Introduction to First-principles and DMFT

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Contents

Many-electron problem. Why is it so hard?

Hamiltonian

$$\hat{H} = -\frac{\hbar^2}{2m_e}\sum_i \nabla_i^2 - \sum_{i,I}\frac{Z_I e^2}{|\mathbf{r}_i - \mathbf{R}_I|} + \frac{1}{2}\sum_{i\neq j}\frac{e^2}{|\mathbf{r}_i - \mathbf{r}_j|} - \sum_I \frac{\hbar^2}{2M_I}\nabla_I^2 + \frac{1}{2}\sum_{I\neq J}\frac{Z_I Z_J e^2}{|\mathbf{R}_I - \mathbf{R}_J|},$$

The curse of dimensionality

The number of single particle basis function: M
The number of electrons: N
The number of slater's determinant:

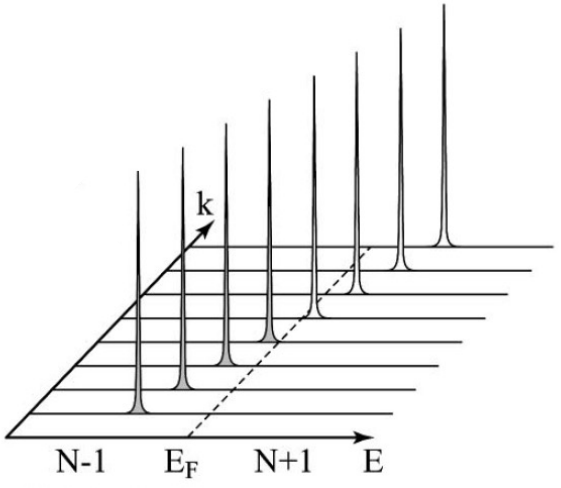
$$\begin{pmatrix} M \\ N \end{pmatrix} = \frac{M!}{N!(M-N)!} \approx e^{CN}, C > 0$$

Ex) Two carbon atom(N=12, M=36): 10^9 determinants

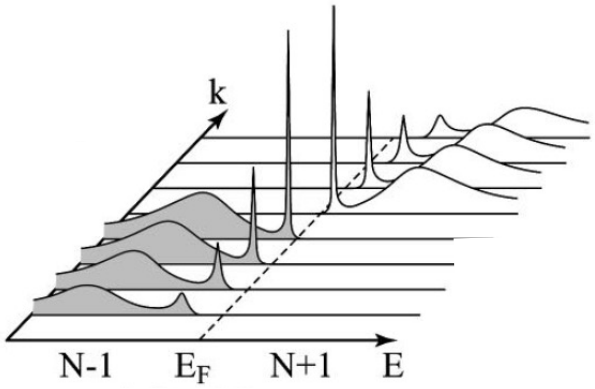
[1] R. M. Martin, L. Reining, and D. M. Ceperley, Interacting Electrons (Cambridge University Press, 2016).

ARPES measurement of simple metals

Theory (noninteracting)



Measurement



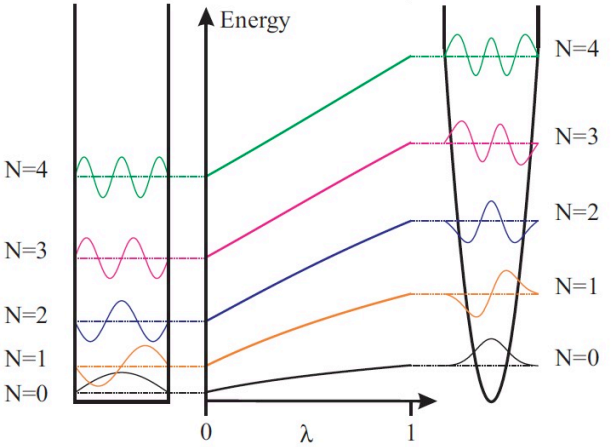
- Particle like peak with renormalized mass and finite lifetime
- One-to-one correspondence between the states near Fermi level

[1] A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys. 75, 473 (2003)

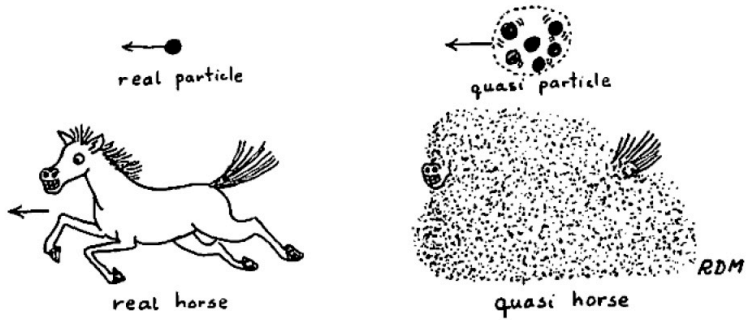
Fermi liquid theory

Adiabatic Continuity

$$-\frac{1}{2}\frac{d^2\psi}{dx^2} + V(x)\psi = E\psi \qquad V(x) = \begin{cases} \frac{1}{2}\lambda x^2 & |x| < \pi, \\ \infty & |x| \geq \pi. \end{cases}$$



Quasiparticles

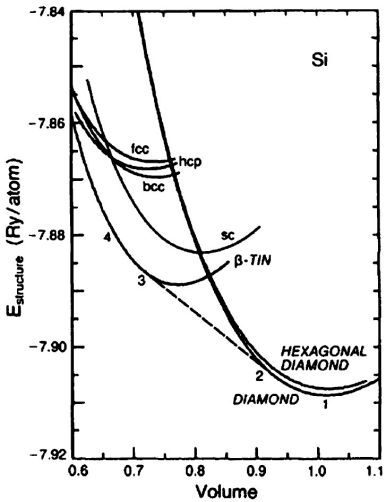


- Adiabatic continuity (conservation law unchanged by the interaction)
- Electron is independent for the requirement of the exclusion principles
- Working horses: DFT and GW

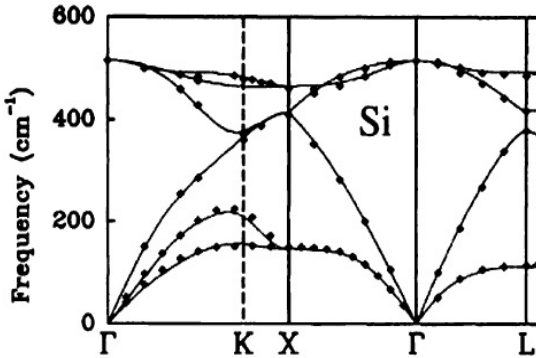
[1] L. D. Landau, JETP 3, 920 (1956) [2] L. D. Landau, JETP 5, 101 (1957) [3] L. D. Landau, JETP 8, 70 (1959)
[4] A. Damascelli, Z. Hussain, and Z.-X. Shen, Rev. Mod. Phys. 75, 473 (2003). [5] A. J. Schofield, Contemporary Physics 40, 95 (1999).

DFT for weakly correlated materials

Crystal structure



Phonon



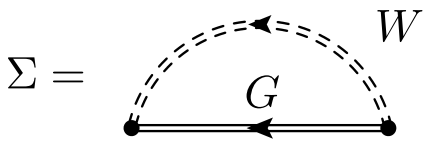
- Total energy calculation by solving auxiliary non-interacting system

$$\left\{ \frac{p^2}{2m} + V_{KS}(\mathbf{r}) \right\} \psi_i(\mathbf{r}) = \varepsilon_i \psi_i(\mathbf{r}), \;\; \rho(\mathbf{r}) = \sum_i^{occ} |\psi_i(\mathbf{r})|^2$$

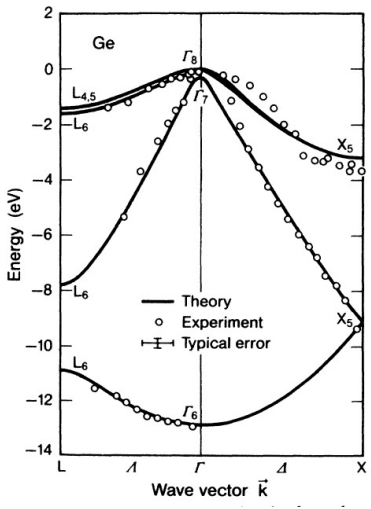
- For the ground state properties of weakly correlated materials
- Kohn-Sham energy is not a quasiparticle energy.

[1] M.T. Yin and M.L. Cohen, Phys. Rev. B 26, 5568 (1982).
[2] P. Giannozzi, S. Degironcoli, P. Pavone and S. Baroni, Phys. Rev. B 43, 7231 (1991).

GW for weakly correlated materials



Spectral function



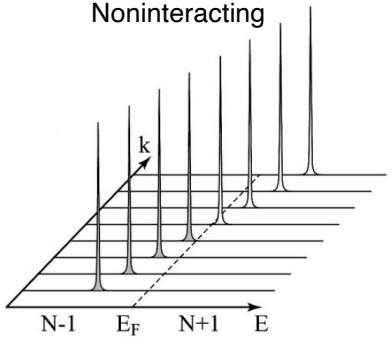
- The first order expansion of self-energy in W
- For the excited state properties of weakly correlated materials

[1] M. S. Hybertsen and S. G. Louie, Phys. Rev. B 34, 5390 (1986).
[2] M. Rohlfing and S.G. Louie, Phys. Rev. B 62, 4927 (2000).

Electron correlation

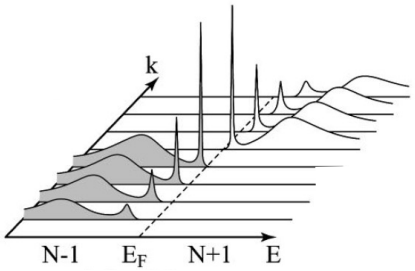
Practically, what is not captured within a mean-field theory (e.g. HF, DFT): $\left|\Sigma - \Sigma_{MF}\right|$

Noninteracting



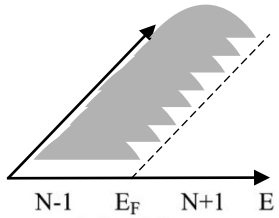
- Pauli-exclusion principle

Weakly correlated



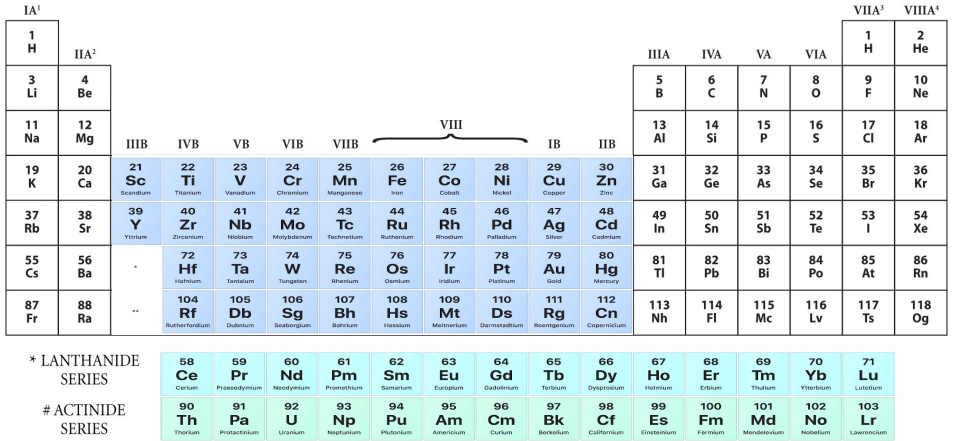
- a mean-field approximation (HF, DFT) or a low-order perturbation expansion around it suffices
- Adiabatic continuity
- quasiparticle

Strongly correlated



- Perturbative approach to a mean field fails
- In the extreme case, no continuity
- Sensitive to small changes in their control parameters, resulting in large responses.
- Potential applications such as oxide electronics, high-temperature superconductors, and spintronic devices

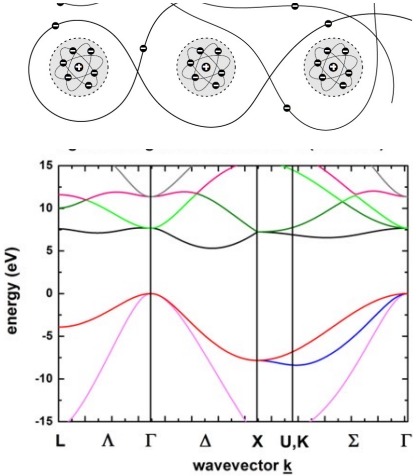
Electron correlation



- Electronic correlation becomes important typically in materials with open d- and f-subshell elements

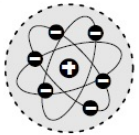
Beyond one-particle picture

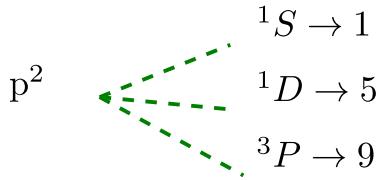
Itinerant electrons: bands



- Nondegenerate ground states
- Single slater determinants

Localized electrons: multiplets

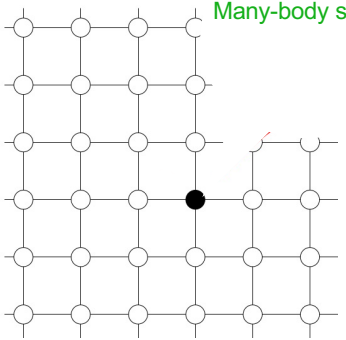




- Degenerate ground states
- Linear combination of many Slater determinants

- Electrons In open d and f shell systems are neither localized nor itinerant
- Beyond one-particle picture to retain multiplet structure

Dynamical Mean Field Theory



- Atom-like: multiplets at each sites
- Band-like: electron hopping between sites

- Mapping quantum many electron problem to quantum impurity problem in an effective field

$$S_{eff} = -\int_0^\beta d\tau d\tau'\, d^+_{o\sigma}(\tau)\, \mathcal{G}_0^{-1}(\tau,\tau')\, d_{o\sigma}(\tau') + \int_0^\beta d\tau\, U n_{o\uparrow}(\tau) n_{o\downarrow}(\tau)$$

Electron hopping
Band-like

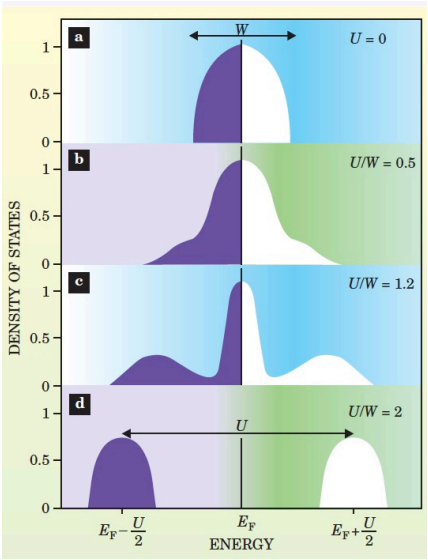
Multiplets
Atom-like

- Local approximation (no k dependence), but non-local extension are possible

[1] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Rev. Mod. Phys. 68, 13 (1996)

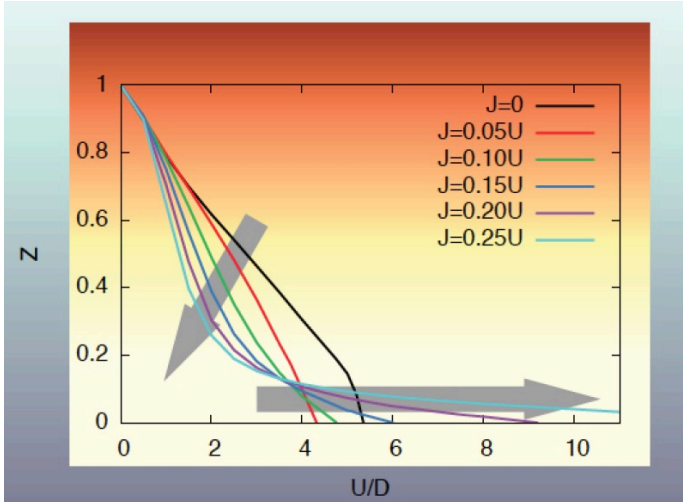
DMFT for the Mott and Hund physics

Mott insulator



A reference picture to understand cuprate physics

Hund metal

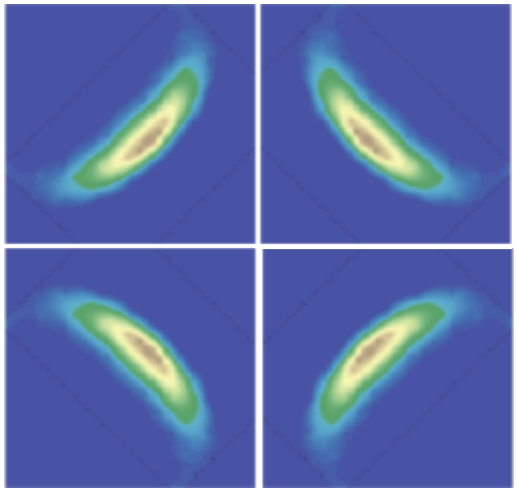


A reference picture to understand
Fe-based superconductors and ruthenates

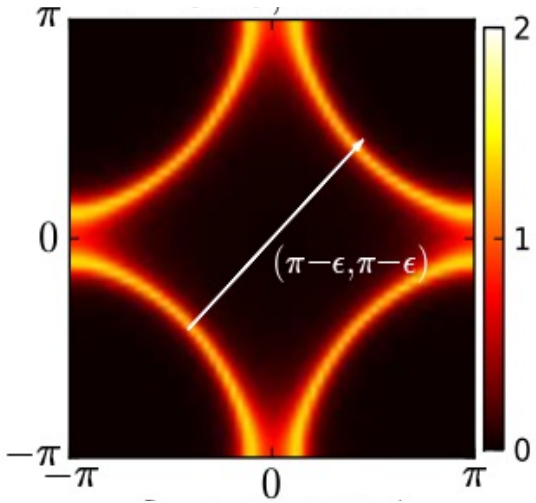
[1] G. Kotliar and D. Vollhardt, Physics Today 57, 53 (2004).
[2] L. de' Medici, J. Mravlje, and A. Georges, Phys. Rev. Lett. 107, 256401 (2011).

