GW+EDMFT:
a diagrammatically controlled ab initio theory of strong correlation in real materials

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COMSCOPE
from codes to spectroscopies

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Contents

1. Introduction

Standard model of solids (Fermi liquid theory)

- Density Functional Theory (Kohn & Hohenberg 1964, Kohn & Sham 1965)

$$\Gamma\ [\ \rho\ (r)\ ] = \Gamma_{univ}\ [\ \rho\ (r)\ ] + \int dr V_{cryst}(r)\rho\ (r)$$

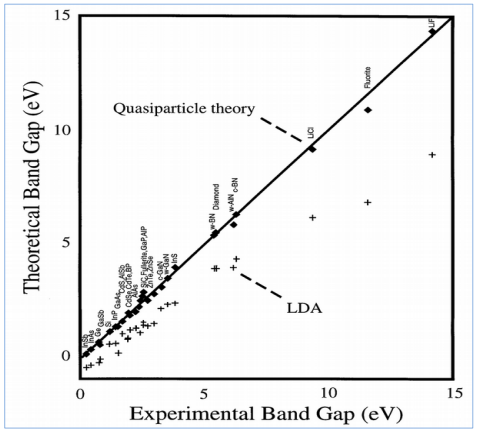
$$-\nabla^2 / 2 + V_{KS}(r)[\rho]\ \psi_{kj} = \varepsilon_{kj}\psi_{kj}$$

$$\rho(r) = \sum_{\varepsilon_{kj}<0} \psi_{kj}{}^*(r)\psi_{kj}(r)$$

$$V_{KS} = V_{Hartree} + V_{cryst} + V_{xc}$$

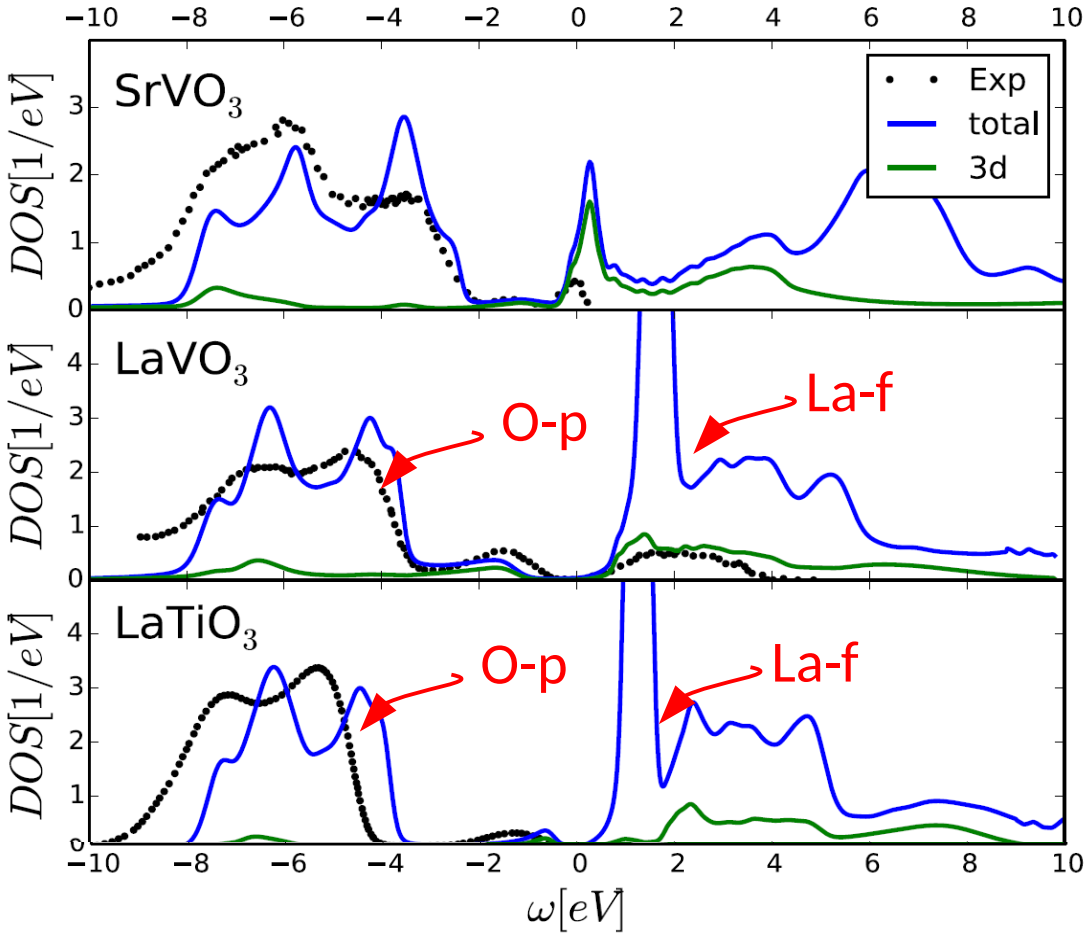
Starting point for perturbation theory in the screened Coulomb interactions (Hedin 1965) G0W0

$$G^{-1} = G_{0KS}^{-1} + [\ \ \ - V_{xc}\ ]$$



- This paradigm now enables material design and theoretical spectroscopy for weakly correlated electrons. It fails in a broad class of interesting materials. Correlated Electron Systems. Non perturbative problem. Need new starting point.

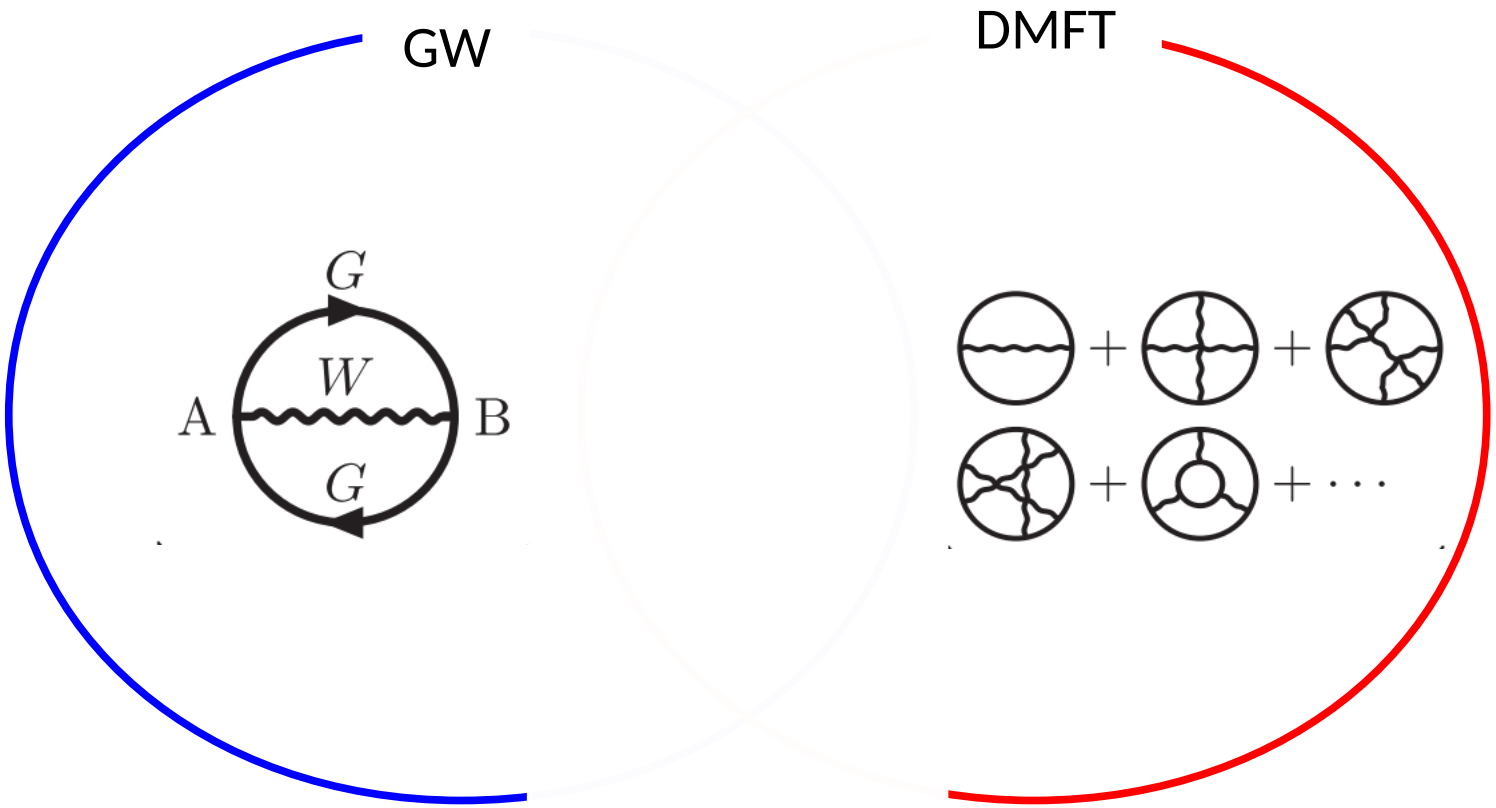
First-principles +DMFT: LDA+DMFT



- Many successful stories of LDA+DMFT including metal insulator transition in transition metal oxides
- Many new features including total energy, forces, exact double-counting energy
- Position of La-f, O-p

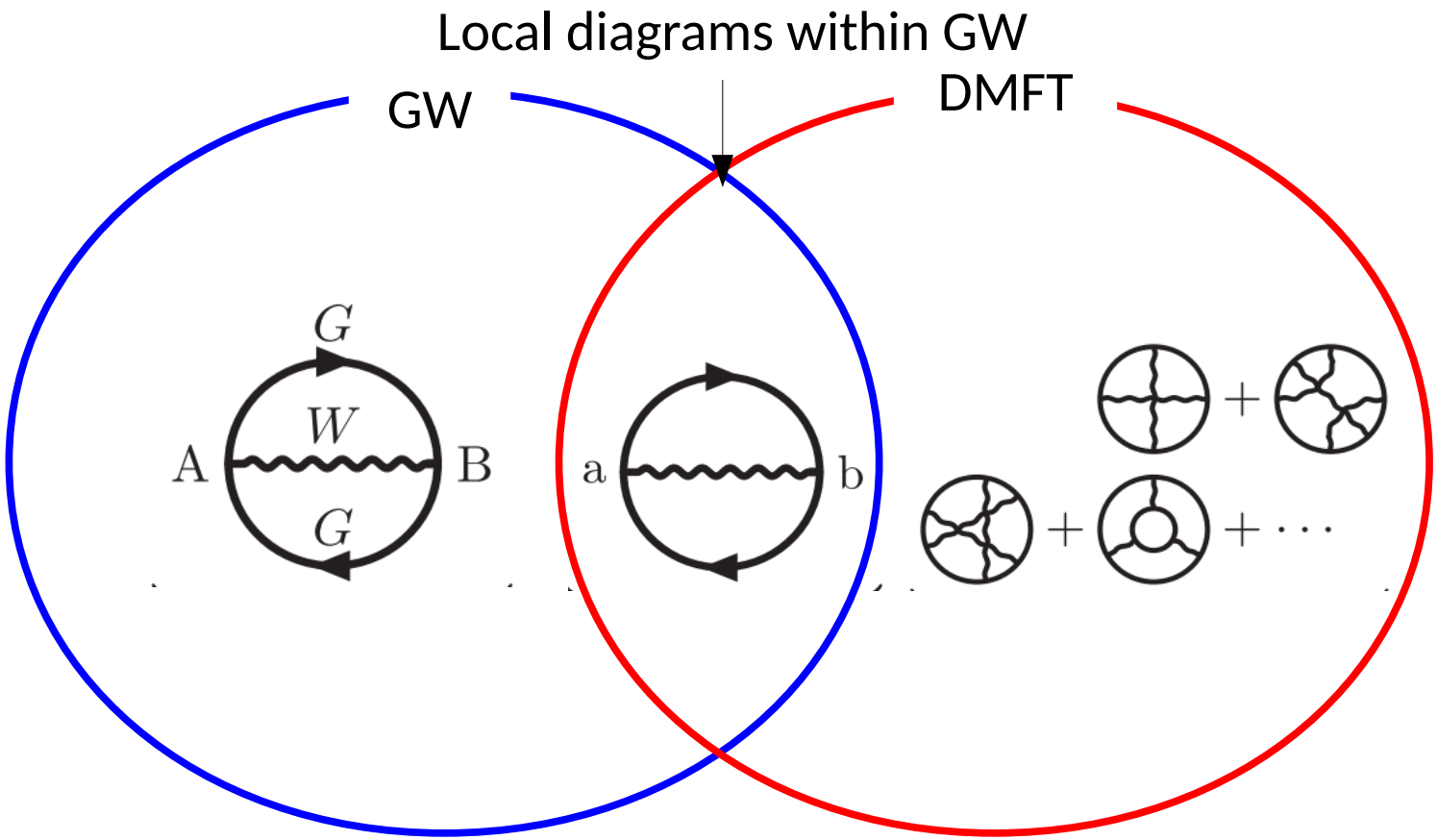
[1] K. Haule, T. Birol, and G. Kotliar, Phys. Rev. B 90, 075136 (2014).
[2]G. Kotliar, S. Y. Savrasov, K. Haule, V. S. Oudovenko, O. Parcollet, and C. A. Marianetti, Rev. Mod. Phys. 78, 865 (2006).

GW and 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).

GW+EDMFT: diagrammatically controlled *ab initio* theory



- a diagrammatical route to calculate on-site strong correlation and non-local correlation
- first-order non-trivial correction to DMFT or local correction to GW

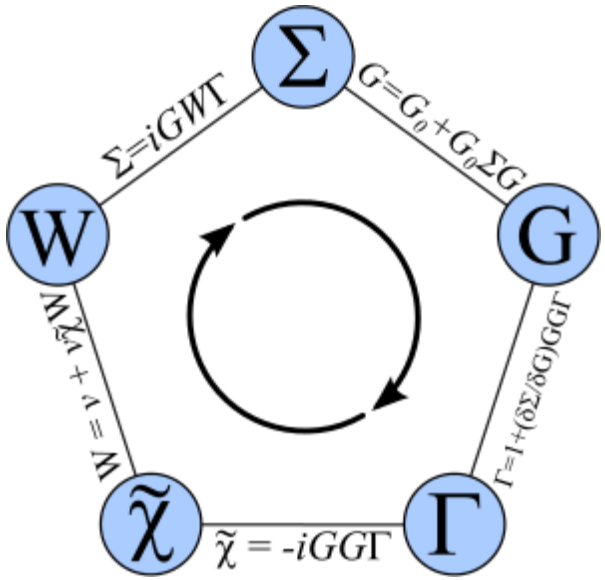
[1] P. Sun and G. Kotliar, Phys. Rev. B 66, 085120 (2002).
[2] S. Biermann, F. Aryasetiawan, and A. Georges, Phys. Rev. Lett. 90, 086402 (2003)
[3]F. Nilsson, L. Boehnke, P. Werner, and F. Aryasetiawan, Phys. Rev. Materials 1, 043803 (2017).

2. GW+EDMFT

[1] T. Ayral, S. Biermann, and P. Werner, Phys. Rev. B 87, 125149 (2013).

Hedin's equation

- Hedin's equation



$$\Sigma(1,2) = -\int d(34)G(1,3^+)W(1,4)\Gamma(3,2,4), \quad (1)$$

$$G(1,2) = G_0(1,2) + \int d(34)G_0(1,3)\Sigma(3,4)G(4,2), \quad (2)$$

$$\Gamma(1,2,3) = \delta(1-2)\delta(2-3), + \int d(4567)\frac{\partial\Sigma(1,2)}{\partial G(4,5)}G(4,6)G(7,5)\Gamma(6,7,3), \quad (3)$$

$$\Pi(1,2) = \int d(34)G(1,3)\Gamma(3,4,2)G(4,1^+), \quad (4)$$

$$W(1,2) = v(1,2) + \int d(34)v(1,3)\Pi(3,4)W(4,2). \quad (5)$$

- If $\Gamma(1,2,3) \approx \delta(1–2)\delta(2–3)$

$$\Sigma^{GW} = -G(1,2)W(1,2) \quad \Pi(1,2) = G(1,2)G(2,1^+)$$

[1]L. Hedin, Phys. Rev. 139, A796 (1965).

GW loop

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

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

$\Longleftrightarrow$

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

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

$\mathcal{G}$: fermionic Weiss field
$\mathcal{U}$: bosonic Weiss field
G_{imp}: impurity Green's function
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Extended Dynamical Mean Field Theory for U-V Hamiltonian

- DMFT formalism for models with spatially non-local interactions
- Model Hamiltonian example: U-V Hubbard model

$$H = -t \sum_{\langle ij\rangle\sigma} (c^{\dagger}_{i\sigma} c_{j\sigma} + \text{H.c.}) - \mu \sum_i n_i \; + U \sum_i n_{i\uparrow} n_{i\downarrow} + V \sum_{\langle ij\rangle} n_i n_j ,$$

- Actions formulation

$$S[c^*,c] = \int_0^\beta d\tau \Bigg\{ \sum_{ij\sigma} c^*_{i\sigma}(\tau)[(\partial_\tau - \mu)\delta_{ij} + t_{ij}]c_{j\sigma}(\tau) + U \sum_i n_{i\uparrow}(\tau) n_{i\downarrow}(\tau) + \frac{1}{2} \sum_{ij} v^{nl}_{ij} n_i(\tau) n_j(\tau) \Bigg\}.$$

- Rewriting the action using the identity $n_i n_i = 2 n_{i\uparrow} n_{i\downarrow} + n_i$

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

where $v_{ij} = U\delta_{ij} + V\delta_{\langle ij\rangle}$ and $\tilde{\mu} = \mu + U/2$

[1] Q. Si and J. L. Smith, Phys. Rev. Lett. 77, 3391 (1996).
[2] H. Kajueter, Ph.D. thesis, Rutgers University, New Brunswick,(1996).
[3]R. Chitra and G. Kotliar, Phys. Rev. Lett. 84, 3678 (2000).
[4] T. Ayral, S. Biermann, and P. Werner, Phys. Rev. B 87, 125149 (2013).

Extended Dynamical Mean Field Theory

• To map this problem onto single-site impurity model, we need to decouple spatially non-local interactions by using a Hubbard-stratonovich transformation for a real and ß-periodic field.

$$\exp\left(\frac{1}{2}\int_0^\beta d\tau\, b_i(\tau)A_{ij}b_j(\tau)\right)$$
$$= \int \frac{\mathcal{D}[x_1(\tau),x_2(\tau),\ldots]}{\sqrt{(2\pi)^N \det A}} \exp\left(\int_0^\beta d\tau\left\{-\frac{1}{2}x_i(\tau)[A^{-1}]_{ij}x_j(\tau) \mp x_i(\tau)b_i(\tau)\right\}\right)$$

• Then the action becomes

$$S[c^*,c,\phi] = \int_0^\beta d\tau\left\{-\sum_{ij\sigma} c^*_{i\sigma}(\tau)\left[\left(G_0^H\right)^{-1}\right]_{ij}c_{j\sigma}(\tau)\right\} + \int_0^\beta d\tau$$
$$\times\left\{\frac{1}{2}\sum_{ij}\phi_i(\tau)[v^{-1}]_{ij}\phi_j(\tau) + i\alpha\sum_i \phi_i(\tau)n_i(\tau)\right\},$$

where $[G_0^{H-1}]_{ij} \equiv [(-\partial_\tau + \mu + \frac{U}{2})\delta_{ij} - t_{ij}.]$

Bare fermionic and bosonic fields coupled by a local coupling

The physically relavant case corresponds to $\alpha = 1$

Extended Dynamical Mean Field Theory

- By integrating out all sites but one in the action in the infinite dimension limit

$$\begin{aligned} S^{\mathrm{EDMFT}} &= -\int_0^\beta d\tau\, d\tau' \sum_\sigma c_\sigma^*(\tau)\mathcal{G}^{-1}(\tau-\tau')c_\sigma(\tau') \\ &+ \frac{1}{2}\int_0^\beta d\tau\, d\tau' \phi(\tau)\mathcal{U}^{-1}(\tau-\tau')\phi(\tau') \\ &+ i\int_0^\beta d\tau \phi(\tau) n(\tau), \end{aligned}$$

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

- by integrating out the ϕ field

$$\begin{aligned} S^{\mathrm{EDMFT}} &= -\int_0^\beta d\tau\, d\tau' \sum_\sigma c_\sigma^*(\tau)\mathcal{G}^{-1}(\tau-\tau')c_\sigma(\tau') \\ &+ \frac{1}{2}\int_0^\beta d\tau\, d\tau' n(\tau)\mathcal{U}(\tau-\tau')n(\tau') - \frac{1}{2}\mathrm{Tr}\ln\mathcal{U}, \end{aligned}$$

- Green's function to be computed within EDMFT

$$G_{imp} = -\langle T c(\tau) c^*(0)\rangle$$

$$W_{imp} = \langle T\phi(\tau)\phi(0)\rangle$$

Extended Dynamical Mean Field Theory

- How to compute W_{imp}

$$W_{imp} = 2\frac{\delta lnZ}{\delta \mathcal{U}^{-1}} = 2\frac{\delta lnZ}{\delta \mathcal{U}}\frac{\delta \mathcal{U}}{\delta \mathcal{U}^{-1}} = -2\mathcal{U}\frac{\delta lnZ}{\delta \mathcal{U}}\mathcal{U} = -\mathcal{U}\langle Tn(\tau)n(0)\rangle\mathcal{U} + \mathcal{U}$$

$$W_{imp} = \mathcal{U} - \mathcal{U}\chi_{imp}\mathcal{U}$$

EDMFT loop

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

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

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

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

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

$\mathcal{G}$: fermionic Weiss field
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Σ_{DC}: double-counted self-energy
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GW+EDMFT as a electronic structure theory

- Within EDMFT, fermionic as well as bosonic self energy are local.
- a way to restore the non-local self energy by adding the first non-trivial non-local diagrammatic correction

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

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

A new Notation from now on

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

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

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

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

[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]P. Sun and G. Kotliar, Phys. Rev. Lett. 92, 196402 (2004).

