FLATIRON INSTITUTE

CoQuí: Electronic Structure Beyond DFT

— Why and What?

TRIQS School 2025, Paris
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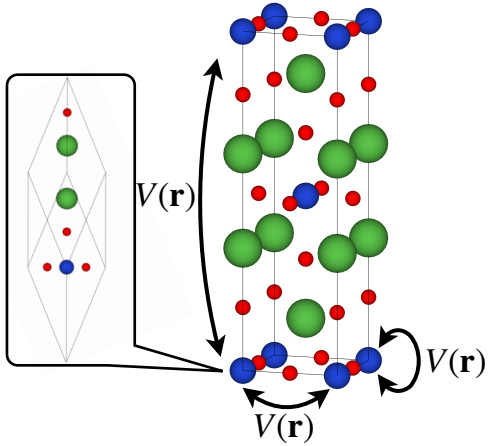
Center for Computational Quantum Physics (CCQ),
Flatiron Institute

- Github repository:
  https://github.com/AbInitioQHub/coqui
- Tutorials:
  https://github.com/AbInitioQHub/coqui-tutorial

Many-Electron Problem at Different Scales

Quantum materials



- All electrons
- Bare interaction ~ $O(10)$ eV

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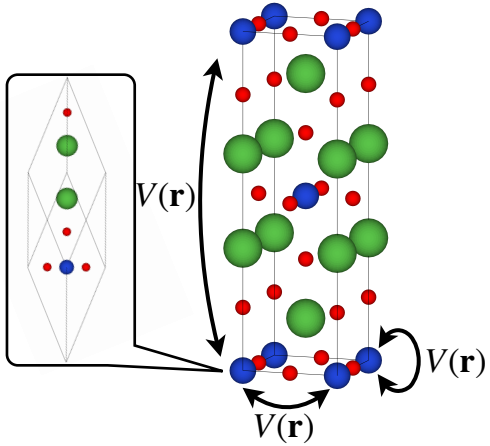

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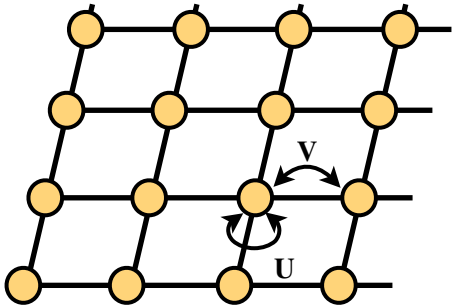
Effective lattice model (e.g. Hubbard model)



- Low-energy orbitals (near Fermi level)
- Short-range screened interactions ~ $O(1)$ eV
- Methods: QMC, DMRG, DMFT, etc.

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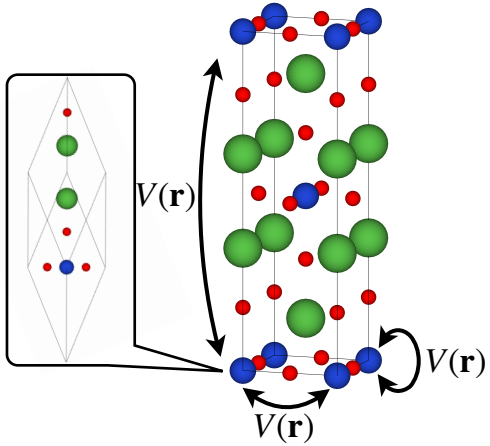

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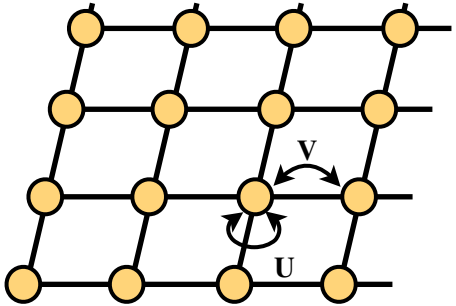

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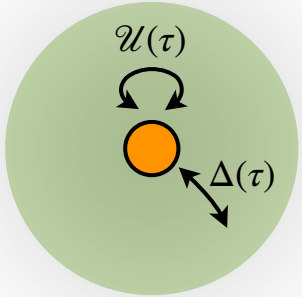

- Single site + baths
- Local interactions
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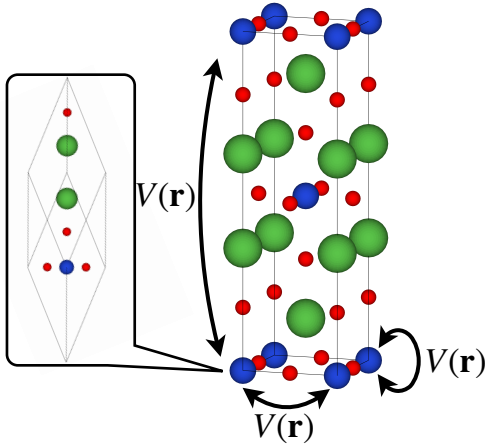

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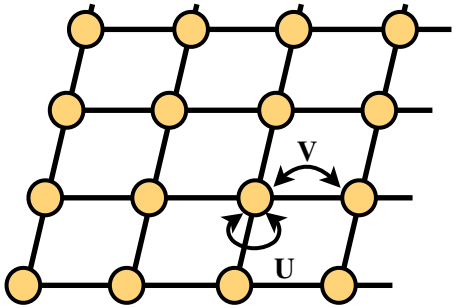

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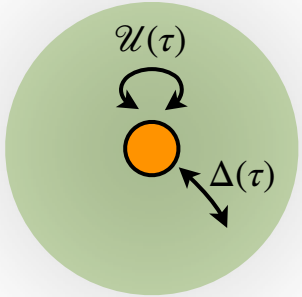

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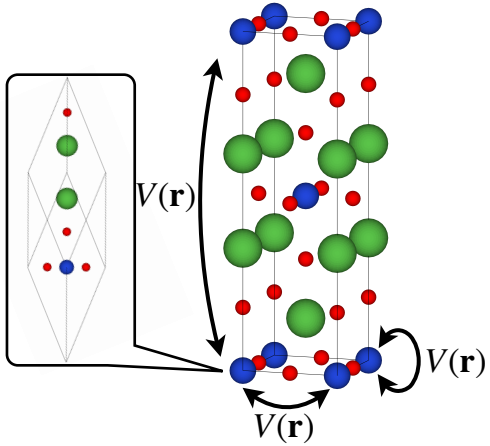

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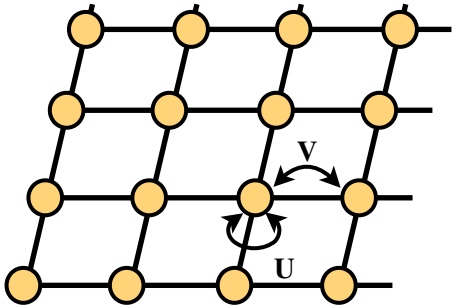

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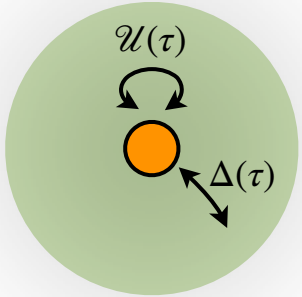

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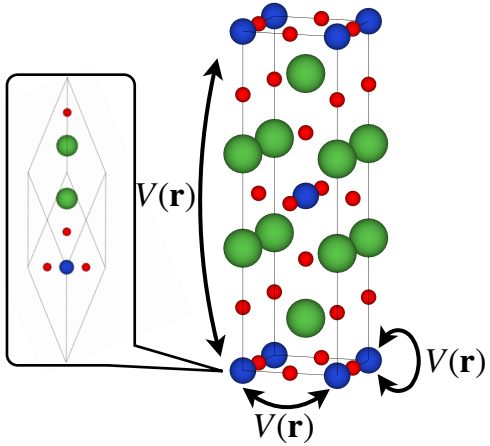

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$$\left[-\frac{1}{2}\nabla^2 + v_{\text{ext}}(\mathbf{r}) + v_{\text{H}}(\mathbf{r}) + v_{\text{xc}}(\mathbf{r})\right]\phi_i(\mathbf{r}) = \epsilon_i\phi_i(\mathbf{r})$$

- **Balance between computational efficiency and accuracy**
- **Broad applicability:** Molecules, materials, surfaces.
- **Dominance:**

  Primary *ab initio* tool across physics, chemistry and materials science.
- **Mature community codes**

Outline

- *Ab Initio* Many-Body Electronic Structure
- CoQuí: **Co**rrelated **Qu**antum ì**i**nterface

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- *Ab Initio* Many-Body Electronic Structure
  - Density functional theory: Challenges
  - *Ab initio* GW method in a nutshell
  - *Ab initio* GW+EDMFT in a nutshell
  - Numerical challenges
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Density Functional Theory: Challenges

Energy functional: $E_0[v_{\text{ext}}] = \langle \Psi \,|\, H \,|\, \Psi \rangle = T + V_{\text{e-e}} + \int d\mathbf{r} \rho(\mathbf{r}) v_{\text{ext}}(\mathbf{r})$

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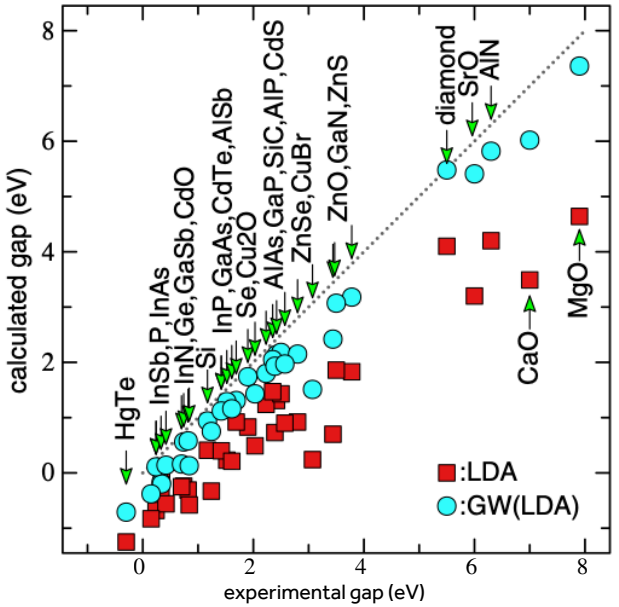
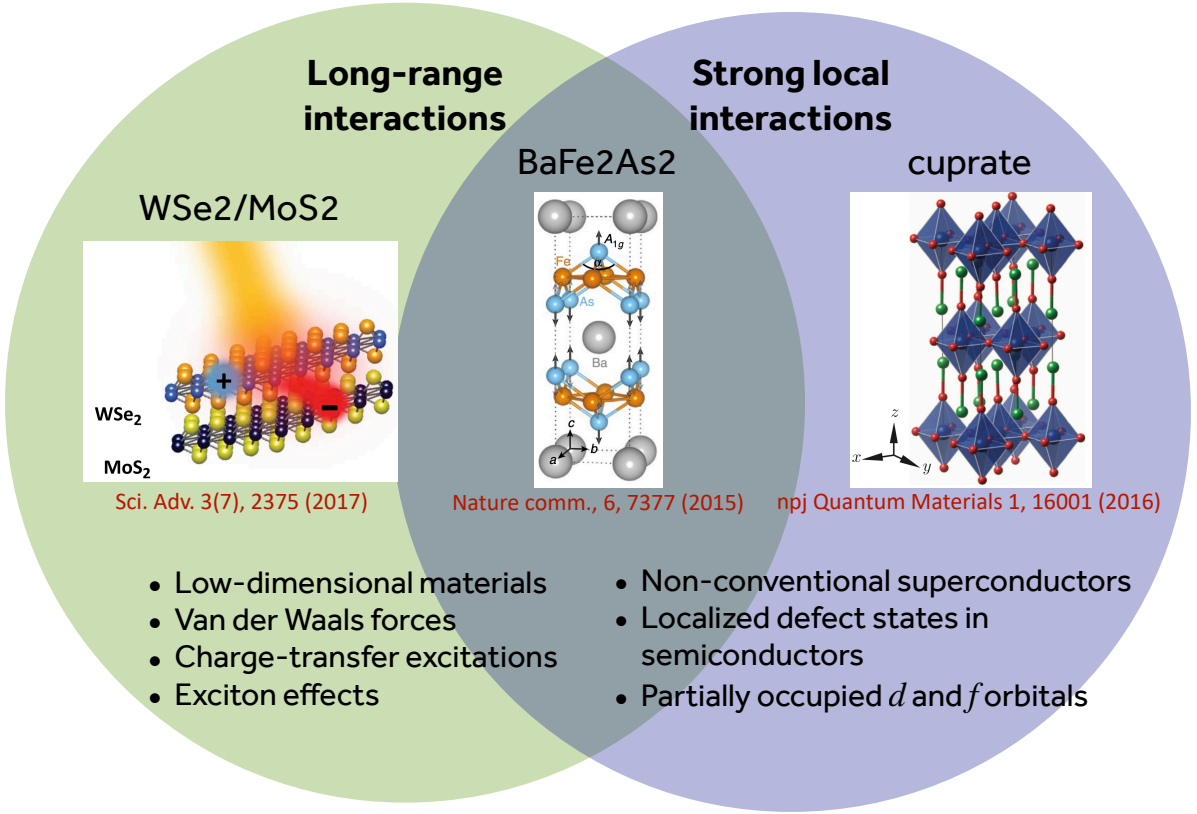
Band-gap Issue

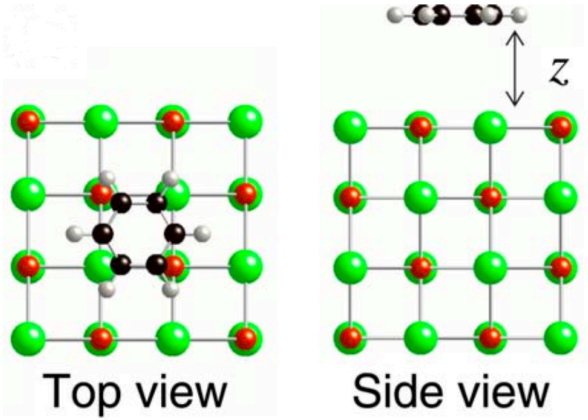


Density Functional Theory: Challenges



Density Functional Theory: Challenges

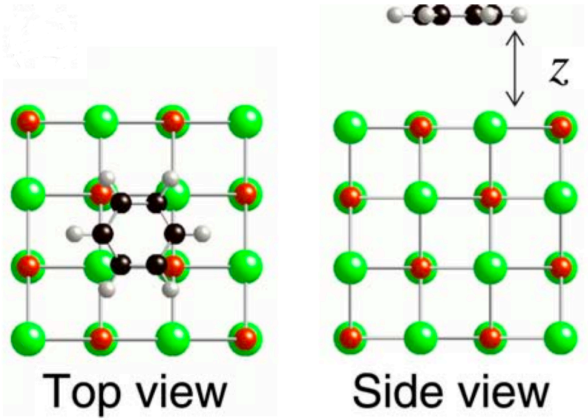
Example: substrate-induced long-range screening in graphene

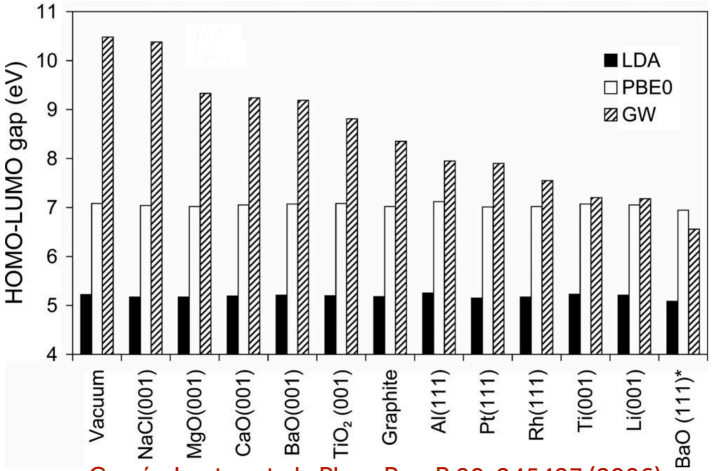


Density Functional Theory: Challenges

Example: substrate-induced long-range screening in graphene

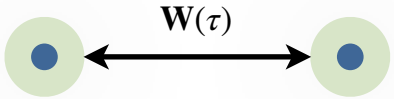
- Local and semi-local functionals rely only on local density properties ☹
- Hybrid functional predicts substrate-independent screening ☹





García-Lastra et al., Phys. Rev. B 80, 245427 (2006)

Ab Initio GW Method

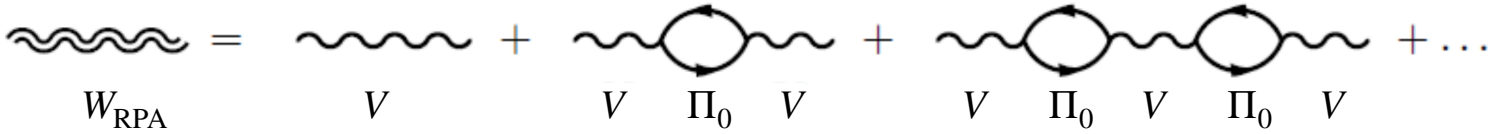


Dressed electrons

"Super" Hartree-Fock with RPA screened interactions

GW approximation: $\Sigma^{\mathrm{GW}}(\tau) = -G(\tau)W_{\mathrm{RPA}}(\tau)$

Long-range screening effects via RPA



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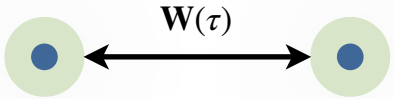

