Aalto University
School of Science

GW precision: comparison between different software

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Towards exascale solutions in Green function methods and advanced DFT

Paphos, Cyprus, October 3-8, 2023

Participants

Novel Materials Discovery - Center of Excellence (NOMAD-CoE)

Work Package 2: Exascale Green-Function-Based Methods

- Abinit: Maryam Azizi, Mateo Giantomassi, Xavier Gonze
- Exciting: Alexander Bucheri, Claudia Draxl, Andris Gulans, Davis Zavickis
- FHI-aims: Francisco Delesma, Dorothea Golze, Patrick Rinke
- GPAW: Mikael Kuisma, Kristian Thyghesen

Outline

Outline

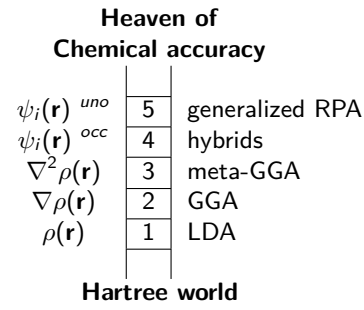
To be considered

With respect to the computer-generated data

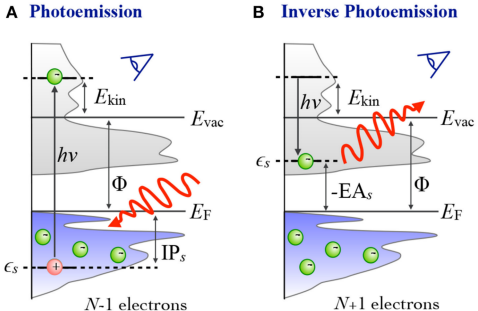
- How much can we rely on it?
- How to evaluate it?
- How to improve it?

In practice

- Validation/Accuracy
  -Comparison with high-level of theory
  -Comparison with experiment
- Verification/Precision
  - Comparison between other implementations

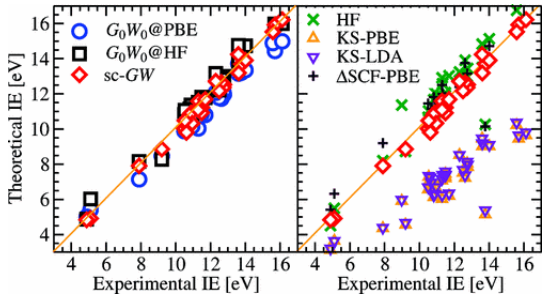


Theoretical Spectroscopy (The success of the GW approach)



D. Golze et. al., Front. Chem. 7, 377 (2019)

Ionization energies of molecular sytems



F. Caruso, et. al., Phys. Rev. B 86, 081102 (2012)

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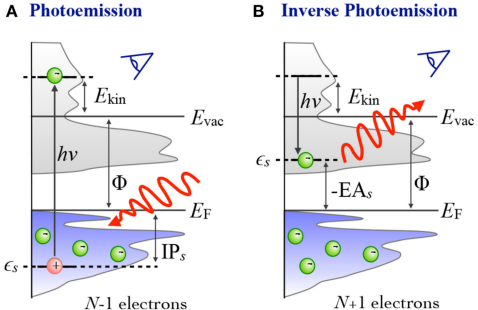
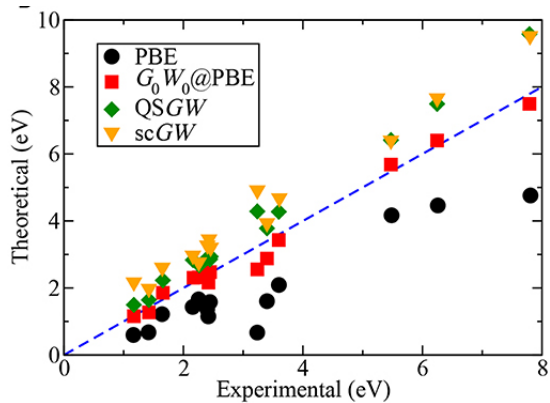