GW+EDMFT loop

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

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

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

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

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

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

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

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

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

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

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

G: Green's function
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$\mathcal{G}$: fermionic Weiss field
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3. partial self-consistency within LQSGW+DMFT

GW+EDMFT loop

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

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

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

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

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

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

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

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

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

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

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

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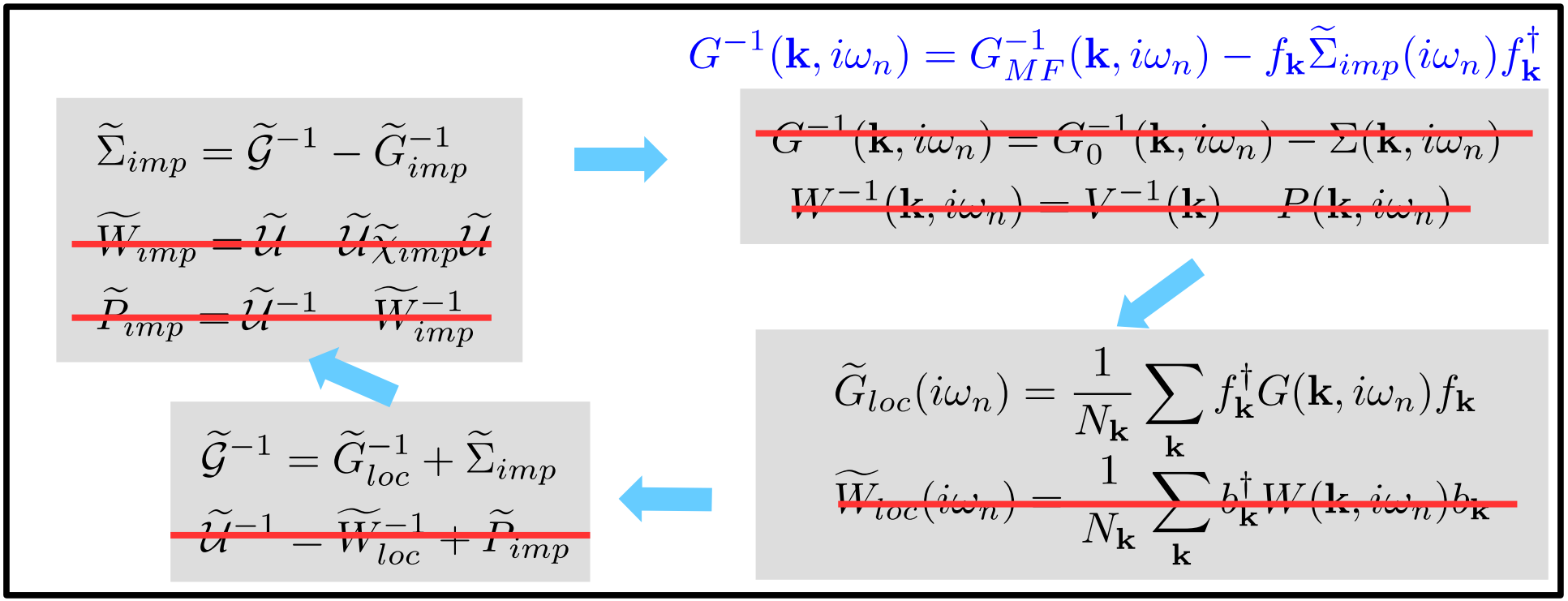
Various simplified approaches to GW+EDMFT

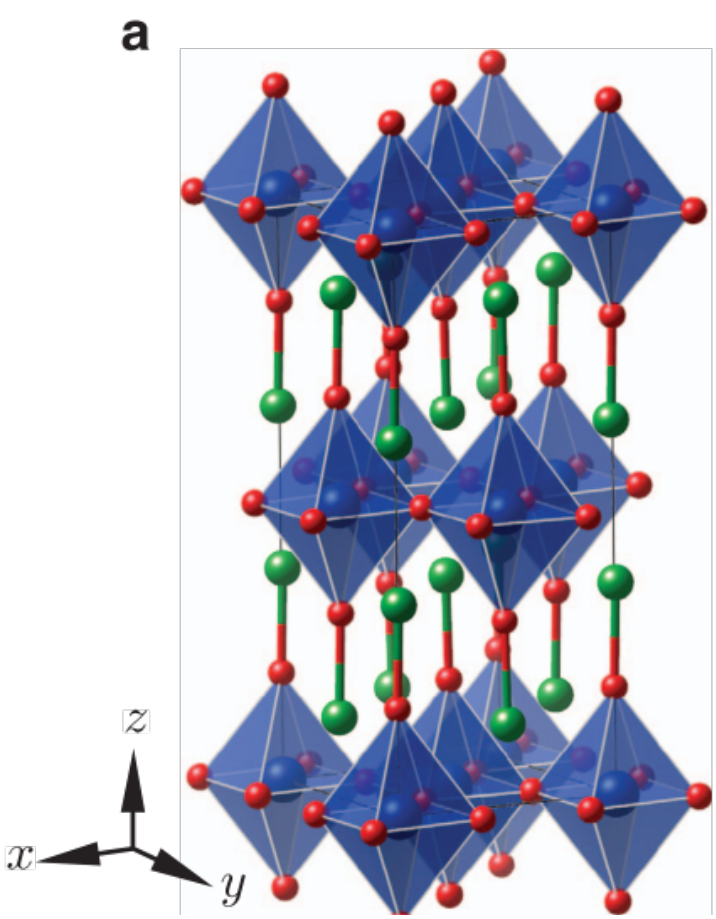
- fixing Bosonic quantities at the GW level
- U from constrained random phase approximation (cRPA) [1] and its extension [2]
- One-shot DMFT approach: G_{MF} is fixed
- For the construction of G_{MF}: one-shot GW [6,7], Screened Exchange [3], QSGW [4] and LQSGW [2], non-local QSGW and LQSGW [5]

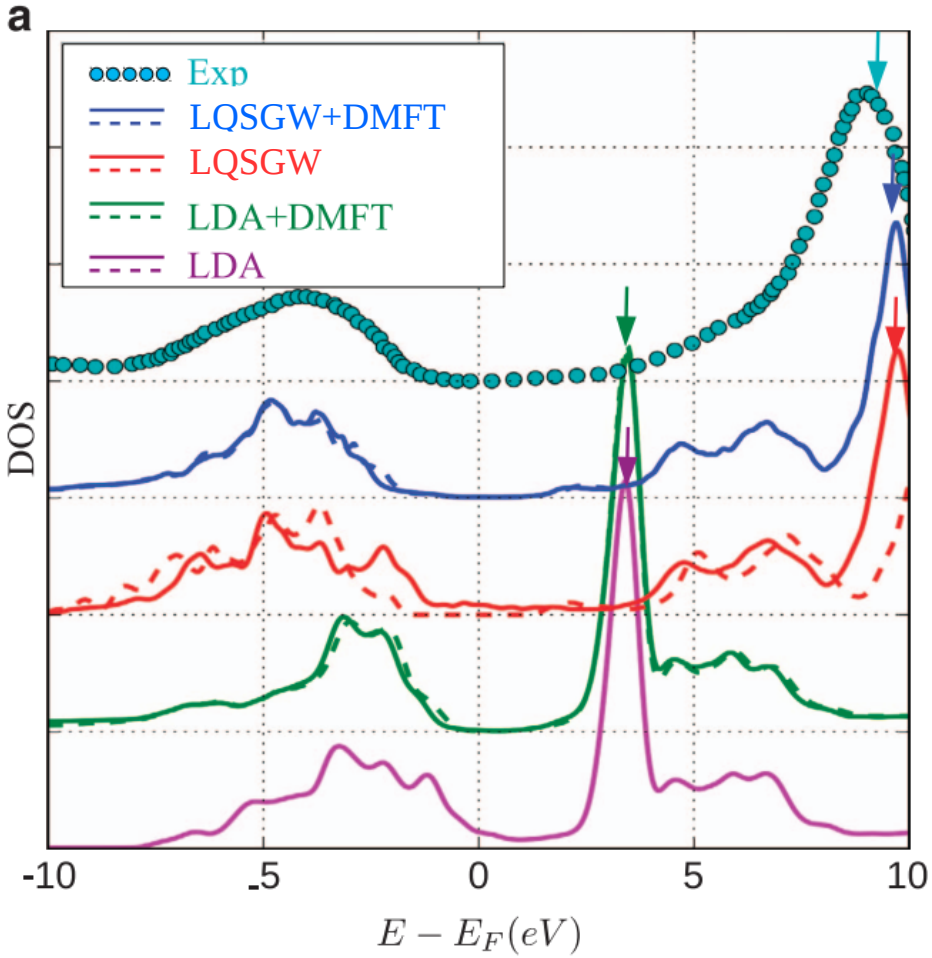


*) Multitier-GW+EDMFT [8] approach has been proposed: full GW+EDMFT approach in the low-energy regions

[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. Sponza et al., Phys. Rev. B 95, 041112 (2017).
[5] J. M. Tomczak, J. Phys.: Conf. Ser. 592, 012055 (2015). [6] J. M. Tomczak et al., Phys. Rev. B 90, 165138 (2014).
[7] C. Taranto et al., Phys. Rev. B 88, 165119 (2013). [8] F. Nilsson et al., Phys. Rev. Materials 1, 043803 (2017).

LQSGW+DMFT validation on La2CuO4

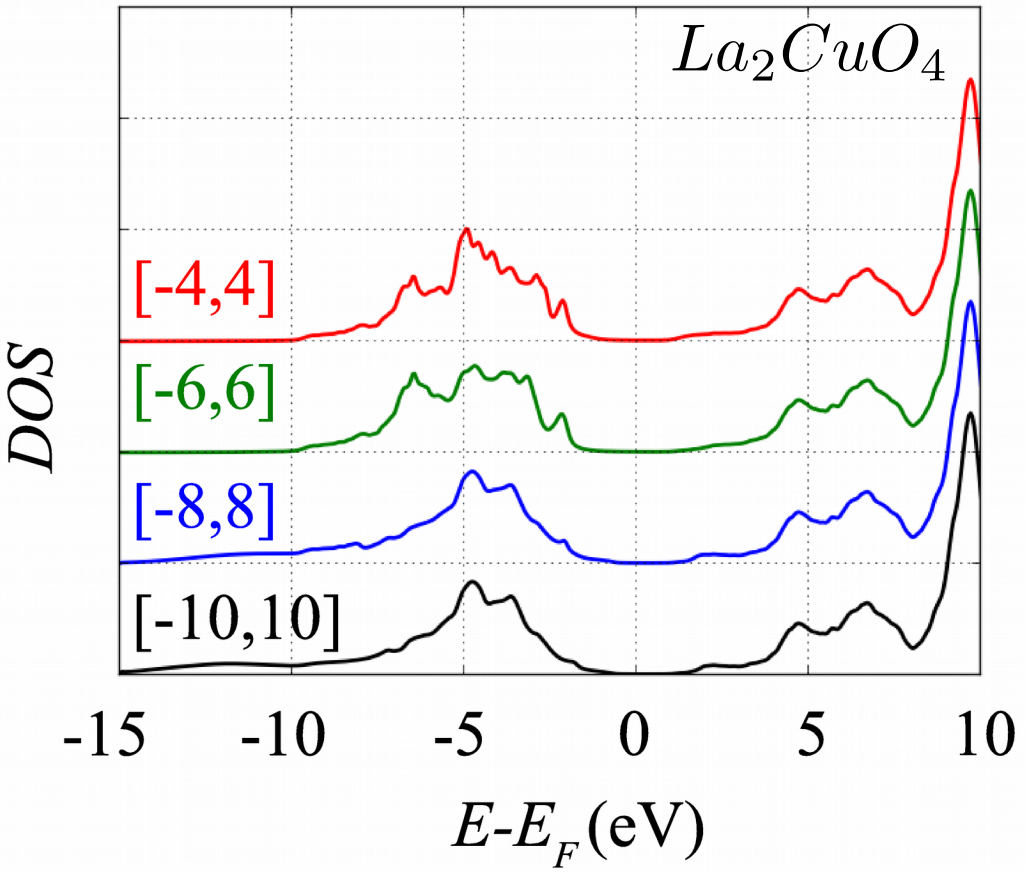


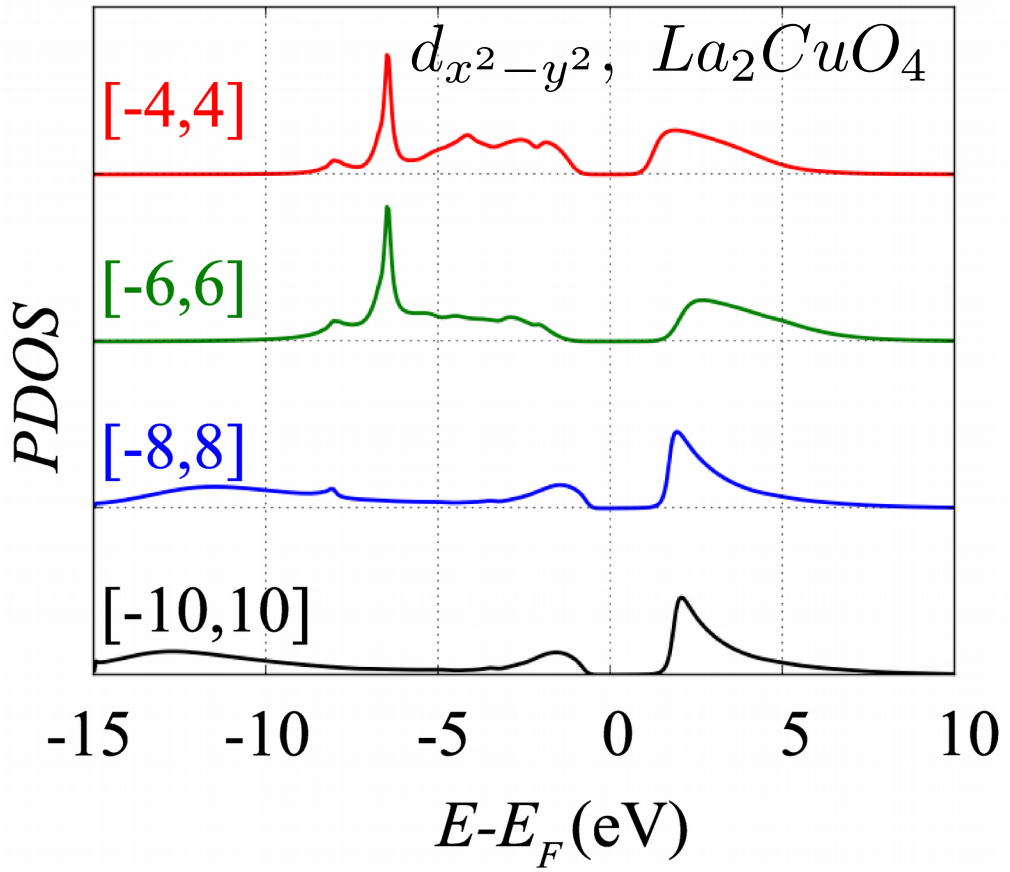


- LDA+DMFT and LQSGW+DMFT opens a paramagnetic Mott gap
- LQSGW predict a metal in non-magnetic calculation and an insulator in spin-polarized calculation. But its gap is too big (3.4 eV vs ~2 eV)
- LDA predict a metallic phase
- better O-p and La-f position within LQSGW+DMFT

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

Choice of local orbitals

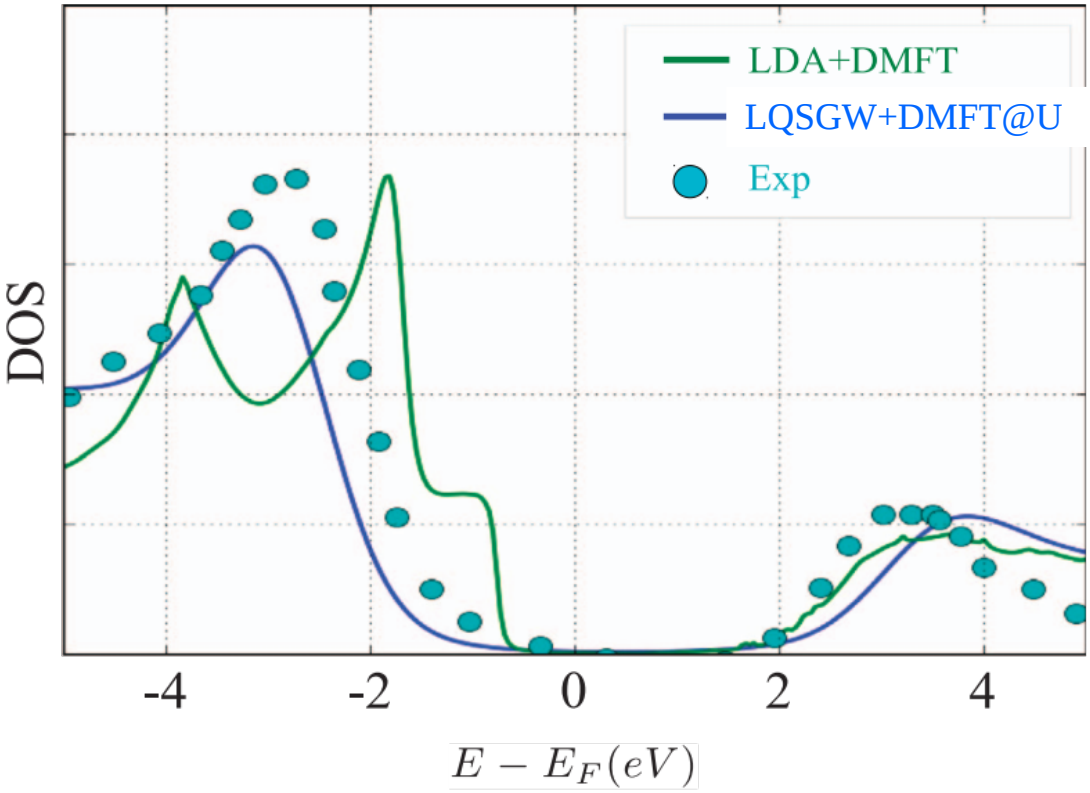




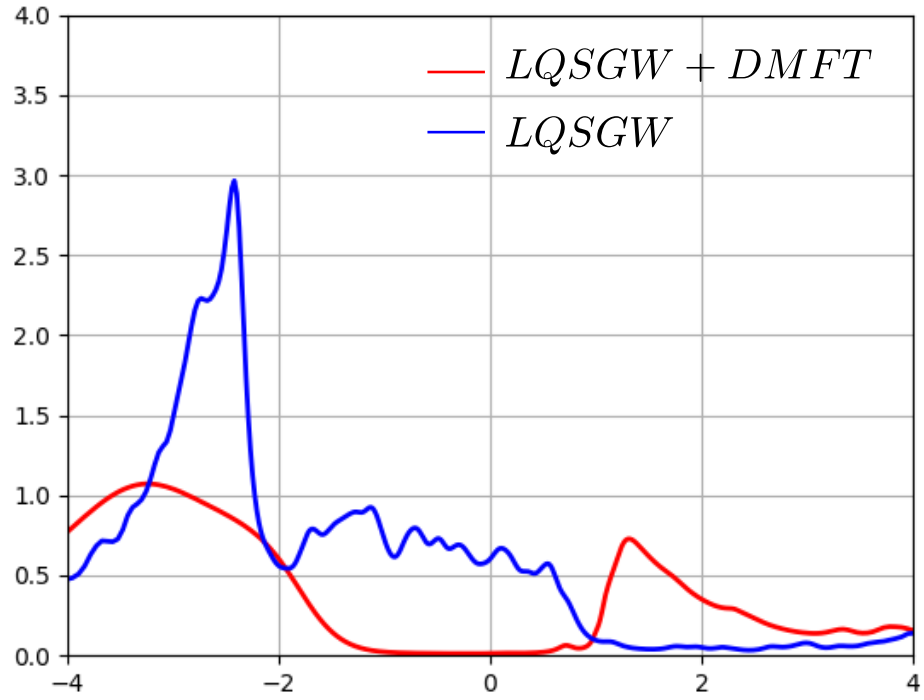
- U and Σ_{DC} are self-consistently determined
- Identical Mott gap regardless of the choice of orbitals
- The position of Lower Hubbard band is sensitive to the choice

LQSGW+DMFT validation on NiO

Static U



Dynamical U



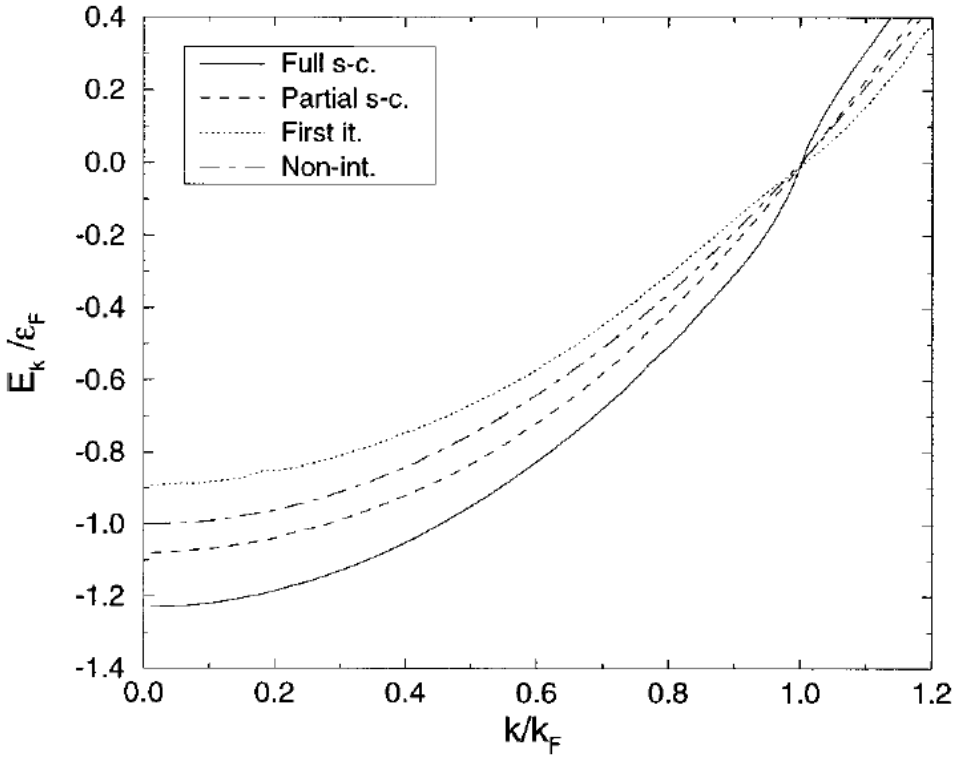
- a proper choice of static U gives similar spectral function to Exp and full LQSGW+DMFT

Linearized Quasiparticle Self-consistent GW (LQSGW)

- Why LQSGW, not one-shot GW or Full GW?

Full GW

Electron gas



- G_0W_0: band narrowing (consistent with exp)
- GW: band widening. Worse than G_0W_0

[1]B. Holm and U. von Barth, Phys. Rev. B 57, 2108 (1998).

