LQSGW+DMFT and fully self-consistent GW+EDMFT

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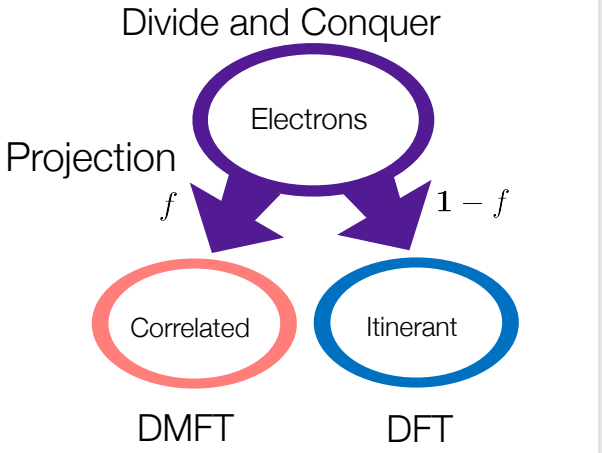
Outline

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: SrVO3, NiO

1. Successes and challenges of DFT+DMFT

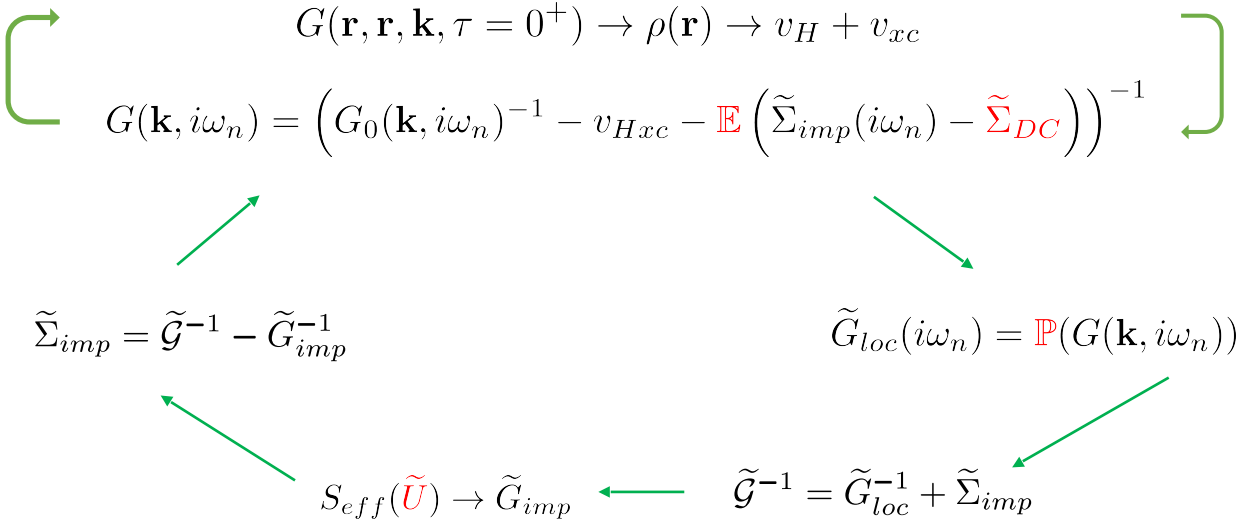
DFT + DMFT is a work horse methodology for correlated quantum materials



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

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

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



$\widetilde{U}$ 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

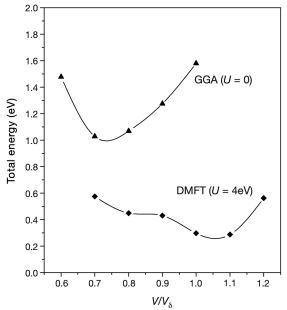
HOME About AMULET FEATURES Why it is special DOCUMENTA Inputs and HOWT

AMULET

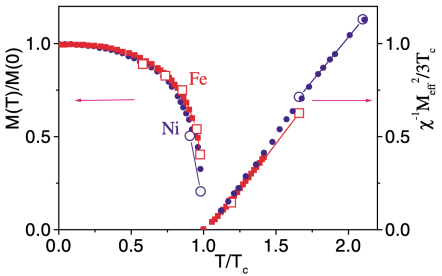
The AMULET is a free software for scientific and/or educational purposes and it is distributed under FreeBSD License. To
description of a planned research has to be submitted to AMULET Developers Team. In the description one needs to indi
and explain shortly why conventional band structure methods do 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)

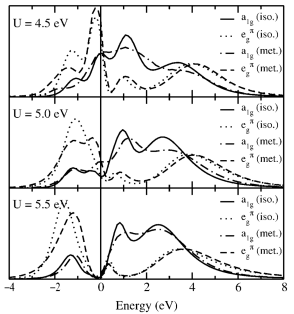
Volume collapse in Pu



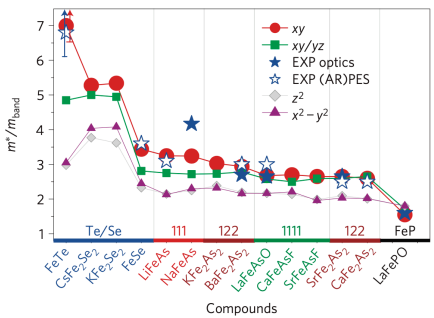
Finite T ferromagnetism



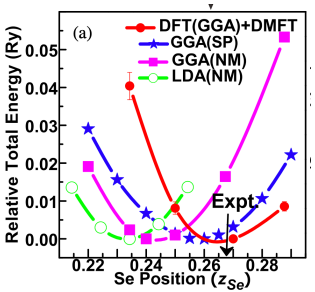
MIT in V2O3



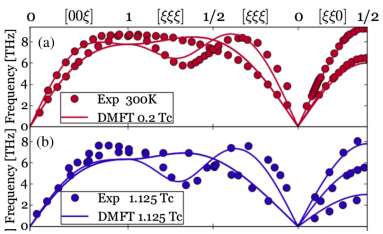
Hund metal physics



Structure prediction (FeSe)



Phonon



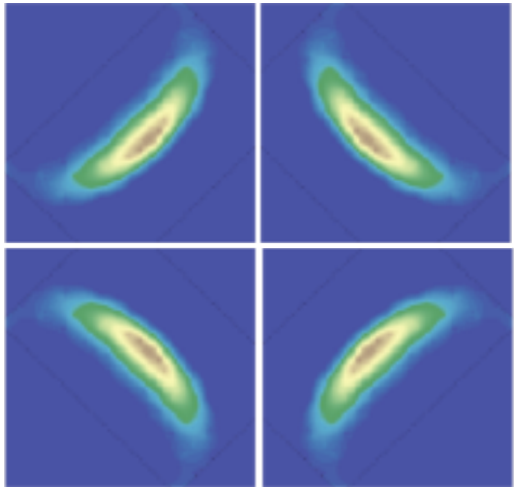
[1] S. Y. Savrasov, G. Kotliar, and E. Abrahams, Nature 410, 793 (2001). [2] A. I. Lichtenstein, M. I. Katsnelson, and G. Kotliar, Phys. Rev. Lett. 87, 067205 (2001). [3]K. Held, G. Keller, V. Eyert, D. Vollhardt, and V. I. Anisimov, Phys. Rev. Lett. 86, 5345 (2001).[4] Zhiping. P. Yin, K. Haule, and 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?

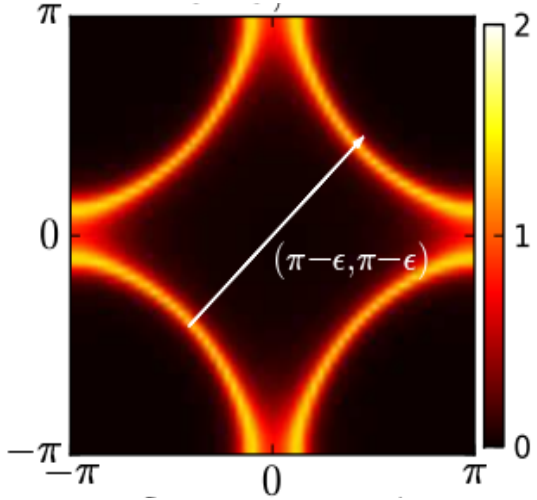
DFT + single-site DMFT: local (no k dependence) and dynamical (frequency-dependent) self-energy

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

Pseudo-gap in Cuprates



Single-site DMFT, Hubbard model

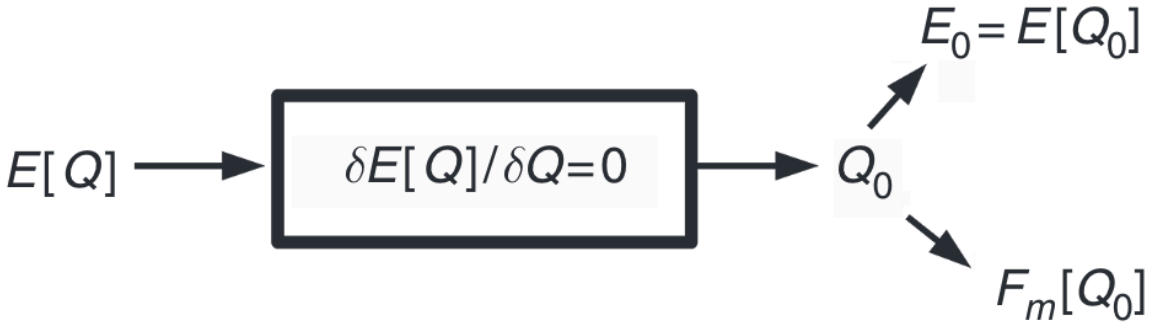


For the system where non-local (k-dependent) correlation is important, we need a tool beyond DFT + single site DMFT

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

2. Introduction of GW+EDMFT

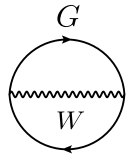
Functional approaches to quantum many-body problems



Brachistochrone curve

$$\frac{\delta T[y(x)]}{\delta y(x)} = 0$$

Free energy functional and Ψ functional (instead of density functional theory)



Any two particle irreducible diagram in G and W, e.g.

- Free Energy Functional

$$\Gamma[G,\Sigma,W,\Pi] = \underbrace{-\operatorname{Tr}[log(-G_H^{-1})] - \operatorname{Tr}[log(1-G_H\Sigma)] - \operatorname{Tr}[G\Sigma]}_{\text{Fermionic}} + \underbrace{\frac{1}{2}\operatorname{Tr}[log(1-V\Pi)] + \frac{1}{2}\operatorname{Tr}[\Pi W]}_{\text{Bosonic}} + \underbrace{\Psi[G,W]}_{\text{Fermion-boson coupling}}.$$

- Stationary Condition

$$\frac{\delta\Gamma}{\delta G} = 0 \rightarrow \Sigma = \frac{\delta\Psi}{\delta G}$$

$$\frac{\delta\Gamma}{\delta W} = 0 \rightarrow \Pi = -2\frac{\delta\Psi}{\delta W}$$

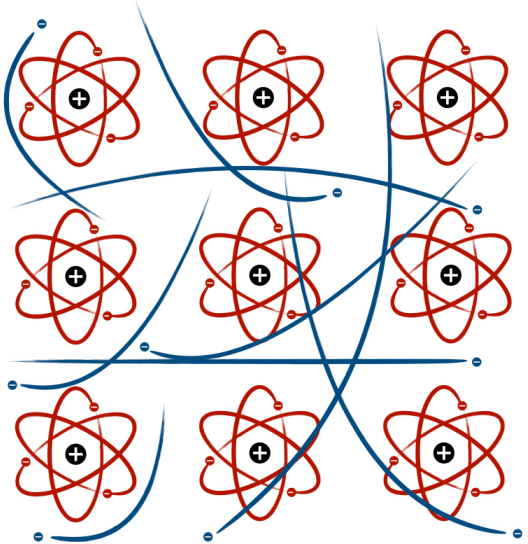
$$\frac{\delta\Gamma}{\delta\Sigma} = 0 \rightarrow G = \left(G_H^{-1} - \Sigma\right)^{-1}$$

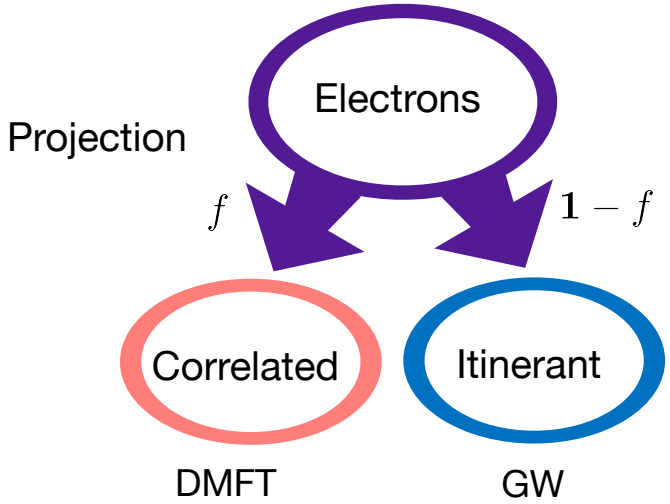
$$\frac{\delta\Gamma}{\delta\Pi} = 0 \rightarrow W = \left(V^{-1} - \Pi\right)^{-1}$$

[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).

Quantum embedding and projection

Divide and Conquer





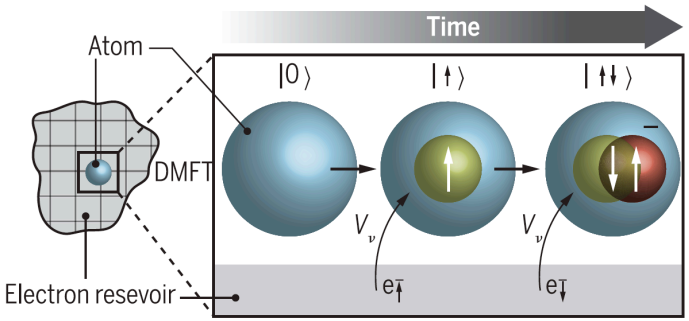
$$A = \begin{pmatrix} \widetilde{A}_{cc} & A_{ci} \\ A_{ic} & A_{ii} \end{pmatrix} \qquad f = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

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

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

Dynamical mean field theory provides a reference frame for correlated electrons

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



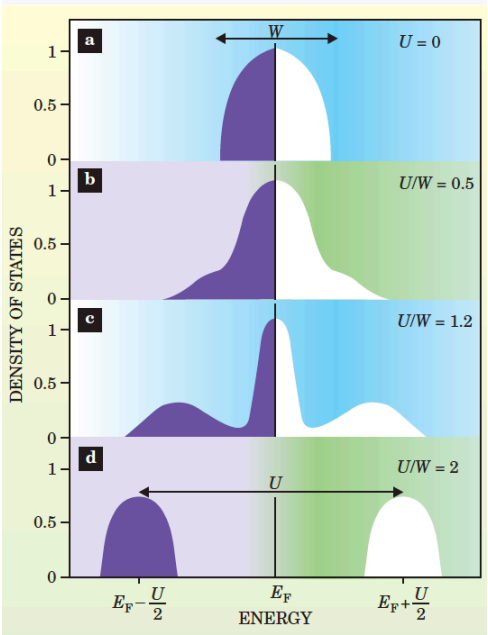
$$H = \sum_{ij,\sigma} t_{ij} c^{\dagger}_{i\sigma} c_{j\sigma} + \sum_{i} U n_{i\uparrow} n_{i\downarrow}$$

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

Electron hopping
Band-like

Multiplets
Atom-like

Single Band Hubbard Model



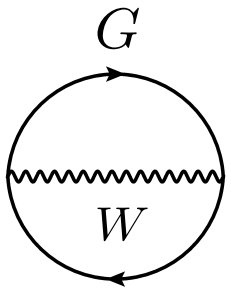
Band-like

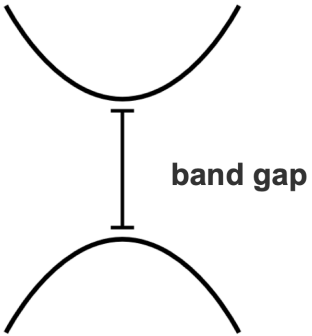
Atom-like

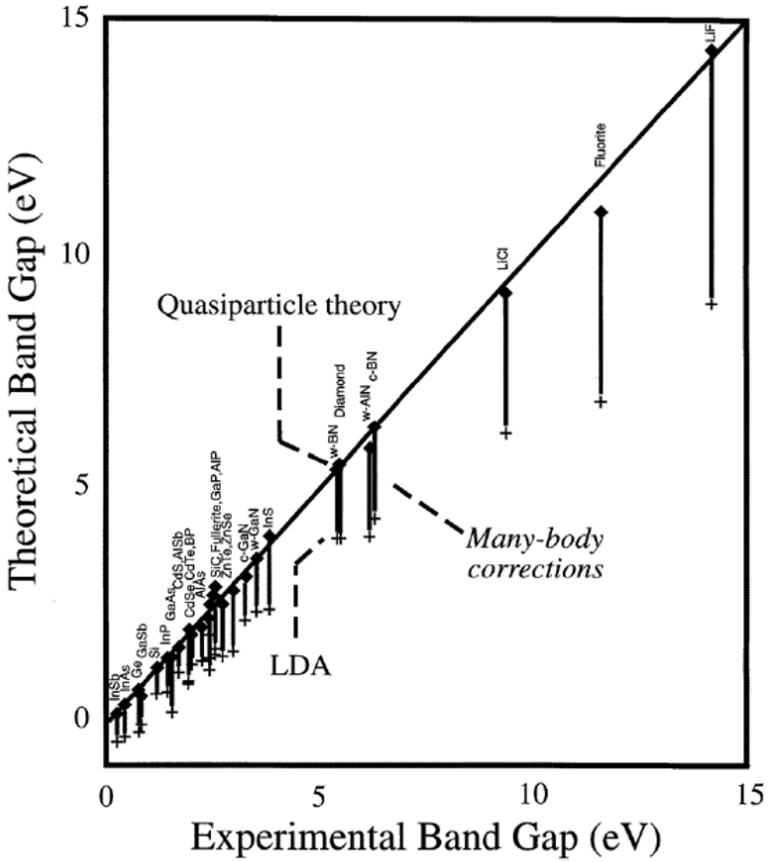
[1] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Rev. Mod. Phys. 68, 13 (1996) [2] G. Kotliar and D. Vollhardt, Physics Today 57, 53 (2004).