D. Golze et. al., Front. Chem. 7, 377 (2019)

Band-gap for periodic systems



M. Grumet, et al. Phys. Rev. B 98, 155143 (2018).

G_0W_0 approach

Solve iteratively the quasi-particle equation (QPE)

$$\epsilon_{n\mathbf{k}}^{QP} = \epsilon_{n\mathbf{k}}^{KS} + \langle \psi_n^{\mathbf{k}} | \Sigma(\epsilon_{n\mathbf{k}}^{QP}) - v_{xc}^{KS} | \psi_n^{\mathbf{k}} \rangle$$

or by the linealized QPE

$$\epsilon_{n\mathbf{k}}^{QP} = \epsilon_{n\mathbf{k}}^{KS} + Z_{n\mathbf{k}} \langle \psi_n^{\mathbf{k}} | \Sigma(\epsilon_{n\mathbf{k}}^{QP}) - v_{xc}^{KS} | \psi_n^{\mathbf{k}} \rangle$$

$$Z_{n\mathbf{k}} = \left[1 - \frac{d}{d\omega} \langle \psi_n^{\mathbf{k}} | \Sigma(\omega) | \psi_n^{\mathbf{k}} \rangle \Big|_{\omega = \epsilon_{n\mathbf{k}}^{KS}} \right]^{-1}$$

The self-energy

$$\Sigma(\omega) = \frac{\mathrm{i}}{2\pi} \int d\omega G(\omega + \omega') W(\omega') e^{\mathrm{i}\omega\eta}$$

Σ is analytically continued from the imaginary to real frequency axis

Previous works (representatives)

GW100: Benchmarking G_0W_0 for Molecular Systems
M. Van Setten et al, J. Chem. Theory Comput. 11, 5665−5687 (2015)
Software: FHI-aims, Turbomole, Berkeley-GW
All-electron codes agree withing 1.0 meV range for QP-HOMO and QP-LUMO
Selection of the basis set, analytical continuation

Reproducibility in G_0W_0 Calculations for Solids
T. Rangel et al. Computer Phys. Commun. 255, 107242 (2020)
Softwares: Abinit, Yambo, Berkeley-GW
Method: pseudopotential + plane waves
Systems: Au, Si, TiO_2, ZnO
For Si, TiO_2 all codes agree within 50 meV

Motivation

Provide G_0W_0 calculations for solids with different software

To know the state-of-art of the canonical G_0W_0 implementations

Impulse for the development of low-scaling GW algorithms
Please follow Thursday morning's session.

Towards exascale calculations of Green-function-based methods
Aim of this school

Demonstrator Codes

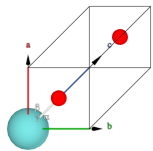
Demonstrator Codes

Characteristics of the codes

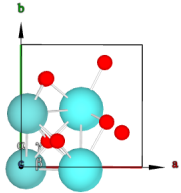
Abinit	Pseudopotentials-planewaves (PW) + projector augmented waves (PAW)
exciting	Linearized augmented planewave + local-orbital (LAPW + lo)
FHI-aims	Numeric atom-centered orbitals (NAOs)
GPAW	Projector-augmented wave (PAW)

Systems

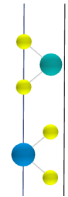
ZrO_2



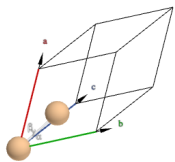
$Zr_2Y_2O_7$



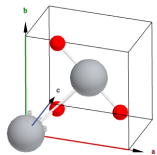
$MoWS_4$



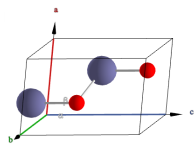
Si



TiO_2

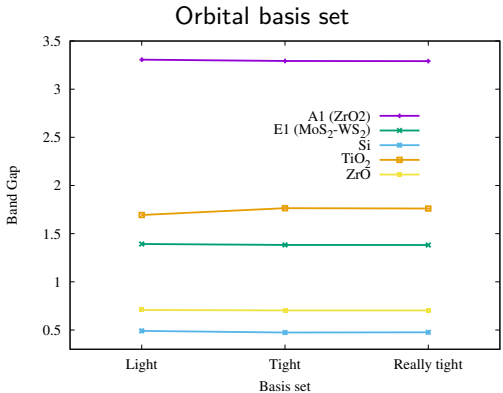


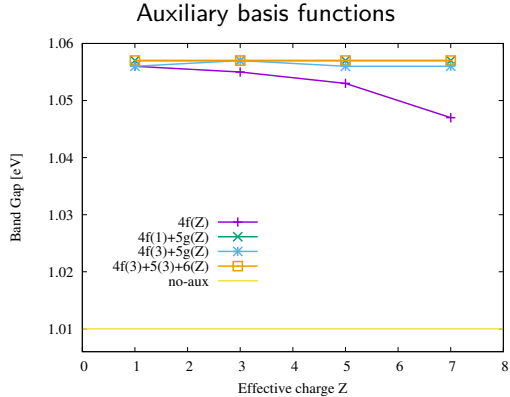
ZnO



Outline

Basis set quality (FHI-aims)





