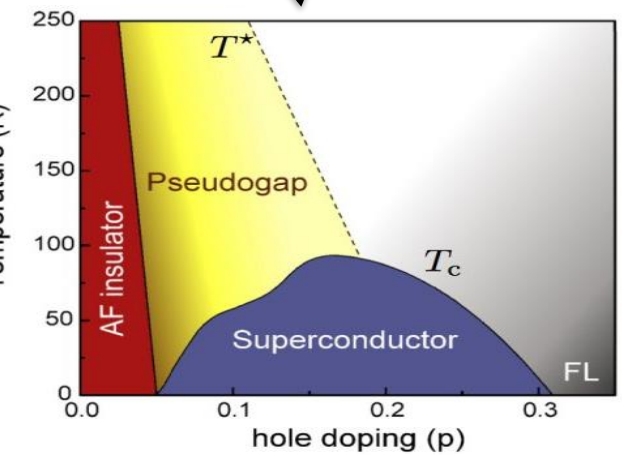
21	22	23	24	25	26	27	28	29	30
Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn
44.956	47.867	50.942	51.996	54.938	55.845	58.933	58.693	63.546	65.38
39	40	41	42	43	44	45	46	47	48
Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd
88.906	91.224	92.906	95.96	[97.91]	101.07	102.91	106.42	107.87	112.41
71	72	73	74	75	76	77	78	79	80
Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg
174.97	178.49	180.95	183.84	186.21	190.23	192.22	195.08	196.97	200.59



Vanadate:
Metal-insulator transition



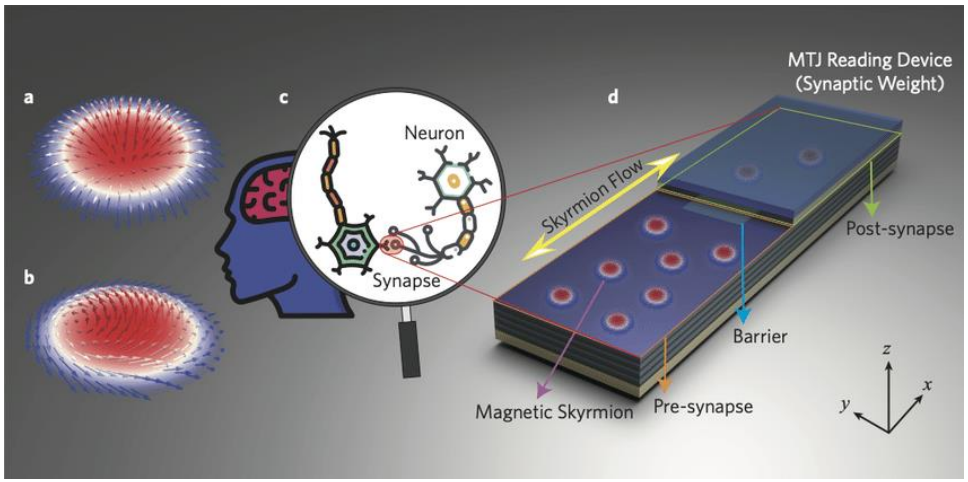
Nickelate:
Site-selective Mott transition
with electron-lattice coupling



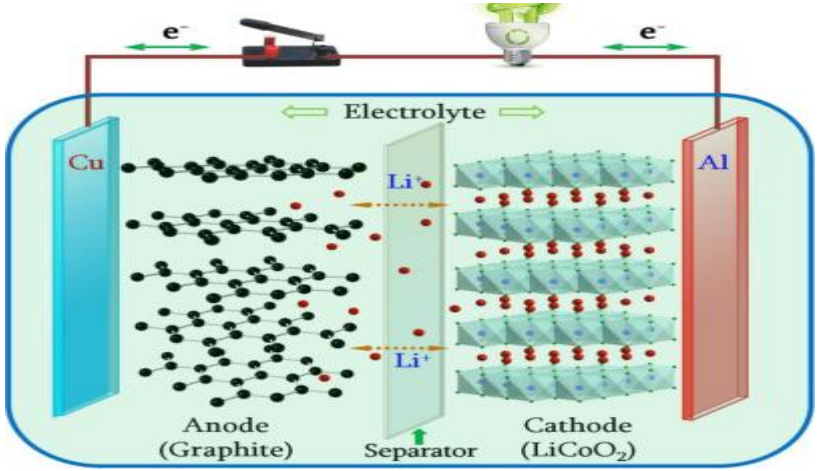
Cuprates:
High Tc superconductivity

Applications of strong correlated materials

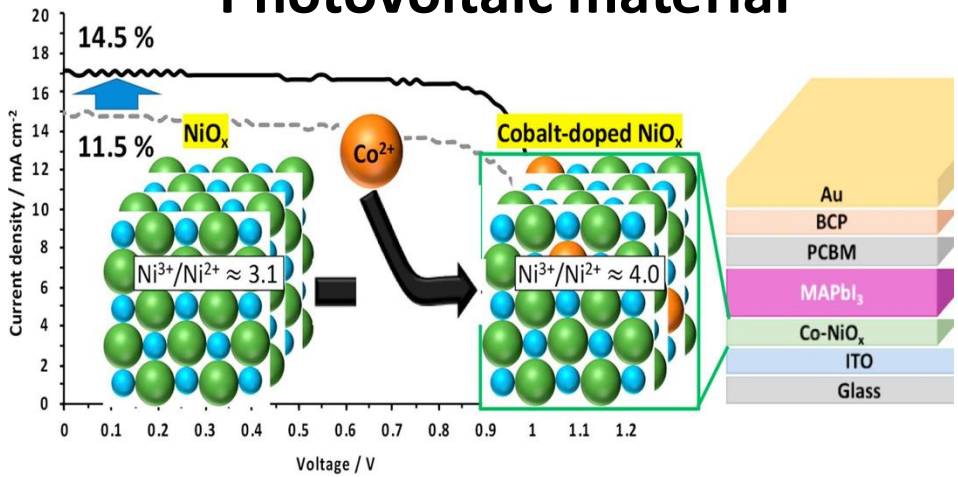
Magnetic skyrmion device



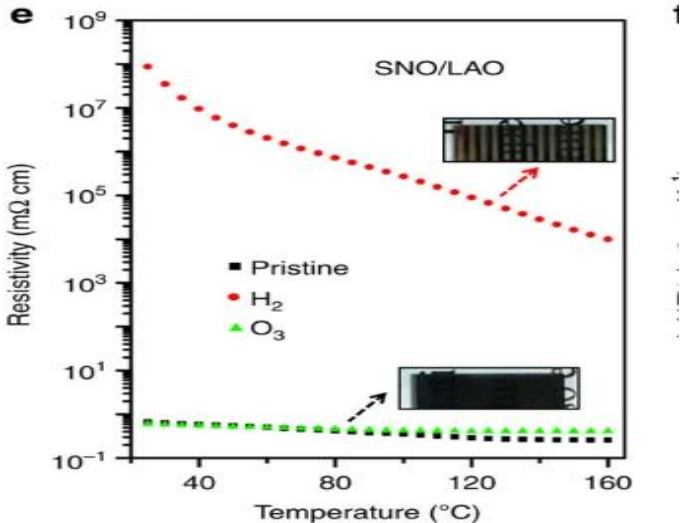
Batteries



Photovoltaic material



Resistivity switching devices



Theory of everything in solids

Microscopic Hamiltonian

$$\hat{H} = -\frac{\hbar^2}{2m}\sum_i \nabla_i^2 + \sum_i V_{ext}(\mathbf{r}_i) + \frac{1}{2}\sum_{i\neq j}\frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|}$$

Schrodinger Equation : $\hat{H}\,\Psi = i\hbar\frac{\partial\Psi}{\partial t}$ Observable=<**Ψ**|O|**Ψ**>

$\Psi = \Psi(\vec{x}_1, \vec{x}_2 \ldots \vec{x}_N, t)$ (Dimension : 3N+1 (N~10^{23}))

Some approximations are inevitable !