GW approximation provides a reference frame for itinerant electrons

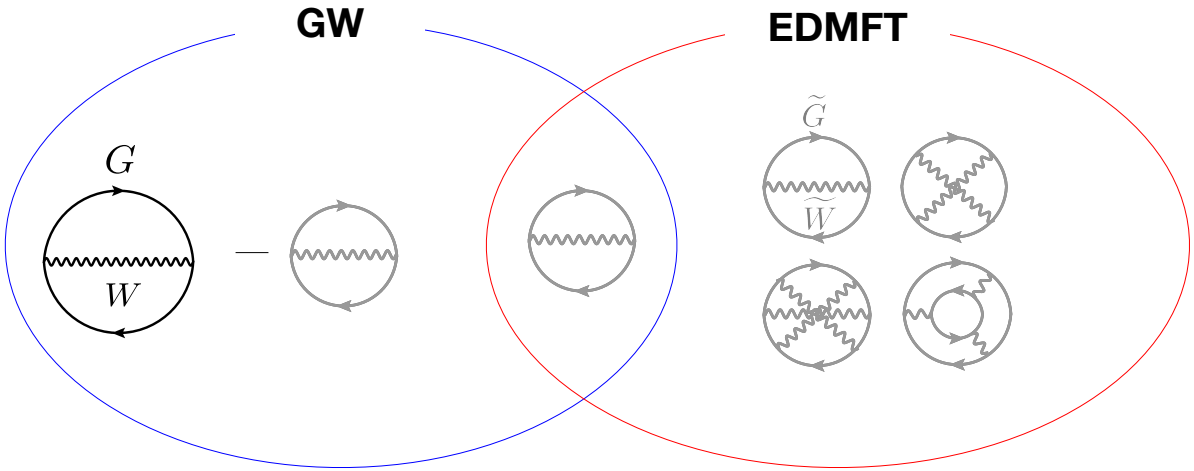
$$\Psi_{GW} =$$







$\Psi_{GW+EDMFT}$



$$\widetilde{G} = \hat{P}(G)$$
$$\widetilde{W} = \hat{P}(W)$$

$$\Psi_{GW+EDMFT}(G,W) = -\frac{1}{2}\,\mathrm{Tr}\,GWG + \Psi_{EDMFT}(\widetilde{G},\widetilde{W}) + \frac{1}{2}\,\mathrm{Tr}\,\widetilde{G}\widetilde{W}\widetilde{G}.$$

$$\Sigma(1,2) = -G(1,2)W(1,2) + \frac{\delta\Psi_{EDMFT}}{\delta\widetilde{G}(2,1)} + \widetilde{G}(1,2)\widetilde{W}(1,2).$$

$$\Pi(1,2) = G(1,2)G(2,1) - 2\frac{\delta\Psi_{EDMFT}}{\delta\widetilde{W}(2,1)} - \widetilde{G}(1,2)\widetilde{G}(2,1).$$

[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).

Three different self-consistent loop in GW+EDMFT

Non-local correlation within GW
Local correlation within EDMFT

$$\Sigma_H = GV$$
$$\Sigma_{GW} = -GW$$
$$\Pi_{GW} = GG$$

$$G = \{G_H^{-1} - \Sigma_{GW} - \hat{E}^f(\Sigma_{EDMFT} - \Sigma_{DC})\}^{-1}$$
$$W = \{V^{-1} - \Pi_{GW} - \hat{E}^b(\Pi_{EDMFT} - \Pi_{DC})\}^{-1}$$

$$\widetilde{G}_{loc} = \hat{P}^f(G)$$
$$\widetilde{W}_{loc} = \hat{P}^b(W)$$

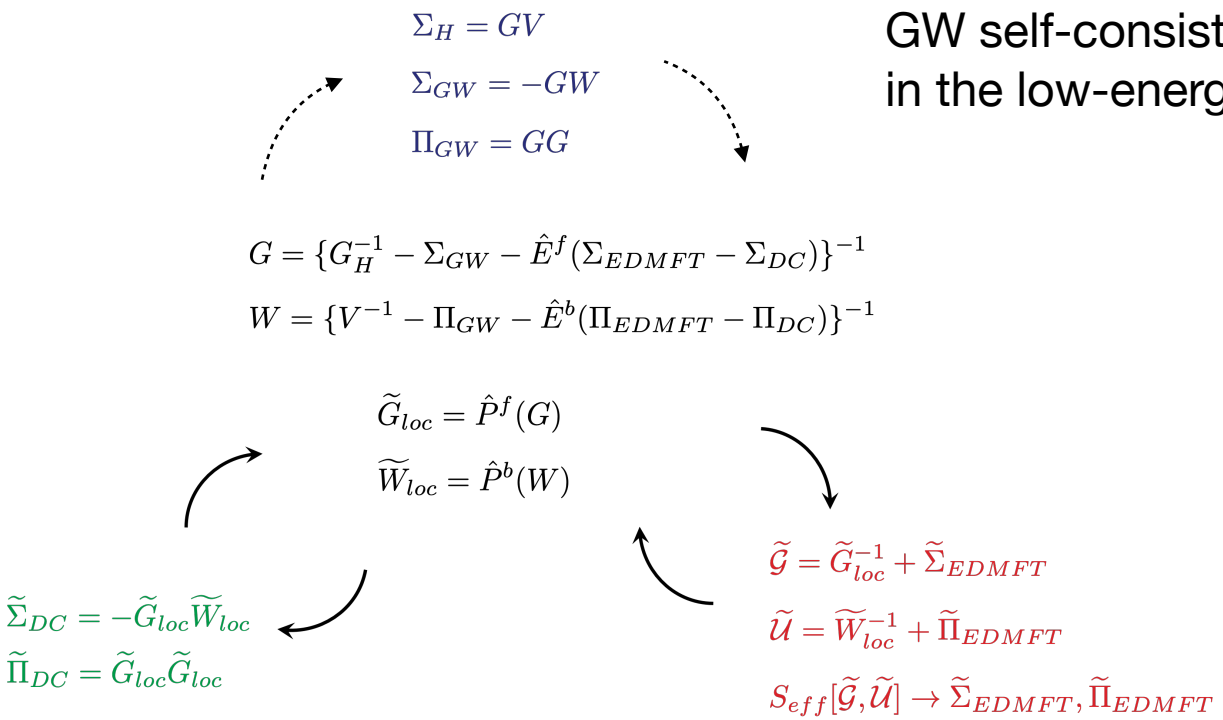
$$\widetilde{\Sigma}_{DC} = -\widetilde{G}_{loc}\widetilde{W}_{loc}$$
$$\widetilde{\Pi}_{DC} = \widetilde{G}_{loc}\widetilde{G}_{loc}$$

$$\widetilde{\mathcal{G}} = \widetilde{G}_{loc}^{-1} + \widetilde{\Sigma}_{EDMFT}$$
$$\widetilde{\mathcal{U}} = \widetilde{W}_{loc}^{-1} + \widetilde{\Pi}_{EDMFT}$$
$$S_{eff}[\widetilde{\mathcal{G}}, \widetilde{\mathcal{U}}] \rightarrow \widetilde{\Sigma}_{EDMFT}, \widetilde{\Pi}_{EDMFT}$$

$$\frac{\delta\Gamma_{GW+EDMFT}}{\delta W} = 0 \rightarrow P = 2GG + \frac{\delta\Psi_{EDMFT}}{\delta W} - 2\mathbb{E}(\widetilde{G}\widetilde{G})$$
$$\frac{\delta\Gamma_{GW+EDMFT}}{\delta P} = 0 \rightarrow W^{-1} = v^{-1} - P$$
$$\frac{\delta\Gamma_{GW+EDMFT}}{\delta G} = 0 \rightarrow \Sigma_{xc} = -GW + \frac{\delta\Psi_{EDMFT}}{\delta G} + \mathbb{E}(\widetilde{G}\widetilde{W})$$
$$\frac{\delta\Gamma_{GW+EDMFT}}{\delta\Sigma} = 0 \rightarrow G^{-1} = G_0^{-1} - \Sigma_H - \Sigma_{xc}$$

3. Approximation to GW+EDMFT: multitier scheme

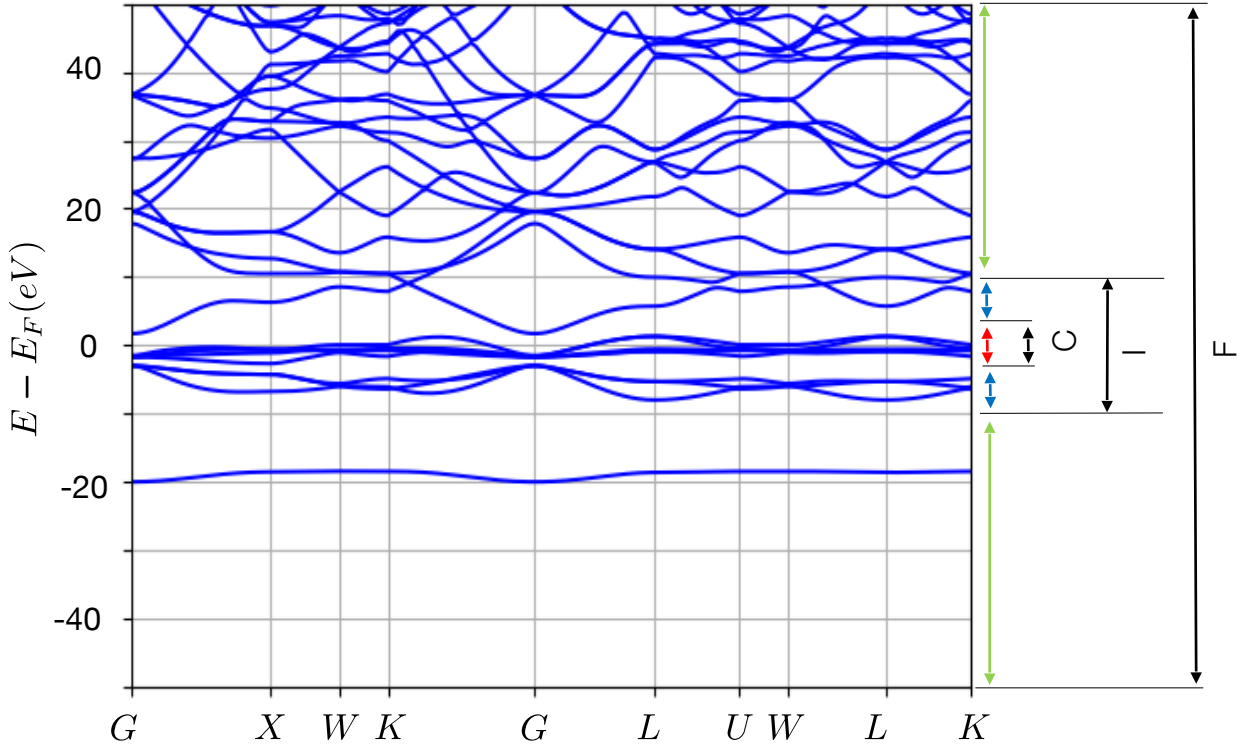
Simplification-I: multitier GW+EDMFT



GW self-consistency
in the low-energy Hilbert space

[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).

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



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

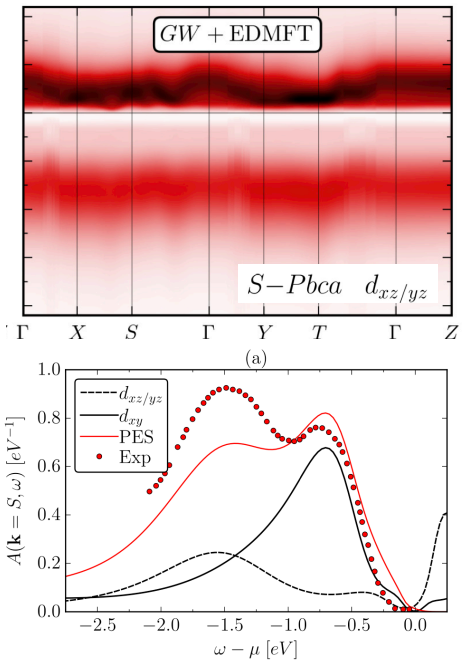
C=Tier-I
I=Tier-I ⊕ Tier-II
F= Tier-I ⊕ Tier-II ⊕ Tier-III

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

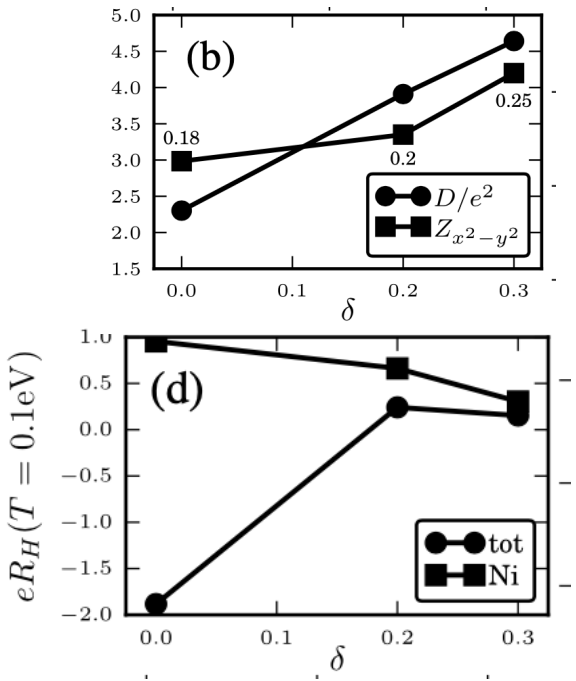
[1] F. Nilsson, L. Boehnke, P. Werner, and F. Aryasetiawan, Phys. Rev. Materials 1, 043803 (2017).

Application of multitier GW+EDMFT to real materials

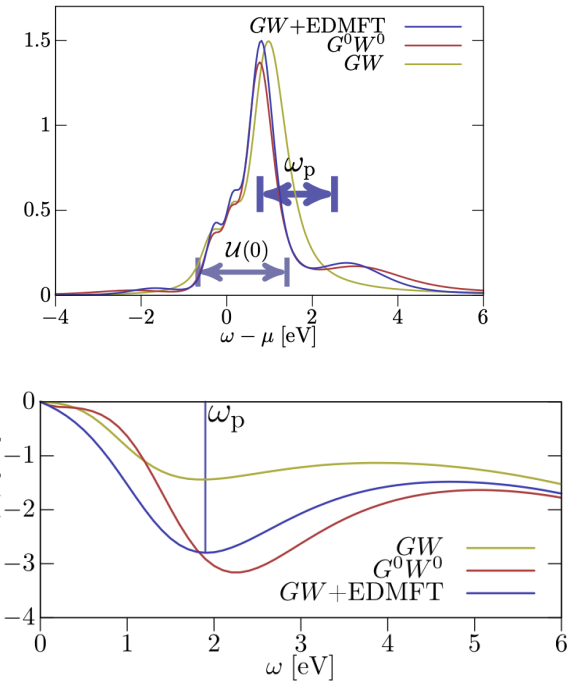
MIT in Ca_2RuO_4



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

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

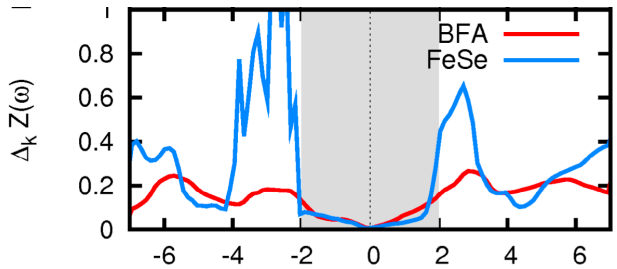
Validation on Fe-based superconductors quasiparticle bands

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

Within QSGW

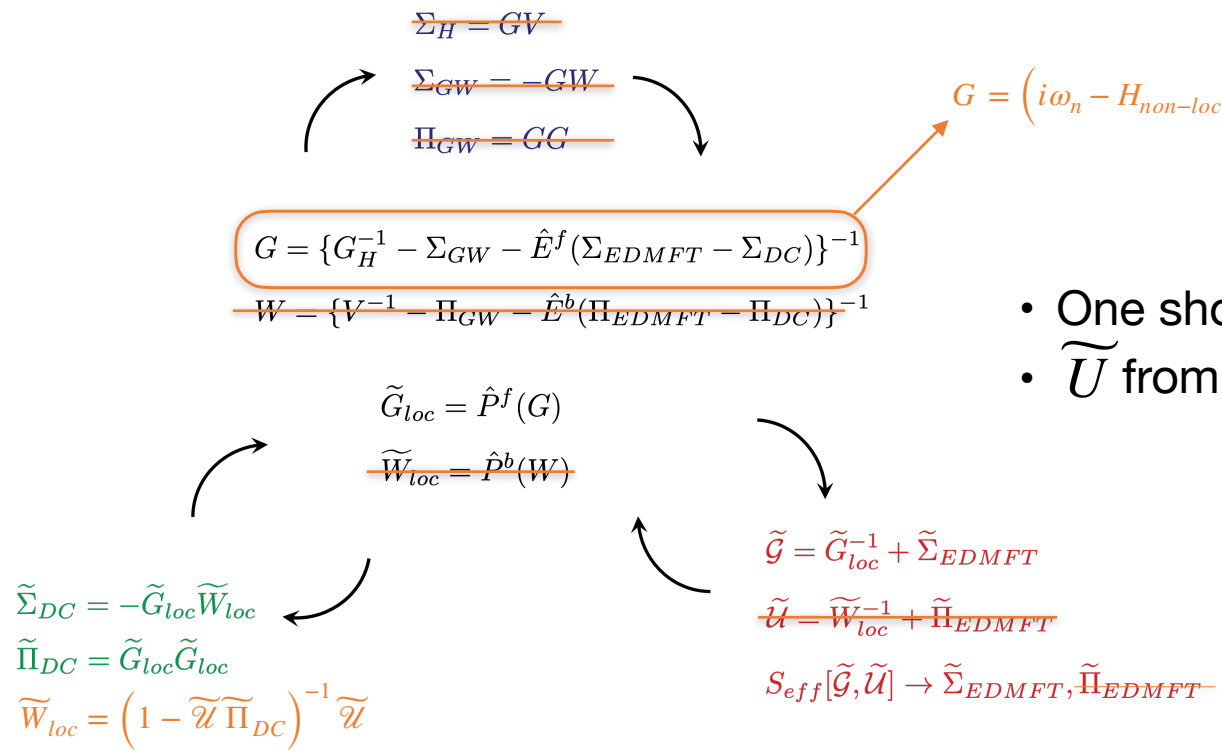


1) $H^{non-local}(\mathbf{k}) = H^{LQSGW}(\mathbf{k})$
2) Determine Z(k) by fitting ARPES spectra of LiFeAs

	$Z_m(\Gamma)$	$Z_m(M)$
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).