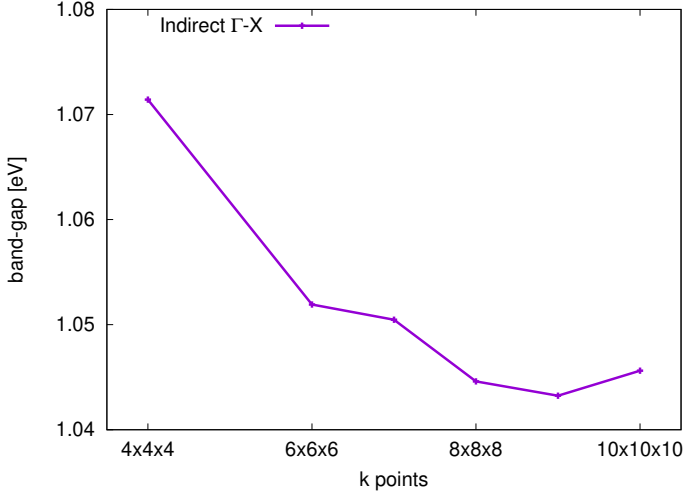
In other codes:

-Choose the pseudo potential, plane-wave cutoff

-Converge the LAPW+localized orbitals

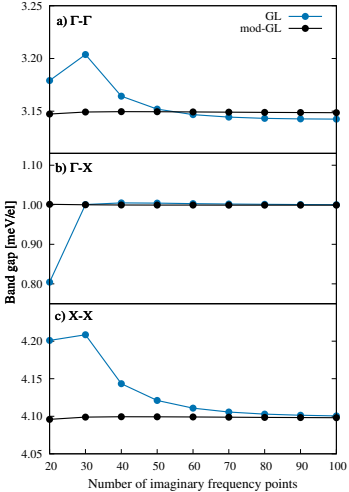
-SCF convergence

k-point convergence (FHI-aims)

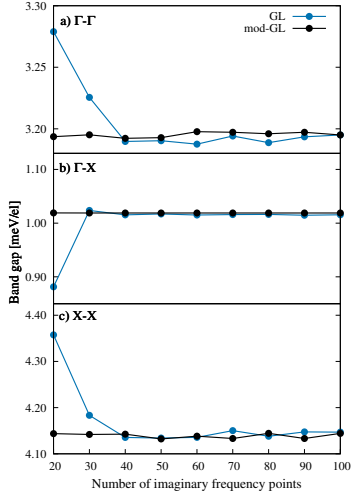


Analytic continuation (FHI-aims)

2-pole model



Pade model



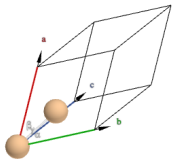
Outline

Starting point: PBEsol (Kohn–Sham band-gap)

System	k-point	Abinit	exciting	FHI-aims	GPAW
ZrO_3 (A1)	6x6x6	3.320	3.321	3.309	3.32
$Zr_2Y_2O_7$ (C1)	4x4x4	2.769	2.754	2.742	2.75
$MoWS_4$ (E1)	12x12x1	1.215		1.212	1.25
Si	8x8x8	0.460	0.482	0.459	0.51
TiO_2	6x6x10	1.830	1.809	1.794	1.74
ZnO	8x8x5	0.763	0.714	0.713	0.67

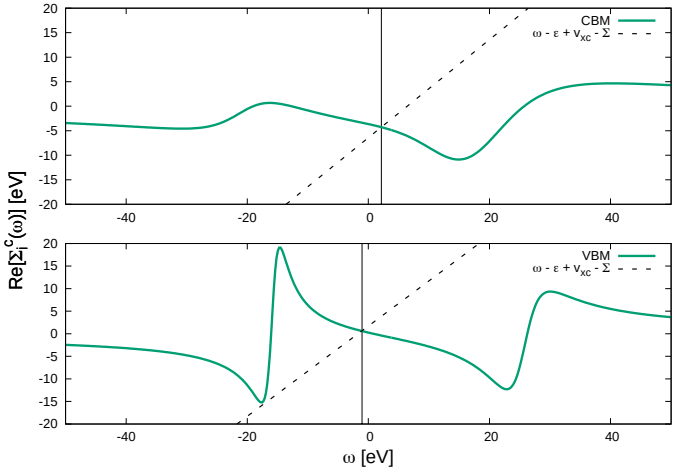
Real part of the self-energy

Silicon



xc	PBEsol
k-points	8x8x8
grid	Gauss-Legendre
ω	60
ω_{max}	10 Ha
AC	Pade (16)