Macroscopic Materials

- ✓ Complexity : Macroscopic arrays of atoms
- ✓ Bulk, Nanostructure, Superlattice, Thin Film, …
- ✓ Strong Correlation : Coulomb E ~ Kinetic E (Non-perturbative)
- ✓ Emergent Behavior: Magnetism, Metal-insulator transition, …

First-principles approach to solids

Chemistry
1998

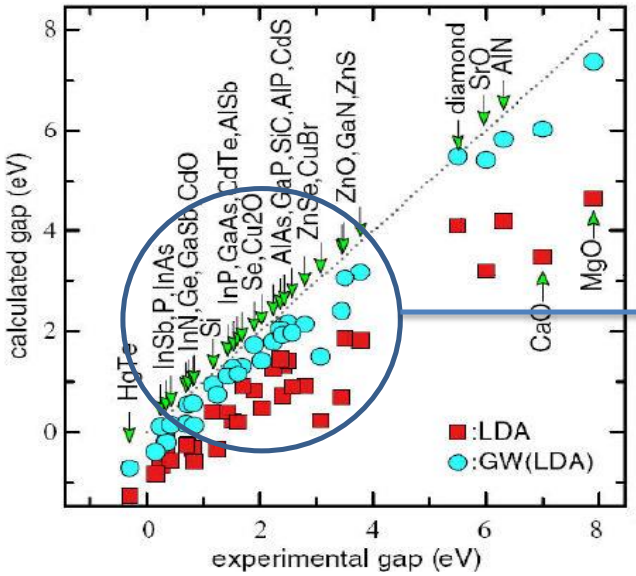
W. Kohn

Ground State Energy **E** = **E[n(r)]**

n(r)& Total electronic charge density

- Kohn-Sham Ansatz : Many body problem -> one-body problem (quasi-particles)

$$n(\mathbf{r}) = \sum_{i,\sigma} |\psi_i^\sigma(\mathbf{r})|^2$$

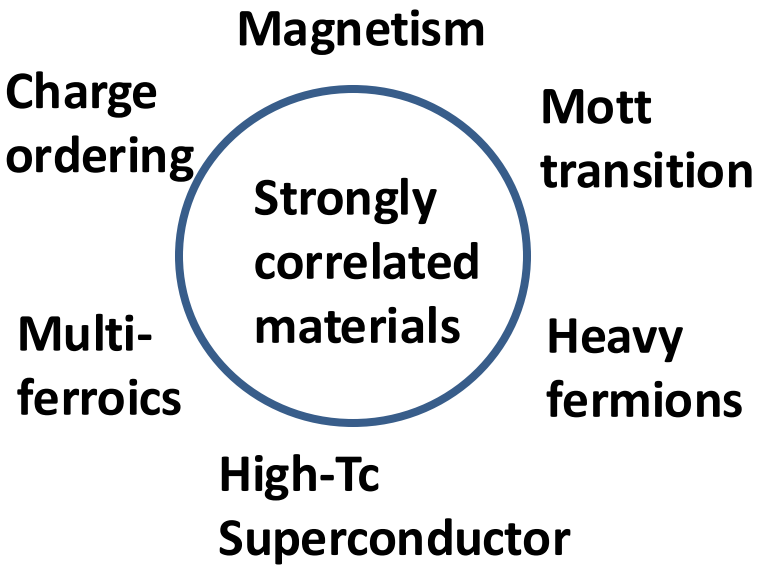


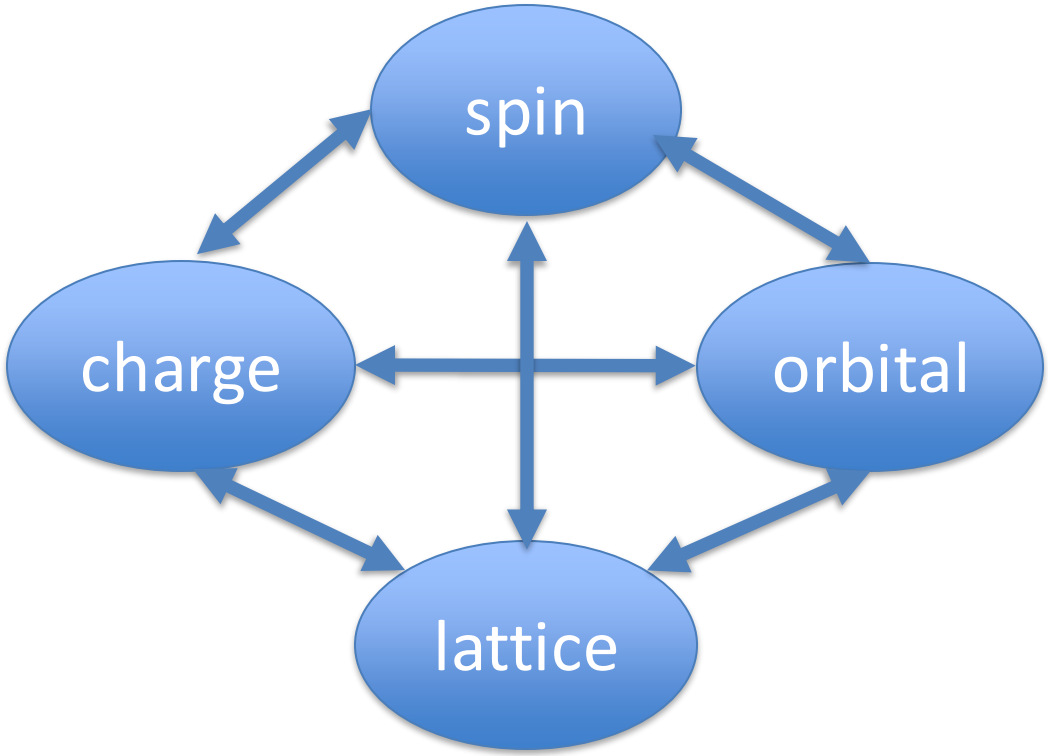
Practically Efficient :
e^N -> N^3 (N: number of atom)

Works reasonably in semiconductors, wide-gap band insulators

Ref) M. Schilfgaarde et al, PRL**&96&**26402 (2006)

Failure of DFT to Strongly correlated materials





. Strong correlations in materials lead to novel material properties due to spin, charge, orbital, and lattice coupling.

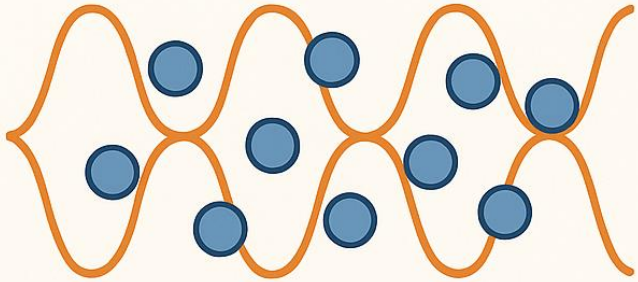
. Conventional density function theory (DFT) usually fails.

Dynamical Mean Field Theory (DMFT)

is a computational and conceptual framework developed to study the behavior of electrons in materials where their mutual interactions are so strong that the simple independent-particle picture taught in introductory quantum mechanics is no longer valid.

Independent-particle picture

Strongly interacting electrons



Independent-particle picture

Strongly interacting electrons

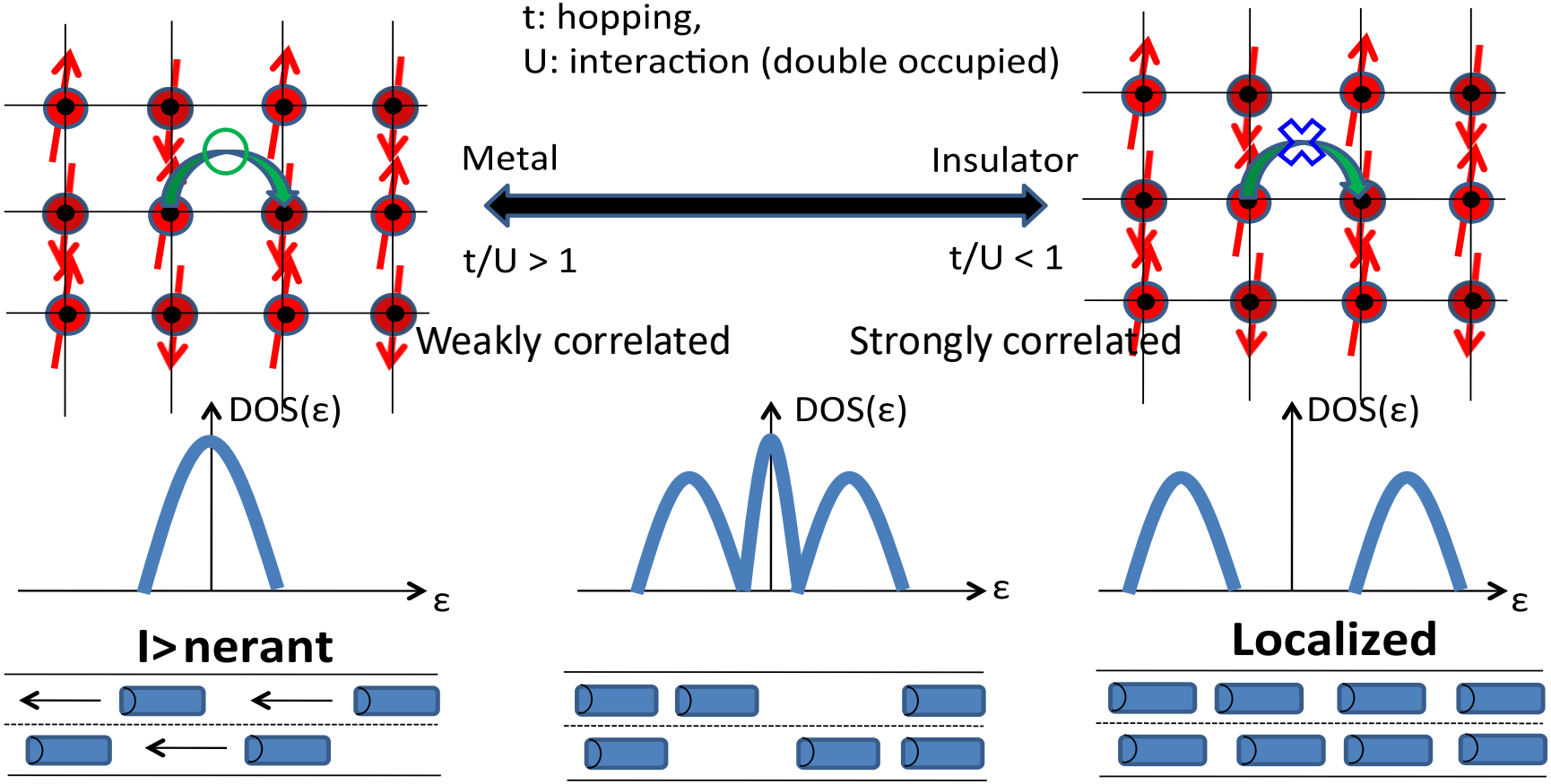
Anisimov et al. *Phys. Rev. B* **44**, 943–954 (1991).