Non-local correlation and DMFT

Pseudo-gap in Cuprates



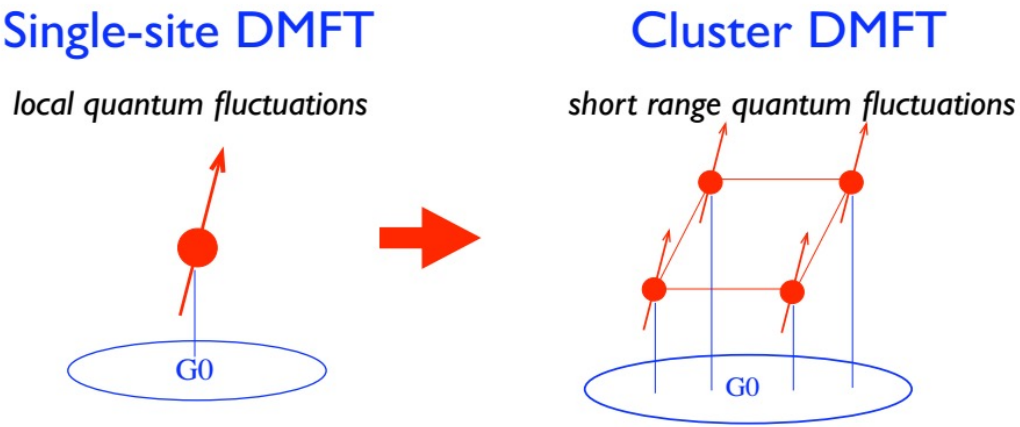
DMFT, Hubbard model



Non-local correlation is important for many fascinating phenomena including
- Pseudogap in cuprates
- Paramagnon near magnetic phase transition

[1] K. M. Shen, et al., Science 307, 901 (2005).
[2] H. Park, Ph.D thesis

Nonlocal self-energy: cluster extension

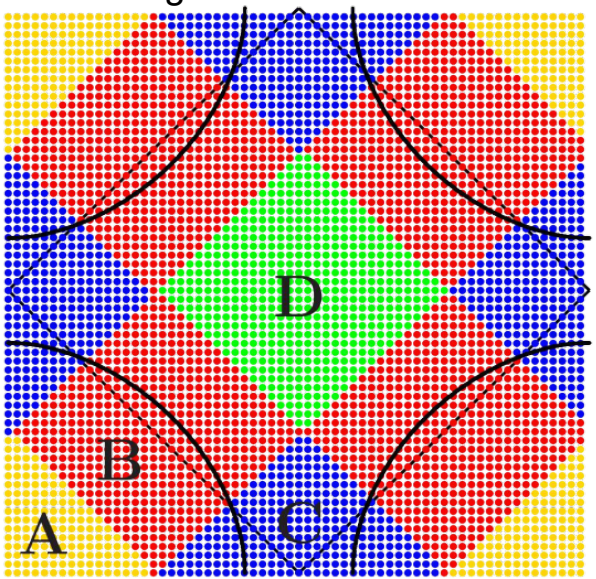


- Correlated orbitals on one site → correlated orbitals on several sites (cluster)
- Dynamical cluster approximation (in momentum space) and cellular DMFT (in real space)

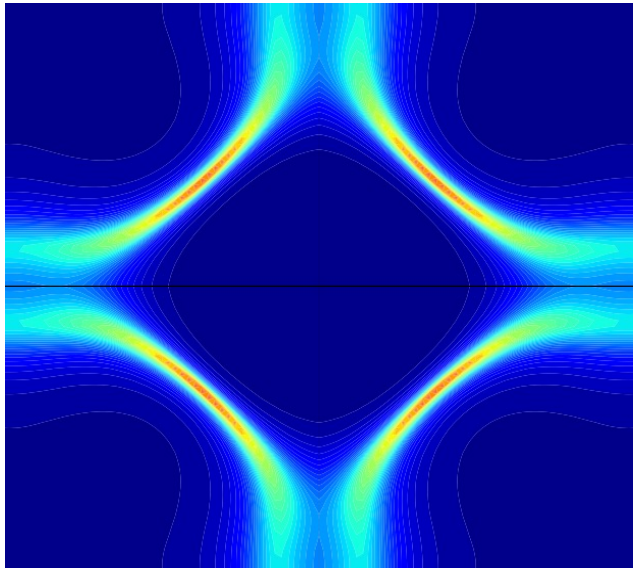
[1]M. H. Hettler, A. N. Tahvildar-Zadeh, M. Jarrell, T. Pruschke, and H. R. Krishnamurthy, Phys. Rev. B 58, R7475 (1998).
[2]A. I. Lichtenstein and M. I. Katsnelson, Phys. Rev. B 62, R9283 (2000).
[3]G. Kotliar, S. Y. Savrasov, G. Pálsson, and G. Biroli, Phys. Rev. Lett. 87, 186401 (2001).
Images from O.Parcollet's slide

Nonlocal self-energy: diagrammatic extension

Momentum resolution for cluster and diagrammatic extension



GW+EDMFT, Hubbard model



- GW+EDMFT, TRILEX, QUADRILEX, D\Gamma A, dual fermions, dual boson...
- local approximation to one physical quantity (such as self-energy, three point vertex…) and non-local self-energy through Feynman diagrams
- finer momentum resolution is possible than cluster extension.

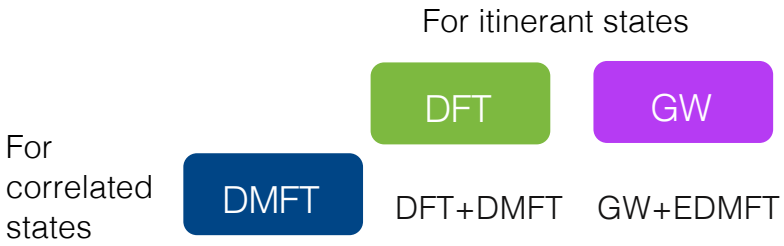
[1] G. Rohringer, et al., Rev. Mod. Phys. 90, 025003 (2018).
[2]J. Vucicevic, T. Ayral, and O. Parcollet, Physical Review B 96, (2017).

A Formal Viewpoint on First-principles and DMFT

[1] G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. 78, 865 (2006).
[2] G. Kotliar and S. Y. Savrasov, arXiv:Cond-Mat/0208241 (2002).
[3] R. M. Martin, L. Reining, and D. M. Ceperley, Interacting Electrons (Cambridge University Press, 2016).
[4] R. Chitra and G. Kotliar, Effective-Action Approach to Strongly Correlated Fermion Systems, Phys. Rev. B 63, 115110 (2001).

First principles +DMFT: quantum embedding

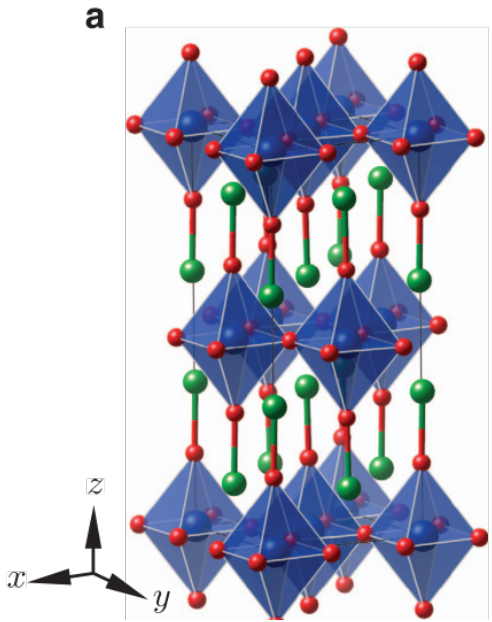
"Divide and Conquer"

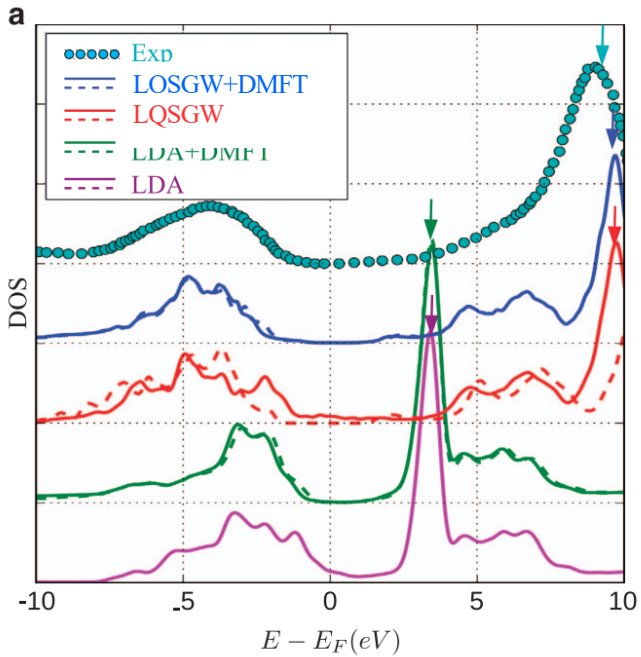


- Single particle Hilbert space partition into ``correlated'' and ``itinerant'' (e.g. Wannier functions)
- Correlated subspace within DMFT
- itinerant subspace from first-principles (DFT, GW…)
- Parameters to setup the quantum impurity problem(Coulomb interaction parameters and double-counting self-energy) should be chosen accordingly.
- Tradeoff between speed and accuracy

[1] G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. 78, 865 (2006).

LQSGW+DMFT validation on La2CuO4

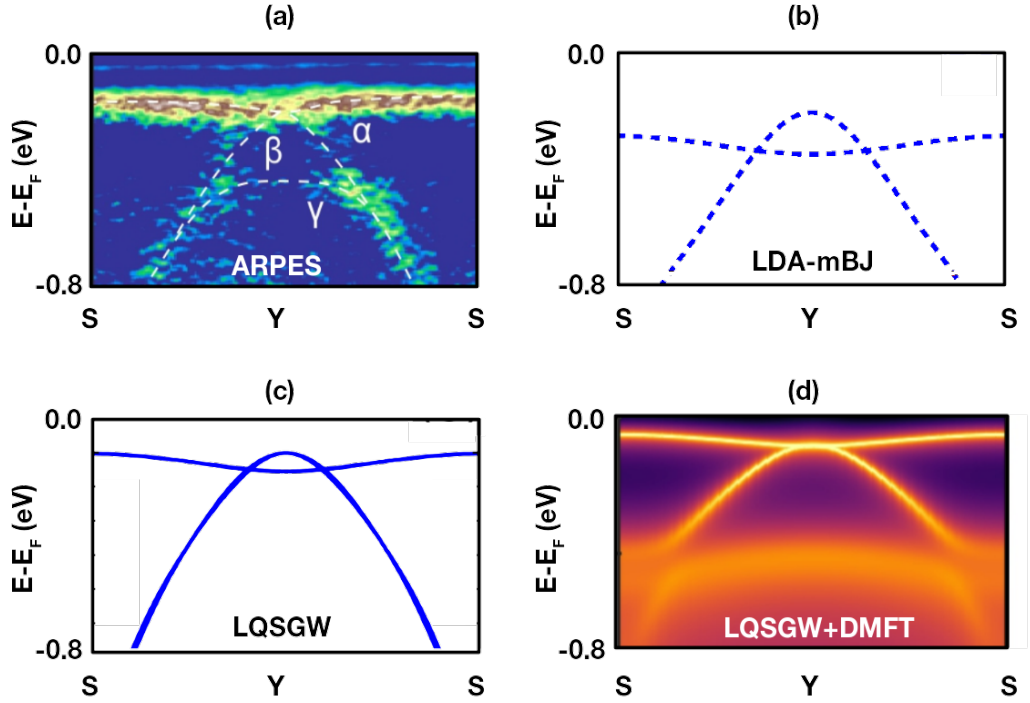




- LDA+DMFT and LQSGW+DMFT opens a paramagnetic Mott gap
- LQSGW predict a metal in non-magnetic calculation and an insulator in spin-polarized calculation.
- LDA predict a metallic phase
- better O-p and La-f position within LQSGW+DMFT

[1]S. Choi, A. Kutepov, K. Haule, M. van Schilfgaarde, and G. Kotliar, Npj Quantum Materials 1, 16001 (2016).

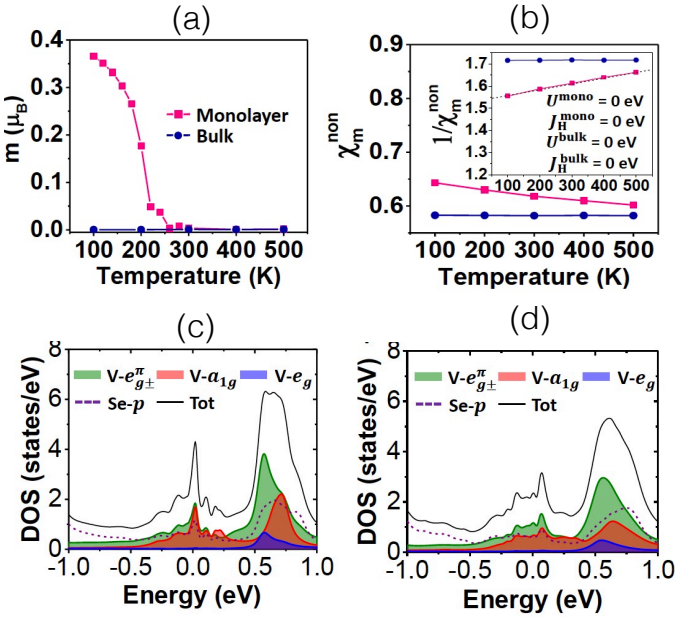
Theory Validation: $FeSb_2$ bandstructure



- $FeSb_2$: narrow-gap correlated semiconductor
- Colossal thermopower up to 45mV/K at 10K and a record-high thermoelectric power factor of 2300μW/K^{2}cm

[1] A. Chikina, et al., Phys. Rev. Research 2, 023190 (2020).

Ferromagnetism in suspended VSe2 monolayers



(a) Ferromagnetic order parameter (b) non-interacting local susceptibility (c) density of states in monolayer (d) density of states n bulk

Scientific Achievement

By using charge self-consistent LDA+DMFT approach,, we show the emergence of ferromagnetism in VSe2 in its monolayer limit below 250K, which is distinct from CrI3.

Significance and Impact

This study provides an important example

where the reduced dimensionality is an essential factor to form local moments and subsequent magnetic orderings. Our work will be useful in the understanding and design of possible magnetic devices based on 2D heterostructures

Research Details

- Magnetism is due to the 2D saddle point van hove singularity at the fermi level
- Interlayer coupling ruins the saddle point.
- Doping effect also hinders ferromagnetism.

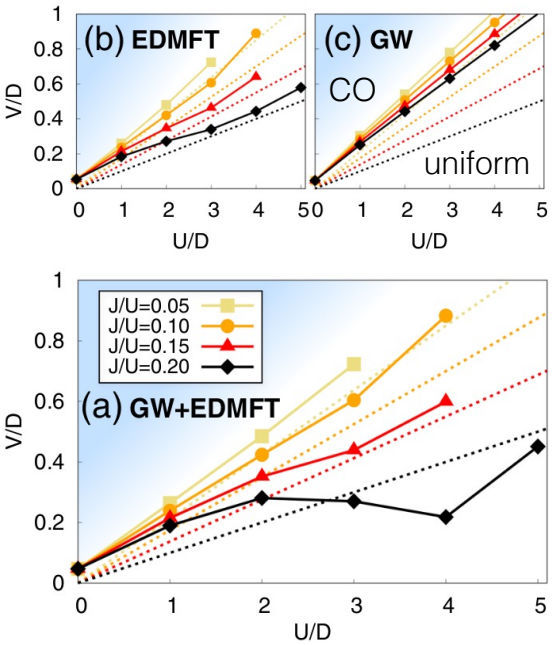
[1] T. J. Kim, S. Ryee, M. J. Han, and S. Choi,, 2D Mater. 7, 035023 (2020).

Does GW+EDMFT make a difference? Yes

$$\mathcal{H} = -t \sum_{\langle ij\rangle,\gamma,\sigma} \left(c^{\dagger}_{i\gamma\sigma} c_{j\gamma\sigma} + \text{H.c.}\right) - \mu \sum_{i,\gamma,\sigma} n_{i\gamma\sigma} + H_{\text{loc}} + H_{\text{nonloc}},$$

$$H_{\text{loc}} = U \sum_{i,\gamma,\sigma} n_{i\gamma\uparrow} n_{i\gamma\downarrow} + (U-2J) \sum_{i,\gamma,\gamma'}^{\gamma\neq\gamma'} n_{i\gamma\uparrow} n_{i\gamma'\downarrow} + (U-3J) \sum_{i,\gamma,\gamma',\sigma}^{\gamma<\gamma'} n_{i\gamma\sigma} n_{i\gamma'\sigma} - J \sum_{i,\gamma,\gamma'}^{\gamma\neq\gamma'} \left(c^{\dagger}_{i\gamma\uparrow} c_{i\gamma\downarrow} c^{\dagger}_{i\gamma'\downarrow} c_{i\gamma'\uparrow} + c^{\dagger}_{i\gamma\uparrow} c^{\dagger}_{i\gamma\downarrow} c_{i\gamma'\uparrow} c_{i\gamma'\downarrow}\right).$$

$$H_{\text{nonloc}} = \sum_{\langle ij\rangle\,\gamma,\gamma',\sigma,\sigma'} V n_{i\gamma\sigma} n_{j\gamma'\sigma'}.$$



- Hund's coupling induced valence-skipping (negative U) charge order instability enhancement
- Only visible within GW+EDMFT

[1]S. Ryee, P. Sémon, M. J. Han, and **S. Choi**, Npj Quantum Mater. 5, 1 (2020).

A Formal viewpoint on DFT+DMFT

Functional Approach to DFT

DFT Free energy functional

$$\Gamma_{DFT}[\rho(\mathbf{r}), v_{xc}(\mathbf{r})] = -\mathfrak{Tr}\left[\log(-G_0^{-1})\right] + \mathfrak{Tr}\left[\log(1 - G_0(v_{Hxc})\right] - \mathrm{Tr}\left[\rho v_{Hxc}\right] + \Phi_{DFT}[\rho]$$

Free energy functional for independent Kohn-Sham particles

Contribution to the K-S particles terms due to e-e interaction

e-e interaction

$$G_0^{-1}(\mathbf{r}, i\omega_n) = \left(i\omega_n + \mu + \nabla^2/2 - v_{ext}(\mathbf{r})\right))$$

Stationary condition

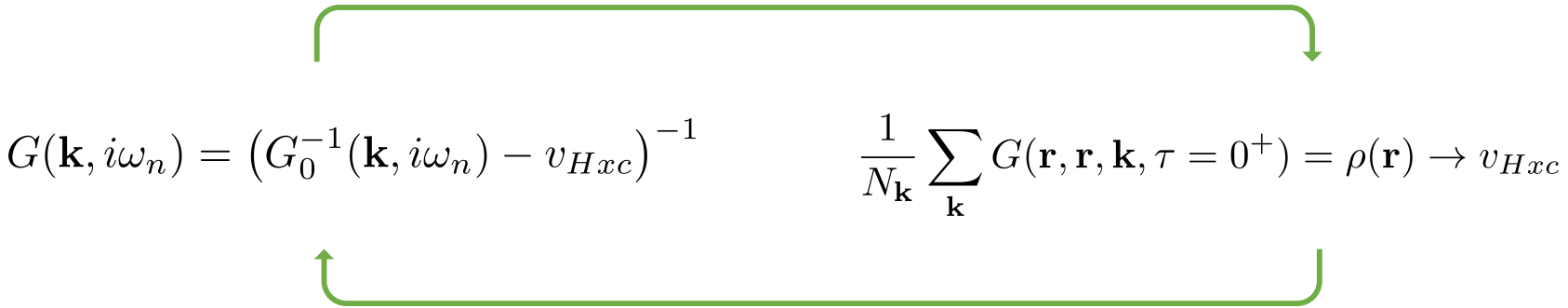
$$\frac{\delta\Gamma_{DFT}}{\delta\rho(\mathbf{r})} = 0 \rightarrow v_{Hxc}(\mathbf{r}) = \frac{\delta\Phi_{DFT}}{\delta\rho(\mathbf{r})}$$

$$\frac{\delta\Gamma_{DFT}}{\delta v_{Hxc}(\mathbf{r})} = 0 \rightarrow \underbrace{\left(G_0^{-1}(\mathbf{r}, \tau = 0^+) - v_{Hxc}(\mathbf{r})\right)^{-1}}_{= G(\mathbf{r}, \tau = 0^+)} = \rho(\mathbf{r})$$

$\Phi_{DFT}[\rho(\mathbf{r})]$ is unknown: local density approximation(LDA), generalized gradient approximation(GGA), …….

