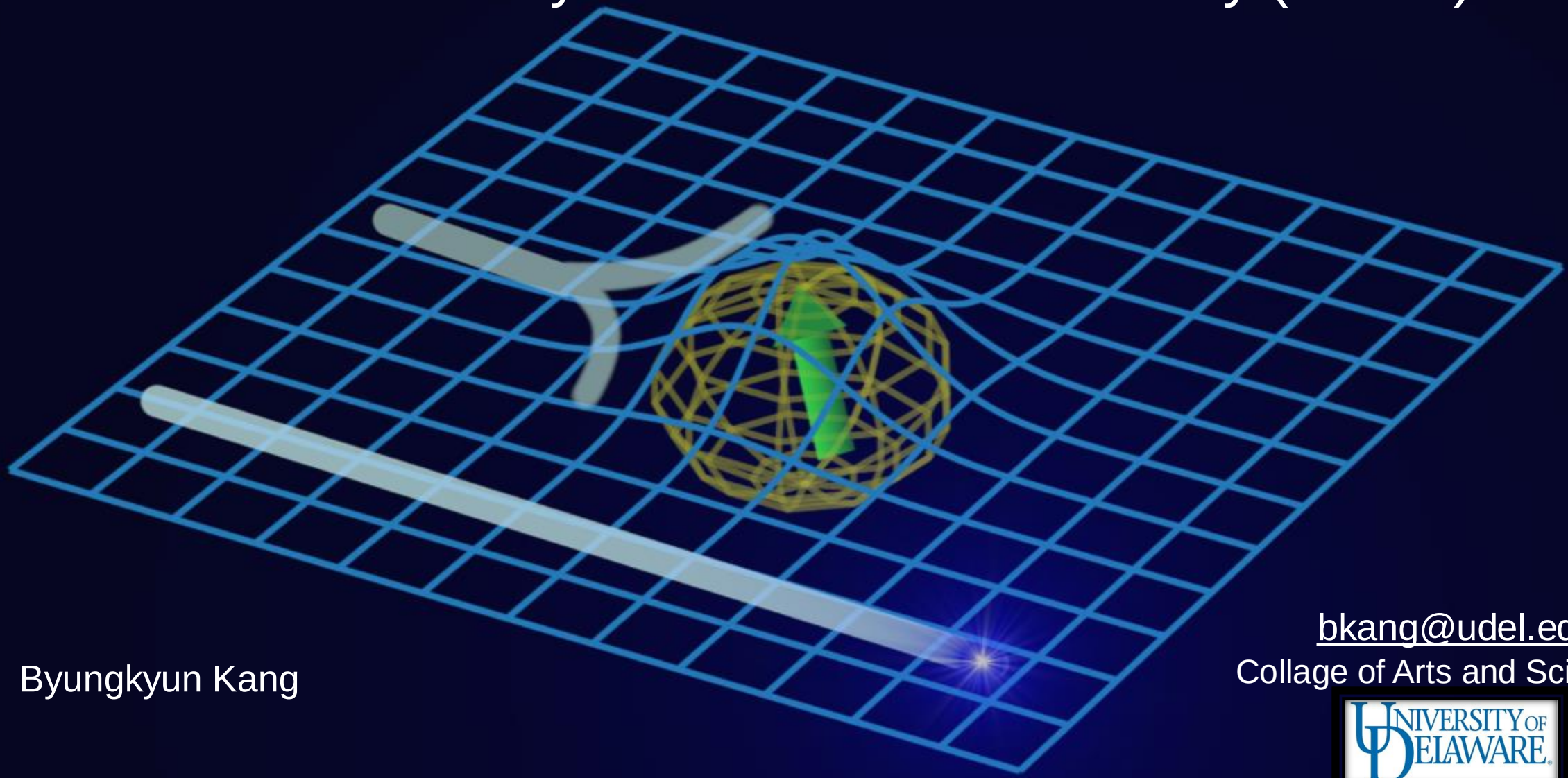
# Many-body perturbation theory (GW) combined with dynamical mean field theory (DMFT)



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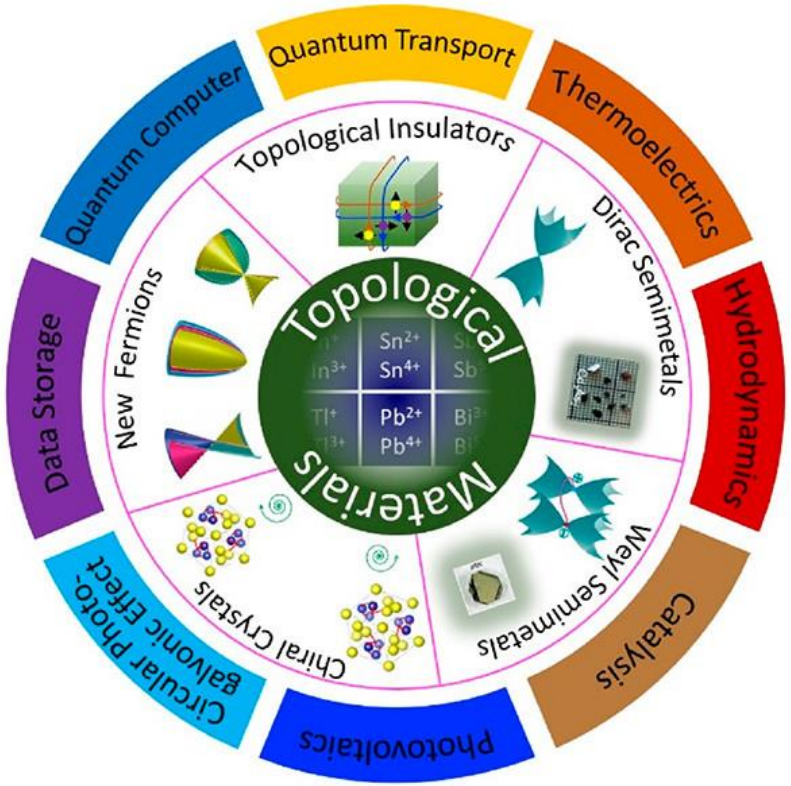
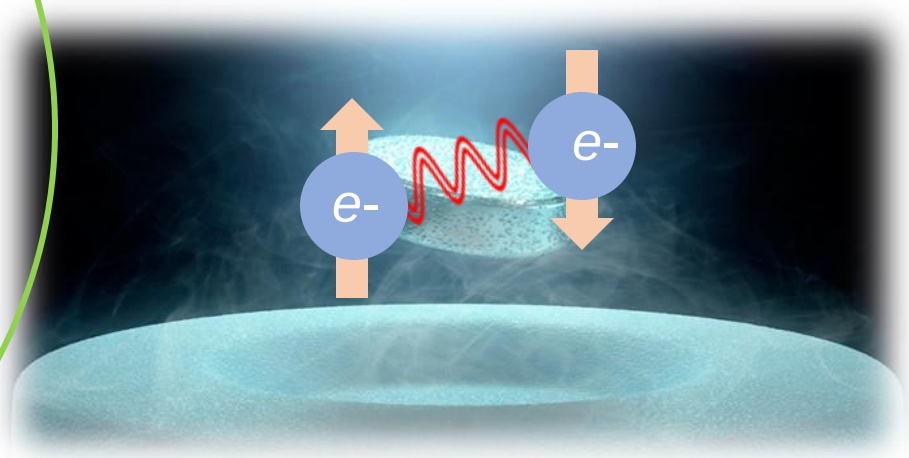
# Contents

- Quantum materials
- Many-body methods
- LQSGW+DMFT
- Fully self-consistent GW+EDMFT

# Quantum materials

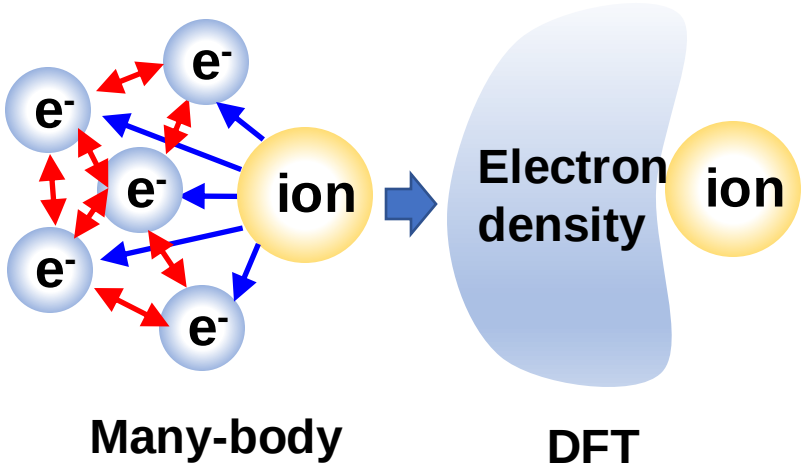



Correlated
Quantum Materials




[Chem. Rev. 121,5,2780,(2021)]

# Density Functional Theory



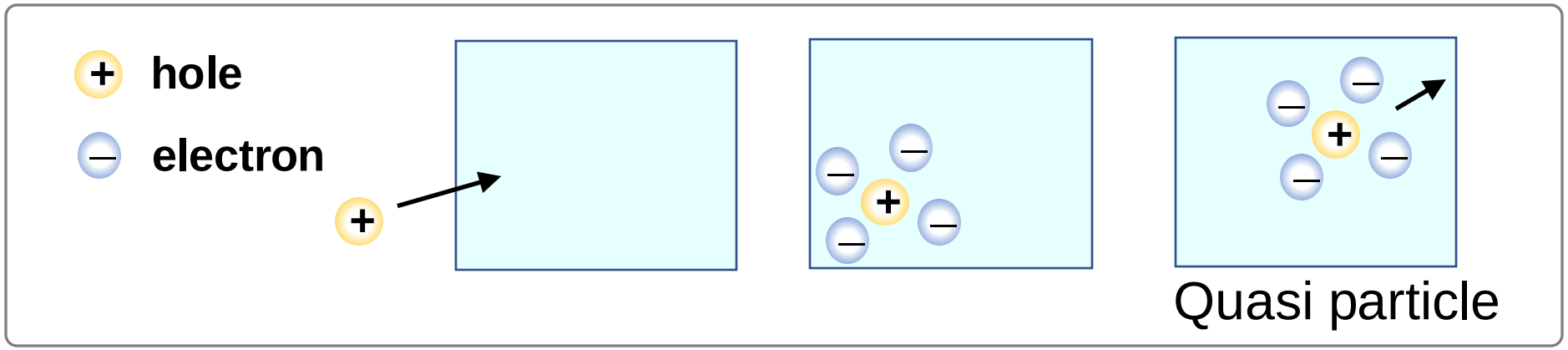

- DFT solve electronic structure from first-principles
- Ground state is unique with self-consistent
- W. Kohn 1999 Nobel Prize for chemistry.