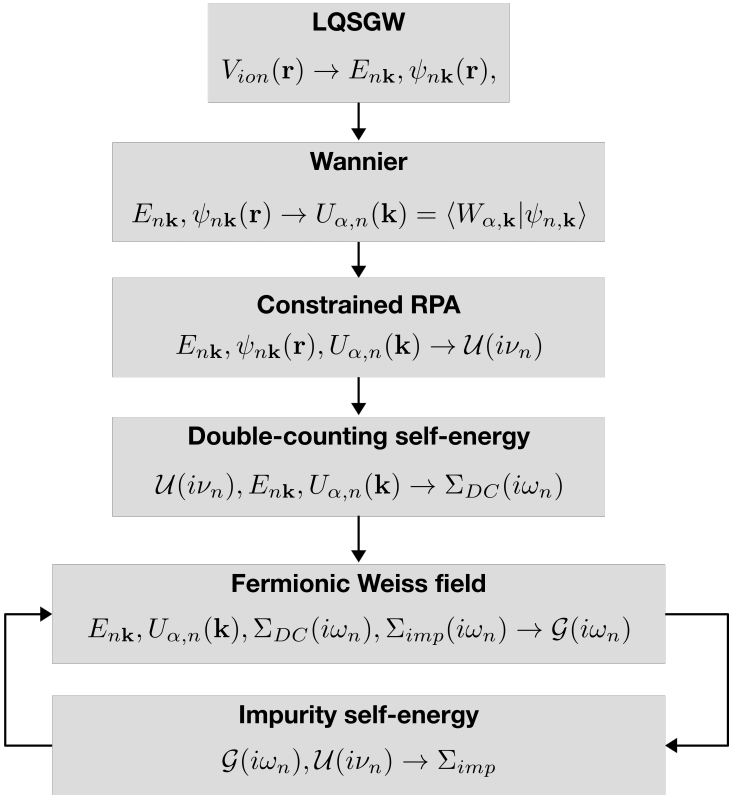
Simplification-I: partial self-consistency scheme



- One shot DMFT correction to $H_{non\text{-}local}$
- $\widetilde{U}$ from cRPA

[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).

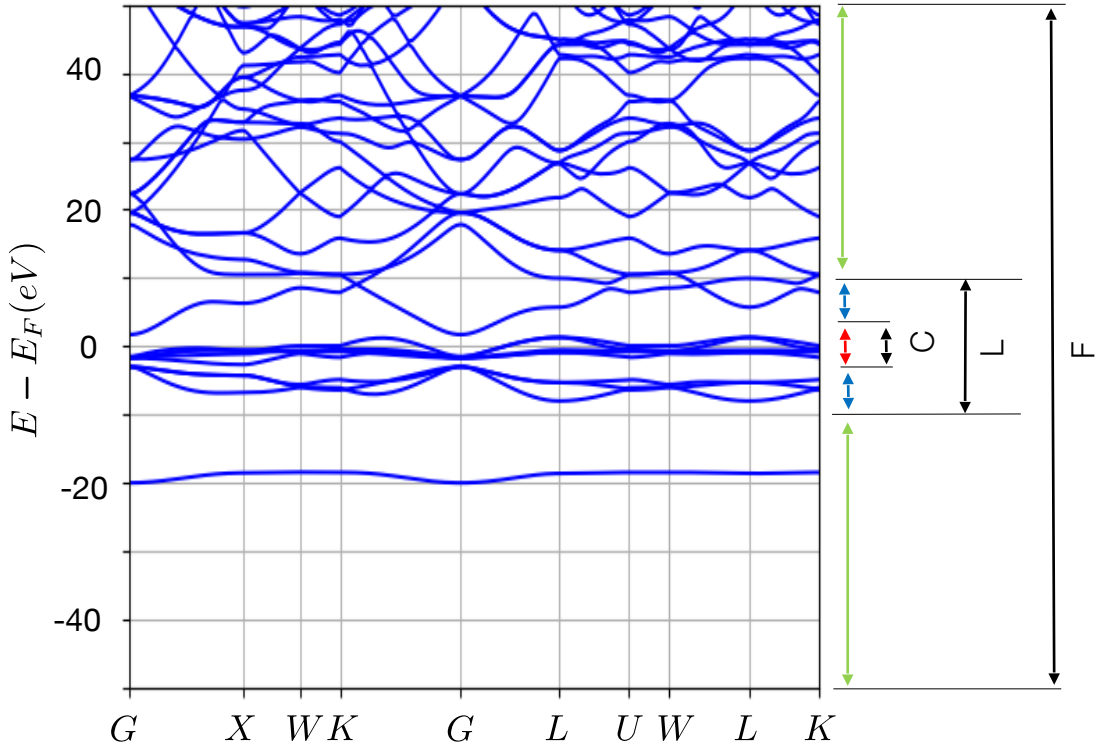
Ab initio LQSGW+DMFT



- 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



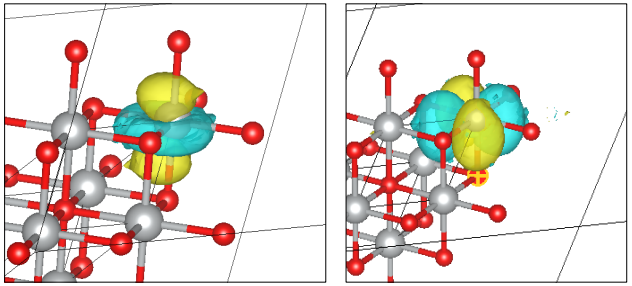
- F: full space
  - F= Tier-I $\bigoplus$ Tier-II $\bigoplus$ Tier-III
  - notation: $A\ (r, r')$

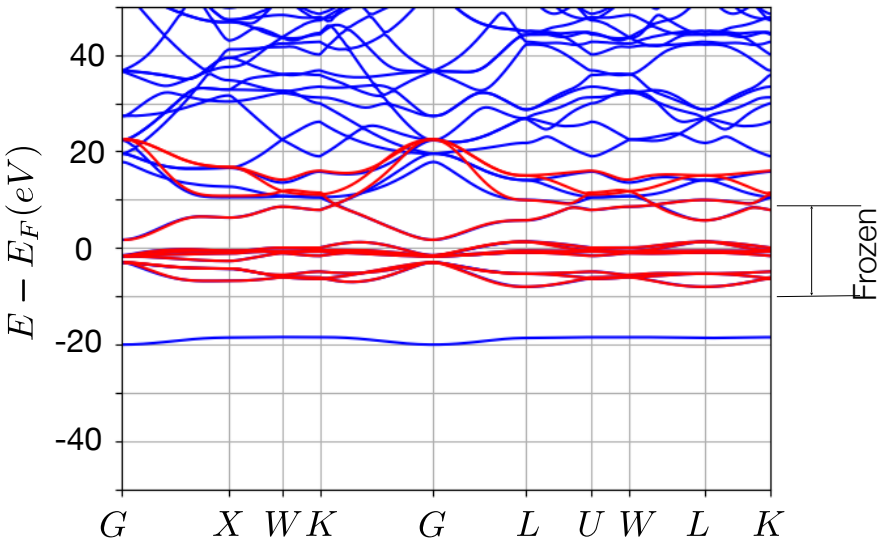
- L: Low-energy subspace defined by Wannier functions spanning an energy window ($\mathrm{E_F} \pm 10eV$)
  - L=Tier-I $\bigoplus$ Tier-II
  - notation: $\bar{A}_{ij}$, i,j=> Wannier functions

- 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

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

→ 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)

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

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

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

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

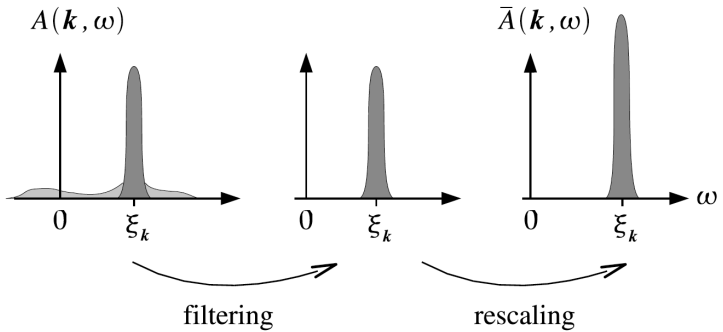
$$P_{GW}(\mathbf{k}^c, i\nu_n^c) = -\sum_{\mathbf{R}^c}\int_0^{\beta^c} d\tau G^{QP}(\mathbf{R}^c, \tau) \circ G^{QP}(-\mathbf{R}^c, -\tau) e^{-i(\mathbf{k}^c\cdot\mathbf{R}^c - \nu_n^c\tau)}$$

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

- 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: $6 \times 6 \times 6$)

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

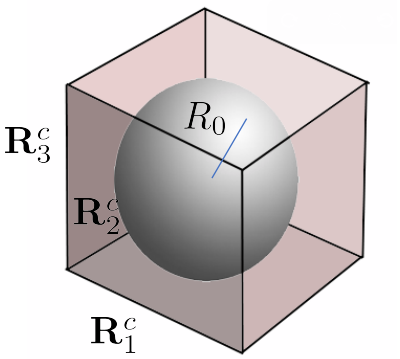
Quasiparticle Hamiltonian: $$H_{QP} = \sqrt{Z}\left(\varepsilon_0 + \Sigma(0)\right)\sqrt{Z}$$

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 $\boldsymbol{\Sigma^{dyn} = \Sigma^{imp} = \Sigma^{DC}}$

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

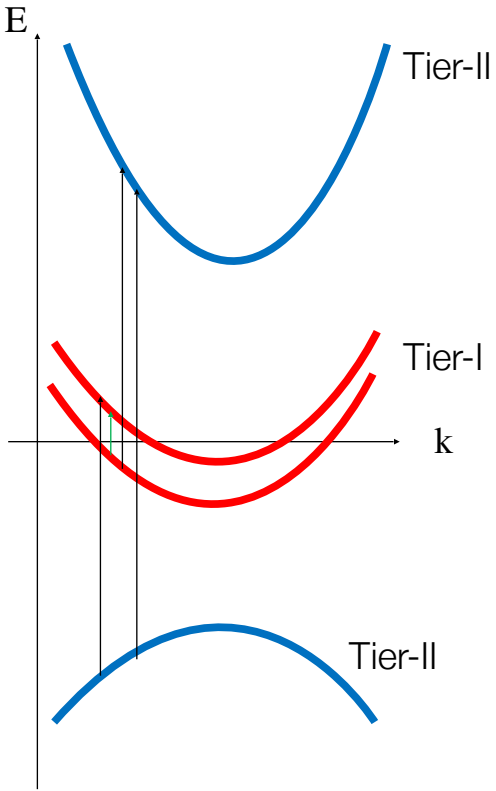
$$\widetilde{Z}_{DC}^{-1} = 1 - \left. \frac{\partial \widetilde{\Sigma}_{DC}(i\omega_n)}{\partial (i\omega_n)} \right|_{i\omega_n = 0}$$

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

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

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

Bosonic Weiss Field: constrained random phase approximation



Partially screened Coulomb interaction by neglecting transition between correlated bands

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

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

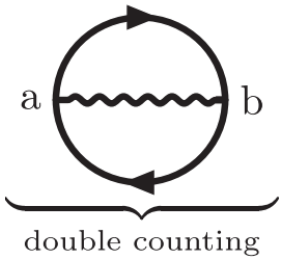
$$W_r^{-1}(\mathbf{k}^c,i\omega_n^c) = W^{-1}(\mathbf{k}^c,i\omega_n^c) + P_{QP}^{low}(\mathbf{k}^c,i\omega_n^c)$$

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

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

[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

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

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

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

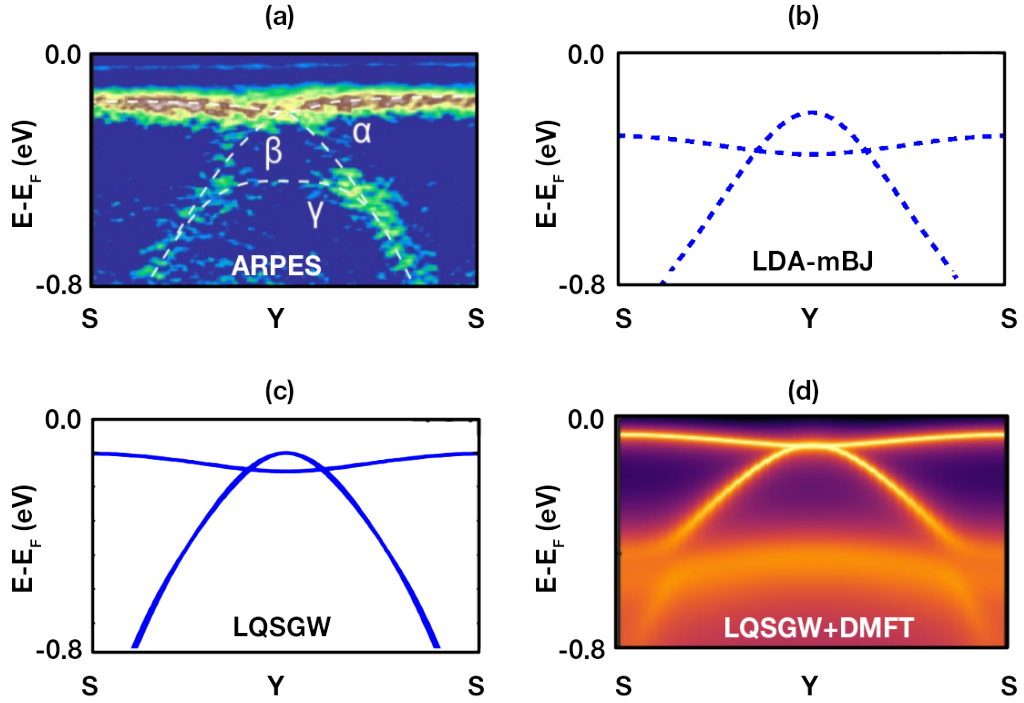
$$\longrightarrow \quad \widetilde{G}_{loc} = \mathbb{P}(G)$$

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

$$\downarrow$$

$$S_{eff}(\widetilde{\mathcal{G}}, \widetilde{\mathcal{U}}) \rightarrow \widetilde{\Sigma}_{imp} \quad \longleftarrow \quad \widetilde{\mathcal{G}}^{-1} = \widetilde{G}_{loc}^{-1} + \widetilde{\Sigma}_{imp}$$

