From DFT+DMFT to LQSGW+DMFT and Full GW+EDMFT: application to materials.

Sangkook Choi

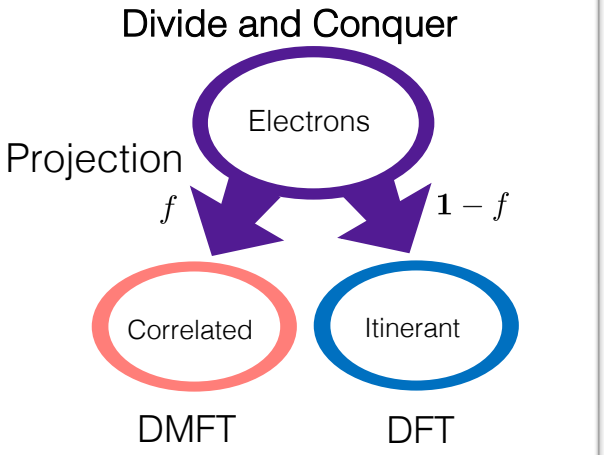
sachoi@bnl.gov
https://sites.google.com/view/sangkookchoi

Outline

1. Successes and challenges of DFT+DMFT
2. Introduction of GW+EDMFT
3. Approximation to GW+EDMFT: multitier scheme
4. Approximation to GW+EDMFT: partial self-consistency scheme
5. Full GW+EDMFT
6. Examples: $SrVO_3$, $LaNiO_2$
7. Hands-on (LQSGW+DMFT on Na)

1. Successes and challenges of DFT+DMFT

DFT + DMFT is a work horse methodology for correlated quantum materials



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

$$\mathbb{P}(A) = f^\dagger A f = \widetilde{A}_{cc}$$

$$\mathbb{E}(\widetilde{A}_{cc}) = f \widetilde{A}_{cc} f^\dagger = \begin{pmatrix} \widetilde{A}_{cc} & 0 \\ 0 & 0 \end{pmatrix}$$

$$G(\mathbf{r},\mathbf{r},\mathbf{k},\tau=0^+) \rightarrow \rho(\mathbf{r}) \rightarrow v_H + v_{xc}$$

$$G(\mathbf{k},i\omega_n) = \left(G_0(\mathbf{k},i\omega_n)^{-1} - v_{Hxc} - \mathbb{E}\left(\widetilde{\Sigma}_{imp}(i\omega_n) - \widetilde{\Sigma}_{DC} \right) \right)^{-1}$$

$$\widetilde{G}_{loc}(i\omega_n) = \mathbb{P}(G(\mathbf{k},i\omega_n))$$

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

$$S_{eff}(\widetilde{U}) \rightarrow \widetilde{G}_{imp}$$

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

$\widetilde{U}$ and $\widetilde{\Sigma}_{DC}$ should be determined by the choice of the projectors ($\mathbb{P}, \mathbb{E}$)

[1] V. I. Anisimov, A. I. Poteryaev, M. A. Korotin, A. O. Anokhin, and G. Kotliar, J. Phys.: Condens. Matter 9, 7359 (1997). [2] A. I. Lichtenstein and M. I. Katsnelson, Phys. Rev. B 57, 6884 (1998). [3] G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. 78, 865 (2006). [4] G. Kotliar and S. Y. Savrasov, arXiv:Cond-Mat/0208241 (2002). [5] R. M. Martin, L. Reining, and D. M. Ceperley, Interacting Electrons (Cambridge University Press, 2016). [6] R. Chitra and G. Kotliar, Phys. Rev. B 63, 115110 (2001). [7] S. Y. Savrasov and G. Kotliar, Spectral Density Functionals for Electronic Structure Calculations, Phys. Rev. B 69, 245101 (2004).

Numerous implementation (incomplete list)

DFT + embedded DMFT Functional*

Developed by Kristjan Haule at Rutgers University, ©Copyright 2007-2020.

TRIQS/DFTTools: A TRIQS application for *ab initio* calculations of correlated materials ☆

DCore

DCore = Integrated **D**MFT software for **Cor**related **e**lectrons.

RSPt

RSPt is a code for electronic structure calculations and its acronym stands for Relativistic Spin Polarized toolkit. RSPt offers a robust and flexible set of tools to calculate total energies, magnetic moments, band structures, Fermi surfaces and densities of states for all elements, and combinations thereof, over a wide range of volumes and structures.

DMFTwDFT: An open-source code combining Dynamical Mean Field Theory with various density functional theory packages☆,☆☆

Vijay Singh [a,b,*], Uthpala Herath [b], Benny Wah [a], Xingyu Liao [a], Aldo H. Romero [b], Hyowon Park [a]

ONETEP + TOSCAM: Uniting Dynamical Mean Field Theory and Linear-Scaling Density Functional Theory

Edward B. Linscott, Daniel J. Cole, Nicholas D. M. Hine, Michael C. Payne, and Cédric Weber*

Cite This: *J. Chem. Theory Comput.* 2020, 16, 4899–4911 Read Online

abinit

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:

QUESTAAL

Quasiparticle Electronic Structure and Augmented LMTOs

AMULET

Advanced Materials simULation Ekaterinburg's Toolbox

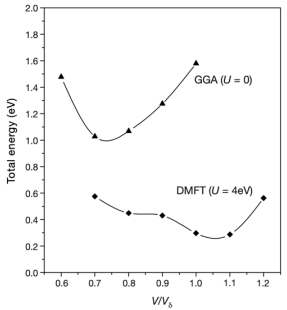
HOME FEATURES DOCUMENTA

AMULET

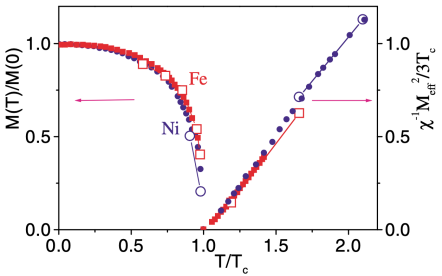
The AMULET is a free software for scientific and/or educational purposes and it is distributed under FreeBSD License. To description of a planned research has to be submitted to AMULET Developers Team. In the description one needs to indi and explain shortly why conventional band structure methods do not work for your problems/compounds and what k expecting from DMFT. The source code will be e-mailed to you if it is suitable for your study.

Numerous successes of DFT+DMFT (very incomplete list)

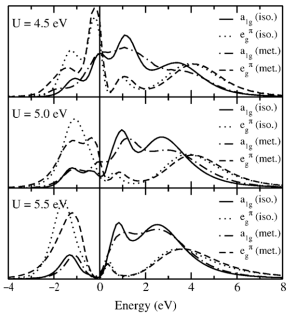
Volume collapse in Pu



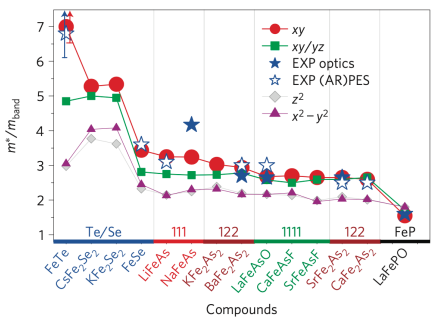
Finite T ferromagnetism



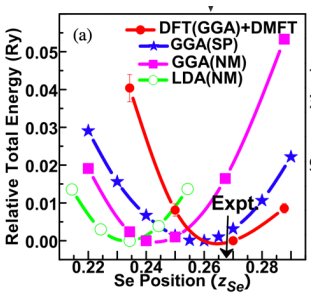
MIT in V2O3



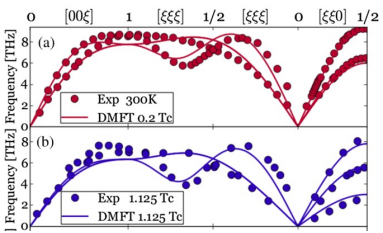
Hund metal physics



Structure prediction (FeSe)



Phonon



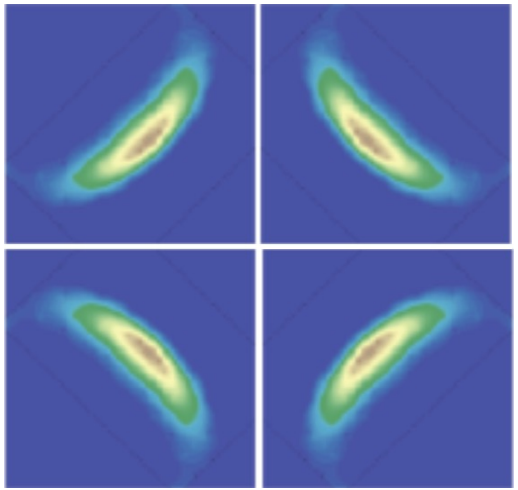
[1] S. Y. Savrasov, G. Kotliar, and E. Abrahams, Nature 410, 793 (2001). [2] A. I. Lichtenstein, M. I. Katsnelson, and G. Kotliar, Phys. Rev. Lett. 87, 067205 (2001). [3]K. Held, G. Keller, V. Eyert, D. Vollhardt, and V. I. Anisimov, Phys. Rev. Lett. 86, 5345 (2001).[4] Zhiping. P. Yin, K. Haule, and G. Kotliar, Nature Materials 10, 932 (2011). [5] Q. Han, T. Birol, and K. Haule, Phys. Rev. Lett. 120, 187203 (2018).[6] K. Haule, J. Phys. Soc. Jpn. 87, 041005 (2018).

What if nonlocal correlation is essential?

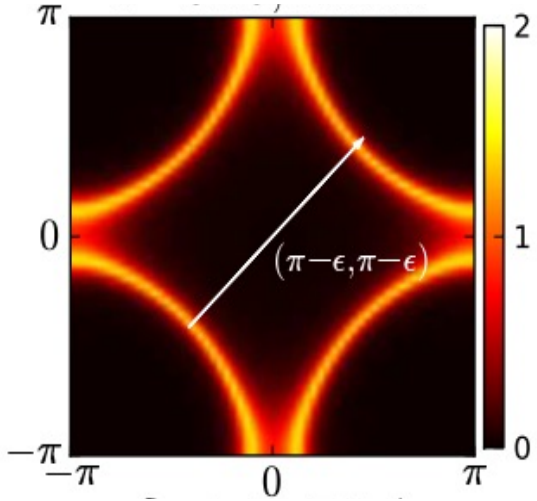
DFT + single-site DMFT: local (no k dependence) and dynamical (frequency-dependent) self-energy

$$\widetilde{\Sigma}(\mathbf{k}, i\omega_n) \simeq \widetilde{\Sigma}(i\omega_n)$$