Coulomb interaction among electrons is considerably greater than their kinetic energy, rendering the single-particle approximation inadequate

Effects of strong correlations

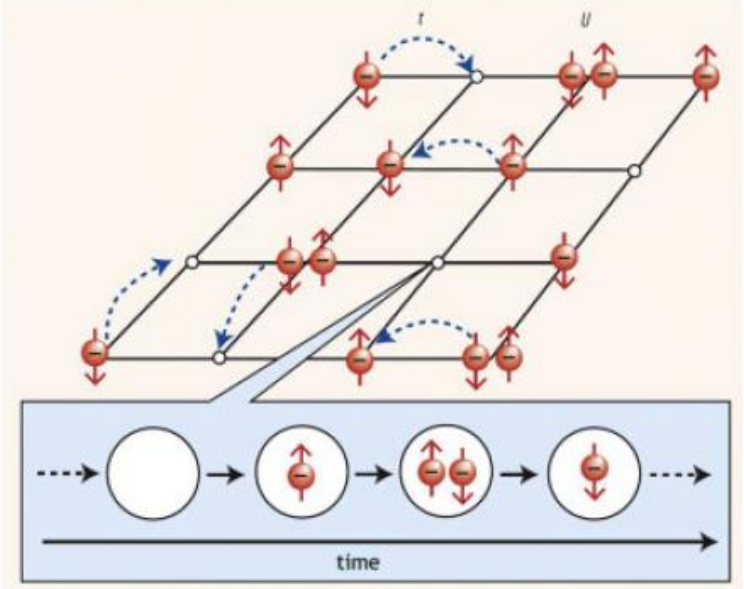
Hubbard Model: $H = -\sum_{\langle i,j\rangle,\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + U \sum_{i} c^{\dagger}_{i\uparrow} c_{i\uparrow} c^{\dagger}_{i\downarrow} c_{i\downarrow}$

t: hopping,
U: interaction (double occupied)



Mott insulator : partially filled electrons (Interaction driven)

c.f.) Band insulator : fully filled or empty electrons



In particular, for strong repulsion U double occupations are energetically unfavorable and are therefore suppressed.

In this situation the local correlation function $\langle n_{i\downarrow} n_{i\uparrow} \rangle$ *must not be factorized since* $\langle n_{i\downarrow} n_{i\uparrow} \rangle \neq \langle n_{i\downarrow} \rangle \langle n_{i\uparrow} \rangle$

The Hubbard model looks deceptively simple. However, the competition between the kinetic energy and the interaction leads to a complicated many-body problem.

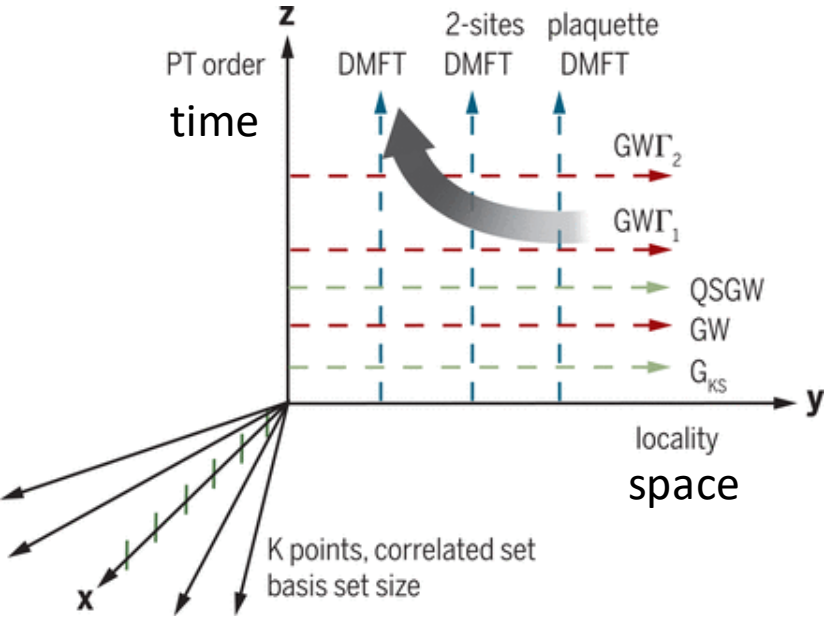
Assumed some parameters to be large (in fact, infinite), e.g., the length of the spins S, the spin degeneracy N, the number Z of nearest neighbors of a lattice site (the coordination number), or the spatial dimension d.

The limit of high spatial dimension d or coordination number Z enables a mean-field theory for Hubbard-type lattice fermions with kinetic energy and local interaction, yielding a diagrammatically controlled theory with a well-behaved free energy.

Vollhart, *Ann. Phys. (Berlin) 524, No. 1, 1–19 (2012)*

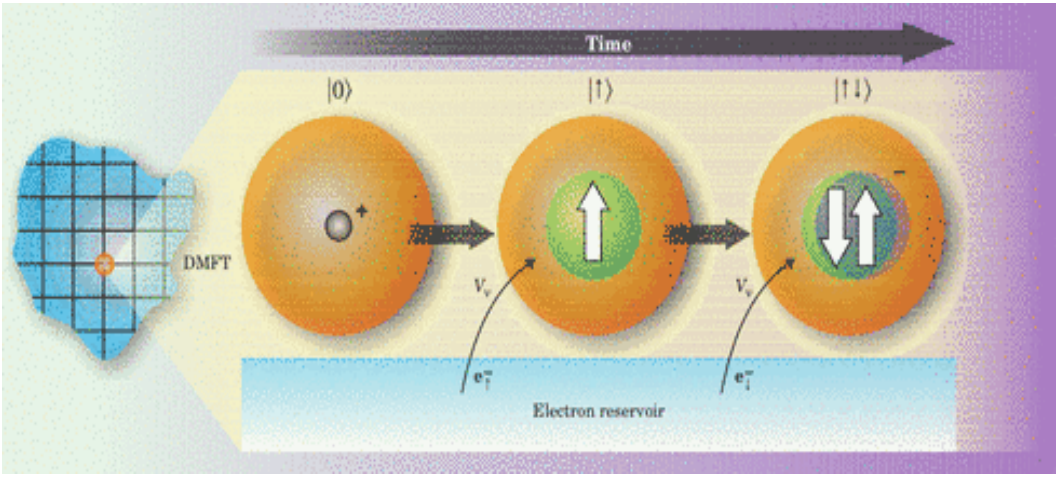
Beyond-DFT methods

Strong correlation problem



Ref) P. Kent and G. Kotliar, Science 361 348 (2018)

Dynamical mean field theory (DMFT)

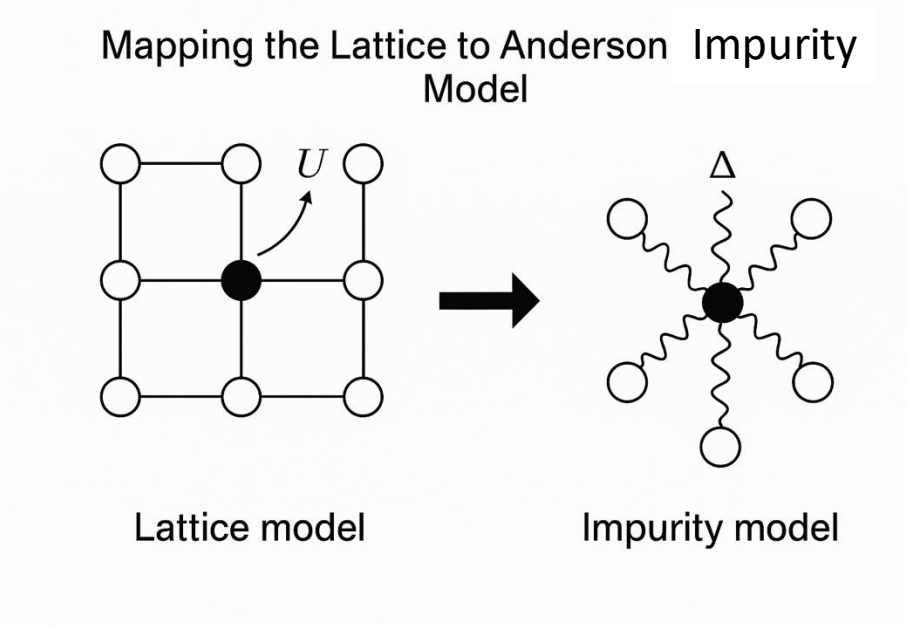


Ref) Georges et al, Rev. Mod. Phys. **68** 13 (1996)
Kotliar et al, Rev. Mod. Phys. **78** 865 (2006)