Oneshot GW

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

- Starting point dependent (PBE vs PBE0)

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

Projector (f_k) construction by using Wannier functions

- From tight-binding model

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

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

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

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

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

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

- then a projector to correlated orbitals

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

Product basis (b_k) construction by using Wannier functions

- With products of Wannier functions for correlated orbitals

$$B_{\tau,\tau'}(\mathbf{r}) = W_{\mathbf{R}=\mathbf{0}\tau}(\mathbf{r}) W^*_{\mathbf{R}=\mathbf{0}\tau'}(\mathbf{r})$$

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

$$\langle \mathbf{r} | c_b \rangle = \sum_{\tau,\tau'} X_{\tau,\tau';b} B_{\tau,\tau'}(\mathbf{r})$$

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

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

$$\sum_{\tau_3,\tau_4} O_{\tau_1,\tau_2;\tau_3,\tau_4} D_{\tau_3,\tau_4;b} = F_b D_{\tau_1,\tau_2;b}$$

$$X_{\tau_1,\tau_2;b} = \frac{1}{\sqrt{F_b}} D_{\tau_1,\tau_2;b}$$

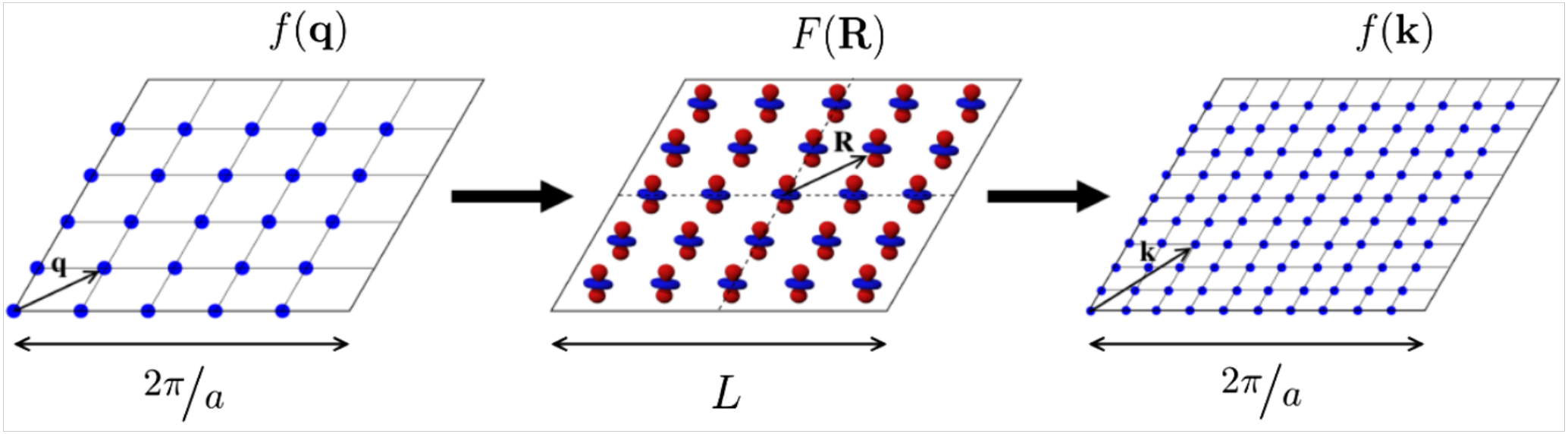
Wannier-interpolation of LQSGW bandstructure (H_{QP})

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

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

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

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



Coulomb interaction matrix from cRPA

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

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

Aim: calculate the effective interaction between the correlated orbitals.

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

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

How to define P_{QP}^{low}

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

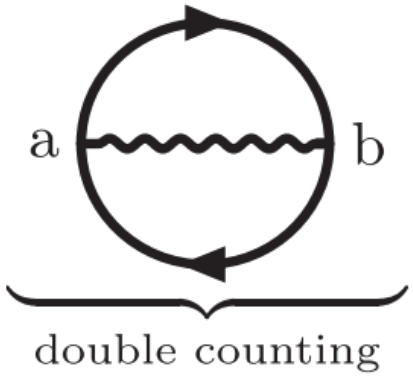
$$P_{QP} = P_{QP}^{low} + P_{QP}^{high}$$

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

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

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

Double counting energy



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

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

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

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

LQSGW+DMFT loop

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

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

$\Leftrightarrow$

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

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

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

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

$\Downarrow$

$$f_k = \langle \mathbf{r}|\tau\mathbf{k}\rangle = \sum_n U^*_{n\tau}(\mathbf{k})\langle \mathbf{r}|n\mathbf{k}\rangle \qquad b_{\mathbf{k}} = \sum_{\tau,\tau'} X_{\tau,\tau';b}\langle \mathbf{r}|\mathbf{R}=0,\tau\rangle\langle \mathbf{R}=0,\tau'|\mathbf{r}\rangle$$

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

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

LQSGW+DMFT loop

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

$$\widetilde{\mathcal{U}}(i\omega_n) = \widetilde{W}_r(\mathbf{R} = 0, i\omega_n)$$

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

LQSGW+DMFT loop

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

$$G^{-1}(\mathbf{k}, i\omega_n) = i\omega_n - H^{nl}_{QP}(\mathbf{k}) - f_{\mathbf{k}}\widetilde{\Sigma}_{imp}(i\omega_n)f^{\dagger}_{\mathbf{k}}$$

$$H^{nl}_{QP}(\mathbf{k}) = \sqrt{Z^{-1}_{DC}(\mathbf{k})}H_{QP}\sqrt{Z^{-1}_{DC}(\mathbf{k})} - f_{\mathbf{k}}\widetilde{\Sigma}_{DC}(\omega=0)f^{\dagger}_{\mathbf{k}}$$

$$Z^{-1}_{DC}(\mathbf{k}) = f_{\mathbf{k}}\left(1 - \frac{\partial\Sigma_{DC}(\mathbf{k},\omega=0)}{i\omega_n}\right)f^{\dagger}_{\mathbf{k}}$$

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

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

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

CTQMC with dynamic interactions and GPU acceleration

CTQMC

$$S = -\sum_{ij} \iint c_i^\dagger(\tau)\mathcal{G}^{-1}_{0,ij}(\tau-\tau')c_i(\tau')d\tau d\tau' + \sum_{ijkl} \iint c_i^\dagger(\tau^+)c_j^\dagger(\tau'^+)\mathcal{U}_{ijkl}(\tau-\tau')c_k(\tau')c_l(\tau)d\tau d\tau'$$

- **Split** Impurity Model action $S = S_A + S_B$
- **Expand** $Z = \int D[c^\dagger, c]e^{-S_A - S_B} = \sum_n \int D[c^\dagger, c]e^{-S_A}(-S_B)^n$
- **Sample** Z and get observables

$$\Rightarrow \quad \int D[c^\dagger, c]e^{-S_A} \circ \quad \text{should be } \textbf{easy} \text{ to evaluate !}$$

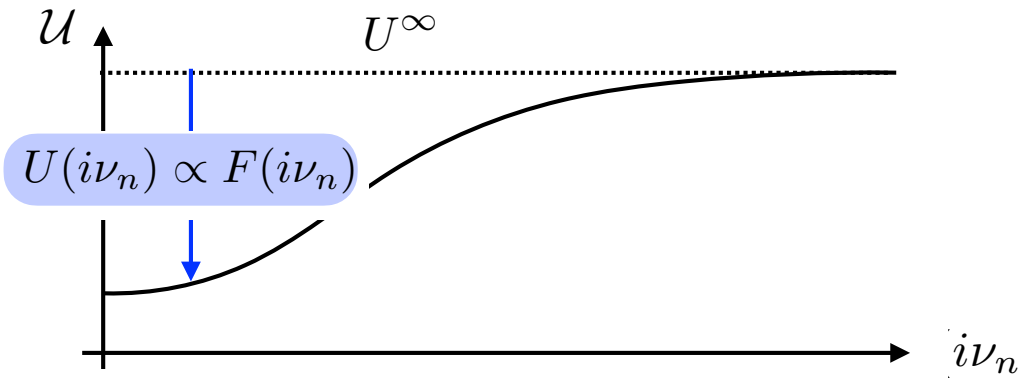
	S_B	S_A	$\int D[c^\dagger, c]e^{-S_A} \circ$	retarded interactions
CT-INT	interactions	quadratic	Wick	✓
CT-HYB	hybridisation	few degrees of freedom	matrices or „segments“	?

Split the action

$$S = -\sum_{ij} \iint c_i^\dagger(\tau)\mathcal{G}^{-1}_{0,ij}(\tau-\tau')c_i(\tau')d\tau d\tau' + \sum_{ijkl} \iint c_i^\dagger(\tau^+)c_j^\dagger(\tau'^+)\mathcal{U}_{ijkl}(\tau-\tau')c_k(\tau')c_l(\tau)d\tau d\tau'$$

- $\mathcal{G}^{-1}_{0,ij}(i\omega_n) = i\omega_n + \mu - \epsilon_{ij} - \Delta_{ij}(i\omega_n)$ $\Rightarrow$ $S_{\text{hyb}} := \sum_{ij} \iint c_i^\dagger(\tau)\Delta_{ij}(\tau-\tau')c_j(\tau')d\tau d\tau'$

- $\mathcal{U}_{ijkl}(i\nu_n) = U^\infty_{ijkl} + U_{ijkl}(i\nu_n)$ $\Rightarrow$ $S_{\text{dyn}} := \sum_{IJ} \iint Q_I^\dagger(\tau)F_{IJ}(\tau-\tau')Q_J(\tau')d\tau d\tau'$



$$Q_I^\dagger := \sum_{il} c_i^\dagger c_l \langle i,l|I\rangle$$

$$U_{ijkl}(i\nu_n) = \sum_{IJ} \langle i,l|I\rangle F_{IJ}(i\nu_n)\langle J|k,j\rangle$$

- $H_{\text{loc}} := \sum_{ij} c_i^\dagger(\epsilon_{ij} - \mu\delta_{ij})c_j + \sum_{ijkl} c_i^\dagger c_j^\dagger U^\infty_{ijkl}c_k c_l$ $\Rightarrow$ $S_{\text{loc}} := \int \sum_i c_i^\dagger(\tau)\partial_\tau c_i(\tau) + H_{\text{loc}}[c^\dagger(\tau), c(\tau)]d\tau$

$$S = S_{\text{loc}} + S_{\text{hyb}} + S_{\text{dyn}}$$

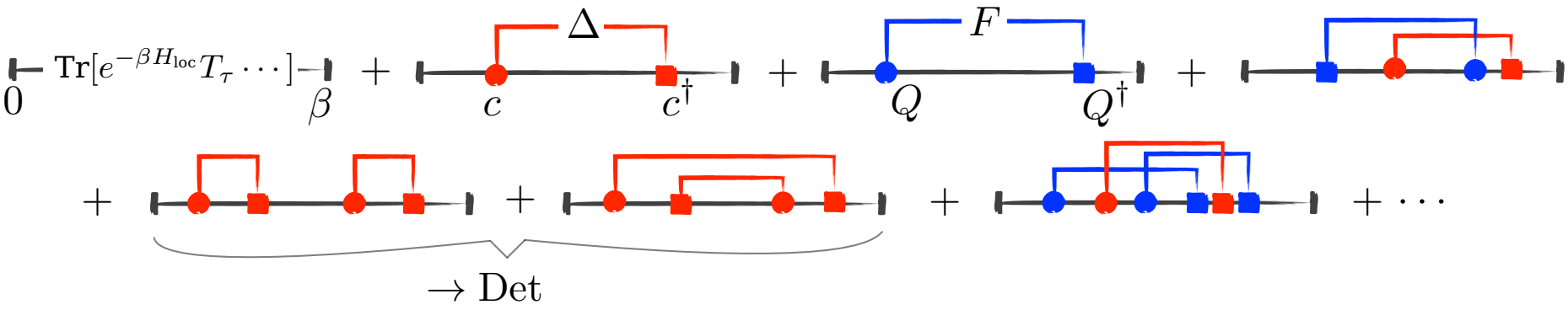
Expand the partition function

$$S_{\rm hyb} := \sum_{ij} \iint c_i^\dagger(\tau)\Delta_{ij}(\tau-\tau')c_j(\tau')d\tau d\tau' \qquad S_{\rm dyn} := \sum_{IJ} \iint Q_I^\dagger(\tau)F_{IJ}(\tau-\tau')Q_J(\tau')d\tau d\tau'$$

- $$Z = \int D[c,c^\dagger]e^{-S_{\rm loc}-S_{\rm hyb}-S_{\rm dyn}} = \sum_{nm}\frac{1}{n!m!}\int D[c,c^\dagger]e^{-S_{\rm loc}}(-S_{\rm hyb})^n(-S_{\rm dyn})^m$$

$$= \sum_{nm}\frac{(-1)^{n+m}}{m!n!}\sum_{\substack{i_1\cdots i_n\\ j_1\cdots j_n}}\int_0^\beta d\tau_1\cdots d\tau_n\int_0^\beta d\tau_1'\cdots d\tau_n' \sum_{\substack{I_1\cdots I_m\\ J_1\cdots J_m}}\int_0^\beta d\tilde\tau_1\cdots d\tilde\tau_m\int_0^\beta d\tilde\tau_1'\cdots d\tilde\tau_m'$$

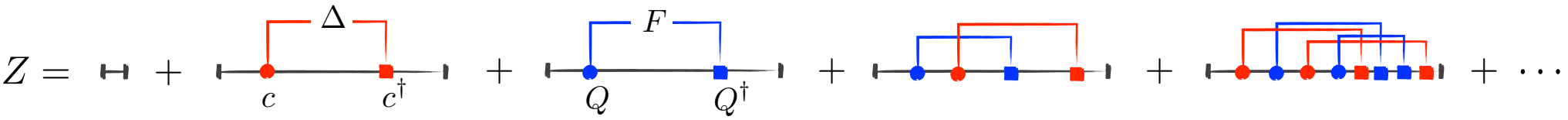
$$\times\,{\rm Tr}[e^{-\beta H_{\rm loc}}T_\tau\prod_{r=1}^n c_{i_r}^\dagger(\tau_r)c_{j_r}(\tau_r')\prod_{s=1}^m Q_{I_s}^\dagger(\tilde\tau_s)Q_{J_s}(\tilde\tau_s')]\times\prod_{r=1}^n\Delta_{i_rj_r}(\tau_r-\tau_r')\times\prod_{s=1}^m F_{I_sJ_s}(\tilde\tau_s-\tilde\tau_s')$$

$=$ 

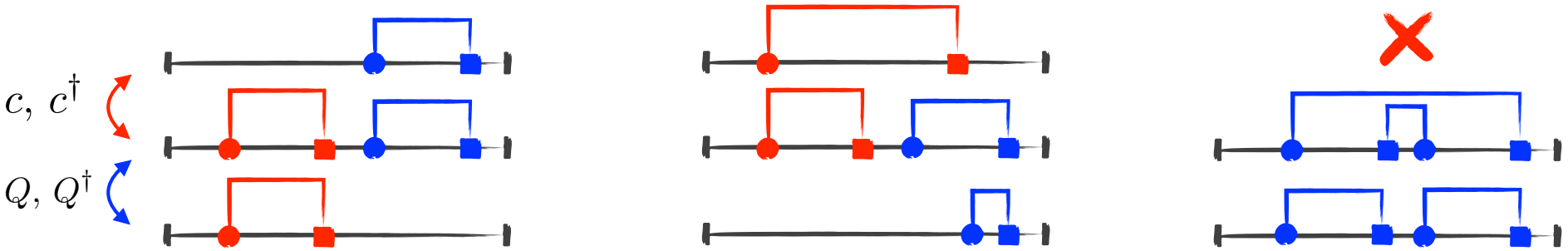
- $\prod_{r=1}^n\Delta_{i_rj_r}(\tau_r-\tau_r') \quad\to\quad {\rm Det}[\Delta_{i_kj_l}(\tau_k-\tau_l')]$ **crucial !**

$\prod_{s=1}^m F_{I_sJ_s}(\tilde\tau_s-\tilde\tau_s') \quad\to\quad {\rm Perm}[F_{I_kJ_l}(\tilde\tau_k-\tilde\tau_l')]$ **not convenient !**

(Dress and) Sample

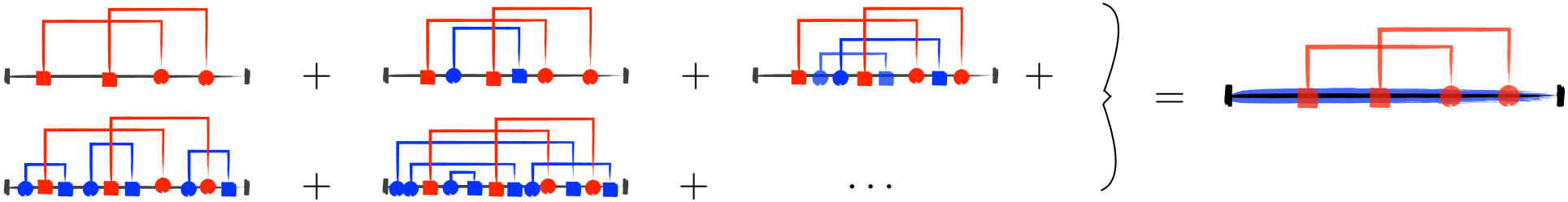


$[H_{\rm loc}, Q] \neq 0$: **Sample** by updating $c,\ c^\dagger$ **or** $Q,\ Q^\dagger,$ for example



(insert/remove) (shift) (reconnect)

$[H_{\rm loc}, Q] = 0$:$^{(*)}$ **a) Dress**



b) Sample by updating $c,\ c^\dagger$ **only**

(*): Additional terms and conditions apply ….

Dress the expansion

- $$Z = \sum_{nm} \frac{(-1)^m}{m!} \sum_{\substack{i_1 \cdots i_n \\ j_1 \cdots j_n}} \int_> d\tau_1 \cdots d\tau_n \int_> d\tau'_1 \cdots d\tau'_n \sum_{\substack{I_1 \cdots I_m \\ J_1 \cdots J_m}} \int d\tilde{\tau}_1 \cdots d\tilde{\tau}_m \int d\tilde{\tau}'_1 \cdots d\tilde{\tau}'_m$$
$$\times \mathrm{Tr}[e^{-\beta H_{\mathrm{loc}}} T_\tau \prod_{r=1}^{n} c_{j_r}(\tau'_r) c^\dagger_{i_r}(\tau_r) \prod_{s=1}^{m} Q^\dagger_{I_s}(\tilde{\tau}_s) Q_{J_s}(\tilde{\tau}'_s)] \times \mathrm{Det}[\Delta_{i_l j_k}(\tau_k - \tau'_l)] \times \prod_{s=1}^{m} F_{I_s J_s}(\tilde{\tau}_s - \tilde{\tau}'_s)$$

- Assume

(i) $[H_{\mathrm{loc}}, Q_J] = 0$

(ii) $[Q^\dagger_I, c^\dagger_i] = q_{iI} \cdot c^\dagger_i$ and $[Q^\dagger_I, c_i] = -q_{iI} \cdot c_i$

(iii) $[Q_I, Q_J] = [Q^\dagger_I, Q_J] = 0$

(iv) $\exists K_{IJ}$ β-periodic such that $K''_{IJ} = F_{IJ}$

$$\Rightarrow Z = \sum_{n} \sum_{\substack{i_1 \cdots i_n \\ j_1 \cdots j_n}} \int_> d\tau_1 \cdots d\tau_n \int_> d\tau'_1 \cdots d\tau'_n \mathrm{Tr}[e^{-\beta H_{\mathrm{loc}}} T_\tau \prod_{r=1}^{n} c_{j_r}(\tau'_r) c^\dagger_{i_r}(\tau_r)] \times \mathrm{Det}[\Delta(\tau_k - \tau'_l)]$$
$$\times \exp\Big[\sum_{r,s=1}^{n} \sum_{IJ} q_{i_r I} q_{j_s J} \big(K_{IJ}(\tau_r - \tau_s) - K_{IJ}(\tau_r - \tau'_s) - K_{IJ}(\tau'_r - \tau_s) + K_{IJ}(\tau'_r - \tau'_s) \big) \Big]$$

Werner, P., and A. J. Millis (2010), Phys. Rev. Lett. 104 (14), 146401

T. Ayral, S. Biermann, and P. Werner, Phys. Rev. B 87, 125149

Dress the expansion

$(i)\quad [H_{\rm loc}, Q_J] = 0$ $\qquad$ $(ii)\quad [Q_I^\dagger, c_i^\dagger] = q_{iI}\cdot c_i^\dagger \quad \text{and} \quad [Q_I^\dagger, c_i] = -q_{iI}\cdot c_i$

$(iii)\quad [Q_I, Q_J] = [Q_I^\dagger, Q_J] = 0$ $\qquad$ $(iv)\quad \exists K_{IJ}\ \beta\text{-periodic such that } K''_{IJ} = F_{IJ}$

$$\Rightarrow \frac{d}{d\tilde{\boldsymbol{\tau}}}\mathrm{Tr}[e^{-\beta H_{\rm loc}} T_\tau c(\tau') c^\dagger(\tau) \boldsymbol{Q^\dagger(\tilde{\tau})} Q(\tilde{\tau}')] =$$

$$\big(\boldsymbol{q}\cdot\delta(\tilde{\boldsymbol{\tau}}-\tau) - \boldsymbol{q}\cdot\delta(\tilde{\boldsymbol{\tau}}-\tau')\big)\mathrm{Tr}[e^{-\beta H_{\rm loc}} T_\tau c(\tau') c^\dagger(\tau) Q(\tilde{\tau}')]$$

$$\Rightarrow \int_0^\beta \overbrace{F(\tilde{\boldsymbol{\tau}}-\tilde{\tau}')}^{f'}\overbrace{\mathrm{Tr}[\cdots \boldsymbol{Q^\dagger(\tilde{\tau})} Q(\tilde{\tau}')]}^{g} d\tilde{\boldsymbol{\tau}} = K'(\tilde{\boldsymbol{\tau}}-\tilde{\tau}')\mathrm{Tr}[\cdots \boldsymbol{Q^\dagger(\tilde{\tau})} Q(\tilde{\tau}')]\Big|_{\tilde{\boldsymbol{\tau}}=0}^{\tilde{\boldsymbol{\tau}}=\beta}$$

vanish by (iv)

$$-\boldsymbol{q}(K'(\tau-\tilde{\tau}') - K'(\tau'-\tilde{\tau}'))\mathrm{Tr}[\cdots Q(\tilde{\tau}')]$$

$$\Rightarrow \iint_0^\beta F(\tilde{\boldsymbol{\tau}}-\tilde{\boldsymbol{\tau}}')\mathrm{Tr}[\cdots \boldsymbol{Q^\dagger(\tilde{\tau})Q(\tilde{\tau}')}] d\tilde{\boldsymbol{\tau}} d\tilde{\boldsymbol{\tau}}' = \{\ \text{boundary terms}\ \}\Big|_{\tilde{\boldsymbol{\tau}}'=0}^{\tilde{\boldsymbol{\tau}}'=\beta}$$

vanish by (iv)

$$-\boldsymbol{q^2}(K(\tau-\tau) - K(\tau-\tau') - K(\tau'-\tau) + K(\tau'-\tau'))\mathrm{Tr}[\cdots]$$