Pseudo-gap in Cuprates



Single-site DMFT, Hubbard model

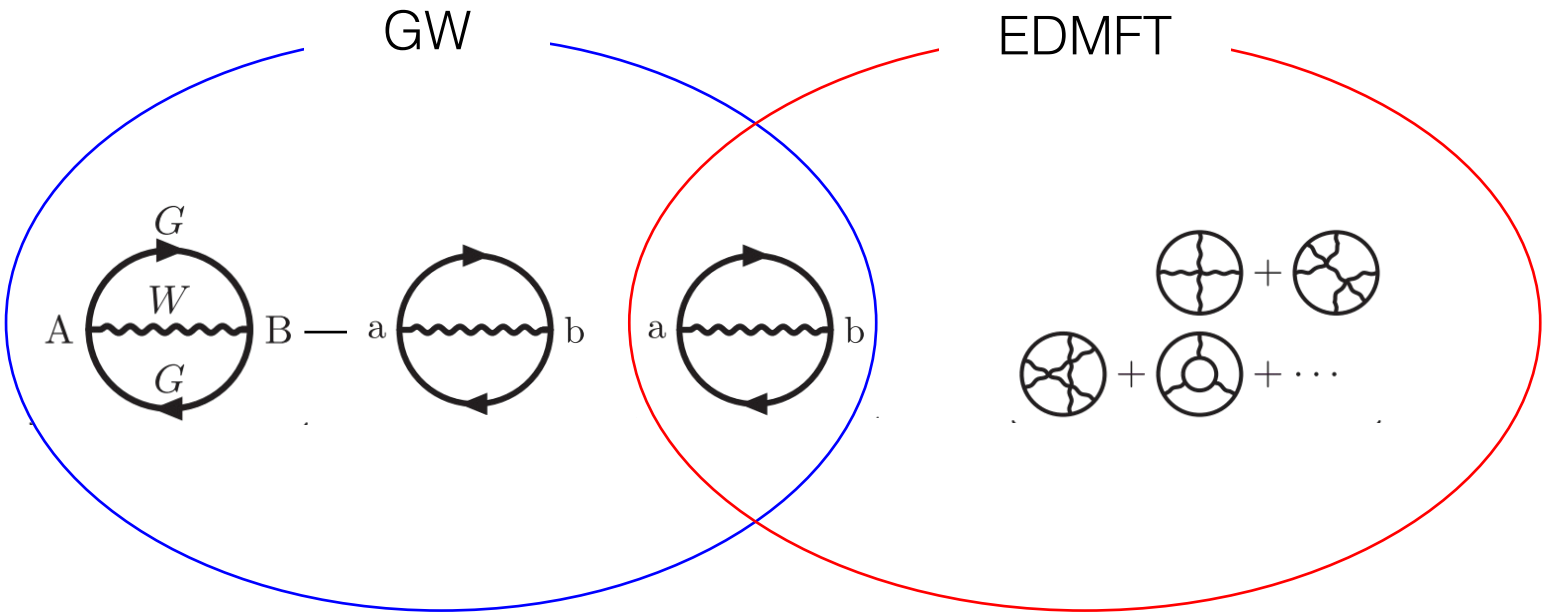


For the system where non-local (k-dependent) correlation is important, we need a tool beyond DFT + single site DMFT

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

2. Introduction of GW+EDMFT

GW+EDMFT, a diagrammatic extension of DMFT



- local correlation within EDMFT and non-local correlation within GW
- itinerant states within GW
- correlated states within GW+EDMFT

[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).

Functional Approach to GW+EDMFT

- Free energy Functional

$$G_0 = (i\omega_n - T - V_n)^{-1}$$

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

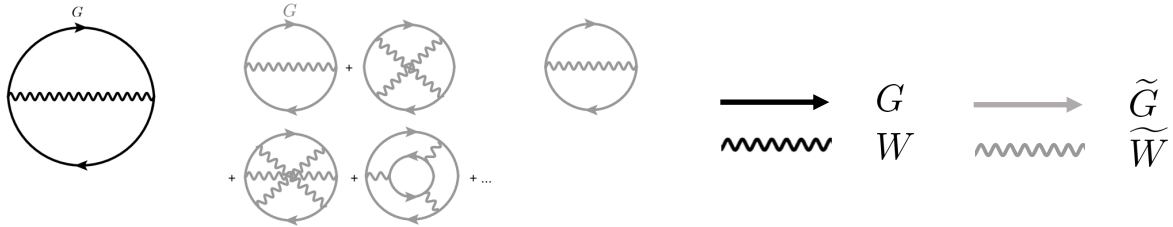
$$= -\mathfrak{Tr}\left[\log(-G_0^{-1})\right] + \mathfrak{Tr}\left[\log(1 - G_0\Sigma)\right] - \mathfrak{Tr}\left[G\Sigma\right] + \Phi_H[G] + \frac{1}{2}\mathfrak{Tr}\left[\log(1 - vP)\right] + \frac{1}{2}\mathfrak{Tr}\left[PW\right] + \Psi_{GW+EDMFT}[G, W]$$

Free energy functional due to G

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

e-e interaction

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



[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).

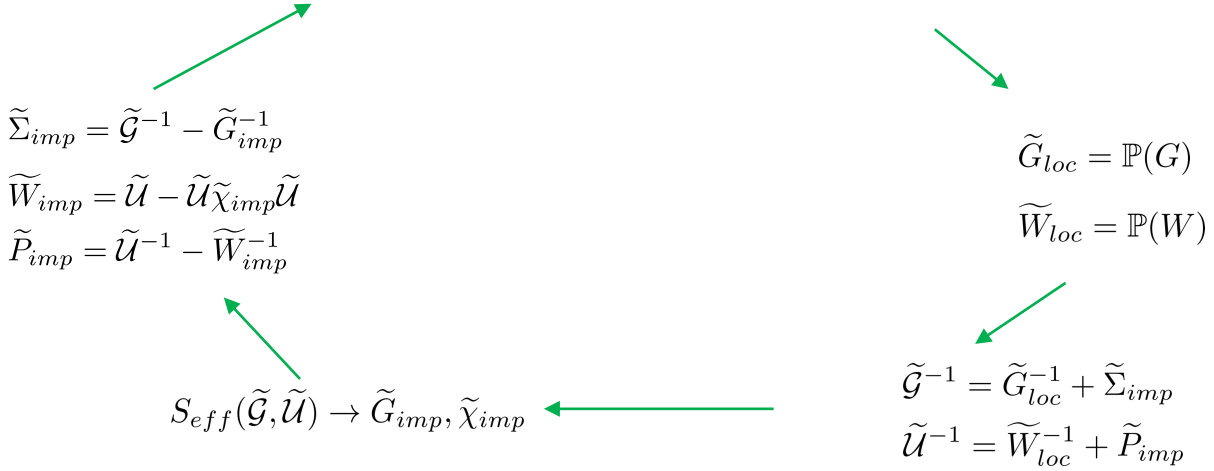
GW+EDMFT loop

Stationary condition

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

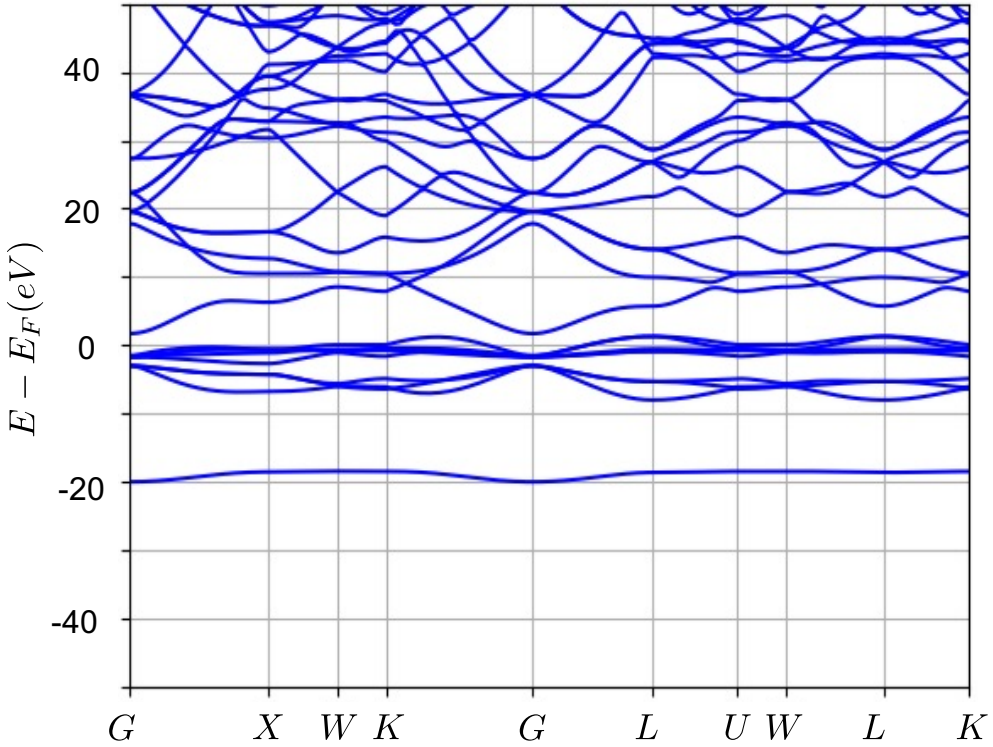
$$\frac{\delta\Gamma_{GW+EDMFT}}{\delta W} = 0 \rightarrow P = 2GG + \frac{\delta\Psi_{EDMFT}}{\delta W} - 2\mathbb{E}(\widetilde{G}\widetilde{G}) \qquad \frac{\delta\Gamma_{GW+EDMFT}}{\delta P} = 0 \rightarrow W^{-1} = v^{-1} - P$$

$$G^{-1}(\mathbf{k}, i\omega_n) = G_0^{-1}(\mathbf{k}, i\omega_n) - \Sigma_H - \Sigma_{GW}(\mathbf{k}, i\omega_n) - \mathbb{E}\left(\widetilde{\Sigma}_{imp}(i\omega_n) - \widetilde{\Sigma}_{DC}(i\omega_n)\right)$$
$$W^{-1}(\mathbf{k}, i\nu_n) = V^{-1}(\mathbf{k}) - P_{GW}(\mathbf{k}, i\omega_n) - \mathbb{E}\left(\widetilde{P}_{imp}(i\omega_n) - \widetilde{P}_{DC}(i\omega_n)\right)$$



3. Approximation to GW+EDMFT: multitier scheme

Simplification I: Multitier scheme
GW+EDMFT in the intermediate space



C — F

C: correlated subspace
I: intermediate subspace (5-8 orbital)
F: full space

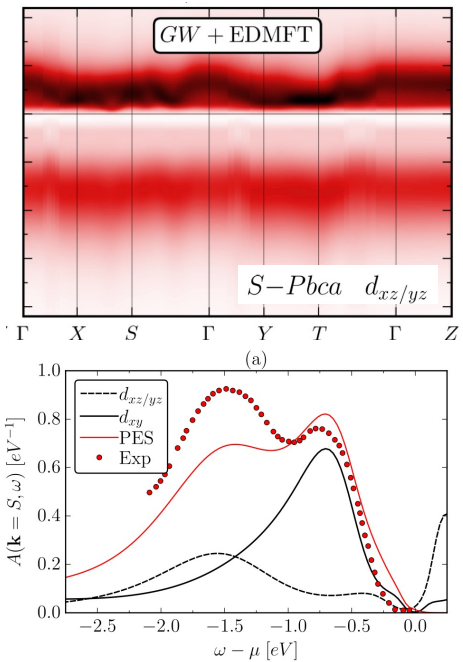
C=Tier-I
I=Tier-I ⊕ Tier-II
F= Tier-I ⊕ Tier-II ⊕ Tier-III

Tier-I: GW+EDMFT
Tier-II: GW
Tier-III: one-shot GW, not GW

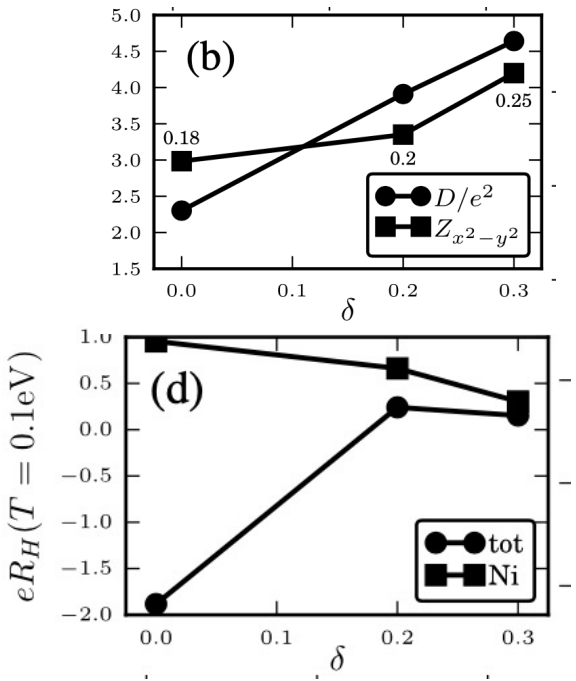
[1] F. Nilsson, L. Boehnke, P. Werner, and F. Aryasetiawan, Phys. Rev. Materials 1, 043803 (2017).