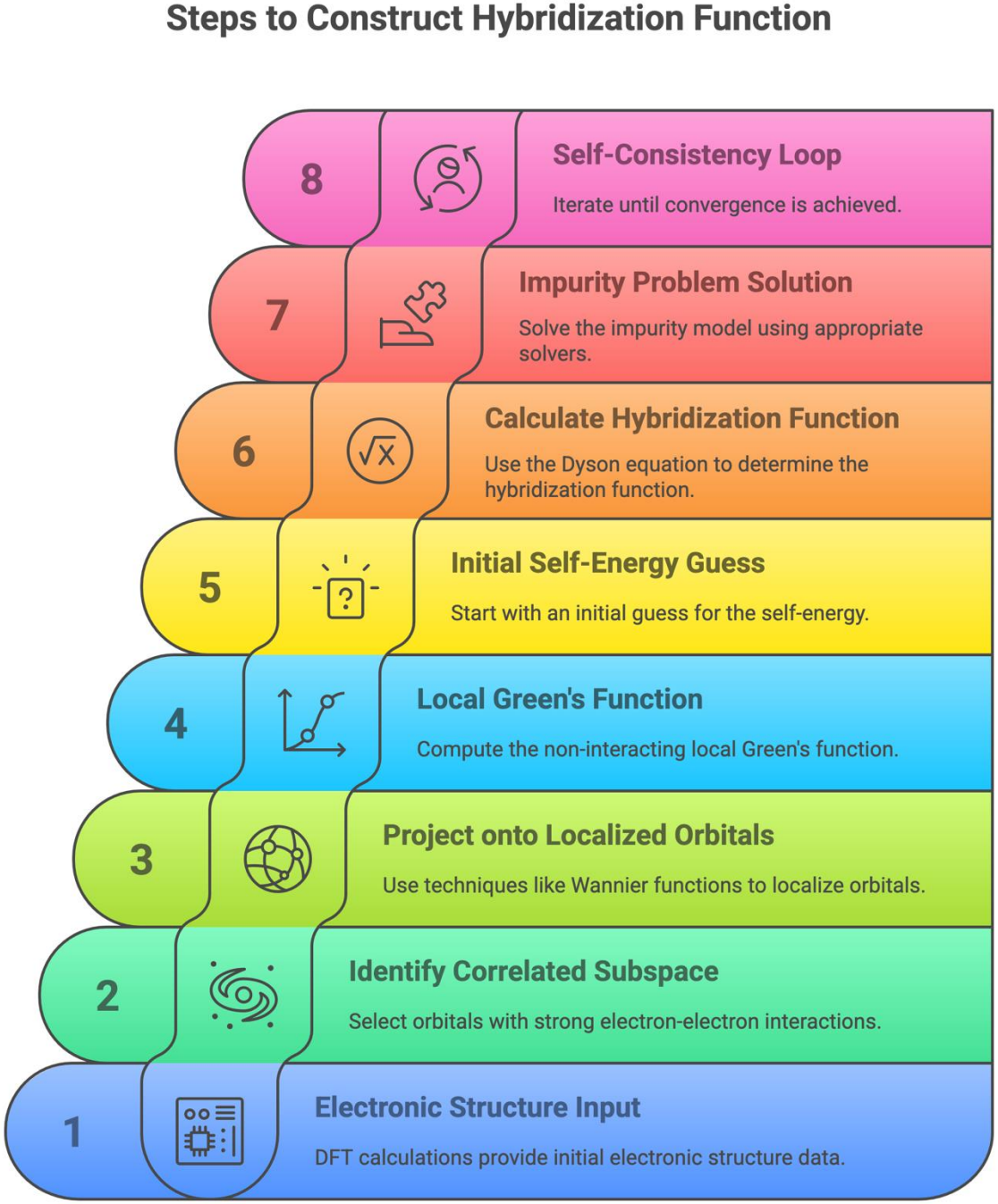
- DMFT variable: time-dependent but local Green's function: $G^{loc}(t-t')$ and self-energy $\Sigma^{loc}(t-t')$
- DMFT can capture the dynamical correlation beyond DFT.

Conceptual Framework of DMFT

- Reduction of the complex lattice problem to a single-site impurity problem.
- Focusing on a single lattice site (“impurity”) and its interaction wit an effective medium (“bath”) that represents the rest of the lattice.
- Treatment of local interactions exactly while approximating the rest of the lattice as a dynamic mean field.
- Self-consistent determination of the impurity’s environment.



The DMFT Self-Consistency Loop



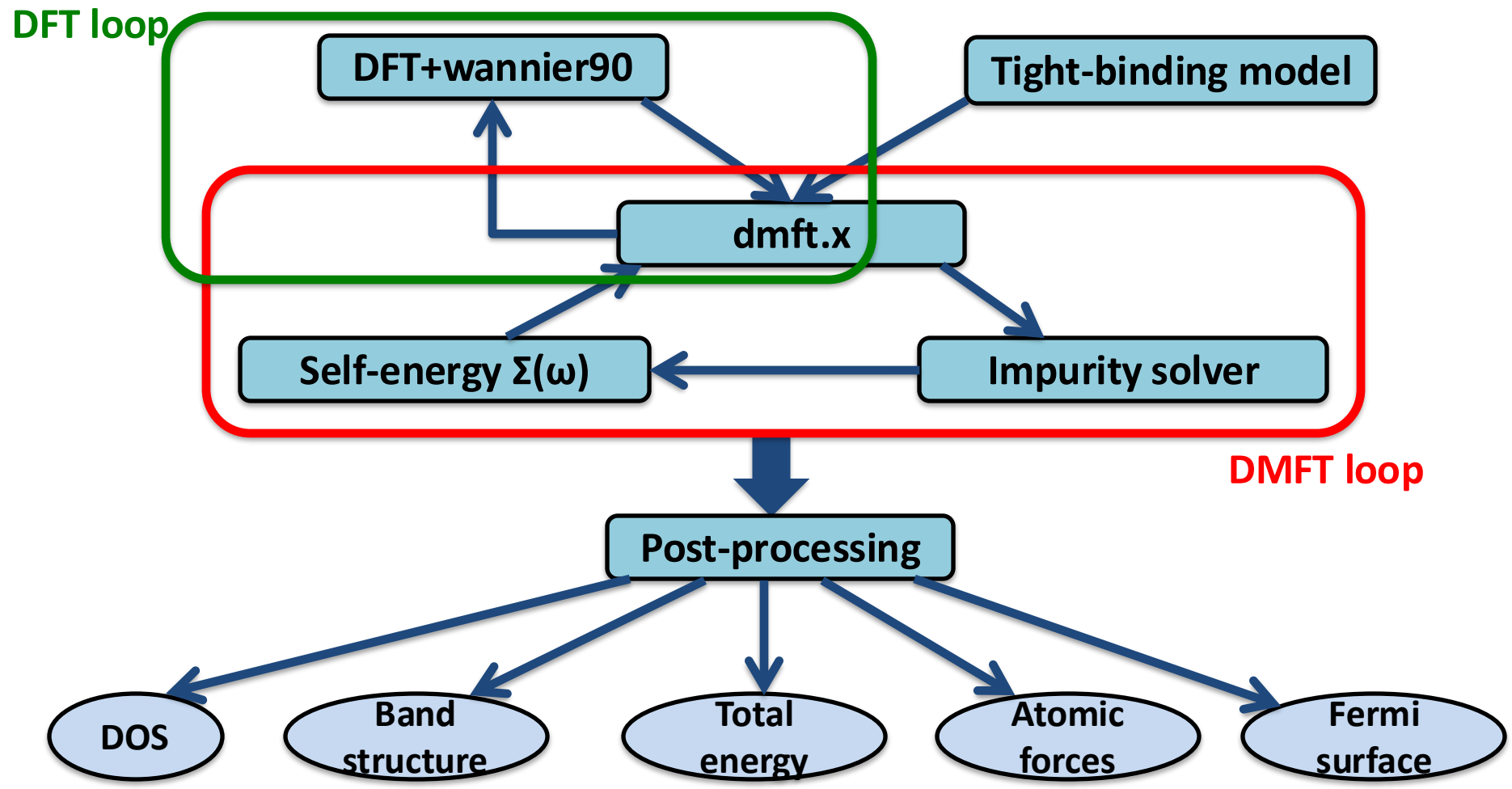
Big picture

- You begin with a standard LDA calculation using any DFT code: $\varepsilon_{k,n}, \varphi_{k,n}, H_{LDA}$ (non interactive Hamiltonian)
- Identify correlated subspace
- Construct the low energy Hamiltonian , H_{LDA}^{proj}
- Add an **on-site Hubbard-like interaction term**

$$\mathrm{H} = H_{LDA}^{proj} + H_{int} - H_{DC}$$

- Map the correlated lattice problem onto an impurity model and solve the impurity problem using a DMFT solver (QMC, ED, NRG, etc.)
- **Construct Lattice Green's Function, Compute Local Green's Function, Update the Impurity Model, Solve again.**
- New density->DFT->DMFT-> until convergence!