DFT self-consistent loop

Under a periodic potential



DMFT effective action

Action of Hubbard model

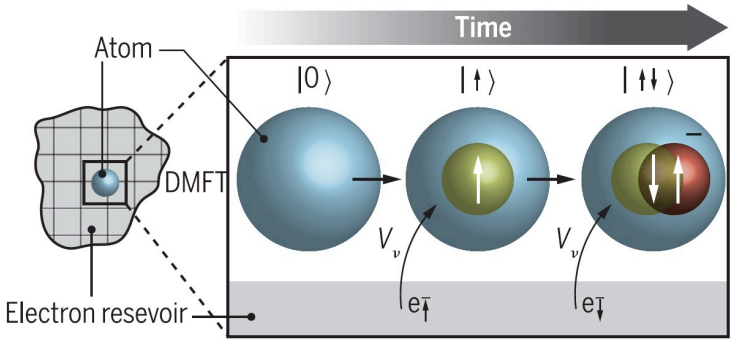
$$S = \int_0^\beta \left[\sum_{i,\sigma} c^\dagger_{i\sigma}(\tau)(\frac{\partial}{\partial\tau} - \mu)c_{i\sigma}(\tau) - \sum_{ij,\sigma} t_{ij}(c^\dagger_{i\sigma}(\tau)c_{j\sigma}(\tau) + h.c.) + U\sum_i n_{i\uparrow}(\tau)n_{i\downarrow}(\tau) \right]$$

One-site effective action (integrating out all the degree of freedom on all other lattice sites in the infinite dimension limit)

$$S_{eff} = S_{at} + S_{hyb}$$

$$S_{at} = \int_0^\beta d\tau \left[\sum_\sigma c^\dagger_\sigma(\tau) \left(-\frac{\partial}{\partial\tau} + \varepsilon \right) c_\sigma(\tau) + U n_\uparrow(\tau) n_\downarrow(\tau) \right]$$

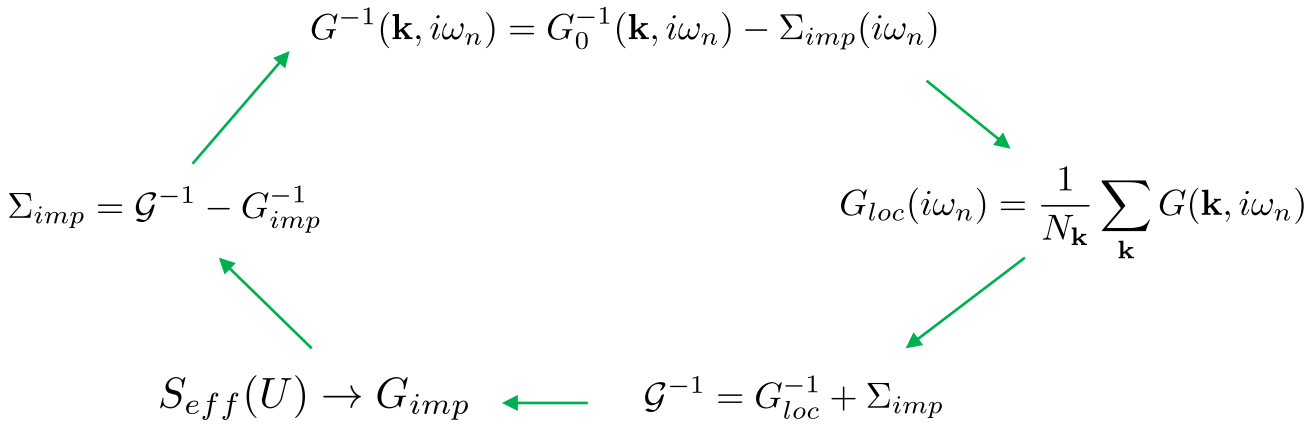
$$S_{hyb} = \int_0^\beta d\tau \int_0^\beta d\tau' \sum_\sigma c^\dagger_\sigma(\tau)\Delta(\tau - \tau')c_\sigma(\tau')$$



bare green's function: $\mathcal{G}_0(i\omega_n)^{-1} = i\omega_n - \varepsilon - \Delta(i\omega_n)$

DMFT self-consistent loop

Under a periodic potential



DMFT effective action

- Providing an interpolation from weak to strong coupling

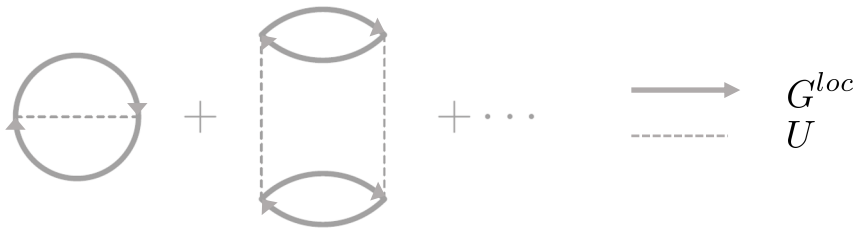
In the non-interacting limit

$$U = 0 \rightarrow \Sigma = 0$$

In the atomic limit

$$t = 0 \rightarrow \Delta = 0$$

- Dynamical but local self-energy
- Only single particles hop on and off the site
- Δ(τ) is self-consistently determined.
- U should be given
- $\Phi_{DMFT}[G^{loc}]$: sum of all possible two-particle irreducible local diagrams



Quantum projection and embedding

$A \rightarrow \langle \mathbf{r}|A|\mathbf{r}'\rangle$

$f = \langle \mathbf{r}|c_f\rangle \rightarrow$ a orthonormal basis set for correlated orbitals

$b = \langle \mathbf{r}|c_b\rangle \rightarrow$ a orthonormal product basis set composed of correlated orbitals

$\widetilde{A} \rightarrow \langle c_f|A|c'_f\rangle$ for a fermionic quantity and $\langle c_b|A|c'_b\rangle$ for a bosonic quantity

For example, if

$$A = \begin{pmatrix} A_{cc} & A_{ci} \\ A_{ic} & A_{ii} \end{pmatrix} \qquad f = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

Then, projection operation is given by

$$\widetilde{A} = f^\dagger A f = A_{cc}$$

Then, embedding operation is given by

$$f A_{cc} f^\dagger = \begin{pmatrix} A_{cc} & 0 \\ 0 & 0 \end{pmatrix}$$

Functional Approach to DFT+DMFT

DFT+DMFT Free energy Functional

$$\Gamma_{DFT+DMFT}[\rho(\mathbf{r}), v_{Hxc}(\mathbf{r}), \widetilde{G}_{\alpha\beta}(i\omega_n), \widetilde{\Sigma}_{\alpha\beta}(i\omega_n)]$$
$$= -\mathfrak{Tr}\left[\log(-G_0^{-1})\right] + \mathfrak{Tr}\left[\log(1 - G_0\{v_{Hxc} + f\widetilde{\Sigma}f^\dagger\})\right]$$
$$- \mathrm{Tr}\left[\rho v_{Hxc}\right] - \mathfrak{Tr}\left[\widetilde{G}\widetilde{\Sigma}\right] + \Phi_{DFT+DMFT}[\rho, \widetilde{G}]$$

Stationary condition

$$\frac{\delta\Gamma_{DFT+DMFT}}{\delta\rho(\mathbf{r})} = 0 \rightarrow v_{Hxc}(\mathbf{r}) = \frac{\delta\Phi_{DFT+DMFT}}{\delta\rho(\mathbf{r})}$$

$$\frac{\delta\Gamma_{DFT+DMFT}}{\delta\widetilde{G}_{\alpha\beta}(i\omega_n)} = 0 \rightarrow \frac{1}{\beta}\widetilde{\Sigma}_{\beta\alpha}(i\omega_n) = \frac{\delta\Phi_{DFT+DMFT}}{\delta\widetilde{G}_{\alpha\beta}(i\omega_n)}$$

$$\frac{\delta\Gamma_{DFT}}{\delta v_{KS}(\mathbf{r})} = 0 \rightarrow \underbrace{\mathfrak{Tr}\left[\left(G_0^{-1} - v_{Hxc} - f\widetilde{\Sigma}f^\dagger\right)\right]}_{= G(\mathbf{r}, \mathbf{r}, \tau = 0^+)} = \rho(\mathbf{r})$$

$$\frac{\delta\Gamma_{DFT+DMFT}}{\delta\widetilde{\Sigma}_{\alpha\beta}(i\omega_n)} = 0 \rightarrow f^\dagger \underbrace{\left(G_0^{-1} - v_{Hxc} - f\widetilde{\Sigma}f^\dagger\right)^{-1}}_{= G} f = \widetilde{G}$$

Functional Approach to DFT+DMFT

Approximation to $\Phi_{DFT+DMFT}$

$$\Phi_{DFT+DMFT}[\rho, \widetilde{G}] = \Phi_{DFT}[\rho] + \Phi_{DMFT}[\widetilde{G}] - \Phi_{DC}[\widetilde{G}]$$

$$\frac{\delta \Gamma_{DFT+DMFT}}{\delta \rho(\mathbf{r})} = 0 \rightarrow v_{Hxc}(\mathbf{r}) = \frac{\delta \Phi_{DFT}}{\delta \rho(\mathbf{r})}$$

$$\frac{\delta \Gamma_{DFT+DMFT}}{\delta \widetilde{G}_{\alpha\beta}(i\omega_n)} = 0 \rightarrow \frac{1}{\beta}\widetilde{\Sigma}_{\beta\alpha}(i\omega_n) = \frac{\delta \Phi_{DMFT}}{\delta \widetilde{G}_{\alpha\beta}(i\omega_n)} - \frac{\delta \Phi_{DC}}{\delta \widetilde{G}_{\alpha\beta}(i\omega_n)}$$

DMFT self-energy by solving DMFT effective action with a given Coulomb interaction tensor

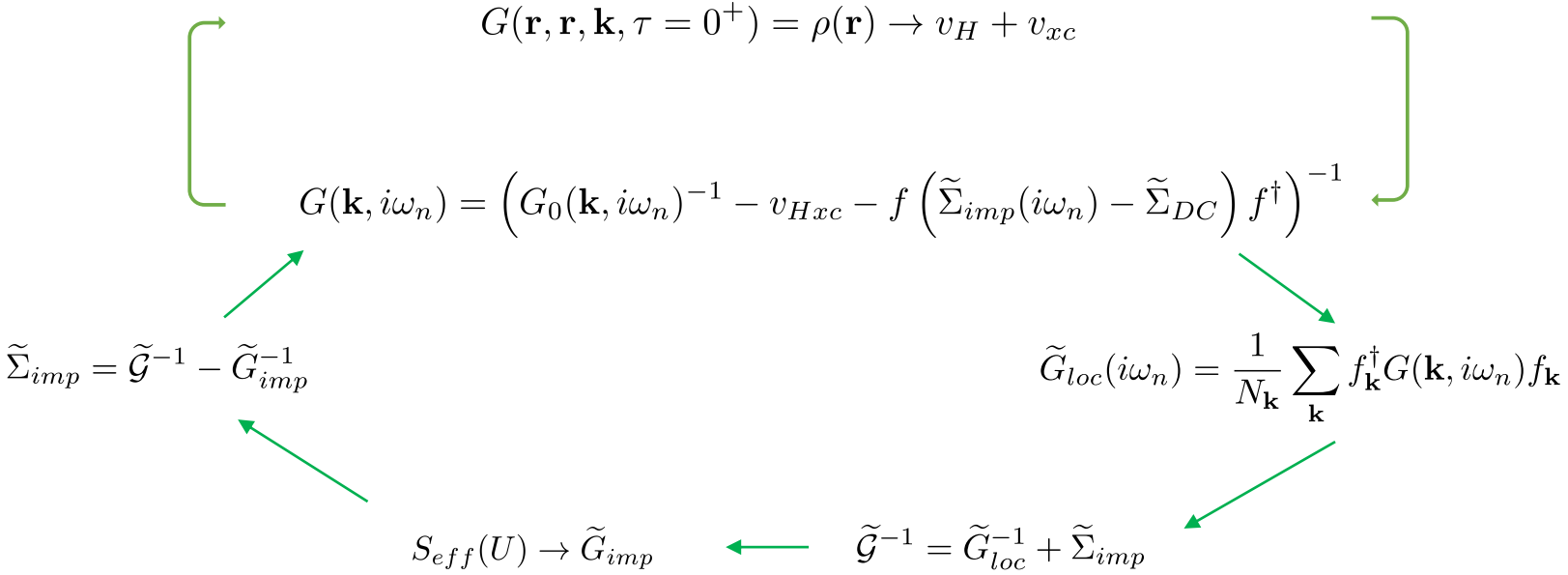
$$\widetilde{\Sigma}^{imp} = \frac{\delta \Phi_{DMFT}}{\delta \widetilde{G}_{\alpha\beta}(i\omega_n)} = \widetilde{\mathcal{G}}_0^{-1} - \widetilde{G}^{-1}$$

$$\widetilde{\Sigma}^{DC} = \frac{\delta \Phi_{DC}}{\delta \widetilde{G}_{\alpha\beta}} \simeq U(N - \frac{1}{2}) - J/2(N-1)$$

[1] K. Haule, Phys. Rev. Lett. 115, 196403 (2015).

Charge self-consistent DFT+DMFT loop

Under a periodic potential



U and $\widetilde{\Sigma}_{DC}$ as external parameters

A Formal viewpoint on GW+EDMFT

Functional Approach to GW

DFT Free energy functional

$$\Gamma_{GW}[G(\mathbf{r},\mathbf{r}',i\omega_n),\Sigma(\mathbf{r},\mathbf{r}',i\omega_n),W(\mathbf{r},\mathbf{r}',i\nu_n),P(\mathbf{r},\mathbf{r}',i\nu_n)]$$

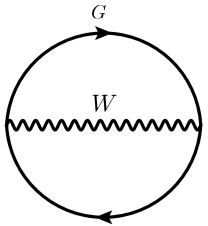
$$= -\mathfrak{Tr}\left[\log(-G_0^{-1})\right] + \mathfrak{Tr}\left[\log(1-G_0\Sigma)\right] - \mathfrak{Tr}\left[G\Sigma\right] + \Phi_H[G] + \frac{1}{2}\mathfrak{Tr}\left[\log(1-vP)\right] + \frac{1}{2}\mathfrak{Tr}\left[PW\right] + \Psi_{GW}[G,W]$$

Free energy functional due to G

Contribution to the first two terms due to e-e interaction

e-e interaction

$$\Psi_{GW}[G,W] = -\frac{1}{2}\mathfrak{Tr}\left[GWG\right]$$



Stationary condition

$$\frac{\delta\Gamma_{GW}}{\delta G} = 0 \rightarrow \Sigma_{xc} = -GW \qquad \frac{\delta\Gamma_{GW}}{\delta\Sigma} = 0 \rightarrow G^{-1} = G_0^{-1} - \Sigma_H - \Sigma$$

$$-2\frac{\delta\Gamma_{GW}}{\delta W} = 0 \rightarrow P = 2GG \qquad \frac{\delta\Gamma_{GW}}{\delta P} = 0 \rightarrow W^{-1} = v^{-1} - P$$

[1] C.-O. Almbladh, U. V. Barth, and R. V. Leeuwen, Int. J. Mod. Phys. B 13, 535 (1999).

GW loop

$$G^{-1}(\mathbf{k}, i\omega_n) = G_0^{-1}(\mathbf{k}, i\omega_n) - \Sigma(\mathbf{k}, i\omega_n)$$
$$W^{-1}(\mathbf{k}, i\omega_n) = V^{-1}(\mathbf{k}) - P(\mathbf{k}, i\omega_n)$$

$$\Sigma(\mathbf{k}, i\omega_n) = -\int_0^\beta d\tau \sum_{\mathbf{R}} G(\mathbf{R}, \tau) \cdot W(\mathbf{R}, \tau) e^{i(\mathbf{k}\cdot\mathbf{R} - \omega_n \tau)}$$

$$P(\mathbf{k}, i\omega_n) = \int_0^\beta d\tau \sum_{\mathbf{R}} G(\mathbf{R}, \tau) \cdot G(-\mathbf{R}, -\tau) e^{i(\mathbf{k}\cdot\mathbf{R} - \omega_n \tau)}$$

G: Green's function
Σ: electron self-energy
P: polarizability
W: screened Coulomb interaction

$\mathcal{G}$: fermionic Weiss field
$\mathcal{U}$: bosonic Weiss field
G_{imp}: impurity Green's function
P_{imp}: impurity polarizability
χ_{imp}: impurity susceptiblity
Σ_{DC}: double-counted self-energy
P_{DC}: double-counted polarizability

Extended DMFT action

Action of extended Hubbard model

$$S[c^*,c] = \int_0^\beta d\tau \left\{ \sum_{ij\sigma} c^*_{i\sigma}(\tau) \left[(\partial_\tau - \tilde{\mu})\,\delta_{ij} + t_{ij} \right] c_{j\sigma}(\tau) + \frac{1}{2} \sum_{ij} v_{ij} n_i(\tau) n_j(\tau) \right\}.$$

One-site effective action (infinite dimension limit)

$$S^{EDMFT} = S_{at} + S_{hyb} + S_{dyn}$$

$$S_{at} = \int_0^\beta d\tau \left[\sum_\sigma c^\dagger_\sigma(\tau) \left(-\frac{\partial}{\partial\tau} + \varepsilon \right) c_\sigma(\tau) + \mathcal{U}^\infty \sum_{\sigma,\sigma'} n_\sigma(\tau) n'_\sigma(\tau) \right]$$

$$S_{hyb} = \int_0^\beta d\tau \int_0^\beta d\tau' \sum_\sigma c^\dagger_\sigma(\tau) \Delta(\tau - \tau') c_\sigma(\tau')$$

$$S_{dyn} = \int_0^\beta d\tau \int_0^\beta d\tau' \sum_{\sigma,\sigma'} n_\sigma(\tau) F(\tau - \tau') n'_\sigma(\tau')$$

Fermionic bare green's function: $\mathcal{G}_0(i\omega_n)^{-1} = i\omega_n - \varepsilon - \Delta(i\omega_n)$

Bosonic bare green's function: $\mathcal{U}_0(i\nu_n) = F(i\nu_n) + \mathcal{U}^\infty$

EDMFT loop

$$G^{-1}(\mathbf{k}, i\omega_n) = G_0^{-1}(\mathbf{k}, i\omega_n) - \Sigma_{imp}(i\omega_n)$$
$$W^{-1}(\mathbf{k}, i\omega_n) = V^{-1}(\mathbf{k}) - P_{imp}(i\omega_n)$$

$$G_{loc}(i\omega_n) = \frac{1}{N_{\mathbf{k}}} \sum_{\mathbf{k}} G(\mathbf{k}, i\omega_n)$$
$$W_{loc}(i\omega_n) = \frac{1}{N_{\mathbf{k}}} \sum_{\mathbf{k}} W(\mathbf{k}, i\omega_n)$$

$$\mathcal{G}^{-1} = G_{loc}^{-1} + \Sigma_{imp}$$
$$\mathcal{U}^{-1} = W_{loc}^{-1} + P_{imp}$$

$$S_{eff} \rightarrow G_{imp}, \chi_{imp}$$

$$\Sigma_{imp} = \mathcal{G}^{-1} - G_{imp}^{-1}$$
$$W_{imp} = \mathcal{U} - \mathcal{U}\chi_{imp}\mathcal{U}$$
$$P_{imp} = \mathcal{U}^{-1} - W_{imp}^{-1}$$

G: Green's function
Σ: electron self-energy
P: polarizability
W: screened Coulomb interaction

$\mathcal{G}$: fermionic Weiss field
$\mathcal{U}$: bosonic Weiss field
G_{imp}: impurity Green's function
P_{imp}: impurity polarizability
χ_{imp}: impurity susceptiblity
Σ_{DC}: double-counted self-energy
P_{DC}: double-counted polarizability