Application of multitier GW+EDMFT to real materials

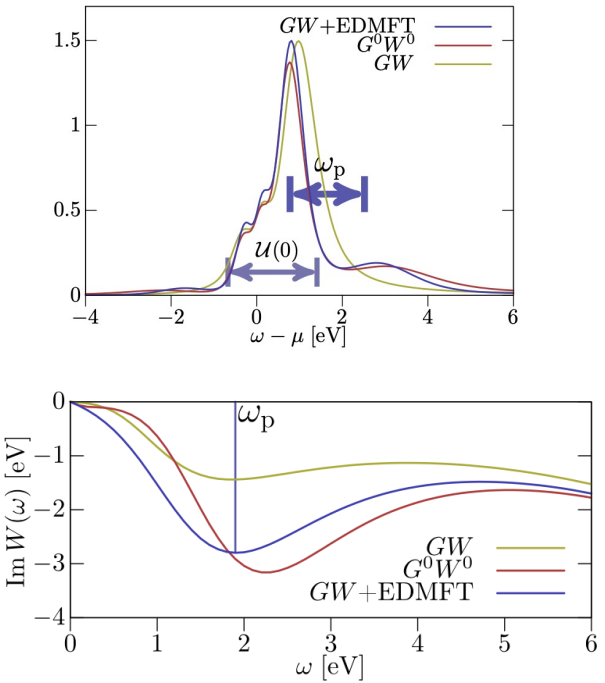
MIT in Ca_2RuO_4



Multiorbital nature of infinite-layer nickelates



Plasmon satellites in SrVO3



[1] F. Petocchi, V. Christiansson, and P. Werner, Phys. Rev. B 104, 195146 (2021). [2] F. Petocchi, V. Christiansson, F. Nilsson, F. Aryasetiawan, and P. Werner, Phys. Rev. X 10, 041047 (2020). [3] F. Petocchi, F. Nilsson, F. Aryasetiawan, and P. Werner, Phys. Rev. Research 2, 013191 (2020). [4] L. Boehnke, F. Nilsson, F. Aryasetiawan, and P. Werner, Phys. Rev. B 94, 201106 (2016).

4. Approximation to GW+EDMFT: partial self-consistency scheme

Important ansatz for simplification II: separable self-energy

$$\widetilde{\Sigma}(\mathbf{k}, i\omega_n) \simeq \widetilde{\Sigma}^{non-local}(\mathbf{k}) + \widetilde{\Sigma}^{dyn}(i\omega_n) \rightarrow G^{-1}(\mathbf{k}, i\omega_n) = i\omega_n - \Big(\underbrace{H_0 + \mathbb{E}(\widetilde{\Sigma}^{non-local}(\mathbf{k}))}_{= H^{non-local}(\mathbf{k})} + \mathbb{E}(\widetilde{\Sigma}^{dyn}(i\omega_n)) \Big)$$

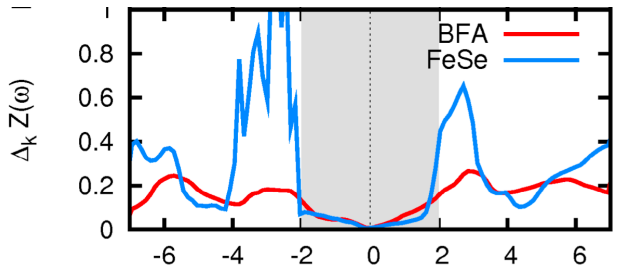
Validation on Fe-based superconductors quasiparticle bands

- Up to linear term in frequency

$$\widetilde{\Sigma}(\mathbf{k}, i\omega_n) \simeq \widetilde{\Sigma}(k, 0) + \left(1 - \widetilde{Z}^{-1}(\mathbf{k})\right) i\omega_n \qquad \widetilde{Z}^{-1}(\mathbf{k}) = 1 - \left. \frac{\partial \widetilde{\Sigma}(\mathbf{k}, i\omega_n)}{\partial (i\omega_n)} \right|_{i\omega_n = 0}$$

- If $\tilde{Z}(\mathbf{k})$ is k-independent, self-energy is separable

Within QSGW



1) $H^{non-local}(\mathbf{k}) = H^{LQSGW}(\mathbf{k})$
2) Determine Z(k) by fitting ARPES spectra of LiFeAs

	$Z_m(\Gamma)$	$Z_m(M)$
xy	0.21±0.01	0.18±0.01
xz/yz	0.38±0.01	0.30±0.04

[1] J. M. Tomczak, M. van Schilfgaarde, and G. Kotliar, Phys. Rev. Lett. 109, 237010 (2012). [2] J. M. Tomczak, J. Phys.: Conf. Ser. 592, 012055 (2015). [3] M. Kim, H. Miao, S. Choi, M. Zingl, A. Georges, and G. Kotliar, Phys. Rev. B 103, 155107 (2021).

Simplification II: partial self-consistency scheme

$$G^{-1}(\mathbf{k}, i\omega_n) = G_0^{-1}(\mathbf{k}, i\omega_n) - \Sigma_{GW}(\mathbf{k}, i\omega_n) - \mathbb{E}\left(\widetilde{\Sigma}_{imp}(i\omega_n) - \widetilde{\Sigma}_{DC}(i\omega_n)\right)$$

$$W^{-1}(\mathbf{k}, i\nu_n) = V^{-1}(\mathbf{k}) - P_{GW}(\mathbf{k}, i\omega_n) - \mathbb{E}\left(\widetilde{P}_{imp}(i\omega_n) - \widetilde{P}_{DC}(i\omega_n)\right)$$

$$G^{-1}(\mathbf{k}, i\omega_n) = i\omega_n - \left(H^{non-local}(\mathbf{k}) + \mathbb{E}(\widetilde{\Sigma}^{imp}(i\omega_n))\right)$$

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

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

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

$$\widetilde{G}_{loc} = \mathbb{P}(G)$$

$$\widetilde{W}_{loc} = \mathbb{P}(W)$$

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

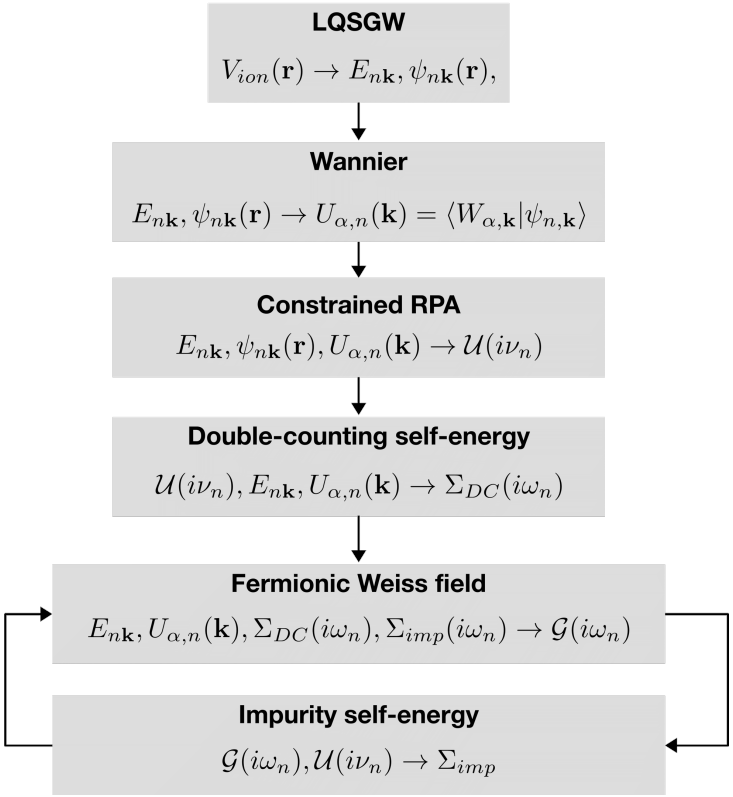
$$\widetilde{\mathcal{U}}^{-1} = \widetilde{W}_{loc}^{-1} + \widetilde{P}_{imp}$$

$$S_{eff}(\widetilde{\mathcal{G}}, \widetilde{\mathcal{U}}) \rightarrow \widetilde{G}_{imp}, \widetilde{\chi}_{imp}$$

- 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: $H^{non-local}$ is fixed
- For $H^{non-local}$: one-shot GW [7,8], Screened Exchange [3], QSGW [4,5] and LQSGW [2], non-local QSGW and LQSGW [6]
- One example is LQSGW+DMFT

[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).

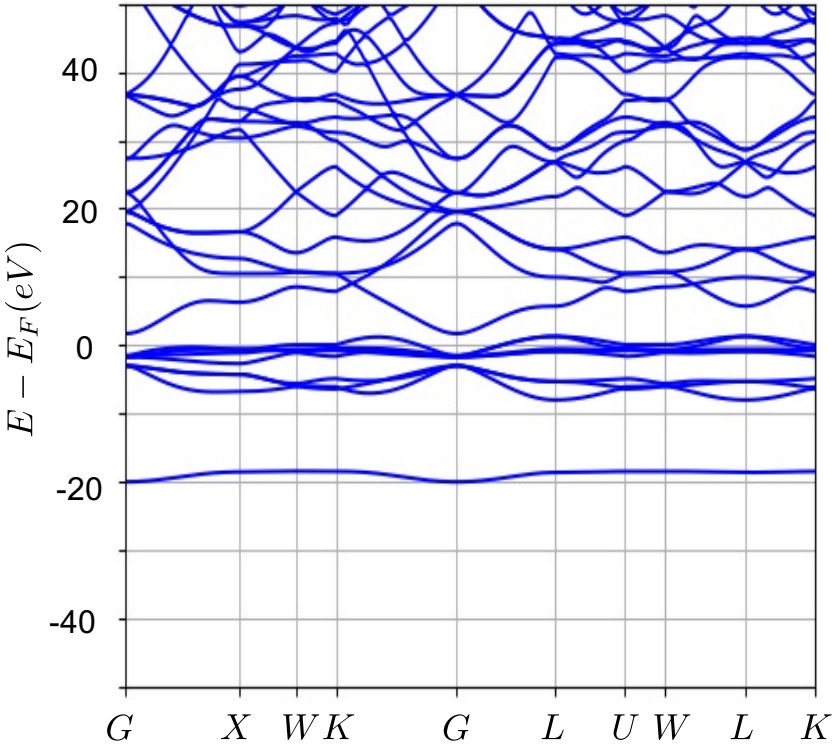
Ab initio LQSGW+DMFT



- A simplification of full GW+EDMFT
- One-shot DMFT correction to ab initio LQSGW
- Interaction tensor and double-counting energy are calculated (within cRPA and local-GW)
- a parameter-free method
- Validation on charge transfer insulators, Fe-based superconductors, and narrow-gap correlated insulators

[1] J. M. Tomczak, J. Phys.: Conf. Ser. 592, 012055 (2015).
[2] **S. Choi**, A. Kutepov, K. Haule, M. van Schilfgaarde, and G. Kotliar, npj Quantum Materials 1, 16001 (2016).
[3] **S. Choi**, P. Semon, B. Kang, A. Kutepov, and G. Kotliar,, Computer Physics Communications 244, 277 (2019).