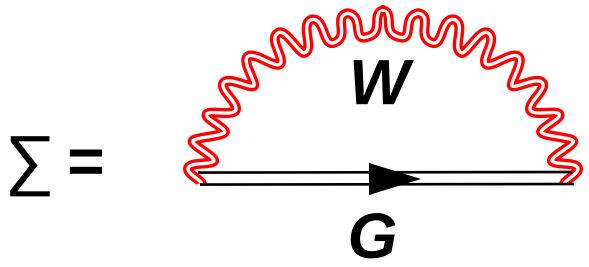
## DFT+U (hybrid functional) for strong correlation

- Add electron correlation U on static mean-field
- Work for magnetic ground state
- Practical for supercell

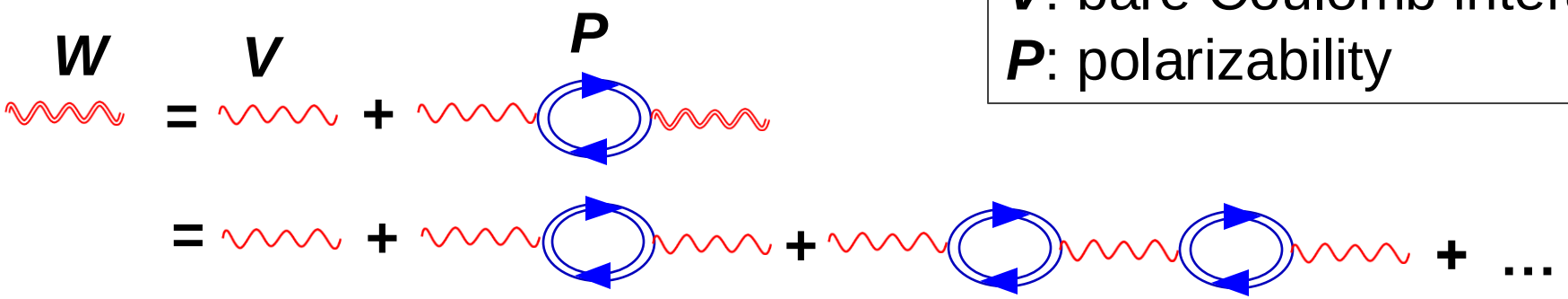
[1] P. Hohenberg, W. Kohn, Phys. Rev. B 136, 864 (1964).
[2] W. Kohn, L. J. Sham, Phys. Rev. B 140, 1133 (1965).

# Many-Body perturbation theory : GW method




∑ : self-energy
***G***: Green's function
***W***: screened Coulomb interaction
***V***: bare Coulomb interaction
***P***: polarizability

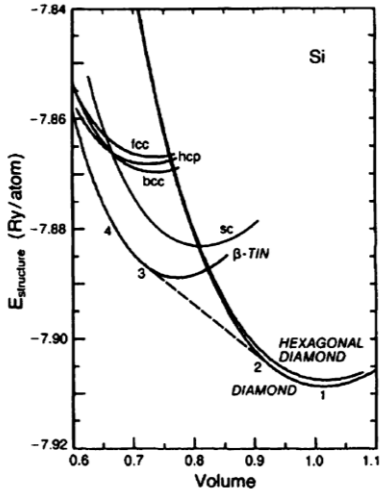


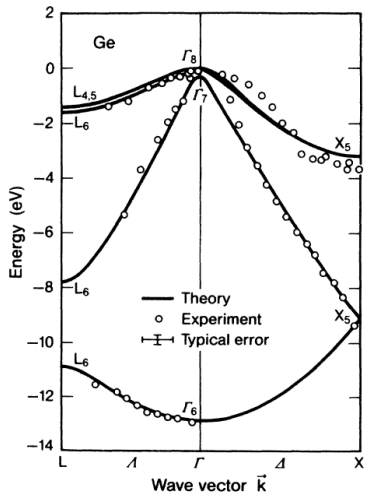

[1] L. Hedin, Phys. Rev. A 139, 796 (1965)

# DFT and GW are good theory tools for conventional materials

DFT for Ground states

GW for Excitations



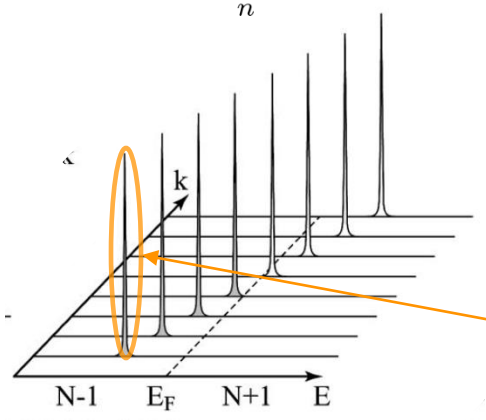

$$H = \sum_i \frac{\mathbf{p}_i^2}{2m_e} + V_n(\mathbf{r}_i) + V_H(\mathbf{r}_i) + V_{eff}(\mathbf{r}_i)$$

$$V_{eff} = V_{KS}$$

$$V_{eff} = W \circlearrowleft G$$

DFT: One-particle picture

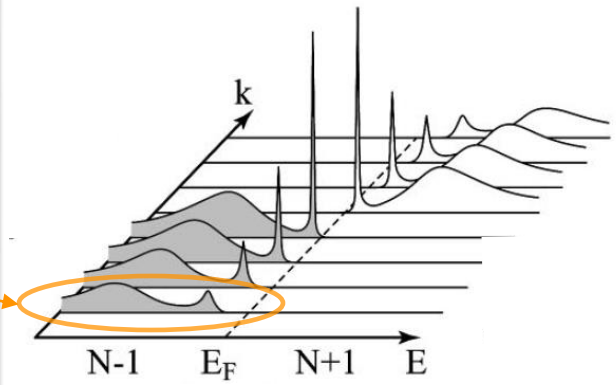
$$A(\mathbf{k}, E) = \sum_n \delta(E - E_{n\mathbf{k}})$$




Itinerant electron
Nondegenerate Ground States

GW: Green's function

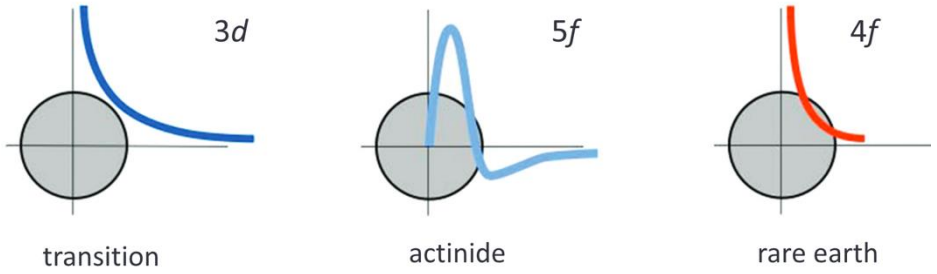
$$A(\mathbf{k}, E) = -\frac{1}{\pi} Im\, \mathrm{Tr}\, G(\mathbf{k}, E)$$



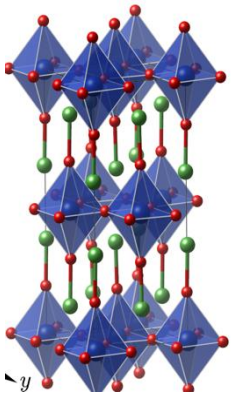

Quasiparticle
One-to-one correspondence


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

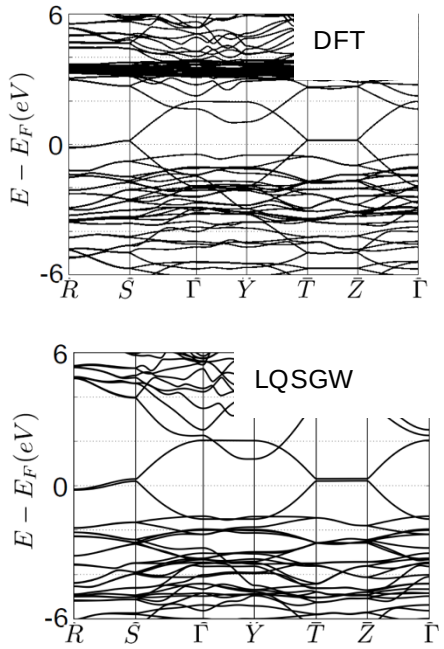
# DFT and GW often fail for correlated quantum materials