Dress the expansion

- $$Z = \sum_{nm} \frac{(-1)^m}{m!} \sum_{\substack{i_1 \cdots i_n \\ j_1 \cdots j_n}} \int_> d\tau_1 \cdots d\tau_n \int_> d\tau'_1 \cdots d\tau'_n \sum_{\substack{I_1 \cdots I_m \\ J_1 \cdots J_m}} \int d\tilde{\tau}_1 \cdots d\tilde{\tau}_m \int d\tilde{\tau}'_1 \cdots d\tilde{\tau}'_m$$
$$\times \mathrm{Tr}[e^{-\beta H_{\mathrm{loc}}} T_\tau \prod_{r=1}^{n} c_{j_r}(\tau'_r) c^\dagger_{i_r}(\tau_r) \prod_{s=1}^{m} Q^\dagger_{I_s}(\tilde{\tau}_s) Q_{J_s}(\tilde{\tau}'_s)] \times \mathrm{Det}[\Delta_{i_l j_k}(\tau_k - \tau'_l)] \times \prod_{s=1}^{m} F_{I_s J_s}(\tilde{\tau}_s - \tilde{\tau}'_s)$$

- $$\int\int_0^\beta d\boldsymbol{\tau} d\tilde{\boldsymbol{\tau}} \mathrm{Tr}[e^{-\beta H_{\mathrm{loc}}} T_\tau \prod_{r=1}^{n} c(\tau'_r) c^\dagger(\tau_r) \boldsymbol{Q^\dagger(\tilde{\tau}) Q(\tilde{\tau}')}] F(\tilde{\boldsymbol{\tau}} - \tilde{\boldsymbol{\tau}}') =$$
$$- \sum_{r,s=1}^{n} \boldsymbol{q^2} \big(K(\tau_r - \tau_s) - K(\tau_r - \tau'_s) - K(\tau'_r - \tau_s) + K(\tau'_r - \tau'_s)\big) \mathrm{Tr}[e^{-\beta H_{\mathrm{loc}}} T_\tau \prod_{r=1}^{n} c(\tau'_r) c^\dagger(\tau_r)]$$

$$\Rightarrow Z = \sum_{n} \sum_{\substack{i_1 \cdots i_n \\ j_1 \cdots j_n}} \int_> d\tau_1 \cdots d\tau_n \int_> d\tau'_1 \cdots d\tau'_n \mathrm{Tr}[e^{-\beta H_{\mathrm{loc}}} T_\tau \prod_{r=1}^{n} c_{j_r}(\tau'_r) c^\dagger_{i_r}(\tau_r)] \times \mathrm{Det}[\Delta_{i_k j_l}(\tau_k - \tau'_l)]$$
$$\times \exp\Big[\sum_{r,s=1}^{n} \sum_{IJ} q_{i_r I} q_{j_s J} \big(K_{IJ}(\tau_r - \tau_s) - K_{IJ}(\tau_r - \tau'_s) - K_{IJ}(\tau'_r - \tau_s) + K_{IJ}(\tau'_r - \tau'_s)\big) \Big]$$

Assumptions (i-iii)

$(i)\quad [H_{\rm loc}, Q_J] = 0 \qquad (ii)\quad [Q_I^\dagger, c_i^\dagger] = q_{iI}\cdot c_i^\dagger \quad \text{and} \quad [Q_I^\dagger, c_i] = -q_{iI}\cdot c_i \qquad (iii)\quad [Q_I, Q_J] = [Q_I^\dagger, Q_J] = 0$

- **Density-Density**

$$H_{\rm loc} = \sum_i (\epsilon_i - \mu) n_i + \sum_{ij} U_{ij} n_i n_j \qquad \Rightarrow \qquad Q \in \{n_i\} \qquad (n_i := c_i^\dagger c_i)$$

$$S = -\int \sum_i c_i^\dagger(\tau) \mathcal{G}_{0,i}^{-1}(\tau - \tau') c_i(\tau') d\tau d\tau' + \int \sum_{ij} n_i(\tau) \mathcal{U}_{ij}(\tau - \tau') n_j(\tau') d\tau d\tau'$$

- **General**

$$H_{\rm loc} = \sum_{ij} c_i^\dagger (\epsilon_{ij} - \mu\delta_{ij}) c_j + \sum_{ijkl} c_i^\dagger c_j^\dagger U_{ijkl} c_k c_l \quad \overset{\text{always}}{\Rightarrow} \quad Q = N := \sum_i c_i^\dagger c_i$$

$SU(2)\times SO(3):\quad S_z,\quad L_z$ (depends on basis c_i) (ii): ~~S^2, L^2~~

$SO(3):\quad J_z$ (depends on basis c_i) (ii): ~~J^2~~

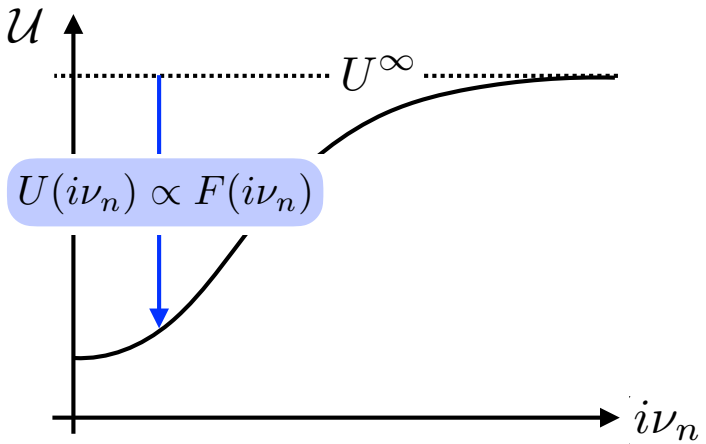
Assumption (iv)

(iv) $\exists K_{IJ}$ β-periodic such that $K''_{IJ} = F_{IJ}$

- $S_{\text{dyn}} := \iint Q^\dagger(\tau) F(\tau - \tau') Q(\tau') d\tau d\tau'$

  $\Rightarrow F(\tau) = \beta^{-1} \sum_n F(i\nu_n) e^{-i\nu_n \tau}$ $\qquad F(i\nu_n) \sim (i\nu_n)^{-2}$

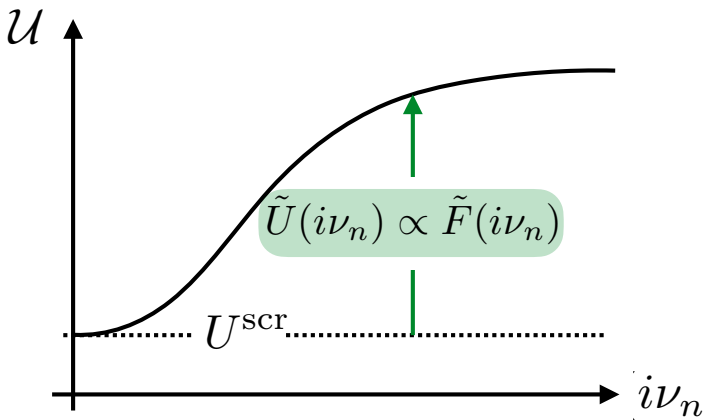
  $\Rightarrow K(\tau) = \frac{1}{2\beta} F(i\nu_n = 0)\tau^2 + A\tau + B + \sum_{n\neq 0} \frac{F(i\nu_n)}{\beta (i\nu_n)^2} e^{-i\nu_n \tau}$ ✗ ✓



- $\tilde{S}_{\text{dyn}} := S_{\text{dyn}} - \int Q^\dagger(\tau^+) F(i\nu_n = 0) Q(\tau) d\tau$

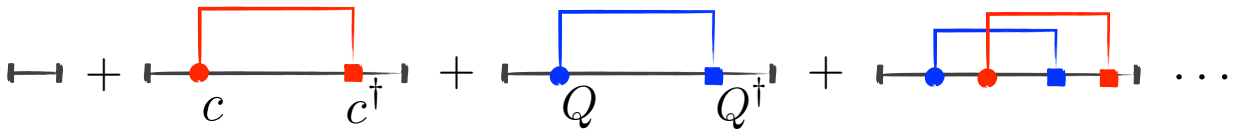
  $\Rightarrow \tilde{F}(\tau) = F(\tau) - F(i\nu_n = 0)\delta(\tau)$

  $\Rightarrow \tilde{K}(\tau) = \frac{1}{2\beta} F(i\nu_n = 0)|\tau|(|\tau| - \beta) + \sum_{n\neq 0} \frac{F(i\nu_n)}{\beta (i\nu_n)^2} e^{-i\nu_n \tau}$ ✓ ✓



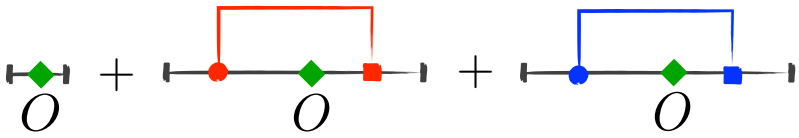
$\Rightarrow \tilde{H}_{\text{loc}} := H_{\text{loc}} + Q^\dagger F(i\nu_n = 0) Q = \sum_{ij} c_i^\dagger (\epsilon_{ij}^{\text{scr}} - \mu\delta_{ij}) c_j + \sum_{ijkl} c_i^\dagger c_j^\dagger U_{ijkl}^{\text{scr}} c_k c_l$

Observables

$$Z = \int D[c^\dagger, c] e^{-S} = \sum_{\mathcal{C},\mathcal{Q}} w(\mathcal{C},\mathcal{Q}) =$$ 

$S = S_{\rm loc} + c^\dagger \Delta c + Q^\dagger F Q$ $\quad \mathcal{C} \leftrightarrow c, c^\dagger$ configuration $\quad \mathcal{Q} \leftrightarrow Q, Q^\dagger$ configuration

- Expand observables as

$$\langle O \rangle = Z^{-1} \int D[c, c^\dagger] e^{-S} O = Z^{-1} \times ($$ 

$$)$$

or use source fields

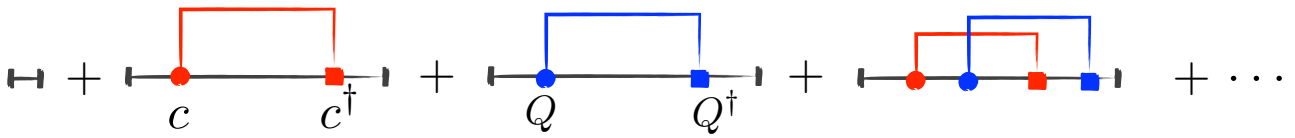
$$-\frac{\partial}{\partial \Delta} \ln Z = -\langle c\, c^\dagger \rangle = G \quad \Rightarrow \quad G = -Z^{-1} \sum_{\mathcal{C},\mathcal{Q}} \frac{\partial}{\partial \Delta} w(\mathcal{C},\mathcal{Q}) \quad \text{(Green Function)}$$

$$-\frac{\partial}{\partial F} \ln Z = \langle Q\, Q^\dagger \rangle = \chi \quad \Rightarrow \quad \chi = -Z^{-1} \sum_{\mathcal{C},\mathcal{Q}} \frac{\partial}{\partial F} w(\mathcal{C},\mathcal{Q}) \quad \text{(Susceptibility)}$$

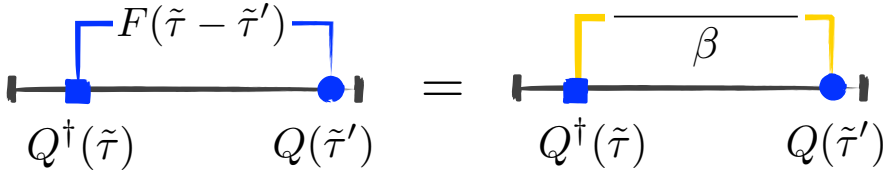
- Write $\langle O \rangle = Z^{-1} \sum_{\mathcal{C},\mathcal{Q}} o(\mathcal{C},\mathcal{Q}) w(\mathcal{C},\mathcal{Q})$ if possible !

Susceptibilities

$$\chi(i\nu_n) = -Z^{-1} \sum_{\mathcal{C},\mathcal{Q}} \frac{\partial w(\mathcal{C},\mathcal{Q})}{\partial F(i\nu_n)}$$

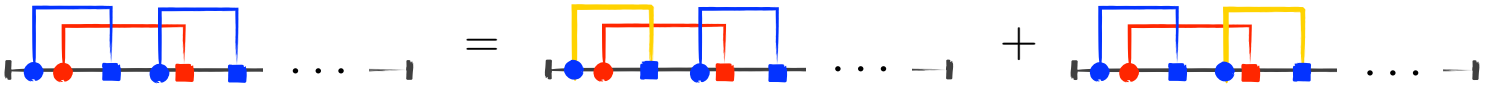
- $Z = \sum_{\mathcal{C},\mathcal{Q}} w(\mathcal{C},\mathcal{Q}) =$ 

$\mathcal{C} \leftrightarrow c, c^\dagger$ configuration $\quad \mathcal{Q} \leftrightarrow Q, Q^\dagger$ configuration

- $\frac{\partial}{\partial F(i\nu_n)}$ 

$=$

$$F(\tau) = \beta^{-1} \sum_n F(i\nu_n) e^{-i\nu_n \tau}$$

$\Rightarrow$ $\frac{\partial}{\partial F(i\nu_n)}$ 

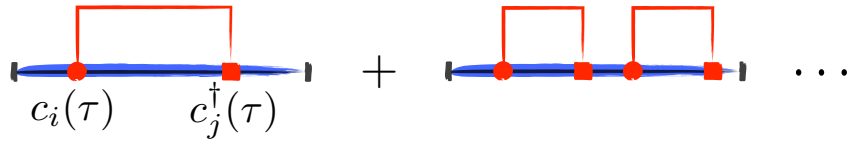
$$\mathbf{F \neq 0}: \quad \frac{\partial}{\partial F(i\nu_n)} \equiv \times \frac{e^{i\nu_n(\tilde{\tau}'-\tilde{\tau})}}{\beta F(\tilde{\tau}-\tilde{\tau}')} \Rightarrow \chi(i\nu_n) = -Z^{-1} \sum_{\mathcal{C},\mathcal{Q}} w(\mathcal{C},\mathcal{Q}) \sum_{s=1}^{|\mathcal{Q}|} \frac{e^{i\nu_n(\tilde{\tau}'_s-\tilde{\tau}_s)}}{\beta F(\tilde{\tau}_s-\tilde{\tau}'_s)}$$

$$\mathbf{F = 0}: \quad \chi(i\nu_n) = -Z^{-1} \sum_{\mathcal{C},\tilde{\tau}\tilde{\tau}'} \mathrm{Tr}_{\mathcal{C},Q^\dagger Q} \times \mathrm{Det}_{\mathcal{C}} \times \frac{e^{i\nu_n(\tilde{\tau}'-\tilde{\tau})}}{\beta} \quad \textbf{v.s.} \quad Z = \sum_{\mathcal{C}} \mathrm{Tr}_{\mathcal{C}} \cdot \mathrm{Det}_{\mathcal{C}}$$

Susceptibilities: Dressed expansion

$$\chi(i\nu_n) = -Z^{-1} \sum_{\mathcal{C}} \frac{\partial w(\mathcal{C})}{\partial F(i\nu_n)}$$

$$Z = \sum_{\mathcal{C}} w(\mathcal{C}) = \sum_{\mathcal{C}} \mathrm{Tr}_{\mathcal{C}} \times \mathrm{Det}_{\mathcal{C}} \times \mathrm{Exp}_{\mathcal{C}} =$$



$$\mathrm{Exp}_{\mathcal{C}} = \exp\Big(\sum_{r,s=1}^{|\mathcal{C}|} q_{i_r} q_{j_s} \big(\tilde{K}(\tau_r - \tau_s) - \tilde{K}(\tau_r - \tau'_s) - \tilde{K}(\tau'_r - \tau_s) + \tilde{K}(\tau'_r - \tau'_s) \big)$$

$$\tilde{K}(\tau) = \frac{1}{2\beta} F(i\nu_n = 0)|\tau|(|\tau| - \beta) + \sum_{n \neq 0} \frac{F(i\nu_n)}{\beta (i\nu_n)^2} e^{-i\nu_n \tau}$$

$\boldsymbol{i\nu_n \neq 0}$ **:** $\quad \dfrac{\partial w(\mathcal{C})}{\partial F(i\nu_n)} = \mathrm{Tr}_{\mathcal{C}} \times \mathrm{Det}_{\mathcal{C}} \times \dfrac{\partial \mathrm{Exp}_{\mathcal{C}}}{\partial F(i\nu_n)}$

$$\Rightarrow \quad \chi(i\nu_n) = Z^{-1} \sum_{\mathcal{C}} w(\mathcal{C}) \sum_{r,s=1}^{|\mathcal{C}|} \frac{q_{i_r} q_{j_s}}{\beta \nu_n^2} \left(e^{i\nu_n(\tau_s - \tau_r)} - e^{i\nu_n(\tau'_s - \tau_r)} - e^{i\nu_n(\tau_s - \tau'_r)} + e^{i\nu_n(\tau'_s - \tau'_r)} \right)$$

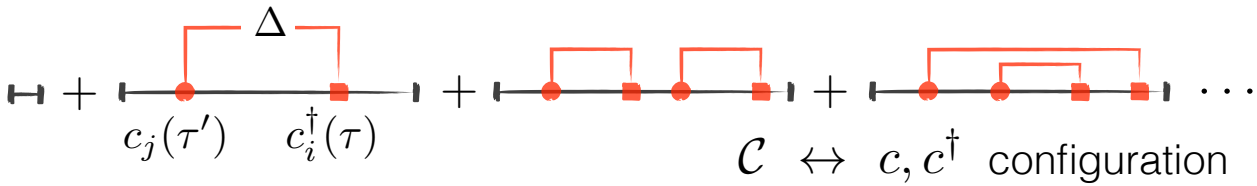
$\boldsymbol{i\nu_n = 0}$ **:** $\quad \dfrac{\partial w(\mathcal{C})}{\partial F(i\nu_n)} = \mathrm{Tr}_{\mathcal{C}} \times \mathrm{Det}_{\mathcal{C}} \times \dfrac{\partial \mathrm{Exp}_{\mathcal{C}}}{\partial F(i\nu_n)} + \dfrac{\partial \mathrm{Tr}_{\mathcal{C}}}{\partial F(i\nu_n)} \times \mathrm{Det}_{\mathcal{C}} \times \mathrm{Exp}_{\mathcal{C}}$

$\Rightarrow \quad \chi(i\nu_n) =$ do it yourself ….

$(\tilde{H}_{\mathrm{loc}} := H_{\mathrm{loc}} + Q^\dagger F(i\nu_n = 0) Q)$

Transition towards numerics …

$$Z = \sum_{\mathcal{C}} w(\mathcal{C}) = \sum_{\mathcal{C}} \mathrm{Tr}_{\mathcal{C}} \times \mathrm{Det}_{\mathcal{C}} =$$



$\mathcal{C} \leftrightarrow c, c^\dagger$ configuration

- Choose impurity **Basis** $\{|a\rangle\}$: $(\mathbf{M}_i)_{ab} := \langle a|c_i|b\rangle$ $(\mathbf{H})_{ab} := \langle a|H_{\mathrm{loc}}|b\rangle \propto \delta_{ab}$

$$\Rightarrow \quad w(\mathcal{C}) = \mathrm{Tr}[e^{-\beta\mathbf{H}}\mathrm{T}_\tau \prod_{r=1}^{|\mathcal{C}|} \mathbf{M}_{j_r}(\tau'_r)\mathbf{M}^\dagger_{i_r}(\tau_r)] \times \mathrm{Det}_{\mathcal{C}} \qquad \left(\mathbf{M}(\tau) := e^{\tau\mathbf{H}}\mathbf{M}e^{-\tau\mathbf{H}}\right)$$

- Sample $w(\mathcal{C})$ with **Markov-Chain** $\mathcal{C}_1 \to \mathcal{C}_2 \to \cdots \to \mathcal{C}_t \to \mathcal{C}_{t+1} \to \cdots$

$$\Rightarrow \quad \langle O\rangle = Z^{-1}\sum_{\mathcal{C}} o(\mathcal{C})w(\mathcal{C}) \approx N^{-1}\sum_{t=1}^{N} o(\mathcal{C}_t)$$

- $P(\mathcal{C}_{t+1}|\mathcal{C}_1\cdots\mathcal{C}_t) = P(\mathcal{C}_{t+1}|\mathcal{C}_t)$ from **Metropolis-Hasting**

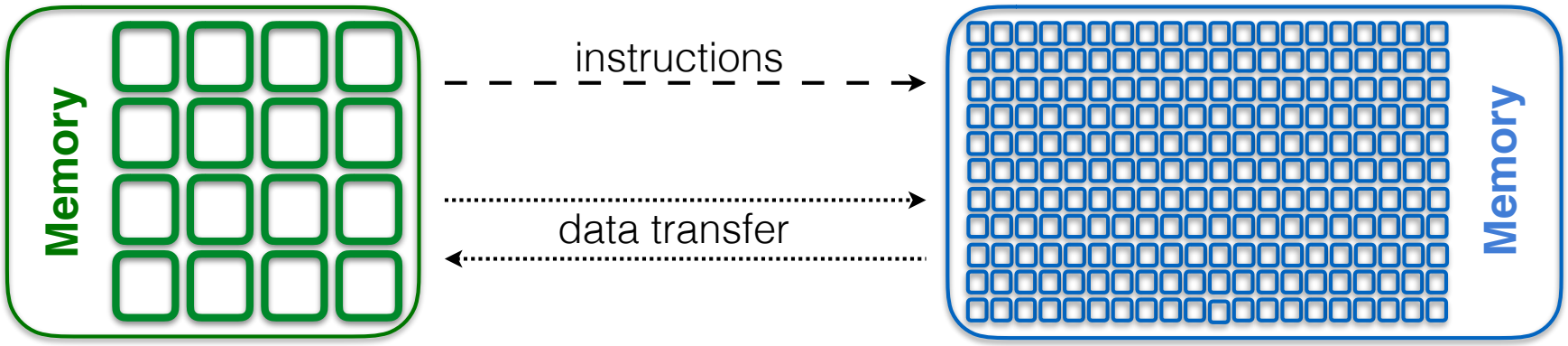
$P(\mathcal{C}_{t+1}|\mathcal{C}_t)$

1) Propose $\mathcal{C}'$ (e.g. $\mathcal{C}' := \mathcal{C}_t \pm c, c^\dagger$)

2) Set $\mathcal{C}_{t+1} := \mathcal{C}'$ with prob. $\dfrac{q(\mathcal{C}' \to \mathcal{C}_t)w(\mathcal{C}')}{q(\mathcal{C}_t \to \mathcal{C}')w(\mathcal{C}_t)}$, otherwise $\mathcal{C}_{t+1} := \mathcal{C}_t$

CPU

GPU



- **Few** but **powerful** compute cores
- Optimised for **sequential** processing

  ➡ minimise time per operation

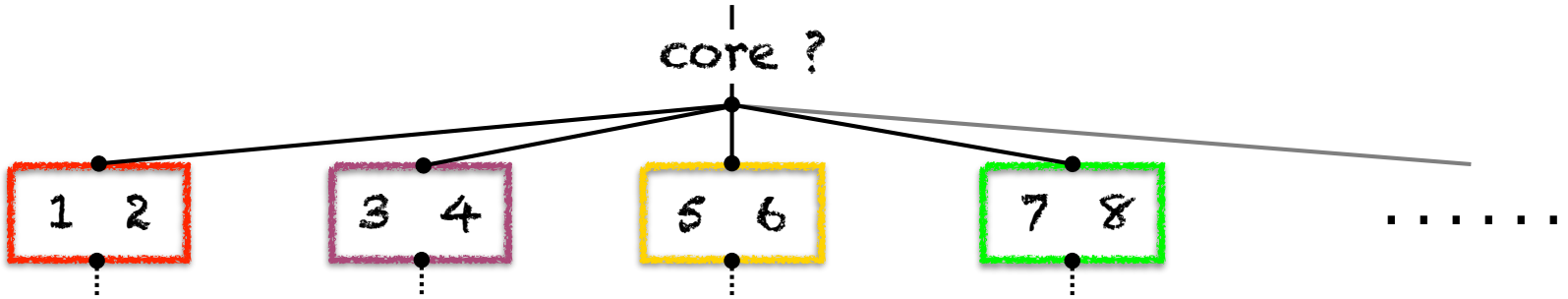
- **Master**

- **Many** but **slow** compute cores
- Optimised for **parallel** processing

  ➡ maximise operations per time

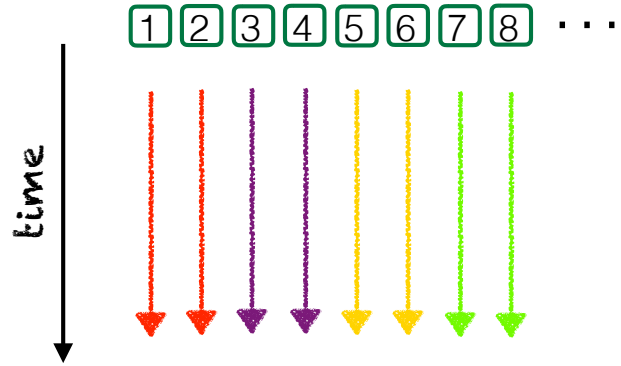
- **Slave**

What means „parallel processing“ ?