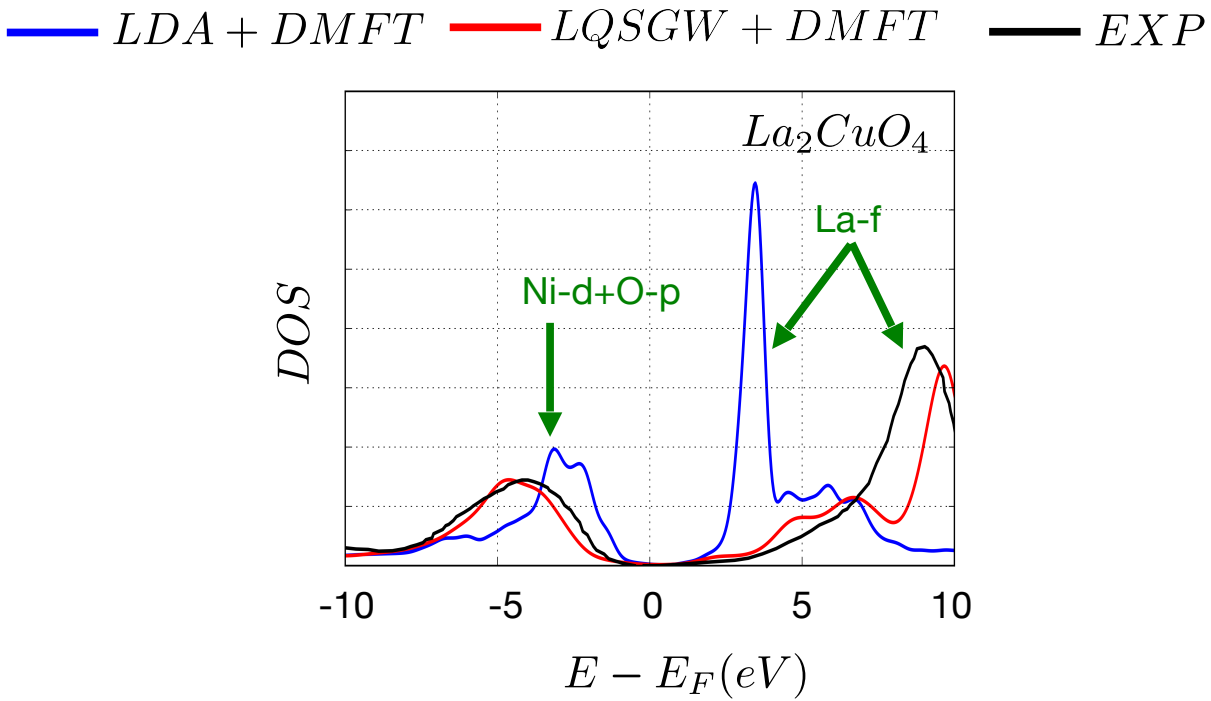
Validation: $FeSb_2$ bandstructure



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

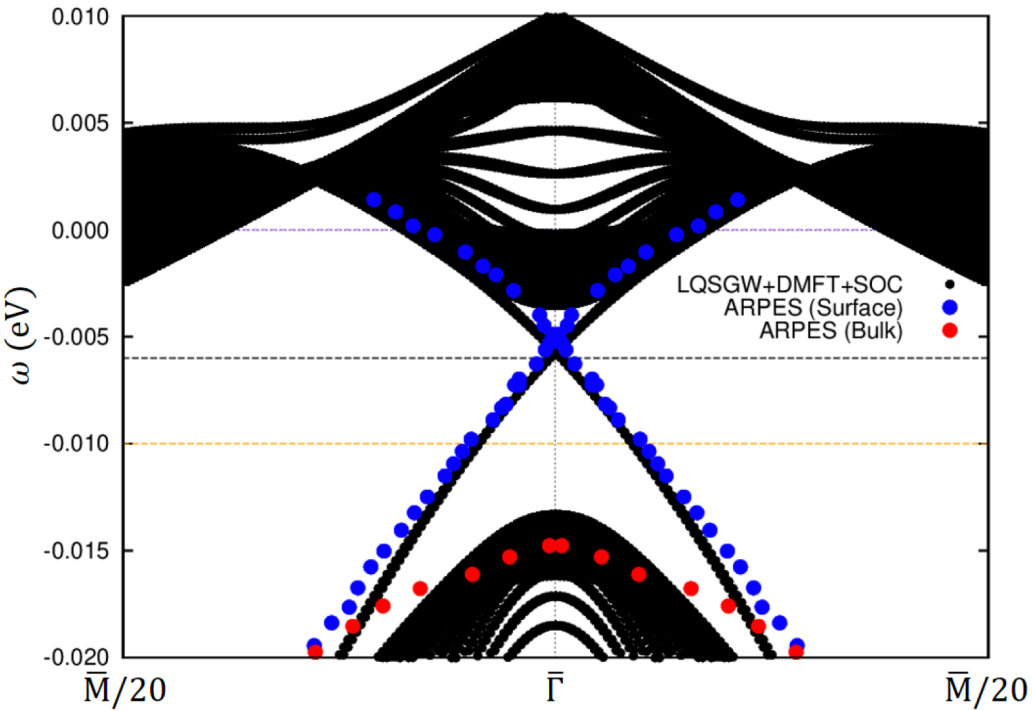
[1] A. Chikina, … S.Choi, et al., Phys. Rev. Research 2, 023190 (2020).

Validation: La2CuO4



[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).

Validation: Fe-based superconductors

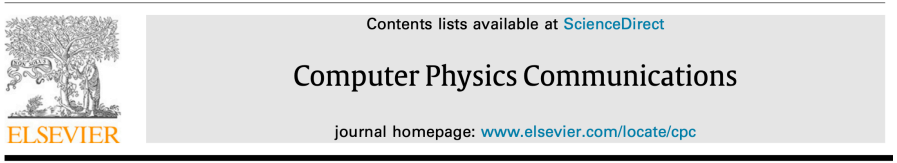


KIAS

Minjae Kim

[1] M. Kim, S. Choi, W. H. Brito, and G. Kotliar, Phys. Rev. Lett. 132, 136504 (2024).

We released open-source ab initio package for correlated quantum materials


Contents lists available at ScienceDirect

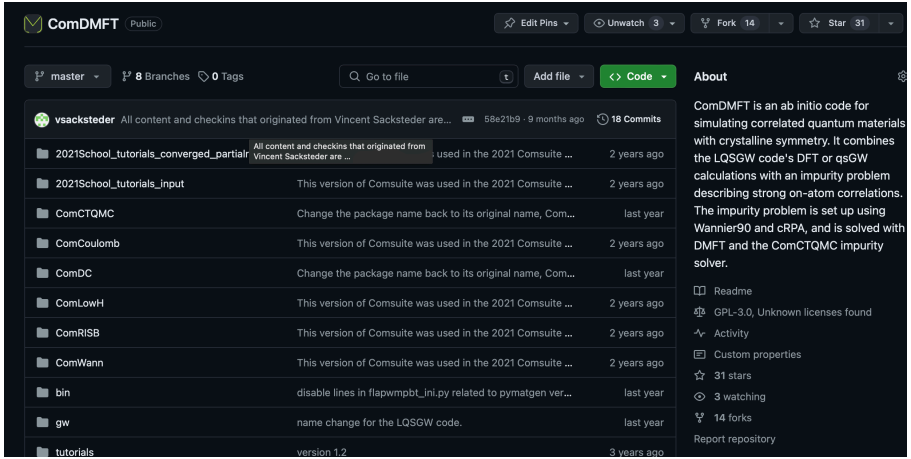
Computer Physics Communications

journal homepage: www.elsevier.com/locate/cpc

ComDMFT: A massively parallel computer package for the electronic structure of correlated-electron systems☆

Sangkook Choi [a,*], Patrick Semon [a], Byungkyun Kang [a], Andrey Kutepov [a], Gabriel Kotliar [a,b]

[a] Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, NY 11973, USA
[b] Department of Physics and Astronomy, Rutgers University, NJ 08854, USA



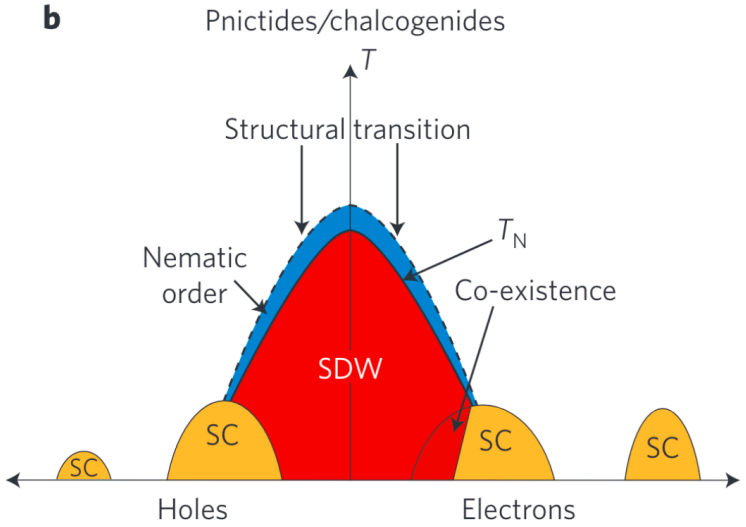
- Various flavors (DFT+DMFT and LQSGW+DMFT, tradeoff between speed and accuracy)
- GPU-accelerated
- For the GW/LDA part of the GW+DMFT/LDA+DMFT scheme, the code FlapwMBPT was used.

[1] S. Choi+, P. Semon, B. Kang, A. Kutepov, and G. Kotliar, CPC. 244, 277 (2019) [2] A. L. Kutepov, V. S. Oudovenko, and G. Kotliar, CPC 219, 407 (2017). [3] A. Kutepov, S. Y. Savrasov, and G. Kotliar, PRB 80, 041103 (2009). [4] A. Kutepov, K. Haule, S. Y. Savrasov, and G. Kotliar, PRB 85, 155129 (2012).[4] B. Kang, P. Semon, C. Melnick, G. Kotliar, and S. Choi, arXiv:2310.04613.

5. Full GW+EDMFT

Demonstration using model Hamiltonian: Charge Order and Hund Metal Physics

Charged ordered phase could be a common neighbor of a SC phase



Extended Hund-Hubbard model (3 orbital and 2 electron)

Hund metal physics

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

Charge order physics

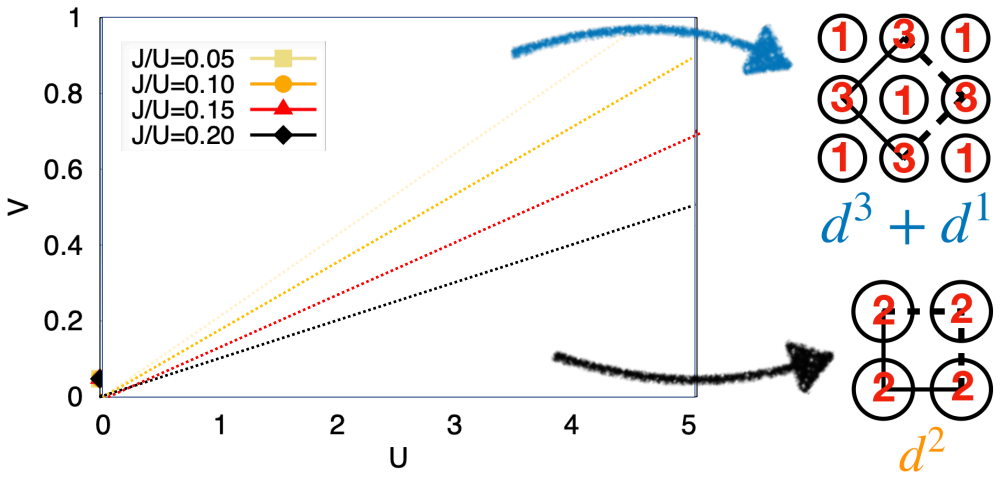
$$H_{\text{loc}} = U\sum_{i,\gamma,\sigma} n_{i\gamma\uparrow}n_{i\gamma\downarrow} + (U-2J)\sum_{i,\gamma,\gamma'}^{\gamma\neq\gamma'} n_{i\gamma\uparrow}n_{i\gamma'\downarrow} + (U-3J)\sum_{i,\gamma,\gamma',\sigma}^{\gamma<\gamma'} n_{i\gamma\sigma}n_{i\gamma'\sigma}$$

$$-J\sum_{i,\gamma,\gamma'}^{\gamma\neq\gamma'}\left(c^{\dagger}_{i\gamma\uparrow}c_{i\gamma\downarrow}c^{\dagger}_{i\gamma'\downarrow}c_{i\gamma'\uparrow} + c^{\dagger}_{i\gamma\uparrow}c^{\dagger}_{i\gamma\downarrow}c_{i\gamma'\uparrow}c_{i\gamma'\downarrow}\right).$$

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

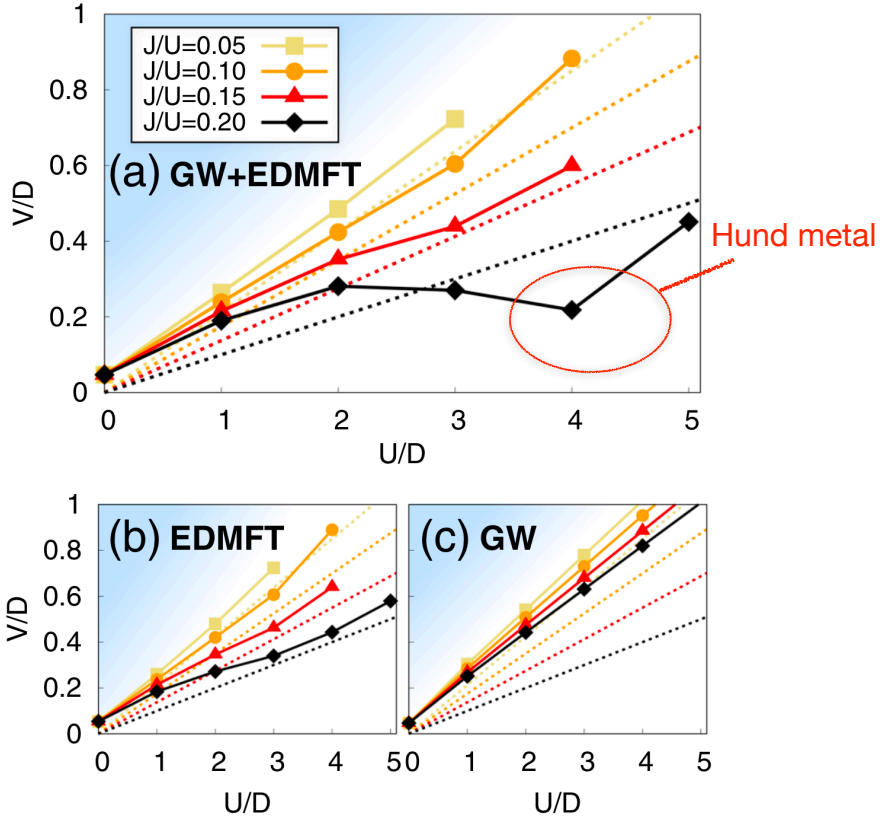
[1] S. Ryee, P. Sémon, M. J. Han, and **S. Choi**, Npj Quantum Mater. **5**, 1 (2020).

Phase boundary of H_{int}



Valence-skipping charge order
- negative effective U
- Implication for unconventional superconductor pairing

Phase boundary of full Hamiltonian within GW+EDMFT



Enhancement of the charge-order instability due to Hund metal physics

only observable within GW+EDMFT

Importance of treating local and non-local correlation on equal footing.

Charge-order=> nonlocal correlation
Hund metal=> local correlation

Three different self-consistent loops in GW+EDMFT

$$\Sigma_H = GV$$
$$\Sigma_{GW} = -GW$$
$$\Pi_{GW} = GG$$

$$G = \{G_H^{-1} - \Sigma_{GW} - \hat{E}^f(\Sigma_{EDMFT} - \Sigma_{DC})\}^{-1}$$
$$W = \{V^{-1} - \Pi_{GW} - \hat{E}^b(\Pi_{EDMFT} - \Pi_{DC})\}^{-1}$$

$$\widetilde{G}_{loc} = \hat{P}^f(G)$$
$$\widetilde{W}_{loc} = \hat{P}^b(W)$$

$$\widetilde{\Sigma}_{DC} = -\widetilde{G}_{loc}\widetilde{W}_{loc}$$
$$\widetilde{\Pi}_{DC} = \widetilde{G}_{loc}\widetilde{G}_{loc}$$

$$\widetilde{\mathcal{G}} = \widetilde{G}_{loc}^{-1} + \widetilde{\Sigma}_{EDMFT}$$
$$\widetilde{\mathcal{U}} = \widetilde{W}_{loc}^{-1} + \widetilde{\Pi}_{EDMFT}$$
$$S_{eff}[\widetilde{\mathcal{G}}, \widetilde{\mathcal{U}}] \rightarrow \widetilde{\Sigma}_{EDMFT}, \widetilde{\Pi}_{EDMFT}$$

Fully self-consistent GW+EDMFT is here, finally!!!

ComDMFT v.2.0: Fully Self-Consistent *ab initio* GW+EDMFT for the Electronic Structure of Correlated Quantum Materials

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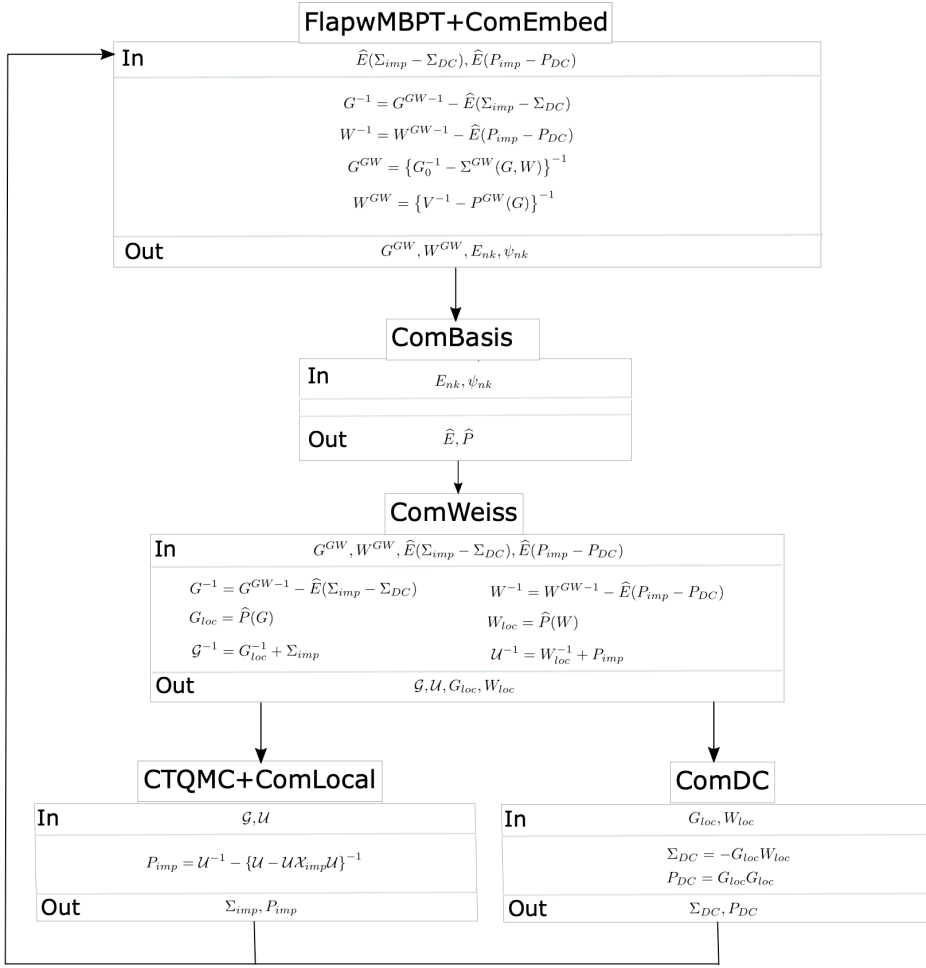
[1] B. Kang, P. Semon, C. Melnick, G. Kotliar, and S. Choi, ComDMFT v.2.0: Fully Self-Consistent Ab Initio GW+EDMFT for the Electronic Structure of Correlated Quantum Materials, arXiv:2310.04613.