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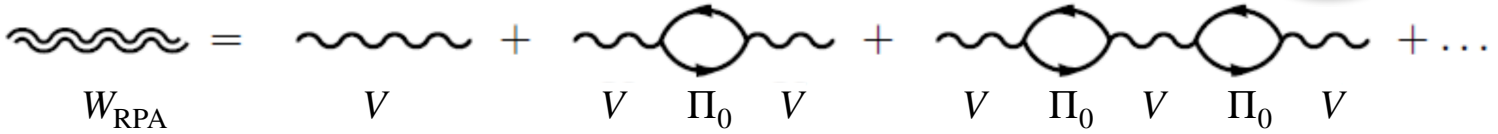

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- **Real Materials:** Hybertsen and Louie, PRL (1985)
- Gold standard for band gap calculations

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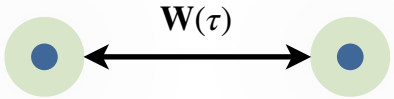

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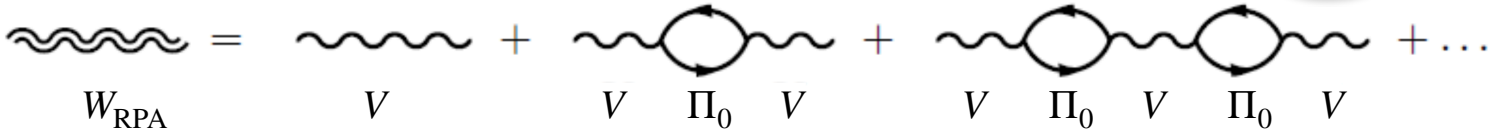

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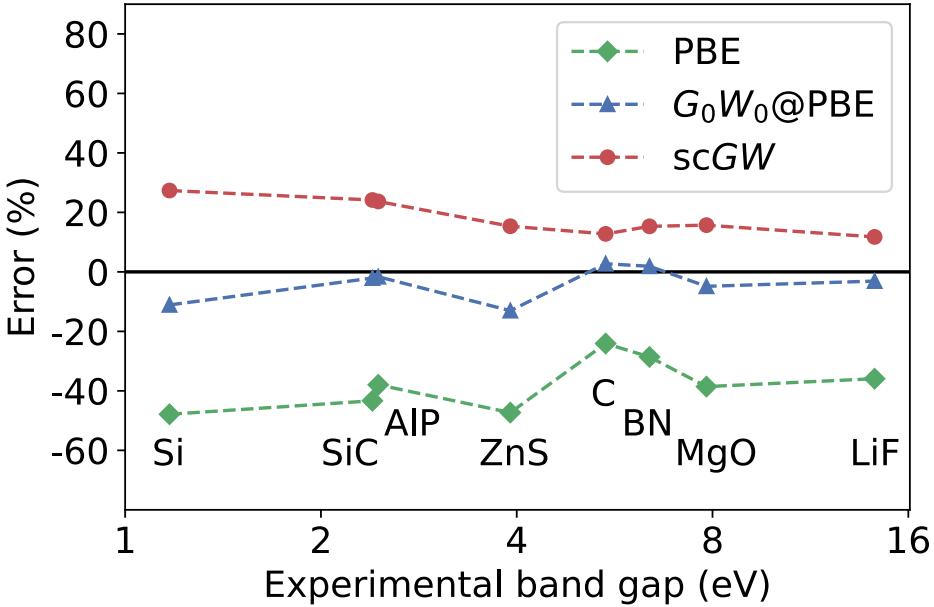
The **standard** implementation of Π_0 in *ab initio* codes is Lindhard equation:

$$\Pi_0 = G_0 * G_0 \rightarrow \text{so-called } G_0W_0 \text{ method}$$

Self-consistent GW using the LW functional $\Phi[G]$is essentially unavailable, except in VASP

Ab Initio GW Method



- Single-shot GW is *surprisingly* accurate, predicting better band gaps for semiconductors.
  a. **Error cancellation** between the vertex corrections and non-self-consistency errors
  b. **Starting-point dependence**
- scGW slightly overestimates the band gaps

Ab Initio GW Method - Summary

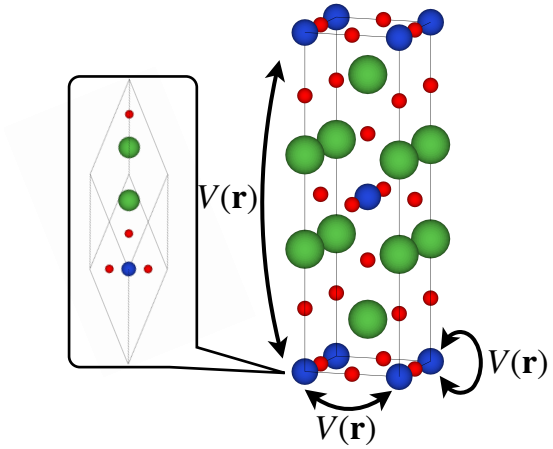
- GW approximation treats long-range interactions using RPA.
- Derived from the approximate Luttinger–Ward functional $\Phi[G]$
  → yields well-defined quasiparticle excitations.
- Most applications reply on single-shot GW with the Lindhard form of Π_0, because:
  - Empirically, "error cancellation" makes it more accurate than self-consistent GW.
  - Self-consistency poses numerical challenges.
- Still insufficient for strong local correlations due to its perturbative nature.

GW+EDMFT in a nutshell

Formal derivation: see Antoine's GW+EDMFT lecture

***Ab initio* GW Toward the Complete Basis Limit**:

- Long-range correlations
- Renormalization from high- and low-lying orbitals



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$$\mathbf{\Sigma}^{\mathbf{k}} = (\mathbf{\Sigma}^{GW})^{\mathbf{k}}$$

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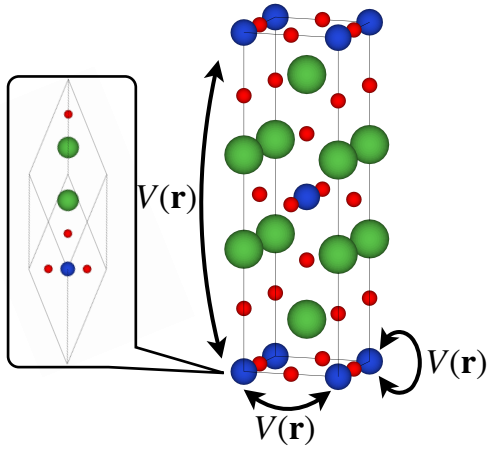

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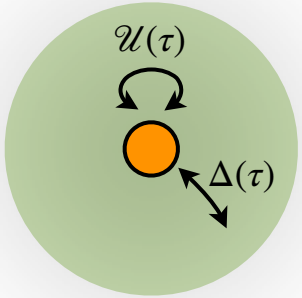
$\mathcal{U}(\tau)$ and $\Delta(\tau)$



$\mathcal{U}(\tau)$: Local dynamic screened interactions

$\Delta(\tau)$: Embedded in an effective bath

S. Biermann *et al.*, Phys. Rev. Lett. 90, 086402 (2003)
P. Sun *et al.*, Phys. Rev. B 66, 085120 (2002)

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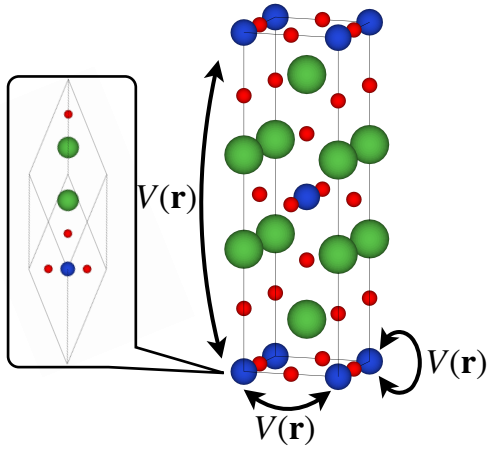
Non-Perturbative Impurity Solver, e.g. CT-SEG

- Σ^{imp}: band structure renormalization beyond GW
- Π^{imp}: Screening beyond RPA



Σ^{imp} and Π^{imp}

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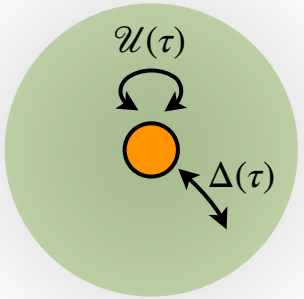

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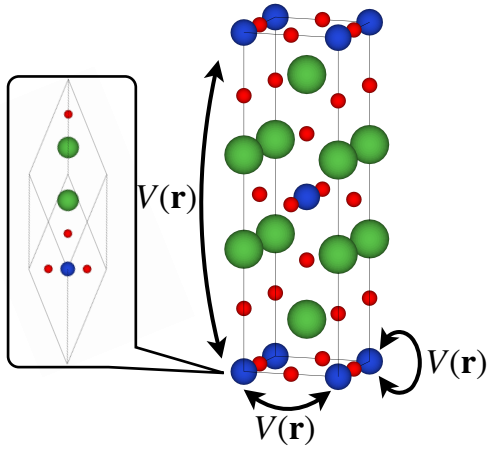

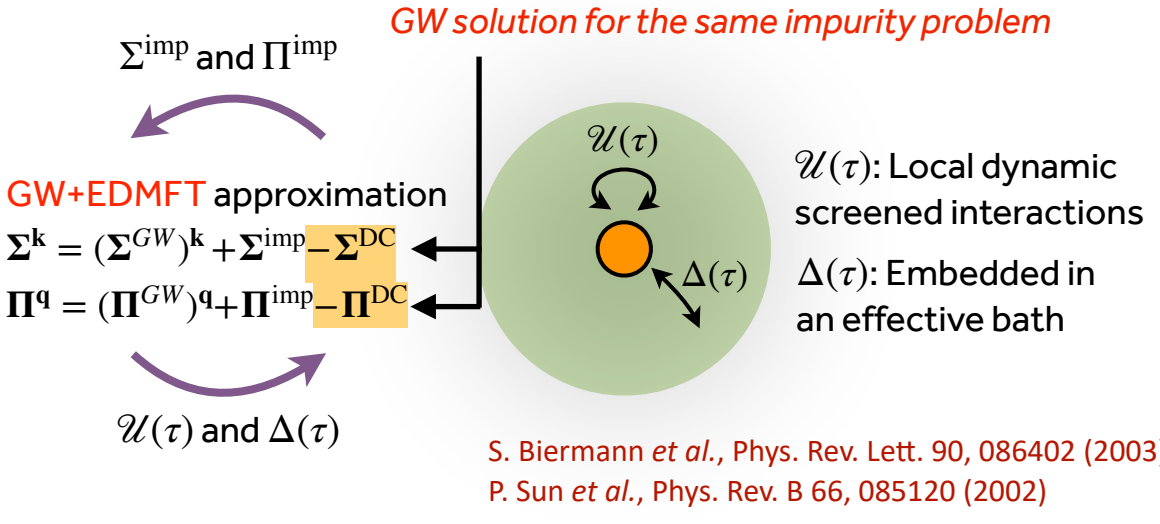

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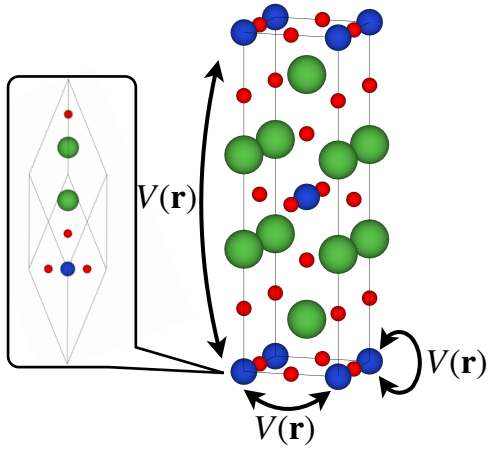
Iterate until $G_{\mathrm{loc}} = G_{\mathrm{imp}}$ *and* $W_{\mathrm{loc}} = W_{\mathrm{loc}}$



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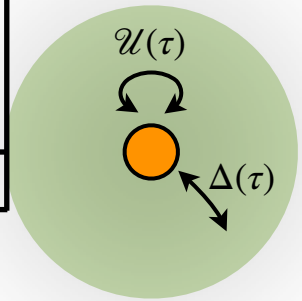
$\mathcal{U}(\tau)$ and $\Delta(\tau)$

GW solution for the same impurity problem



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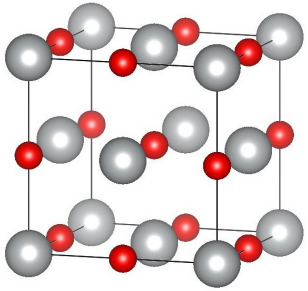
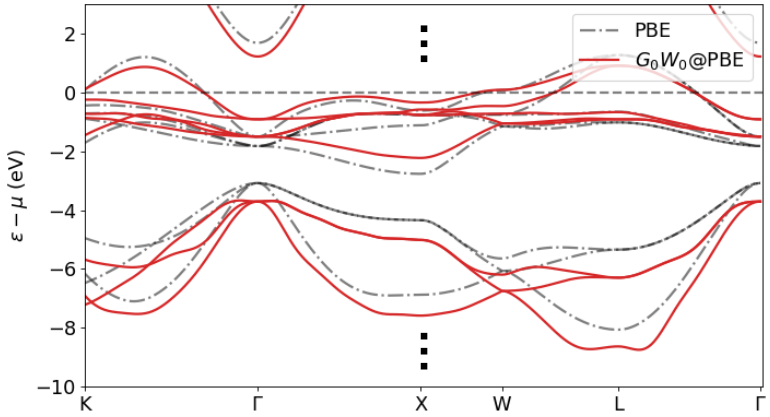
GW+EDMFT example: Paramagnetic NiO

CoQuí × TRIQS



- k-mesh: 9x9x9
- Temperature: 1100 K



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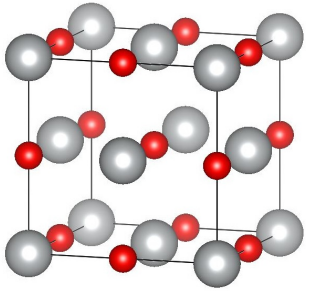

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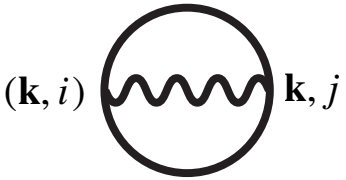
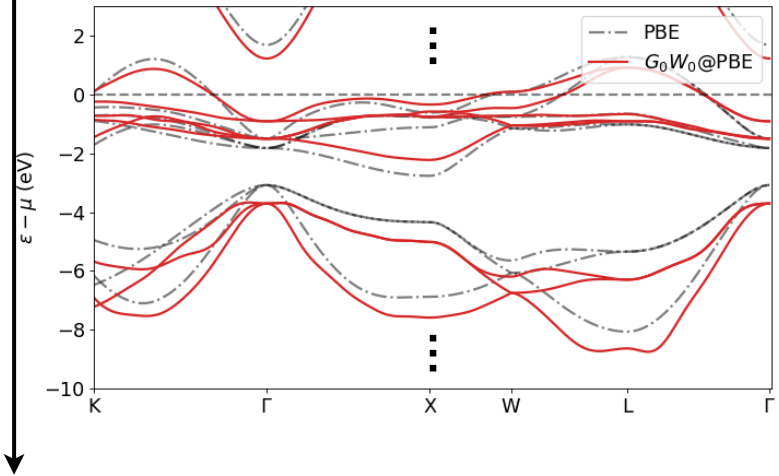
GW Hilbert space



360 eV above E_F



GW+EDMFT example: Paramagnetic NiO

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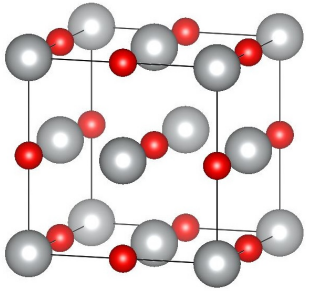

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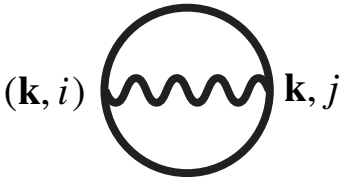
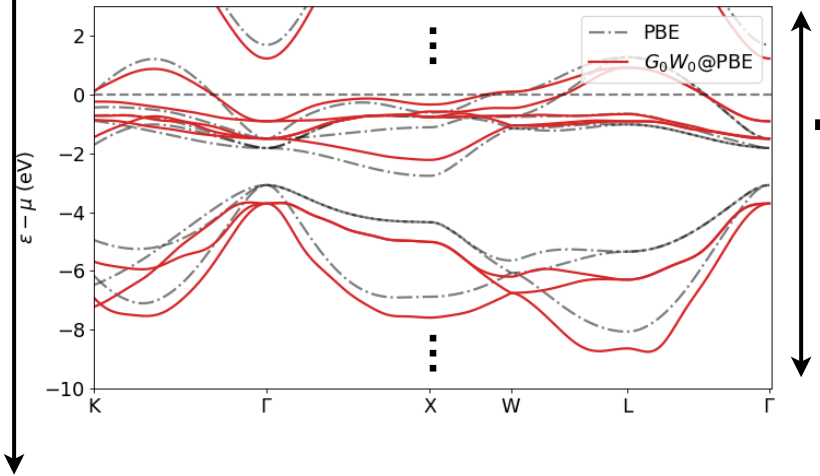
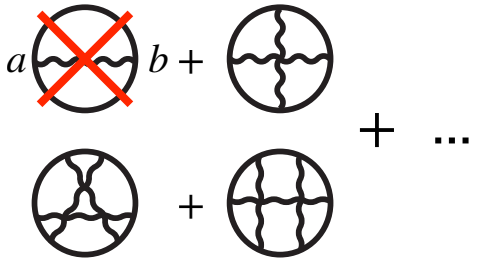