Hilbert space and its subspaces for LQSGW+DMFT

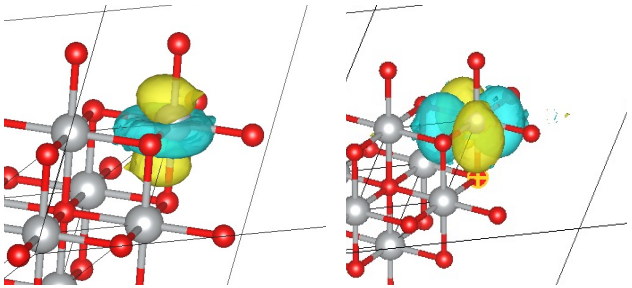


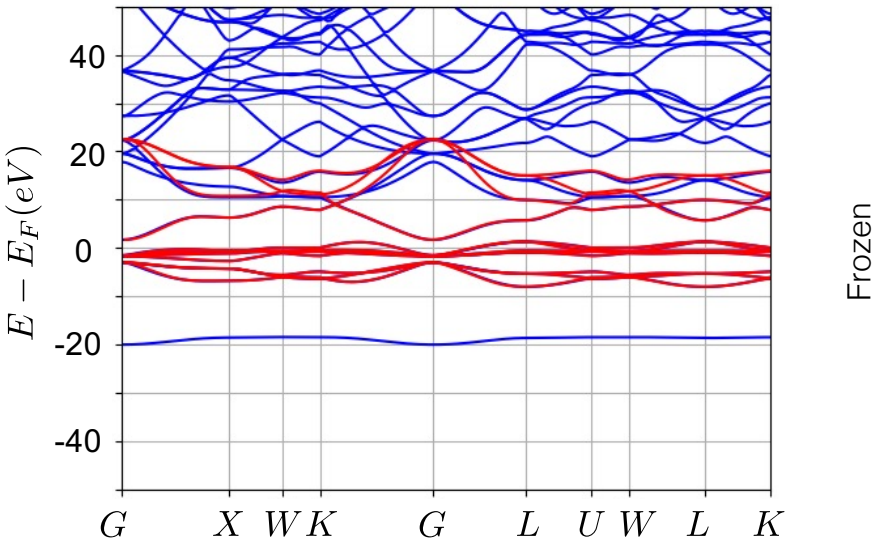
C ⊂ L ⊂ F

- F: full space
  - F= Tier-I ⊕ Tier-II ⊕ Tier-III
  - notation: $A\ (r, r')$
- L: Low-energy subspace defined by Wannier functions spanning an energy window $(\mathbf{E_F} \pm 10eV)$
  - L=Tier-I ⊕ Tier-II
  - notation: $\bar{A}_{ij}$, i,j=> Wannier functions
- C: correlated subspace
  - C=Tier-I
  - notation: $\tilde{A}_{ij}$, i,j=> Wannier functions

Tier-I: one-shot DMFT correction to LQSGW
Tier-II: LQSGW
Tier-III: LQSGW

Basis set in the low-energy space





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

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

- 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 in the inner (frozen) window $E_{n\mathbf{k}}$

- 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).

Nonlocal Hamiltonian 1: Linearized quasiparticle self-consistent GW (LQSGW)

$$Z^{-1}(\mathbf{k}^c) = 1 - \left.\frac{\partial \Sigma(\mathbf{k}^c, i\omega_n)}{\partial(i\omega_n)}\right|_{i\omega_n=0}$$

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

$$G^{QP}(\mathbf{k}^c, i\omega_n^c) = \left(i\omega_n^c - H^{QP}(\mathbf{k}^c)\right)^{-1}$$

$$W_{GW}(\mathbf{k}^c, i\omega_n^c) = \left(V^{-1}(\mathbf{k}^c) - P_{GW}(\mathbf{k}^c, i\omega_n^c)\right)^{-1}$$

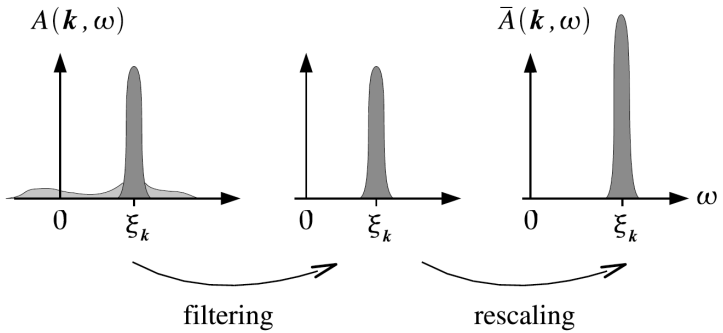
$$P_{GW}(\mathbf{k}^c, i\nu_n^c) = -\sum_{\mathbf{R}^c}\int_0^{\beta^c} d\tau G^{QP}(\mathbf{R}^c, \tau) \circ G^{QP}(-\mathbf{R}^c, -\tau) e^{-i(\mathbf{k}^c\cdot\mathbf{R}^c - \nu_n^c\tau)}$$

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

- The developer: Andrey Kutepov
- LAPW basis set
- space-time methods (to avoid convolution)
- calculation in a coarse Matsubara frequency grid (typical simulation temperature ~ 1000K)
- calculation in a coarse momentum space grid (e.g. NiO: 6×6×6)

[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 self-energy and quasiparticle Hamiltonian



$$
\begin{aligned}
G(i\omega_n) &= \frac{1}{i\omega_n - \varepsilon_0 - \Sigma(i\omega_n)} \qquad \text{filtering} \\
&\simeq \frac{1}{i\omega_n - \varepsilon_0 - \Sigma(0) - i\omega_n \frac{\partial \Sigma(i\omega_n)}{\partial(i\omega_n)}\Big|_{\omega=0}} \\
&= \frac{1}{i\omega_n \left(1 - \frac{\partial \Sigma(i\omega_n)}{\partial(i\omega_n)}\Big|_{\omega=0}\right) - \varepsilon_0 - \Sigma(0)} \\
&= \frac{Z}{i\omega_n - \sqrt{Z}\left(\varepsilon_0 + \Sigma(0)\right)\sqrt{Z}} \qquad \text{rescaling} \\
&\simeq \frac{1}{i\omega_n - \sqrt{Z}\left(\varepsilon_0 + \Sigma(0)\right)\sqrt{Z}}
\end{aligned}
$$

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

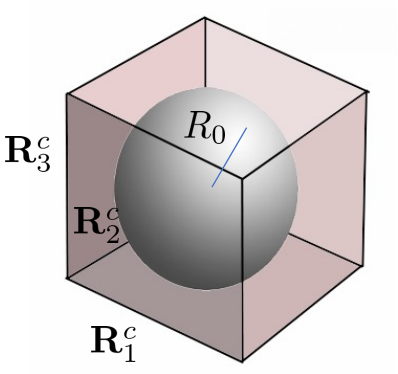
Quasiparticle Hamiltonian: $H_{QP} = \sqrt{Z}\left(\varepsilon_0 + \Sigma(0)\right)\sqrt{Z}$

Nonlocal Hamiltonian II: Wannier-interpolation of 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 (R_0), we can interpolate the bands at an arbitrary k point

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

$$\begin{aligned}
&H_{\tau,\tau'}(\mathbf{k}^f) \\
&= \sum_{\mathbf{R}^f} H_{\tau,\tau'}(\mathbf{R}^f) e^{i\mathbf{k}^f \cdot \mathbf{R}^f} \\
&= \sum_{|\mathbf{R}^f| \le R_0} H_{\tau,\tau'}(\mathbf{R}^f) e^{i\mathbf{k}^f \cdot \mathbf{R}^f} + \sum_{|\mathbf{R}^f| > R_0} H_{\tau,\tau'}(\mathbf{R}^f) e^{i\mathbf{k}^f \cdot \mathbf{R}^f} \\
&= \sum_{|\mathbf{R}^c| \le R_0} H_{\tau,\tau'}(\mathbf{R}^c) e^{i\mathbf{k}^f \cdot \mathbf{R}^c}
\end{aligned}$$



[1] N. Marzari, A. A. Mostofi, J. R. Yates, I. Souza, and D. Vanderbilt, Rev. Mod. Phys. 84, 1419 (2012).

Nonlocal Hamiltonian III: Nonlocal LQSGW

$$\widetilde{\Sigma}(\mathbf{k}, i\omega_n) \simeq \widetilde{\Sigma}^{non-local}(\mathbf{k}) + \widetilde{\Sigma}^{dyn}(i\omega_n) \rightarrow G^{-1}(\mathbf{k}, i\omega_n) = i\omega_n - \Big(\underbrace{H_0 + \mathbb{E}(\widetilde{\Sigma}^{non-local}(\mathbf{k}))}_{= H^{non-local}(\mathbf{k})} + \mathbb{E}(\widetilde{\Sigma}^{dyn}(i\omega_n)) \Big)$$

We choose nonlocal Hamiltonian by requiring that G is G_{LQSGW} when $\Sigma^{dyn} = \Sigma^{imp} = \Sigma^{DC}$

$$\begin{aligned}
G &\simeq \frac{1}{i\omega_n - \Big(H^{non-local}(\mathbf{k}) + \mathbb{E}(\Sigma^{DC}(\omega=0)) + \mathbb{E}((1-\widetilde{Z}_{DC}^{-1}) i\omega_n) \Big)} \\
&= \frac{1}{Z_{DC}^{-1} i\omega_n - (H^{non-local}(\mathbf{k}) + \mathbb{E}(\Sigma^{DC}(\omega=0)))} \\
&= \frac{Z_{DC}}{i\omega_n - \sqrt{Z_{DC}}\,(H^{non-local}(\mathbf{k}) + \mathbb{E}(\Sigma^{DC}(\omega=0)))\,\sqrt{Z_{DC}}} \\
&\simeq \frac{1}{i\omega_n - \sqrt{Z_{DC}}\,(H^{non-local}(\mathbf{k}) + \mathbb{E}(\Sigma^{DC}(\omega=0)))\,\sqrt{Z_{DC}}} \\
&= \frac{1}{i\omega_n - H^{LQSGW}(\mathbf{k})}
\end{aligned}$$

$$\widetilde{Z}_{DC}^{-1} = 1 - \left. \frac{\partial \widetilde{\Sigma}_{DC}(i\omega_n)}{\partial (i\omega_n)} \right|_{i\omega_n = 0}$$

$$Z_{DC}^{-1} = \mathbb{E}(\widetilde{Z}_{DC}^{-1}) + \Big(1 - \mathbb{E}(\widetilde{1})\Big)$$

$$H^{non-local}(\mathbf{k}) = Z_{DC}^{-1/2} H^{LQSGW}(\mathbf{k}) Z_{DC}^{-1/2} - \mathbb{E}(\Sigma^{DC}(\omega=0))$$

[1] J. M. Tomczak, J. Phys.: Conf. Ser. 592, 012055 (2015).