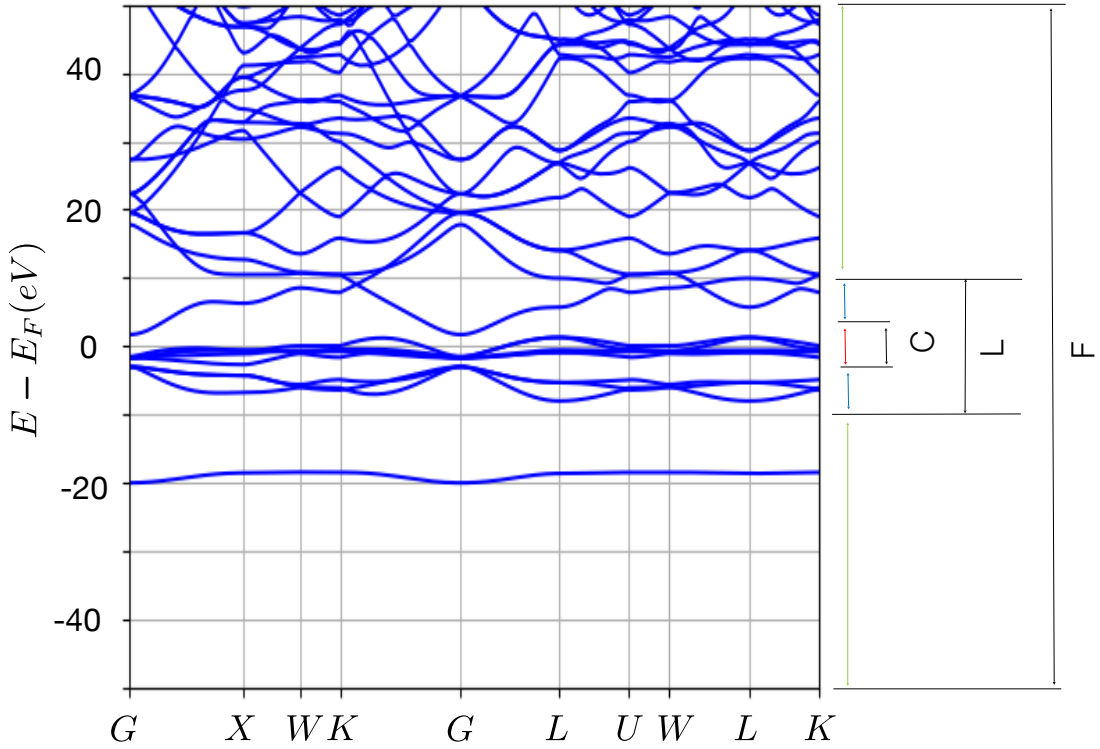
Full GW+EDMFT

- Full self-consistency
- No adjustable parameters
- G and W interpolation
- Wannier functions and their orthonormalized product basis
- Causal optimization on $\widetilde{P}_{imp}$ and $\widetilde{U}$
- Built on top of FlapwMBPT, Wannier90 and ComCTQMC



[1] A. L. Kutepov, V. S. Oudovenko, and G. Kotliar, Comp. Phy. Comm. **219**, 407 (2017).
[2] C. Melnick, P. Sémon, K. Yu, N. D'Imperio, A.-M. Tremblay, and G. Kotliar, Comp. Phys. Comm. **267**, 108075 (2021).
[3] M. Han and H. J. Choi, Phys. Rev. B **104**, 115112 (2021).

Hilbert space and its subspaces for Full GW+EDMFT

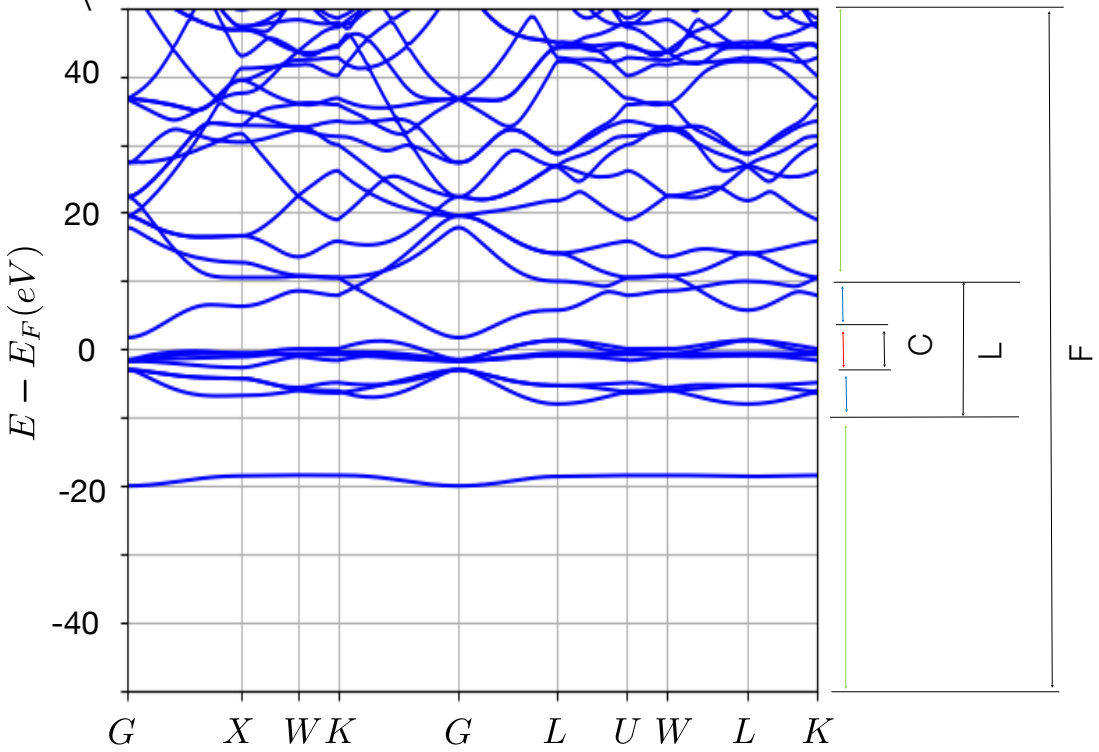


- F: full space
  - F= Tier-I ⊕ Tier-II ⊕ Tier-III
  - notation: $A\ (r, r')$
- L: Low-energy subspace defined by Wannier function spanning an energy window ($E_F \pm 10eV$)
  - L=Tier-I ⊕ Tier-II
  - notation: $\bar{A}_{ij}$, i,j=> Wannier functions
- C: correlated subspace
  - C=Tier-I
  - notation: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

G_{GW} and W_{GW} in coarse k-grid

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

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

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

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

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

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

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

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

- Modification on FlapwMBPT (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: $6 \times 6 \times 6$)

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

$\leftarrow$

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

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

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

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

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

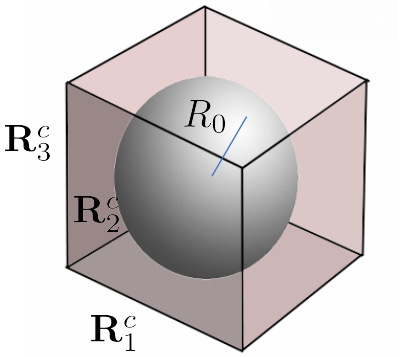
$$X_{\tau_1,\tau_2;I} = \frac{1}{\sqrt{F_I}} V_{\tau_1,\tau_2;I}$$

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

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

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

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

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

For screened Coulomb interaction

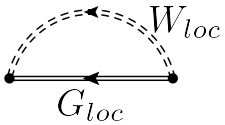
$$\overline{W}_{GW}(\mathbf{k}^c, i\nu_n^c) = \overline{b}^{\dagger}(\mathbf{k}^c) W_{GW}(\mathbf{k}^c, i\nu_n^c) \overline{b}(\mathbf{k}^c)$$

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

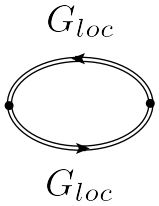
$$\overline{W}(\mathbf{k}^f, i\nu_n^f) = \left(\overline{W}^{-1}_{GW}(\mathbf{k}^f, i\nu_n^f) + \widetilde{b}(\mathbf{k}^c) \left(\widetilde{P}_{imp}(i\nu_n^f) - \widetilde{P}_{DC}(i\nu_n^f) \right) \widetilde{b}^{\dagger}(\mathbf{k}^c) \right)^{-1}$$

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

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

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

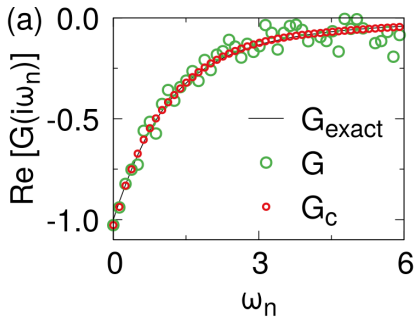
$$\widetilde{W}_{imp}(i\nu_n^f) = \widetilde{\mathcal{U}}(i\nu_n^f) - \widetilde{\mathcal{U}}(i\nu_n^f)\widetilde{\chi}_{imp}(i\nu_n^f)\widetilde{\mathcal{U}}(i\nu_n^f)$$

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

- 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

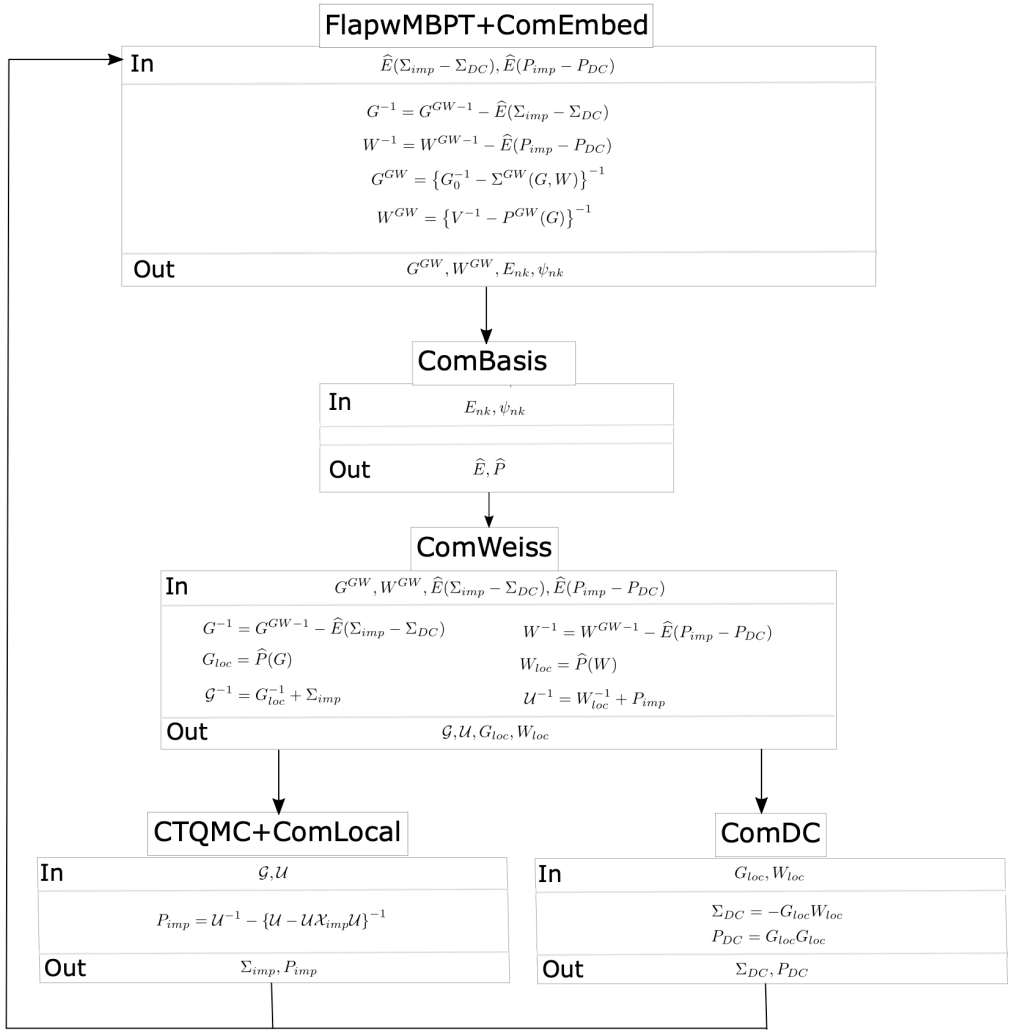
$$\widetilde{\Sigma}_{imp}(i\omega_n^f) \to \widetilde{\Sigma}_{imp}(i\omega_n^c) \qquad \widetilde{\Sigma}_{DC}(i\omega_n^f) \to \widetilde{\Sigma}_{DC}(i\omega_n^c)$$
$$\widetilde{P}_{imp}(i\omega_n^f) \to \widetilde{P}_{imp}(i\omega_n^c) \qquad \widetilde{P}_{DC}(i\omega_n^f) \to \widetilde{P}_{DC}(i\omega_n^c)$$

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

$$P(\mathbf{k}^c, i\nu_n^c) = P_{GW}(\mathbf{k}^c, i\nu_n^c) + \mathbb{E}\left(\widetilde{P}_{imp}(i\nu_n^c) - \widetilde{P}_{DC}(i\nu_n^c)\right)$$
$$P_{GW}(\mathbf{k}^c, i\nu_n^c) = -\sum_{\mathbf{R}^c} \int_0^{\beta^c} d\tau G(\mathbf{R}^c, \tau) \circ G(-\mathbf{R}^c, -\tau) e^{-i(\mathbf{k}^c \cdot \mathbf{R}^c - \nu_n^c \tau)}$$

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

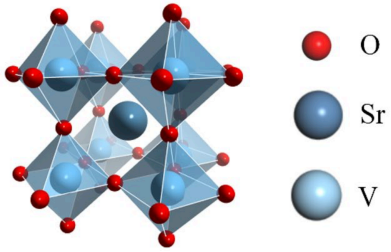
$$\Sigma_{GW}(\mathbf{k}^c, i\omega_n^c) = -\sum_{\mathbf{R}^c} \int_0^{\beta^c} d\tau G(\mathbf{R}^c, \tau) \circ W(-\mathbf{R}^c, -\tau) e^{-i(\mathbf{k}^c \cdot \mathbf{R}^c - \omega_n^c \tau)}$$
$$\Sigma(\mathbf{k}^c, i\omega_n^c) = \Sigma_{GW}(\mathbf{k}^c, i\omega_n^c) + \mathbb{E}\left(\widetilde{\Sigma}_{imp}(i\omega_n^c) - \widetilde{\Sigma}_{DC}(i\omega_n^c)\right)$$


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}

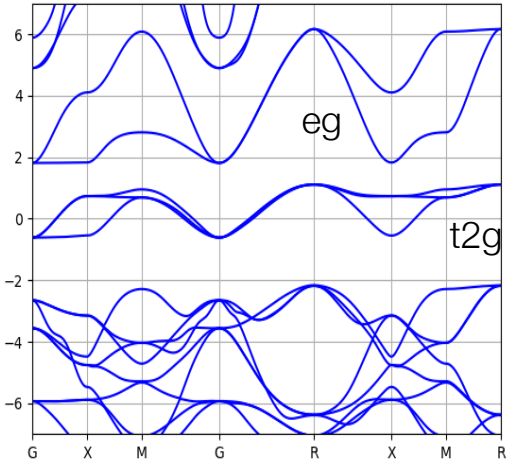
Validation-SrVO3

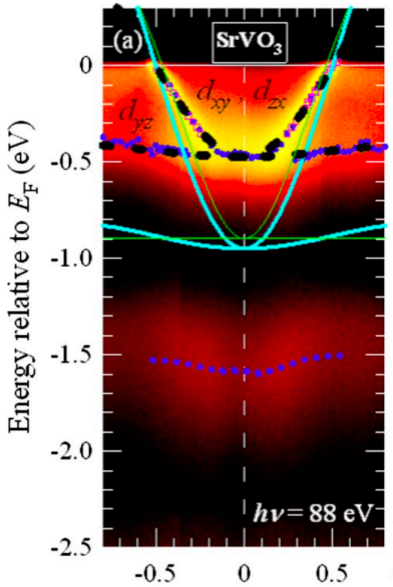
T: 1000K
Correlated space: V-3d
Low-energy space: $E_F \pm 10$ eV (20 orbitals)
K-grid: $10 \times 10 \times 10$