CPU

- Cores can work **independently**

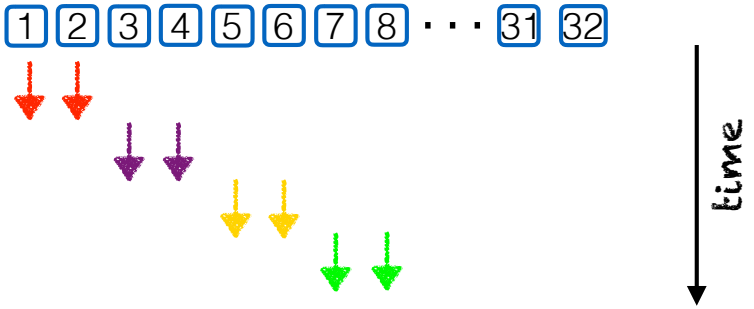


- **one** QMC-simulation per **CPU** core **?**

➡ Yes, good idea !!

GPU

- Cores are **tied** by groups of 32 (Warps)**:** execute same instruction or wait



- **one** QMC-simulation per **GPU** core **?**

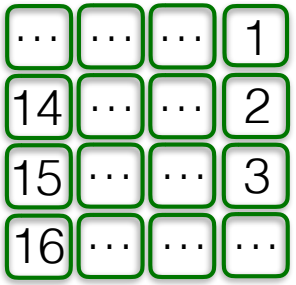
➡ No, bad idea !!

Conclusion: **algorithm** (**CPU**) → divide work → **sequential part** (**CPU**) + **parallel part** (**GPU**)

Profile CPU code

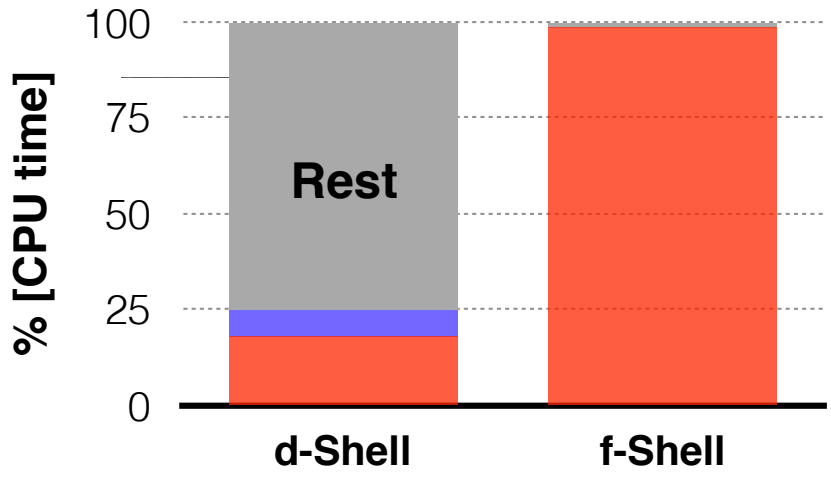
Implementation

- **one** Monte-Carlo simulation per **CPU core** (using MPI)
- several algorithmic optimisations

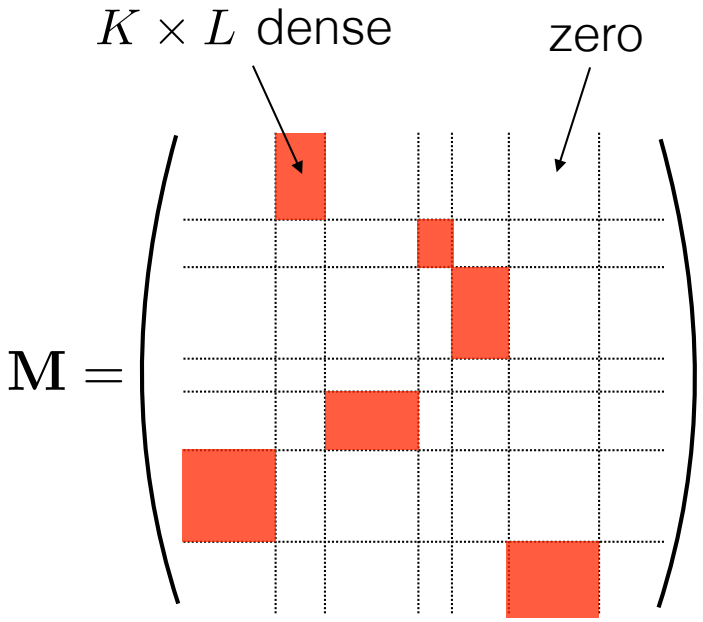


Profiling

$$w(\mathcal{C}) = \mathrm{Tr}[e^{-\beta \mathbf{H}} T_\tau \prod_{\mathcal{C}} \mathbf{M}\mathbf{M}^\dagger] \times \mathrm{Det}_{\mathcal{C}}$$

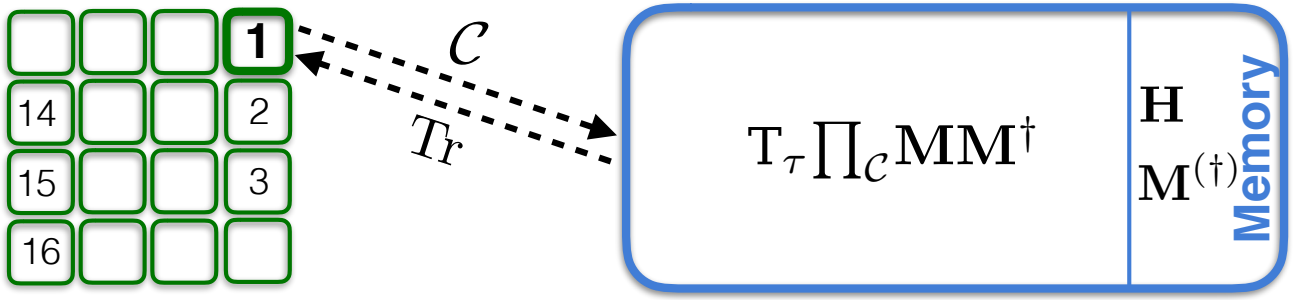


$K, L \leq 30$ $K, L \leq 300$



Matrix-Multiplication

$$w(\mathcal{C}) = \mathrm{Tr}[e^{-\beta \mathbf{H}} T_\tau \prod_{\mathcal{C}} \mathbf{M}\mathbf{M}^\dagger] \times \mathrm{Det}_{\mathcal{C}}$$



- **one** Monte-Carlo simulation (i.e. **CPU core**) uses **GPU** for matrix-multiplication

  ➡ speed-up for this Monte-Carlo simulation: $x \approx 6$ 😊

- **other** Monte-Carlo simulations (i.e. **CPU cores**) work as usual

  ➡ overall speed-up := $\dfrac{\text{\#MC samples CPU + GPU}}{\text{\#MC samples CPU}} = \dfrac{N-1+x}{N} \approx 1.3$ ☹

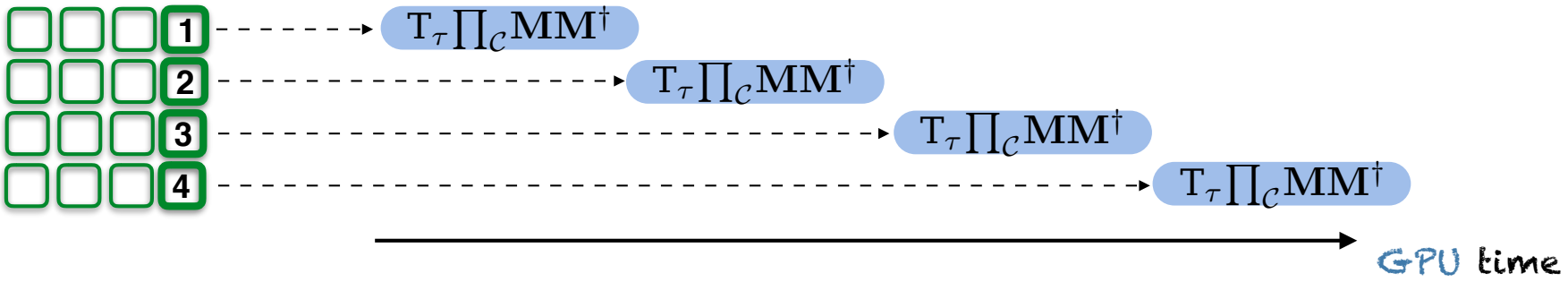
  N : #CPU cores

- Matrices to small too small to keep **GPU** busy

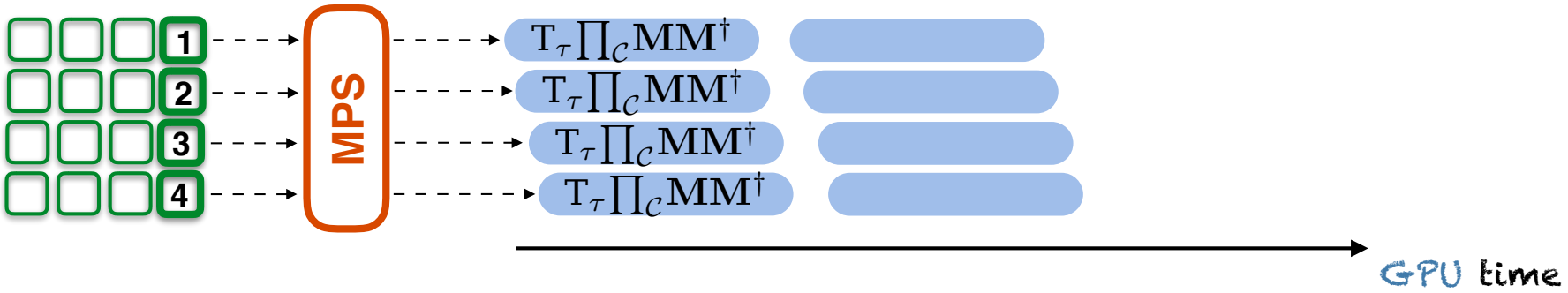
➡ need more parallelism

Multiple (MPI) processes and GPU

- By default, **GPU** access from different MPI processes is **sequential**



- **Concurrent GPU** access with **MPS** (multi-process service)



- Disadvantages of using **MPS**:

  (i) Different MPI processes can **not share** GPU memory

  ➔ Waste of **GPU**-memory (storing $\mathbf{M}$-matrices takes 0.6 GB)

  (ii) **CPU** cores using **GPU** for matrix-multiplication are essentially idle

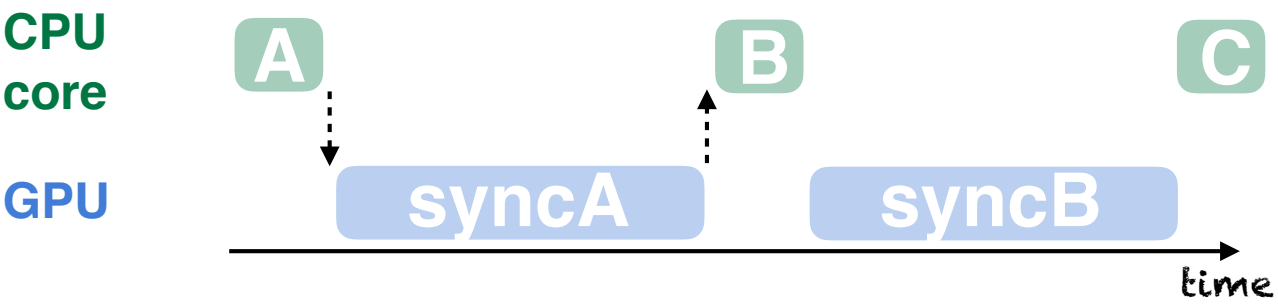
  ➔ Waste of computational resources

➔ Multiple MC-Simulations per MPI-Process (i.e. CPU core) ?

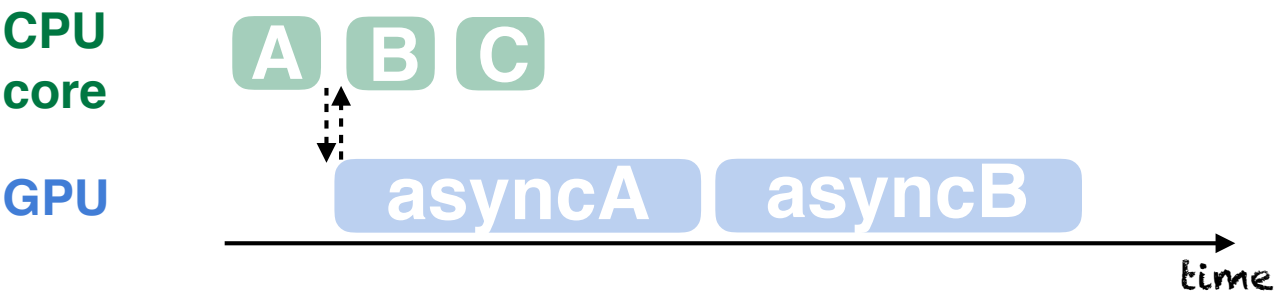
Single (MPI) process and GPU

- Synchronous (blocking) v.s. asynchronous (non-blocking) calls:

```
A();
syncA();
B();
syncB();
C();
```

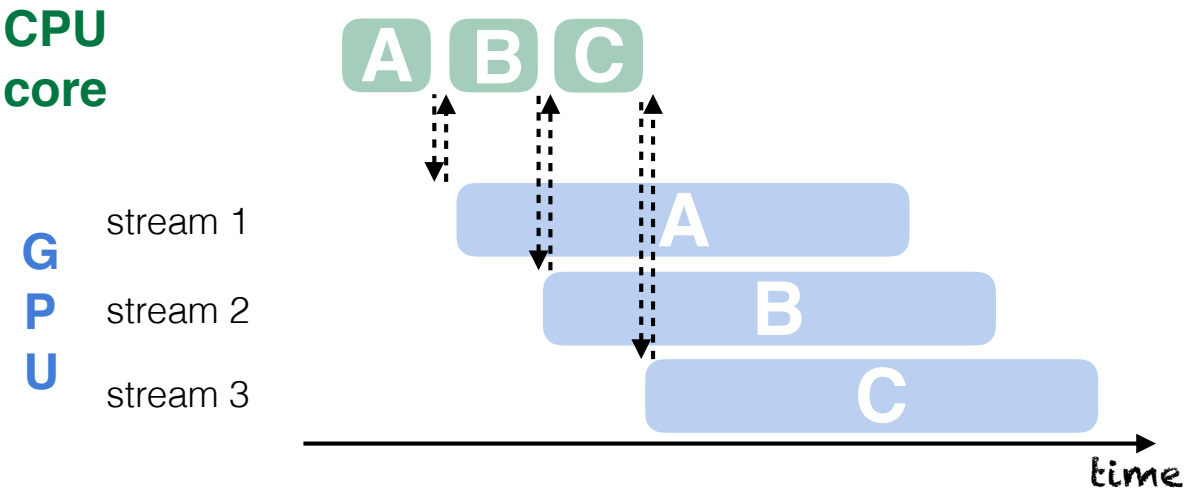



```
A();
asyncA();
B();
asyncB();
C();
```

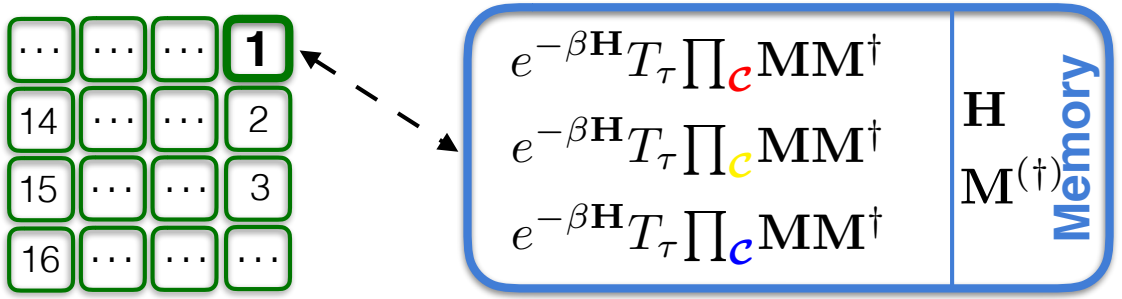


- Streams + Asynchronous calls:

```
A();
asyncA<<<…,1>>>();
B();
asyncB<<<…,2>>>();
C();
asyncC<<<…,3>>>();
```



Asynchronous CPU-GPU Algorithm



Loop over Monte-Carlo Simulations

CPU core 1

GPU

Stream 1 $\mathbf{M}_{\alpha_1}\mathbf{M}_{\alpha_2}\cdots\mathbf{M}_{\alpha_{2k}}$

Stream 2 $\mathbf{M}_{\alpha_1}\mathbf{M}_{\alpha_2}\cdots\mathbf{M}_{\alpha_{2k}}$

Stream 3 $\mathbf{M}_{\alpha_1}\mathbf{M}_{\alpha_2}\cdots\mathbf{M}_{\alpha_{2k}}$

time

Results

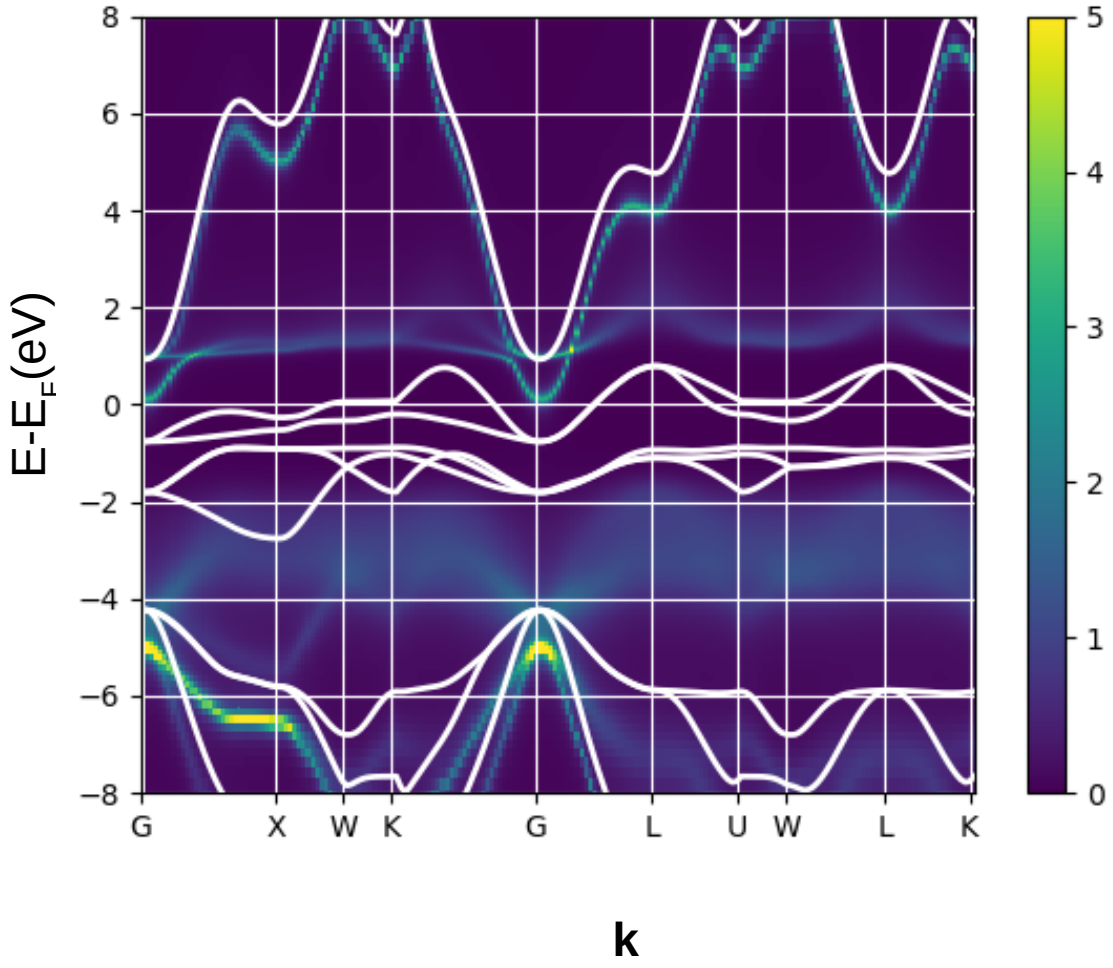
node configuration		**speed-up** (CPU+GPU over CPU)
2 NVidia K80 2 Intel Xeon Processor E5-2697 v4 (36 cores)		**3.5x**
2 NVidia P100 2 Intel Xeon Processor E5-2697 v4 (36 cores)		**5x**
1 NVidia K20 1 AMD Opteron (16 cores)	@ Titan, Oak Ridge	**5x**
4 NVidia P100 2 IBM Power8 (20 cores)	@ SummitDev, Oak Ridge	**12.5x**