The overall structure of DFT+DMFT



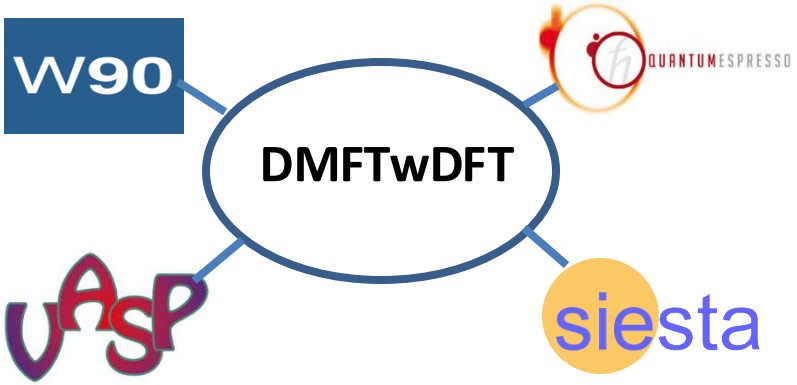
Our open-source DMFT software (DMFTwDFT) interfaced to various DFT and Wannier90 codes

Real life Software Development

Features of DMFTwDFT code

DMFTwDFT website: github.com/DMFTwDFT-project/DMFTwDFT

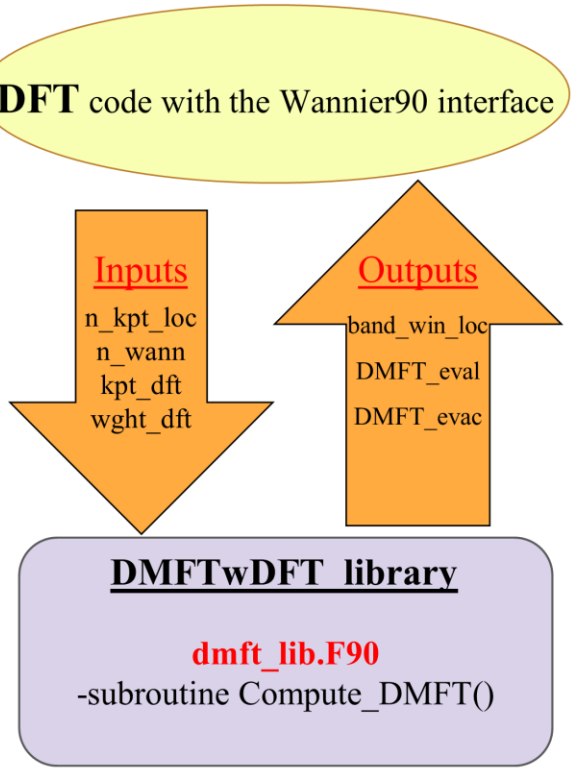
- **Interfaced to various electronic structure packages**



- **Parallelization using MPI**
  : k-point mesh parallelization

Ref) Vijay Singh *et al*, Computer Physics Communications, 261,107778 (2021)

- **Library mode**



. Efficient charge density update from the DMFT density matrix

Installation: *git clone https://github.com/DMFTwDFT-project/DMFTwDFT.git*

Dependencies: *pip install matplotlib numpy scipy weave mpi4py requests*
Note: Python 2.x due to CTQMC's reliance on the weave library.

Building: *python setup.py {gfortran, intel}*

After compilation:

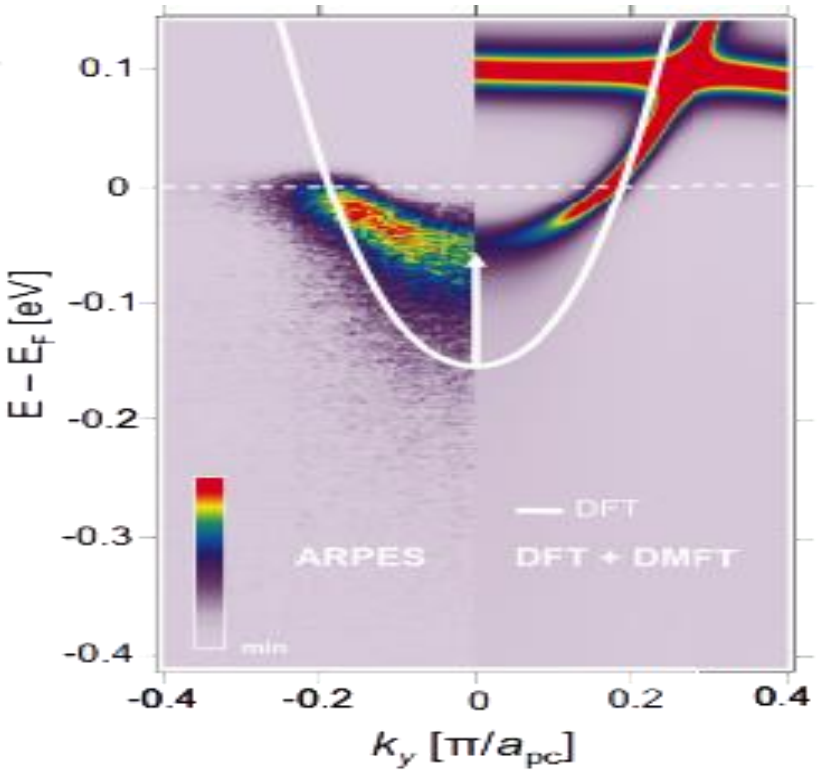
dmft.x - This module is for achieving DMFT self-consistency. It performs the k-point sum and and computes the
local Green's function (G_loc.out) and hybridization function (Delta.inp).
dmft_dos.x - Performs DOS calculation
dmft_ksum_band - Performs band structure calculation
dmft_ksum_partial_band - Performs projected band structure calculation
fort_kpt_tools.so - Fortran based k-points calculation module

Note: *CTQMC impurity solver and Max-entropy routines developed by Professor Haule at Rutgers University available with the EDMFT package*

DMFTwDFT requires wanner90.x and w90chk2chk.x to be available in the bin directory