$La_2CuO_4$

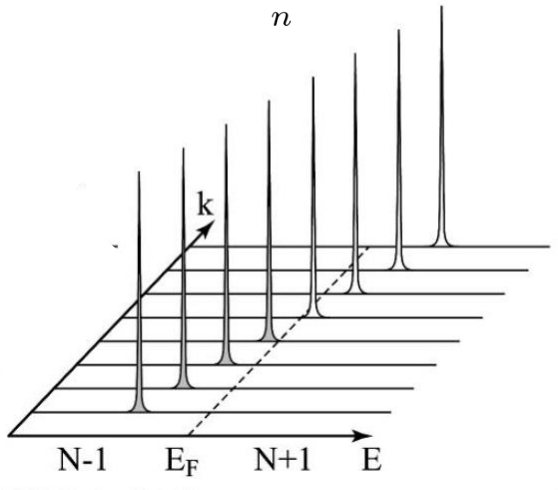


Paramagentic insulator



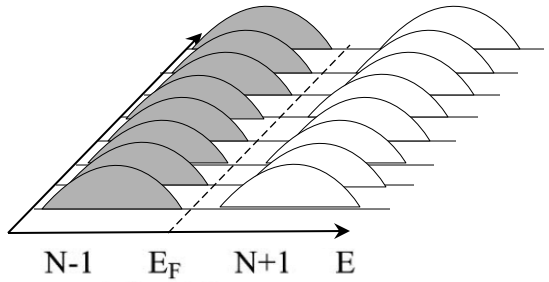

## One-particle picture

$$A(\mathbf{k}, E) = \sum_n \delta(E - E_{n\mathbf{k}})$$




Itinerant electron
Nondegenerate Ground States

## Correlated Quantum Materials

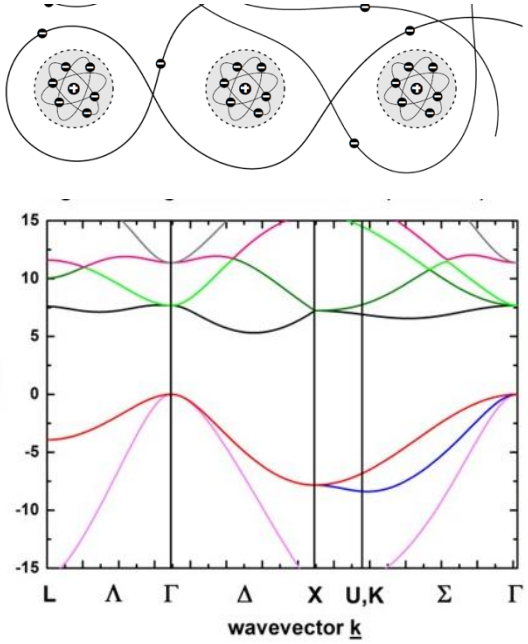



Perturbation to a mean-field fails
In the extreme case, no continuity

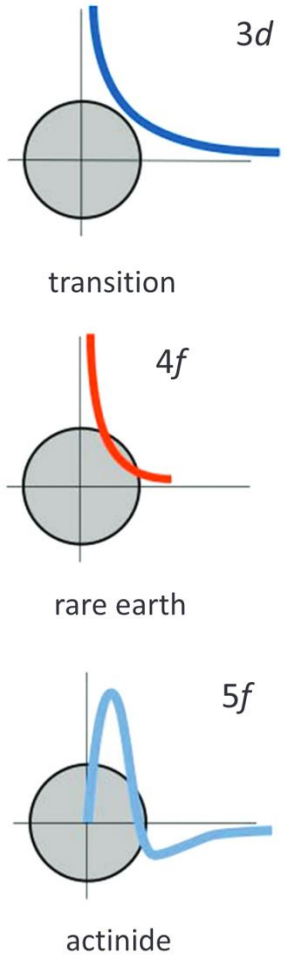
[npj Quantum Materials 1, 16001 (2016)]

# Ab initio theory beyond one-particle picture is needed!!!

## Itinerant electrons: bands

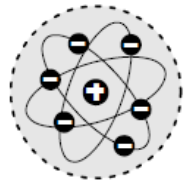


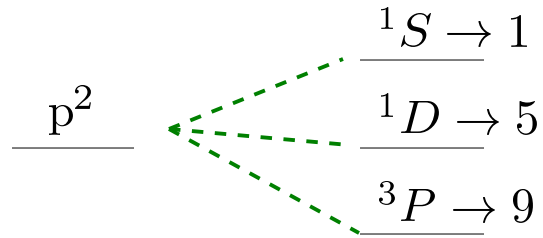

- Nondegenerate ground states
- Single slater determinants




Neither band-like nor atom-like

## Localized electrons: multiplets

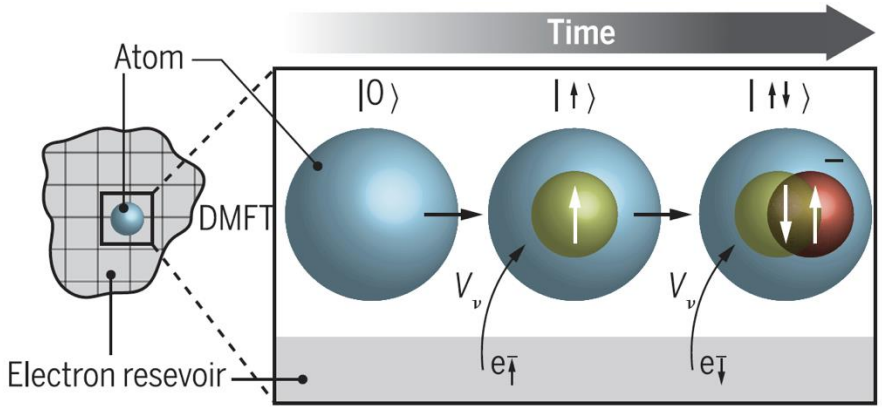





- Degenerate ground states
- Linear combination of many Slater determinants

# Dynamical mean field theory provides a reference frame for correlated electrons

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

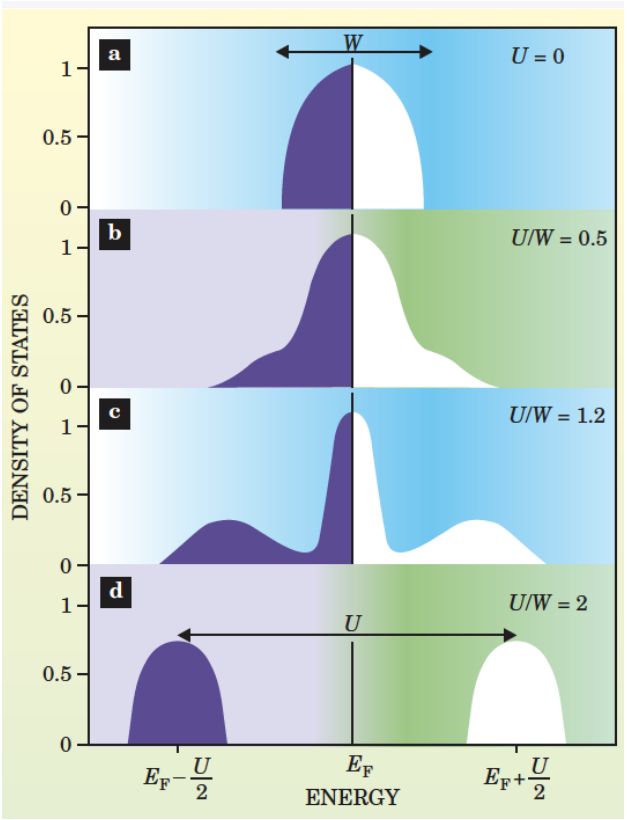



$$S_{eff} = -\int_0^\beta d\tau d\tau'\, d^+_{o\sigma}(\tau)\, \mathcal{G}_0^{-1}(\tau,\tau')\, d_{o\sigma}(\tau') + \int_0^\beta d\tau\, U n_{o\uparrow}(\tau) n_{o\downarrow}(\tau)$$

Electron hopping
Band-like

Multiplets
Atom-like

Single Band Hubbard Model

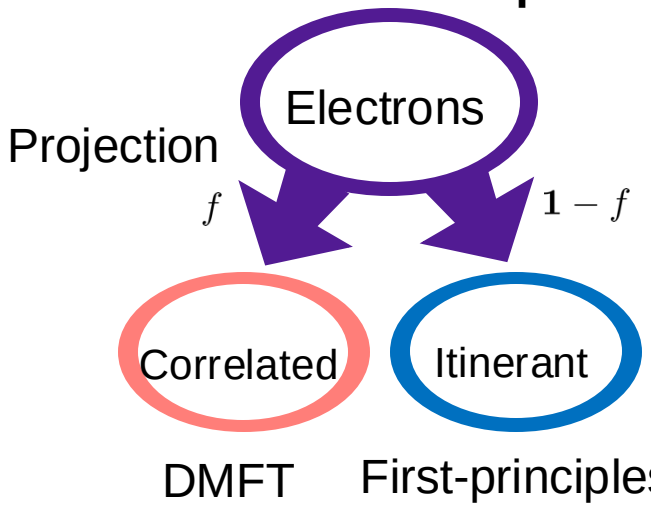



Band-like

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).

# Parameter-free first-principles + DMFT is a challenging task




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

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

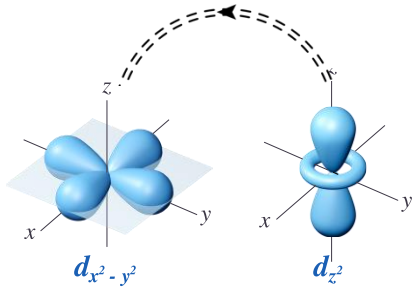
$$\widetilde{\Sigma}_{imp} = \widetilde{\mathcal{G}}^{-1} - \widetilde{G}_{imp}^{-1}$$

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

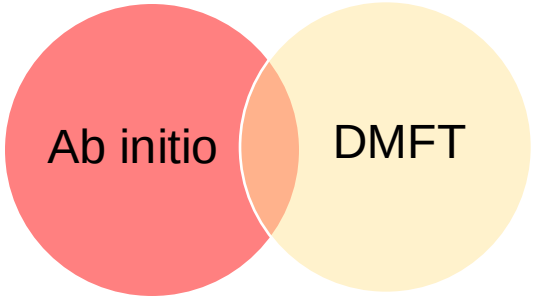
$$S_{eff}(U) \rightarrow \widetilde{G}_{imp} \longleftarrow \widetilde{\mathcal{G}}^{-1} = \widetilde{G}_{loc}^{-1} + \widetilde{\Sigma}_{imp}$$