WANNIER90

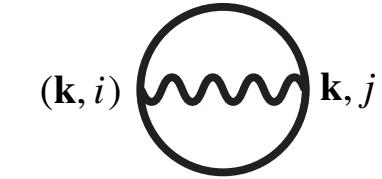
Wannier d orbitals

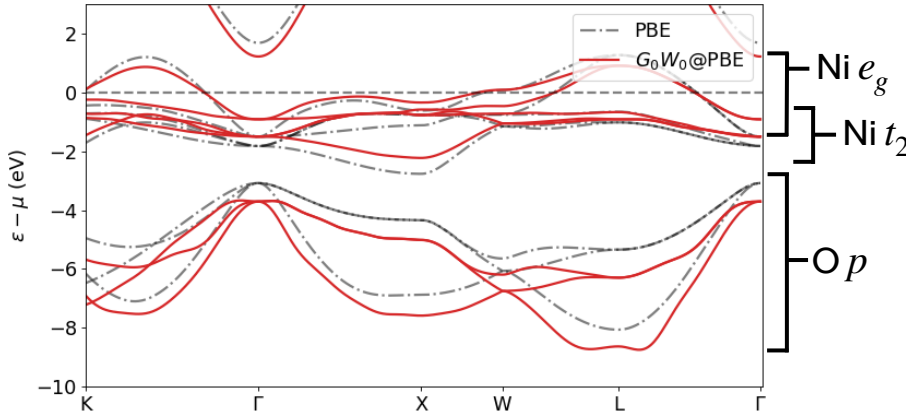


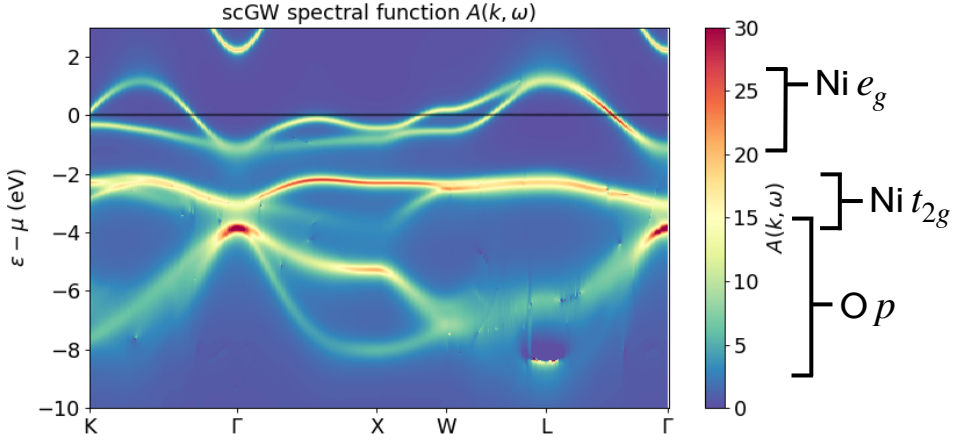
GW+EDMFT example: Paramagnetic NiO

CoQuí × TRIQS

- **Non-local** scGW enhances the t_{2g}-e_g splitting and facilitate hybridization between Ni t_{2g} and O p orbitals



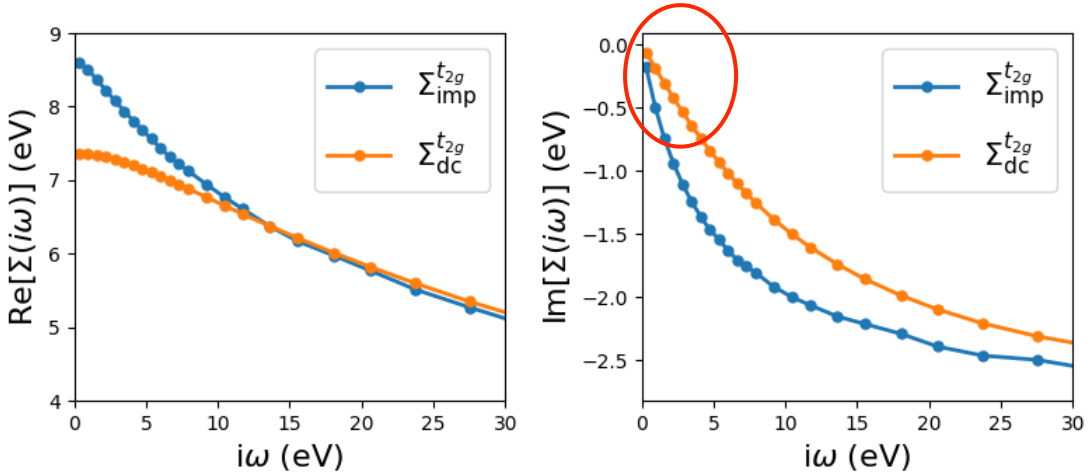




GW+EDMFT example: Paramagnetic NiO

CoQuí × TRIQS

- EDMFT correction to the self-energy $\Delta\Sigma^{GW+\mathrm{EDMFT}} = \Sigma_{\mathrm{imp}} - \Sigma_{\mathrm{dc}}$
  a. Enhances mass renormalization for Ni t_{2g} orbitals: $Z_{\mathrm{GW}} = 0.76 \rightarrow Z_{\mathrm{GW+EDMFT}} = 0.66$



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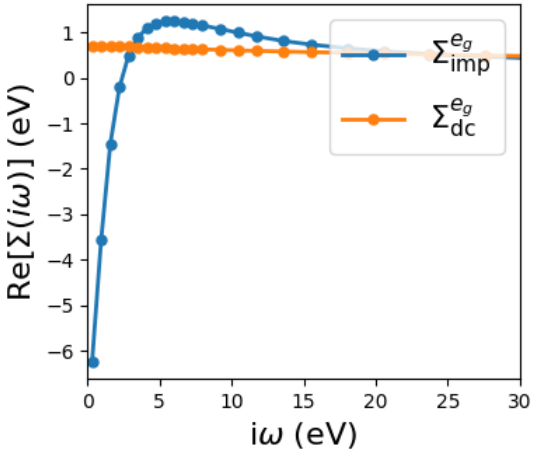
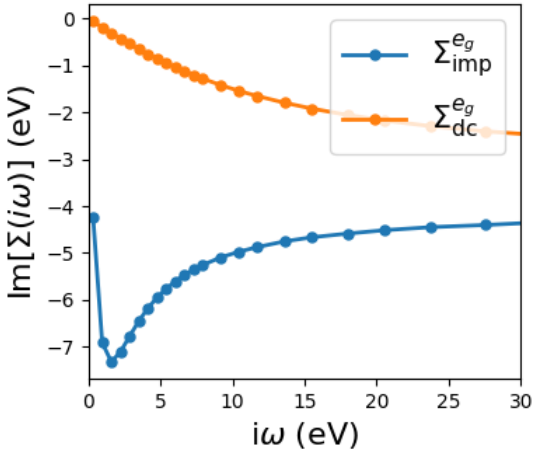
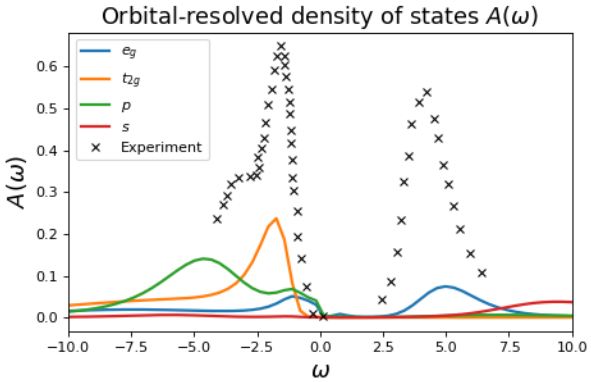
  b. Induces a Mott gap at Ni e_g orbitals







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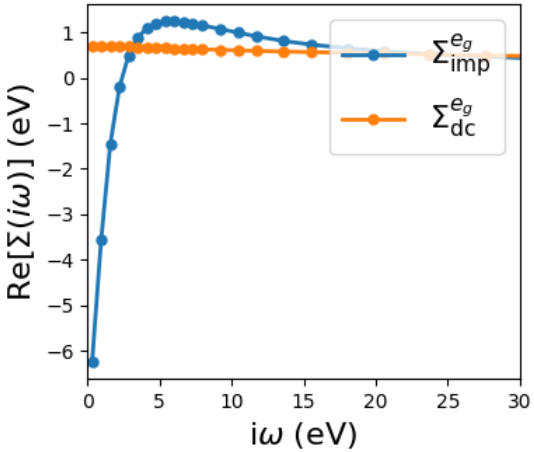
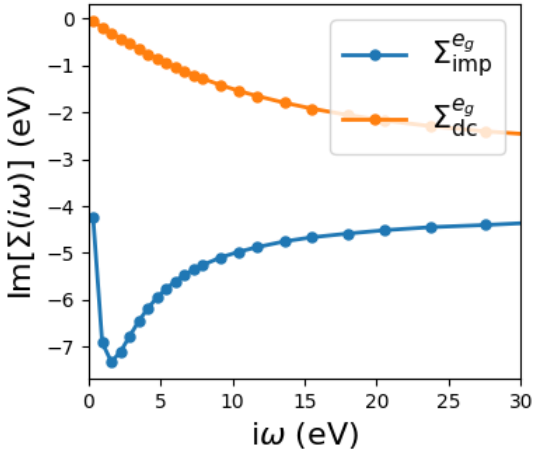
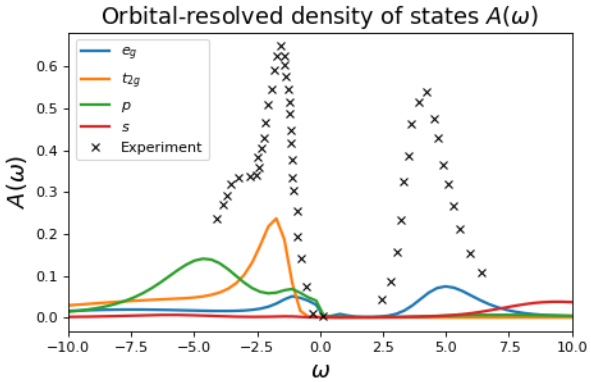
  b. Induces a Mott gap at Ni e_g orbitals
- Occupations: $(N_{e_g}, N_{t_{2g}}) = (2.40,\ 5.89) \rightarrow (2.17,\ 5.91)$

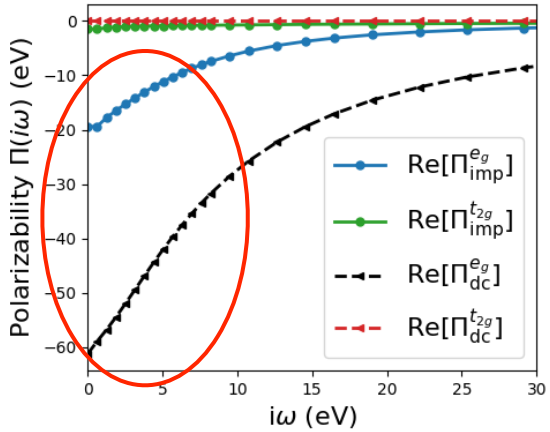






GW+EDMFT example: Paramagnetic NiO

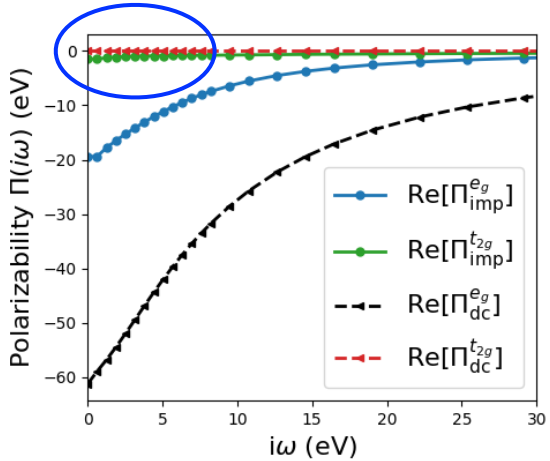
- Local vertex correction $\Delta\Pi^{GW+\mathrm{EDMFT}} = \Pi_{\mathrm{imp}} - \Pi_{\mathrm{dc}}$
  - Suppresses the screening in Ni e_g orbitals



GW+EDMFT example: Paramagnetic NiO

CoQuí × TRIQS

- Local vertex correction $\Delta\Pi^{GW+\mathrm{EDMFT}} = \Pi_{\mathrm{imp}} - \Pi_{\mathrm{dc}}$
  - Suppresses the screening in Ni e_g orbitals
  - Enhances the screening in Ni t_{2g} orbitals



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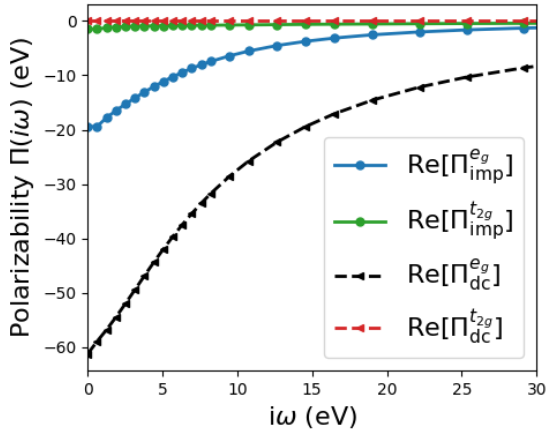
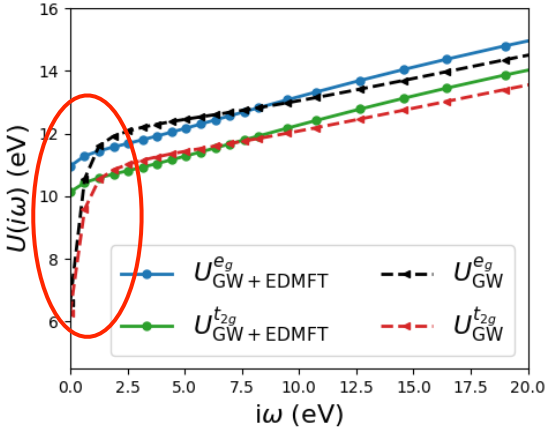
- Local vertex correction $\Delta\Pi^{GW+\mathrm{EDMFT}} = \Pi_{\mathrm{imp}} - \Pi_{\mathrm{dc}}$
  - Suppresses the screening in Ni e_g orbitals
  - Enhances the screening in Ni t_{2g} orbitals
- Strong enhancement of $\mathscr{U}(i\omega)$ in the low-energy regime: ~ 6 → ~ 11 eV





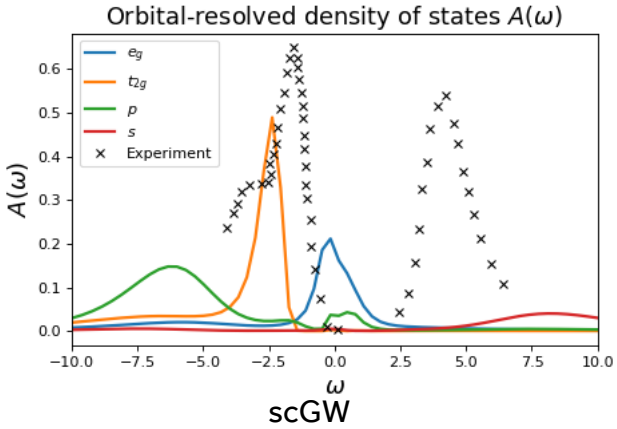
GW+EDMFT example: Paramagnetic NiO

CoQuí × TRIQS

Experiment: Sawatzky, and Allen, PRL. 53 (24) 2339–2342 (1984)

$$\mathbf{\Sigma}^{\mathbf{k}} = (\mathbf{\Sigma}^{GW})^{\mathbf{k}} + \Sigma^{\mathrm{imp}} - \Sigma^{\mathrm{DC}}$$

$$\mathbf{\Pi}^{\mathbf{q}} = (\mathbf{\Pi}^{GW})^{\mathbf{q}} + \Pi^{\mathrm{imp}} - \Pi^{\mathrm{DC}}$$



scGW

	U	U’
e_g	6.62	4.37
t_{2g}	6.31	4.63

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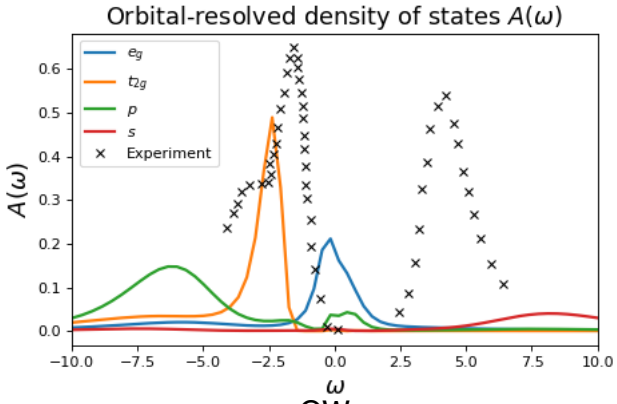
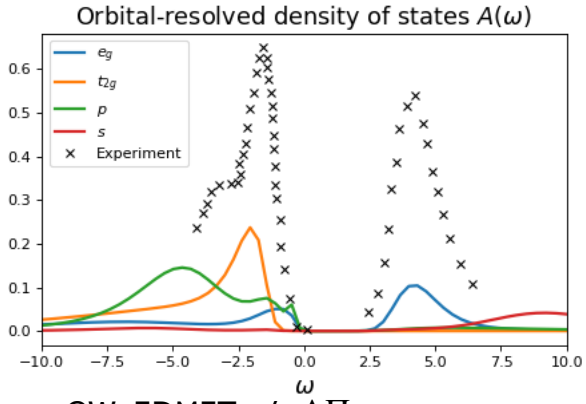


scGW



GW+EDMFT w/o $\Delta\Pi_{\mathrm{GW+EDMFT}}$

	U	U’
e_g	6.62	4.37
t_{2g}	6.31	4.63

	U	U’
e_g	10.47	8.29
t_{2g}	9.68	7.95

GW+EDMFT example: Paramagnetic NiO

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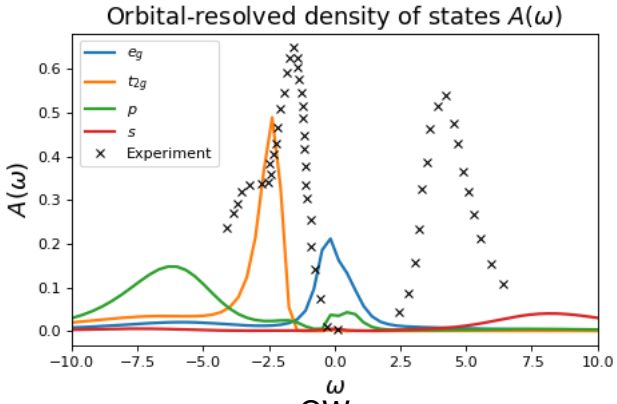