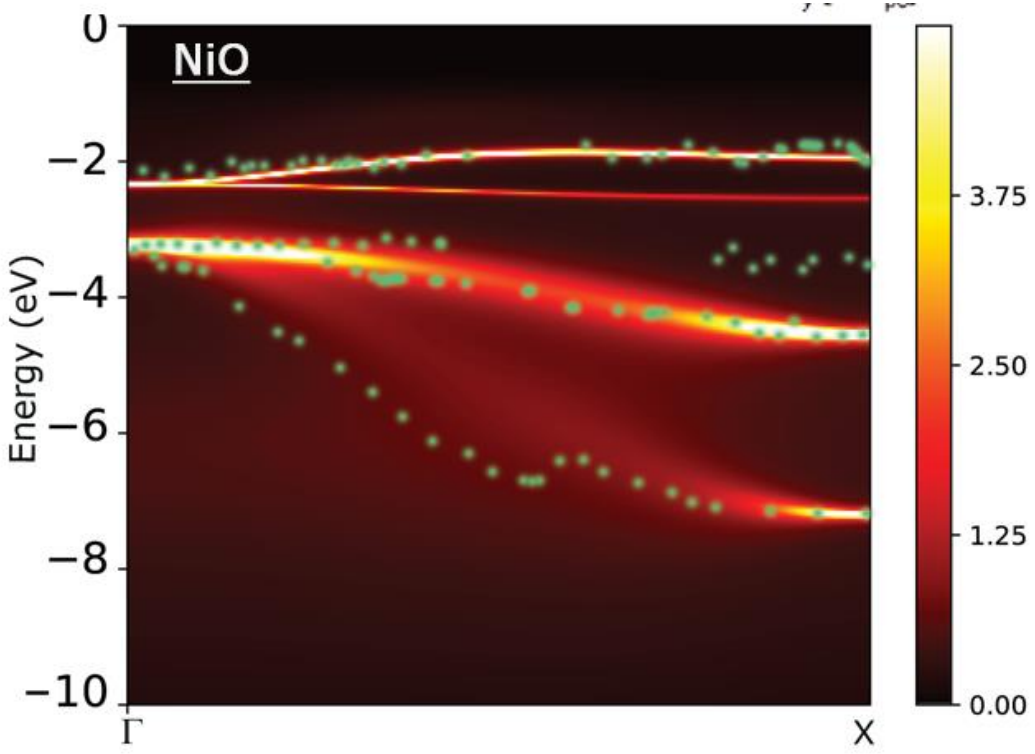
Results of DMFTwDFT – Compared to ARPES

$LaNiO_3$



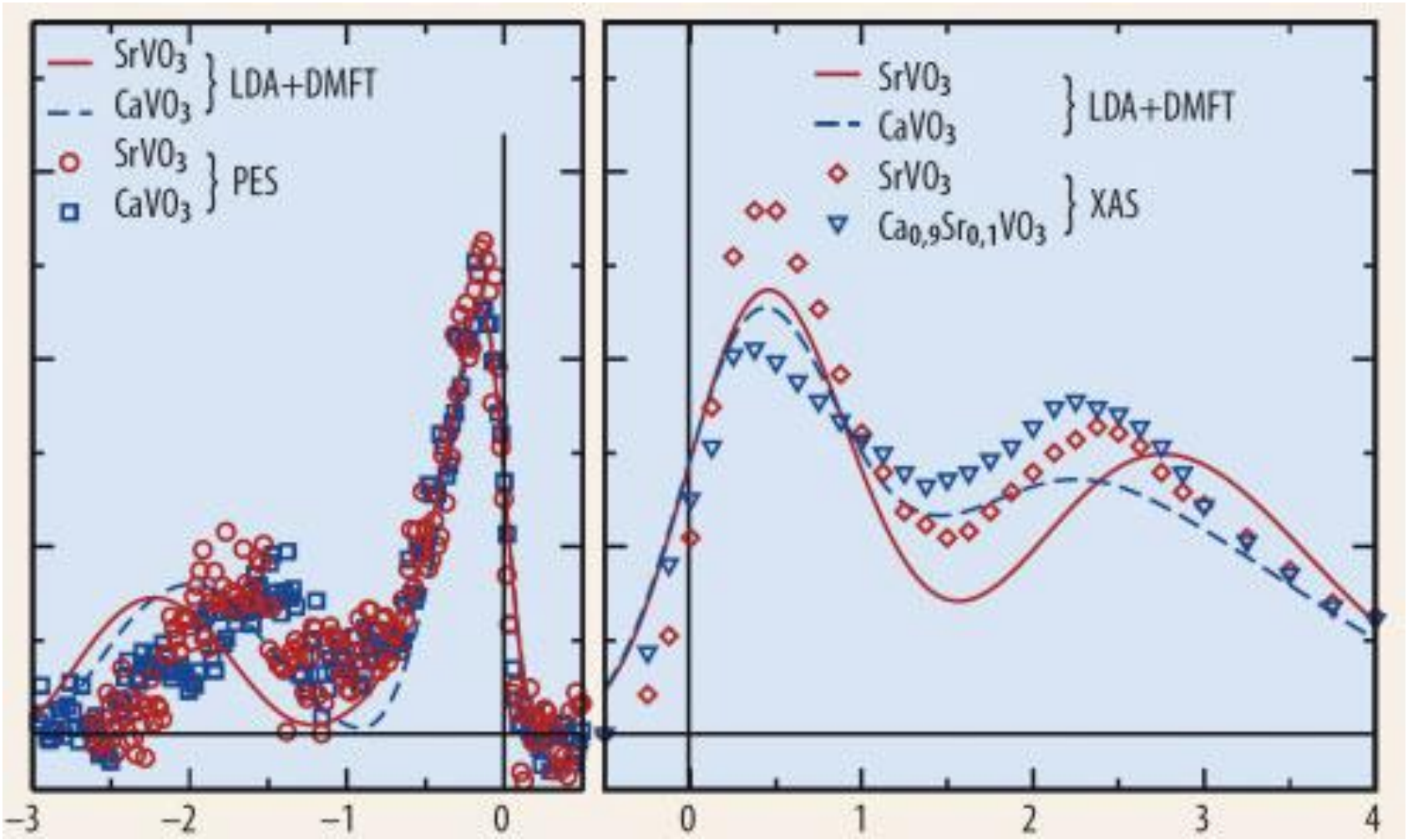
ARPES Ref) Nowadnick et al. PRB 92, 245109, 2015

NiO



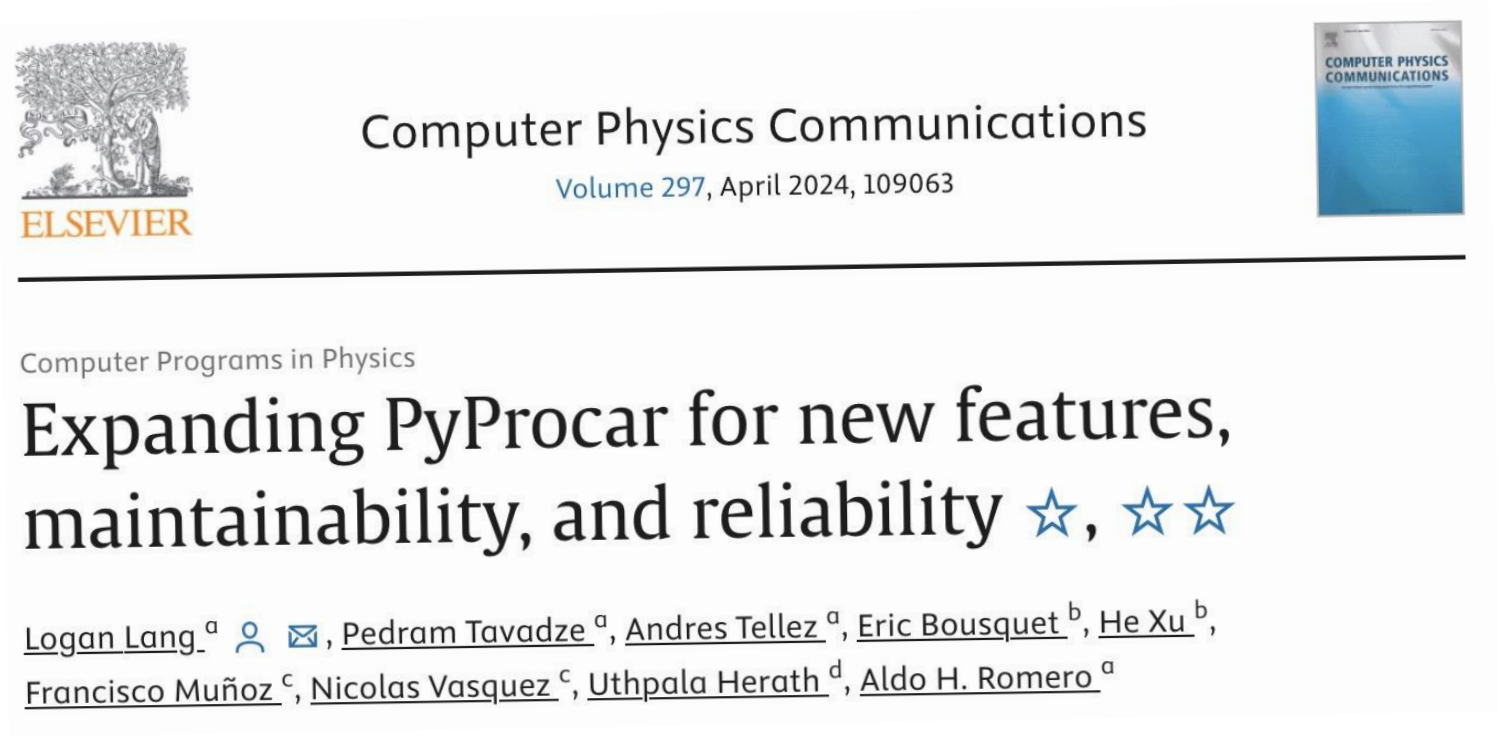
ARPES Ref) Shen et al. PRB 44 3604, 1991

DMFT band structure is consistent with ARPES measurement



I. A. Nekrasov et al., Phys. Rev. B 72, 155106 (2005).

Advertisement: PyProcar



Computer Physics Communications
Volume 297, April 2024, 109063

Computer Programs in Physics

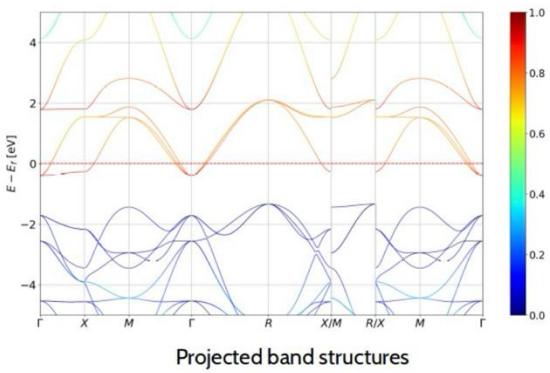
Expanding PyProcar for new features, maintainability, and reliability ☆, ☆☆

Logan Lang [a], Pedram Tavadze [a], Andres Tellez [a], Eric Bousquet [b], He Xu [b], Francisco Muñoz [c], Nicolas Vasquez [c], Uthpala Herath [d], Aldo H. Romero [a]