EDMFT effective action

- Providing an interpolation from weak to strong coupling

In the non-interacting limit

$$v = 0 \rightarrow \Sigma = 0 \;\&\; P = 0$$

In the atomic limit

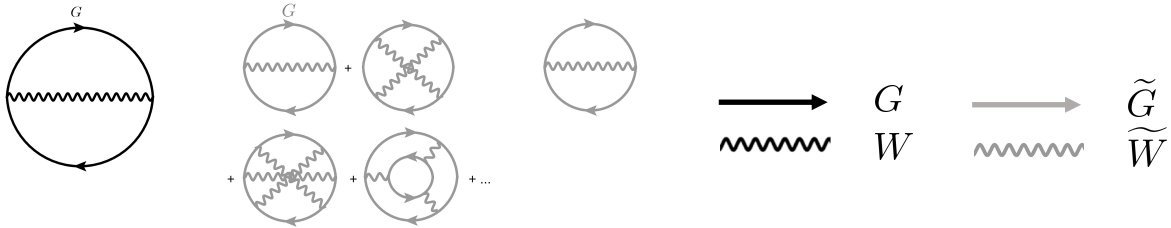
$$t = 0 \;\&\; v_{i \neq j} = 0 \rightarrow \Delta = 0 \;\&\; F = 0$$

- Dynamical but local self-energy and polarizability
- Δ(τ) and $P(\tau)$ are self-consistently determined.
- $\Phi_{DMFT}[G^{loc}, W^{loc}]$: sum of all possible two-particle irreducible local diagrams

Functional Approach to GW+EDMFT

Free energy Functional

$$\Gamma_{GW+EDMFT}[G(\mathbf{r},\mathbf{r}',i\omega_n),\Sigma(\mathbf{r},\mathbf{r}',i\omega_n),W(\mathbf{r},\mathbf{r}',i\nu_n),P(\mathbf{r},\mathbf{r}',i\nu_n)]$$
$$= -\mathfrak{Tr}\left[\log(-G_0^{-1})\right] + \mathfrak{Tr}\left[\log(1-G_0\Sigma)\right] - \mathfrak{Tr}\left[G\Sigma\right] + \Phi_H[G] + \frac{1}{2}\mathfrak{Tr}\left[\log(1-vP)\right] + \frac{1}{2}\mathfrak{Tr}\left[PW\right] + \Psi_{GW+EDMFT}[G,W]$$



$$\Psi_{GW+EDMFT}[G,W] = -\frac{1}{2}\mathfrak{Tr}\left[GWG\right] + \Psi_{EDMFT}[\widetilde{G},\widetilde{W}] + \frac{1}{2}\mathfrak{Tr}\left[\widetilde{G}\widetilde{W}\widetilde{G}\right]$$

Stationary condition

$$\frac{\delta\Gamma_{GW+EDMFT}}{\delta G} = 0 \rightarrow \Sigma_{xc} = -GW + \frac{\delta\Psi_{EDMFT}}{\delta G} + \widetilde{G}\widetilde{W} \qquad \frac{\delta\Gamma_{GW+EDMFT}}{\delta\Sigma} = 0 \rightarrow G^{-1} = G_0^{-1} - \Sigma_H - \Sigma_{xc}$$

$$\frac{\delta\Gamma_{GW+EDMFT}}{\delta W} = 0 \rightarrow P = 2GG + \frac{\delta\Psi_{EDMFT}}{\delta W} - 2\widetilde{G}\widetilde{G} \qquad \frac{\delta\Gamma_{GW+EDMFT}}{\delta P} = 0 \rightarrow W^{-1} = v^{-1} - P$$

[1] P. Sun and G. Kotliar, Phys. Rev. B 66, 085120 (2002). [2] S. Biermann, F. Aryasetiawan, and A. Georges, Phys. Rev. Lett. 90, 086402 (2003) [3]F. Nilsson, L. Boehnke, P. Werner, and F. Aryasetiawan, Phys. Rev. Materials 1, 043803 (2017).

GW+EDMFT loop

$$\Sigma(\mathbf{k}, i\omega_n) = \Sigma_{GW}(\mathbf{k}, i\omega_n) + f_{\mathbf{k}} \left(\widetilde{\Sigma}_{imp}(i\omega_n) - \widetilde{\Sigma}_{DC}(i\omega_n) \right) f_{\mathbf{k}}^{\dagger}$$

$$P(\mathbf{k}, i\nu_n) = P_{GW}(\mathbf{k}, i\nu_n) + b_{\mathbf{k}} \left(\widetilde{P}_{imp}(i\nu_n) - \widetilde{P}_{DC}(i\nu_n) \right) b_{\mathbf{k}}^{\dagger}$$

$$G^{-1}(\mathbf{k}, i\omega_n) = G_0^{-1}(\mathbf{k}, i\omega_n) - \Sigma(\mathbf{k}, i\omega_n)$$

$$W^{-1}(\mathbf{k}, i\omega_n) = V^{-1}(\mathbf{k}) - P(\mathbf{k}, i\omega_n)$$

$$\widetilde{G}_{loc}(i\omega_n) = \frac{1}{N_{\mathbf{k}}} \sum_{\mathbf{k}} f_{\mathbf{k}}^{\dagger} G(\mathbf{k}, i\omega_n) f_{\mathbf{k}}$$

$$\widetilde{W}_{loc}(i\omega_n) = \frac{1}{N_{\mathbf{k}}} \sum_{\mathbf{k}} b_{\mathbf{k}}^{\dagger} W(\mathbf{k}, i\omega_n) b_{\mathbf{k}}$$

$$\widetilde{\mathcal{G}}^{-1} = \widetilde{G}_{loc}^{-1} + \widetilde{\Sigma}_{imp}$$

$$\widetilde{\mathcal{U}}^{-1} = \widetilde{W}_{loc}^{-1} + \widetilde{P}_{imp}$$

$$S_{eff} \rightarrow \widetilde{G}_{imp}, \widetilde{\chi}_{imp}$$

$$\widetilde{\Sigma}_{imp} = \widetilde{\mathcal{G}}^{-1} - \widetilde{G}_{imp}^{-1}$$

$$\widetilde{W}_{imp} = \widetilde{\mathcal{U}} - \widetilde{\mathcal{U}} \widetilde{\chi}_{imp} \widetilde{\mathcal{U}}$$

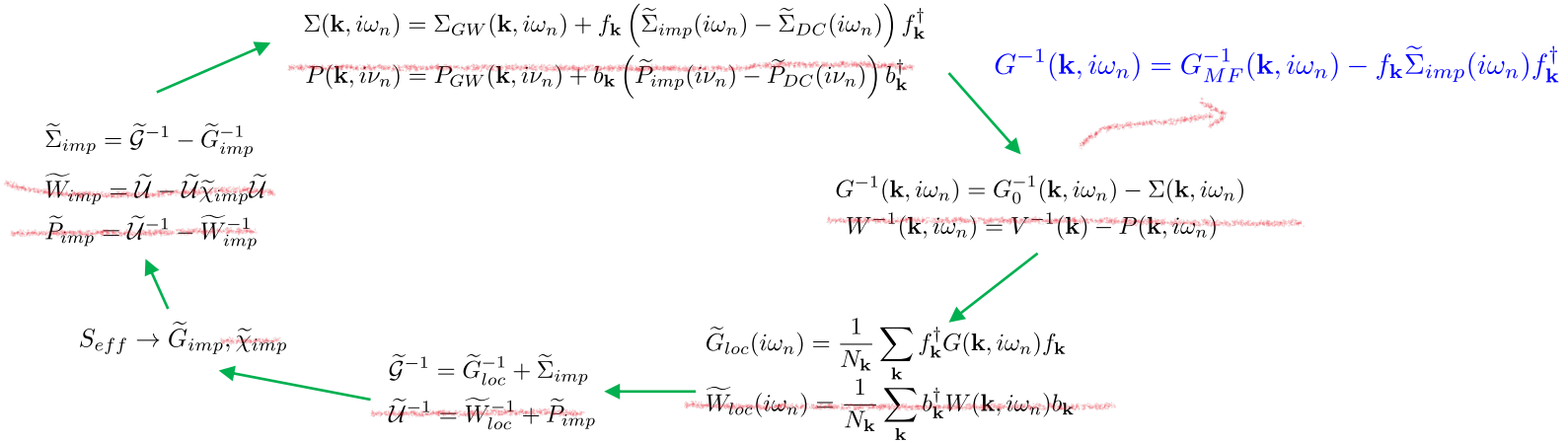
$$\widetilde{P}_{imp} = \widetilde{\mathcal{U}}^{-1} - \widetilde{W}_{imp}^{-1}$$

G: Green's function
Σ: electron self-energy
P: polarizability
W: screened Coulomb interaction

$\mathcal{G}$: fermionic Weiss field
$\mathcal{U}$: bosonic Weiss field
G_{imp}: impurity Green's function
P_{imp}: impurity polarizability
χ_{imp}: impurity susceptiblity
Σ_{DC}: double-counted self-energy
P_{DC}: double-counted polarizability

Partial Self-Consistency within LQSGW+DMFT

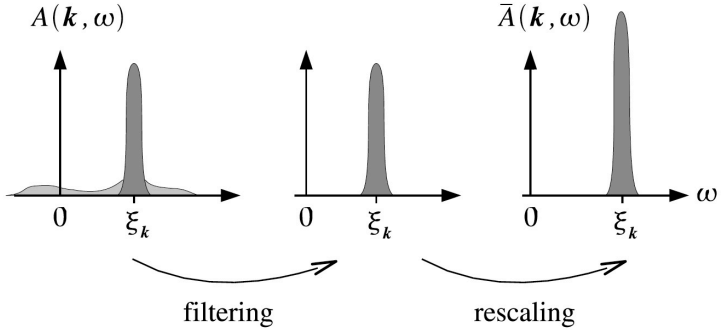
Various simplified approaches to GW+EDMFT



- fixing Bosonic quantities at the GW level
- U from constrained random phase approximation (cRPA) [1] and its extension [2] or constrained DFT(cDFT)
- One-shot DMFT approach: G_{MF} is fixed
- For G_{MF} : one-shot GW [7,8], Screened Exchange [3], QSGW [4,5] and LQSGW [2], non-local QSGW and LQSGW [6
- Multitier-GW+EDMFT [9] approach has been proposed: full GW+EDMFT approach in the low-energy space

[1] F. Aryasetiawan et al., Phys. Rev. B 70, 195104 (2004). [2] S. Choi, et al., Npj Quantum Materials 1, 16001 (2016).[3] A. van Roekeghem, et al., Phys. Rev. Lett. 113, 266403 (2014). [4] L. Sponzaet al., Phys. Rev. B 95, 041112 (2017). [5] D. Pashov et al., Computer Physics Communications 249, 107065 (2020).[6] J. M. Tomczak, J. Phys.: Conf. Ser. 592, 012055 (2015). [7] J. M. Tomczak et al., Phys. Rev. B 90, 165138 (2014).[8] C. Taranto et al., Phys. Rev. B 88, 165119 (2013). [9] F. Nilsson et al., Phys. Rev. Materials 1, 043803 (2017).

Mean-field starting point: quasiparticle Hamiltonian



$$G(i\omega_n) = \frac{1}{i\omega_n - \varepsilon_0 - \Sigma(i\omega_n)}$$

filtering

$$\simeq \frac{1}{i\omega_n - \varepsilon_0 - \Sigma(0) - i\omega_n \frac{\partial\Sigma(i\omega_n)}{\partial(i\omega_n)}\Big|_{\omega=0}}$$

$$= \frac{1}{i\omega_n \left(1 - \frac{\partial\Sigma(i\omega_n)}{\partial(i\omega_n)}\Big|_{\omega=0}\right) - \varepsilon_0 - \Sigma(0)}$$

$$= \frac{Z}{i\omega_n - \sqrt{Z}\left(\varepsilon_0 + \Sigma(0)\right)\sqrt{Z}}$$

rescaling

$$\simeq \frac{1}{i\omega_n - \sqrt{Z}\left(\varepsilon_0 + \Sigma(0)\right)\sqrt{Z}}$$

Quasiparticle weight: $$Z = \frac{1}{1 - \frac{\partial\Sigma(i\omega_n)}{\partial(i\omega_n)}\Big|_{\omega=0}}$$

Quasiparticle Hamiltonian: $$H_{QP} = \sqrt{Z}\left(\varepsilon_0 + \Sigma(0)\right)\sqrt{Z}$$

Linearized quasiparticle self-consistent GW method (LQSGW)

$$\Sigma(\mathbf{k}, i\omega_n) = -\int_0^\beta d\tau \sum_{\mathbf{R}} G_{QP}(\mathbf{R}, \tau) \cdot W(\mathbf{R}, \tau) e^{i(\mathbf{k}\cdot\mathbf{R} - \omega_n \tau)}$$

$$P(\mathbf{k}, i\omega_n) = \int_0^\beta d\tau \sum_{\mathbf{R}} G_{QP}(\mathbf{R}, \tau) \cdot G_{QP}(-\mathbf{R}, -\tau) e^{i(\mathbf{k}\cdot\mathbf{R} - \omega_n \tau)}$$

$$Z(\mathbf{k})^{-1} = 1 - \frac{\partial \Sigma(\mathbf{k}, \omega = 0)}{i\omega_n}$$

$$H_{QP}(\mathbf{k}) = \sqrt{Z(\mathbf{k})}\left(H_0(\mathbf{k}) + \Sigma(\mathbf{k}, \omega = 0)\right)\sqrt{Z(\mathbf{k})}$$

$$G_{QP}^{-1}(\mathbf{k}, i\omega_n) = i\omega_n - H_{QP}(\mathbf{k})$$

$$W^{-1}(\mathbf{k}, i\omega_n) = V^{-1}(\mathbf{k}) - P(\mathbf{k}, i\omega_n)$$

[1]A. L. Kutepov, V. S. Oudovenko, and G. Kotliar, Computer Physics Communications 219, 407 (2017).
[2]A. Kutepov, S. Y. Savrasov, and G. Kotliar, Phys. Rev. B 80, 041103 (2009).
[3]A. Kutepov, K. Haule, S. Y. Savrasov, and G. Kotliar, Phys. Rev. B 85, 155129 (2012).

Linearized Quasiparticle Self-consistent GW (LQSGW)

- Why LQSGW, not one-shot GW?

Oneshot GW

	$G_0W_0(0)$	$G_0W_0(0.25)$
Si	1.26	1.75
Ge	0.63	1.00
SiC	3.08	3.74
AlN	4.81	5.79
C	5.62	6.34
GaAs	1.21	1.83
GaP	2.42	2.97
GaN	2.75	3.67
ZnO	2.02	3.32
ZnSe	2.28	3.06
MoS_2	1.39	1.69
TiO_2	3.27	3.96
HfO_2	5.67	6.63
SiO_2	8.36	9.39
MgO	6.71	8.02
LiF	13.13	14.43

- Starting point dependent (PBE vs PBE0)

[1]W. Chen and A. Pasquarello, Phys. Rev. B 90, 165133 (2014).

Projector (f_k) construction by using Wannier functions

- From tight-binding model

$$|n\mathbf{k}\rangle = \frac{1}{\sqrt{N_{\mathbf{k}}}} \sum_{\mathbf{R},\tau} U_{n\tau}(\mathbf{k}) e^{i\mathbf{k}\cdot\mathbf{R}} |\tau\mathbf{R}\rangle$$

$$|\tau\mathbf{R}\rangle = \frac{1}{\sqrt{N_{\mathbf{k}}}} \sum_{\mathbf{k},n} U^*_{n\tau}(\mathbf{k}) e^{-i\mathbf{k}\cdot\mathbf{R}} |n\mathbf{k}\rangle$$

- One way to construct orthonormal basis set of $|\tau\mathbf{R}\rangle$ from $|n\mathbf{k}\rangle$, or to determine $U_{n\tau}(\mathbf{k})$
  → by minimizing total spread

$$\Omega = \sum_{\tau\mathbf{R}} \langle \mathbf{r}^2 - \langle \mathbf{r} \rangle^2_{\tau\mathbf{R}} \rangle_{\tau\mathbf{R}}$$, where $\langle A \rangle_{\tau\mathbf{R}} = \langle \mathbf{R}\tau | A | \mathbf{R}\tau \rangle$

→ Under the constraint that it preserves band eigenvalues $E_{n\mathbf{k}}$ in the inner (frozen) window

- Our default choice of inner (frozen) window: $E_F \pm$ 10eV

- then a projector to correlated orbitals

$$f_k = \langle \mathbf{r} | \tau\mathbf{k} \rangle = \sum_n U^*_{n\tau}(\mathbf{k}) \langle \mathbf{r} | n\mathbf{k} \rangle$$

[1] N. Marzari, A. A. Mostofi, J. R. Yates, I. Souza, and D. Vanderbilt, Maximally Localized Wannier Functions: Theory and Applications, Rev. Mod. Phys. 84, 1419 (2012).

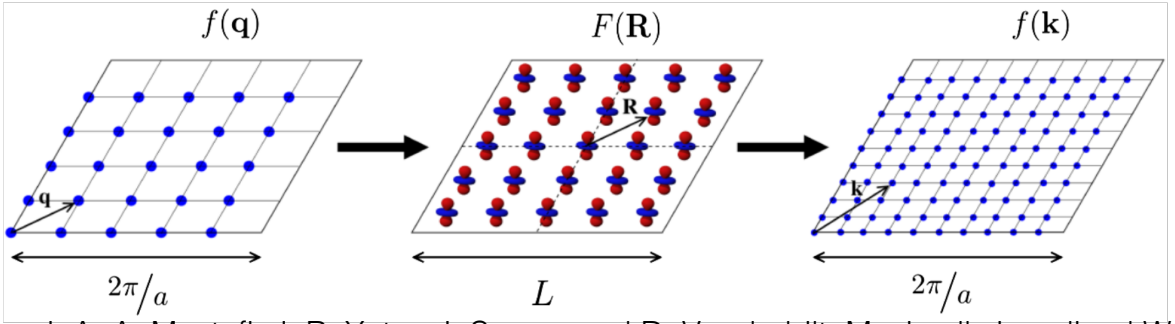
Wannier-interpolation of LQSGW bandstructure (H_{QP})

- The more localized orthonormal basis set → the sparser H matrix in the R space
- With localized basis set, hopping energy are essentially 0 beyond a few neighbours.
- If the supercell defined by k-grid is larger than the hopping range, we can interpolate the bands at a arbitrary k point

$$H_{\tau,\tau'}(\mathbf{k}) = \sum_{n,m} \langle \tau\mathbf{k}|n\mathbf{k}\rangle H_{n,m}(\mathbf{k}) \langle m\mathbf{k}|\tau'\mathbf{k}\rangle$$

$$H_{\tau,\tau'}(\mathbf{R}) = \frac{1}{N_{\mathbf{k}}} \sum_{\mathbf{k}} H_{\tau,\tau'}(\mathbf{k}) e^{-i\mathbf{k}\cdot\mathbf{R}}$$

$$H_{\tau,\tau'}(\mathbf{k}') = \sum_{\mathbf{R}} H_{\tau,\tau'}(\mathbf{R}) e^{i\mathbf{k}'\cdot\mathbf{R}}$$



[1] N. Marzari, A. A. Mostofi, J. R. Yates, I. Souza, and D. Vanderbilt, Maximally Localized Wannier Functions: Theory and Applications, Rev. Mod. Phys. 84, 1419 (2012).

Coulomb interaction matrix from cRPA

Suppose that the band-structure of a given solid can be separated in to a narrow band near the Fermi level and the rest.

Divide the complete Hilbert space into the subspace for the correlated orbitals and the rest

Aim: calculate the effective interaction between the correlated orbitals.