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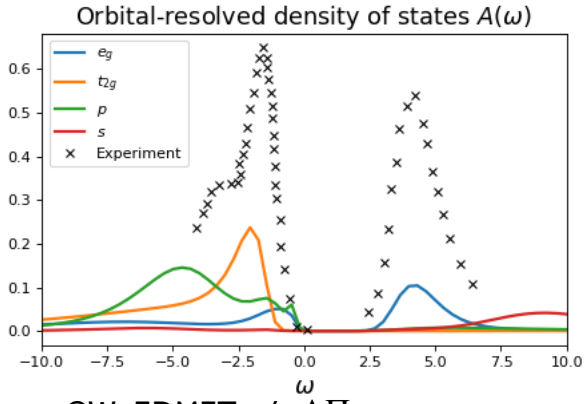

GW+EDMFT w/o $\Delta\Pi_{\mathrm{GW+EDMFT}}$

	U	U’
e_g	10.47	8.29
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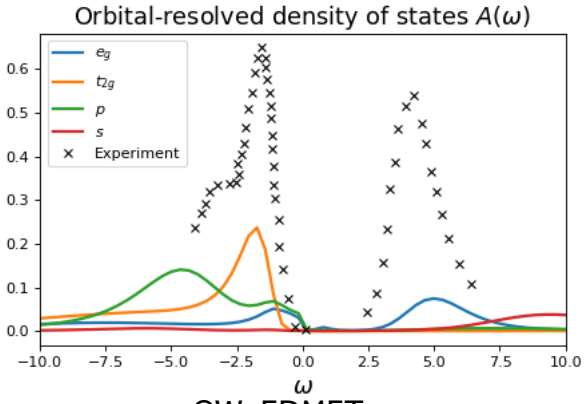


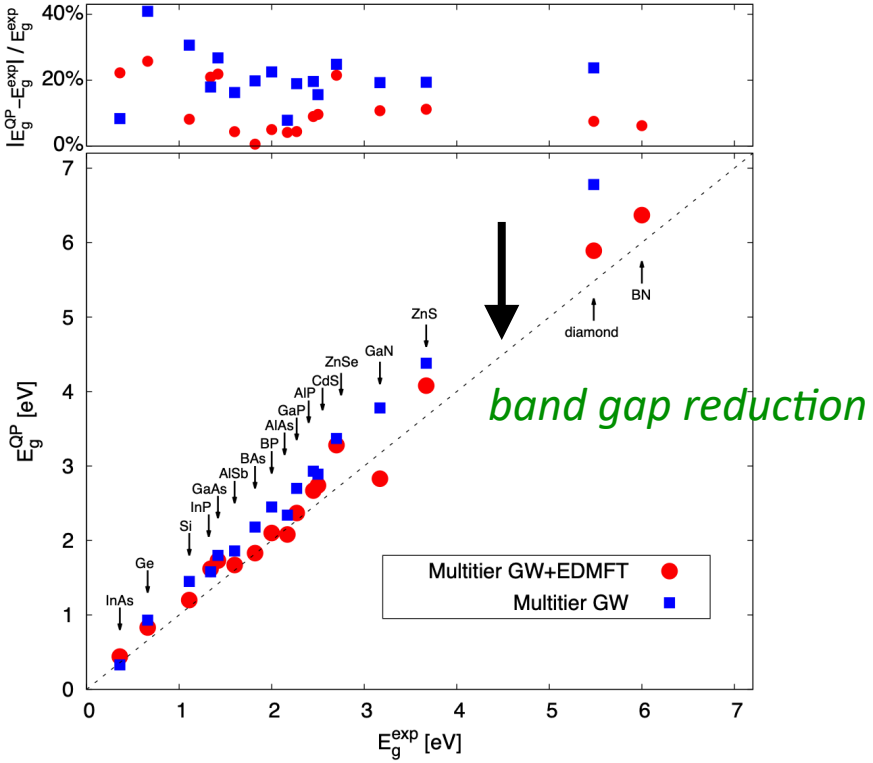
GW+EDMFT

	U	U’
e_g	11.02	8.94
t_{2g}	10.20	8.47

GW+EDMFT example: Semiconductors

Christiansson, Petocchi, Werner, arXiv:2410.19579

- *Multi-tier* GW+EDMFT study on semiconductors and insulators
- $s + p$ Wannier functions on each atomic sites
- Self-consistency within the Wannier space
- $\Delta\Pi^{GW+\mathrm{EDMFT}}$ enhances the screening, resulting in improvements compared to sc*GW*



GW+EDMFT - Summary

Issues of LDA+DMFT:

a. *Ad hoc* Hubbard U

b. Ill-defined double counting issue

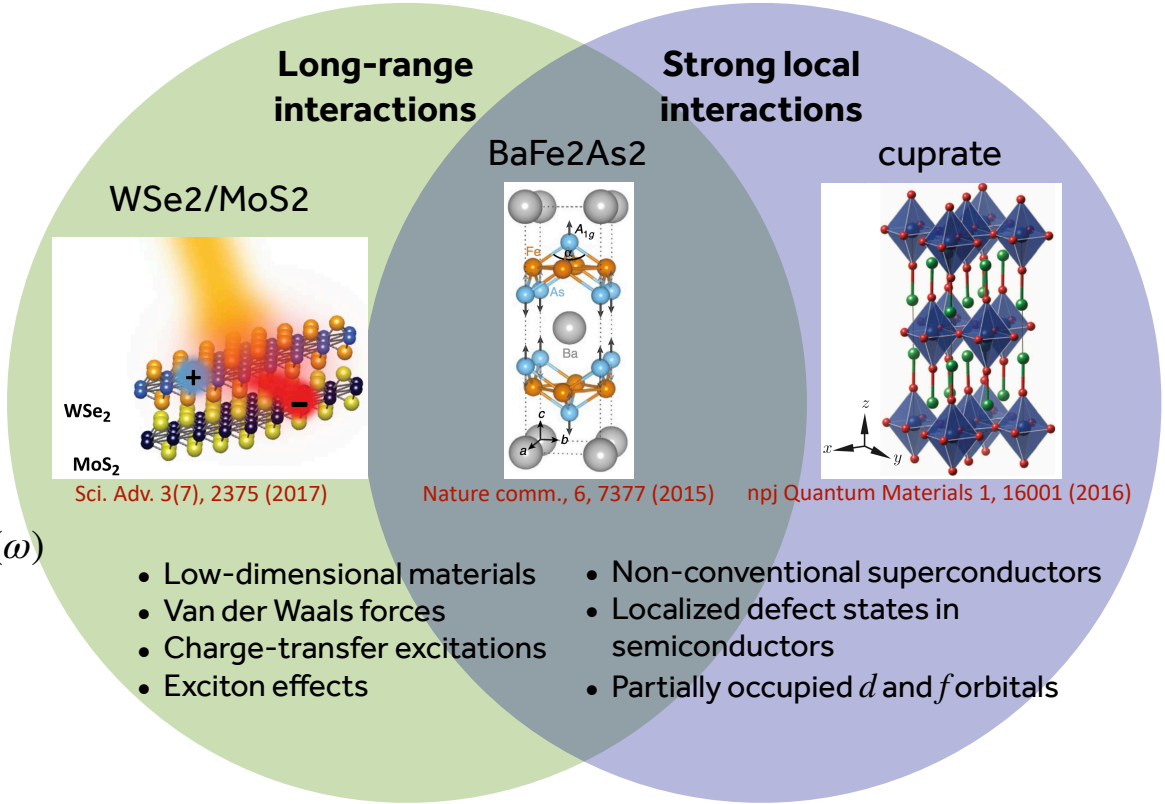
c. Missing long-range correlations

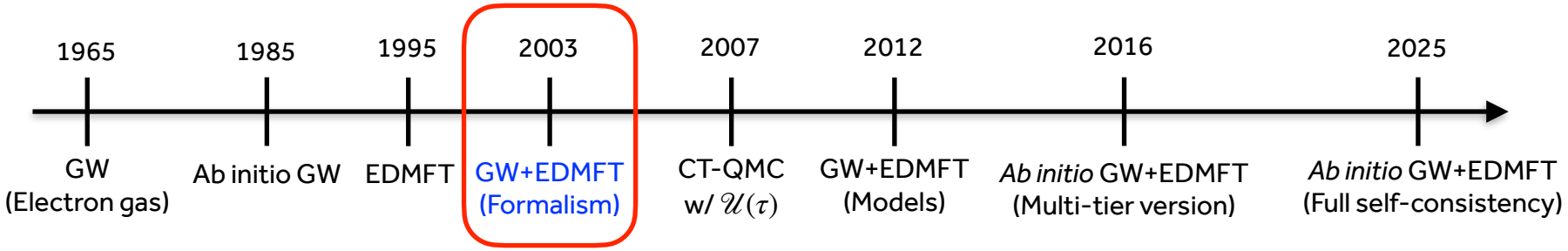
GW+EDMFT

a. **Self-consistent** bosonic Weiss field $\mathcal{U}(\omega)$

b. Double counting rigorously defined via $\Psi[G, W]$ **functional**

c. Long-range correlations at **GW** level





Formalism

S. Biermann *et al.*, Phys. Rev. Lett. 90, 086402 (2003)

P. Sun *et al.*, Phys. Rev. B 66, 085120 (2002)

Models

Phys. Rev. B **87**, 125149 (2013)

Ab initio multi-tier GW+EDMFT

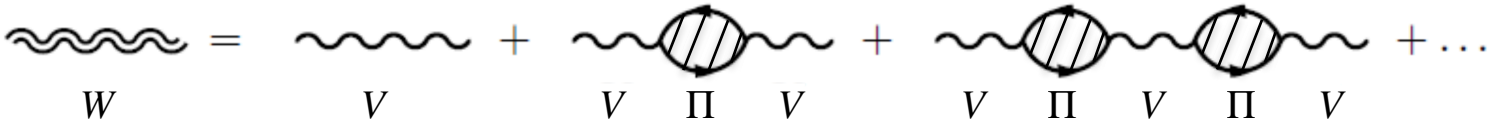
PRM **1**, 043803 (2017)

Ab initio GW+EDMFT

Comput. Phys. Commun. 308 109447 (2025)

Ab initio Many-Body Simulations: Numerical Challenges

Computing and storing *ab initio* self-consistent screened interactions in GW+EDMFT:



$$W_{ijkl}^{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3\mathbf{k}_4}(i\omega) = \int\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \phi_i^{\mathbf{k}_1 *}(\mathbf{r}) \phi_j^{\mathbf{k}_2}(\mathbf{r}) W(\mathbf{r},\mathbf{r}';i\omega) \phi_k^{\mathbf{k}_3 *}(\mathbf{r}') \phi_l^{\mathbf{k}_4}(\mathbf{r}')$$

V: bare interactions

Π: polarizability

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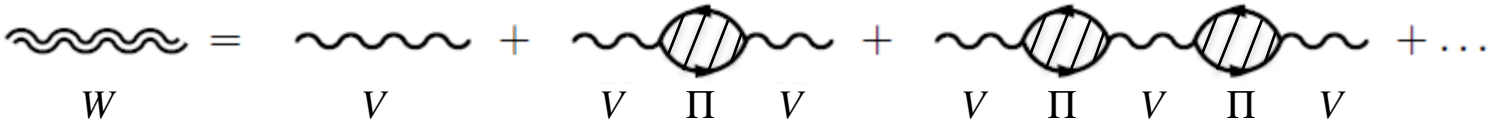

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V: bare interactions
Π: polarizability

1. Explicit Coulomb interaction → high computational cost and memory usage as the system size scales

$$V_{ijkl}^{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3\mathbf{k}_4} \sim O(N_k^3 N_{\mathrm{orb}}^4)$$

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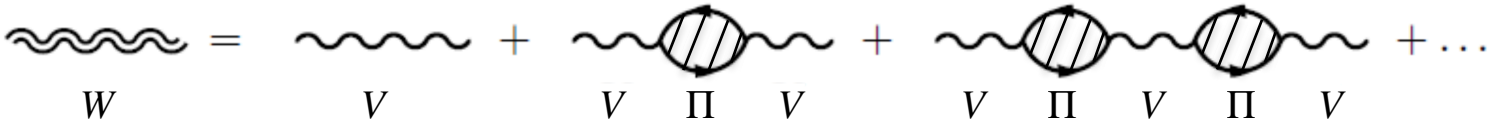

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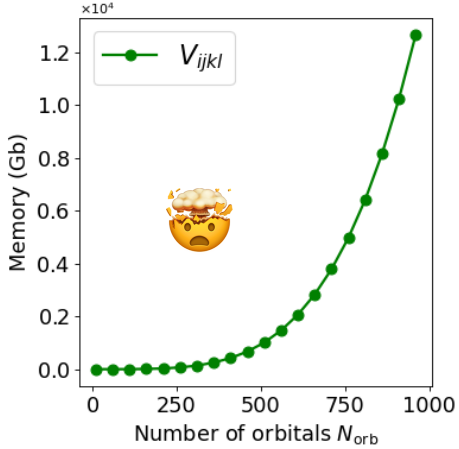

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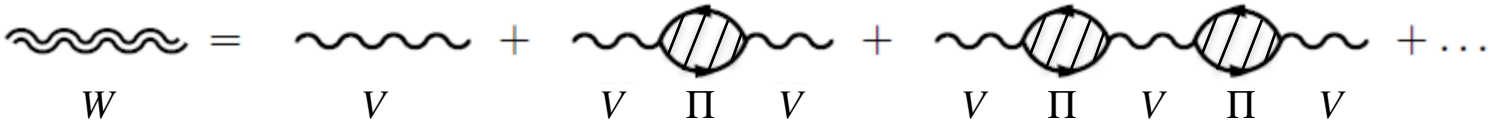


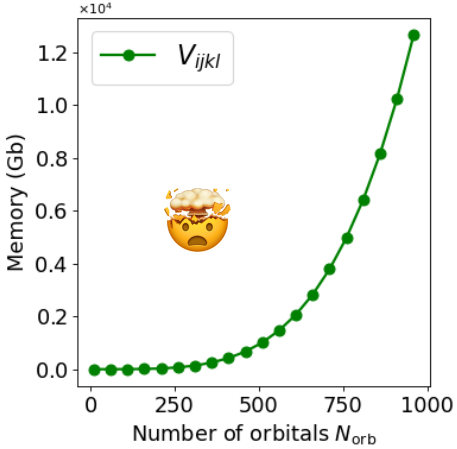
$$W^{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3\mathbf{k}_4}_{ijkl}(i\omega) = \int\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \phi_i^{\mathbf{k}_1 *}(\mathbf{r})\phi_j^{\mathbf{k}_2}(\mathbf{r}) W(\mathbf{r},\mathbf{r}';i\omega)\phi_k^{\mathbf{k}_3 *}(\mathbf{r}')\phi_l^{\mathbf{k}_4}(\mathbf{r}')$$

V: bare interactions

Π: polarizability

2. Representing dynamic correlation functions

$$W^{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3\mathbf{k}_4}_{ijkl}(i\omega) \sim O(N_\omega N_k^3 N_{\mathrm{orb}}^4) \longrightarrow O(N_\omega) \times$$



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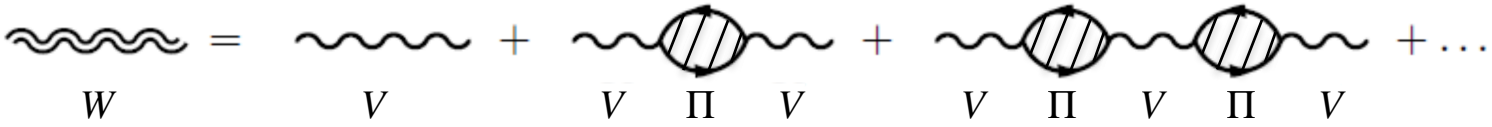

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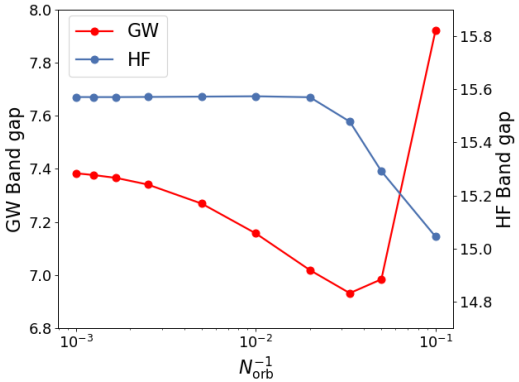
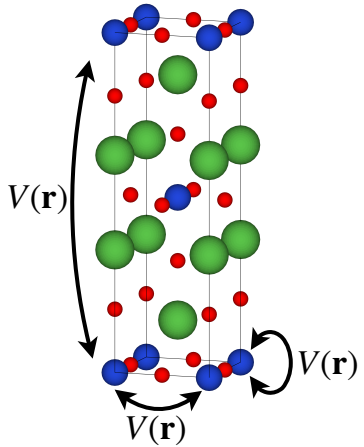
V: bare interactions

Π: polarizability

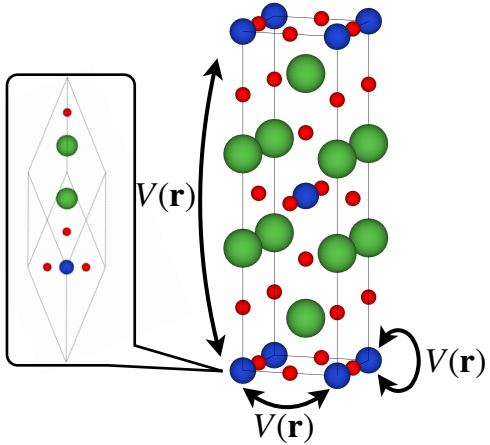
3. Slow convergence in both **basis-set** and **finite-size** errors.