Bosonic Weiss Field: constrained random phase approximation

$$G^{-1}(\mathbf{k}, i\omega_n) = G_0^{-1}(\mathbf{k}, i\omega_n) - \Sigma_{GW}(\mathbf{k}, i\omega_n) - \mathbb{E}\left(\widetilde{\Sigma}_{imp}(i\omega_n) - \widetilde{\Sigma}_{DC}(i\omega_n)\right)$$

$$W^{-1}(\mathbf{k}, i\nu_n) = V^{-1}(\mathbf{k}) - P_{GW}(\mathbf{k}, i\omega_n) - \mathbb{E}\left(\widetilde{P}_{imp}(i\omega_n) - \widetilde{P}_{DC}(i\omega_n)\right)$$

$$G^{-1}(\mathbf{k}, i\omega_n) = i\omega_n - \left(H^{non-local}(\mathbf{k}) + \mathbb{E}(\Sigma^{loc}(i\omega_n))\right)$$

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

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

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

$$\widetilde{G}_{loc} = \mathbb{P}(G)$$

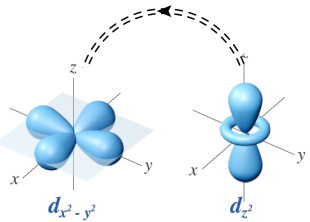
$$\widetilde{W}_{loc} = \mathbb{P}(W)$$

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

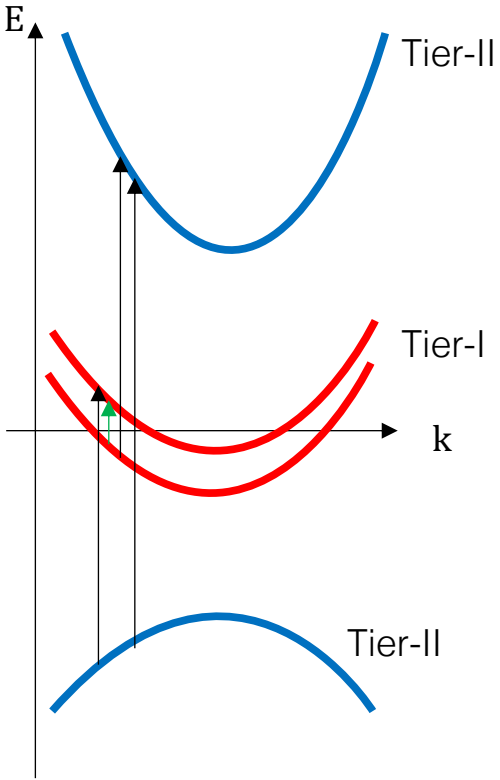
$$\widetilde{\mathcal{U}}^{-1} = \widetilde{W}_{loc}^{-1} + \widetilde{P}_{imp}$$

$$S_{eff}(\widetilde{\mathcal{G}}, \widetilde{\mathcal{U}}) \rightarrow \widetilde{G}_{imp}, \widetilde{\chi}_{imp}$$

- U: effective Coulomb interaction (bare with respect to the correlated orbitals but renormalized with respect to the rest)
- How can we calculate U within GW?



Bosonic Weiss Field: constrained random phase approximation



Partially screened Coulomb interaction by neglecting transition between correlated bands

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

$$P_{QP}^{low}(\mathbf{r},\mathbf{r}',\mathbf{k}^c,i\omega_n^c) = -N_s \sum_{\mathbf{k}^{\mathcal{C}}} \sum_{n}^{\substack{\text{unocc} \\ \text{in the space}}} \sum_{m}^{\substack{\text{occ} \\ \text{in the space}}} \psi_{n\mathbf{k}^{\mathcal{C}}}(\mathbf{r})\psi^*_{m\mathbf{k}^{\mathcal{C}}+\mathbf{k}^c}(\mathbf{r})\psi^*_{n\mathbf{k}^{\mathcal{C}}}(\mathbf{r}')\psi_{m\mathbf{k}^{\mathcal{C}}+\mathbf{k}}(\mathbf{r}')\frac{2(E^c_{n\mathbf{k}^{\mathcal{C}}} - E^c_{n\mathbf{k}^{\mathcal{C}}+\mathbf{k}^c})}{\omega_n^{c2} - (E^c_{n\mathbf{k}^{\mathcal{C}}} - E^c_{n\mathbf{k}^{\mathcal{C}}+\mathbf{k}^c})^2},$$

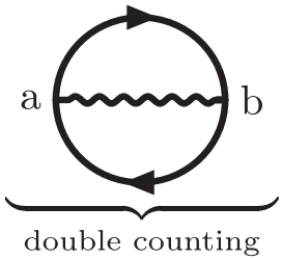
$$W_r^{-1}(\mathbf{k}^c,i\omega_n^c) = W^{-1}(\mathbf{k}^c,i\omega_n^c) + P_{QP}^{low}(\mathbf{k}^c,i\omega_n^c)$$

$$\widetilde{\mathcal{U}}_{ijkl}(i\omega_n^c) = \int d\mathbf{r}d\mathbf{r}' W^*_{\mathbf{R}^c=0,i}(\mathbf{r})W^*_{\mathbf{R}^c=0,j}(\mathbf{r}')W_r(\mathbf{r},\mathbf{r}',\mathbf{R}^c=0,i\omega_n^c)W_{\mathbf{R}^c=0,k}(\mathbf{r}')W_{\mathbf{R}^c=0,l}(\mathbf{r})$$

$$\widetilde{\mathcal{U}}_{ijkl}(i\omega_n^c) \rightarrow \widetilde{\mathcal{U}}_{ijkl}(i\omega_n^f)$$

[1] F. Aryasetiawan, M. Imada, A. Georges, G. Kotliar, S. Biermann, A.I. Lichtenstein, Phys. Rev. B 70 (19) (2004) 195104, [2] F. Aryasetiawan, K. Karlsson, O. Jepsen, U. Schönberger, Phys. Rev. B 74 (12) (2006) 125106 [3] T. Miyake, F. Aryasetiawan, Phys. Rev. B 77 (8) (2008) 085122, [4] P. Werner, M. Casula, T. Miyake, F. Aryasetiawan, A.J. Millis, S. Biermann, Nat. Phys. 8 (4) (2012) 331–337

Double counting self-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 self-consistent equation

$$G^{-1}(\mathbf{k}, i\omega_n) = i\omega_n - \left(H^{non-local}(\mathbf{k}) + \mathbb{E}(\widetilde{\Sigma}^{imp}(i\omega_n)) \right)$$

$$H^{non-local}(\mathbf{k}) = Z_{DC}^{-1/2} H^{LQSGW}(\mathbf{k}) Z_{DC}^{-1/2} - \mathbb{E}(\widetilde{\Sigma}^{DC}(\omega = 0))$$

$$\widetilde{Z}_{DC}^{-1} = 1 - \left. \frac{\partial \widetilde{\Sigma}_{DC}(i\omega_n)}{\partial (i\omega_n)} \right|_{i\omega_n = 0}$$

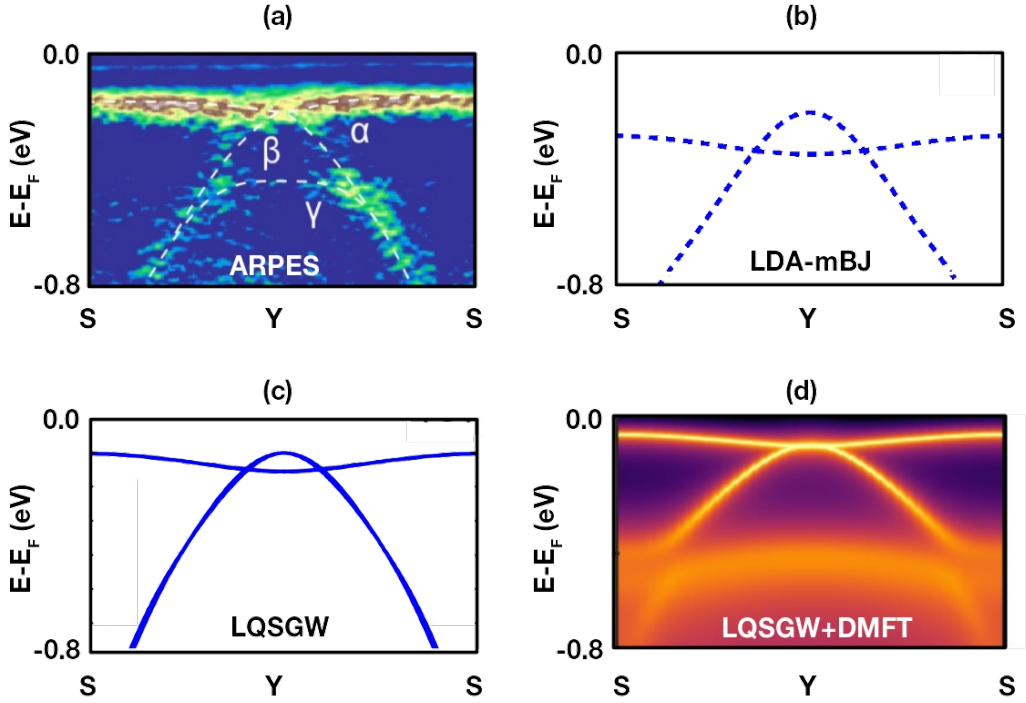
$$\widetilde{G}_{loc} = \mathbb{P}(G)$$

$$Z_{DC}^{-1} = \mathbb{E}(\widetilde{Z}_{DC}^{-1}) + \left(1 - \mathbb{E}(\widetilde{1}) \right)$$

$$S_{eff}(\widetilde{\mathcal{G}}, \widetilde{\mathcal{U}}) \rightarrow \widetilde{\Sigma}_{imp}$$

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

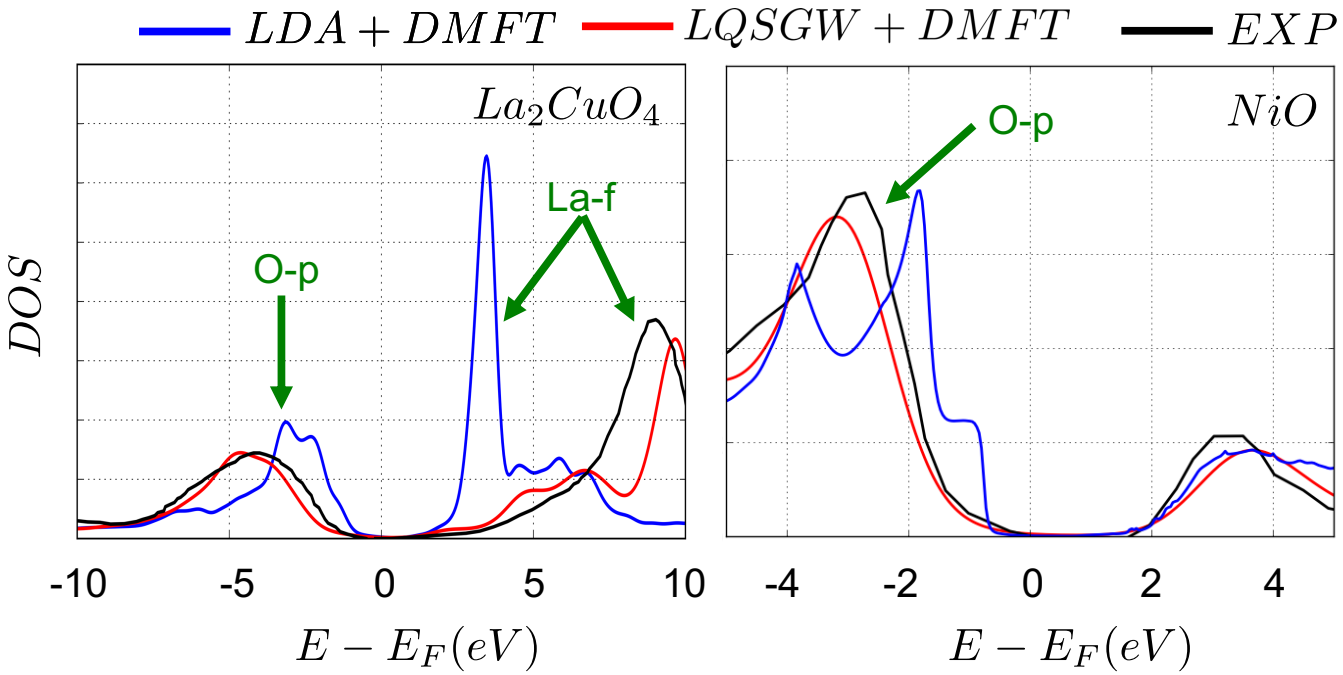
Validation: $FeSb_2$ bandstructure



- $FeSb_2$: 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, … **S.Choi,** et al., Phys. Rev. Research 2, 023190 (2020).