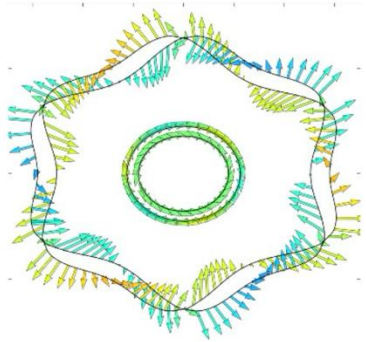
https://romerogroup.github.io/pyprocar/

https://github.com/romerogroup/pyprocar

Aside note: PyProcar

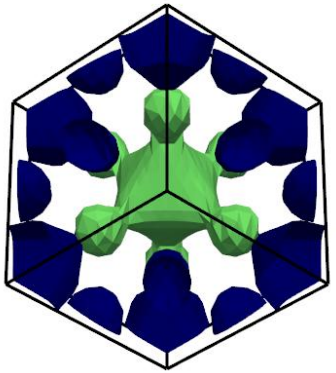


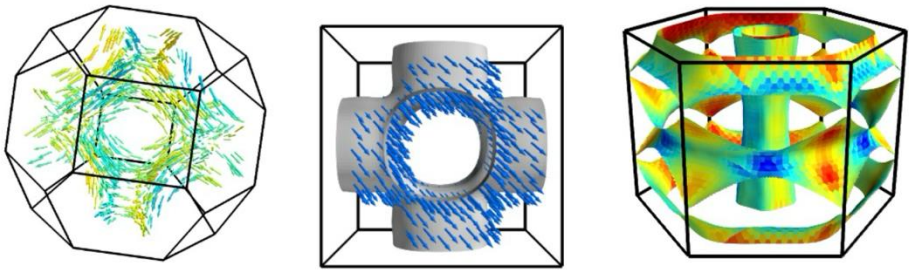
Projected band structures



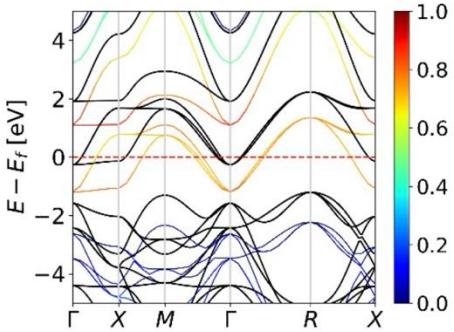
Spin texture plots

Energy Value : 17.2398 eV

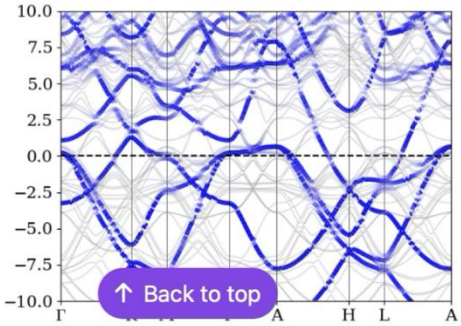




Projected Fermi surfaces



Band structure comparison



Band unfolding

Fermi Cross section Slicer – Used to find extremal orbits for dHvA frequency

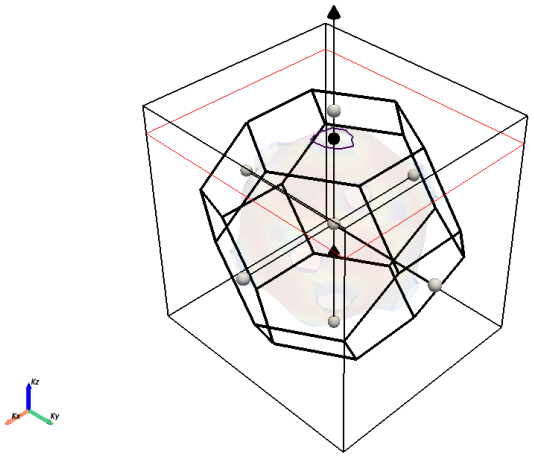
$$F = \frac{c\hbar A}{2\pi e}!(cgs)$$
$$e = 4.768e^{-10}!statcoulombs$$
$$c = 3.0e^{10}!cm/s$$
$$\hbar = 1.0546e^{-27}!erg * s$$

```
fermiHandler = pyprocar.FermiHandler(
                                    code="vasp",
                                    dirname=data_dir,
                                    apply_symmetry=True)


fermiHandler.plot_fermi_cross_section_box_widget(
                                bands=[5],
                                slice_normal=(0,0,1),
                                slice_origin=(0,0,0),
                                surface_opacity=0.05,
                                add_scalar_bar=False,
                                cross_section_slice_linewidth=False,
                                cross_section_slice_show_area=True,
                                mode="parametric",
                                # save_2d=f'{figure_dir}{os.sep}belly_001_slice.png',
                                show=True)
```

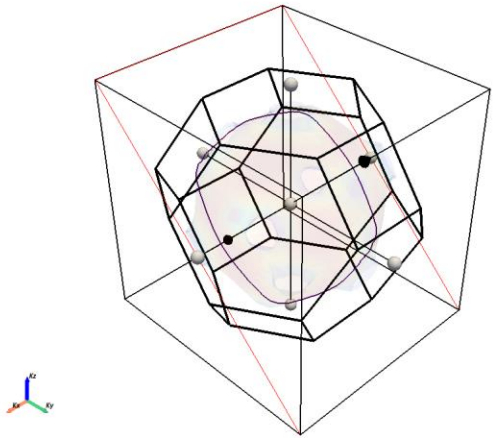
neck_001

Cross sectional area : 0.1597 Ang^-2



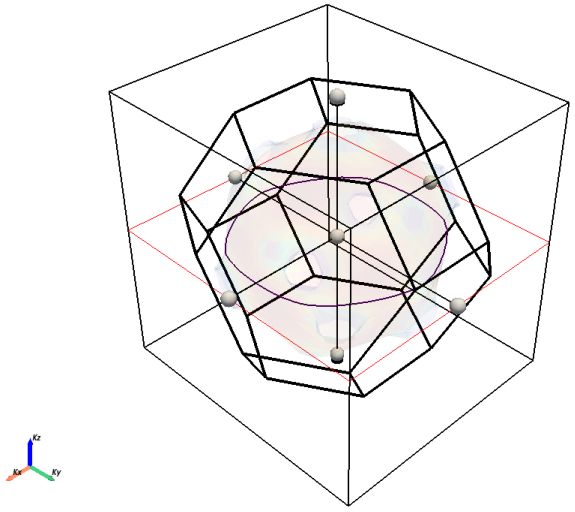
belly_011

Cross sectional area : 4.3956 Ang^-2



belly_001

Cross sectional area : 4.1586 Ang^-2



Running one-shot DMFT calculations

- Within siesta input:

  Siesta2Wannier90.WriteMmn .true.

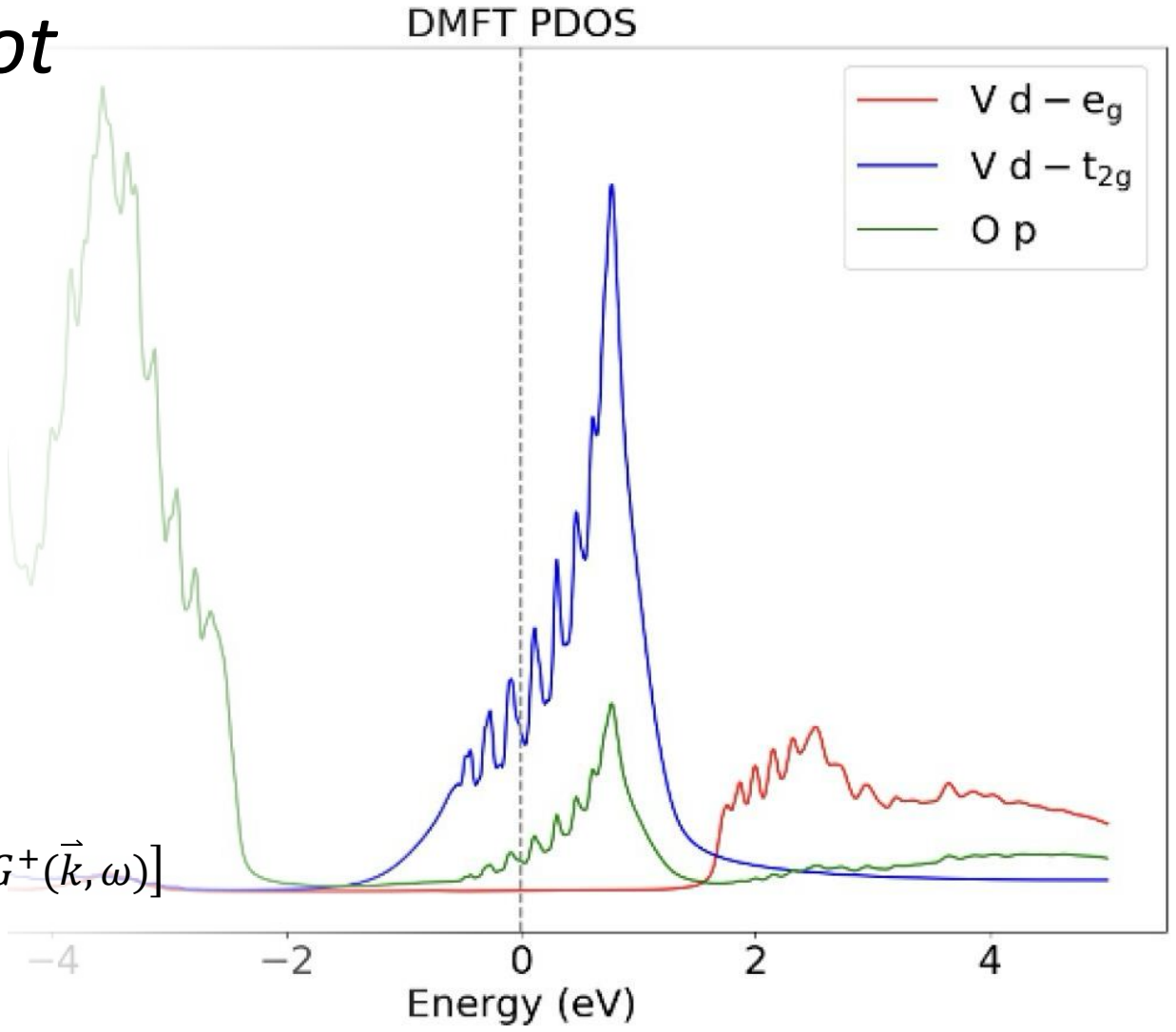
  Siesta2Wannier90.WriteAmn .true.