**Challenges: U and $\widetilde{\Sigma}_{DC}$ should be determined by the choice of correlated orbitals ($f$)**

Effective (renormalized) Coulomb interaction

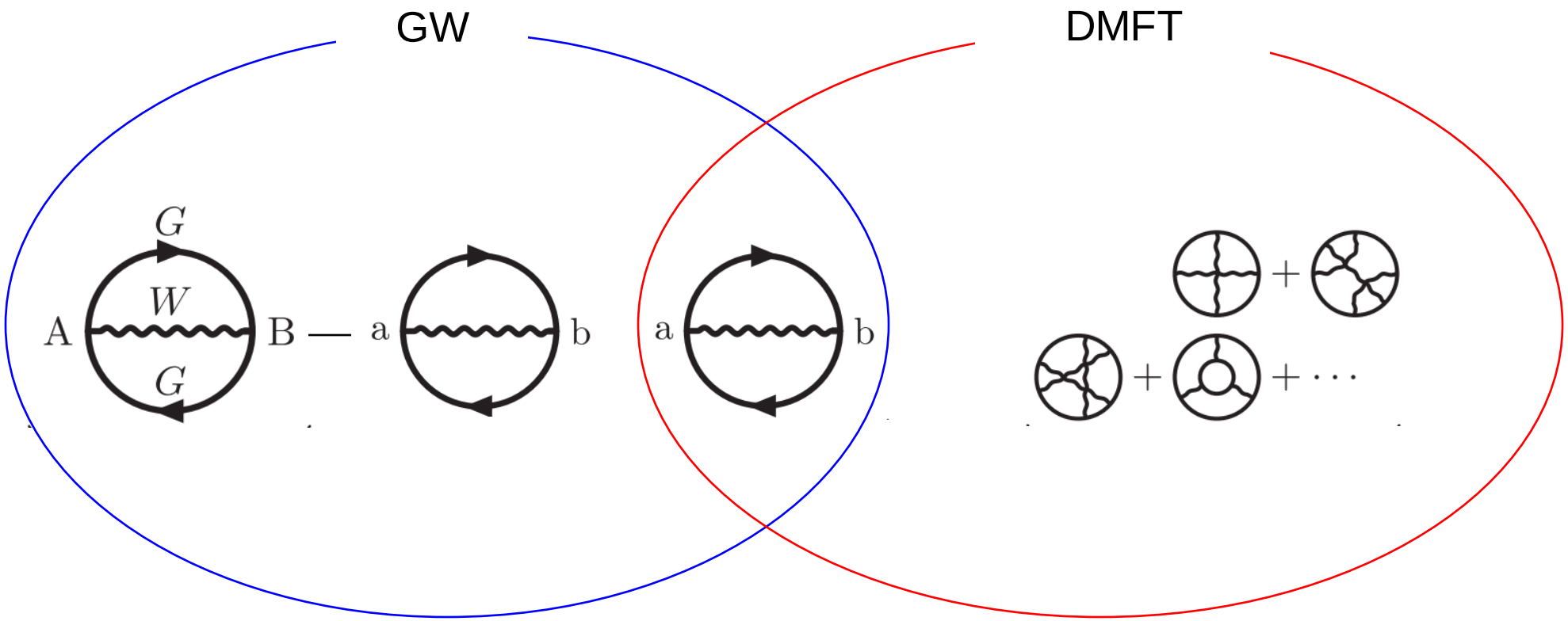



Double counted exchange and correlation




[1] A. Georges, et al., Rev. Mod. Phys. 68, 13 (1996), [2] G. Kotliar et.al., Rev. Mod. Phys. 78, 865 (2006).

# Diagrammatic Approaches can be a solution

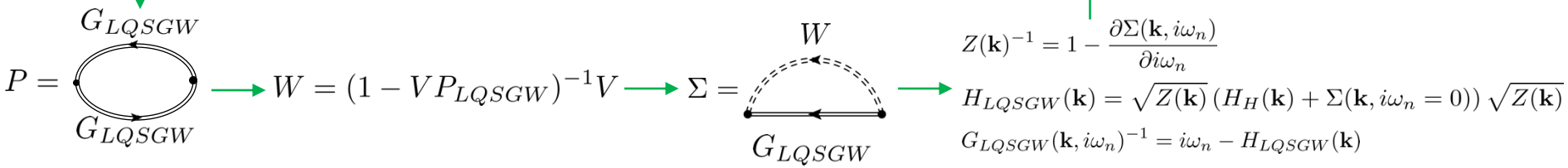



Double counted self-energy: Local GW
Effective Coulomb interaction: $U = (1 + W_{loc}P_{imp})^{-1}W_{loc}$
Full version: fully self-consistent GW+EDMFT

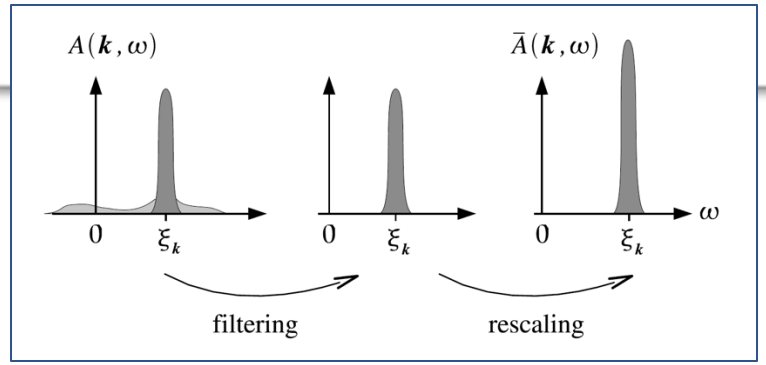
[1] P. Sun and G. Kotliar, Phys. Rev. B 66, 085120 (2002). [2] S. Biermann, F. Aryasetiawan, and A. Georges, Phys. Rev. Lett. 90, 086402 (2003) [3]F. Nilsson, L. Boehnke, P. Werner, and F. Aryasetiawan, Phys. Rev. Materials 1, 043803 (2017).

# We proposed LQSGW+DMFT, a parameter free ab initio Method

## For $H_{abinitio}$: Linearized quasiparticle self-consistent GW




free from the starting point dependence of one-shot GW



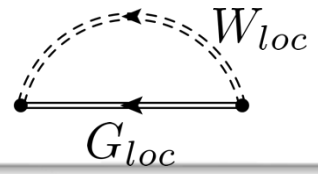

## For U: cRPA for effective Coulomb interaction

bare with respect to the correlated orbitals
but renormalized with respect to the rest

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

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

## For $\Sigma_{DC}$: local GW




- A simplification of full GW+EDMFT
- a parameter-free method (advantage over DFT+DMFT)
- Validation on charge transfer insulators, Fe-based superconductors, and narrow-gap correlated insulators

[1] J. Phys.: Conf. Ser. 592, 012055 (2015). [2] npj QM 1, 16001 (2016). [3] S. Choi, P. Semon, B. Kang, A. Kutepov, and G. Kotliar, CPC. 244, 277 (2019)

# ComDMFT: LQSGW+DMFT through HPC

Computer Physics Communications 244 (2019) 277–294

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]

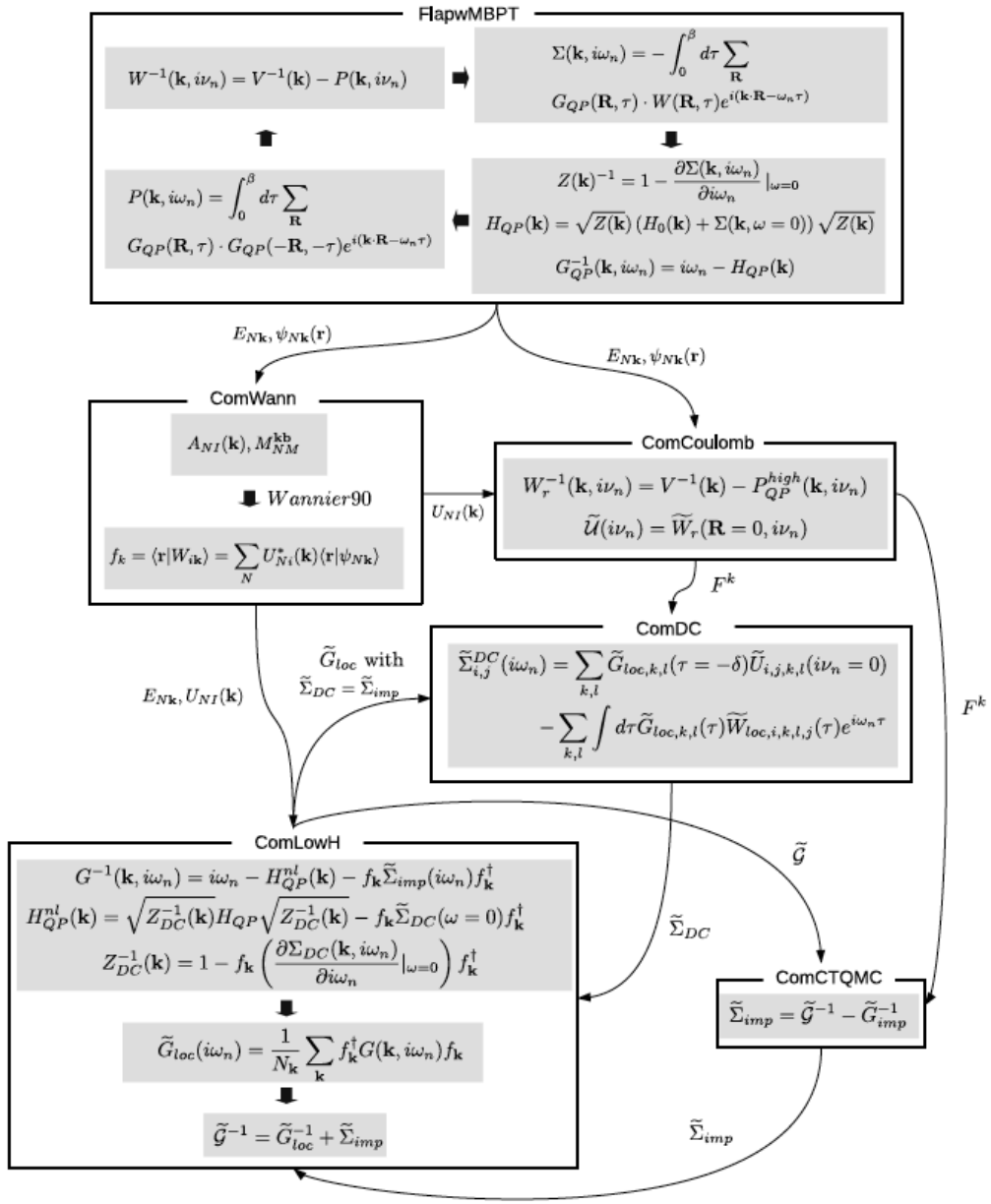
[a] Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, NY 11973, USA
[b] Department of Physics and Astronomy, Rutgers University, NJ 08854, USA

$$\widetilde{\Sigma}_{imp} = \widetilde{\mathcal{G}}^{-1} - \widetilde{G}^{-1}_{imp} \quad \Rightarrow \quad G^{-1}(\mathbf{k}, i\omega_n) = G^{-1}_{MF}(\mathbf{k}, i\omega_n) - f_{\mathbf{k}}\widetilde{\Sigma}_{imp}(i\omega_n)f^{\dagger}_{\mathbf{k}}$$