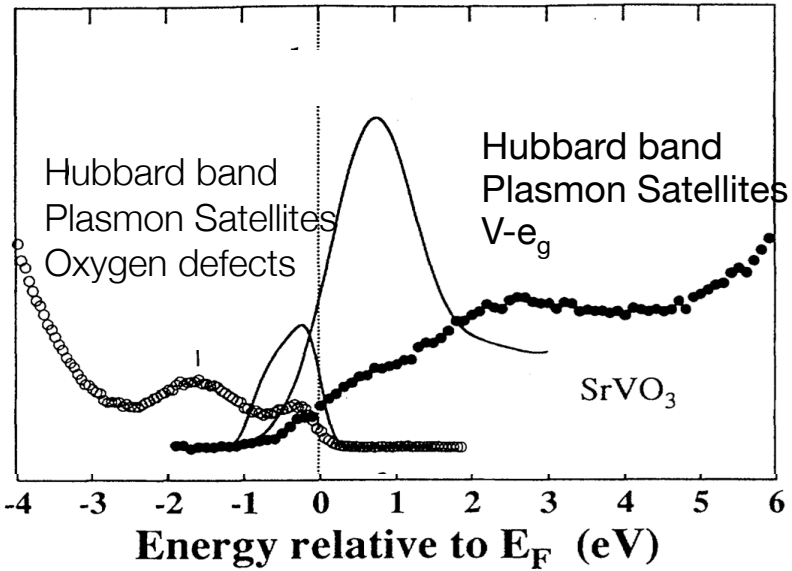
SrVO3, a classical test material for electron correlation



$$\frac{\gamma}{\gamma^{LDA}} = 2$$

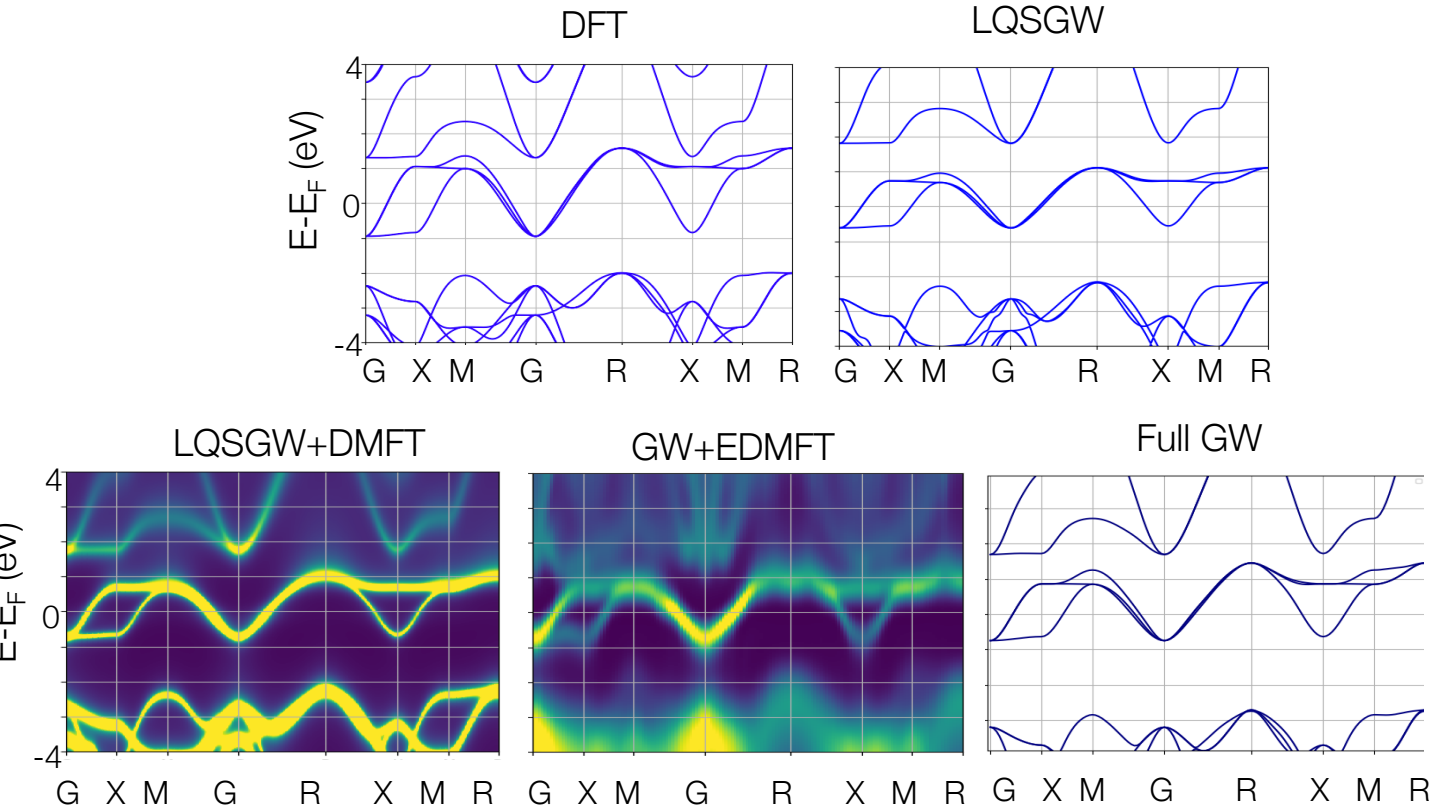


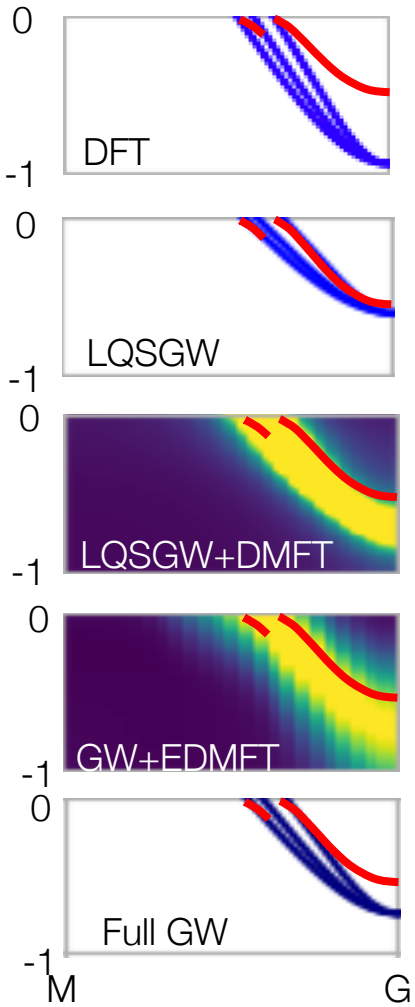




[1] Europhys. Lett. 100, 67001 (2012). [2] Phys. Rev. B 90, 165138 (2014). [3] Phys. Rev. B 94, 201106(R) (2016). [4] Phys. Rev. Mater. 1, 043803 (2017). [5] Phys. Rev. B 52, 13711 (1995). [6] Phys. Rev. Lett. 93, 156402 (2004). [7] Phys. Rev. B 82, 085119 (2010). [8] Phys. Rev. Lett. 92, 176403 (2004). [9] Phys. Rev. B 73, 155112 (2006). [10] Phys. Rev. B 74, 125120 (2006). [11] Phys. Rev. B 94, 241110 (2016). [12] Phys. Rev. B 88, 235110 (2013). [13] Phys. Rev. B 87, 155147 (2013). [14] Phys. Rev. Research 2, 013191 (2020).

Spectral Function



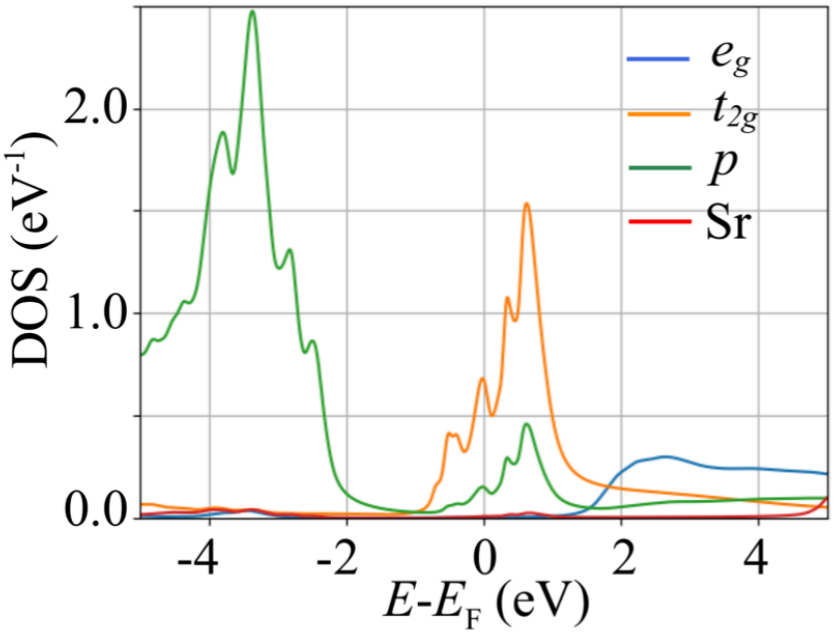


Group velocity agrees with exp. Bandwidth is overestimated.
Full GW+EDMFT shows almost identical spectra to full GW

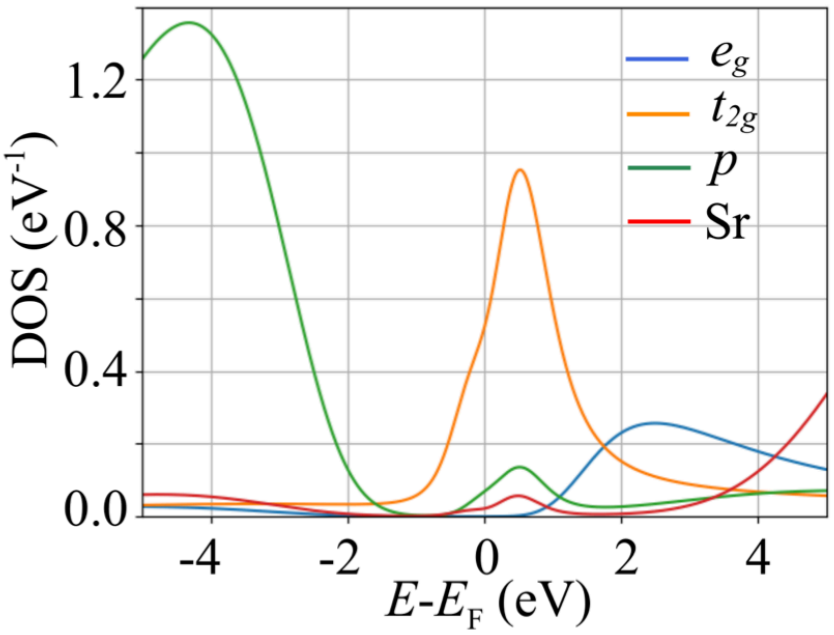
[1] T. Mitsuhashi, M. Minohara, R. Yukawa, M. Kitamura, K. Horiba, M. Kobayashi, and H. Kumigashira, Phys. Rev. B 94, 125148 (2016).

PDOS

LQSGW+DMFT



GW+EDMFT

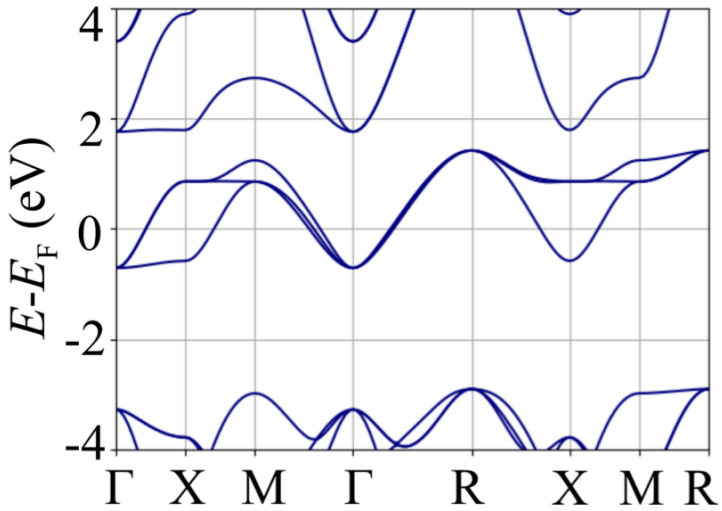


LQSGW+DMFT and GW+EDMFT show quantitatively similar results

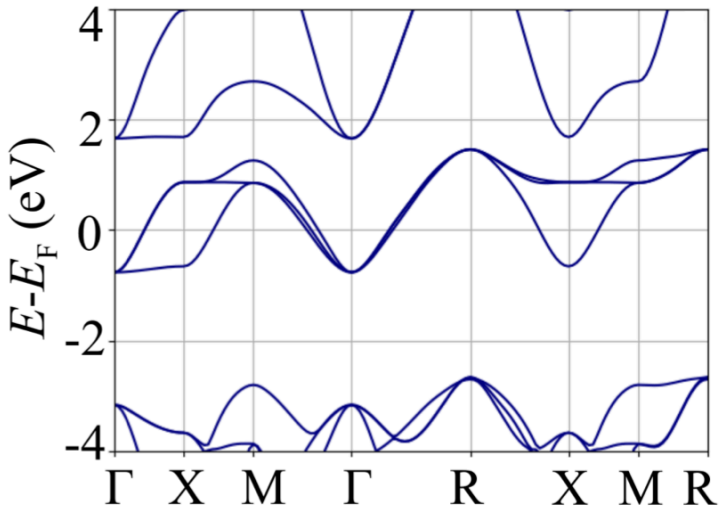
Within GW+EDMFT, no sign of low-energy t_{2g} subpeak and no sign of high-energy t_{2g} subpeak

Effect of GW self-consistency

GW band at 1st iteration



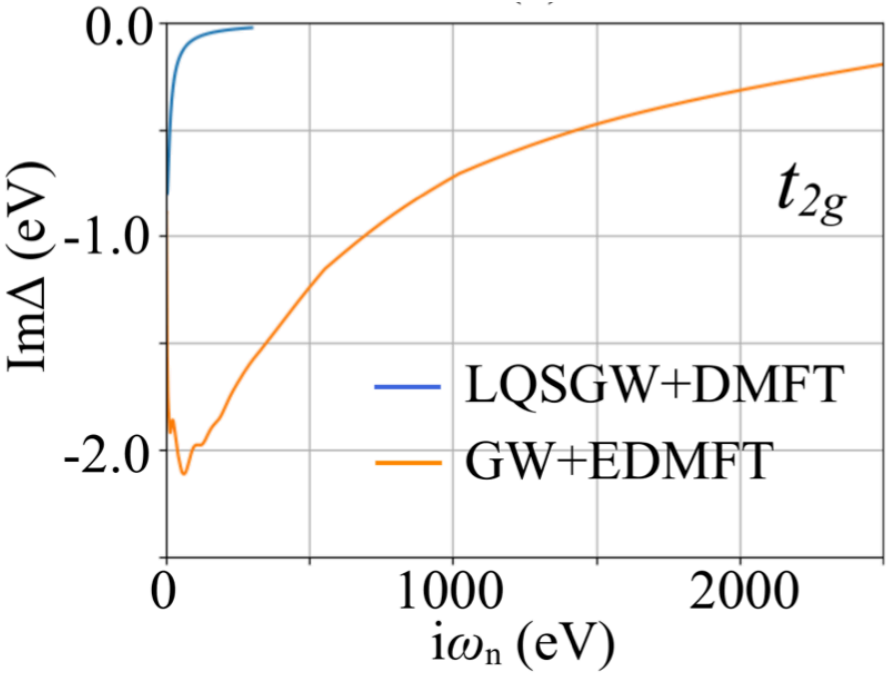
GW band at the last iteration

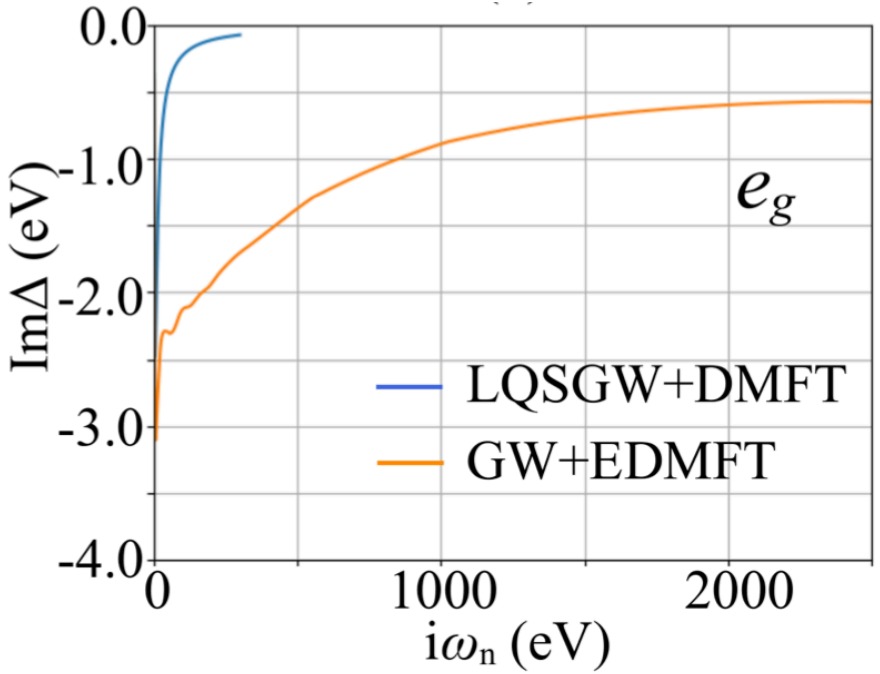


$$H^{QP}(\mathbf{k}^c) = Z^{1/2}(\mathbf{k}^c)\left(H_0(\mathbf{k}^c) + \Sigma_{GW}(\mathbf{k}^c, \omega = 0)\right) Z^{1/2}(\mathbf{k}^c)$$

$\boldsymbol{G^{GW}}$ didn't change much from the first iteration=> The effect of GW self-consistency is small