4. LQSGW+DMFT code tutorial with NiO

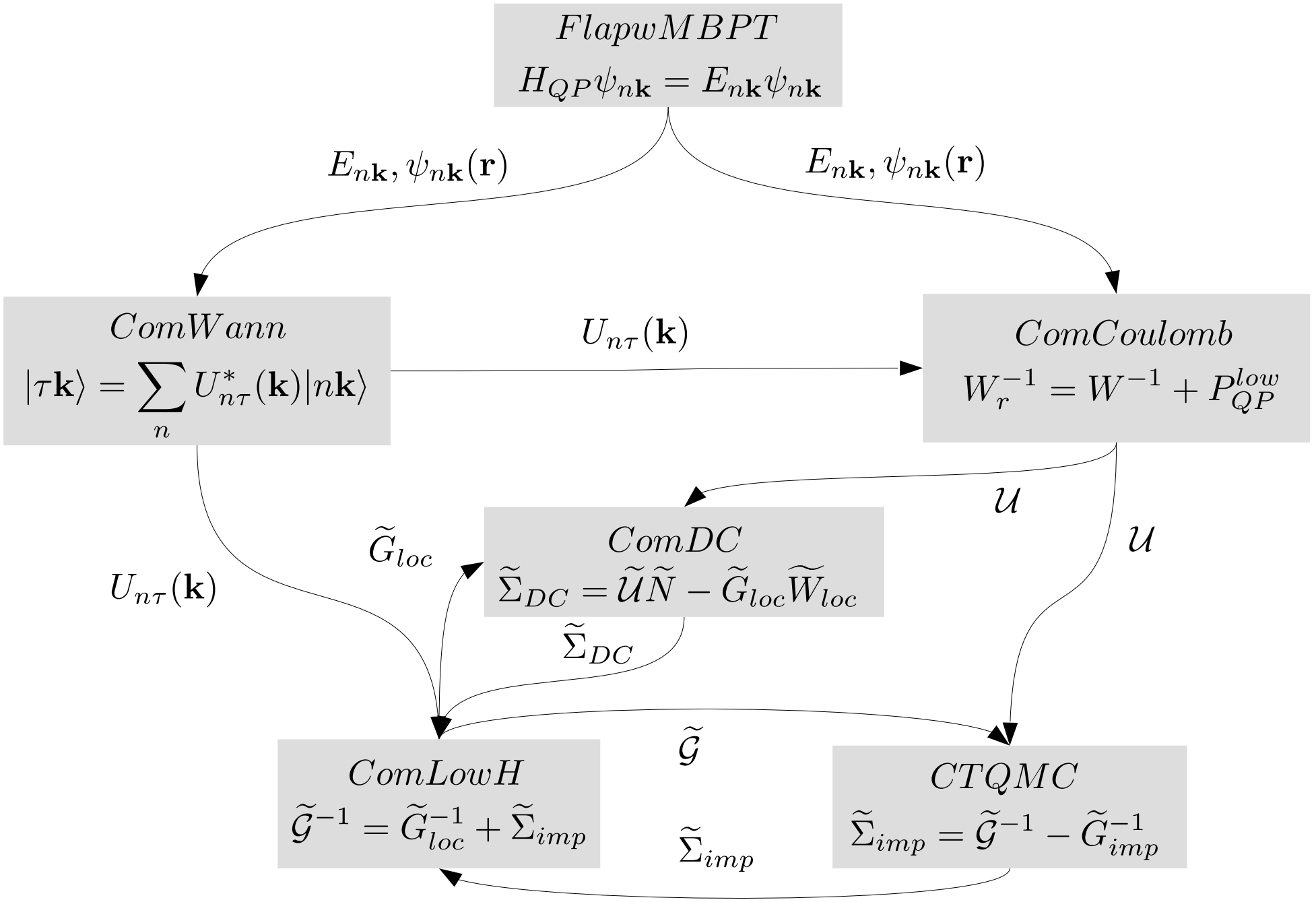
Goal

1. DMFT self-consistent solution (starting from a prerun)
2. DOS and PDOS calculation
3. Spectral function calculation

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



LQSGW+DMFT in COMSUITE



Directories

Located at /soft/public_soft/gw_dmft

```
.
..
bin
ComCoulomb
ComDC
ComLowH
ComWann
CTQMC
example
gw
wannier90_2.1
```

: where we have all the excutibles
: program to calculate bosonic Weiss field
: program for double counting self-energy
: program to calculate fermionic Weiss field, DOS, spectral functions
: program to construct Wannier functions
: CTQMC impurity solver
: examples
: FlapwMBPT code by Andrey Kutepov
: Wannier90 package

Let's load the gw_dmft module

- **Make sure that you logged in to the cluster with "ssh -X" (for the visualization)**
- **load gw_dmft modules**

  $ module load gw_dmft/2018

- **All loaded modules**

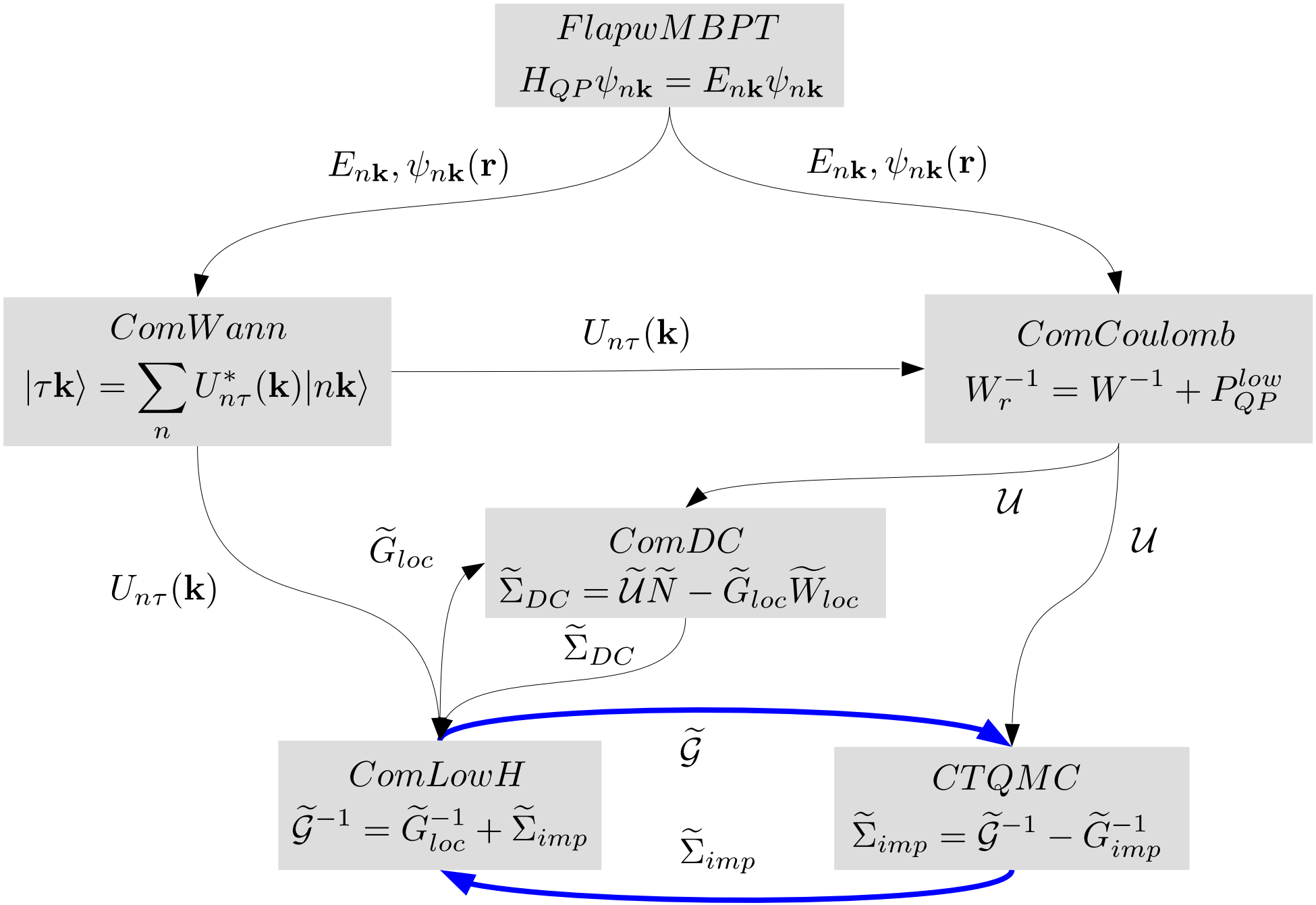
  $ module list

Currently Loaded Modules:
1) gw_dmft/2018
2) slurm/17.11
3) intel/18.2
4) conda/2

- **If there is any other modules are loaded, please unload them to avoid conflicts.**

  $ module unload ??? (if necessary)

Let's resume our calculations from almost converged results



NiO LQSGW prerun

- copy examples/NiO to the location you would like to run
  $ cp -r $COMSUITE_BIN/../example/NiO .

- move to the lqsgw directory
  $ cd NiO/
  $ cd lqsgw

- you can see the job script (please don't run this example but use the prerun outputs)

```
#!/bin/bash
#
#SBATCH --partition=c-apc
#SBATCH --nodes=1
#SBATCH -c 6
#SBATCH --time=24:00:00
#SBATCH -J temp
#SBATCH -o temp.out
#SBATCH --error=temp.err
module load gw_dmft/2018
mpirun -np 6 $COMSUITE_BIN/rspflapw.exe
```

NiO LQSGW+DMFT run

- move to the lqsgw+dmft directory
  $ cd ../lqsgw_dmft

- see the job submission script (llscript) and run the calculation
  $ sbatch llscript

```
#!/bin/bash
#
#SBATCH –partition = c-apc
#SBATCH –nodes=1
#SBATCH –c=1
#SBATCH –time=01:00:00
#SBATCH -e temp.err
#SBATCH -o temp.out
#SBATCH -j temp
module load gw_dmft/2018
$COMSUITE_BIN/comdmft.py
```

comdmft.ini

```
control={'initial_lattice_dir'         : '../lqsgw',
          'method'                 : 'lqsgw+dmft',
          'spin_orbit'             : False,
          'mpi_prefix': "srun -n 1",
          'impurity_problem':[[2, 'd']],
          'impurity_problem_equivalence':[1],
          'restart': True
          'sigma_mix_ratio': 0.05
         }
wan_hmat={'kgrid': [10, 10, 10],
              'froz_win_min': -10.0,
              'froz_win_max': 10.0,
            }
imp={'temperature'           : 300,
        '1':{
       'impurity_matrix': [[1,0,0,0,0],
                           [0,1,0,0,0],
                           [0,0,2,0,0],
                           [0,0,0,1,0],
                           [0,0,0,0,2]],
       'thermalization_time': 1,
       'measurement_time': 15,
       'green_cutoff':  50,
       'coulomb': 'full',
        }}
```

- LQSGW+DMFT input file in python dirctionary format
- all dictionary keys in small letters
- composed of three python dictionaries of “control”, “wan_hmat” and “imp”

In control

- **'methods': 'lqsgw+dmft'**
- **'initial_lattice_dir': '../lqsgw"**
  - the path to LQSGW output directory
- **'impurity_problem': [[2,'d']]**
  - a python list to specify correlated orbitals. The first and second indices are for the atom index and shell type.
  - atom index: in the order listed in the "../lqsgw/coord.xsf"

O
Ni

```
PRIMCOORD
   2      1
   8      0.000000        0.000000        0.000000
  28      2.097129        2.097129        2.097129
```

  - shell index: either "d" or "f"
- **'impurity_problem_equivalence': [1]**
  - equivalence of each impurity problem.
  - identified by an integer starting from 1. If this value is the same, they are equivalent.
  - If this value is negative, it is the time-reversal symmetry pair to the one with the same absolute value**.**
- **'spin_orbit': True or False**
  - if False, correlated orbitals correspond to cubic spherical harmonics

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

where Y_l^m is a spherical harmonics.

In control

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

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

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

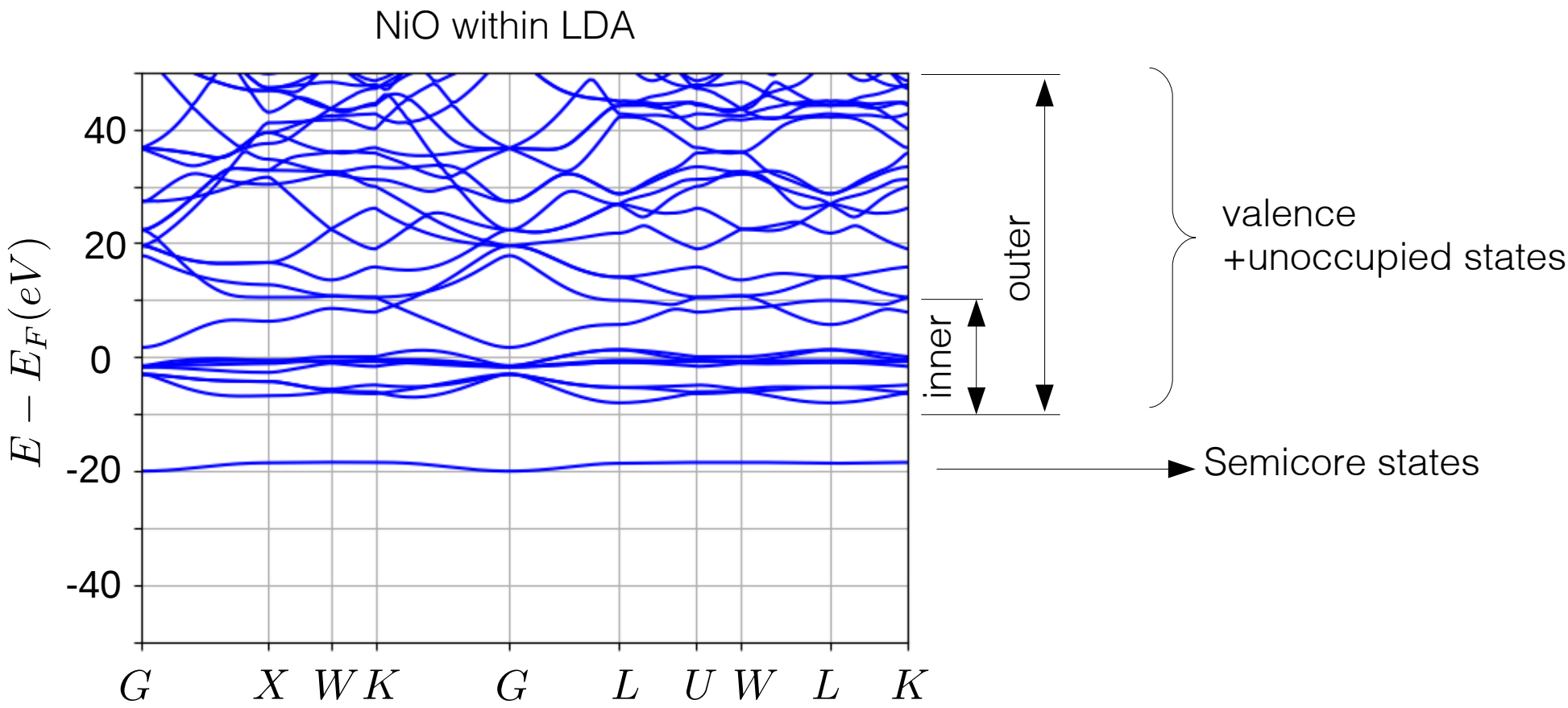
- **'mpi_prefix': : 'srun -n 1'**
  - MPI prefix commonly used for ComCoulomb, ComDC, ComLowH, ComWann, and CTQMC.
  - If a different MPI prefixs from this prefix is necessary for a program, use 'mpi_prefix_coulomb', 'mpi_prefix_lowh', 'mpi_prefix_dc', 'mpi_prefix_wannier', and 'mpi_prefix_impurity',
- **'restart': : True**
  - True or False. If True, It will resume the calculation from the prerun.
  - default value: False
- **'mpi_prefix_lowh':**
  - MPI prefix for ComLowH
  - default value: control['mpi_prefix']
- **'mpi_prefix_impurity':**
  - MPI prefix for the impurity solver
  - default value: control['mpi_prefix']
- **'mpi_prefix_wannier'**
  - MPI prefix for ComWann
  - default value: control['mpi_prefix']

In control

- **'mpi_prefix_coulomb':**
  - MPI prefix for ComCoulomb
  - default value: control['mpi_prefix']
- **'mpi_prefix_dc':**
  - MPI prefix for ComDC
  - default value: control['mpi_prefix']
- **'sigma_mix_ratio'**
  - Self-energy linear mixing ratio.
  - default value: 0.1
- **'max_iter_num_impurity':**
  - maximum iteration for the DMFT self-consistent loop.
  - default value: 50
- **'proj_win_min':**
  - low-energy cutoff to renormalize the projectors
  - default value: wan_hmat[‘dis_win_min’]
- **'proj_win_max':**
  - high-energy cutoff to renormalize the projectors
  - default value: wan_hmat[‘dis_win_max’]

Important concepts for wan_hmat

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



Important concept for wan_hmat

- The choice of initial trial orbitals $|\tau\mathbf{R}=0\rangle_t$
  - MT orbitals with desired angular momentum character
  - Radial function of $|\tau\mathbf{R}=0\rangle_t$ is chosen in such a way which maximize

$$\frac{1}{N_{\mathbf{k}}} \sum_{n\mathbf{k}}^{E_{min}^{inner}<E_{n\mathbf{k}}<E_{min}^{inner}} |\langle n\mathbf{k}|\tau\mathbf{k}\rangle_t|^2$$

where $|\tau\mathbf{k}\rangle_t = \frac{1}{N_{\mathbf{k}}} \sum_{\mathbf{R}} |\tau\mathbf{R}\rangle_t e^{i\mathbf{k}\cdot\mathbf{R}}$

  - Among MT orbitals we choose ones

$$\frac{1}{N_{\mathbf{k}}} \sum_{n\mathbf{k}}^{E_{min}^{inner}<E_{n\mathbf{k}}<E_{min}^{inner}} |\langle n\mathbf{k}|\tau\mathbf{k}\rangle_t|^2 > 0.2$$

  - For correlated orbitals, final wannier function $|\tau\mathbf{R}=0\rangle_f$ usually

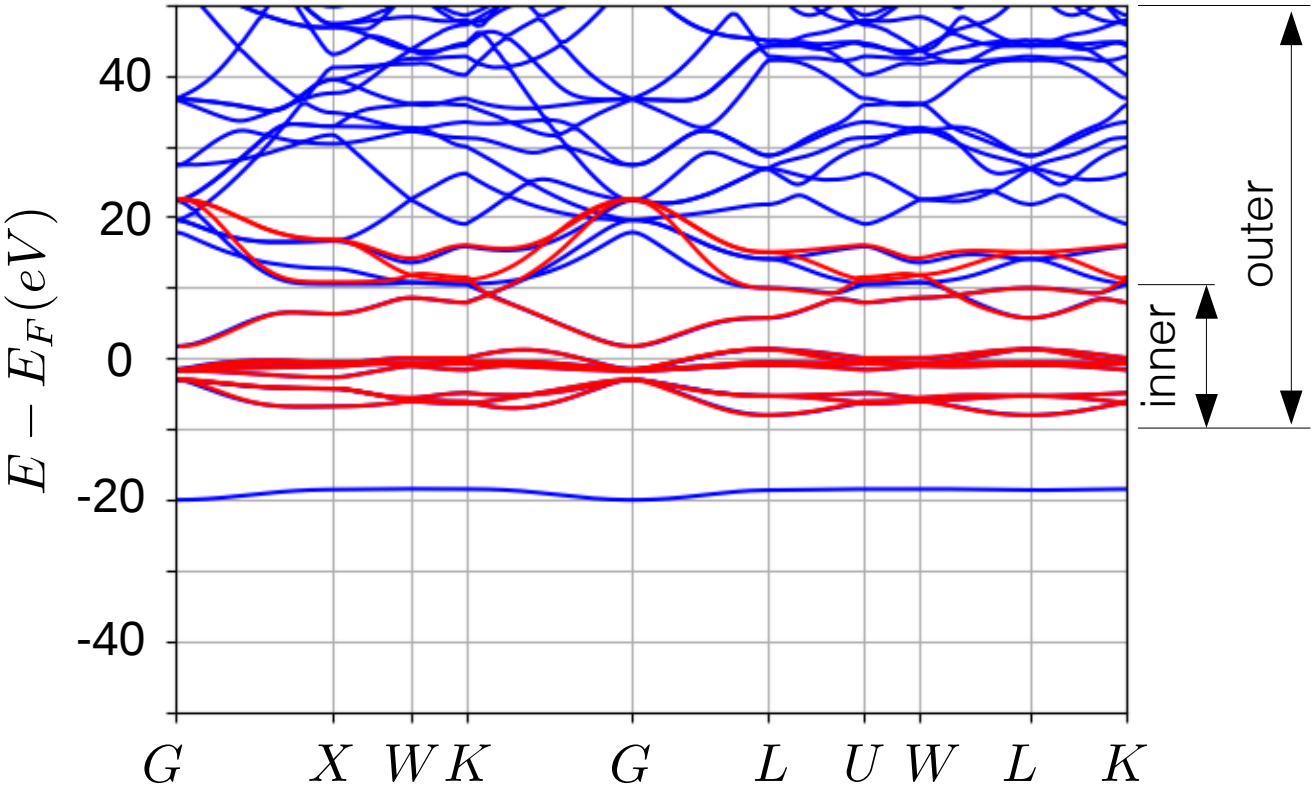
$${}_f\langle\tau\mathbf{R}=0|\tau\mathbf{R}=0\rangle_t > 0.95$$

which means $|\tau\mathbf{R}=0\rangle_f$ is strongly localized and a atom-like wavefunction

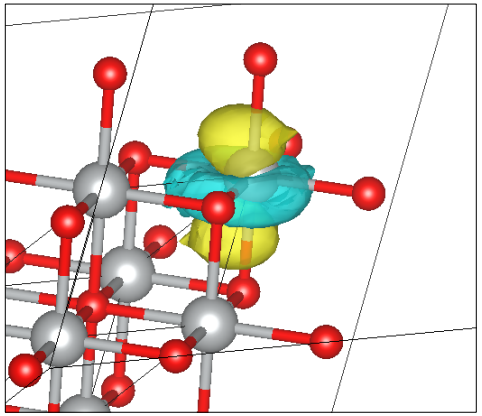
Wannier functions and interpolated bandstructure of NiO

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

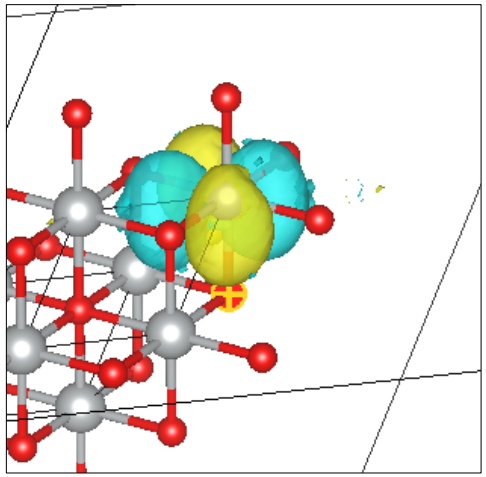
NiO within LDA



$Ni - d_{z^2}$



$Ni - d_{xy}$



In wan_hmat

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

In imp

- **'temperature': 300**
  - simulation temperature in K
- **for each distinct impurity problem indexed by the value in control[“impurity_problem_equivalence”]**
  - **'impurity_matrix': [**
    [1,0,0,0,0],
    [0,1,0,0,0], **1: t2g**
    [0,0,2,0,0], **2:eg**
    [0,0,0,1,0],
    [0,0,0,0,2]
    ],

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

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

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

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

--If control[‘spin_orbit’]==False, “m” is sorted in ascending order.
To illustrate for “d” orbitals, in this order: |xy>,|yz>,|z^2>,|xz>, |x^2-y^2>

--if control[‘spin_orbit’]==True, the most rapidly changing index is “m” and the next one is “i”. They are sorted in ascending order,
To illustrate for “f” orbitals, in this order: |3,-0.5, -2.5>,|3,-0.5, -1.5>,|3,-0.5, -0.5>, |3,-0.5, 0.5>,|3,-0.5, 1.5>,|3,-0.5, 2.5>, |3,0.5, -3.5>,|3,0.5, -2.5>,|3,0.5, -1.5>, |3,0.5, -0.5>,|3,0.5, 0.5>,|3,0.5, 1.5>,|3,0.5, 2.5>, |3,0.5, 3.5>,

In imp

- 'Coulomb': ‘full’,

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

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

--If 'full', no additional approximation is considered.