This interaction has to be bare with respect to the correlated orbitals but renormalized with respect to the rest

[1]F. Aryasetiawan, M. Imada, A. Georges, G. Kotliar, S. Biermann, and A. I. Lichtenstein, Phys. Rev. B 70, 195104 (2004).

How to define P_{QP}^{low}

- How to pick bands in the correlated subspace:
  - We pick the same number of bands as the number of correlated orbitals
  - The orbital character of the selected bands are mostly from the correlated orbitals

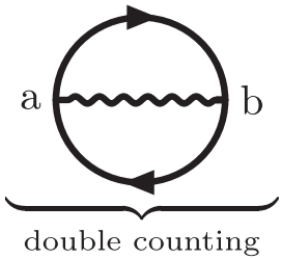
$$P_{QP} = P_{QP}^{low} + P_{QP}^{high}$$

$$P_{QP}^{low}(\mathbf{r}, \mathbf{r}', \mathbf{k}, i\omega_n) = -N_s \sum_{\mathbf{k}'} \sum_{n}^{\substack{\text{unocc} \\ \text{in the space}}} \sum_{m}^{\substack{\text{occ} \\ \text{in the space}}} \psi_{n\mathbf{k}'}(\mathbf{r}) \psi^*_{m\mathbf{k}'+\mathbf{k}}(\mathbf{r}) \psi^*_{n\mathbf{k}'}(\mathbf{r}') \psi_{m\mathbf{k}'+\mathbf{k}}(\mathbf{r}') \frac{2(E_{n\mathbf{k}'} - E_{n\mathbf{k}'+\mathbf{k}})}{\omega_n^2 - (E_{n\mathbf{k}'} - E_{n\mathbf{k}'+\mathbf{k}})^2},$$

$$W_r^{-1}(\mathbf{k}, i\omega_n) = W^{-1}(\mathbf{k}, i\omega_n) + P_{QP}^{low}(\mathbf{k}, i\omega_n)$$

$$\mathcal{U}(i\omega_n) = W_r(\mathbf{R} = 0, i\omega_n)$$

Double counting energy



$$\widetilde{\Sigma}^{DC}_{i,j}(i\omega_n) = -\sum_{k,l} 2\widetilde{G}_{k,l}(\tau = -\delta)\widetilde{U}_{i,j,k,l}(i\nu_n = 0)$$

$$-\sum_{k,l}\int d\tau \widetilde{G}_{k,l}(\tau)\widetilde{W}_{loc,i,k,l,j}(\tau)e^{i\omega_n\tau},$$

$$\widetilde{W}_{i,j,k,l}(i\nu_n) = \widetilde{U}_{i,j,k,l}(i\nu_n) + \sum_{m,n,p,q} \widetilde{U}_{i,j,m,n}(i\nu_n)\widetilde{P}_{m,n,p,q}(i\omega_n)\widetilde{W}_{p,q,k,l}(i\omega_n)$$

$$\widetilde{P}_{i,j,k,l}(i\omega_n) = 2\int d\tau \widetilde{G}_{i,l}(\tau)\widetilde{G}_{j,k}(-\tau)e^{i\omega\tau}$$

LQSGW+DMFT loop

$$\Sigma(\mathbf{k}, i\omega_n) = -\int_0^\beta d\tau \sum_{\mathbf{R}} G_{QP}(\mathbf{R}, \tau) \cdot W(\mathbf{R}, \tau) e^{i(\mathbf{k}\cdot\mathbf{R} - \omega_n \tau)}$$

$$P(\mathbf{k}, i\omega_n) = \int_0^\beta d\tau \sum_{\mathbf{R}} G_{QP}(\mathbf{R}, \tau) \cdot G_{QP}(-\mathbf{R}, -\tau) e^{i(\mathbf{k}\cdot\mathbf{R} - \omega_n \tau)}$$

$\Leftrightarrow$

$$Z(\mathbf{k})^{-1} = 1 - \frac{\partial \Sigma(\mathbf{k}, \omega = 0)}{i\omega_n}$$

$$H_{QP}(\mathbf{k}) = \sqrt{Z(\mathbf{k})}\,(H_0(\mathbf{k}) + \Sigma(\mathbf{k}, \omega = 0))\,\sqrt{Z(\mathbf{k})}$$

$$G_{QP}^{-1}(\mathbf{k}, i\omega_n) = i\omega_n - H_{QP}(\mathbf{k})$$

$$W^{-1}(\mathbf{k}, i\omega_n) = V^{-1}(\mathbf{k}) - P(\mathbf{k}, i\omega_n)$$

$$f_k = \langle \mathbf{r} | \tau \mathbf{k} \rangle = \sum_n U^*_{n\tau}(\mathbf{k}) \langle \mathbf{r} | n\mathbf{k} \rangle$$

For the GW/LDA part of the GW+DMFT/LDA+DMFT scheme, the code FlapwMBPT was used.

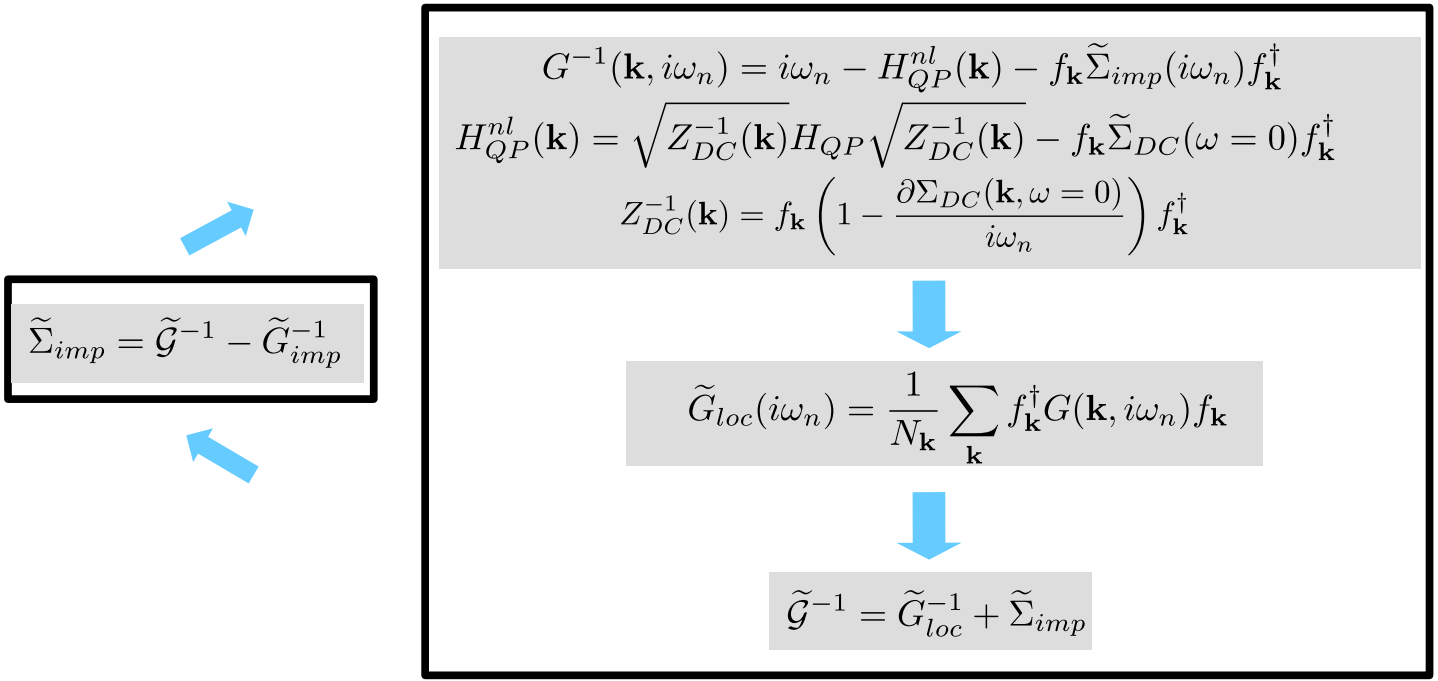
[1]A. L. Kutepov, V. S. Oudovenko, and G. Kotliar, Computer Physics Communications 219, 407 (2017).
[2]A. Kutepov, S. Y. Savrasov, and G. Kotliar, Phys. Rev. B 80, 041103 (2009).
[3]A. Kutepov, K. Haule, S. Y. Savrasov, and G. Kotliar, Phys. Rev. B 85, 155129 (2012).

LQSGW+DMFT loop

$$W_r^{-1}(\mathbf{k}, i\omega_n) = W^{-1}(\mathbf{k}, i\omega_n) + P_{QP}^{low}(\mathbf{k}, i\omega_n)$$
$$\widetilde{\mathcal{U}}(i\omega_n) = \widetilde{W}_r(\mathbf{R} = 0, i\omega_n)$$

$$\widetilde{\Sigma}_{i,j}^{DC}(i\omega_n) = -\sum_{k,l} 2\widetilde{G}_{k,l}(\tau = -\delta)\widetilde{U}_{i,j,k,l}(i\nu_n = 0) - \sum_{k,l} \int d\tau \widetilde{G}_{k,l}(\tau)\widetilde{W}_{loc,i,k,l,j}(\tau)e^{i\omega_n\tau}$$

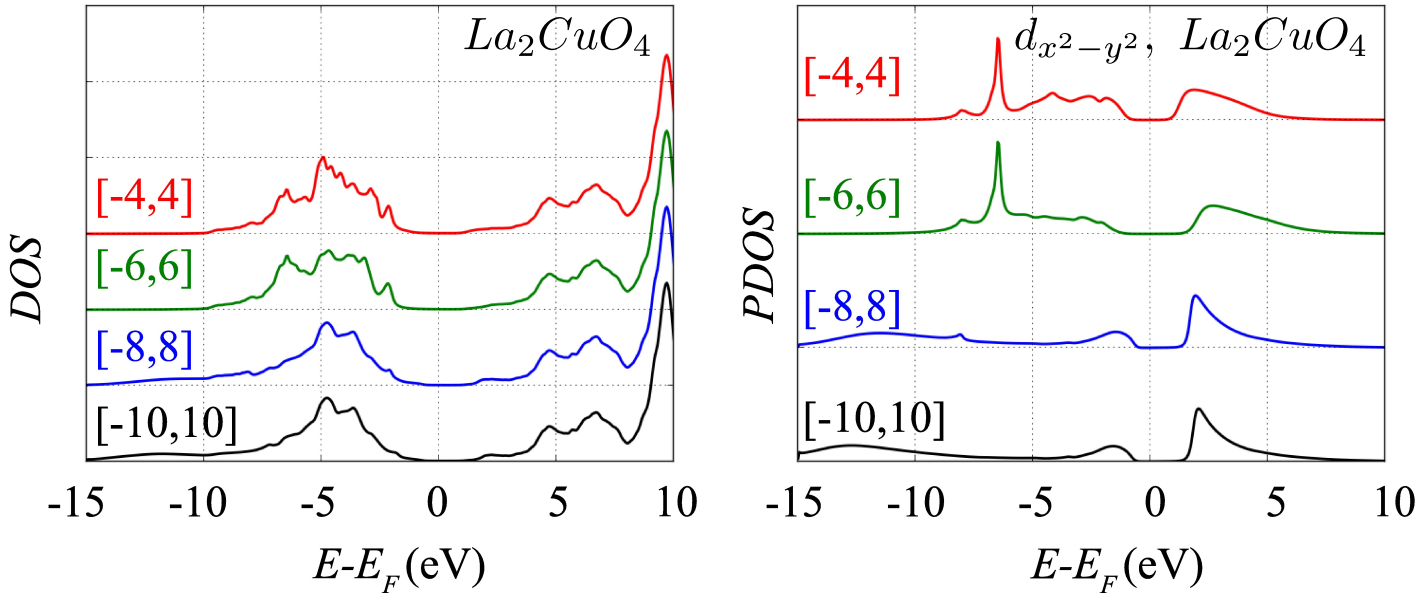
LQSGW+DMFT loop



- fixing Bosonic quantities at the GW level
- U from constrained random phase approximation (cRPA)
- One-shot DMFT approach: G_{MF} is fixed
- For the construction of G_{MF}: non-local LQSGW

[1] S. Choi, P. Semon, B. Kang, A. Kutepov, and G. Kotliar, Computer Physics Communications 244, 277 (2019).

Choice of correlated orbitals



- U and Σ_{DC} are self-consistently determined
- Identical Mott gap regardless of the choice of orbitals
- The position of Lower Hubbard band is sensitive to the choice

Intro to CTQMC solver

Corey Melnick

Hands-on Training

In this tutorial, commands you should run in terminal are marked by red colors

Special thanks to Vincent Sacksteder
for installing and testing Comsuite
in the virtual machine

https://github.com/comscope/comsuite

1. COMSUITE

A computational materials physics code for simulating correlated quantum materials using Dynamic Mean Field Theory (DMFT) and its extension. It can calculate the electronic structure within three different mathods:

- charge self-consistent LDA+Gutzwiller,
- charge self-consistent LDA+DMFT,
- and ab initio LQSGW+DMFT

For the copyright and license information, please see Copyright.txt and license.txt.

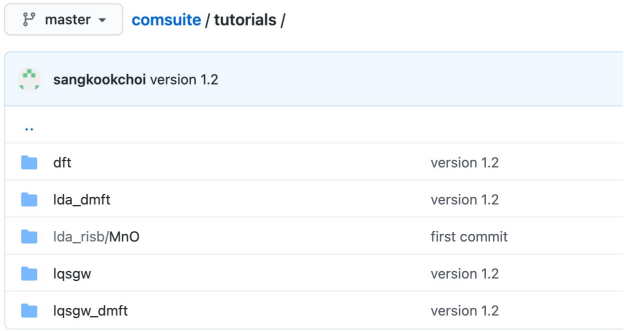
2. New version release announcement

2021. 2. 26

- Updated interface to Flapwmbpt. Now Comsuite requires single input file for FlapwMBPT, Comsuite, and its postprocess.
- Now Comsuite provides an option to calculate quasiparticle bandstructures within LDA+DMFT as well as LQSGW+DMFT.
- Now Comsuite provides options to choose "s"- or "p"-type corrlated orbitals.

2020. 1. 6

- Now Comsuite can calculate antiferromagnetically ordered phase. Please go to tutorial directories (install_directory/tutorials/lda_dmft/NiO_afm and install_directory/tutorials/lqsgw_dmft/NiO_afm). Read pdf f to learn how to calculate the electronic structures of antiferromagnetically ordered NiO. You have two choice of charge self-consistent LDA+DMFT and LQSGW+DMFT.



Compile COMSUITE package.

- First, define the installation directory in the shell. For example in bash shell, use the following command adds $COMSUITE_BIN to your system $PATH

```
export COMSUITE_BIN=install_directory/bin
```

- Then, the compilers, libraries, and flags should be defined in the arch.mk file. An example to install COMSUITE in Cori at NERSC is as follows.

```
##### fortran
F90 = ifort
PF90 = ftn
compfl = -O3

##### f2py
fortran2python = f2py -c --fcompiler=intelem --compiler=intelem

### phdf5
USE_HDF5 = defined  ### comment out this line if you don't want to compile with hdf5 (for LDA+DM
```

[1] S. Choi, P. Semon, B. Kang, A. Kutepov, and G. Kotliar,, Computer Physics Communications 244, 277 (2019).

Rationale of ComDMFT development (as a part of Comsuite)

- **Open source package** under GPL
  (the CPC paper on ComDMFT under review and in arxiv)

- **Multiple methodologies**
  - ab initio LQSGW+DMFT (a simplified version of GW+EDMFT)
  - charge self-consistent LDA+DMFT
  - charge self-consistent LDA+RISB (AKA ComRISB in collaboration with Yongxin)

- **The first open-source package supporting ab initio GW+DMFT**,
  - ab initio LQSGW+DMFT: **a parameter free method**

- **The first open-source package supporting multiple methodology based on first principles+DMFT**

- **GPU-ported impurity solver** by Kwangmin, Patrick and Corey,
  - Asynchronous CPU-GPU algorithm
  - 5X speed-up @TITAN, OLCF, **12.5X speed-up @SUMMIT, OLCF**

[1]S. Choi, P. Semon, B. Kang, A. Kutepov, and G. Kotliar, ArXiv181001679 Cond-Mat (2018).

ComDMFT (as a part of Comsuite)

- Physical observables
  - single particle Green's function
  - local quantities to correlated orbitals
    Impurity self-energy
    Hybridization function
    Double-counting self-energy (for LQSGW+DMFT)
    Bosonic Weiss field within cRPA (for LQSGW+DMFT)

- Paramagnetic phase and AFM phase

- Quasiparticle band within LDA+DMFT and LQSGW+DFMT

- For the LQSGW/LDA part of the LQSGW+DMFT/LDA+DMFT scheme, the code FlapwMBPT was used.

[1]A. L. Kutepov, V. S. Oudovenko, and G. Kotliar, Computer Physics Communications 219, 407 (2017).
[2]A. Kutepov, S. Y. Savrasov, and G. Kotliar, Phys. Rev. B 80, 041103 (2009).
[3]A. Kutepov, K. Haule, S. Y. Savrasov, and G. Kotliar, Phys. Rev. B 85, 155129 (2012).

Source code directory

Located at /home/max/codes/ComsuiteV2/source_code

`ComCTQMC`	: CTQMC impurity solver
`ComCoulomb`	: program to calculate bosonic Weiss field within cRPA
`ComDC`	: program for double counting self-energy
`ComLowH`	: program to calculate fermionic Weiss field, DOS, spectral functions
`ComRISB`	: program for Gutzwiller calculation
`ComWann`	: program to construct Wannier functions
`Copyright.txt`	
`GNUmakefile`	
`README.md`	
`arch.mk`	
`bin`	
`gw`	: FlapwMBPT code by Andrey Kutepov
`license.txt`	
`tutorials`	: tutorials (NiO and FeSe)
`wannier90_2.1`	: Wannier90 package

Environmental variable check

- Check if COMSUITE_BIN has been defined

    echo $COMSUITE_BIN

-If you get "/home/max/codes/Compiled_ComsuitCode/ComsuiteV2/source_code/bin", you are good. Otherwise,

1) Add the following line in the ~/.bashrc

    export COMSUITE_BIN=/home/max/codes/Compiled_ComsuiteCode/ComsuiteV2/source_code/bin

2) Execute the bash setup file

    source ~/.bashrc

3) Check if COMSUITE_BIN has been defined in your shell, again.

    echo $COMSUITE_BIN

Copy directories and input files

- Copy the directory with input files
  cp –r ~/codes/Compiled_ComsuiteCode/ComsuiteV2/tutorials_input ~/

- Move into the directory
  cd ~/tutorials_input

LDA+DMFT tutorial on Na

Warning: parameters in input files are set to run on two cpu cores in an hour.
This means that the results are not fully converged w.r.t. the number of k points,
CTQMC measurements, and LDA+DMFT iteration.