  Siesta2Wannier90.WriteEig .true.

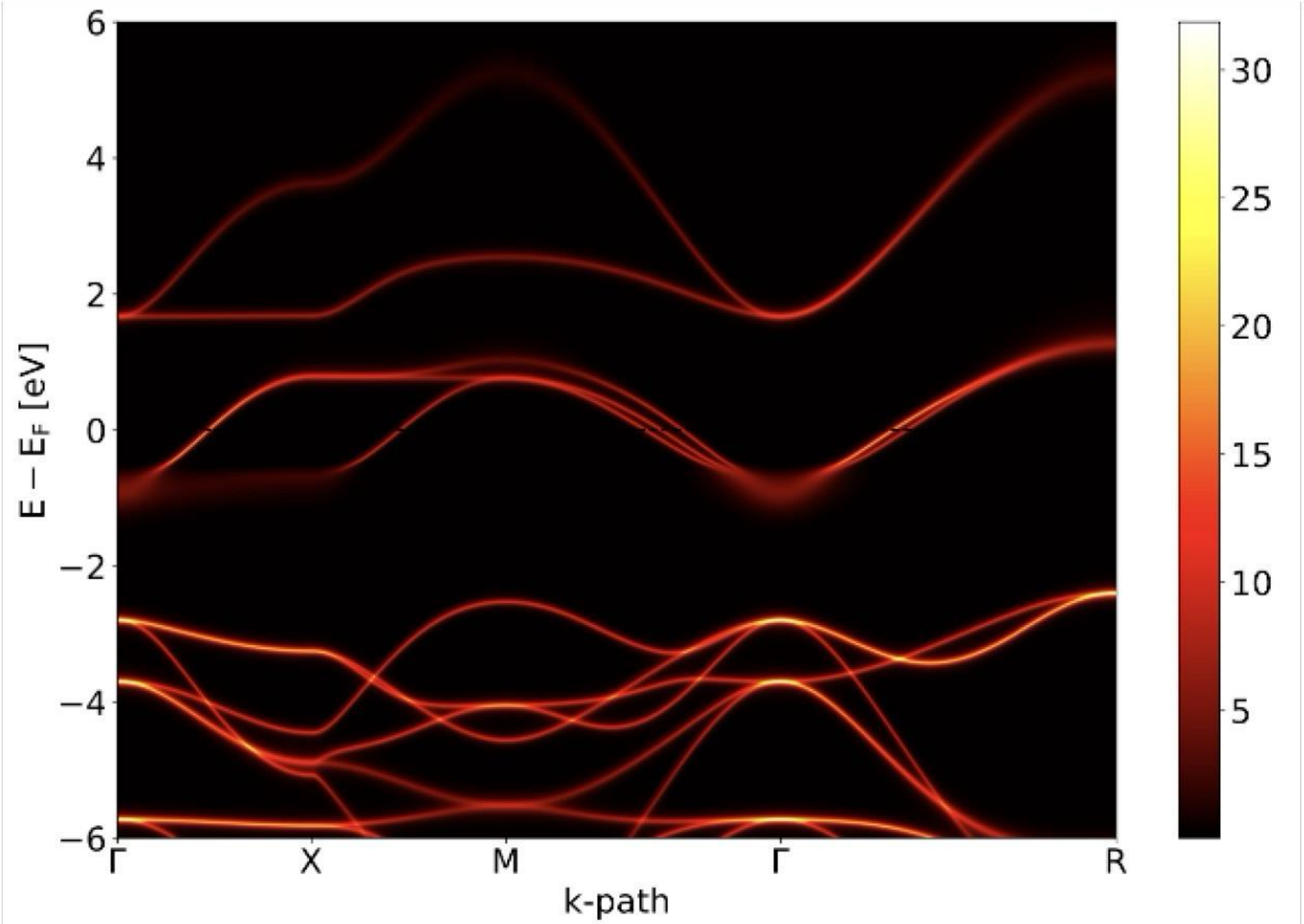
  Siesta2Wannier90.WriteUnk .false.

  Siesta2Wannier90.NumberOfBands 28

- $A(\vec{k},\omega) = \frac{i}{2\pi} Tr\left[G(\vec{k},\omega) - G^{+}(\vec{k},\omega)\right]$



2. The DMFT PDOS of $SrVO_3$ calculated with siesta+DMFT


6
4
2
0
−2
−4
−6
E − E_F [eV]
Γ
X
M
Γ
R
k-path
30
25
20
15
10
5

Full charge self-consistent DFT+DMFT calculations

- *DMFTwDFT build process generates the libdmft.a library which can be used to interface with DFT codes to enable full charge self-consistent DFT+DMFT calculations.*

- *The siesta+DMFT full charge self-consistent DFT+DMFT implementation is still experimental so proceed with caution.*

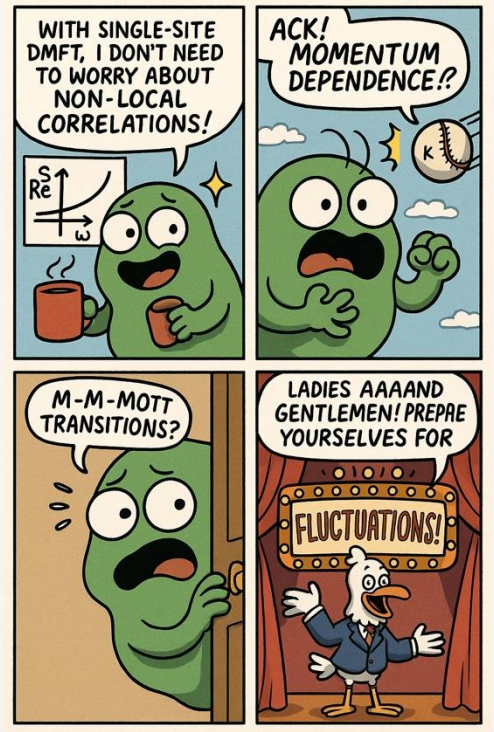
- *Obtain the modified siesta code from the repository https://gitlab.com/uthpalaherath/siesta.git under the dmft branch (thanks to J. Junquera)*

Limitations of the single site DMFT

Neglect of Non-Local Correlations: Single-site DMFT assumes a local self-energy, ignoring spatial correlations between different lattice sites. Exact only in the limit of infinite dimensions or coordination number.

Inability to Capture Momentum-Dependent Phenomena: Phenomena like the pseudogap in underdoped cuprates or d-wave superconductivity require momentum-dependent self-energies, which single-site DMFT cannot provide.

Oversimplification of Phase Transitions: The method may inaccurately predict the nature of phase transitions, such as the Mott metal-insulator transition, due to its neglect of short-range spatial fluctuations.



Cluster Extensions: Enhancing DMFT

To overcome these limitations, cluster extensions of DMFT have been developed. These methods consider a cluster of sites rather than a single site, allowing for the inclusion of short-range spatial correlations.

1. Cellular DMFT (CDMFT)

Divides the lattice into real-space clusters and treats each cluster as an impurity embedded in a self-consistent bath.

Advantages: Captures short-range spatial correlations within the cluster. Suitable for studying systems with strong local interactions.

Considerations: Requires periodization techniques to reconstruct lattice quantities from cluster data. Computationally more demanding than single-site DMFT.

2. Dynamical Cluster Approximation (DCA)

Partitions the Brillouin zone into momentum-space clusters, treating each as an impurity problem.

Advantages: Naturally incorporates momentum-dependent self-energies. Avoids the need for periodization, simplifying the reconstruction of lattice quantities.

Considerations: Assumes periodic boundary conditions within the cluster, which may introduce finite-size effects. Computational cost increases with cluster size.

From Code to Correlations: Final Thoughts

- DMFT implementation in SIESTA unlocks realistic simulations of correlated materials
- Code is scalable, open-source, and ready for community input
- Exciting possibilities for oxides, heterostructures, and quantum materials
- Looking forward to collaborations, benchmarking, and new applications

- *Questions?*

"The best way to predict the future is to invent it."

— Alan Kay