Ab initio MB Electronic Structure – Summary

Quantum materials



- All electrons
- Bare interaction ~ $O(10)$ eV
- Methods: mainly DFT

Scalable frameworks in N_{orb} and N_k, together with efficient software, are essential to make many-body materials simulations practical

CoQuí

Outline

- *Ab Initio* Many-Body Electronic Structure
- CoQuí: **Co**rrelated **Qu**antum ì̲nterface
  - Tensor hypercontraction for Coulomb Hamiltonian
  - *Low-scaling electronic structure methods*

What is CoQuí?

CoQuí, **Co**rrelated **Qu**antum **I**nterface, is a C++/MPI scientific package for *ab initio* electronic structure beyond density functional theory

Goals:

- **Make** many-body methods practical and scalable for large-scale materials simulations
- **Bridge** first-principles simulations and model physics
- **Provide** a flexible platform to test novel algorithms in a first-principles context

- Github repository: https://github.com/AbInitioQHub/coqui
- Discussions page: https://github.com/AbInitioQHub/coqui/discussions
- Tutorials: https://github.com/AbInitioQHub/coqui-tutorial

CoQuí Community

Initiated and hosted at CCQ, Flatiron Institute since 2022

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Main Dev.


Features: GF2, SOSEX, G3W2


TRIQS core libraries: HDF5 C++ interface, NDA, clair/c2py, and ModEST (for embedding)






CoQuí Numerical Setup

CoQuí Numerical Setup

Input:

- Crystal metadata
  (unit cell info, k-points, etc.)

- Single-particle Bloch basis $\phi_i^{\mathbf{k}}(\mathbf{r})$

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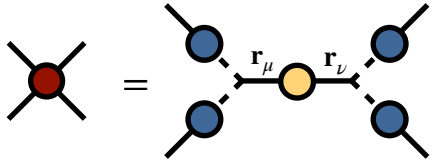
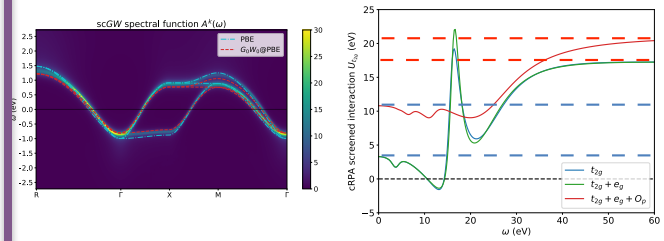
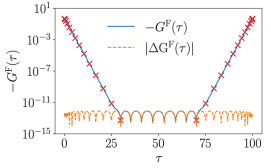
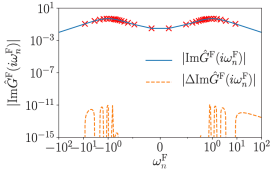




Low-scaling electronic structure beyond DFT

Compact representation of correlation functions





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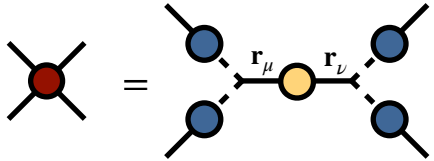

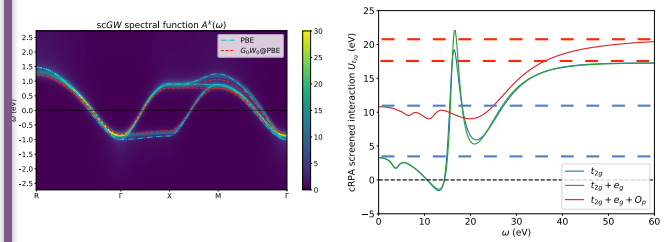

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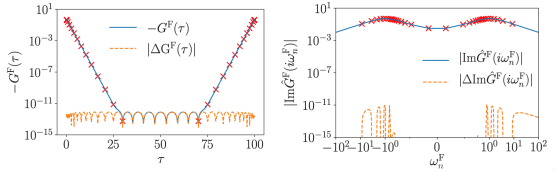

Numerical Pillars of CoQuí's efficiency

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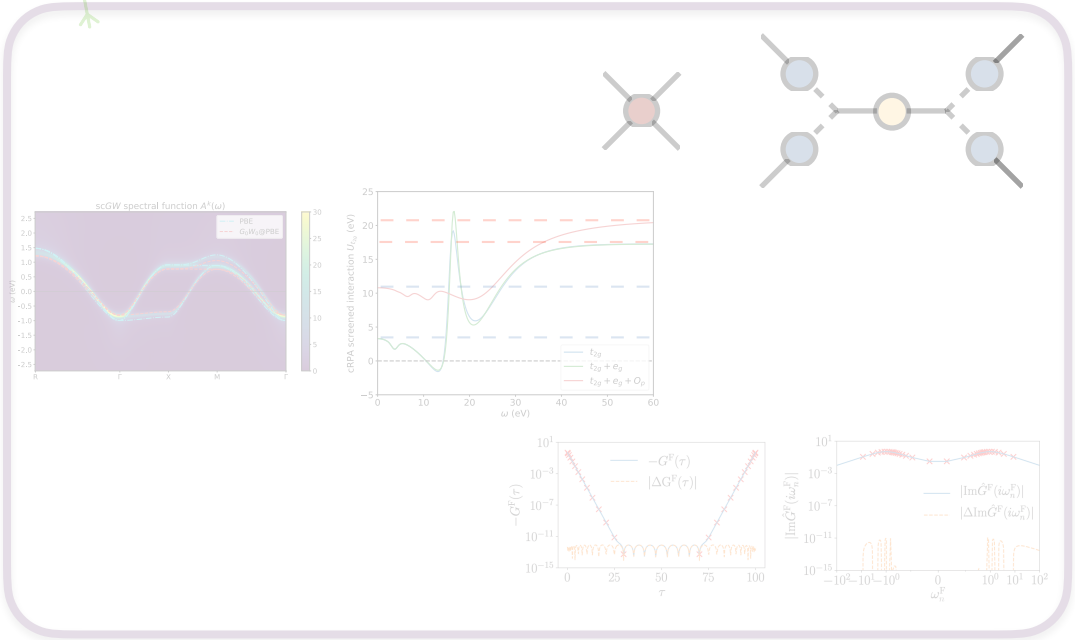

How do we prepare the Bloch basis?

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From DFT to CoQuí

...

From KS-DFT...

$$\left[-\frac{1}{2}\nabla^2 + v_{\text{ext}}(\mathbf{r}) + v_{\text{H}}(\mathbf{r}) + v_{\text{xc}}(\mathbf{r})\right]\phi_i^{\mathbf{k}}(\mathbf{r}) = \epsilon_i^{\mathbf{k}}\phi_i^{\mathbf{k}}(\mathbf{r})$$

We know...

$\{\epsilon_i^{\mathbf{k}}, \phi_i^{\mathbf{k}}(\mathbf{r})\}$ are *auxiliary single-particle states* to describe the the *KS non-interacting* system.

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Use KS orbitals $\phi_i^{\mathbf{k}}(\mathbf{r})$ as a *material-adapted* single-particle basis

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Converters make external DFT results readable in CoQuí

"Agnostic to DFT codes"

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All CoQuí functionality originates from the second quantized Hamiltonian in the user-provided basis:

$$H^{\text{bare}} = \sum_{ij}\sum_{\mathbf{k}} (H_0)_{ij}^{\mathbf{k}} c_i^{\mathbf{k}\dagger} c_j^{\mathbf{k}} + \frac{1}{2}\sum_{ijkl}\sum_{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3\mathbf{k}_4} V_{ijkl}^{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3\mathbf{k}_4} c_i^{\mathbf{k}_1\dagger} c_k^{\mathbf{k}_3\dagger} c_l^{\mathbf{k}_4} c_j^{\mathbf{k}_j}$$

"Bare" Coulomb interactions:

$$V_{ijkl}^{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3\mathbf{k}_4} = \int\int \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}' \phi_i^{\mathbf{k}_1 *}(\mathbf{r})\phi_j^{\mathbf{k}_2}(\mathbf{r})\frac{1}{|\mathbf{r}-\mathbf{r}'|}\phi_k^{\mathbf{k}_3 *}(\mathbf{r}')\phi_l^{\mathbf{k}_4}(\mathbf{r}') \sim O(10)\ \text{eV}$$

From DFT to CoQuí

Converters make external DFT results readable in CoQuí

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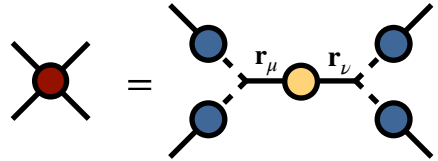
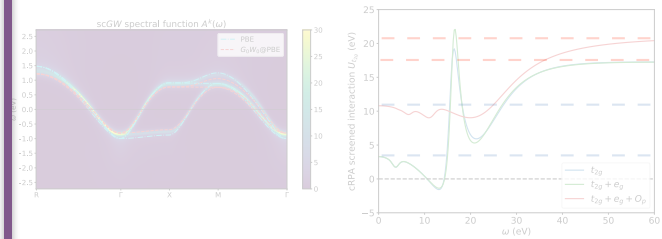
Memory cost quickly becomes unmanageable...

CoQuí Numerical Setup

CoQuí

THC compression for many-body Hamiltonian





Numerical Pillars of CoQuí's efficiency

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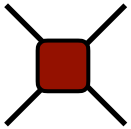
Tensor Hypercontraction (THC)

- **Systematic compression of bare Coulomb integrals:**

$$V_{ijkl}^{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_k\mathbf{k}_l} = \int d\mathbf{r} \int d\mathbf{r}' \phi_i^{\mathbf{k}_i *}(\mathbf{r}) \phi_j^{\mathbf{k}_j}(\mathbf{r}) \frac{1}{|\mathbf{r}-\mathbf{r}'|} \phi_k^{\mathbf{k}_k *}(\mathbf{r}') \phi_l^{\mathbf{k}_l}(\mathbf{r}')$$

Building blocks for microscopic many-body theories



- Memory: $O(N_k^3 N_{\text{orb}}^4)$

ISDF:
Lu and Ying, JCP 302, 329-335 (2015)
Lu and Ying, Ann. Math. Sci. Appl. 1, 321 (2016)
Momentum-dependent ISDF:
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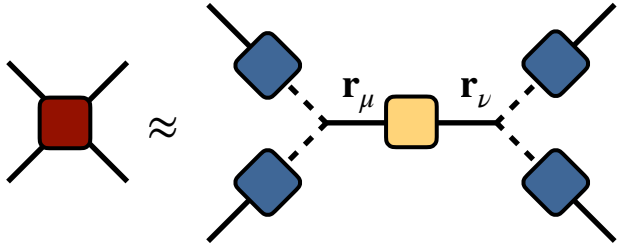
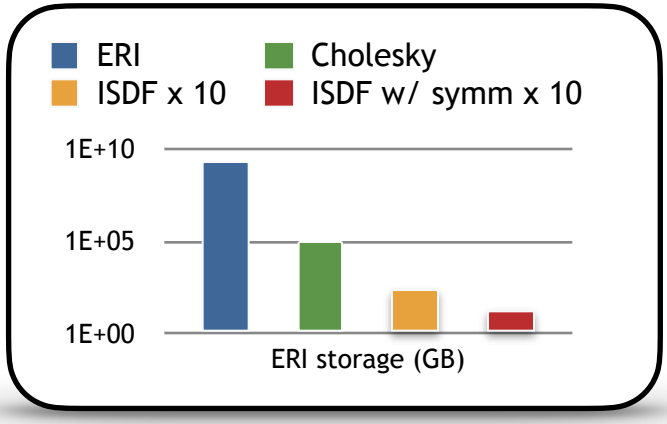


- Memory: $O(N_k^3 N_{\text{orb}}^4) \rightarrow O(N_k N_{orb}^2)$



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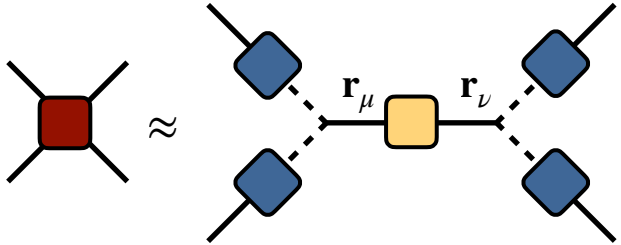
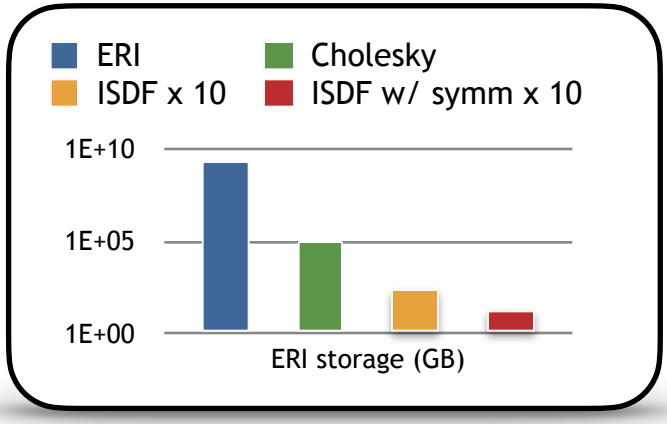


- Memory: $O(N_k^3 N_{\text{orb}}^4) \rightarrow O(N_k N_{orb}^2)$
- Utilize the low-rank structure of $\phi_i^{\mathbf{k}_i *}(\mathbf{r}) \phi_j^{\mathbf{k}_j}(\mathbf{r})$
  - Accuracy is controlled by the size of $\{\mathbf{r}_\mu\}_{\mu=1}^{N_\mu}$, $N_\mu = \alpha N_{\text{orb}} \ll O(N_r)$
  - $N_\mu \approx 10 N_{\text{orb}}$ for 10^{-3} a.u. accuracy



Reformulation of GW in the ISDF representation

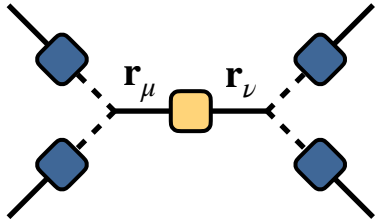
Yeh and Morales, JCTC, 20, 8, 3184–3198 (2024)

GW approximation: $\Sigma^{\mathrm{GW}}(\tau) = -G(\tau)W(\tau)$

Tensor hypercontraction

$$V = \sim\sim\sim$$

$$\approx \sum_{\mu\nu}^{N_\mu} \phi_i^{\mathbf{k}_i *}(\mathbf{r}_\mu)\phi_j^{\mathbf{k}_j}(\mathbf{r}_\mu)V_{\mu\nu}^{\mathbf{q}}\phi_k^{\mathbf{k}_k *}(\mathbf{r}_\nu)\phi_l^{\mathbf{k}_l}(\mathbf{r}_\nu)$$



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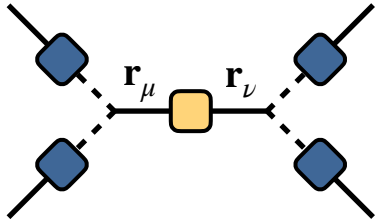
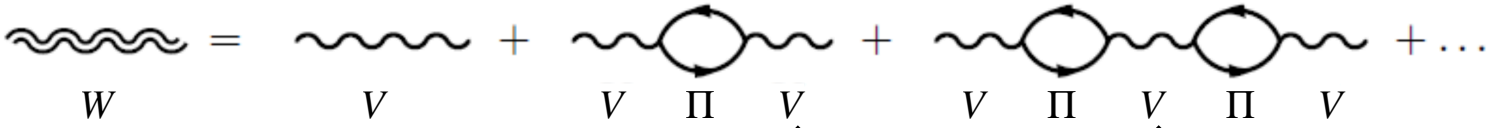
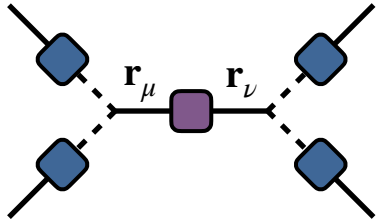