Converged calculation results are located at
“/home/max/codes/Compiled_ComsuiteCode/ComsuiteV2/tutorials_converged”

Simple metal including Na is not the best materials for local self-energy assumption

Goal

1. LDA+DMFT self-consistent calculation
2. Quasiparticle bandstructure calculation

DFT-LDA prerun

- Go to the DFT directory
  cd dft

- Check input files
  ls –al

```
Na.cif
comdmft.ini
```
: Crystallographic information file
: DFT calculation input file

- Run Comsuite
  $COMSUITE_BIN/comdmft.py

comdmft.ini

```
control ={
        'method':'dft',
        'mpi_prefix':'mpirun -np 2',
        'nproc_k_flapwmbpt':2,
        'nproc_tau_flapwmbpt':1,
        }

flapwmbpt={
        'cif':'./Na.cif',
        'iter_dft':50,
        'dft_mix':0.1,
        'rel':1,
        'magn':False,
        'kmesh':[5, 5, 5]
                }
```

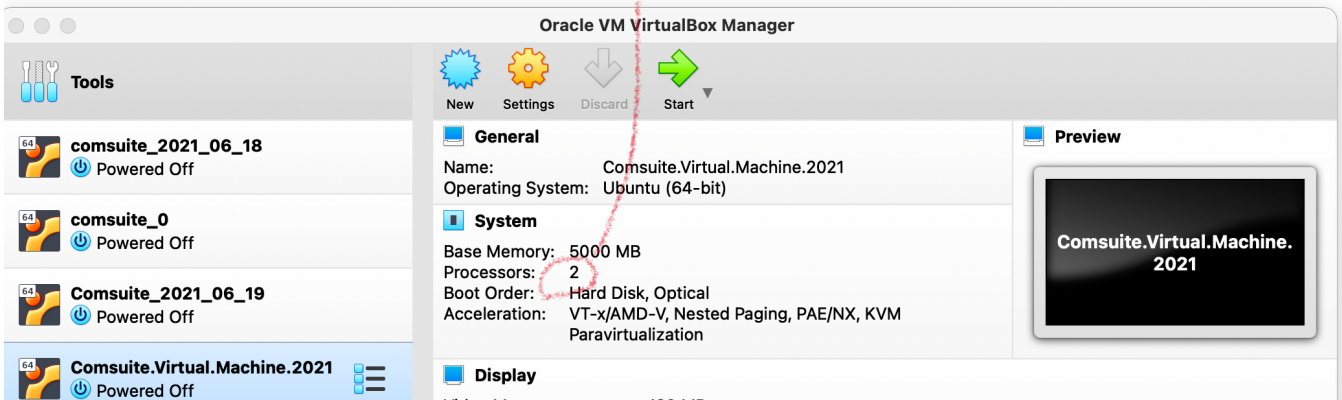
control in comdmft.ini

- method: ‘dft’
  ab initio methodology. Choose among “dft”, “lqsgw”, “lda+dmft”, or “lqsgw+dmft”.

- mpi_prefix: ‘mpirun –np 2’
  MPI prefix used for FlapwMBPT dft run.

- nproc_k_flapwmbpt: ‘2’
  The number of MPI processes for k parallelization

- restart:’false’
  option to resume dft calculation from a checkpoint. Default: False



flapwmbpt in comdmft.ini

- cif: ‘./Na.cif’

  the path to the cif file which contains crystal structure information

- iter_dft: ’50’

  the number of dft iteration

- dft_mix: ‘0.1’

  linear density mixing coefficient.

- rel: 1

  relativisity option (0: nonrelativistic, 1: scalar relativistic, 2: fully relativistic)

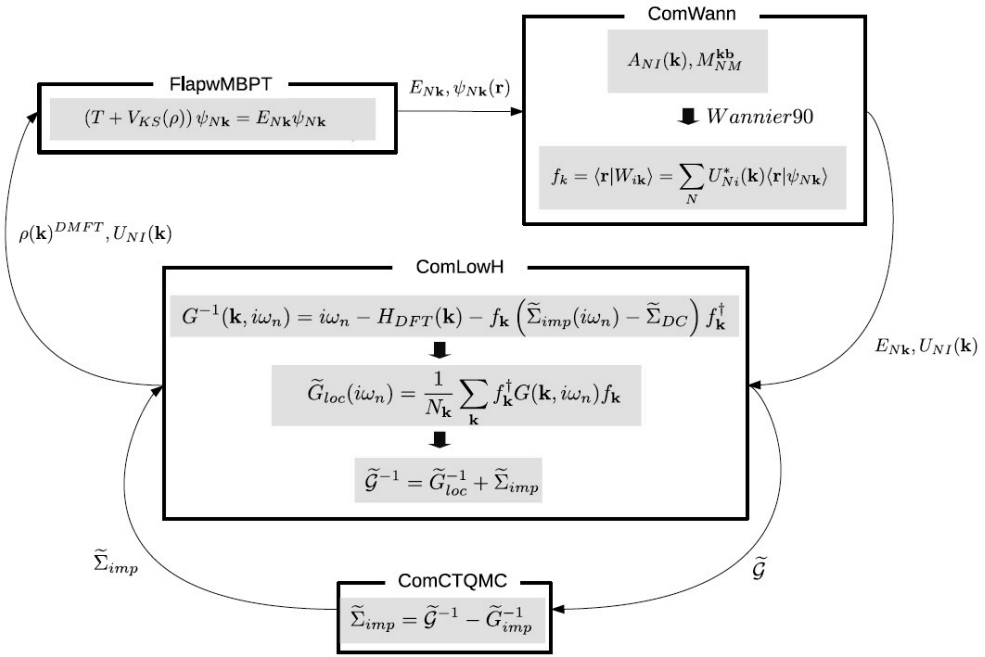
- magn: ‘false’

  spin polarization. ‘true’ or ‘false’

- kmesh: [5, 5, 5]

  k point grid

LDA+DMFT calculation



U and $\widetilde{\Sigma}_{DC}$ as external parameters

LDA+DMFT calculation

- Go to the LDA+DMFT directory
  cd ../lda_dmft

- Check input files
  ls –al

`analysis`	: directory containing python script for the data plots
`band`	: directory for the band plot postprocessing step
`comdmft.ini`	LDA+DMFT input file

- Run Comsuite
  $COMSUITE_BIN/comdmft.py

comdmft.ini for LDA+DMFT calculation

```
control={'initial_lattice_dir'          : '../dft',
         'method'                 : 'lda+dmft',
         'spin_orbit'             : False,
         'mpi_prefix': "mpirun -np 2",
         'impurity_problem':[[1, 's']],
         'impurity_problem_equivalence':[1],
         'restart': False,
         'max_iter_num_outer': 1
         }

wan_hmat={
        'kgrid': [5, 5, 5],
        'froz_win_min': -15.0,
        'froz_win_max': 10.0,
                    }

imp={'temperature'            : 900, # temperature (in K)
     '1':
     {
      'f0': 5.0,
      'nominal_n': 1.0,
     'impurity_matrix': [ # equivalent orbital index matrix. starting from 1.
         [1]
         ],
     'thermalization_time': 1,
     'measurement_time': 2,
     'green_cutoff':  10,
     'coulomb': 'full',
     }
     }
```

In control

- 'methods': 'lda+dmft'
- 'initial_lattice_dir': '../dft'
  - the path to DFT prerun directory
- 'impurity_problem': [[1,'s']]
  - a python list to specify correlated orbitals. The first and second indices are for the atom index and shell type.
  - atom index: in the order listed in the "../dft/crystal_structure.xsf"
  - shell index: "s", "p", "d" or "f"
- 'impurity_problem_equivalence': [1]
  - equivalence of each impurity problem.
  - identified by an integer starting from 1. If this value is the same, they are equivalent.
  - If this value is negative, it is the time-reversal symmetry pair to the one with the same absolute value.
- 'max_iter_num_outer': 1
  - maximum iteration for the charge self-consistent loop.
  - default value: 50
- 'spin_orbit': True or False
  - if False, correlated orbitals correspond to cubic spherical harmonics

$$Y_{lm} = \begin{cases} \frac{i}{\sqrt{2}} Y_l^{-|m|} - (-1)^m Y_l^{|m|}, & m < 0 \\ Y_l^0, & m = 0 \\ \frac{1}{\sqrt{2}} Y_l^{-|m|} + (-1)^m Y_l^{|m|}, & m > 0 \end{cases}$$

where Y_l^m is a spherical harmonics.

In control

- if True, correlated orbitals chosen at each correlated atom correspond spin-angular functions |l,i,m>

$$\Omega_{l,i=\pm\frac{1}{2},m} = \sum_{s\pm 1/2} C^{l,m}_{i,s} Y^{m-s}_{l}(\hat{r}) u_s$$

$$\text{where } u_s \text{ is a spinor, and } C^{l,m}_{i,s} = \langle l, m-s, \tfrac{1}{2}, s | l+i, m \rangle.$$

- 'mpi_prefix': : 'mpirun -np 2'
  - MPI prefix commonly used for ComLowH, ComWann, and ComCTQMC.
  - If a different MPI prefixs from this prefix is necessary for a program, use 'mpi_prefix_lowh', 'mpi_prefix_wannier', and 'mpi_prefix_impurity',
- 'restart': : False
  - True or False. If True, It will resume the calculation from the prerun.
  - default value: False
- 'mpi_prefix_lowh':
  - MPI prefix for ComLowH
  - default value: control['mpi_prefix']
- 'mpi_prefix_impurity':
  - MPI prefix for the impurity solver
  - default value: control['mpi_prefix']
- 'mpi_prefix_wannier'
  - MPI prefix for ComWann
  - default value: control['mpi_prefix']

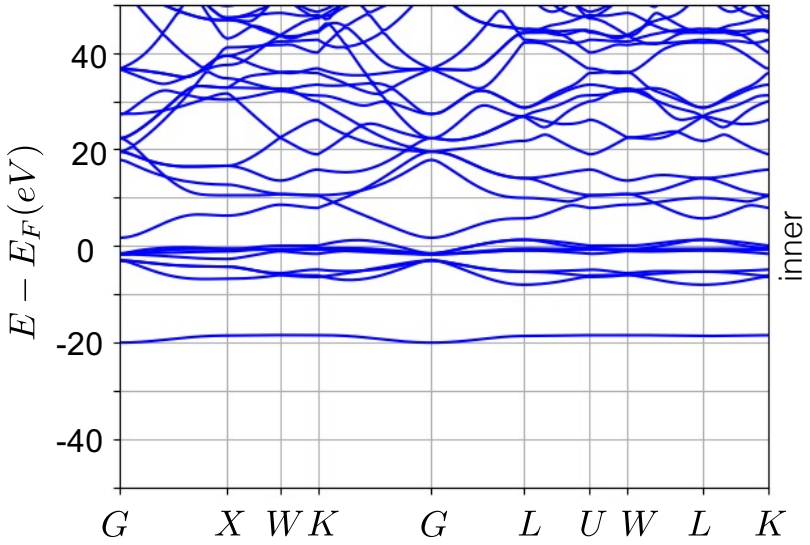
In control

- 'sigma_mix_ratio'
  - impurity self-energy linear mixing ratio.
  - default value: 0.5
- 'proj_win_min':
  - low-energy cutoff to renormalize the projectors
  - default value: wan_hmat['dis_win_min']
- 'proj_win_max':
  - high-energy cutoff to renormalize the projectors
  - default value: wan_hmat['dis_win_max']

Important concepts for wan_hmat

- For Wannier function construction
  - Choice of the inner (frozen) energy window: large energy window in the E_F±10eV
  - Choice of the outer (disentanglement) energy window: from E_F-10eV to E_F+50eV

NiO within LDA

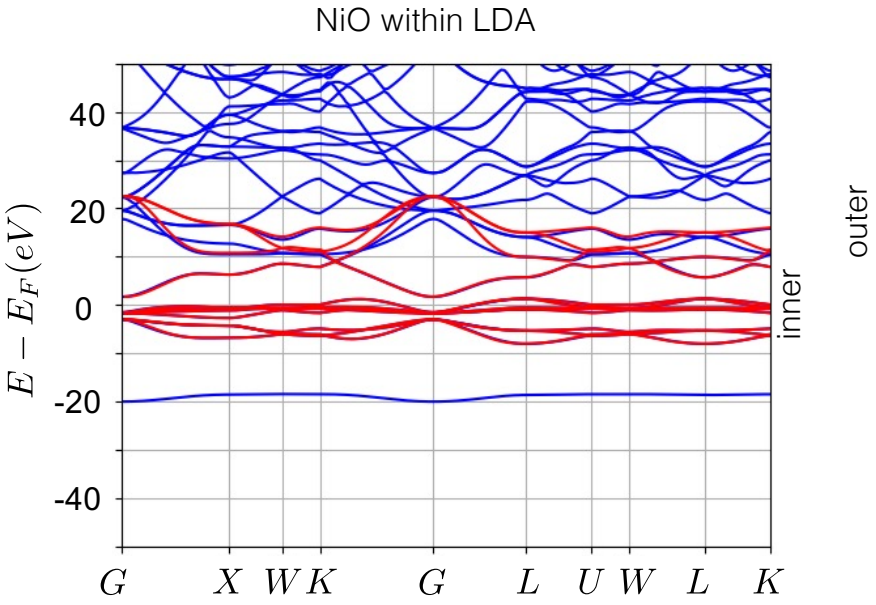


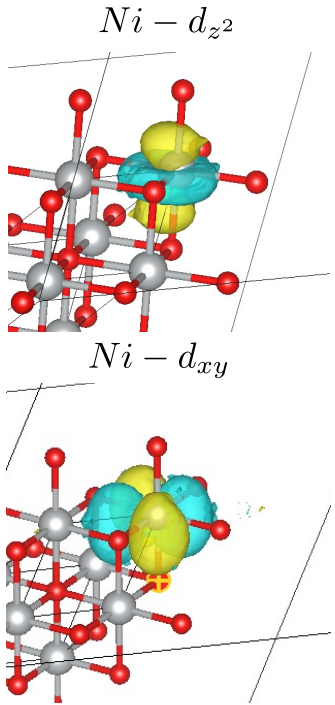
valence
+unoccupied states

Semicore states

Wannier functions and interpolated bandstructure of NiO

- The number of bands in the inner window: 10
- The number of bands in the outer window: 25
- The number of trial orbitals: 12 orbitals (Ni-s, Ni-p, Ni-d, O-p)





In wan_hmat

- 'kgrid': [5,5,5],
  - crystal momentum grid for the wannier interpolation of LDA bandstructure
- 'froz_win_min': -15 eV,
  - lower boundary of the inner (frozen) window in eV
- 'froz_win_max': 10 eV,
  - upper boundary of the inner (frozen) window in eV
- 'dis_win_min':
  - lower boundary of the outer (disentanglement) window in eV.
  - defaule value: froz_win_min
- 'dis_win_max':
  - upper boundary of the outer (disentanglement) window in eV.
  - defaule value: froz_win_max +40.0
- 'num_iter':
  - the number of minization step for the wannierization process. (gauge dependent part of total spreading)
  - default value: 0
- 'dis_num_iter':
  - the number of minization step for the disentanglement process. (gauge independent part of total spreading)
  - defaule value: 100

In imp

- **'temperature': 900**

  - simulation temperature in K

- for each distinct impurity problem indexed by the value in control ["impurity_problem_equivalence"]

  - 'impurity_matrix': [[1]],

    --equivalence of the matrix element of the fermionic Weiss field and impurity self-energy. Starting from "1"

    --if these values are the same, the values of the elements will be assumed to be identical.

    --if the element in the matrix is zero, then it will not be sampled by the impurity solver.

    --each column and row corresponds to the Wannier orbitals in the following order.

    --If control['spin_orbit']==False, "m" is sorted in ascending order.

    To illustrate for "d" orbitals, in this order: $|xy\rangle$,$|yz\rangle$,$|z^2\rangle$,$|xz\rangle$, $|x^2-y^2\rangle$

    --if control['spin_orbit']==True, the most rapidly changing index is "m" and the next one is "i". They are sorted in ascending order,

    To illustrate for "f" orbitals, in this order: |3,-0.5, -2.5>,|3,-0.5, -1.5>,|3,-0.5, -0.5>,
    |3,-0.5, 0.5>,|3,-0.5, 1.5>,|3,-0.5, 2.5>, |3,0.5, -3.5>,|3,0.5, -2.5>,|3,0.5, -1.5>,
    |3,0.5, -0.5>,|3,0.5, 0.5>,|3,0.5, 1.5>,|3,0.5, 2.5>, |3,0.5, 3.5>,

In imp

- 'Coulomb': ‘full’,
  --'full' or 'ising' are available. We construct Coulomb matrix in the following way.

$$U_{m_1,m_2,m_3,m_4} = \sum_{k=0}^{2l,even} \frac{4\pi}{2k+1} F_l^k \sum_{q=-k}^{k} \langle Y_l^{m_1} | Y_k^q Y_l^{m_4} \rangle \langle Y_l^{m_2} Y_k^q | Y_l^{m_3} \rangle$$

  --If 'full', no additional approximation is considered.
  --If 'ising', only U_{abba} or U_{abab} are non-zero.
- ‘f0’: 5
  the monopole term of bosonic Weiss field.
- ‘nominal_n’: 1
  electron occupation in the impurity orbital for the nominal double counting energy $U(N-\frac{1}{2}) - J/2(N-1)$
- 'thermalization_time': 1,
  » wall time for the thermalization in minutes
- 'measurement_time’: 2,
  » wall time for the measurement in minutes
- 'green_cutoff’: 20,
  » cutoff-energy in eV to sample green’s function and self-energy.
  » values beyond this energy will be provided by analytical equations.

Output directory

- in lda_dmft directory

```
analysis
band
cmd.log                 : command log file
comdmft.ini             : input file
convergence.log         : convergence log
dc                      : directory for the double counting calculation
delta.dat               : hybridization function
impurity                : directory for the quantum impurity problem
lattice                 : directory for DFT calculation with updated charge density
lowh                    : directory for fermionic Weiss field calculation
sig.dat                 : impurity self-energy
trans_basis.dat
wannier                 : directory for Wannier function calculation
```

convergence.log

step	i_outer	i_latt	i_imp	causality	delta_rho	w_sp_min	w_sp_max	mu	std_sig	n_imp	histo_1	histo_2	ctqmc_sign
wannier	1					2.83903403	3.16936125						
delta	1		1	good				-0.01645111621					
impurity_1	1		1	good					0.32635444719831463	0.7453301851	165.20786445892114	167.94632639486187	1
dft	2	1			0.003981117								
wannier	2					2.83903413	3.16935564						
delta	2		1	good				-0.157048587494					
impurity_1	2		1	good					0.20118722456537202	0.7152362616	159.4363906949742	166.62052643305128	1
dft	3	1			0.002464323								
wannier	3					2.84203035	3.16923542						
delta	3		1	good				-0.252597980239					
impurity_1	3		1	good					0.12672167809183085	0.6945175952	155.78643805195514	169.74120450338341	1
dft	4	1			0.001383939								
wannier	4					2.8452773	3.17114223						
delta	4		1	good				-0.315051432465					

- keeping track of convergence of some quantities at each iteration
- causality: causality of hybridization function / self-energy
- delta_rho: density changes from previous iteration
- w_sp_min: minimum spreading of the Wannier functions
- w_sp_max: maximum spreading of the Wannier functions
- mu: LDA+DMFT chemical potential w.r.t. LDA chemical potential
- std_sig:

$$\sqrt{\frac{\sum_i (\Sigma_i^j(i\omega_n) - \Sigma_i^{j-1}(i\omega_n))^2}{n_\omega n_{orb}}}$$

- n_imp: occupation in the impurity orbitals
- histo_1: the first moment of the perturbation order histogram
- histo_2: the second moment of the perturbation order histogram
- ctqmc_sign: CTQMC sign

Impurity self-energy

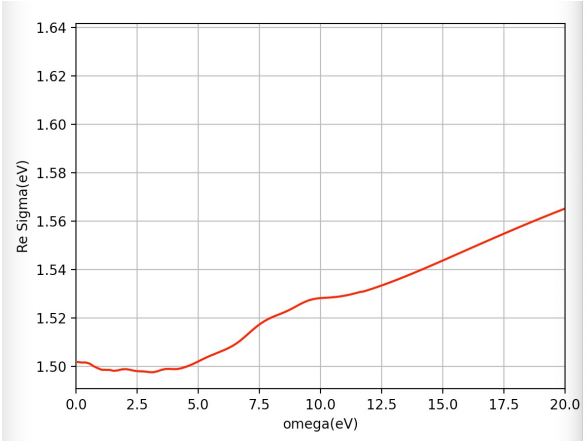
- “sig.dat”
  » Real and imaginary part of impurity self-energy

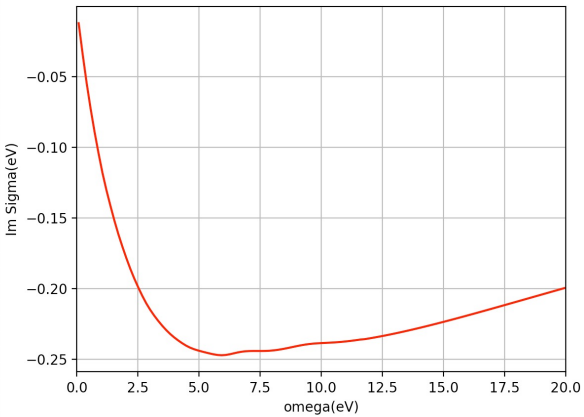
# omega(eV)	Re Sig_{1,1}(eV)	Im Sig_{1,1}(eV)
0.081216424692	1.501800431626	-0.012268251317
0.243649274076	1.501588177463	-0.034063609310
0.406082123461	1.501601337665	-0.054366239901
0.568514972845	1.501110822638	-0.072508424919
0.730947822229	1.500027409077	-0.089041707011
0.893380671613	1.499250948871	-0.104164319516
1.055813520997	1.498605056075	-0.118189374120
1.218246370382	1.498588018057	-0.130392007481
1.380679219766	1.498537768366	-0.141387853780
1.543112069150	1.498168137125	-0.151863364836
1.705544918534	1.498330599474	-0.161486104294

- Let’s plot it
  cd analysis
  python sig.py

Impurity self-energy

For the visualization, I used data at “/home/max/codes/ComsuiteV2/tutorials_converged”





- No divergent self-energy near Fermi-level.
- Up to 2eV, the impurity self-energy shows linear energy dependence.