$\widetilde{\mathcal{U}}(i\nu_n)$ from cRPA
$G_{MF}$ from non-local LQSGW
$f_{\mathbf{k}}$ from Wannier functions

$$\widetilde{\mathcal{G}}^{-1} = \widetilde{G}^{-1}_{loc} + \widetilde{\Sigma}_{imp} \quad \Leftarrow \quad \widetilde{G}_{loc}(i\omega_n) = \frac{1}{N_{\mathbf{k}}}\sum_{\mathbf{k}} f^{\dagger}_{\mathbf{k}} G(\mathbf{k}, i\omega_n) f_{\mathbf{k}}$$

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



# LDA and LQSGW for MnO, NiO

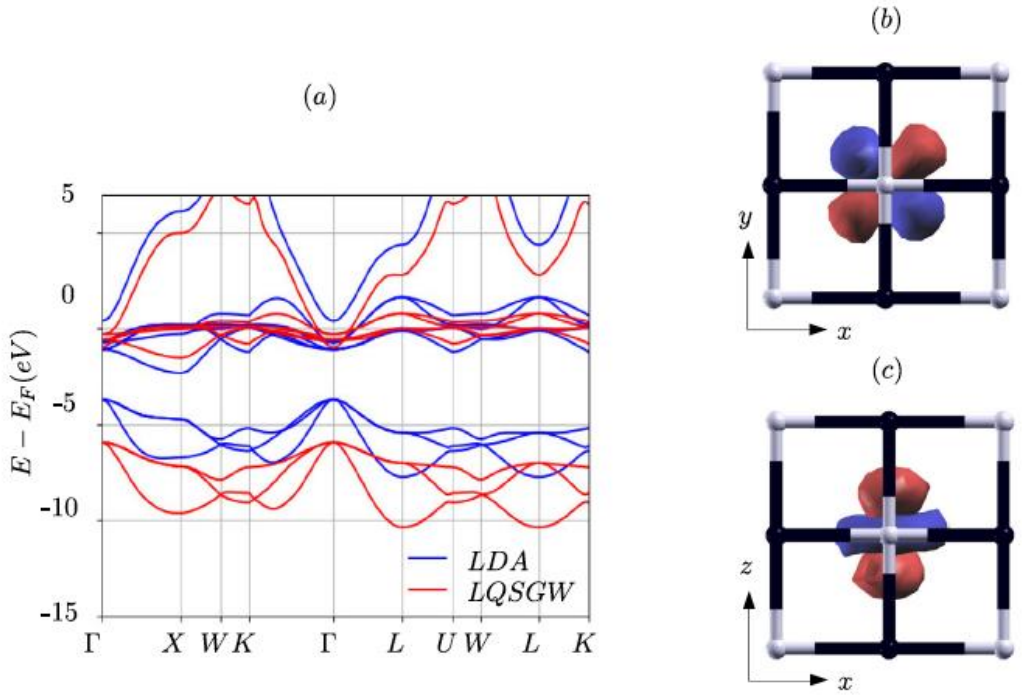


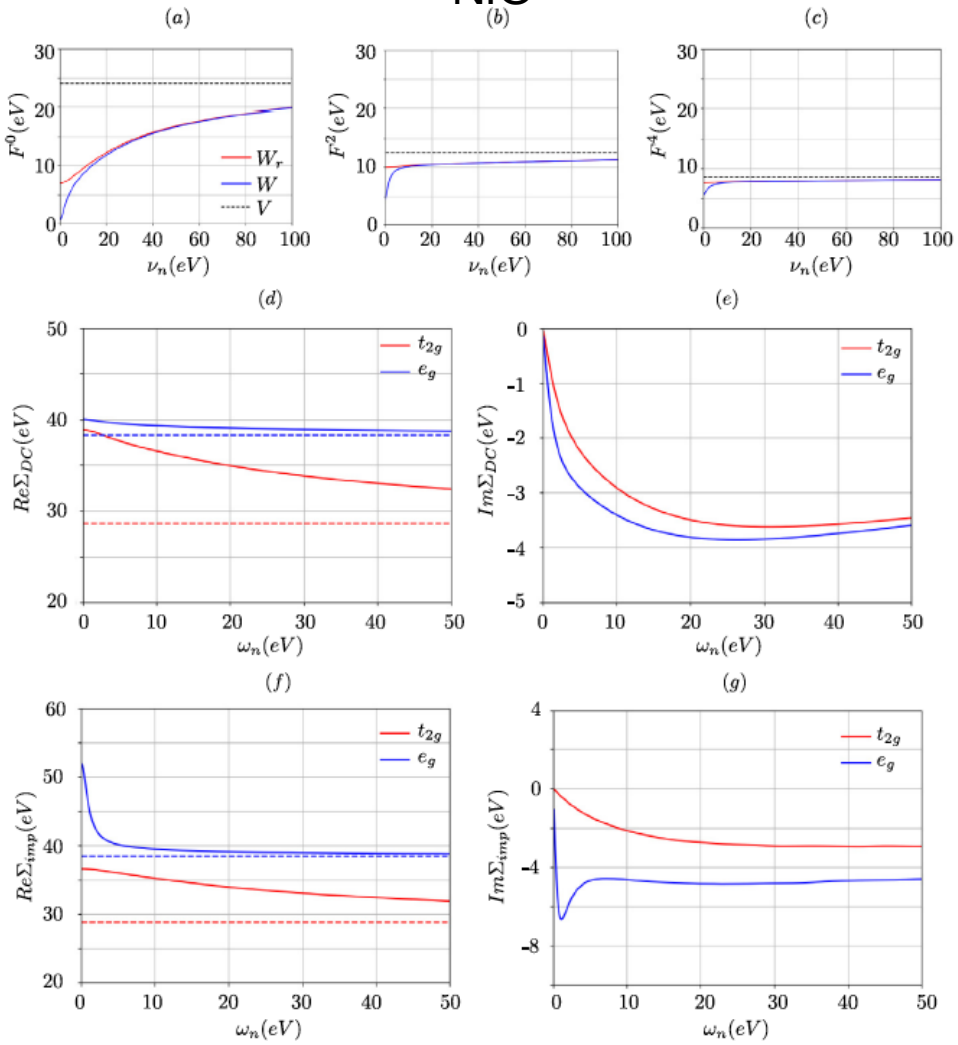

**Fig. 6.** (Color online) (a) Wannier interpolated MnO bandstructures within non spin-polarized LDA and LQSGW. (b) Mn-$d_{xy}$ and (c) Mn-$d_{z^2}$ Wannier functions in a FCC conventional unitcell. White and black spheres represent O and Mn atoms, respectively.

LDA and LQSGW do not capture Mott insulator

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

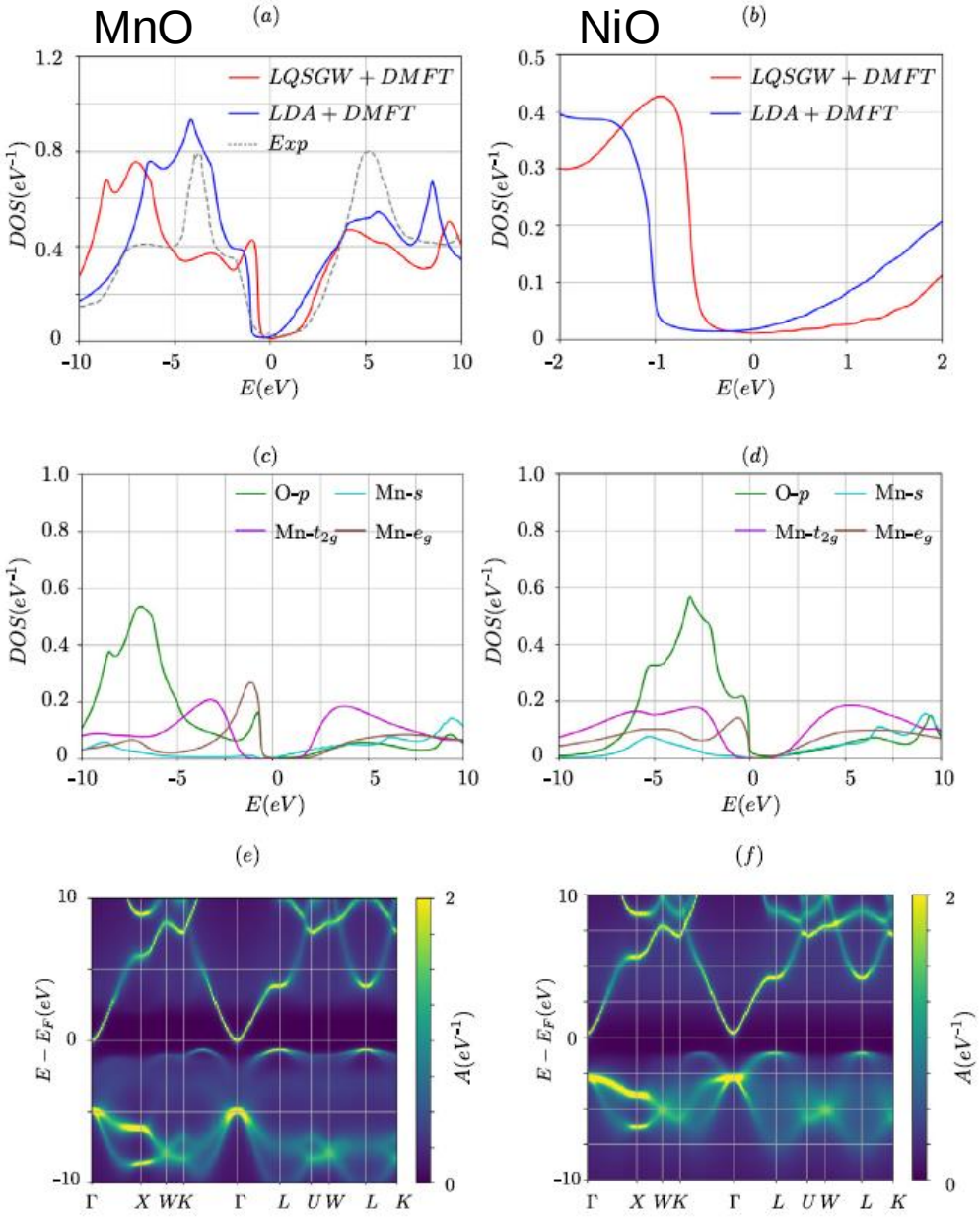
# LQSGW+DMFT for MnO, NiO paramagnetic Mott insulator