Hybridization functions

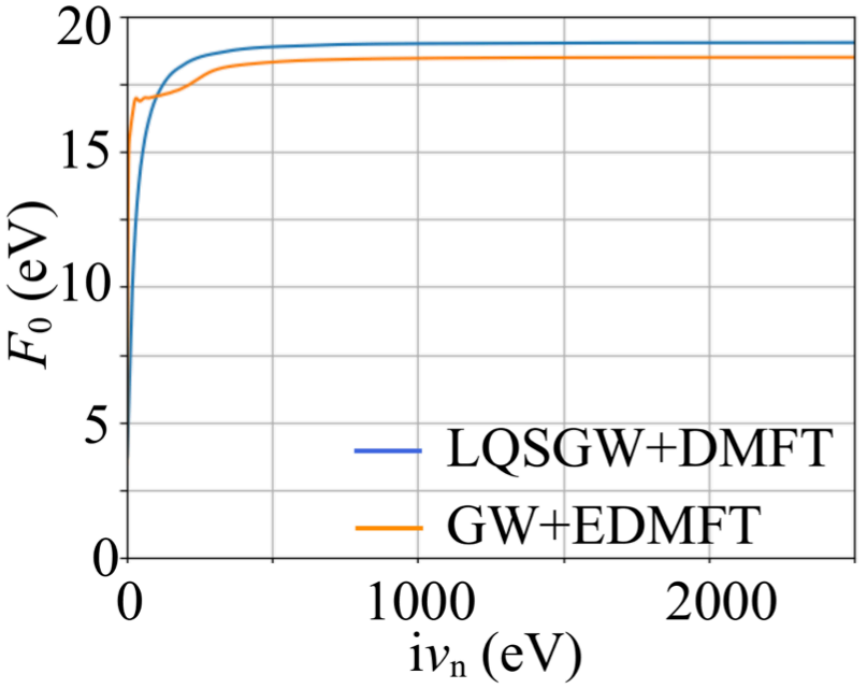


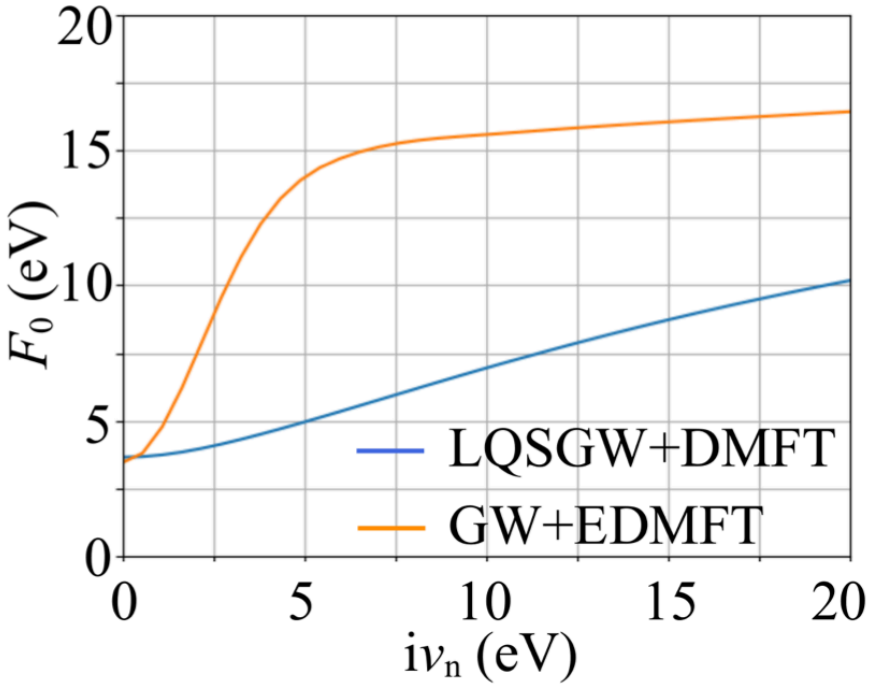


Within GW+EDMFT, Δ is nonzero until $\mathrm{E} > 2.5\mathrm{KeV}$.

In contrast, Δ within LQSGW+DMFT goes to 0 at ~200eV

Bosonic Weiss field

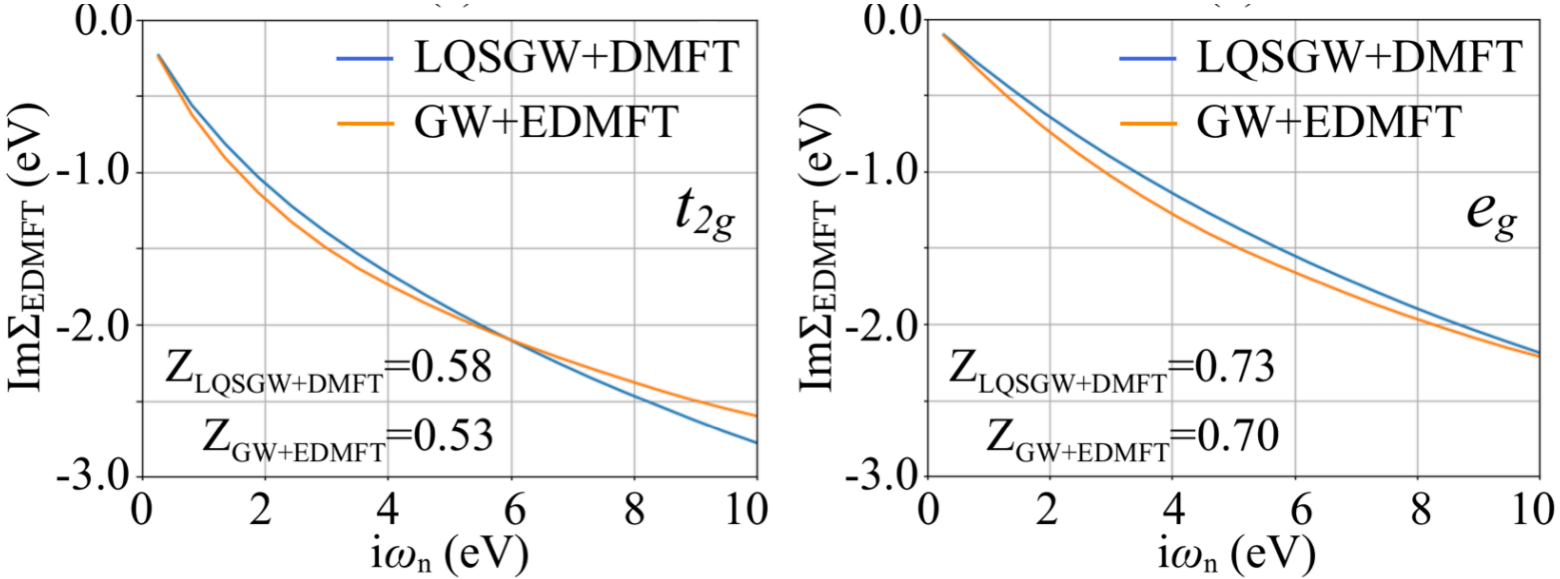




Within GW+EDMFT, $\mathrm{U}\left(\mathrm{i}\omega_n\right)$ reaches to bare value at ~ 10eV.

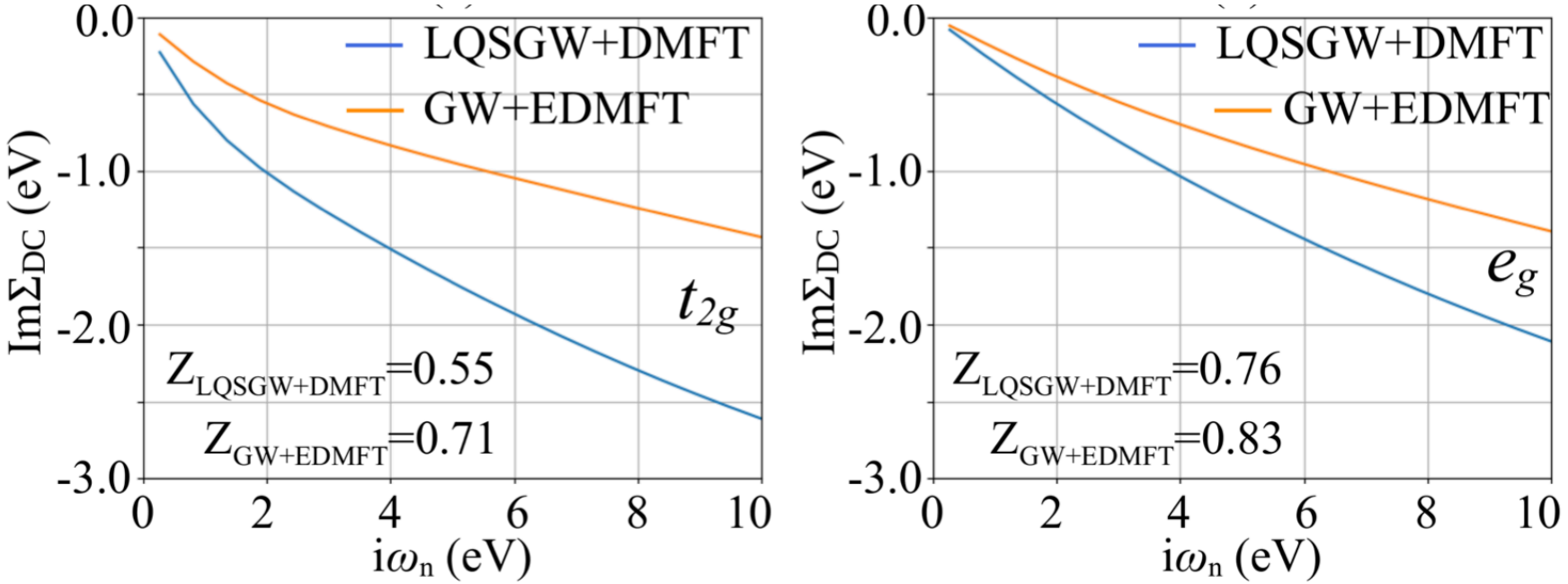
In contrast, $\mathrm{U}\left(\mathrm{i}\omega_n\right)$ within cRPA@LQSGW increases much slowly.

Impurity self-energy



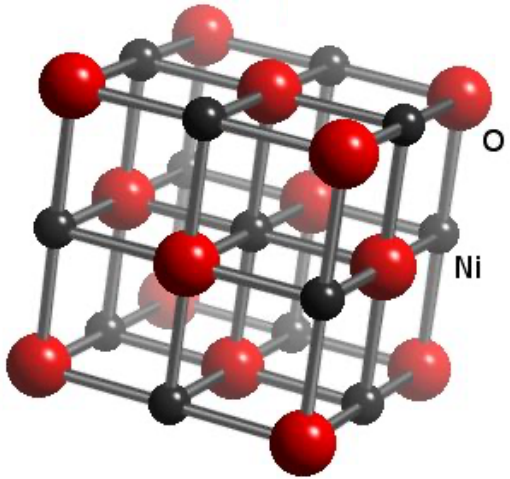
- LQSGW+DMFT as well as GW+EDMFT shows Z=0.5 for t2g orbitals, consistent with Exp
- Despite the discrepancy in Deltal and U between the two methods, their Z factor is surprisingly similar.

Double-counting self-energy

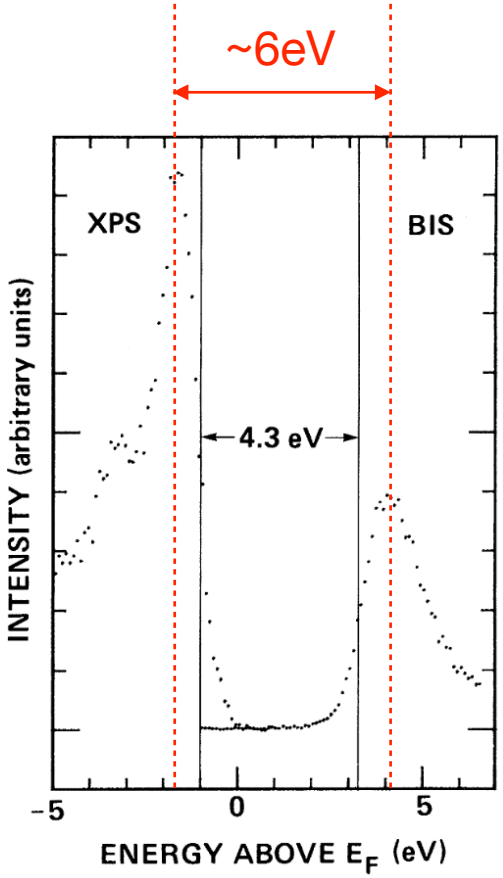


- Within LQSGW+DMFT as well as GW+EDMFT shows $Z_{imp} \sim Z_{DC}$, implying $\Sigma_{imp} \simeq \Sigma_{DC}$

Validation-II: charge transfer insulator NiO

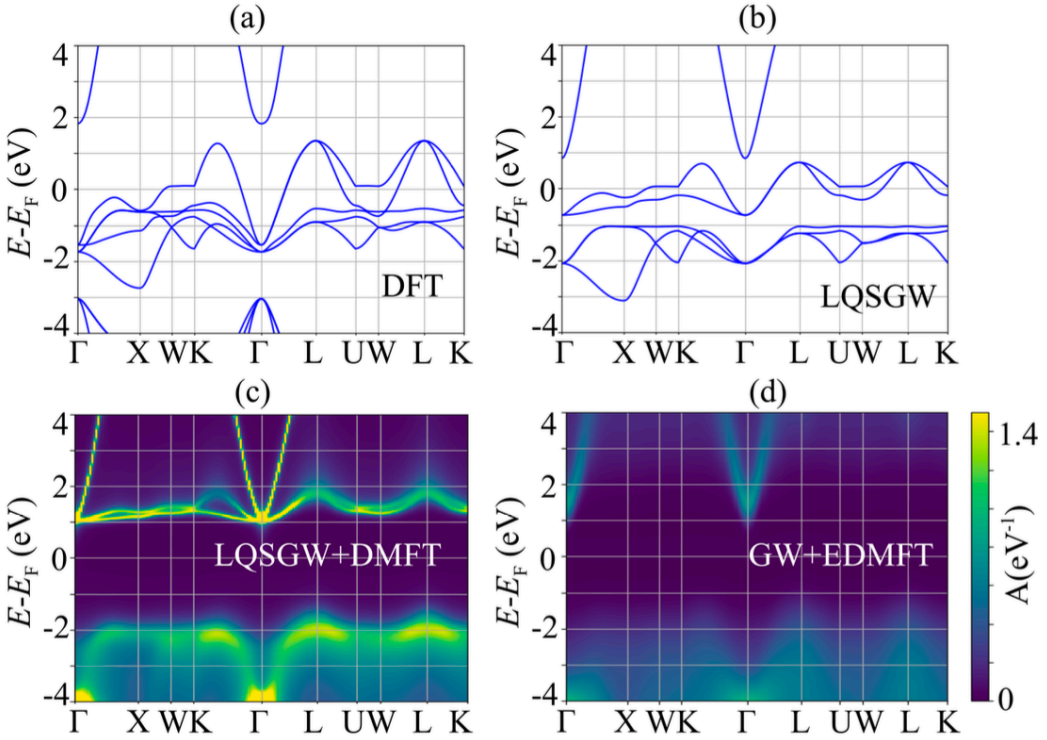


- Archetypical charge-transfer insulator
- Correlated orbitals: 5 Ni-d orbitals
- Simulation temperature: 1000K



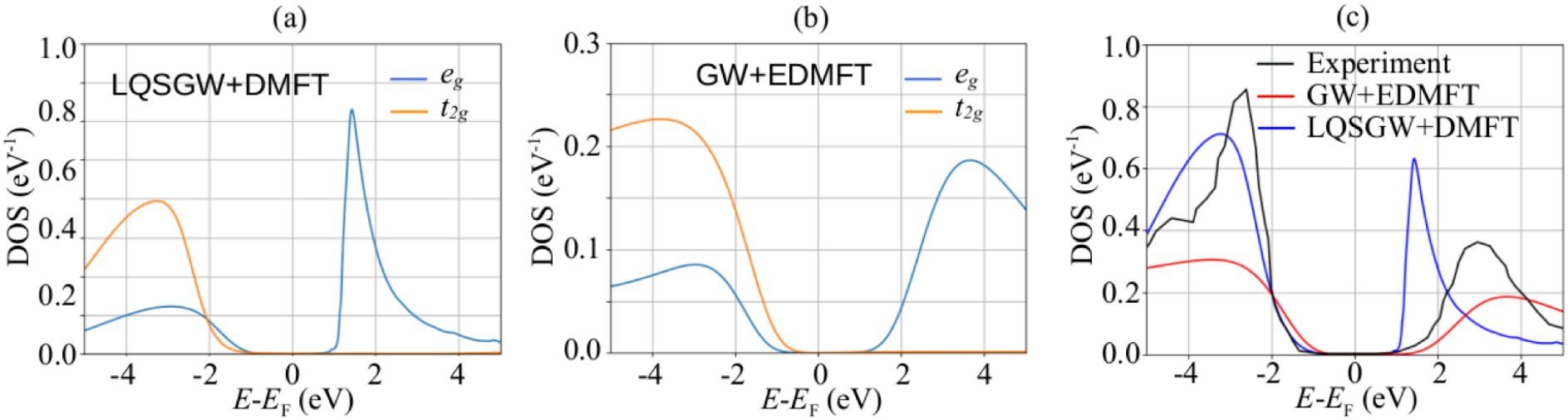
[1] G. A. Sawatzky and J. W. Allen, Phys. Rev. Lett. **53**, 2339 (1984).