RPA long-range screening:



$$\approx \sum_{\mu\nu}^{N_\mu} \phi_i^{\mathbf{k}_i *}(\mathbf{r}_\mu)\phi_j^{\mathbf{k}_j}(\mathbf{r}_\mu)W_{\mu\nu}^{\mathbf{q}}(\tau)\phi_k^{\mathbf{k}_k *}(\mathbf{r}_\nu)\phi_l^{\mathbf{k}_l}(\mathbf{r}_\nu)$$



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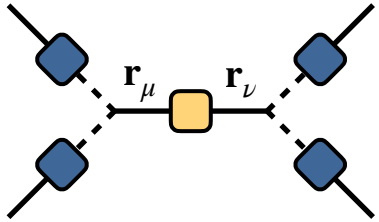

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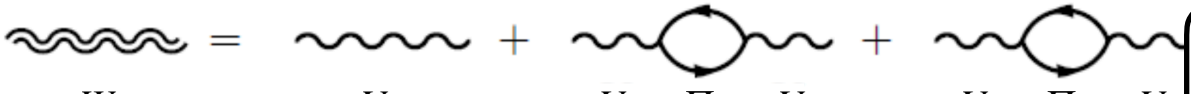

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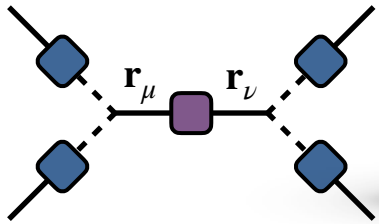
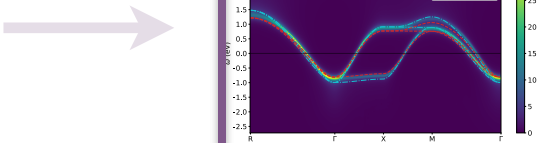
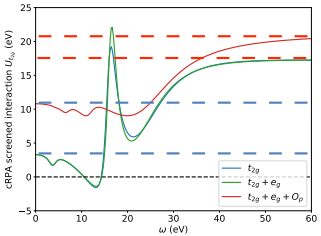


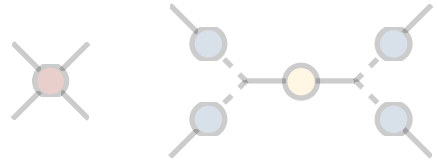
THC-GW:
Low-scaling algorithm at $O(N_\tau N_\mu^3 N_{k_{\mathrm{ibz}}})$ *cost by taking advantages of THC*

CoQuí Numerical Setup

CoQuí







Low-scaling
electronic structure
beyond DFT

Numerical Pillars of CoQuí's efficiency

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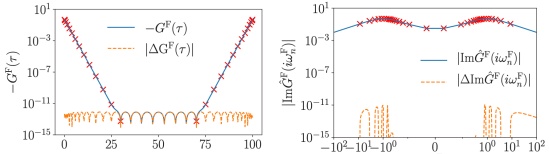
CoQuí Numerical Setup

CoQuí



Compact representation of correlation functions

Similar concept as the DLR basis, cf. Day 1 TRIQS basic tutorial

Numerical Pillars of CoQuí's efficiency

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CoQuí Summary

CoQuí is a package for electronic structure package beyond DFT, built on

- **THC compression** for Coulomb Hamiltonian
- **Low-scaling algorithms** for many-body electronic structure solvers
- **Compact representations** of correlation functions

*CoQuí is not limited to GW+EDMFT — it is a **general, scalable platform** for many-body electronic structure methods beyond DFT*

What can CoQuí do right now?

c.f. CoQuí repository

Compressed Representation for Many-Body Hamiltonians

- THC representation for two-electron Coulomb integrals [ref1, ref2].
- Cholesky decomposition for two-electron Coulomb integrals.

Many-Body Perturbation Theory

- Hartree-Fock [ref]
- RPA correlation energy [ref]
- GW approximation [ref]
- Second-order exchange (SOX) diagram [ref]
- Self-consistency with quasiparticle approximation
- Self-consistency with full frequency dependence

Downfolding for effective low-energy Hamiltonians

- Maximally localized Wannier functions via Wannier90 interface
- Constrained RPA to calculate screened interactions [ref]
- Local effective low-energy Hamiltonian for further correlated calculations [ref]

What can CoQuí do right now?

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- GW approximation [ref] Tutorial 2
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Downfolding for effective low-energy Hamiltonians

- Maximally localized Wannier functions via Wannier90 interface Tutorial 1
- Constrained RPA to calculate screened interactions [ref] Tutorial 3
- Local effective low-energy Hamiltonian for further correlated calculations [ref] Tutorial 4

Thank you.

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Additional slides

CoQuí interface: Python v.s. Input Files

Python interface

```python
# hf.py
# Declare the target system
mf_params = {
    "prefix": "svo",
    "outdir": "./out"
}
svo_mf = coqui.make_mf(mpi, params=mf_params, mf_type="qe")

# Build the Coulomb Hamiltonian
eri_params = {
    "ecut": 40,
    "thresh": 1e-3,
}
svo_thc = coqui.make_thc_coulomb(mf=svo_mf, params=eri_params)

# Hartree-Fock
hf_params = {
    "beta": 300, "wmax": 3.0,
    "output": "svo_hf",
}
coqui.run_hf(params=hf_params, h_int=svo_thc)
```

Input-file-driven interface

```toml
# hf.toml
[mean_field.qe]
name      = "svo_mf"
prefix    = "svo"
outdir    = "./out"

[interaction.thc]
name         = "svo_thc"
mean_field   = "svo_mf"
ecut         = 40
thresh       = 1e-3

[hf]
interaction = "svo_thc"
beta        = 300
wmax        = 3.0
output      = "svo_hf"
```

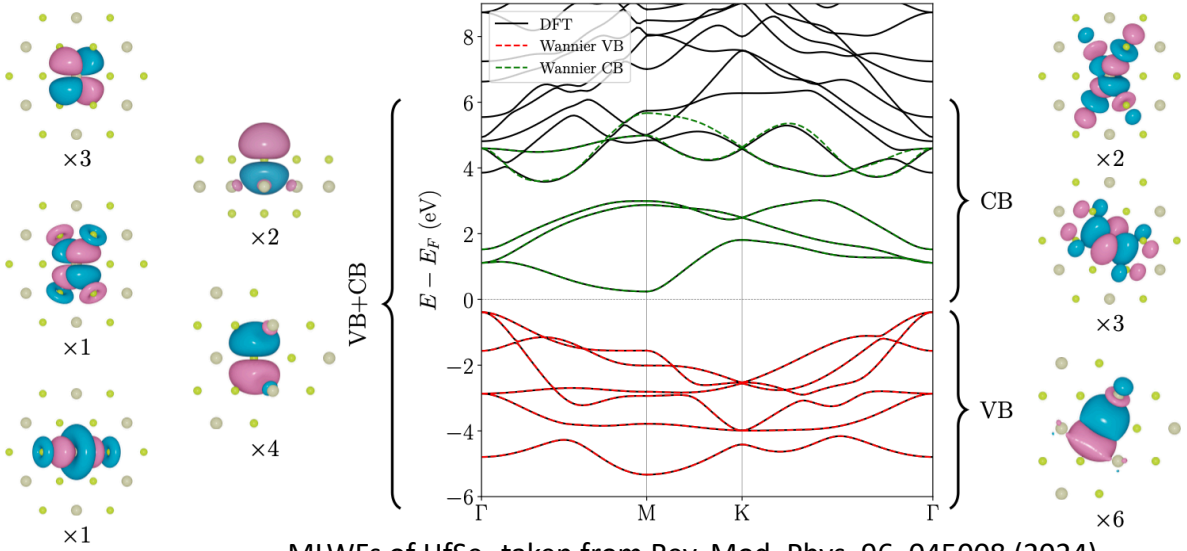
CoQuí Feature: Wannier90 interface

N. Marzari and D. Vanderbilt, PRB 56, 12847 (1997)

Output: MLWFs $\leftarrow |\chi_a^{\mathbf{R}}\rangle = \frac{V}{2\pi^3}\int_{\mathrm{BZ}}\left[\sum_i U_{ai}^{\mathbf{k}}\,|\phi_i^{\mathbf{k}}\rangle\right]e^{-\mathbf{kR}}d\mathbf{k}$

Iteratively optimized for maximal localization

Input: Ab initio KS orbitals



MLWFs of $HfSe_2$ taken from Rev. Mod. Phys. 96, 045008 (2024)

CoQuí Feature: Wannier90 interface

N. Marzari and D. Vanderbilt,
PRB 56, 12847 (1997)

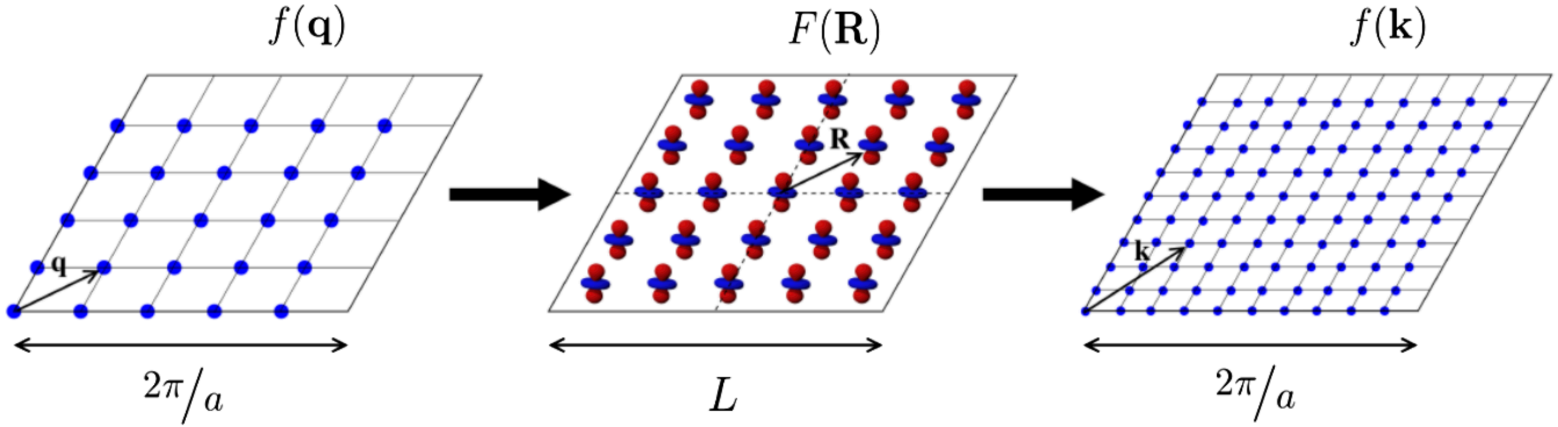
Output: MLWFs $\leftarrow |\chi_a^{\mathbf{R}}\rangle = \frac{V}{2\pi^3}\int_{\mathrm{BZ}}\left[\sum_i U_{ai}^{\mathbf{k}}\,|\phi_i^{\mathbf{k}}\rangle\right]e^{-\mathbf{k}\mathbf{R}}d\mathbf{k}$

Iteratively optimized for maximal localization

Input: Ab initio KS orbitals

- **Wannier interpolation** for many-body solutions at **fine k-resolution**

N. Marzari, *et. al.*
RMP. 84, 1419 (2012)



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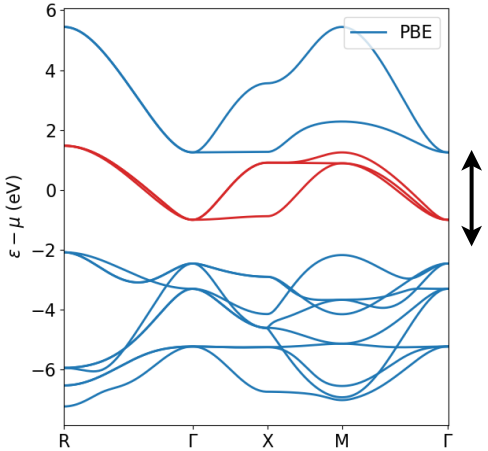
N. Marzari and D. Vanderbilt, PRB 56, 12847 (1997)

Output: MLWFs $\leftarrow |\chi_a^{\mathbf{R}}\rangle = \frac{V}{2\pi^3}\int_{\mathrm{BZ}}\left[\sum_i U_{ai}^{\mathbf{k}} |\phi_i^{\mathbf{k}}\rangle\right] e^{-\mathbf{k}\mathbf{R}} d\mathbf{k}$

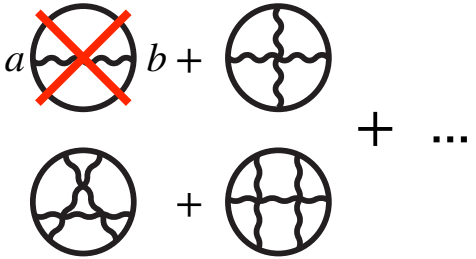
Iteratively optimized for maximal localization

Input: Ab initio KS orbitals

- Physically motivated subspaces for embedding methods such as DMFT

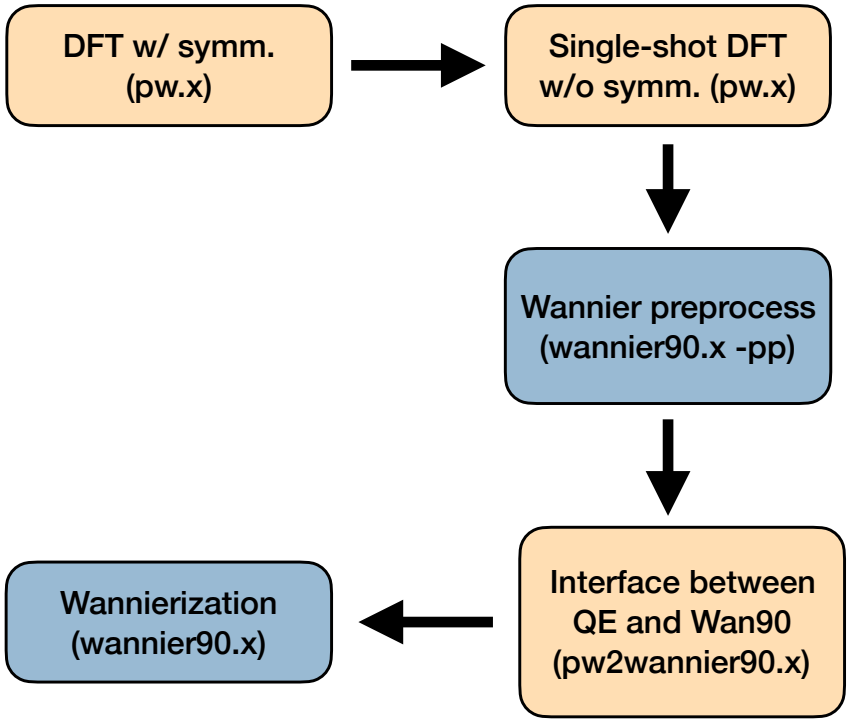


EDMFT subspace:
Ti t_{2g} orbitals



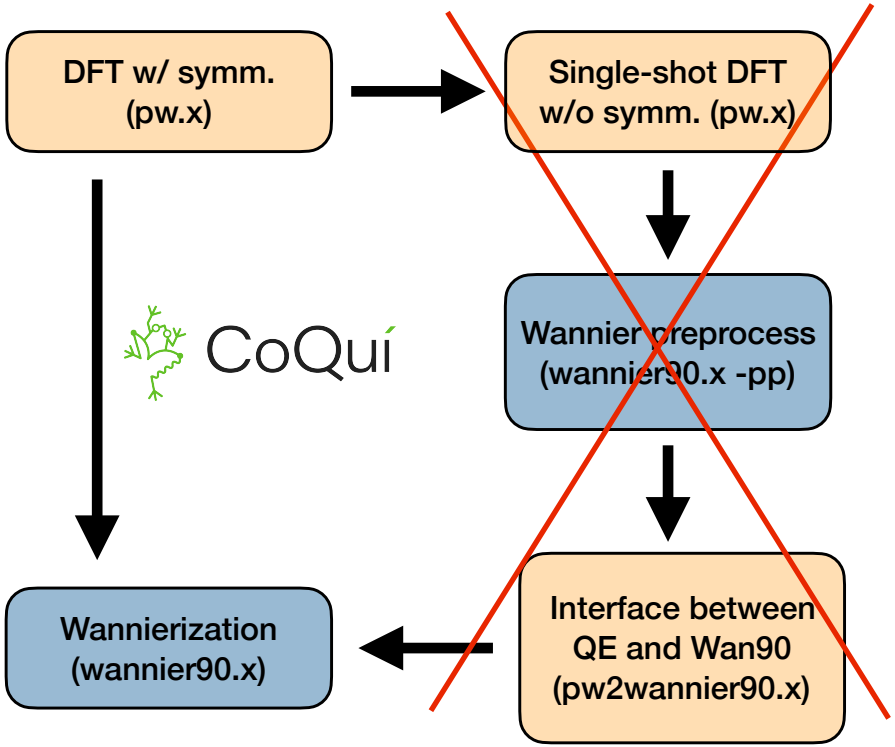
CoQuí Feature: Wannier90 interface

\+ WANNIER90 Interface



CoQuí Feature: Wannier90 interface

QUANTUMESPRESSO + WANNIER90 Interface



Tutorial 1

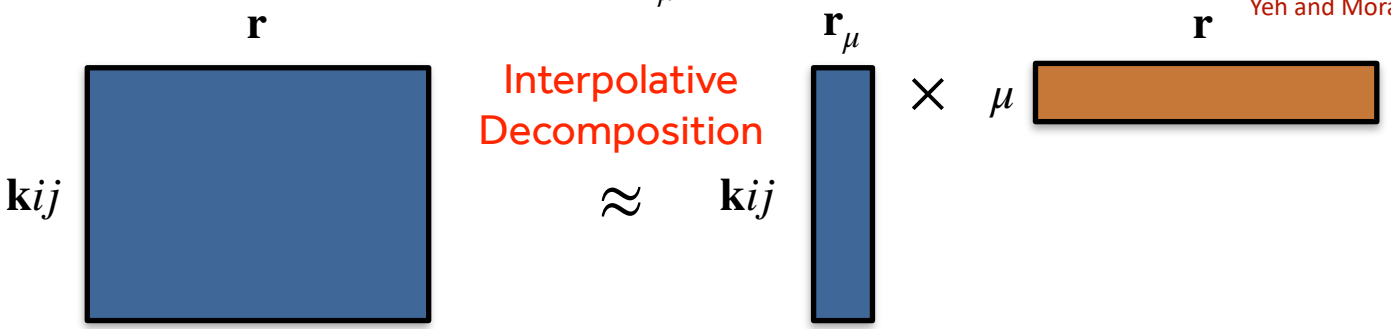
```
# construct MF from a dictionary
mf_params = {
    "prefix": "svo",
    "outdir": "./out",
}
svo_mf = coqui.make_mf(mpi, params=mf_params, mf_type="qe")

# construct MLWFs via calling Wannier90 in the library mode
w90_params = {
    "prefix": "svo",   # equivalent to wannier90's seedname
}
coqui.wannier90(mf=svo_mf, params=w90_params)
```