--If 'ising', only U_{abba} or U_{abab} are non-zero.

- 'thermalization_time': 1,

» wall time for the thermalization in minutes

- 'measurement_time': 5,

» wall time for the measurement in minutes

- 'green_cutoff': 70,

» cutoff-energy in eV to sample green’s function and self-energy.

» values beyond this energy will be provided by analytical equations.

- 'susceptibility_cutoff':

» cutoff-energy to sample susceptibility.

» Default value: 300 eV

Output directory

- in lqsgw_dmft directory

```
cmd.log
comdmft.ini
convergence.log      → convergence log file
coulomb              → Output of ComCoulomb
dc                   → Output of ComDC
delta.dat            → hybridization function
impurity             → Output of CTQMC solver
llscript
lowh                 → Output of ComLowH
sig.dat              → impurity self-energy
sig_dc.dat           → double-counting self-energy
sig_dc_hf.dat        → the high-frequency limit of double-counting self-energy
temp.err
temp.out
u_slater.dat         → bosonic Weiss-field
v_slater.dat         → V_loc
wannier              → Output of ComWann
w_slater.dat         → W_loc
```

convergence.log

step	i_imp	causality	static_f0	w_sp_min	w_sp_max	mu	std_sig	n_imp	histo_1	histo_2
wannier				0.29931962	1.69293343					
coulomb_1			6.54262544803							
delta	0	good				0.029191338307				
impurity_1	0	good					3.33784542527	8.19467	107.860951974	119.601660157
delta	1	good				0.164895902779				
impurity_1	1	good					1.70132143417	8.17458	98.2045831536	97.6973570752
delta	2	good				0.267689886163				
impurity_1	2	good					0.886010804306	8.1662	95.0848573468	90.6707568246
delta	3	good				0.34353949486				
impurity_1	3	good					0.479993764726	8.16272	94.1990718702	89.0756294454
delta	4	good				0.388125192763				

- keeping track of convergence of some quantities at each iteration
- causality: causality of hybridization function / self-energy
- w_sp_min: mininum spreading of the Wannier functions
- w_sp_max: maximum spreading of the Wannier functions
- mu: LQSGW+DMFT chemical potential w.r.t. LQSGW chemical potential
- std_sig:

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

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

Dynamical U

In “lqsgw_dmft”

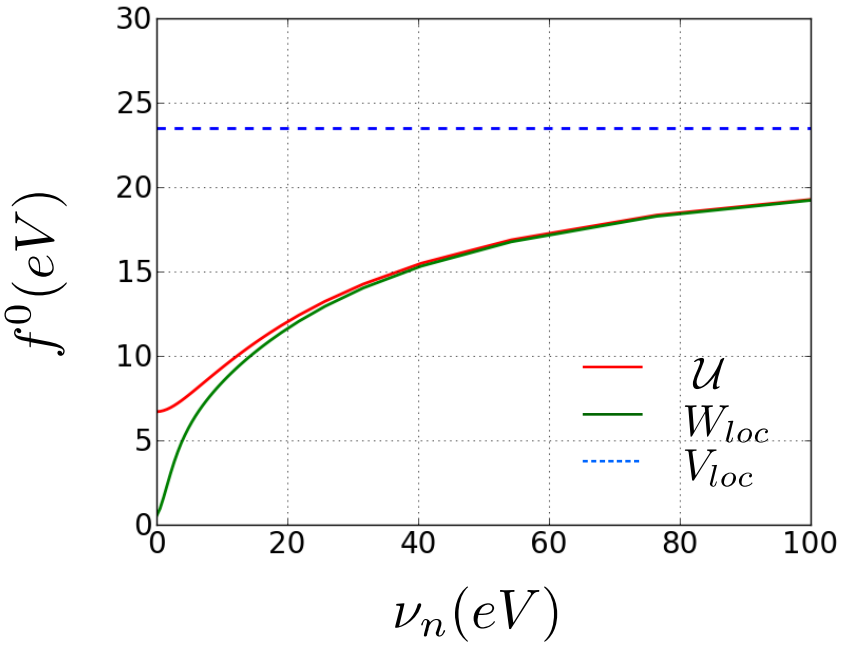
- Bosonic Weiss field in u_slater.dat and W_loc in w_slater.dat

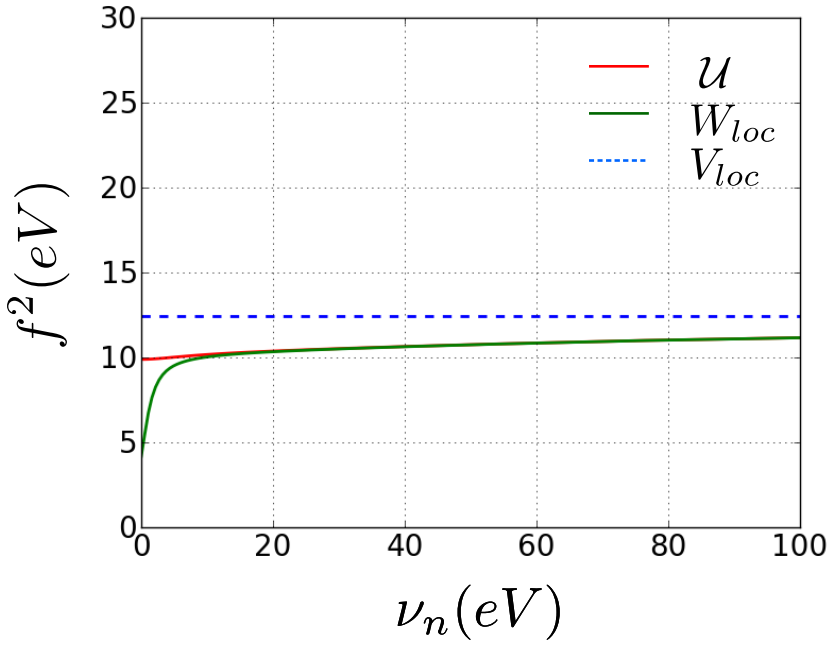
```
      # nu(eV)         1:f0(eV)         1:f2(eV)         1:f4(eV)
  0.000000000000   6.542625448029   9.782007531695  7.349584084243
  0.162432849384   6.555461634129   9.783710903959  7.350340828558
  0.324865698768   6.559451578532   9.784221895288  7.350574481502
  0.487298548153   6.560301567520   9.784309653440  7.350622464232
  0.649731397537   6.568173653669   9.785319774582  7.351044855529
  0.812164246921   6.582296296647   9.787173552298  7.351838790507
  0.974597096305   6.599477786405   9.789473579528  7.352870998503
  1.137029945690   6.619214090634   9.792160035876  7.354123553199
  1.299462795074   6.641396967279   9.795201128364  7.355555800217
  1.461895644458   6.665533537896   9.798509314544  7.357097947324
```

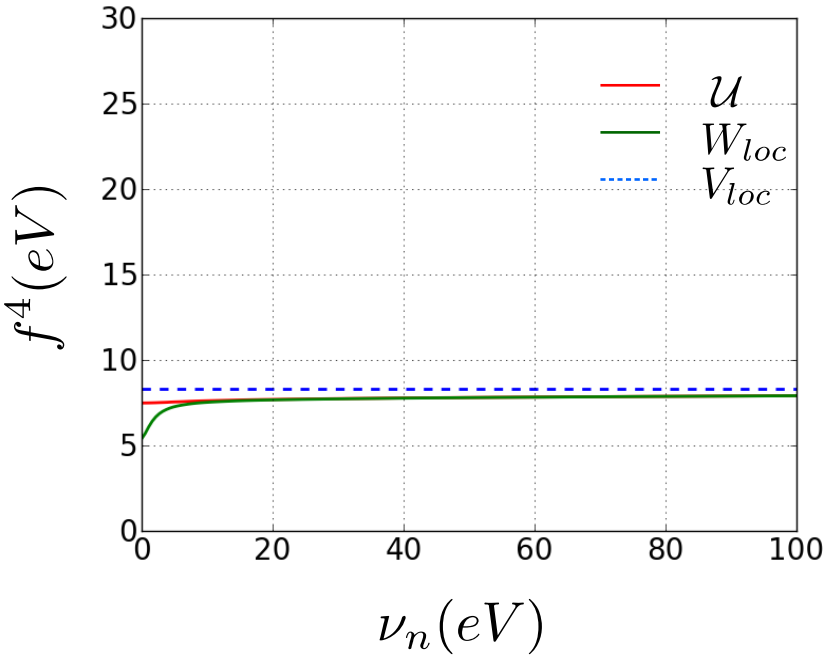
- Bare Coulomb interaction in v_slater.dat

```
      # 1:f0(eV)         1:f2(eV)         1:f4(eV)
   23.277958614497  12.294135887544  8.129215063726
```

Dynamical U







- At high frequency, U and W_{loc} converge to V_{loc}
- In comparison to F^0, F^2 and F^4 shows weaker frequency dependence

Local-GW impurity self-energy

In “lqsgw_dmft”

- “sig_dc.dat”

  » Real and imaginery part of impurity self-energy within local GW approximation are listed

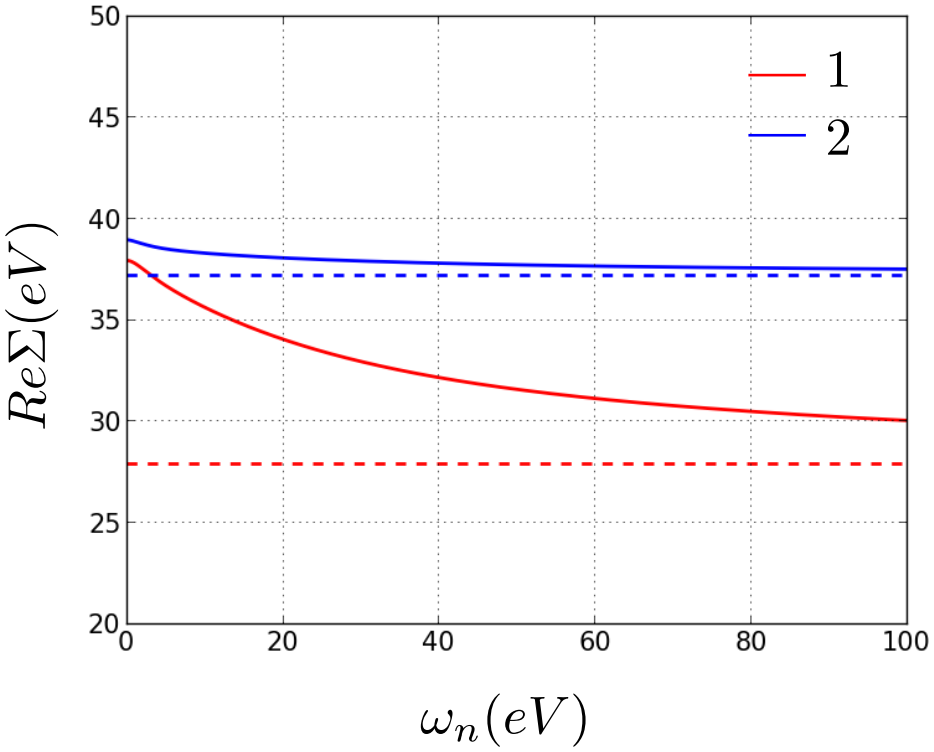
# omega(eV)	Re Sig_{1,1}(eV)	Im Sig_{1,1}(eV)	Re Sig_{1,2}(eV)	Im Sig_{1,2}(eV)
0.081216424692	37.025924683229	-0.065705926967	37.979454934019	-0.136292666459
0.243649274076	37.019196471353	-0.195617699405	37.973962536612	-0.386517235766
0.406082123461	37.006023401056	-0.322618942256	37.964307716952	-0.613800680933
0.568514972845	36.986872376987	-0.445523411213	37.951693682326	-0.819073715258
0.730947822229	36.962320661697	-0.563440899747	37.936870865340	-1.003738945640
0.893380671613	36.933004427524	-0.675791750970	37.920387929100	-1.169379323169
1.055813520997	36.899574763907	-0.782271035711	37.902688726511	-1.317674732094
1.218246370382	36.862666571515	-0.882792506656	37.884146112698	-1.450325215806
1.380679219766	36.822878695458	-0.977432822207	37.865074746593	-1.568988797605

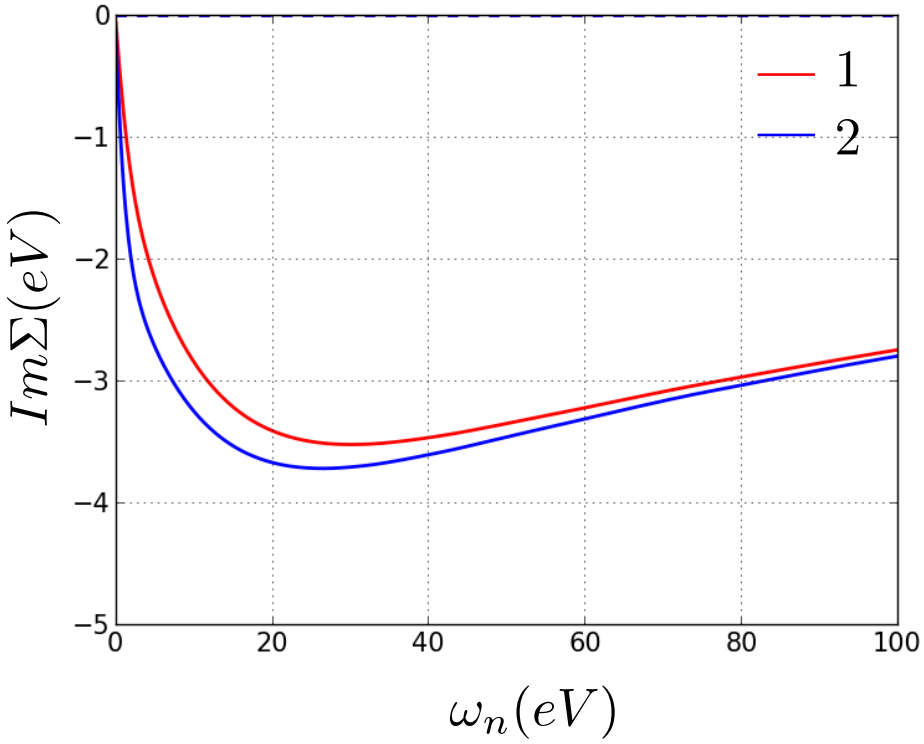
- “sig_hf_dc.dat”

  » Real and imaginery part of the Hartree-Fock contribution to the impurity self-energy within local GW approximation are listed

# Re Sig_{1,1}(eV)	Im Sig_{1,1}(eV)	Re Sig_{1,2}(eV)	Im Sig_{1,2}(eV)
27.069800000000	0.000000000000	36.110988000000	0.000000000000

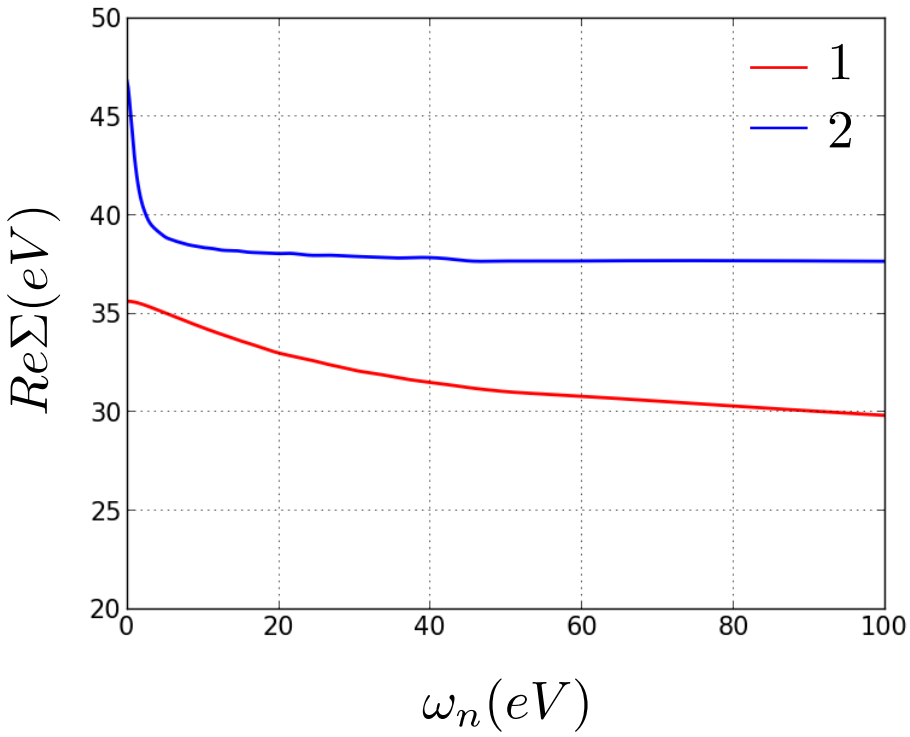
Local-GW impurity self-energy

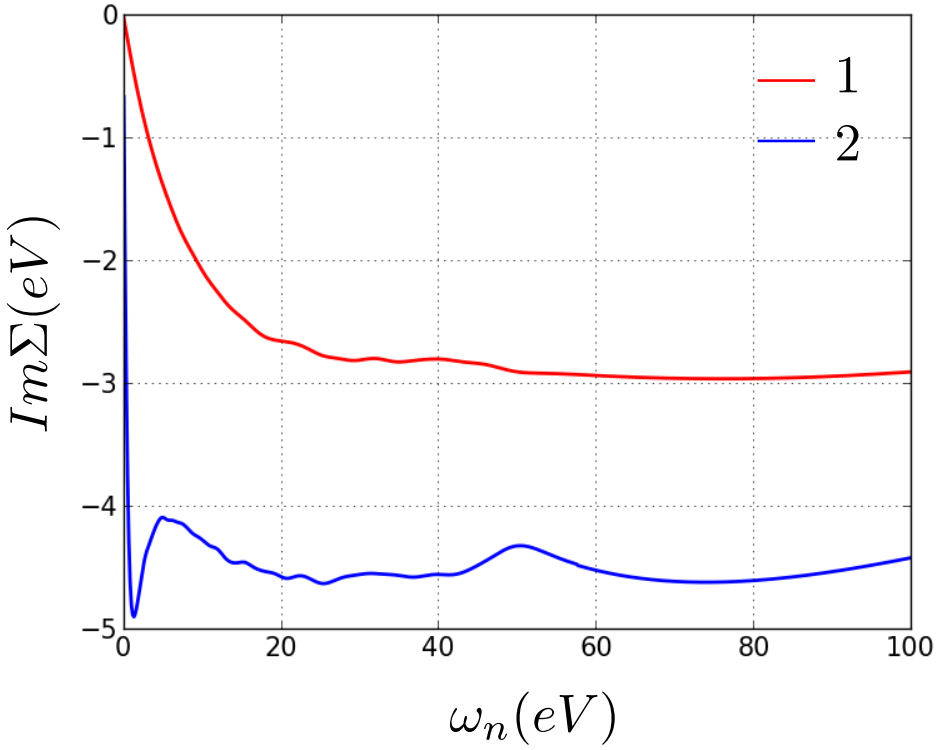




- At high frequency, self-energy converges to Hartree-fock limit
- No divergent self-energy near Fermi-level.