Validation: La2CuO4 and NiO



[1]N. Nucker, et.al., Z. Physik B – Condensed Matter 67, 9 (1987).
[2]R. Zimmermann, et.al., J. Phys.: Condens. Matter 11, 1657 (1999).
[3] S. Choi, et al., npj Quantum Materials 1, 16001 (2016).

20 minutes break

5. Full GW+EDMFT

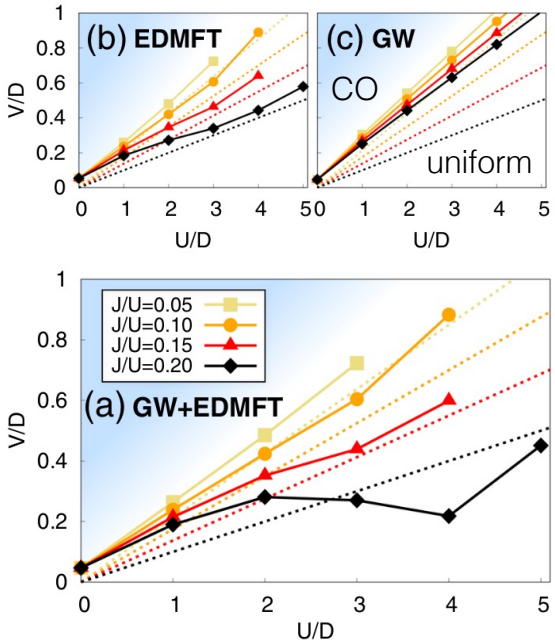
What about full GW+EDMFT? Will full GW+EDMFT make a difference?

- Three orbital extended Hubbard model

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

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

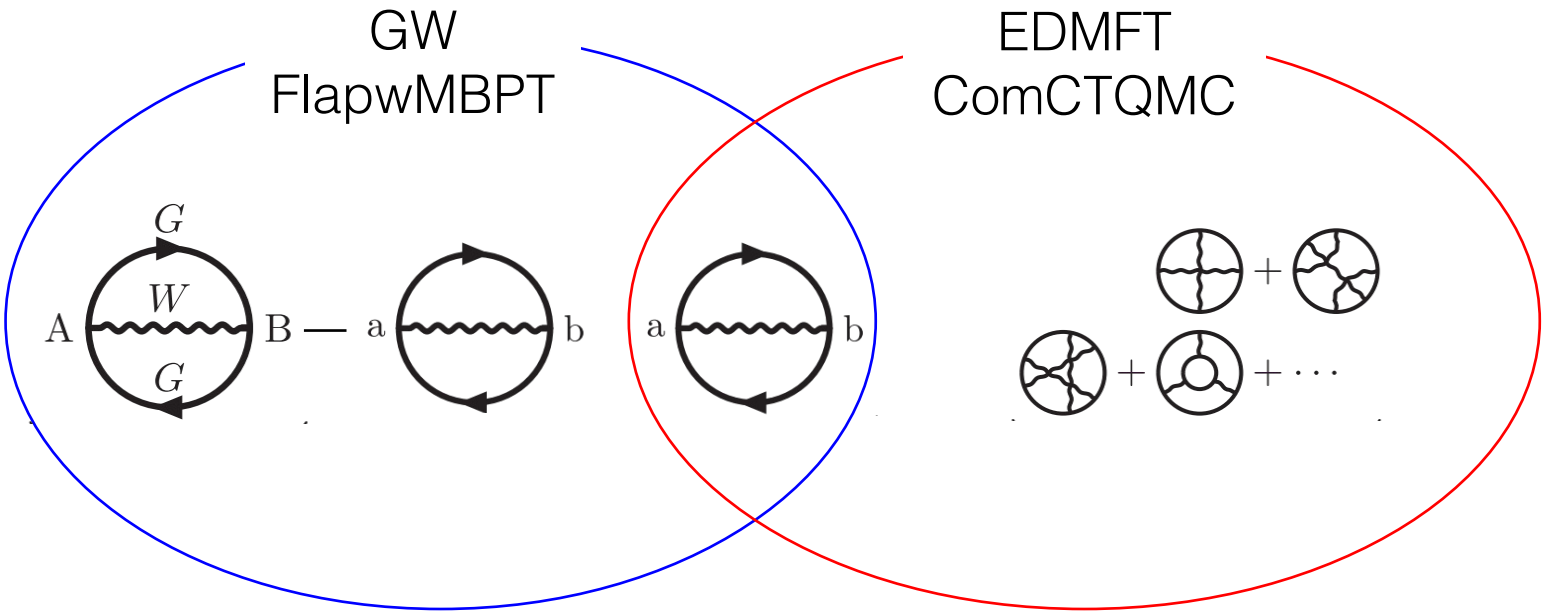
$$H_{\text{nonloc}} = \sum_{\langle ij\rangle\,\gamma,\gamma',\sigma,\sigma'} V n_{i\gamma\sigma} n_{j\gamma'\sigma'}.$$

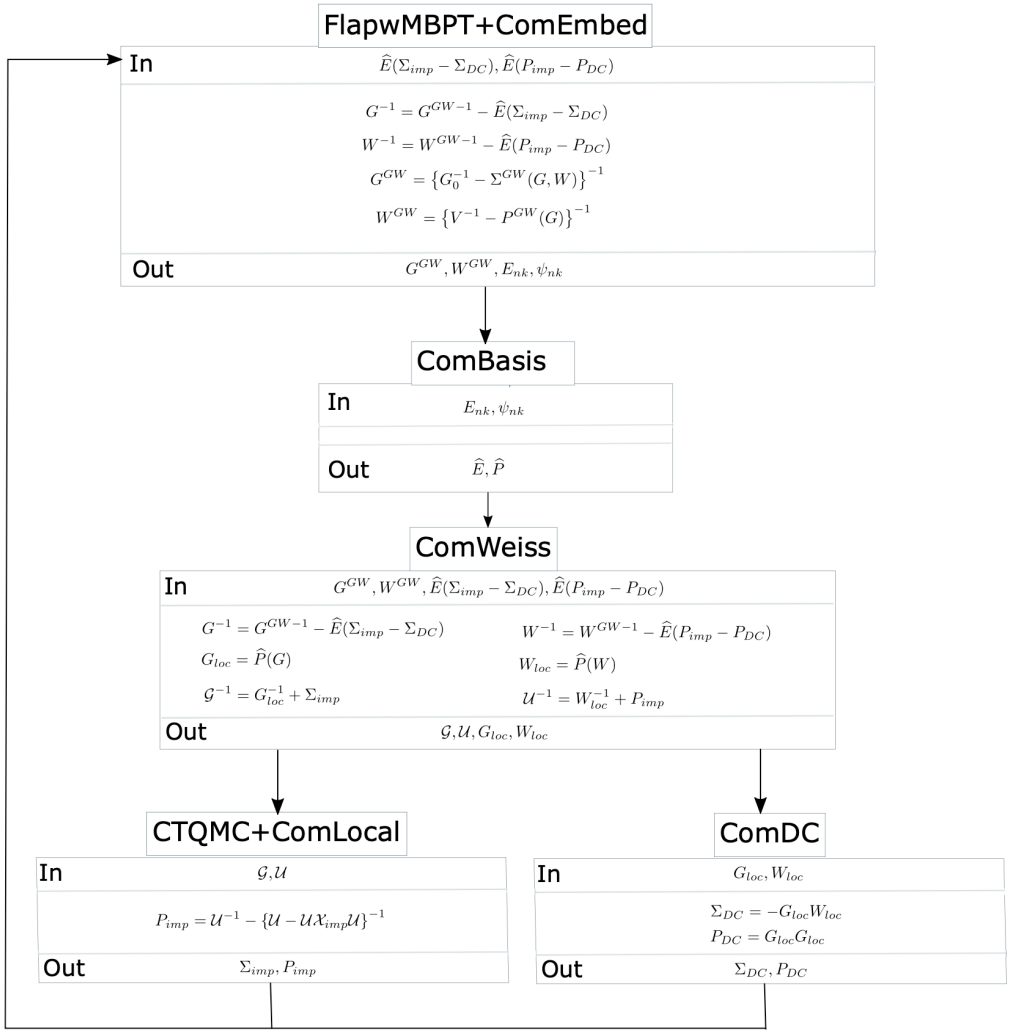


- Hund's coupling induced valence-skipping (negative U) charge order instability enhancement
- Only visible within GW+EDMFT

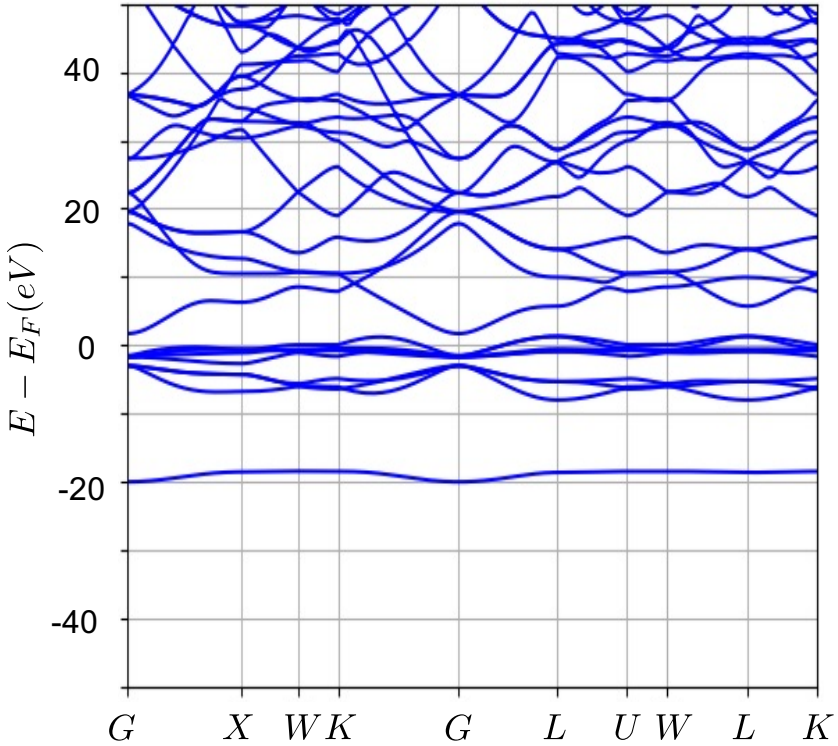
[1]S. Ryee, P. Sémon, M. J. Han, and **S. Choi**, Npj Quantum Mater. 5, 1 (2020).