Validation-I: spectral functions



- Charge transfer gap opening

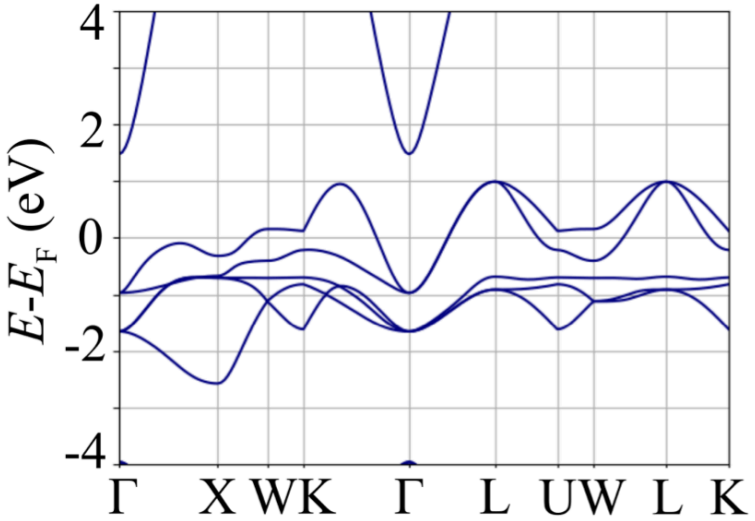
Validation-I: PDOS



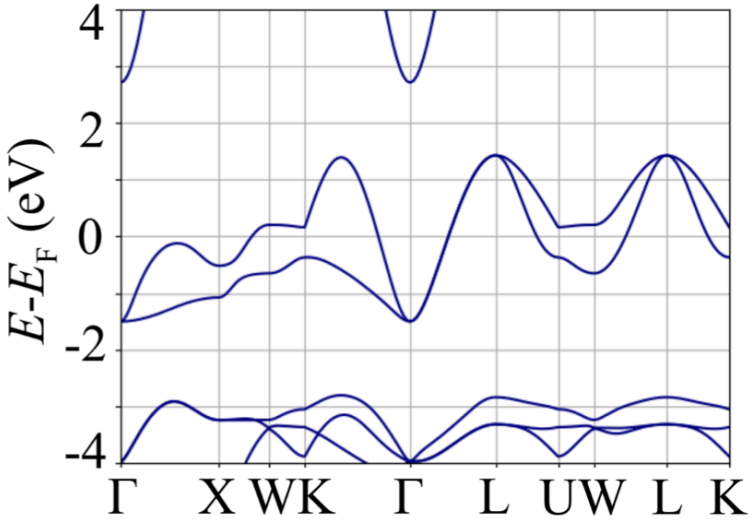
- Peak separation of ~6eV
- Mott gap

Effect of GW self-consistency

GW band at 1st iteration



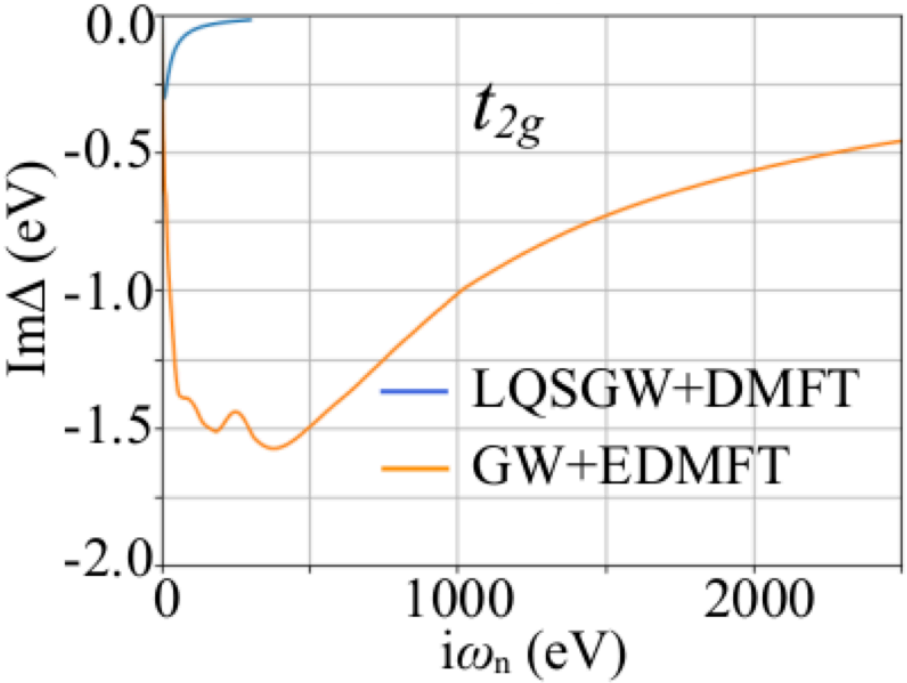
GW band at the last iteration

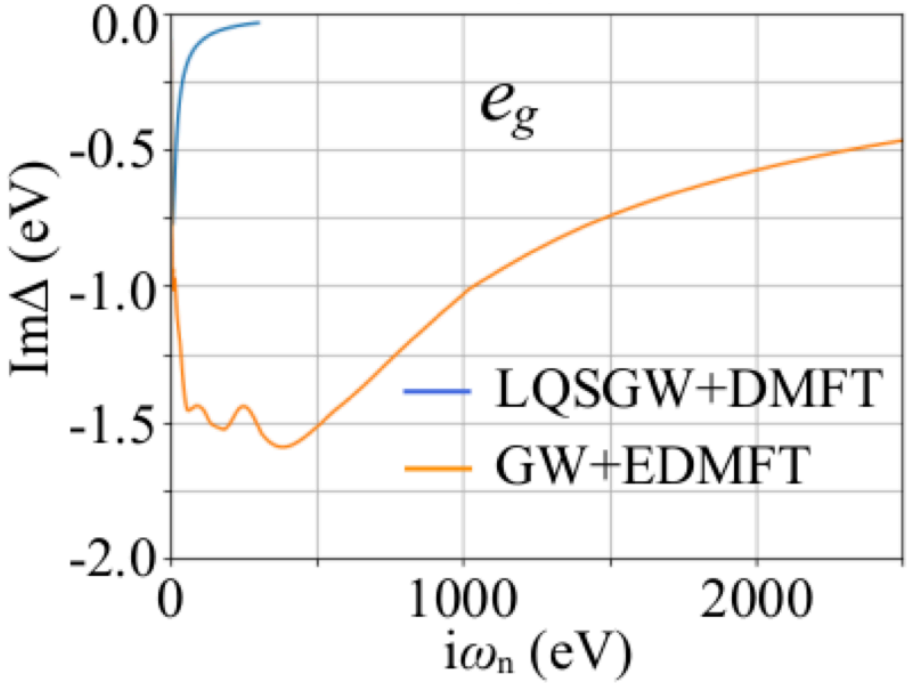


$$H^{QP}(\mathbf{k}^c) = Z^{1/2}(\mathbf{k}^c)\left(H_0(\mathbf{k}^c) + \Sigma_{GW}(\mathbf{k}^c, \omega = 0)\right) Z^{1/2}(\mathbf{k}^c)$$

$\boldsymbol{G^{GW}}$ has been changed substantially

Hybridization functions





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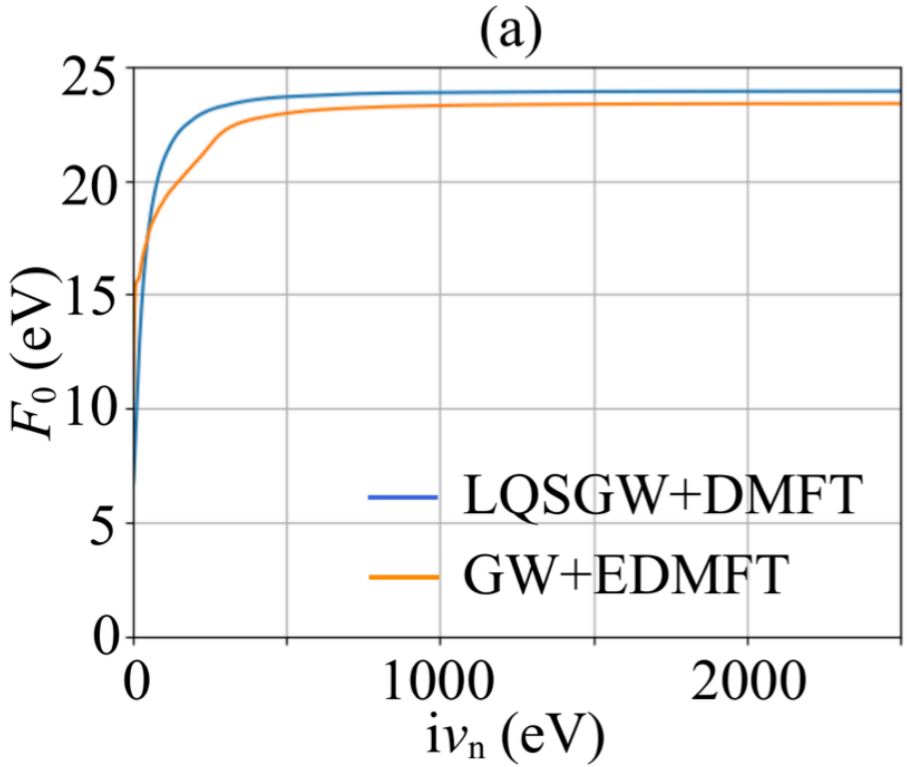
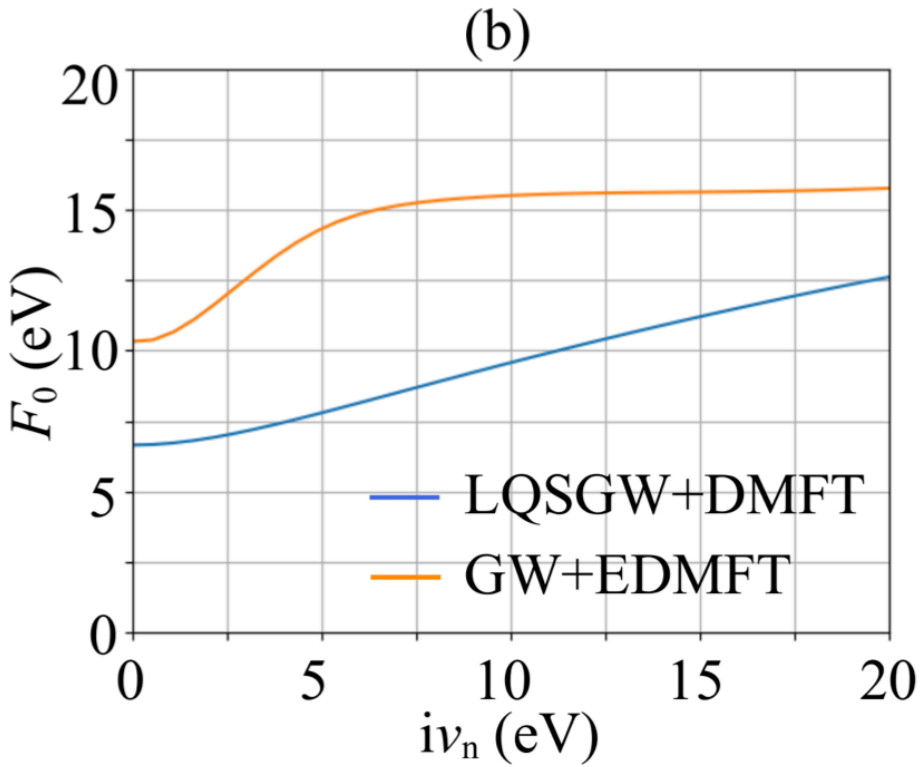
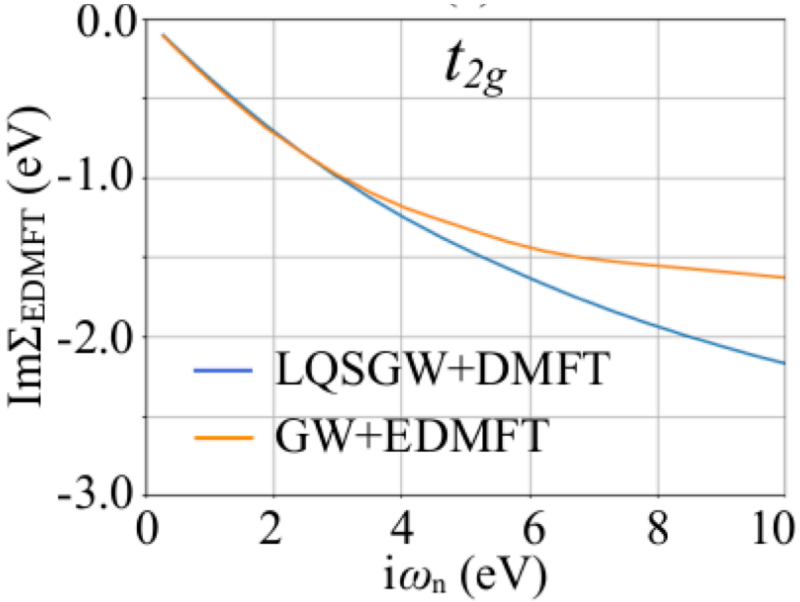
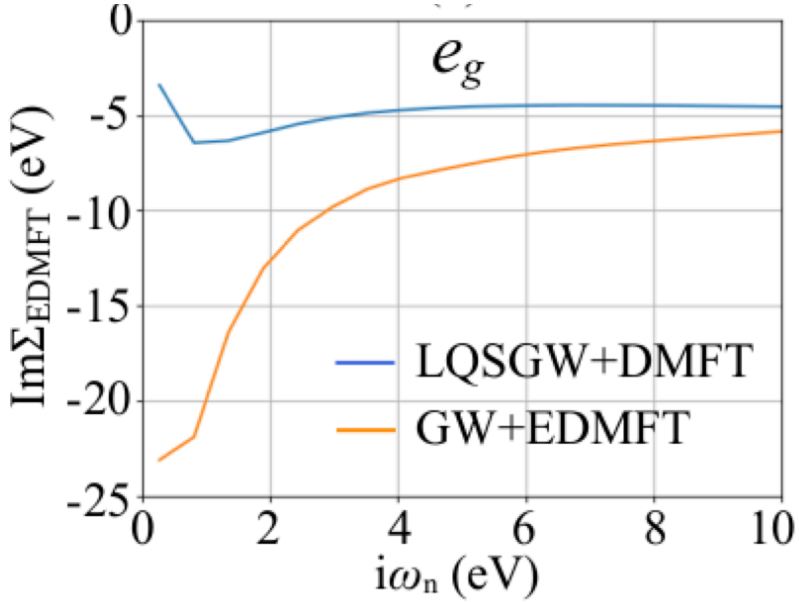




Within GW+EDMFT, $\mathrm{U}(i\omega_n)$ changes more rapidly

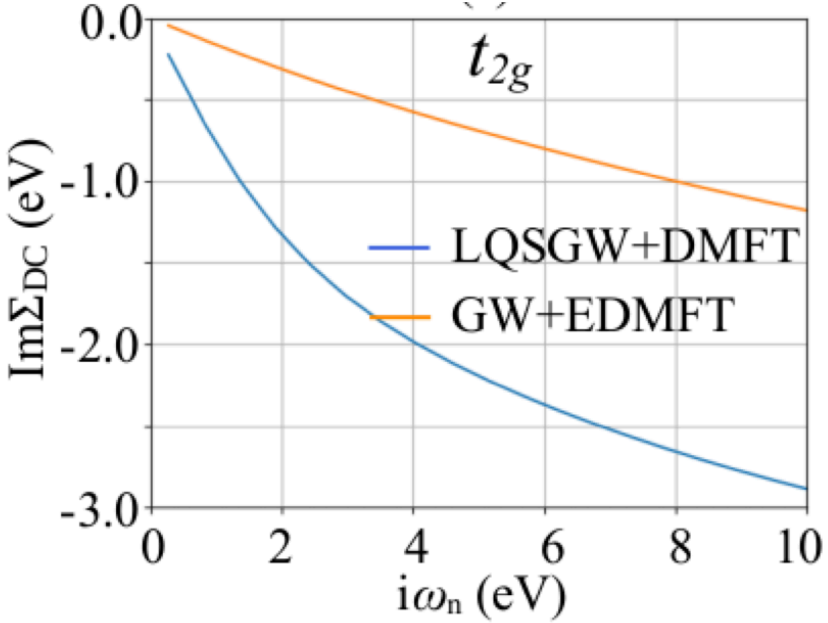
Impurity self-energy

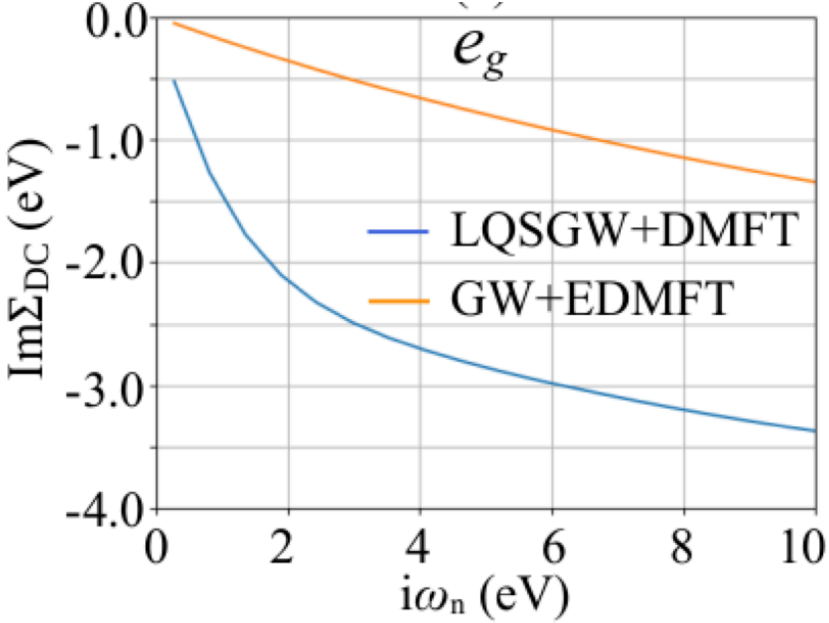




- LQSGW+DMFT as well as GW+EDMFT shows Z=0.7 for t2g orbitals
- Mott gap

Double-counting self-energy





Conclusion

• Full GW+EDMFT is a promising method to understand and predict the properties of correlated quantum materials from first principles.

• ComDMFT package is an *ab initio* package to support ab initio DMFT methodologies and is constantly evolving.

If you are interested in this research direction, feel free to contact me:)

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https://sites.google.com/view/sangkookchoi