Hybridization function

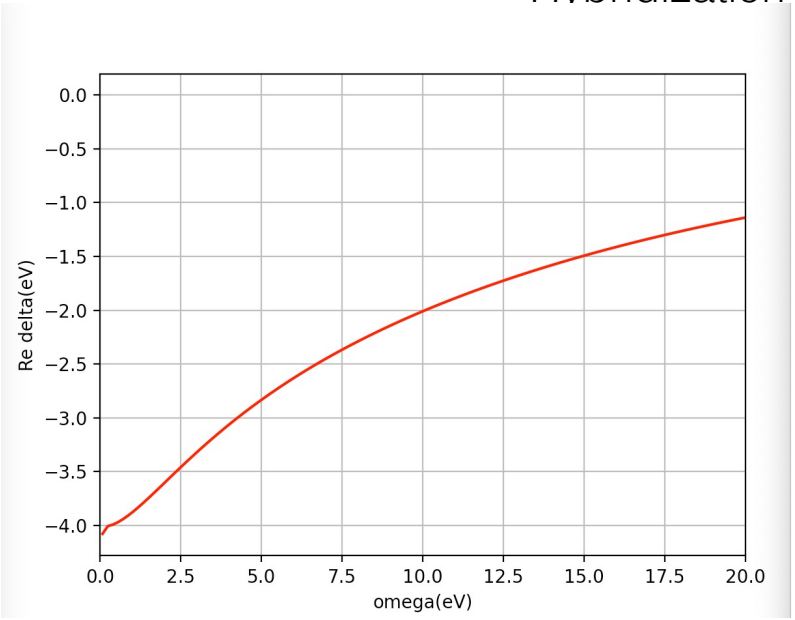
- “delta.dat”
  » Real and imaginary part of hybridization function

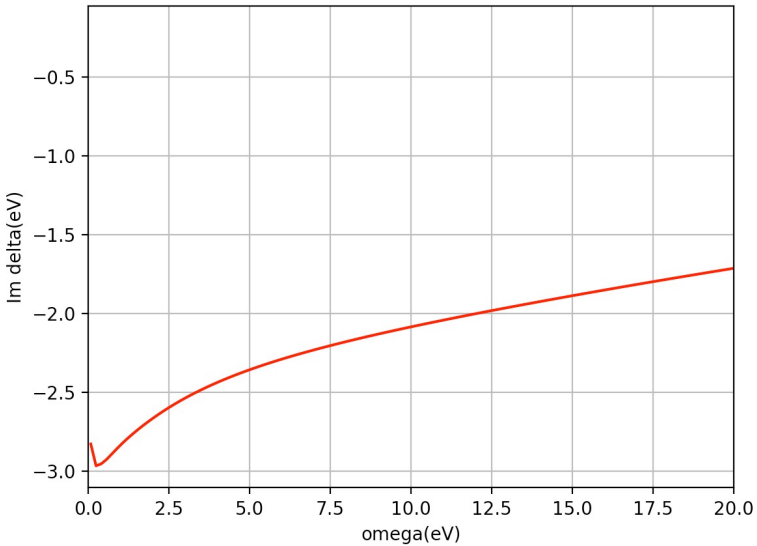
```
0.081216424692     -4.077402095983     -2.828521433978
0.243649274076     -4.006144819843     -2.966827164573
0.406082123461     -3.991149773784     -2.953881842766
0.568514972845     -3.968126243833     -2.926582297915
0.730947822229     -3.938154963765     -2.892171692592
0.893380671613     -3.903350147782     -2.857399777300
1.055813520997     -3.864792500134     -2.824214718055
1.218246370382     -3.823426294363     -2.793440776542
1.380679219766     -3.779812922422     -2.764457834912
1.543112069150     -3.734606699404     -2.736752971151
1.705544918534     -3.688584198508     -2.710484210705
1.867977767919     -3.642016615893     -2.685508905212
2.030410617303     -3.595109067127     -2.661525905159
```

- Let’s plot it

  python delta.py

Hvbridization function





- It shows metallic behavior (imaginary part of delta is nonzero at zero frequency)

LDA+DMFT quasiparticle bandstructure

- Go to the band directory
  cd ../band

- Check input files
  ls –al

`comdmft.ini`	Bandstructure calculation input file

- Run Comsuite
  $COMSUITE_BIN/comdmft.py

comdmft.ini

```
control ={
        'method':'band',
        'mpi_prefix':'mpirun -np 2',
        }

postprocessing={
        'comsuite_dir':'../',
        }
```

- in control
  - ‘method’: ‘band’
    postprocessing calculation. Choose among “band”, “dos”, and “spectral”

  - ‘mpi_prefix’: ‘mpirun –np 2’
    MPI prefix used for bandstructure calculation.

- in postprocessing
  - ‘comsuite_dir’: ‘../’
    The directory where comsuite calculation has been done.
  - ‘kpoints’:
    k point path along which spectral functions are calculated. If not provided, comsuite follow the path defined in Ref[1]

    [1] W. Setyawan and S. Curtarolo, Computational Materials Science 49, 299 (2010).

comdmft.ini

“kpoints” format

- w.r.t. reciprocal lattice vectors

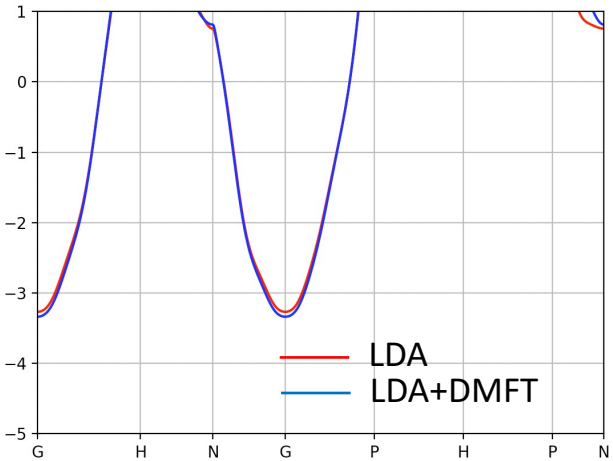
```
frac
  650
    1    0.00000000   0.00000000   0.00000000   G
    2    0.00423729  -0.00423729   0.00423729
    3    0.00847458  -0.00847458   0.00847458
    4    0.01271186  -0.01271186   0.01271186
    5    0.01694915  -0.01694915   0.01694915
    6    0.02118644  -0.02118644   0.02118644
    7    0.02542373  -0.02542373   0.02542373
    8    0.02966102  -0.02966102   0.02966102
    9    0.03389831  -0.03389831   0.03389831
```

LDA+DMFT bandstructure

Let's plot it

```
cd ../analysis
python ./band.py
```

LDA	mBJ	B3LYP	eDMFT	G_0W_0	Expt
3.30	3.29	4.09	2.84	3.15	2.65



[1] S. Mandal, K. Haule, K. M. Rabe, and D. Vanderbilt, ArXiv:2101.03262 [Cond-Mat] (2021).

LQSGW+DMFT tutorial on Na

Warning: parameters in input files are set to run on two cpu cores in an hour.
This means that the results are not fully converged w.r.t. the number of k points, CTQMC measurements, and DMFT iteration.

Converged calculation results are located at
“/home/max/codes/ComsuiteV2/tutorials_converged”

Simple metal including Na is not the best materials for local self-energy assumption

Goal

1. LQSGW+DMFT self-consistent calculation
2. Spectral function calculation

LQSGW prerun

Go to the DFT directory

cd ~/tutorials_input/lqsgw

Check input files

ls –al

`Na.cif`	: Crystallographic information file
`comdmft.ini`	: DFT calculation input file

- Run Comsuite

$COMSUITE_BIN/comdmft.py

comdmft.ini

```
control ={
        'method':'lqsgw',
        'mpi_prefix':'mpirun -np 2',
        'nproc_k_flapwmbpt':1,
        'nproc_tau_flapwmbpt':2,
        }

flapwmbpt={
        'cif':'./Na.cif',
        'iter_dft':50,
        'iter_lqsgw':1,
        'dft_mix':0.1,
        'rel':1,
        'magn':False,
        'kmesh':[5, 5, 5]
                }
```

control in comdmft.ini

- method: ‘lqsgw’
  ab initio methodology. Choose among “dft”, “lqsgw”, “lda+dmft”, or “lqsgw+dmft”.

- mpi_prefix: ‘mpirun –np 2’
  MPI prefix used for FlapwMBPT lqsgw run.

- nproc_k_flapwmbpt: ‘1’
  The number of MPI processes for k parallization

- nproc_tau_flapwmbpt: ‘2’
  The number of MPI processes for τ parallization

- restart:’false’
  option to resume lqsgw calculation from a checkpoint. Default: False

flapwmbpt in comdmft.ini

- cif: ‘./Na.cif’
  the path to the cif file which contains crystal structure information

- iter_dft: ’50’
  the number of dft iteration

- iter_lqwsgw: ’1’
  the number of lqsgw iteration

- dft_mix: ‘0.1’
  linear density mixing coefficient.

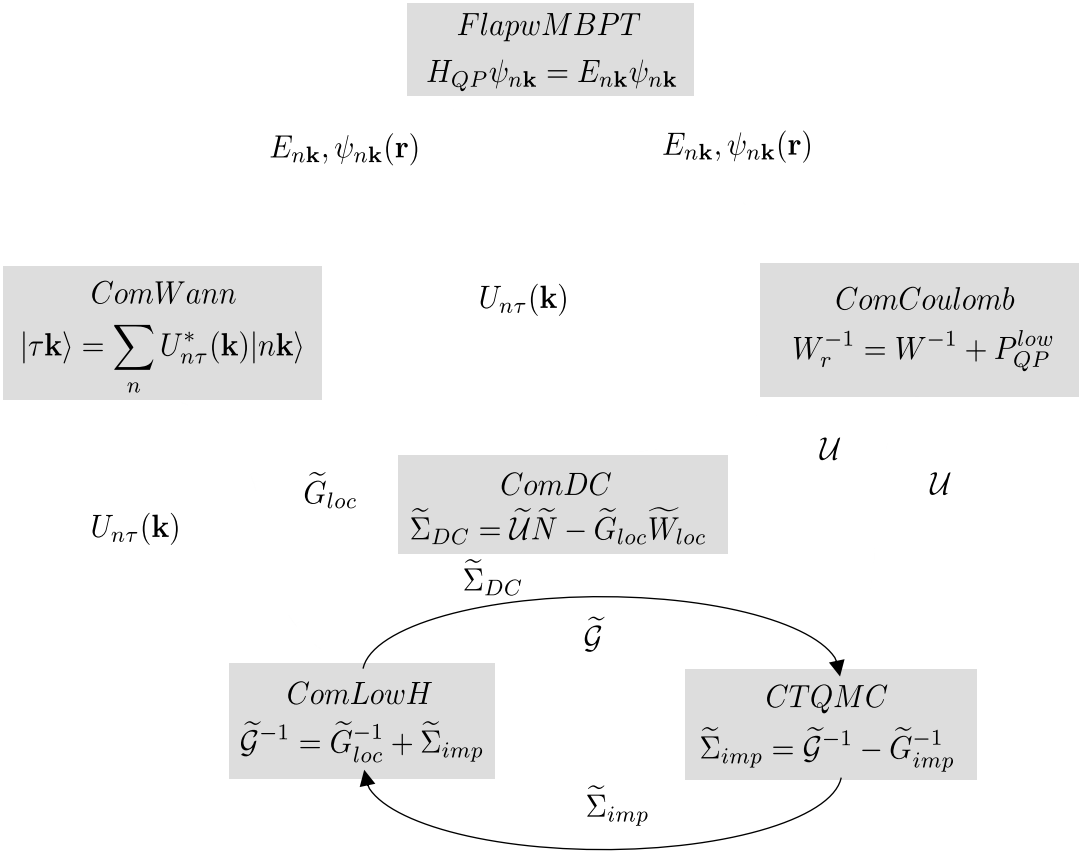
- rel: 1
  relativisity option (0: nonrelativistic, 1: scalar relativistic, 2: fully relativistic)

- magn: ‘false’
  spin polarization. ‘true’ or ‘false’

- kmesh: [5, 5, 5]
  k point grid

- gw_mix: ‘0.1’
  linear self-energy mixing coefficient

LQSGW+DMFT calculation



LQSGW+DMFT calculation

- Go to the LQSGW+DMFT directory
  cd ../lqsgw_dmft

- Check input files
  ls –al

analysis	: directory containing python script for the data plots
comdmft.ini	LQSGW+DMFT input file
realaxis	: directory for the spectral function plot postprocessing step

- Run Comsuite
  $COMSUITE_BIN/comdmft.py

comdmft.ini for LDA+DMFT calculation

```
control={'initial_lattice_dir'          : '../lqsgw',
         'method'                 : 'lqsgw+dmft',
         'spin_orbit'             : False,
         'mpi_prefix': "mpirun -np 2",
         'impurity_problem':[[1, 's']],
         'impurity_problem_equivalence':[1],
         'restart': False,
         'max_iter_num_impurity': 1
         }

wan_hmat={
        'kgrid': [5, 5, 5],
        'froz_win_min': -15.0,
        'froz_win_max': 10.0,
                       }

imp={'temperature'            : 900, # temperature (in K)
     '1':
     {
     'impurity_matrix': [ # equivalent orbital index matrix. starting from 1.
         [1]
         ],
     'thermalization_time': 1,
     'measurement_time': 2,
     'green_cutoff':  20,
     'coulomb': 'full',
     }
     }
```

In control

- 'methods': 'lqsgw+dmft'
- 'initial_lattice_dir': '../lqsgw'
  - the path to LDA output directory
- 'impurity_problem': [[1,'s']]
  - a python list to specify correlated orbitals. The first and second indices are for the atom index and shell type.
  - atom index: in the order listed in the "../dft/crystal_structure.xsf"
  - shell index: "s", "p", "d" or "f"
- 'impurity_problem_equivalence': [1]
  - equivalence of each impurity problem.
  - identified by an integer starting from 1. If this value is the same, they are equivalent.
  - If this value is negative, it is the time-reversal symmetry pair to the one with the same absolute value.
- 'max_iter_num_impurity': 1
  - total number of DMFT self-consistent loop
- 'spin_orbit': True or False
  - if False, correlated orbitals correspond to cubic spherical harmonics

$$Y_{lm} = \begin{cases} \frac{i}{\sqrt{2}} Y_l^{-|m|} - (-1)^m Y_l^{|m|}, & m < 0 \\ Y_l^0, & m = 0 \\ \frac{1}{\sqrt{2}} Y_l^{-|m|} + (-1)^m Y_l^{|m|}, & m > 0 \end{cases}$$

where Y_l^m is a spherical harmonics.

In control

- if True, correlated orbitals chosen at each correlated atom correspond spin-angular functions |l,i,m>

$$\Omega_{l,i=\pm\frac{1}{2},m} = \sum_{s\pm 1/2} C^{l,m}_{i,s} Y^{m-s}_{l}(\hat{r}) u_s$$

$$\text{where } u_s \text{ is a spinor, and } C^{l,m}_{i,s} = \langle l, m-s, \tfrac{1}{2}, s | l+i, m \rangle.$$

- 'mpi_prefix': : 'mpirun -np 2'
  - MPI prefix commonly used for ComLowH, ComWann, and ComCTQMC.
  - If a different MPI prefixs from this prefix is necessary for a program, use 'mpi_prefix_lowh', 'mpi_prefix_wannier', and 'mpi_prefix_impurity',
- 'restart': : False
  - True or False. If True, It will resume the calculation from the prerun.
  - default value: False
- 'mpi_prefix_lowh':
  - MPI prefix for ComLowH
  - default value: control['mpi_prefix']
- 'mpi_prefix_impurity':
  - MPI prefix for the impurity solver
  - default value: control['mpi_prefix']
- 'mpi_prefix_wannier'
  - MPI prefix for ComWann
  - default value: control['mpi_prefix']

In control

- 'sigma_mix_ratio'
  - impurity self-energy linear mixing ratio.
  - default value: 0.5
- 'max_iter_num_outer':
  - maximum iteration for the charge self-consistent loop.
  - default value: 50
- 'proj_win_min':
  - low-energy cutoff to renormalize the projectors
  - default value: wan_hmat[‘dis_win_min’]
- 'proj_win_max':
  - high-energy cutoff to renormalize the projectors
  - default value: wan_hmat[‘dis_win_max’]

In wan_hmat

- 'kgrid': [5,5,5],
  - crystal momentum grid for the wannier interpolation of LDA bandstructure
- 'froz_win_min': -15 eV,
  - lower boundary of the inner (frozen) window in eV
- 'froz_win_max': 10 eV,
  - upper boundary of the inner (frozen) window in eV
- 'dis_win_min':
  - lower boundary of the outer (disentanglement) window in eV.
  - defaule value: froz_win_min
- 'dis_win_max':
  - upper boundary of the outer (disentanglement) window in eV.
  - defaule value: froz_win_max +40.0
- 'num_iter':
  - the number of minization step for the wannierization process. (gauge dependent part of total spreading)
  - default value: 0
- 'dis_num_iter':
  - the number of minization step for the disentanglement process. (gauge independent part of total spreading)
  - defaule value: 100

In imp

- **'temperature': 900**

  - simulation temperature in K

- for each distinct impurity problem indexed by the value in control["impurity_problem_equivalence"]

  - 'impurity_matrix': [[1]],

    --equivalence of the matrix element of the fermionic Weiss field and impurity self-energy. Starting from "1"

    --if these values are the same, the values of the elements will be assumed to be identical.