New features: full GW+EDMFT (experimental, coming soon)




FlapwMBPT+ComEmbed
In
$\widehat{E}(\Sigma_{imp}-\Sigma_{DC}), \widehat{E}(P_{imp}-P_{DC})$
$G^{-1} = G^{GW-1} - \widehat{E}(\Sigma_{imp}-\Sigma_{DC})$
$W^{-1} = W^{GW-1} - \widehat{E}(P_{imp}-P_{DC})$
$G^{GW} = \{G_0^{-1} - \Sigma^{GW}(G,W)\}^{-1}$
$W^{GW} = \{V^{-1} - P^{GW}(G)\}^{-1}$
Out
$G^{GW}, W^{GW}, E_{nk}, \psi_{nk}$
ComBasis
In
E_{nk}, ψ_{nk}
Out
$\widehat{E}, \widehat{P}$
ComWeiss
In
$G^{GW}, W^{GW}, \widehat{E}(\Sigma_{imp}-\Sigma_{DC}), \widehat{E}(P_{imp}-P_{DC})$
$G^{-1} = G^{GW-1} - \widehat{E}(\Sigma_{imp}-\Sigma_{DC})$
$W^{-1} = W^{GW-1} - \widehat{E}(P_{imp}-P_{DC})$
$G_{loc} = \widehat{P}(G)$
$W_{loc} = \widehat{P}(W)$
$\mathcal{G}^{-1} = G_{loc}^{-1} + \Sigma_{imp}$
$\mathcal{U}^{-1} = W_{loc}^{-1} + P_{imp}$
Out
$\mathcal{G}, \mathcal{U}, G_{loc}, W_{loc}$
CTQMC+ComLocal
In
$\mathcal{G}, \mathcal{U}$
$P_{imp} = \mathcal{U}^{-1} - \{\mathcal{U} - \mathcal{U}\chi_{imp}\mathcal{U}\}^{-1}$
Out
Σ_{imp}, P_{imp}
ComDC
In
G_{loc}, W_{loc}
$\Sigma_{DC} = -G_{loc}W_{loc}$
$P_{DC} = G_{loc}G_{loc}$
Out
Σ_{DC}, P_{DC}

Hilbert space and its subspaces for Full GW+EDMFT



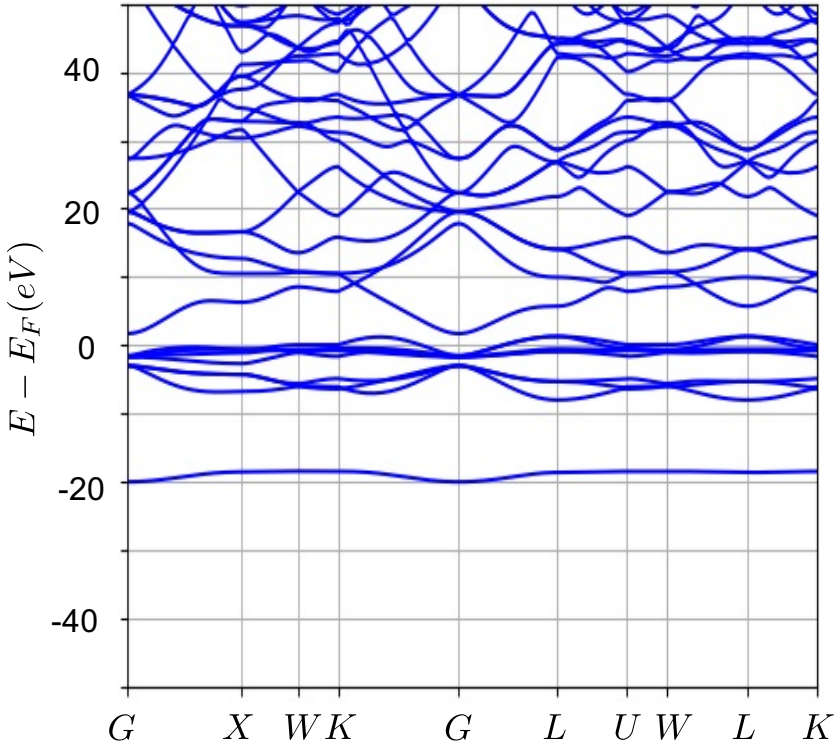
C ⊂ L ⊂ F

- F: full space
  - F= Tier-I ⊕ Tier-II ⊕ Tier-III
  - notation: $A\ (r, r')$
- L: Low-energy subspace defined by Wannier functions spanning an energy window ($\mathrm{E_F} \pm 10eV$)
  - L=Tier-I ⊕ Tier-II
  - notation: $\bar{A}_{ij}$, i,j=> Wannier functions
- C: correlated subspace
  - C=Tier-I
  - notation: $\tilde{A}_{ij}$, i,j=> Wannier functions

Tier-I: GW+EDMFT
Tier-II: GW
Tier-III: GW

Frequency interpolation

- To study materials properties at temperature <1000K, we use two different simulation temperatures



- Calculation in Tier-I and Tier-II will be done at temperature <1000K
- Calculation in Tier-III will be done at temperature ~1000K

GW Green's functions-I: coarse k-grid

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

$$G_{GW}(\mathbf{k}^c, i\omega_n^c) = \left(G_0^{-1}(\mathbf{k}^c, i\omega_n^c) - \Sigma_{GW}(\mathbf{k}^c, i\omega_n^c)\right)^{-1}$$

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

$$P_{GW}(\mathbf{k}^c, i\nu_n^c) = -\sum_{\mathbf{R}^c} \int_0^{\beta^c} d\tau G(\mathbf{R}^c, \tau) \circ G(-\mathbf{R}^c, -\tau) e^{-i(\mathbf{k}^c \cdot \mathbf{R}^c - \nu_n^c \tau)}$$

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

$$W_{GW}(\mathbf{k}^c, i\omega_n^c) = \left(V^{-1}(\mathbf{k}^c) - P_{GW}(\mathbf{k}^c, i\omega_n^c)\right)^{-1}$$

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

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

- The developer: Andrey Kutepov
- LAPW basis set
- space-time methods (to avoid convolution)
- calculation in a coarse Matsubara frequency grid (typical simulation temperature ~ 1000K)
- calculation in a coarse momentum space grid (e.g. NiO: 6×6×6)

Fermionic projection operator

- The formulation within Wannier90 package is based on one-particle picture
- We construct quasiparticle Hamiltonian by linearizing GW self-energy

$$Z_{GW}(\mathbf{k}^c) = \left(1 - \frac{\partial \Sigma_{GW}(\mathbf{k}^c, \omega)}{\partial \omega}\bigg|_{\omega=0}\right)^{-1}$$

$$H^{QP}(\mathbf{k}^c) = Z^{1/2}(\mathbf{k}^c)\left(H_0(\mathbf{k}^c) + \Sigma_{GW}(\mathbf{k}^c, \omega = 0)\right) Z^{1/2}(\mathbf{k}^c)$$

$\rightarrow E^{QP}_{n\mathbf{k^c}}, |n\mathbf{k}^c\rangle \rightarrow A_{mn}(\mathbf{k}^c), U_{mn}(\mathbf{k}^c, \mathbf{b}^c)$

WANNIER90

$$|\tau\mathbf{k}^c\rangle = \frac{1}{\sqrt{N_{\mathbf{k}^c}}} \sum_{R^c} e^{i\mathbf{k}^c \cdot \mathbf{R}^c} |\tau\mathbf{R}^c\rangle$$

$$f(\mathbf{k}^c) = \langle n\mathbf{k}^c | \tau\mathbf{k}^c \rangle$$

Bosonic projection operator

- With products of Wannier functions for correlated orbitals

$$D_{\tau,\tau'}(\mathbf{r}) = W_{R^c=0,\tau}(\mathbf{r}) W^*_{R^c=0,\tau'}(\mathbf{r}) \qquad W_{R^c,\tau}(\mathbf{r}) = \langle \mathbf{r} | \tau \mathbf{R}^c \rangle$$

- Orthonormalized product basis can be represented as a linear combination of the product C

$$\langle \mathbf{r} | B_I \rangle = \sum_{\tau,\tau'} X_{\tau,\tau';I} D_{\tau,\tau'}(\mathbf{r})$$

- The coefficient X can be calculated by diagonalizing the overlap matrix of C

$$O_{\tau_1,\tau_2;\tau_3,\tau_4} = \langle D_{\tau_1,\tau_2} | D_{\tau_3,\tau_4} \rangle$$

$$\sum_{\tau_3,\tau_4} O_{\tau_1,\tau_2;\tau_3,\tau_4} V_{\tau_3,\tau_4;I} = F_I V_{\tau_1,\tau_2;I}$$

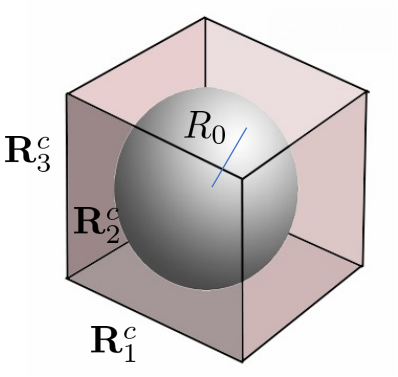
$$X_{\tau_1,\tau_2;I} = \frac{1}{\sqrt{F_I}} V_{\tau_1,\tau_2;I}$$

GW Green's functions-II: fine k-grid

- To improve the momentum space resolution, we interpolate G_{GW} and W_{GW} obtained from FlapwMBPT
- With localized basis set, hopping energy and screened Coulomb interaction are essentially 0 beyond a few neighbors.

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

$$\begin{aligned}
&G^{-1}_{\tau,\tau'}(\mathbf{k}^f, i\omega_n^c) \\
&= \sum_{\mathbf{R}^f} G^{-1}_{\tau,\tau'}(\mathbf{R}^f, i\omega_n^c) e^{i\mathbf{k}^f \cdot \mathbf{R}^f} \\
&= \sum_{|\mathbf{R}^f| \le R_0} G^{-1}_{\tau,\tau'}(\mathbf{R}^f, i\omega_n^c) e^{i\mathbf{k}^f \cdot \mathbf{R}^f} + \sum_{|\mathbf{R}^f| > R_0} G^{-1}_{\tau,\tau'}(\mathbf{R}^f, i\omega_n^c) e^{i\mathbf{k}^f \cdot \mathbf{R}^f} \\
&= \sum_{|\mathbf{R}^c| \le R_0} G^{-1}_{\tau,\tau'}(\mathbf{R}^c, i\omega_n^c) e^{i\mathbf{k}^f \cdot \mathbf{R}^c}
\end{aligned}$$



[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).

Quantum Embedding

For Green's function

$$\overline{G}^{-1}_{GW}(\mathbf{k}^c, i\omega^c_n) = \overline{f}^{\dagger}(\mathbf{k}^c) G^{-1}_{GW}(\mathbf{k}^c, i\omega^c_n) \overline{f}(\mathbf{k}^c)$$

$$\overline{G}^{-1}_{GW}(\mathbf{k}^c, i\omega^c_n) \rightarrow \overline{G}^{-1}_{GW}(\mathbf{k}^f, i\omega^f_n)$$

$$\overline{G}(\mathbf{k}^f, i\omega^f_n) = \left(\overline{G}^{-1}_{GW}(\mathbf{k}^f, i\omega^f_n) + \widetilde{f}(\mathbf{k}^c) \left(\widetilde{\Sigma}_{imp}(i\omega^f_n) - \widetilde{\Sigma}_{DC}(i\omega^f_n) \right) \widetilde{f}^{\dagger}(\mathbf{k}^c) \right)^{-1}$$

$$\widetilde{G}_{loc}(i\omega^f_n) = \frac{1}{N_{\mathbf{k}^f}} \sum_{\mathbf{k}^f} \widetilde{f}^{\dagger}(\mathbf{k}^f) \overline{G}(\mathbf{k}^f, i\omega^f_n) \widetilde{f}(\mathbf{k}^f)$$

For screened Coulomb interaction

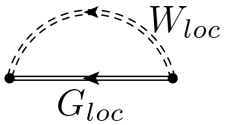
$$\overline{W}_{GW}(\mathbf{k}^c, i\nu^c_n) = \overline{b}^{\dagger}(\mathbf{k}^c) W_{GW}(\mathbf{k}^c, i\nu^c_n) \overline{b}(\mathbf{k}^c)$$

$$\overline{W}_{GW}(\mathbf{k}^c, i\nu^c_n) \rightarrow \overline{W}_{GW}(\mathbf{k}^f, i\nu^f_n)$$