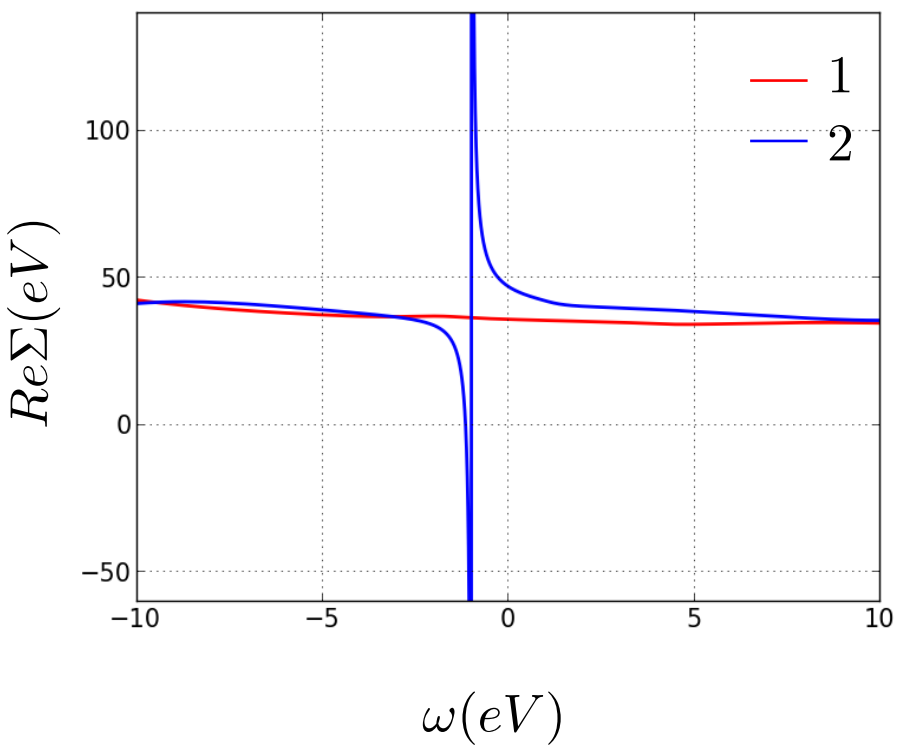
Impurity self-energy

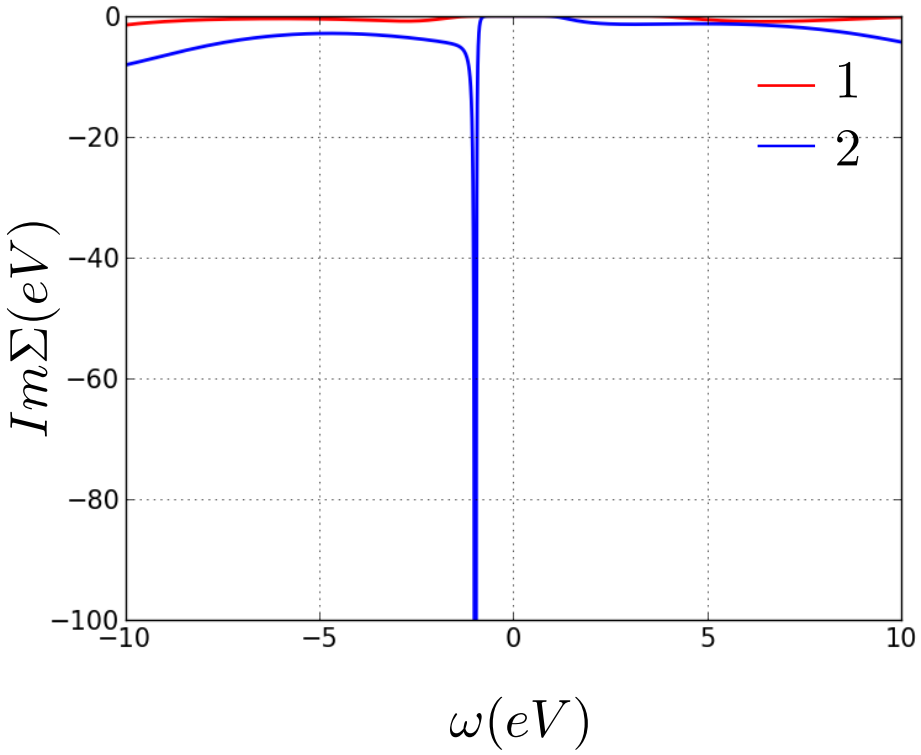




- “lqsgw_dmft/sig.dat”: in the same format as “sig_dc.out”

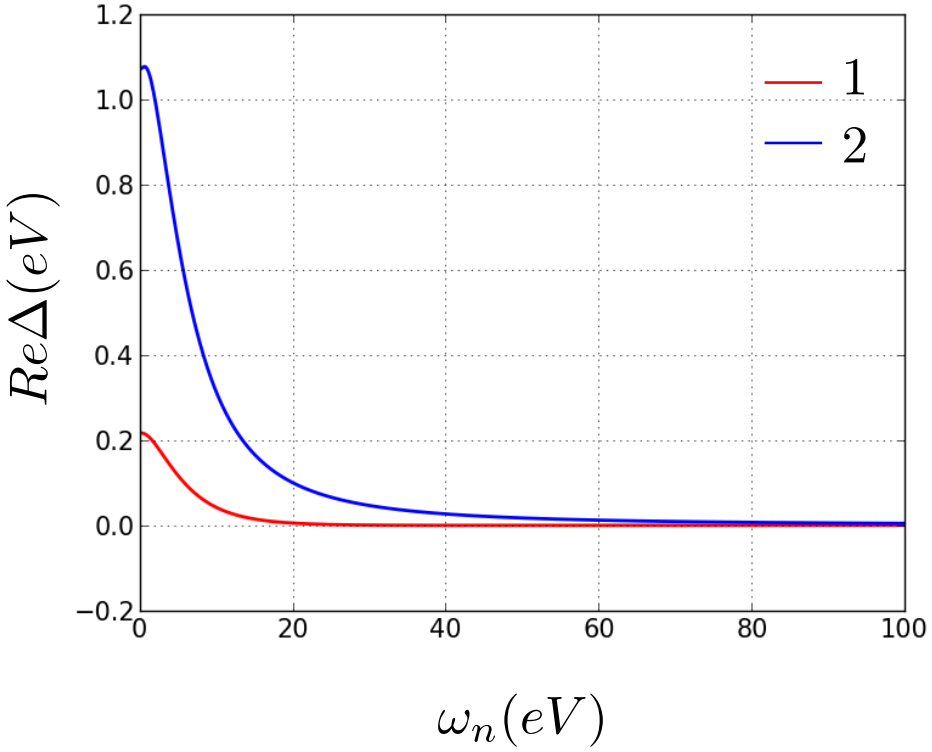
Impurity self-energy on real frequency axis

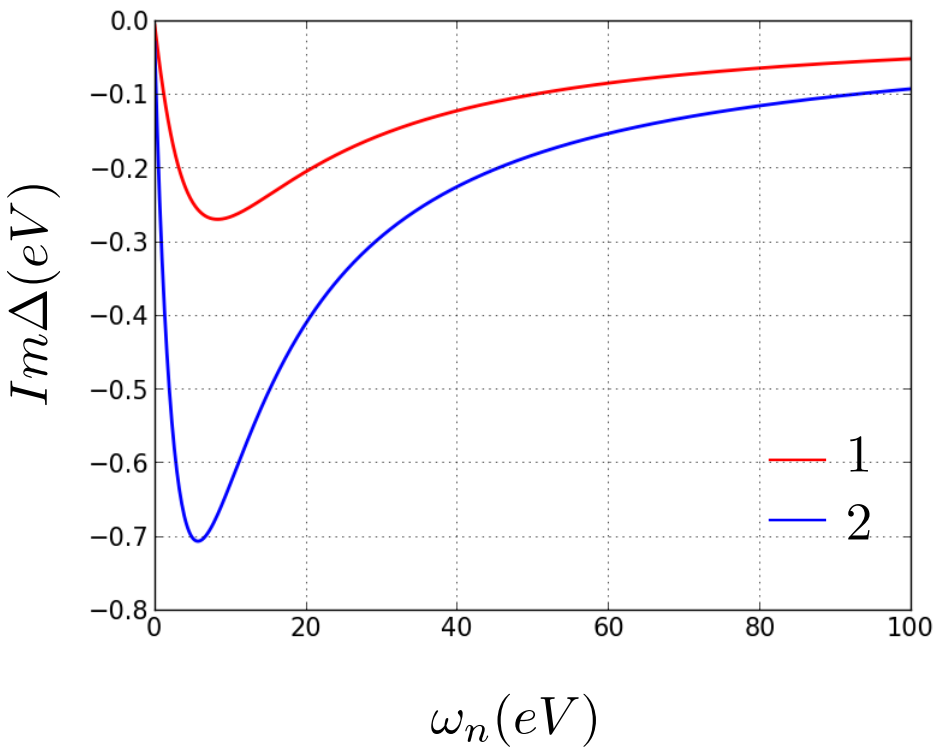




- analytical continuation by maximum entropy method (using Kristjan's code, at lqsgw_dmft/maxent)
- Divergent self-energy near Fermi level for e_g orbitals

Hybridization function





- “lqsgw_dmft/delta.dat”
- in the same format as sig.dat

LQSGW+DMFT DOS

1. move to a directory for the DOS calculation
   $ mkdir realgrid
   $ cd realgrid

2. copy files necessary to calculate DOS and partial DOS by using prepare_realaxis.py
   $ $COMSUITE_BIN/prepare_realaxis.py 0.1 ../lowh/ ../wannier/ ../maxent/sig.dat -m 2

```
[lect18@inter ~]$ /soft/public_soft/gw_dmft/bin/prepare_realaxis.py -h
usage: prepare_realaxis.py [-h] [-m MODE]
                           broadening lowh_directory wan_directory self_energy

prepare inputs of comlowh calculation on real axis

positional arguments:
  broadening            broadening
  lowh_directory        lowh directory
  wan_directory         wannier directory
  self_energy           real-axis self-energy

optional arguments:
  -h, --help            show this help message and exit
  -m MODE, --mode MODE  If 3, code calculates spectral function along the high
                        symmetry line defined in 'kpath.dat'. If it is 2, it
                        calculates projected density of states. Default: 3
```

LQSGW+DMFT DOS

3. prepare job submission script
   (you can copy it from "../realgrid_for_comparison/llscript)

```
#!/bin/bash
#
#SBATCH –partition = c-apc
#SBATCH –nodes=1
#SBATCH –time=01:00:00
#SBATCH -e temp.%j.err
#SBATCH -o temp.%j.out
#SBATCH -j temp
module load gw_dmft/2018
srun -n 1 $COMSUITE_BIN/ComLowH
```

4. submit the script
   $ sbatch llscript

LQSGW+DMFT DOS

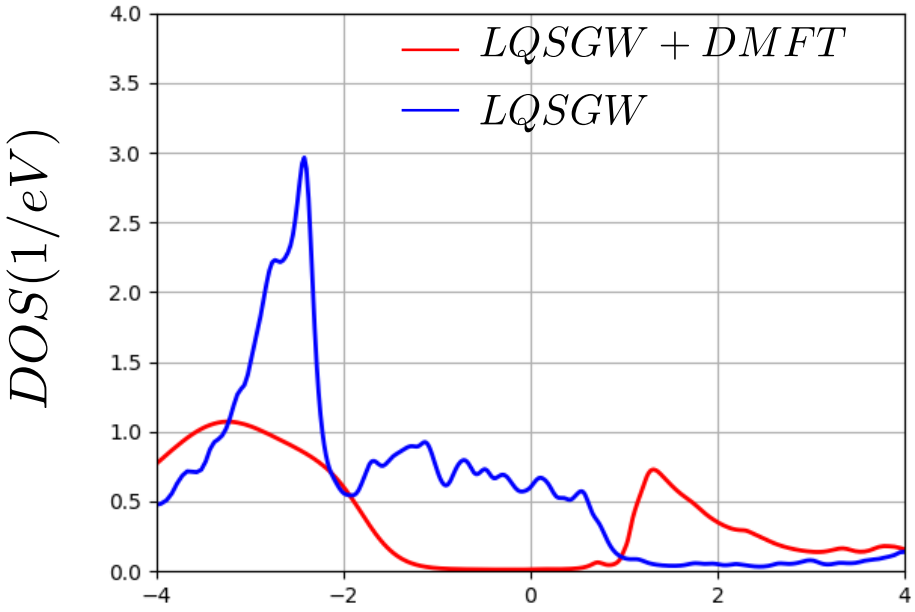
- realaxis/tdos.dat

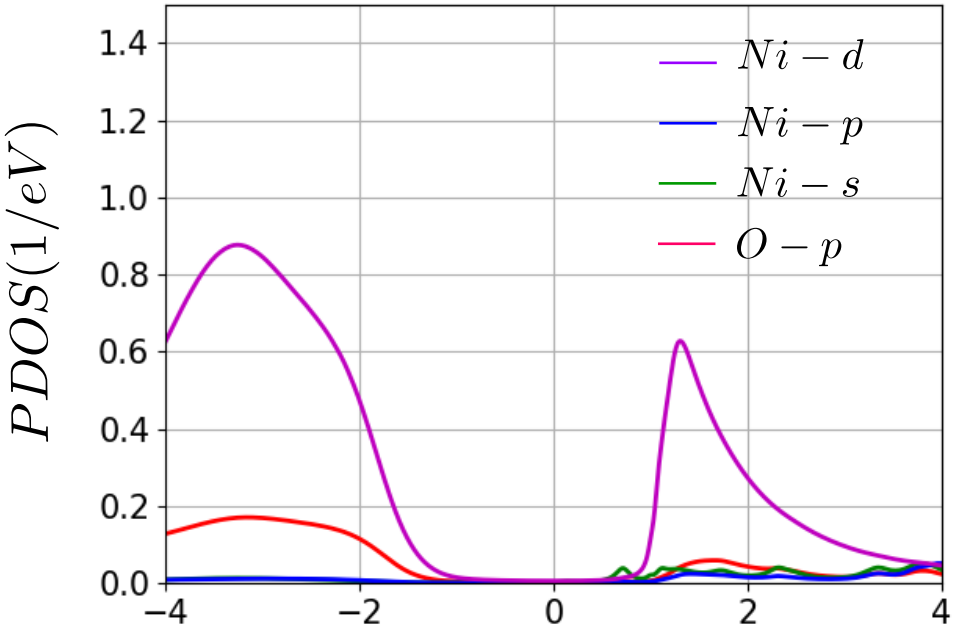
```
#       omega(eV)         DOS(1/eV)
   -50.000000000000     0.000177804560
   -42.023729163000     0.000276918308
   -36.241942949400     0.000627161151
   -31.858487825300     0.005785580105
   -28.420784332200     0.037314234345
   -25.652541304600     0.079763477565
   -23.375538144600     0.081789018970
   -21.469658255600     0.073687474286
   -19.850994524700     0.069423733434
   -18.459161225800     0.068060677542
   -17.249591724000     0.067313675285
   -16.188676770200     0.066102959393
   -15.250591955300     0.064375326169
   -14.415167104500     0.062493362835
```

- realaxis/pdos.dat
  - (atom index, l, m) if spin_orbit==False and (atom index, l, i,m) if spin_orbit==True

```
#       omega(eV)             (1,1,-1)             (1,1,0)              (1,1,1)              (2,0,0)
    -50.000000000000      0.000015753473       0.000015753473       0.000015753473       0.000011950079
   -42.023729163000       0.000023554294       0.000023554294       0.000023554294       0.000017046114
   -36.241942949400       0.000033678323       0.000033678323       0.000033678323       0.000023365976
   -31.858487825300       0.000048541987       0.000048541987       0.000048541987       0.000033820387
   -28.420784332200       0.000083715082       0.000083715082       0.000083715082       0.000068267782
   -25.652541304600       0.000147337328       0.000147337328       0.000147337328       0.000130797612
   -23.375538144600       0.000205012170       0.000205012170       0.000205012170       0.000166506290
   -21.469658255600       0.000274047245       0.000274047245       0.000274047245       0.000189699399
   -19.850994524700       0.000372694520       0.000372694520       0.000372694520       0.000217394738
   -18.459161225800       0.000503176265       0.000503176265       0.000503176265       0.000254764044
   -17.249591724000       0.000661807895       0.000661807895       0.000661807895       0.000304994797
   -16.188676770200       0.000847296191       0.000847296191       0.000847296191       0.000372302995
   -15.250591955300       0.001065018912       0.001065018912       0.001065018912       0.000463759228
   -14.415167104500       0.001328682075       0.001328682075       0.001328682075       0.000591389584
   -13.666419099000       0.001662727242       0.001662727242       0.001662727242       0.000776533107
```

LQSGW+DMFT DOS





realgrid

$ cp ../~~realaxis~~_for_comparison/pdos_plot.py

$ python ./pdos_plot.py

- energy gap opens due to local strong correlation

LQSGW+DMFT spectral function

1. move to a directory for the spectral function calculation
   $ mkdir ../realaxis
   $ cd ../realaxis

2. copy files necessary to calculate spectral functions by using prepare_realaxis.py
   $ $COMSUITE_BIN/prepare_realaxis.py 0.1 ../lowh/ ../wannier/ ../maxent/sig.dat -m 3

```
[lect18@inter ~]$ /soft/public_soft/gw_dmft/bin/prepare_realaxis.py -h
usage: prepare_realaxis.py [-h] [-m MODE]
                           broadening lowh_directory wan_directory self_energy

prepare inputs of comlowh calculation on real axis

positional arguments:
  broadening            broadening
  lowh_directory        lowh directory
  wan_directory         wannier directory
  self_energy           real-axis self-energy

optional arguments:
  -h, --help            show this help message and exit
  -m MODE, --mode MODE  If 3, code calculates spectral function along the high
                        symmetry line defined in 'kpath.dat'. If it is 2, it
                        calculates projected density of states. Default: 3
```

LQSGW+DMFT spectral function

3. create k-path file (kpath.dat)

   $ cp ../realaxis_for_comparison/kpath.dat .

   - k points w.r.t. reciprocal lattice vector

```
186   The number of k points
  0.00000000   0.00000000   0.00000000
  0.01666667   0.00000000   0.01666667
  0.03333333   0.00000000   0.03333333
  0.05000000   0.00000000   0.05000000
  0.06666667   0.00000000   0.06666667
  0.08333333   0.00000000   0.08333333
  0.10000000   0.00000000   0.10000000
  0.11666667   0.00000000   0.11666667
  0.13333333   0.00000000   0.13333333
  0.15000000   0.00000000   0.15000000
  0.16666667   0.00000000   0.16666667
  0.18333333   0.00000000   0.18333333
  0.20000000   0.00000000   0.20000000
  0.21666667   0.00000000   0.21666667
  0.23333333   0.00000000   0.23333333
  0.25000000   0.00000000   0.25000000
  0.26666667   0.00000000   0.26666667
  0.28333333   0.00000000   0.28333333
  0.30000000   0.00000000   0.30000000
  0.31666667   0.00000000   0.31666667
```

LQSGW+DMFT spectral function

3. prepare job submission script
   (you can copy it from "../realaxis_for_comparison/llscript)

```
#!/bin/bash
#
#SBATCH –partition = c-apc
#SBATCH –nodes=1
#SBATCH –time=01:00:00
#SBATCH -e temp.%j.err
#SBATCH -o temp.%j.out
#SBATCH -j temp
module load gw_dmft/2018
srun -n 1 $COMSUITE_BIN/ComLowH
```

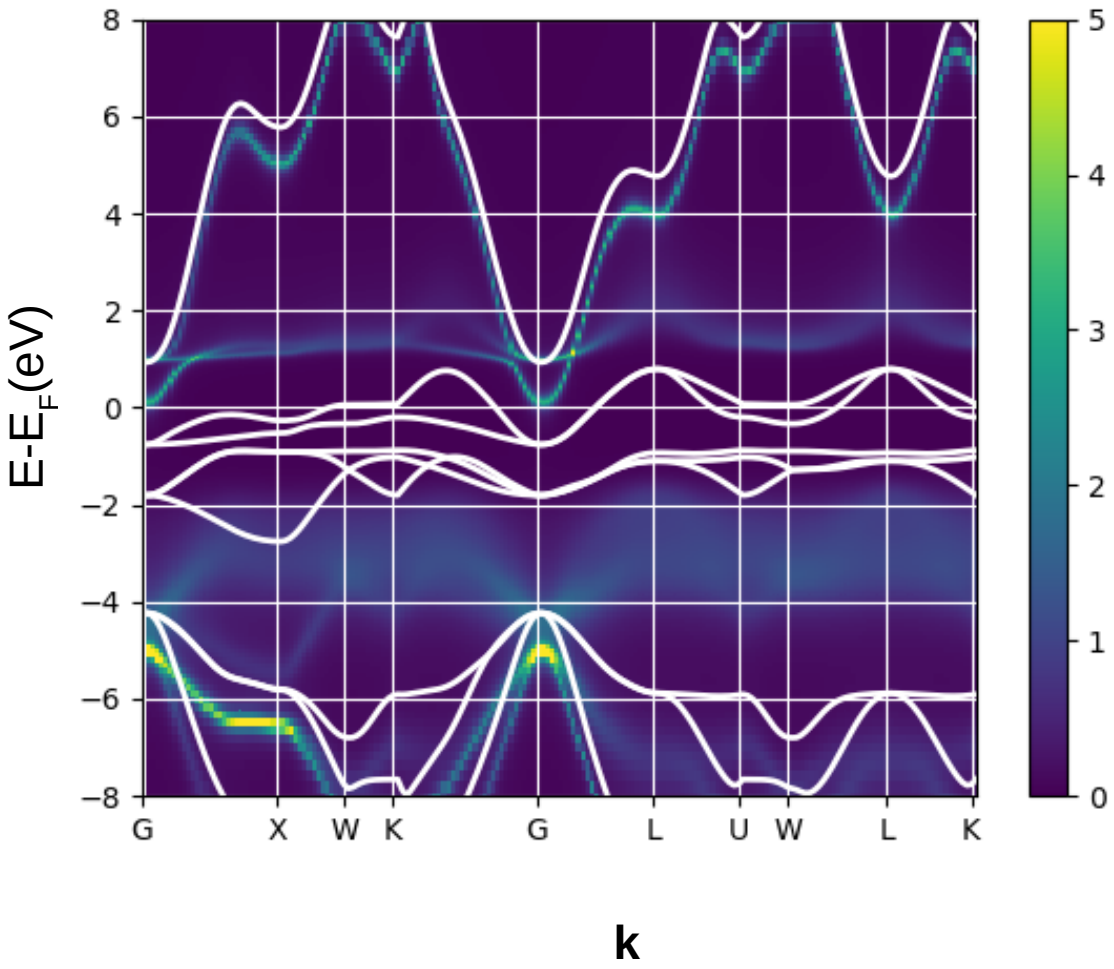
4. submit the script
   $ sbatch llscript

LQSGW+DMFT spectral function

- realaxis/spectral.dat

# kpoint	E(eV)	A(1/eV)
1	-50.000000000000	0.000170826963
2	-50.000000000000	0.000170854205
3	-50.000000000000	0.000170935102
4	-50.000000000000	0.000171067293
5	-50.000000000000	0.000171247218
6	-50.000000000000	0.000171470600
7	-50.000000000000	0.000171733002
8	-50.000000000000	0.000172030282
9	-50.000000000000	0.000172358869
10	-50.000000000000	0.000172715722
11	-50.000000000000	0.000173097979
12	-50.000000000000	0.000173502371
13	-50.000000000000	0.000173924570
14	-50.000000000000	0.000174358701
15	-50.000000000000	0.000174797197

LQSGW+DMFT spectral function



$ cp ../realaxis_for_comparison/spectral.py .
$ python ./spectral.py

- Charge transfer gap opening
- white line: lqsgw bandstructure

LQSGW+DMFT

- Official version of the code will be released under GPL3.0 in August 2018.
- If you have any question, please contact sangkookchoi@gmail.com