    --if the element in the matrix is zero, then it will not be sampled by the impurity solver.

    --each column and row corresponds to the Wannier orbitals in the following order.

    --If control['spin_orbit']==False, "m" is sorted in ascending order.

    To illustrate for "d" orbitals, in this order: |xy>,|yz>,|z^2>,|xz>, |x^2-y^2>

    --if control['spin_orbit']==True, the most rapidly changing index is "m" and the next one is "i". They are sorted in ascending order,

    To illustrate for "f" orbitals, in this order: |3,-0.5, -2.5>,|3,-0.5, -1.5>,|3,-0.5, -0.5>,
    |3,-0.5, 0.5>,|3,-0.5, 1.5>,|3,-0.5, 2.5>, |3,0.5, -3.5>,|3,0.5, -2.5>,|3,0.5, -1.5>,
    |3,0.5, -0.5>,|3,0.5, 0.5>,|3,0.5, 1.5>,|3,0.5, 2.5>, |3,0.5, 3.5>,

In imp

- 'Coulomb': ‘full’,
  --'full' or 'ising' are available. We construct Coulomb matrix in the following way.

$$U_{m_1,m_2,m_3,m_4} = \sum_{k=0}^{2l,even} \frac{4\pi}{2k+1} F_l^k \sum_{q=-k}^{k} \langle Y_l^{m_1} | Y_k^q Y_l^{m_4} \rangle \langle Y_l^{m_2} Y_k^q | Y_l^{m_3} \rangle$$

  --If 'full', no additional approximation is considered.
  --If 'ising', only U_{abba} or U_{abab} are non-zero.
- 'thermalization_time': 1,
  » wall time for the thermalization in minutes
- 'measurement_time’: 2,
  » wall time for the measurement in minutes
- 'green_cutoff’: 20,
  » cutoff-energy in eV to sample green’s function and self-energy.
  » values beyond this energy will be provided by analytical equations.
- 'susceptibility_cutoff':
  » cutoff-energy to sample susceptibility.
  » Default value: 300 eV

Output directory

- in lqsgw_dmft directory

```
analysis
band
cmd.log                 : command log file
comdmft.ini             : input file
convergence.log         : convergence log
coulomb                 : directory for the bosonic Weiss field within cRPA
dc                      : directory for the double counting calculation
delta.dat               : hybridization function
impurity                : directory for the quantum impurity problem
lowh                    : directory for fermionic Weiss field calculation
sig.dat                 : impurity self-energy
sig_dc.dat              : double counting self-energy
sig_dc_hf.dat           : HF contribution in double counting self-energy
trans_basis.dat
u_slater.dat            : bosonic Weiss field within cRPA
v_slater.dat            : local screened Coulomb interaction
w_slater.dat            : local bare Coulomb interaction
wannier                 : directory for Wannier function calculation
```

convergence.log

step	i_imp	causality	static_f0	w_sp_min	w_sp_max	mu	std_sig	n_imp	histo_1	histo_2	ctqmc_sign
wannier				2.47087035	3.9065202						
coulomb_1			2.48540688534606								
dc_1		good									
delta	1	good				0.009788133749					
impurity_1	1	good					0.2237649690574862	0.5721617043	48.63501107725202	63.61860388300722	1
delta	2	good				0.015149694009					
impurity_1	2	good					0.11295662631020237	0.5756367493	48.72429348779647	64.82274549948158	1
delta	3	good				0.020317629267					
impurity_1	3	good					0.05509831723450446	0.5800384964	48.91087647734551	65.05737585460773	1
delta	4	good				0.023045781482					
impurity_1	4	good					0.02796692374404145	0.5759111688	48.480377296698926	66.23955144614358	1
delta	5	good				0.022763114314					

- keeping track of convergence of some quantities at each iteration
- causality: causality of hybridization function / self-energy
- static_f0: static value of bosonic Weiss field
- w_sp_min: minimum spreading of the Wannier functions
- w_sp_max: maximum spreading of the Wannier functions
- mu: LDA+DMFT chemical potential w.r.t. LDA chemical potential
- std_sig:

$$\sqrt{\frac{\sum_i (\Sigma_i^j(i\omega_n) - \Sigma_i^{j-1}(i\omega_n))^2}{n_\omega n_{orb}}}$$

- n_imp: occupation in the impurity orbitals
- histo_1: the first moment of the perturbation order histogram
- histo_2: the second moment of the perturbation order histogram
- ctqmc_sign: CTQMC sign

Dynamical U

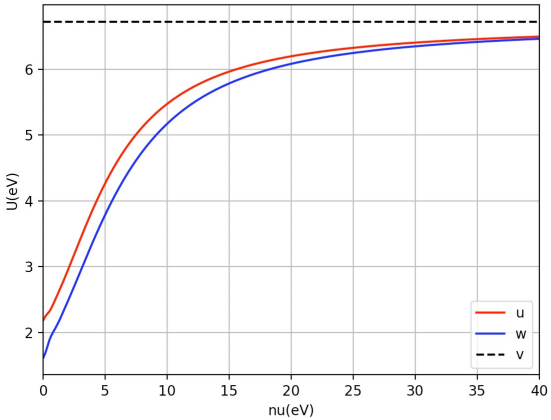
```
      # nu(eV)        1:f0(eV)
0.000000000000  2.178325249978
0.162432849384  2.237485235575
0.324865698768  2.278462848603
0.487298548153  2.312986665643
0.649731397537  2.363690070133
0.812164246921  2.427157767991
0.974597096305  2.494456594169
1.137029945690  2.563385246819
1.299462795074  2.633348723611
1.461895644458  2.703308925822
1.624328493842  2.774400841373
1.786761343226  2.847649955031
1.949194192611  2.922569601886
2.111627041995  2.998565106935
2.274059891379  3.075089392286
```

- Bare Coulomb interaction in v_slater.dat

```
   # 1:f0(eV)
6.725307308007
```

Dynamical U

- Let's plot it
  cd analysis
  python u_omega.py



- At high frequency, U and W_{loc} converge to V_{loc}
- U is larger than W but smaller than V

Impurity self-energy

- “sig.dat”
  » Real and imaginary part of impurity self-energy

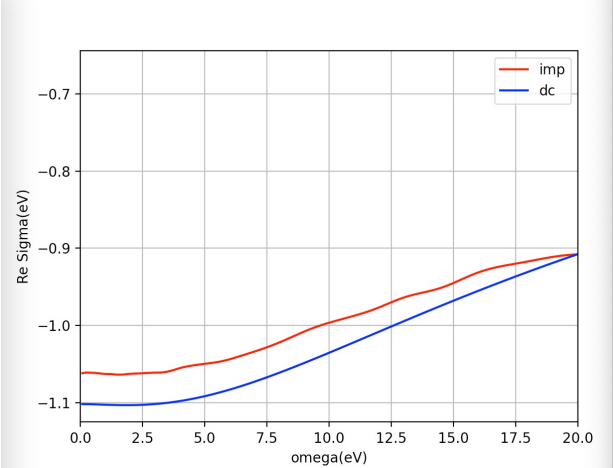
```
   # omega(eV)     Re Sig_{1,1}(eV)     Im Sig_{1,1}(eV)
0.081216424692      1.501800431626      -0.012268251317
0.243649274076      1.501588177463      -0.034063609310
0.406082123461      1.501601337665      -0.054366239901
0.568514972845      1.501110822638      -0.072508424919
0.730947822229      1.500027409077      -0.089041707011
0.893380671613      1.499250948871      -0.104164319516
1.055813520997      1.498605056075      -0.118189374120
1.218246370382      1.498588018057      -0.130392007481
1.380679219766      1.498537768366      -0.141387853780
1.543112069150      1.498168137125      -0.151863364836
1.705544918534      1.498330599474      -0.161486104294
```

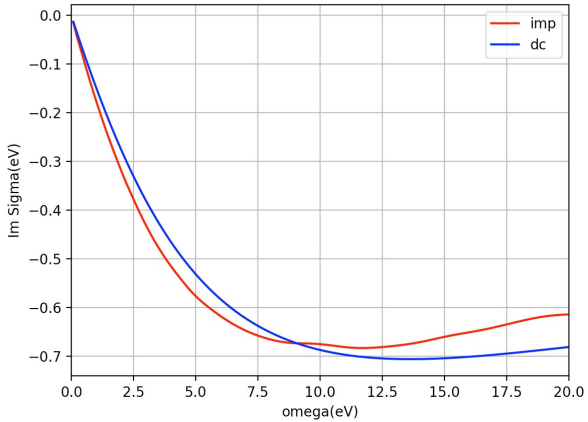
- Let’s plot it

  python sig.py

Local-GW impurity self-energy

For the visualization, I used data at “/home/max/codes/ComsuiteV2/tutorials_converged”





- No divergent self-energy near Fermi-level.
- Up to 5eV, the impurity self-energy shows linear energy dependence.
- smaller z factor within DMFT than GW

Hybridization function

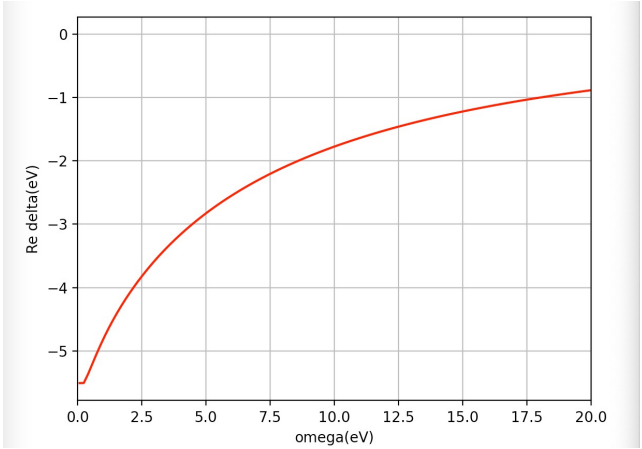
- “delta.dat”
  » Real and imaginary part of hybridization function

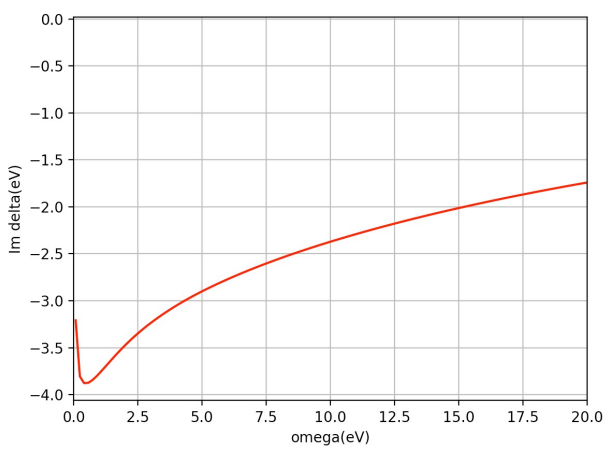
```
0.081216424692     -4.077402095983     -2.828521433978
0.243649274076     -4.006144819843     -2.966827164573
0.406082123461     -3.991149773784     -2.953881842766
0.568514972845     -3.968126243833     -2.926582297915
0.730947822229     -3.938154963765     -2.892171692592
0.893380671613     -3.903350147782     -2.857399777300
1.055813520997     -3.864792500134     -2.824214718055
1.218246370382     -3.823426294363     -2.793440776542
1.380679219766     -3.779812922422     -2.764457834912
1.543112069150     -3.734606699404     -2.736752971151
1.705544918534     -3.688584198508     -2.710484210705
1.867977767919     -3.642016615893     -2.685508905212
2.030410617303     -3.595109067127     -2.661525905159
```

- Let’s plot it

  python delta.py

Hybridization function

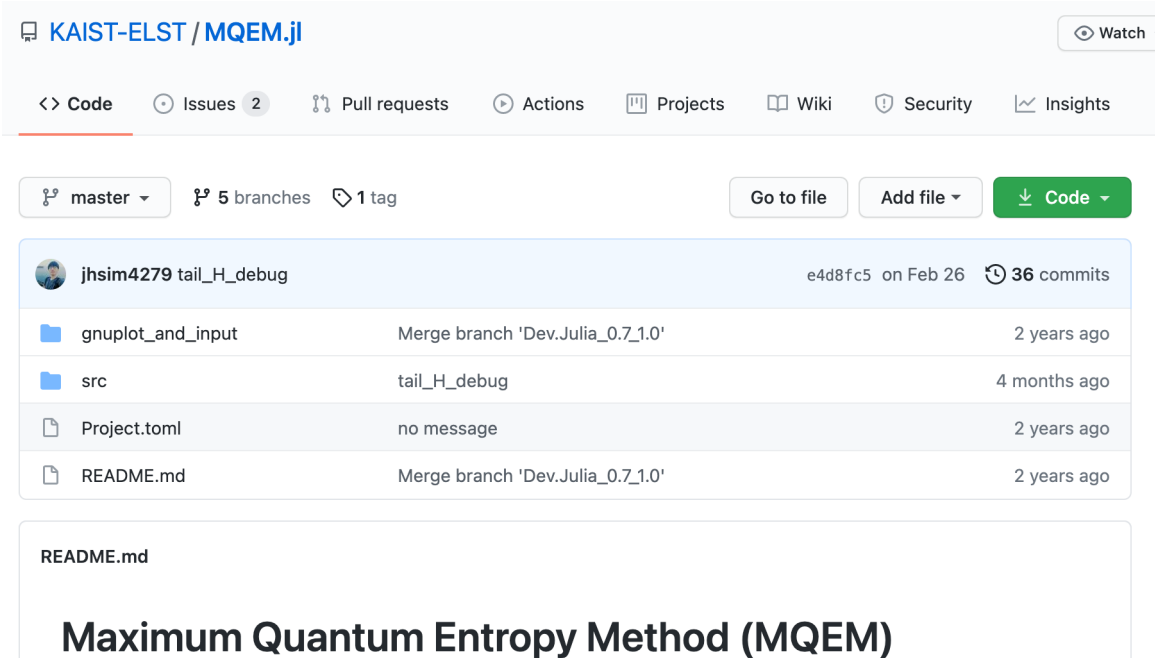




- It shows metallic behavior (imaginary part of delta is nonzero at zero frequency)

Analytical continuation

We will use the maximum entropy (maxent) method for the analytical continuation. For the purposes of this tutorial, we will use MQEM package (https://github.com/KAIST-ELST/MQEM.jl)



Analytical continuation

- To run the maxent code, create the maxent directory and then move to it

  $ cd ../

  $ mkdir maxent

  $ cd maxent

- To run the maxent code (~10 minutes)

  $ COMSUITE_BIN/mqem_wrapper ../sig.dat

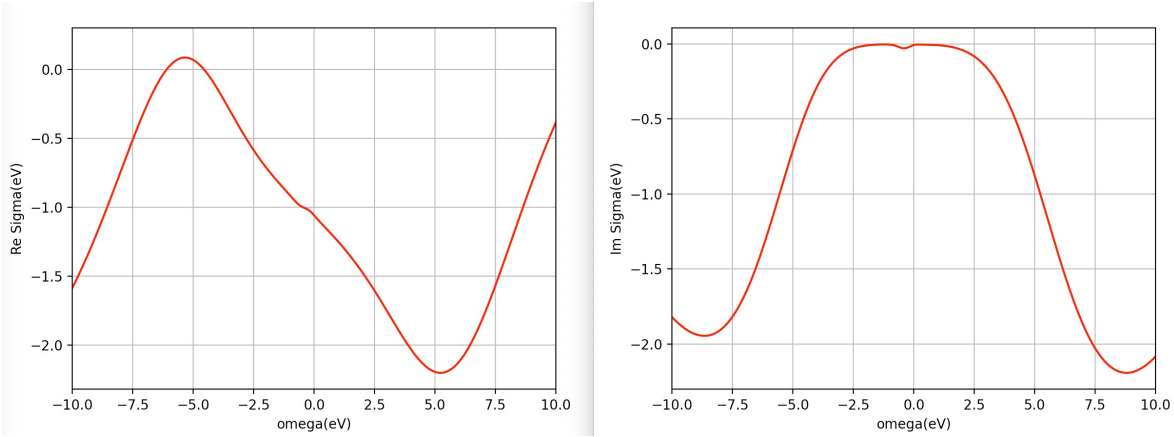
  => We will skip this step due to time limit . We will use the self-energy file I provided instead (located at /home/max/codes/ComsuiteV2/Compiled_ComsuiteCode/tutorials_converged/lqsgw_dmft/maxent/sig_realaxis.dat)

  You may try it as a homework:)

Analytical continuation

Let's plot it

```
cd ../analysis
python ./sig_realaxis.py
```



- Linear self-energy within $\mathrm{E_F} \pm 5\mathrm{eV}$
- Small pole near zero energy

LQSGW+DMFT spectral function calculation

- Go to the spectral function directory
  cd ../realaxis

- Check input files
  ls –al

`comdmft.ini`	Bandstructure calculation input file

- Run Comsuite
  $COMSUITE_BIN/comdmft.py

comdmft.ini

```
control ={
        'method':'spectral',
        'mpi_prefix':'mpirun -np 2',
        }

postprocessing={
        'comsuite_dir':'../',
        'self energy':'/home/max/codes/ComsuiteV2/tutorials_converged/lqsgw_dmft/maxent/sig_realaxis.dat',
        'broadening': 0.01,
        'kpoints': './kpoints'
        }
```

- in control
  - method: ‘spectral’
    postprocessing calculation. Choose among “band”, “dos”, and “spectral”

  - mpi_prefix: ‘mpirun –np 2’
    MPI prefix used for bandstructure calculation.

- in postprocessing
  - comsuite_dir: ‘../’
    The directory where comsuite calculation has been done.
  - self-energy: ‘/home/max/codes/ComsuiteV2/tutorials_converged/lqsgw_dmft/maxent/sig_realaxis.dat’
    real-axis self-energy file.
  - broadening: ‘0.01’
    broadening for the mean-field band
  - kpoints: ‘./kpoints’
    k-path file. If not provided, comsuite follow the path defined in Ref[1]

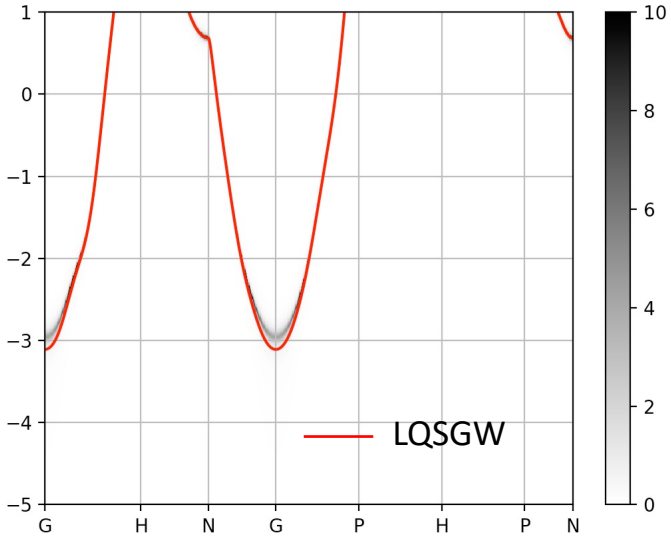
[1] W. Setyawan and S. Curtarolo, Computational Materials Science 49, 299 (2010).

LDA+DMFT bandstructure

Let's plot it

cd ../analysis

python band_spectra.py



LDA	mBJ	B3LYP	eDMFT	G_0W_0	Expt
3.30	3.29	4.09	2.84	3.15	2.65

[1] S. Mandal, K. Haule, K. M. Rabe, and D. Vanderbilt, ArXiv:2101.03262 [Cond-Mat] (2021).