Interpolative Separable Density Fitting (ISDF)

Given a crystal momentum $\mathbf{q}$:

$$\phi_i^{\mathbf{k}-\mathbf{q}^*}(\mathbf{r})\phi_j^{\mathbf{k}}(\mathbf{r}) \approx \sum_{\mu}^{N_\mu} \left[\phi_i^{\mathbf{k}-\mathbf{q}^*}(\mathbf{r}_\mu)\phi_j^{\mathbf{k}}(\mathbf{r}_\mu)\right] \zeta_\mu^{\mathbf{q}}(\mathbf{r})$$



ISDF:
Lu and Ying, JCP 302, 329-335 (2015)
Lu and Ying, Ann. Math. Sci. Appl. 1, 321 (2016)
Momentum-dependent ISDF:
Yeh and Morales, JCTC 19, 6197–6207 (2023)
Symmetry-adapted ISDF:
Yeh and Morales, JCTC, 20, 8, 3184–3198 (2024)

- The column space $\{\mathbf{r}\}$ is represented by selected columns at ***the interpolating points*** $\{\mathbf{r}_\mu\}_{\mu=1}^{N_\mu}$
- $\phi_i^{\mathbf{k}-\mathbf{q}^*}(\mathbf{r})\phi_j^{\mathbf{k}}(\mathbf{r})$ *a*t arbitrary $\mathbf{r}$ is reconstructed through interpolation using the ***interpolating vectors*** $\{\zeta_\mu(\mathbf{r})\}_{\mu=1}^{N_\mu}$
- **Fully separable** in orbital and $\mathbf{k}$-point indices
- Accuracy is controlled by the size of $\{\mathbf{r}_\mu\}_{\mu=1}^{N_\mu}$, $N_\mu = \alpha N_{\text{orb}} \ll O(N_r)$

Interpolative Separable Density Fitting (ISDF)

$$\phi_i^{\mathbf{k}-\mathbf{q}^*}(\mathbf{r})\phi_j^{\mathbf{k}}(\mathbf{r}) \approx \sum_{\mu}^{N_\mu} \left[\phi_i^{\mathbf{k}-\mathbf{q}^*}(\mathbf{r}_\mu)\phi_j^{\mathbf{k}}(\mathbf{r}_\mu)\right] \zeta_\mu^{\mathbf{q}}(\mathbf{r})$$

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Lu and Ying, JCP 302, 329-335 (2015)
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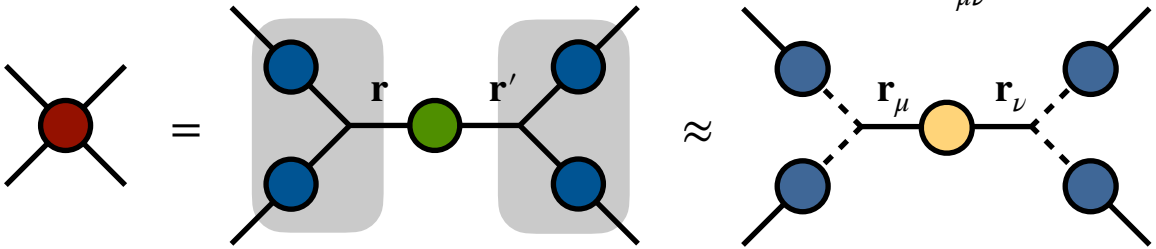
Yeh and Morales, JCTC, 20, 8, 3184–3198 (2024)

- **Tensor hypercontration (THC) representation of ERIs:** $\mathbf{q} = \mathbf{k}_i - \mathbf{k}_j = \mathbf{k}_l - \mathbf{k}_k$

$$V_{ijkl}^{\mathbf{k}_i\mathbf{k}_j\mathbf{k}_k\mathbf{k}_l} = \int d\mathbf{r} \int d\mathbf{r}' \phi_i^{\mathbf{k}_i*}(\mathbf{r})\phi_j^{\mathbf{k}_j}(\mathbf{r}) \frac{1}{|\mathbf{r}-\mathbf{r}'|} \phi_k^{\mathbf{k}_k*}(\mathbf{r}')\phi_l^{\mathbf{k}_l}(\mathbf{r}') \approx \sum_{\mu\nu}^{N_\mu} \phi_i^{\mathbf{k}_i*}(\mathbf{r}_\mu)\phi_j^{\mathbf{k}_j}(\mathbf{r}_\mu) V_{\mu\nu}^{\mathbf{q}} \phi_k^{\mathbf{k}_k*}(\mathbf{r}_\nu)\phi_l^{\mathbf{k}_l}(\mathbf{r}_\nu)$$



$$= V_{\mu\nu}^{\mathbf{q}} = \int d\mathbf{r} \int d\mathbf{r}' \zeta_\mu^{\mathbf{q}*}(\mathbf{r}) \frac{1}{|\mathbf{r}-\mathbf{r}'|} \zeta_\nu^{\mathbf{q}}(\mathbf{r}')$$

Reformulation of GW in the ISDF representation

$$\Sigma_{ij}^{\mathbf{k}}(\tau) = -\sum_{\mathbf{q}}\sum_{ab} G_{ab}^{\mathbf{k}-\mathbf{q}}(\tau) W_{\substack{i\ a\ b\ j}}^{\mathbf{k},\mathbf{k}-\mathbf{q},\mathbf{k}-\mathbf{q},\mathbf{k}}(\tau)$$

$$W_{\substack{i\ a\ b\ j}}^{\mathbf{k},\mathbf{k}-\mathbf{q},\mathbf{k}-\mathbf{q},\mathbf{k}}(\tau) \approx \sum_{\mu\nu}^{N_\mu} \phi_i^{\mathbf{k}*}(\mathbf{r}_\mu)\phi_a^{\mathbf{k}-\mathbf{q}}(\mathbf{r}_\mu) W_{\mu\nu}^{\mathbf{q}}(\tau) \phi_b^{\mathbf{k}-\mathbf{q}*}(\mathbf{r}_\nu)\phi_j^{\mathbf{k}}(\mathbf{r}_\nu)$$

Yeh and Morales, JCTC, 20, 8, 3184–3198 (2024)

$O(N_\tau N_\mu^3 N_{k_{\text{ibz}}})$ cost

w/ space-group symmetries

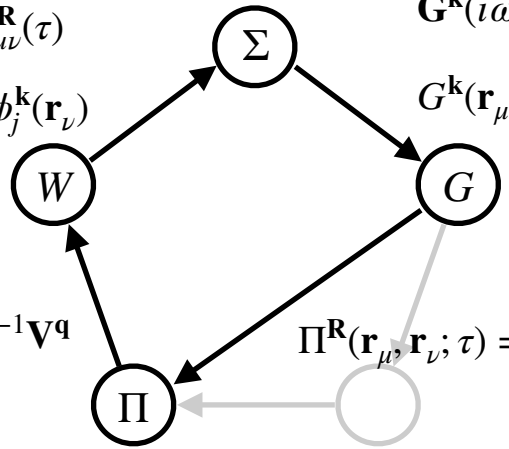
- **Fast algorithms for $\Sigma_{ij}^{\mathbf{k}}(\tau)$ and $W_{\mu\nu}^{\mathbf{q}}(\tau)$**

$$\Sigma_{\mu\nu}^{\mathbf{R}}(\tau) = -G^{\mathbf{R}}(\mathbf{r}_\mu, \mathbf{r}_\nu; \tau) W_{\mu\nu}^{\mathbf{R}}(\tau)$$

$$\Sigma_{ij}^{\mathbf{k}}(\tau) = \sum_{\mu\nu} \phi_i^{\mathbf{k}*}(\mathbf{r}_\nu)\Sigma_{\mu\nu}^{\mathbf{k}}(\tau)\phi_j^{\mathbf{k}}(\mathbf{r}_\nu)$$

$$\mathbf{G}^{\mathbf{k}}(i\omega_n) = [\mathbf{G}_0^{-1}(i\omega_n) - \mathbf{\Sigma}(i\omega_n)]^{-1}$$

$$G^{\mathbf{k}}(\mathbf{r}_\mu, \mathbf{r}_\nu; \tau) = \sum_{ij} \phi_i^{\mathbf{k}}(\mathbf{r}_\mu) G_{ij}^{\mathbf{k}}(\tau) \phi_j^{\mathbf{k}*}(\mathbf{r}_\nu)$$



$$\mathbf{W}^{\mathbf{q}}(i\Omega_n) = [\mathbf{I} - \mathbf{V}^{\mathbf{q}}\mathbf{\Pi}^{\mathbf{q}}(i\Omega_n)]^{-1}\mathbf{V}^{\mathbf{q}}$$

$$\Pi^{\mathbf{R}}(\mathbf{r}_\mu, \mathbf{r}_\nu; \tau) = G^{\mathbf{R}}(\mathbf{r}_\mu, \mathbf{r}_\nu; \tau) G^{-\mathbf{R}}(\mathbf{r}_\nu, \mathbf{r}_\mu; -\tau)$$

Compact Representation of Correlation Functions

Cf. Day 1, MB refresher & TRIQS basic tutorial

Spectral representation of correlation functions:

☹ Analytical continuation issue

$$G(\tau) = \int_{-\omega_{max}}^{\omega_{max}} d\omega \left[\frac{e^{-\tau\omega}}{e^{-\beta\omega} \pm 1}\right] \rho(\omega) = \int_{-\omega_{max}}^{\omega_{max}} d\omega K(\tau, \omega)\rho(\omega) \approx \sum_{l=1}^{r} K(\tau, \omega_l) g_l$$

🙂

Key insight:

ill-defined nature of AC = low-rank structure of $K(\tau, \omega)$

Different low-rank approx. to $K(\tau, \omega)$:

- SVD in CoQuí
- Interpolative decomposition in TRIQS

Despite the choice of low-rank approx. to $K(\tau, \omega)$, the controlled parameters are always:

1. ω_{max} = range of the integrals
2. ϵ = accuracy for the low-rank approx.

Compact Representation of Correlation Functions

Cf. Day 1, MB refresher & TRIQS basic tutorial

Spectral representation of correlation functions:

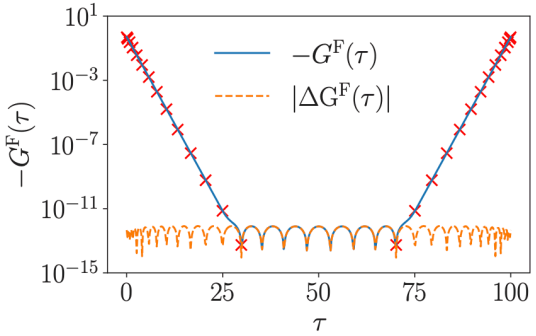
☹ Analytical continuation issue

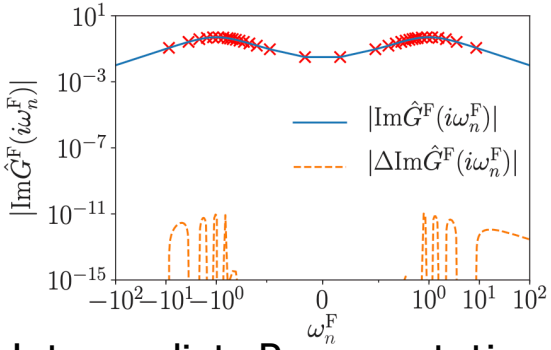
$$G(\tau) = \int_{-\omega_{max}}^{\omega_{max}} d\omega \left[\frac{e^{-\tau\omega}}{e^{-\beta\omega} \pm 1}\right] \rho(\omega) = \int_{-\omega_{max}}^{\omega_{max}} d\omega K(\tau,\omega)\rho(\omega) \approx \sum_{l=1}^{r} K(\tau,\omega_l) g_l$$

🙂

Key insight:

ill-defined nature of AC = low-rank structure of $K(\tau,\omega)$

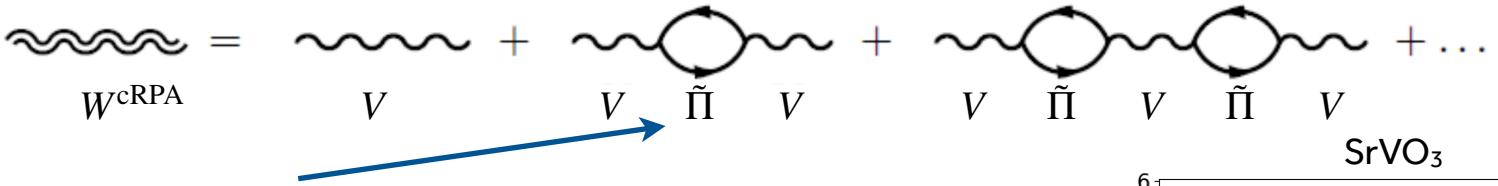




τ and iω_n samplings from the Intermediate Representation

Constrained RPA (cRPA)

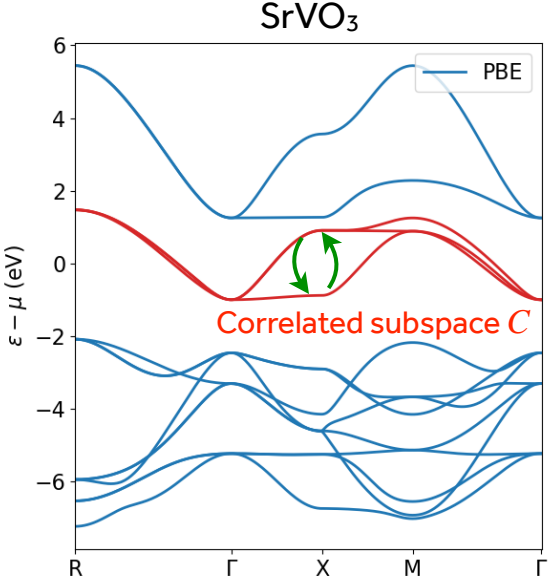
- Effective screened interactions for the correlated subspace C from first principles:



Modified non-interacting polarizability:

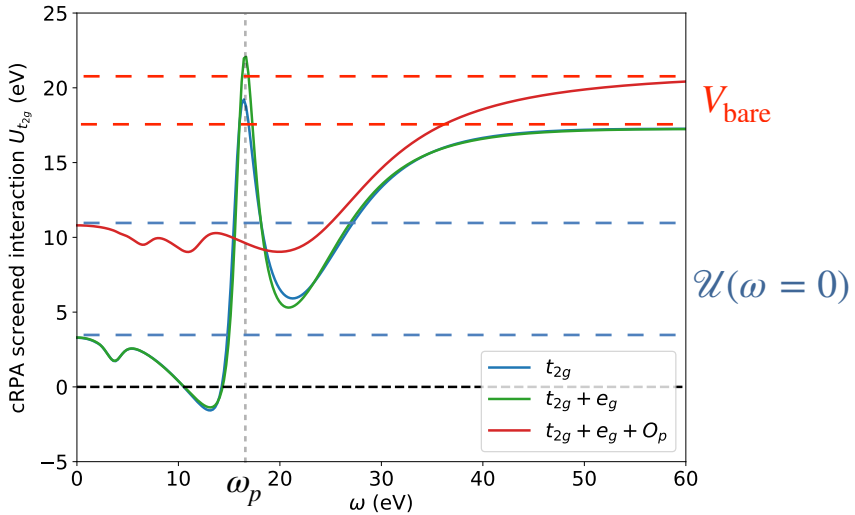
$$\tilde{\Pi}(\mathbf{r},\mathbf{r}';\omega) = \sum_{n}^{\mathrm{occ}'}\sum_{m}^{\mathrm{unocc}'}\left\{\frac{\phi_n(\mathbf{r}')\phi_n^*(\mathbf{r})\phi_m(\mathbf{r})\phi_m^*(\mathbf{r}')}{\omega-\epsilon_m+\epsilon_n+i\delta} - \frac{\phi_n(\mathbf{r})\phi_n^*(\mathbf{r}')\phi_m(\mathbf{r}')\phi_m^*(\mathbf{r})}{\omega+\epsilon_m-\epsilon_n-i\delta}\right\}$$

$$\sum_{n}^{\mathrm{occ}'}\sum_{m}^{\mathrm{unocc}'} = \sum_{n}^{\mathrm{occ}}\sum_{m}^{\mathrm{unocc}} - \sum_{n\in C}^{\mathrm{occ}}\sum_{m\in C}^{\mathrm{unocc}}$$



Constrained RPA (cRPA)

- **Dynamical screened interactions for $SrVO_3$ V t_{2g} orbitals**

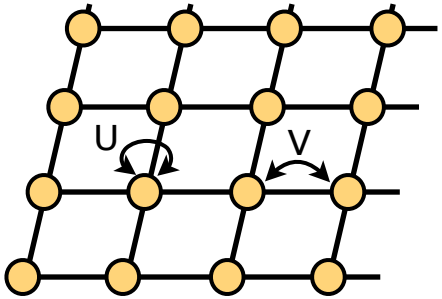


- Frequency dependence ↔ Couplings of bosonic degrees of freedom e.g. particle-hole excitations and plasmons
- Characteristic energy scale ~ 15 eV at the **plasmonic excitation**
- **Two-step renormalizations:**
  1. Renormalization through static $\mathcal{U}$
  2. Further renormalized by the screening bosons.
- The effective static $\mathcal{U}$ needs to be **slightly larger than $\mathcal{U}$ (w=0)** for correct mass renormalization.
  → However, this sometimes put satellites at incorrect positions!