NiO




MnO

NiO




LQSGW+DMFT capture Mott insulator

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

# ComDMFT: fully self-consistent ab initio GW+EDMFT (FGW+EDMFT)

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


Byungkyun Kang[a,b], Patrick Semon[a], Corey Melnick[a], Mancheon Han[d], Seongjun Mo[d,e], Hoonkyung Lee[e], Gabriel Kotliar[a,c], Sangkook Choi[a,d,*]

[a] *Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, NY 11973, USA*
[b] *College of Arts and Sciences, University of Delaware, Newark, DE 19716, USA*
[c] *Department of Physics and Astronomy, Rutgers University, NJ 08854, USA*
[d] *School of Computational Sciences, Korea Institute for Advanced Study, Seoul 02455, Republic of Korea*
[e] *Department of Physics, Konkuk University,Seoul 05029, Korea*

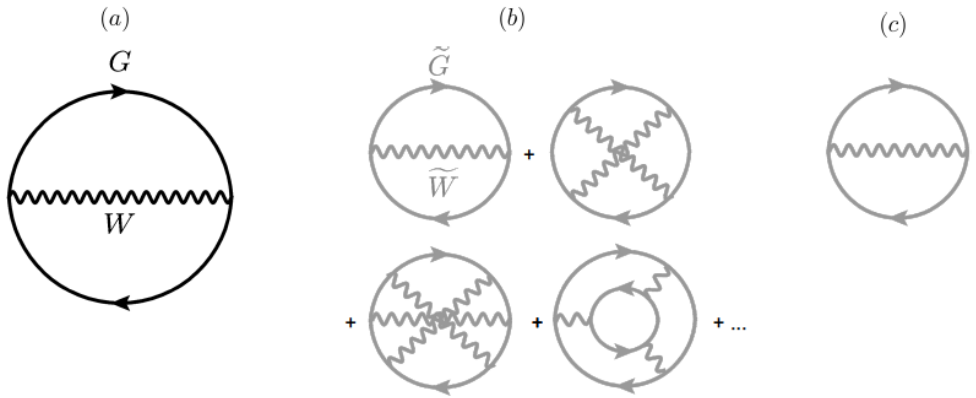




Figure 1: Diagrammatic representations of the Ψ functional within full GW+EDMFT approach. (a) GW Ψ functional (b) EDMFT Ψ functional and (c) local-GW Ψ functional. Full and wiggly lines represent Green's function ($G$) and screened Coulomb interaction ($W$), respectively. Black and gray colors represent quantities in the full Hilbert space and in the correlated subspace, respectively.

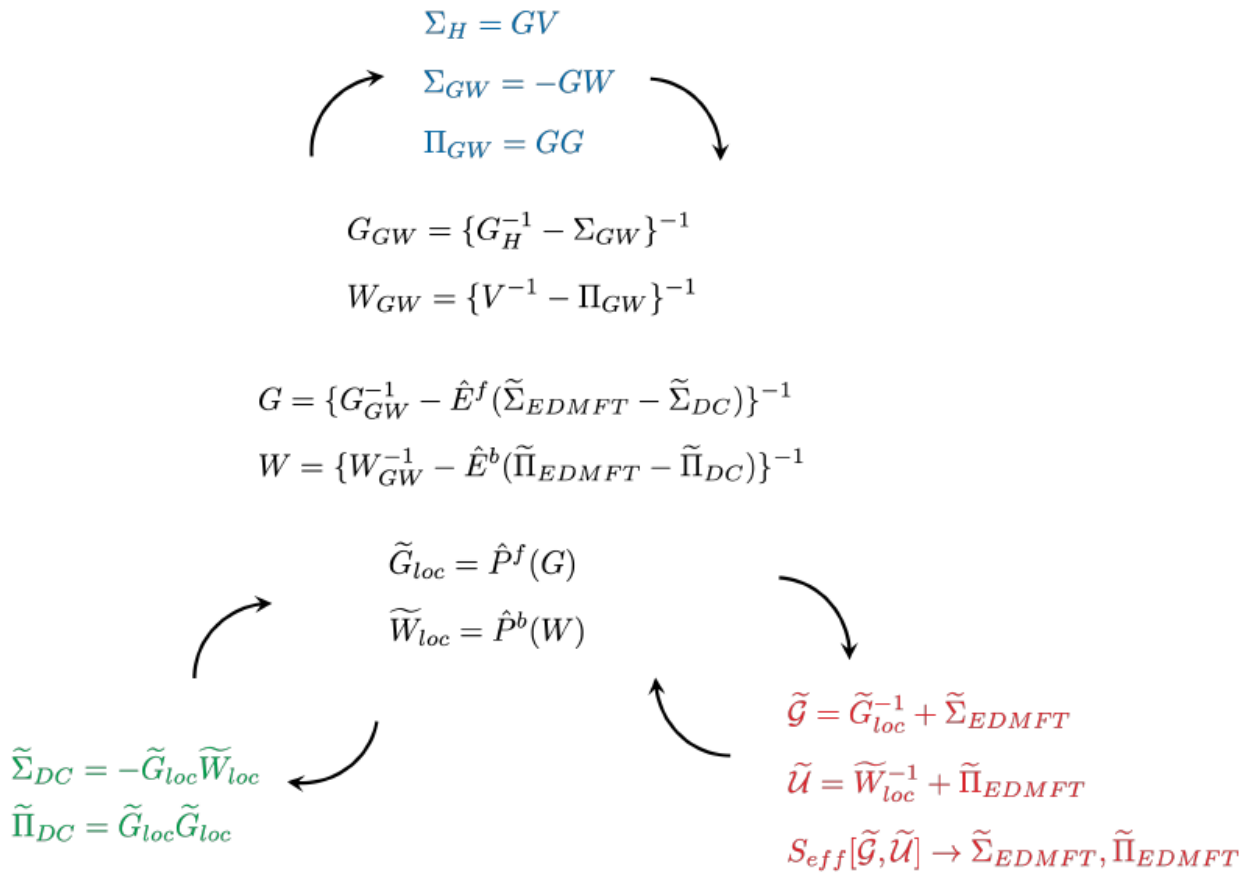



Figure 2: The GW+EDMFT approach involves three self-consistency loops: the GW loop (blue), the EDMFT loop (red), and the double counting loop (green). This diagram seeks to emphasize the need for self-consistent determination of three-different terms in both fermionic and bosonic self-energies. For more information on the practical implementation of the self-consistent loop, please refer to Fig. 4.


[1] B. Kang et al. arXiv:2310.04613 (2024)

# Flowchart

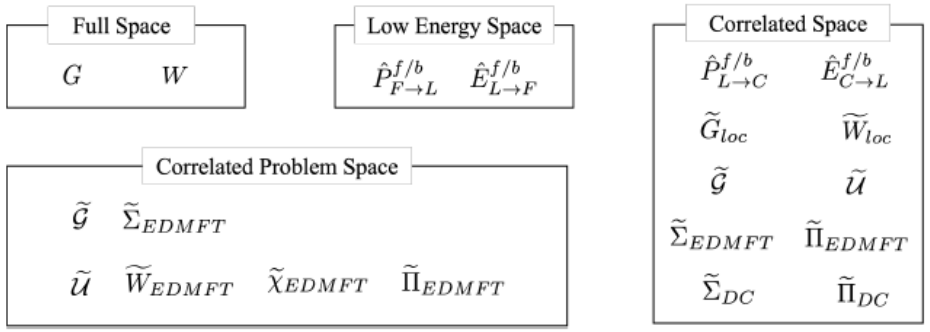
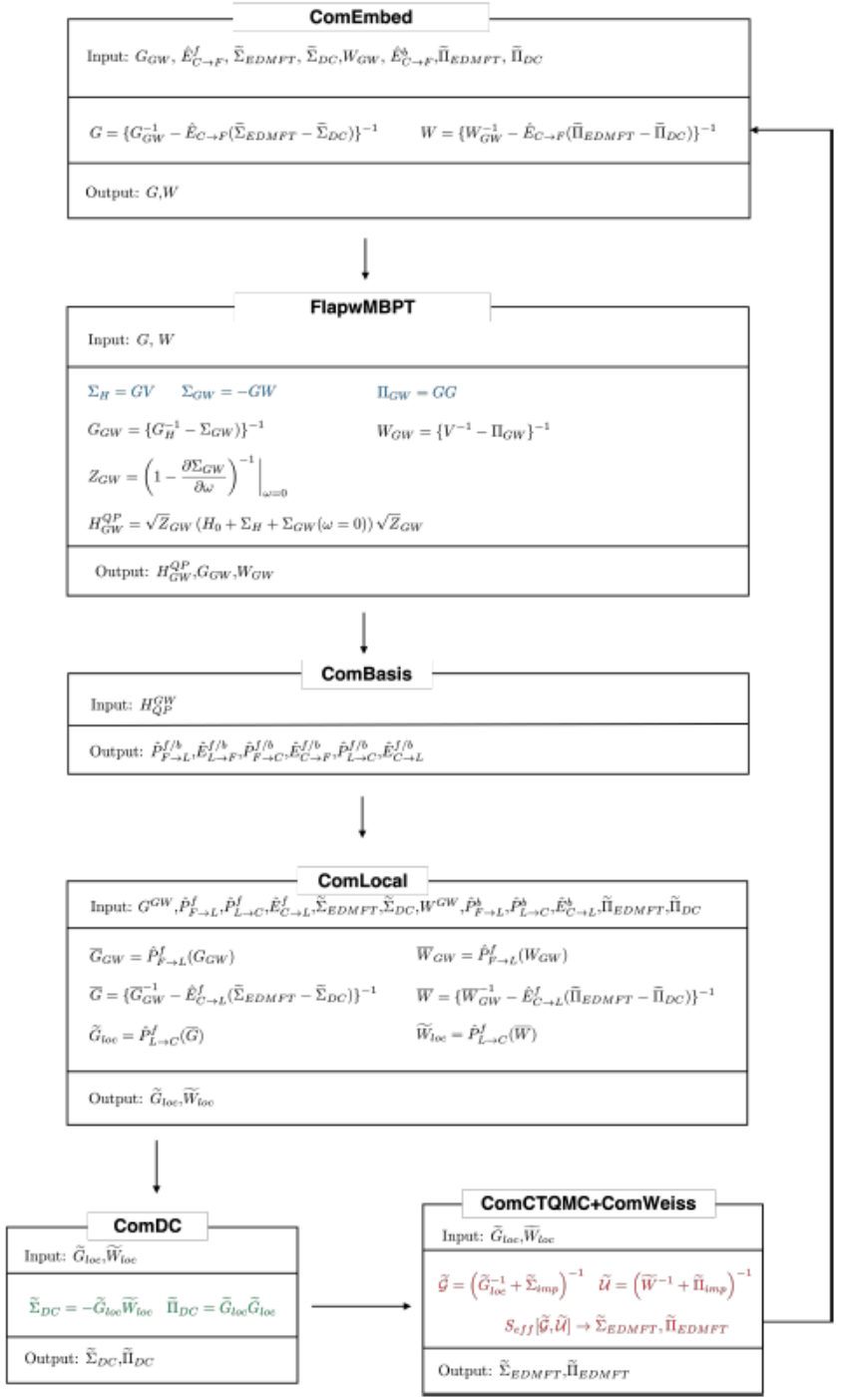