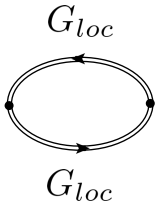
$$\overline{W}(\mathbf{k}^f, i\nu^f_n) = \left(\overline{W}^{-1}_{GW}(\mathbf{k}^f, i\nu^f_n) + \widetilde{b}(\mathbf{k}^c) \left(\widetilde{P}_{imp}(i\nu^f_n) - \widetilde{P}_{DC}(i\nu^f_n) \right) \widetilde{b}^{\dagger}(\mathbf{k}^c) \right)^{-1}$$

$$\widetilde{W}_{loc}(i\omega^f_n) = \frac{1}{N_{\mathbf{k}^f}} \sum_{\mathbf{k}^f} \widetilde{b}^{\dagger}(\mathbf{k}^f) \overline{W}(\mathbf{k}^f, i\nu^f_n) \widetilde{b}(\mathbf{k}^f)$$

Double Counting



$$\widetilde{\Sigma}_{DC}(i\omega_n^f) = -\int_0^{\beta^f} d\tau \widetilde{G}_{loc}(\tau) \circ \widetilde{W}_{loc}(-\tau) e^{i\omega_n^f \tau}$$



$$\widetilde{P}_{DC}(i\omega_n^f) = -\int_0^{\beta^f} d\tau \widetilde{G}_{loc}(\tau) \circ \widetilde{G}_{loc}(-\tau) e^{i\nu_n^f \tau}$$

DMFT effective action

$$\widetilde{\mathcal{G}}(i\omega_n^f) = \left(\widetilde{G}_{loc}^{-1}(i\omega_n^f) + \widetilde{\Sigma}_{imp}(i\omega_n^f)\right)^{-1}$$

$$\widetilde{\mathcal{U}}(i\nu_n^f) = \left(\widetilde{W}_{loc}^{-1}(i\nu_n^f) + \widetilde{P}_{imp}(i\nu_n^f)\right)^{-1}$$

ComCTQMC

$$S_{eff} \rightarrow \widetilde{\Sigma}_{imp}(i\omega_n^f), \widetilde{\chi}_{imp}(i\nu_n^f)$$

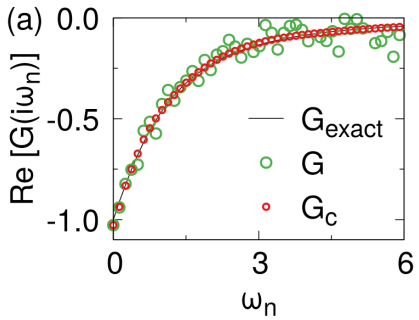
$$\widetilde{W}_{imp}(i\nu_n^f) = \widetilde{\mathcal{U}}(i\nu_n^f) - \widetilde{\mathcal{U}}(i\nu_n^f)\widetilde{\chi}_{imp}(i\nu_n^f)\widetilde{\mathcal{U}}(i\nu_n^f)$$

$$\widetilde{P}_{imp}(i\nu_n^f) = \widetilde{\mathcal{U}}^{-1}(i\nu_n^f) - \widetilde{W}_{imp}^{-1}(i\nu_n^f)$$

- The lead developer: Corey Melnick
- The first developer: Patrick Semon
- continuous-time Monte Carlo solver (hybridization-expansion)
- GPU-accelerated

Causal Optimization of Bosonic Quantities

Statistical noise



Local self-energy assumption breakdown

$$\mathcal{U}^{-1}(i\nu_n) = \left[\langle W\rangle^{-1} + \langle \Pi\rangle\right] + \tilde{\mathcal{U}}_{\mathrm{cor}}^{-1},$$

Causal Bosonic functions

$$G(i\omega_n) = \int \frac{B(x)x}{i\omega_n - x}dx, \quad B(x) \geqslant 0, \quad B(x) = B(-x)$$

$$G(\tau) = -\int B(x)xn_B(x)e^{(\beta-\tau)x}dx, \quad B(x) \geqslant 0,$$

$$G^{(2k)}(\tau) \leqslant 0 \text{ for } k = 0, 1, 2, \ldots. \quad G(\tau) = G(\beta - \tau)$$

For a given non-causal $\mathrm{G}(\mathrm{i}\omega_\mathrm{n})$, search causal $\mathrm{G_c}(\mathrm{i}\omega_\mathrm{n})$ which minimize the distance defined as

$$d = \frac{1}{\beta}\int_0^\beta [G(\tau) - G_c(\tau)]^2 d\tau$$

[1] M. Han and H. J. Choi, Phys. Rev. B 104, 115112 (2021). [2] J. Chen, F. Petocchi, and P. Werner, Phys. Rev. B 105, 085102 (2022).
[3] S. Backes, J.-H. Sim, and S. Biermann, arXiv:2011.05311v1

Feedback to GW Green's functions

$$\widetilde{\Sigma}_{imp}(i\omega_n^f) \to \widetilde{\Sigma}_{imp}(i\omega_n^c) \qquad \widetilde{\Sigma}_{DC}(i\omega_n^f) \to \widetilde{\Sigma}_{DC}(i\omega_n^c)$$
$$\widetilde{P}_{imp}(i\omega_n^f) \to \widetilde{P}_{imp}(i\omega_n^c) \qquad \widetilde{P}_{DC}(i\omega_n^f) \to \widetilde{P}_{DC}(i\omega_n^c)$$

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

$$G_{GW}(\mathbf{k}^c, i\omega_n^c) = \left(G_0^{-1}(\mathbf{k}^c, i\omega_n^c) - \Sigma_{GW}(\mathbf{k}^c, i\omega_n^c)\right)^{-1}$$

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

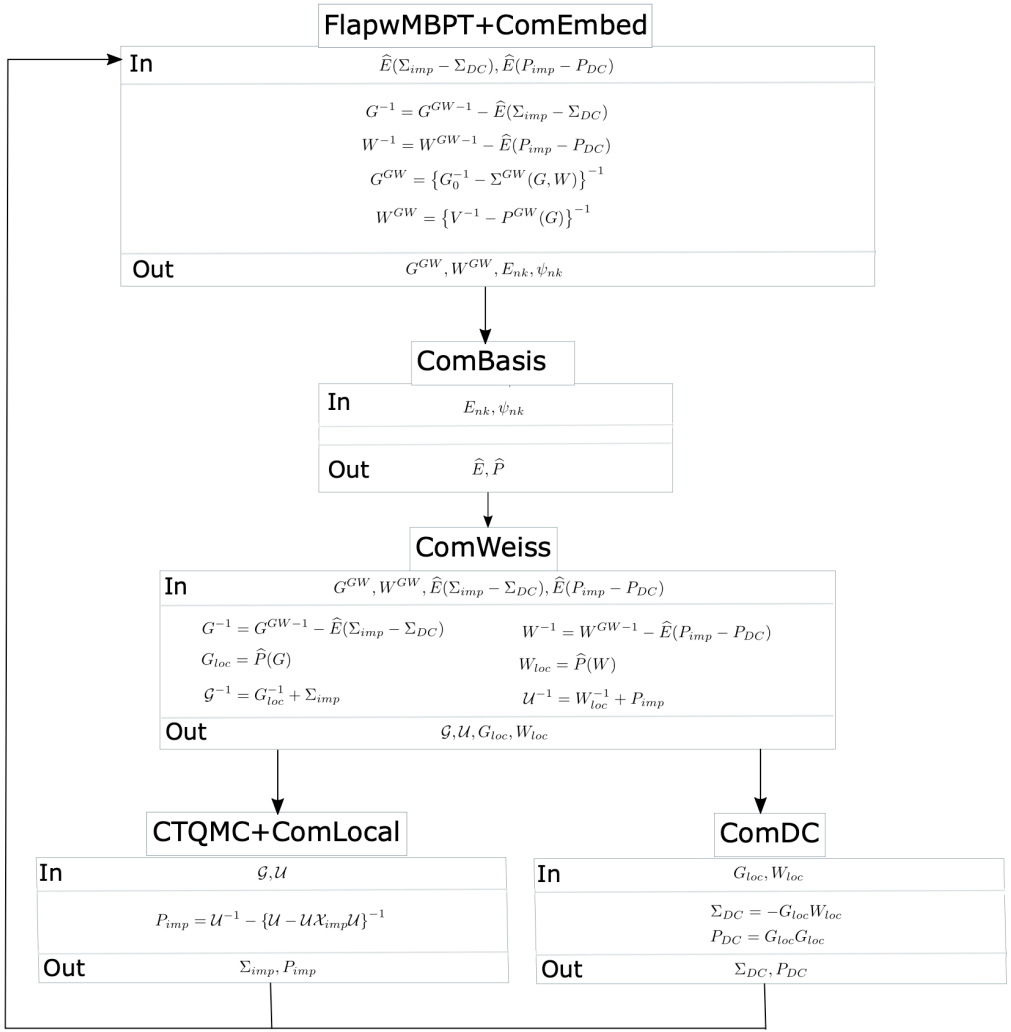
$$P_{GW}(\mathbf{k}^c, i\nu_n^c) = -\sum_{\mathbf{R}^c} \int_0^{\beta^c} d\tau G(\mathbf{R}^c, \tau) \circ G(-\mathbf{R}^c, -\tau) e^{-i(\mathbf{k}^c \cdot \mathbf{R}^c - \nu_n^c \tau)}$$

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

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

$$W_{GW}(\mathbf{k}^c, i\omega_n^c) = \left(V^{-1}(\mathbf{k}^c) - P_{GW}(\mathbf{k}^c, i\omega_n^c)\right)^{-1}$$

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


FlapwMBPT+ComEmbed
In
$\widehat{E}(\Sigma_{imp}-\Sigma_{DC}), \widehat{E}(P_{imp}-P_{DC})$
$G^{-1} = G^{GW-1} - \widehat{E}(\Sigma_{imp}-\Sigma_{DC})$
$W^{-1} = W^{GW-1} - \widehat{E}(P_{imp}-P_{DC})$
$G^{GW} = \{G_0^{-1} - \Sigma^{GW}(G,W)\}^{-1}$
$W^{GW} = \{V^{-1} - P^{GW}(G)\}^{-1}$
Out
$G^{GW}, W^{GW}, E_{nk}, \psi_{nk}$
ComBasis
In
E_{nk}, ψ_{nk}
Out
$\widehat{E}, \widehat{P}$
ComWeiss
In
$G^{GW}, W^{GW}, \widehat{E}(\Sigma_{imp}-\Sigma_{DC}), \widehat{E}(P_{imp}-P_{DC})$
$G^{-1} = G^{GW-1} - \widehat{E}(\Sigma_{imp}-\Sigma_{DC})$
$W^{-1} = W^{GW-1} - \widehat{E}(P_{imp}-P_{DC})$
$G_{loc} = \widehat{P}(G)$
$W_{loc} = \widehat{P}(W)$
$\mathcal{G}^{-1} = G_{loc}^{-1} + \Sigma_{imp}$
$\mathcal{U}^{-1} = W_{loc}^{-1} + P_{imp}$
Out
$\mathcal{G}, \mathcal{U}, G_{loc}, W_{loc}$
CTQMC+ComLocal
In
$\mathcal{G}, \mathcal{U}$
$P_{imp} = \mathcal{U}^{-1} - \{\mathcal{U} - \mathcal{U}\chi_{imp}\mathcal{U}\}^{-1}$
Out
Σ_{imp}, P_{imp}
ComDC
In
G_{loc}, W_{loc}
$\Sigma_{DC} = -G_{loc}W_{loc}$
$P_{DC} = G_{loc}G_{loc}$
Out
Σ_{DC}, P_{DC}