Extended Dynamical Mean-Field Theory (EDMFT)

- Originally for Hubbard model with **inter-site interaction**

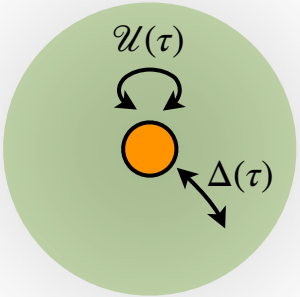
  Sengupta and Georges, PRB 52, 10 295 (1995)
  Si and Smith, PRL 77, 3391 (1996)



EDMFT "local" approximation

$$\Sigma^{\mathbf{k}} = \Sigma^{\mathrm{imp}}$$
$$\Pi^{\mathbf{q}} = \Pi^{\mathrm{imp}}$$

- Impurity problem with **dynamic interactions**



$\mathcal{U}(\tau)$: Local dynamic screened interactions

$\Delta(\tau)$: Embedded in an effective bath

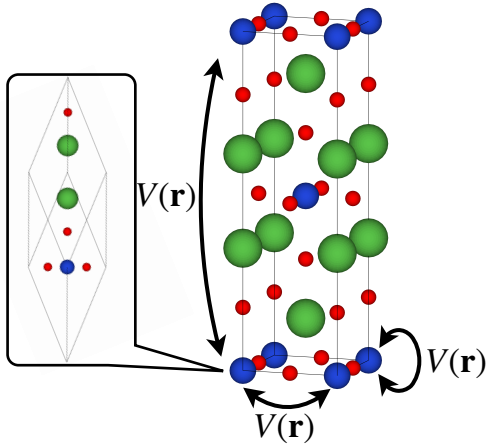
- $\mathcal{U}(\tau)$ induced by screening from the inter-site interaction V
  [Similar to inter-site hopping "t" induces $\Delta(\tau)$]

- Δ and $\mathcal{U}$ are determined **self-consistently** to mimic the lattice environment.

$$G_{\mathrm{loc}} = G_{\mathrm{imp}} = [\mathcal{G}[\Delta]^{-1} - \Sigma_{\mathrm{imp}}]^{-1}$$
$$W_{\mathrm{loc}} = W_{\mathrm{imp}} = [\mathcal{U}^{-1} - \Pi_{\mathrm{imp}}]^{-1}$$

- CT-HYB w/ retarded interaction $\mathcal{U}(\tau)$: Werner and Millis, PRL 99, 146404 (2007)

Extended Dynamical Mean-Field Theory (EDMFT)

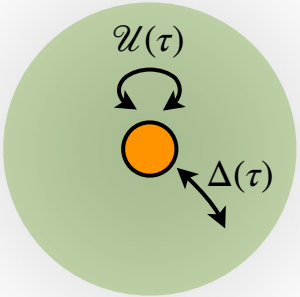


- Impurity problem with **dynamic interactions**

EDMFT "local" approximation

?

$$\mathbf{\Sigma}^{\mathbf{k}} = \mathbf{\Sigma}^{\mathrm{imp}}$$
$$\mathbf{\Pi}^{\mathbf{q}} = \mathbf{\Pi}^{\mathrm{imp}}$$



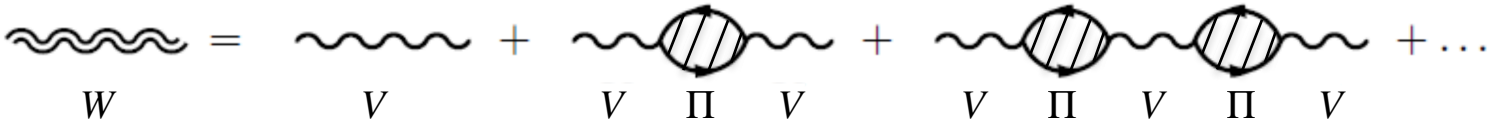
$\mathcal{U}(\tau)$: Local dynamic screened interactions

$\Delta(\tau)$: Embedded in an effective bath

- Only a small subset of electronic degrees of freedom fits into the local EDMFT ansatz
- Non-trivial screening processes:
  a. **Long-range Coulomb interaction** $V(\mathbf{r}) = \dfrac{1}{\mathbf{r}}$
  b. Renormalization from **all-electron** degrees of freedom

Ab initio Many-Body Simulations: Numerical Challenges

Computing and storing *ab initio* self-consistent screened interactions is challenging:



$$W^{\mathbf{k}_1\mathbf{k}_2\mathbf{k}_3\mathbf{k}_4}_{ijkl}(i\omega) = \iint \mathrm{d}\mathbf{r}\mathrm{d}\mathbf{r}'\phi_i^{\mathbf{k}_1*}(\mathbf{r})\phi_j^{\mathbf{k}_2}(\mathbf{r})W(\mathbf{r},\mathbf{r}';i\omega)\phi_k^{\mathbf{k}_3*}(\mathbf{r}')\phi_l^{\mathbf{k}_4}(\mathbf{r}') \quad \sim O(N_\omega N_k^3 N_{\mathrm{orb}}^4)$$

V: bare interactions

Π: polarizability

- Goes beyond Lindblad form of Π_0: $G \approx G_0$

$$\tilde{\Pi}(\mathbf{r},\mathbf{r}';\omega) = \sum_n^{\mathrm{occ}} \sum_m^{\mathrm{unocc}} \left\{ \frac{\phi_n(\mathbf{r}')\phi_n^*(\mathbf{r})\phi_m(\mathbf{r})\phi_m^*(\mathbf{r}')}{\omega - \epsilon_m + \epsilon_n + i\delta} - \frac{\phi_n(\mathbf{r})\phi_n^*(\mathbf{r}')\phi_m(\mathbf{r}')\phi_m^*(\mathbf{r})}{\omega + \epsilon_m - \epsilon_n - i\delta} \right\}$$

$$\Pi = G * G$$

GW+EDMFT — Functional derivations

S. Biermann *et al.*, Phys. Rev. Lett. 90, 086402 (2003)
P. Sun *et al.*, Phys. Rev. B 66, 085120 (2002)

The Almbladh variational free-energy functional of an interacting system:

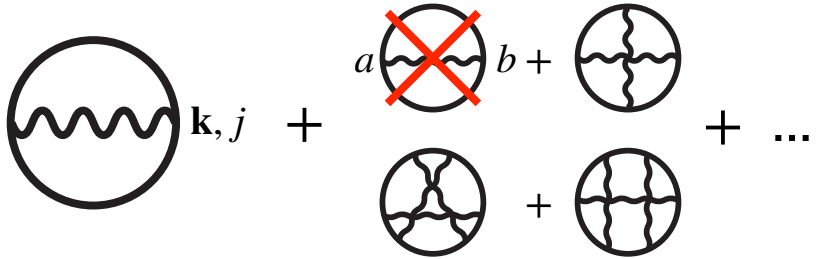
$$F[G,W] = \mathrm{Tr}[\ln G] - \mathrm{Tr}[(G_H^{-1} - G^{-1})G] - \frac{1}{2}\mathrm{Tr}[\ln W] + \frac{1}{2}\mathrm{Tr}[(V^{-1} - W^{-1})W] + \Psi[G,W]$$

- Stationary condition: $\Sigma = \frac{\delta\Psi}{\delta G} = G_H^{-1} - G^{-1}$ ***Sum of all one-particle irreducible diagrams***

$$\Pi[G,W] = -2\frac{\delta\Psi}{\delta W} = V^{-1} - W^{-1}$$ ***Sum of all two-particle irreducible diagrams***

"Self-consistent solution (G, Σ, Π, W) of skeleton diagrams" = Stationary solution

double counting is rigorously defined

$\Psi \approx \Psi^{GW+\mathrm{EDMFT}}[G,W] =$ 

GW+EDMFT — Functional derivations

S. Biermann *et al.*, Phys. Rev. Lett. 90, 086402 (2003)
P. Sun *et al.*, Phys. Rev. B 66, 085120 (2002)

The Almbladh variational free-energy functional of an interacting system:

$$F[G,W] = \mathrm{Tr}[\ln G] - \mathrm{Tr}[(G_H^{-1} - G^{-1})G] - \frac{1}{2}\mathrm{Tr}[\ln W] + \frac{1}{2}\mathrm{Tr}[(V^{-1} - W^{-1})W] + \Psi[G,W]$$

- Stationary condition: $\Sigma = \frac{\delta\Psi}{\delta G} = G_H^{-1} - G^{-1}$ ***Sum of all one-particle irreducible diagrams***

$$\Pi[G,W] = -2\frac{\delta\Psi}{\delta W} = V^{-1} - W^{-1}$$ ***Sum of all two-particle irreducible diagrams***

"Self-consistent solution (G, Σ, Π, W) of skeleton

dou

$\Psi \approx \Psi^{GW+\mathrm{EDMFT}}[G,W] =$ $(\mathbf{k}, i)$  $\mathbf{k}, j$

Many-body analog of how DFT approximates the Hohenberg-Kohn energy functional

$$E^{\mathrm{KS}}[\rho(\mathbf{r})] = T[\rho(\mathbf{r})] + E_{\mathrm{ext}}[\rho(\mathbf{r})] + E_H[\rho(\mathbf{r})] + E_{\mathrm{xc}}[\rho(\mathbf{r})]$$

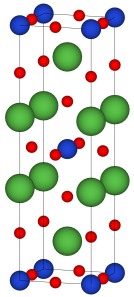
Ab initio GW+EDMFT

Three nested self-consistency loops in different energy scales

TRIQS/CT-SEG
TRIQS/solid_dmft

CoQuí — Complete KS orbitals

TRIQS — MLWFs



DFT + Wannier90

$\{\epsilon_i^{\mathbf{k}}, \phi_i^{\mathbf{k}}\}$ and projector $C_{\alpha i}^{\mathbf{k}}$

$G^{\mathbf{k}} = [G_0^{\mathbf{k},-1} - \Sigma^{\mathbf{k}}]^{-1}$
$W^{\mathbf{q}} = [V^{\mathbf{q},-1} - \Pi^{\mathbf{q}}]^{-1}$

downfolding

$G^{\mathrm{loc}} = \sum_{\mathbf{k}} C^{\mathbf{k}\dagger} G^{\mathbf{k}} C^{\mathbf{k}}$
$W^{\mathrm{loc}} = \sum_{\mathbf{q}} B^{\mathbf{q}\dagger} W^{\mathbf{q}} B^{\mathbf{q}}$

Lattice GW

$(\Sigma^{GW})^{\mathbf{k}}[G^{\mathbf{k}}, W^{\mathbf{q}}]$
$(\Pi^{GW})^{\mathbf{q}}[G^{\mathbf{k}}, W^{\mathbf{q}}]$

Weiss fields

$\mathcal{G} = [G_{\mathrm{loc}}^{-1} + \Sigma^{\mathrm{imp}}]^{-1}$
$\mathcal{U} = [W_{\mathrm{loc}}^{-1} + \Pi^{\mathrm{imp}}]^{-1}$

Impurity solver

$\Sigma^{\mathrm{imp}}[\mathcal{G}, \mathcal{U}]$
$\Pi^{\mathrm{imp}}[\mathcal{G}, \mathcal{U}]$

Double counting

$\Sigma^{\mathrm{DC}}[G^{\mathrm{loc}}, W^{\mathrm{loc}}]$
$\Pi^{\mathrm{DC}}[G^{\mathrm{loc}}, W^{\mathrm{loc}}]$

Upfolding

$\Sigma^{\mathbf{k}} = (\Sigma^{GW})^{\mathbf{k}} + C^{\mathbf{k}}[\Sigma^{\mathrm{imp}} - \Sigma^{\mathrm{DC}}]C^{\mathbf{k}\dagger}$
$\Pi^{\mathbf{q}} = (\Pi^{GW})^{\mathbf{q}} + B^{\mathbf{q}}[\Pi^{\mathrm{imp}} - \Pi^{\mathrm{DC}}]B^{\mathbf{q}\dagger}$

GW+EDMFT approximation

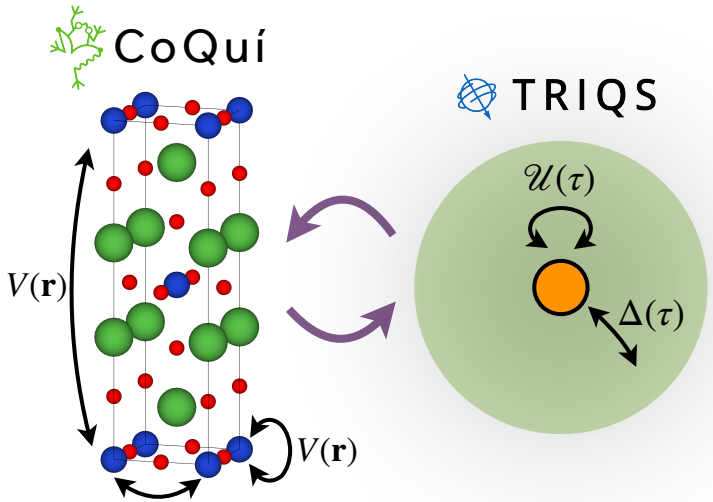
***Ab initio* GW+EDMFT is now available via the integration between CoQuí and TRIQS!**

GW+EDMFT addresses several limitations of LDA+DMFT:

a. Self-consistently determines impurity interactions
b. Well-defined double counting
c. Captures long-range correlations at the RPA level

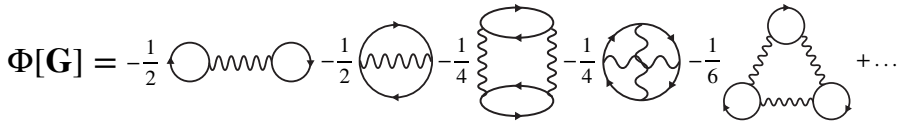
Screening depends sensitively on

a. The reference state (e.g. Metallic v.s. insulating starting point)
b. The orbital character where the local vertex correction $\Delta\Pi_{GW+\mathrm{EDMFT}}$
   - enhances screening for orbitals far from half filling
   - suppresses screening for orbitals near half filling



Many-Body Perturbation Theory

- **Luttinger-Ward functional for free energy**



$$\Sigma = \frac{\delta \Phi[\mathbf{G}]}{\delta \mathbf{G}}$$

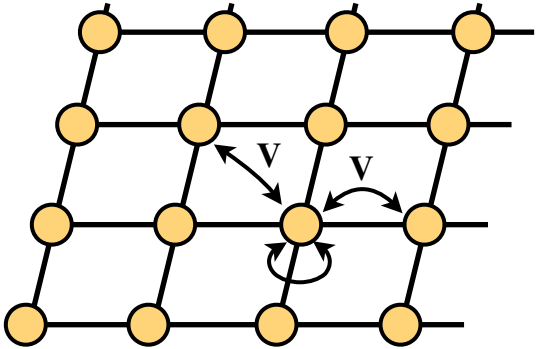
Baym and Kadanoff, Phys. Rev. 124, 287 (1961)
Baym, Phys. Rev. 127, 1391 (1962)

Dyson equation: $\mathbf{G}^{-1} = \mathbf{G}_0^{-1} - \boldsymbol{\Sigma}$

$\mathbf{G}/\mathbf{G}_0$: interacting/non-interacting Green's function

$E_{\mathrm{xc}}[\rho] \leftrightarrow \Phi[G]$: **many-body correlations**

: unit cell (multi-orbitals)



$E_{\mathrm{xc}}[\rho]$: LDA, PBE, etc

$$v_{\mathrm{xc}}(\mathbf{r}) = \frac{\delta E_{\mathrm{xc}}[\rho]}{\delta \rho}$$

$$\rho(\mathbf{r}) = \sum_i^{N_e} |\phi_i(\mathbf{r})|^2$$

Hohenberg and Kohn, Phys. Rev. 136, B864 (1964).
Kohn and Sham, Phys. Rev. 140, A1133 (1965).

$$\left[-\frac{1}{2}\nabla^2 + v_{\mathrm{ext}}(\mathbf{r}) + v_{\mathrm{H}}(\mathbf{r}) + v_{\mathrm{xc}}(\mathbf{r})\right]\phi_i(\mathbf{r}) = \epsilon_i \phi_i(\mathbf{r})$$