Figure 5: Grouped data structure within GW+EDMFT.




[1] B. Kang et al. arXiv:2310.04613 (2024)

# FGW+EDMFT for NiO

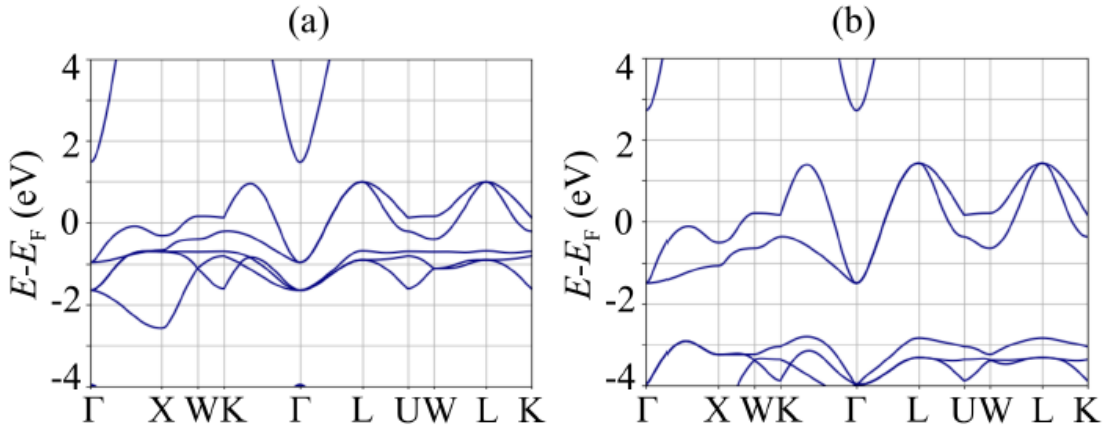



Figure 6: (a) and (b) show the Wannier-interpolated GW bandstructures ($H^{QP}_{GW}$ in eq. (28 of NiO at the first and last iterations of the self-consistent loop in Fig. 4, respectively.

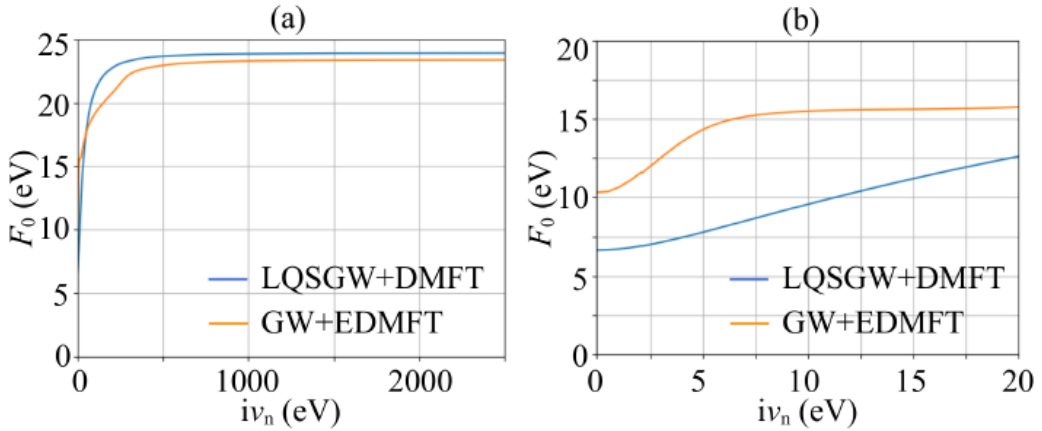



Figure 8: (a) The monopole part ($F^0$ of Slater integrals) of the bosonic Weiss field $F^0$ associated with Ni-$d$ orbitals in NiO. (b) zoom-in view of (a). Orange and blue colors show the results within full GW+EDMFT and LQSGW+DMFT, respectively

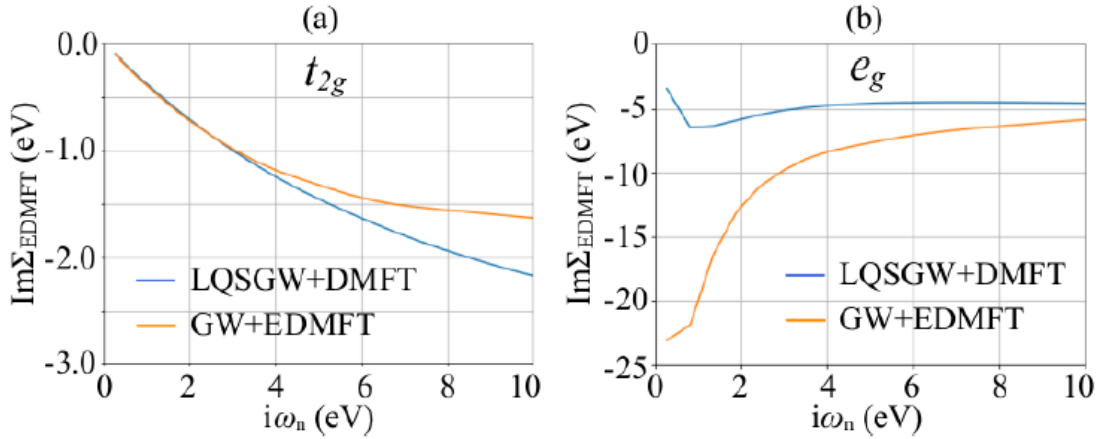
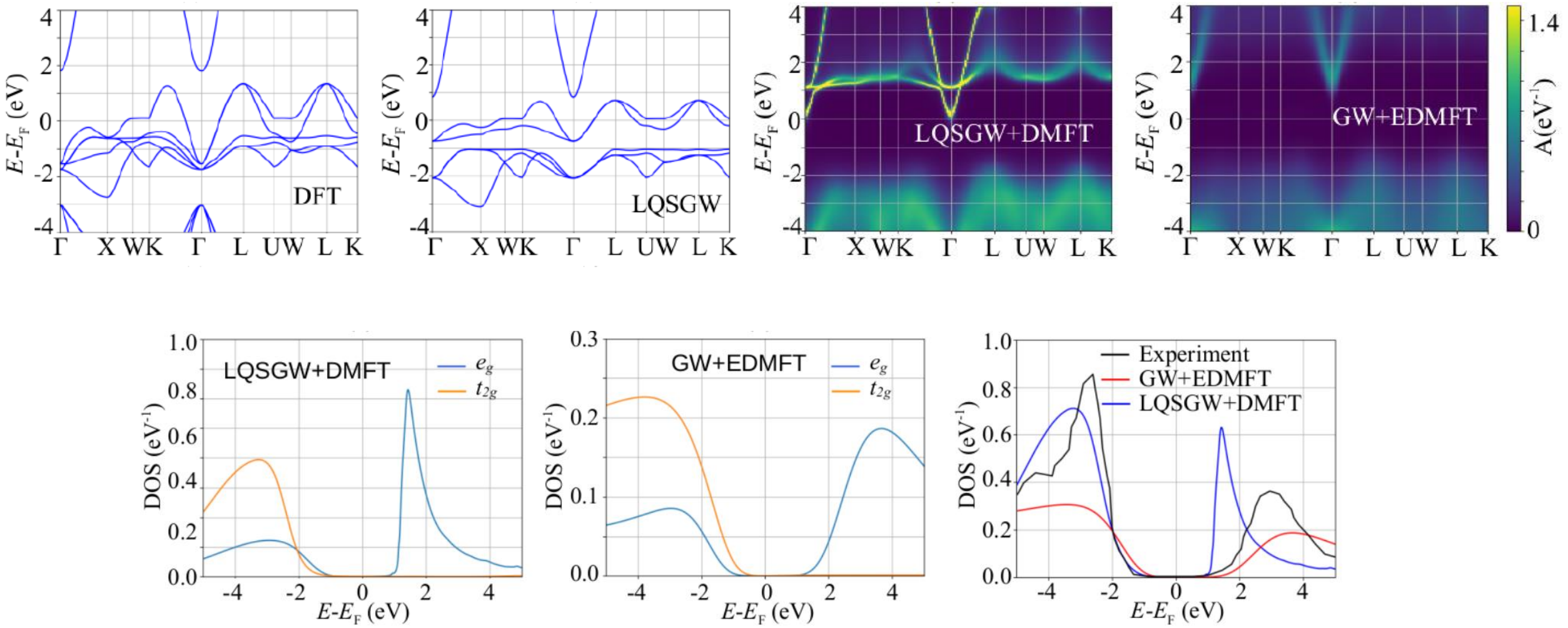



Figure 9: Imaginary parts of EDMFT self-energies associated with (a) Ni-$t_{2g}$ and (b) Ni-$e_g$ in NiO. Orange and blue colors show the results within full GW+EDMFT and LQSGW+DMFT, respectively

GW do not capture Mott insulator.
GW+EDMFT exhibit stronger self-energy.

[1] B. Kang et al. arXiv:2310.04613 (2024)

# FGW+EDMFT for NiO




GW+EDMFT produce peak to peak gap close to experiment.

[1] B. Kang et al. arXiv:2310.04613 (2024)

# FGW+EDMFT for SrVO3 (correlated transition metal oxide)

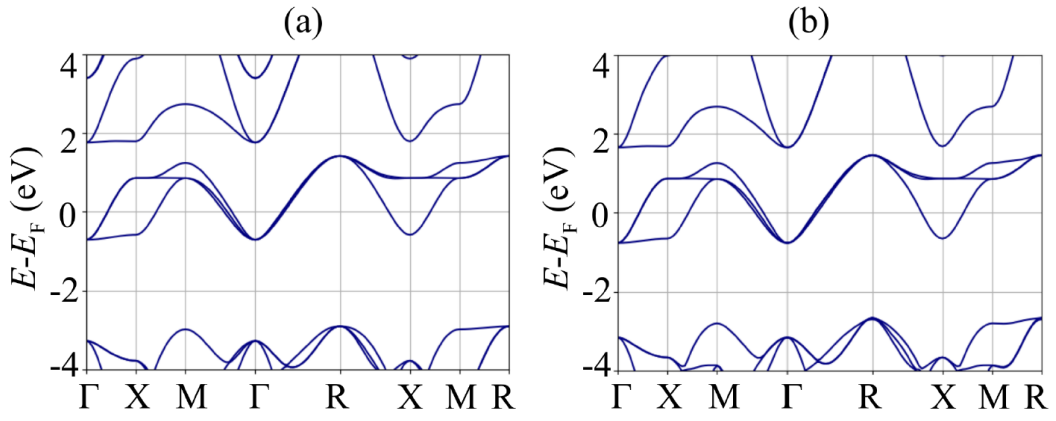



Figure 13: (a) and (b) show the Wannier-interpolated GW bandstructures ($H^{QP}_{GW}$ in e (28)) of $SrVO_3$ at the first and last iterations of the self-consistent loop in Fig. 4, respectivel

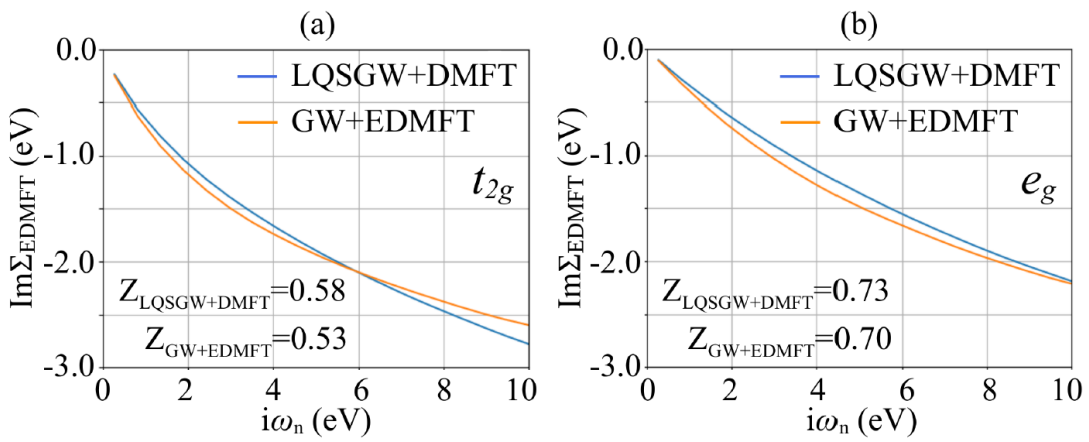



Figure 16: Imaginary parts of EDMFT self-energies associated with (a) V-$t_{2g}$ and (b) V-$e_g$ in $SrVO_3$. Orange and blue colors show the results within full GW+EDMFT and LQSGW+DMFT, respectively

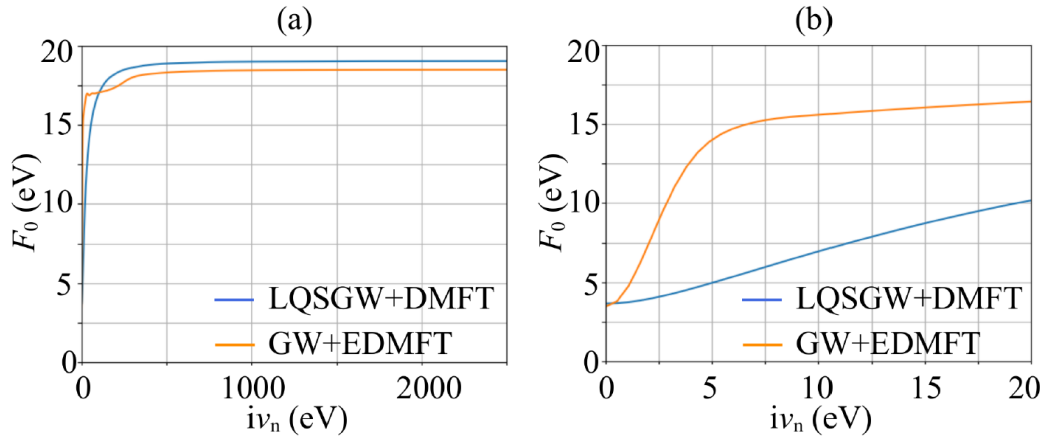
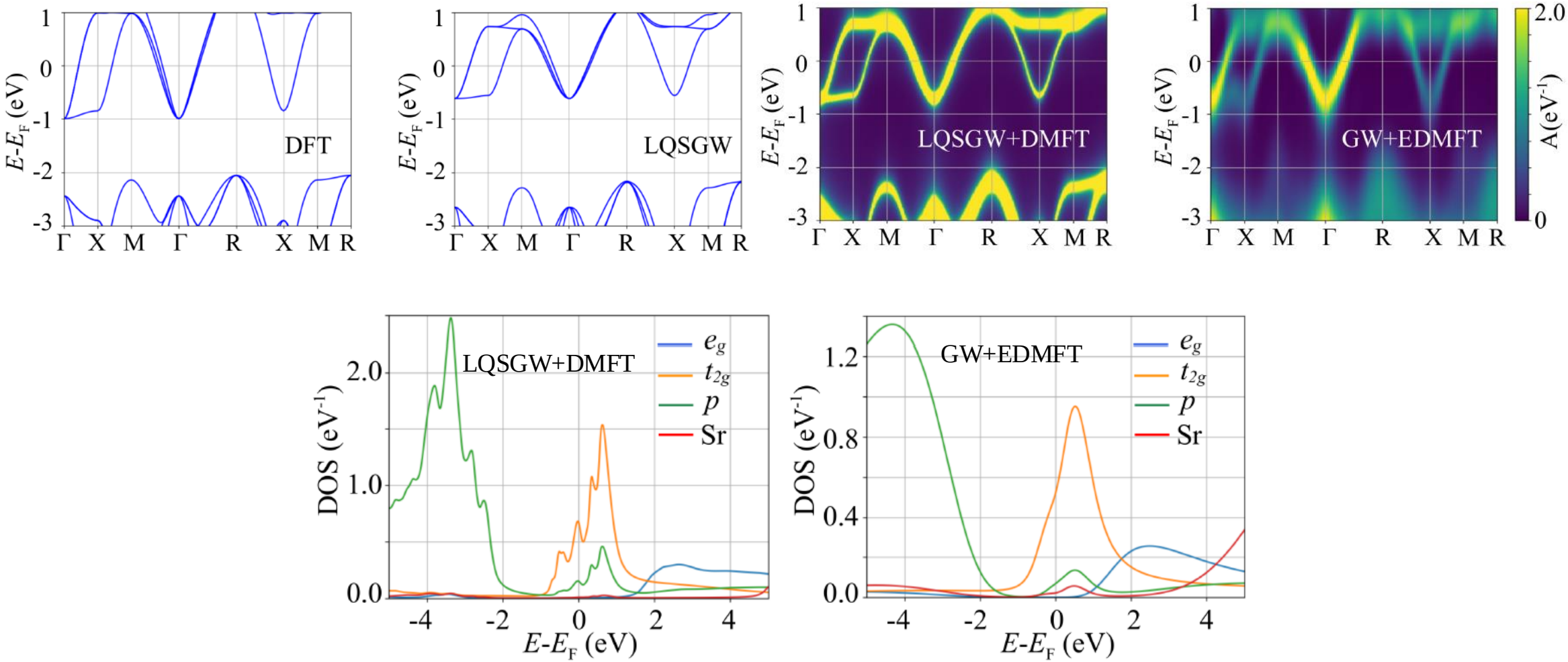



Figure 15: (a) The monopole part ($F^0$ of Slater integrals) of the bosonic Weiss field $F^0$ associated with V-$d$ orbitals in $SrVO_3$. (b) zoom-in view of (a). Orange and blue colors show the results within full GW+EDMFT and LQSGW+DMFT, respectively

GW band structures exhibit metallic. Coulomb interaction slop is larger in GW+EDMFT.

[1] B. Kang et al. arXiv:2310.04613 (2024)

# FGW+EDMFT for SrVO3




GW+EDMFT produce more incoherent electronic structure.

[1] B. Kang et al. arXiv:2310.04613 (2024)

Thank you