Abinit



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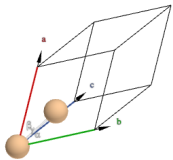

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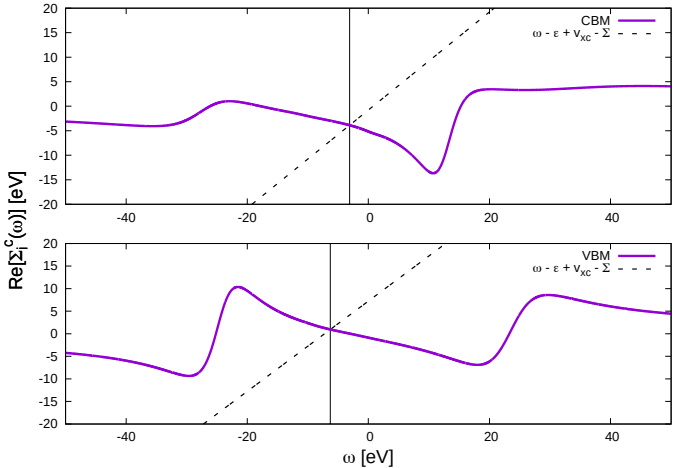
FHI-aims



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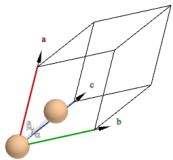

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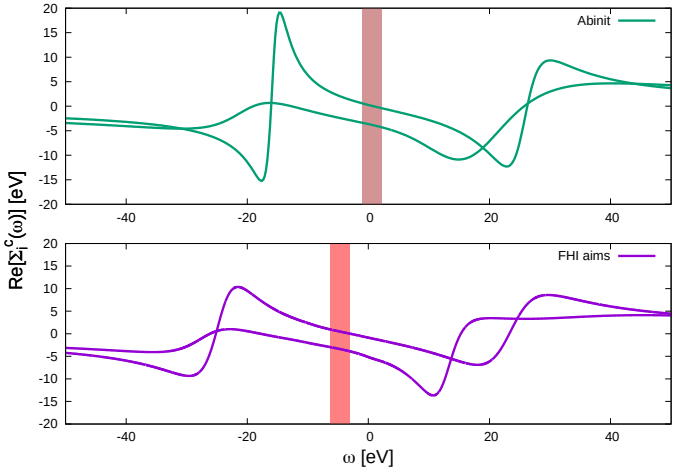
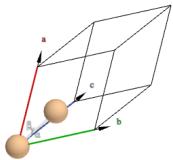
Abinit and FHI-aims



Real part of the self-energy

Silicon



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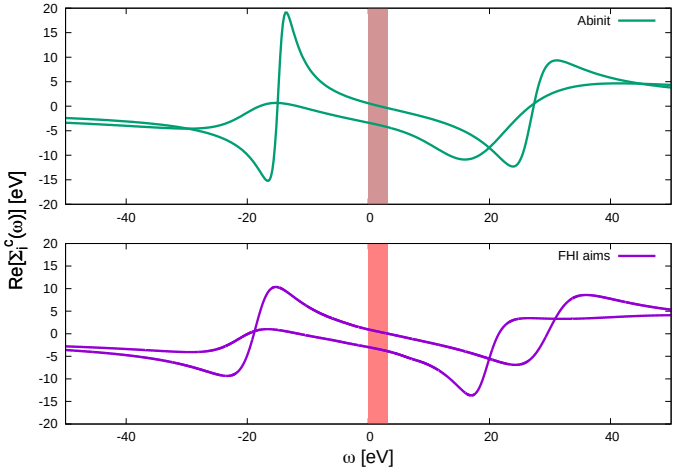
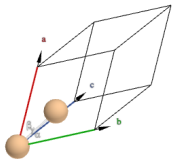


Real part of the self-energy

Silicon



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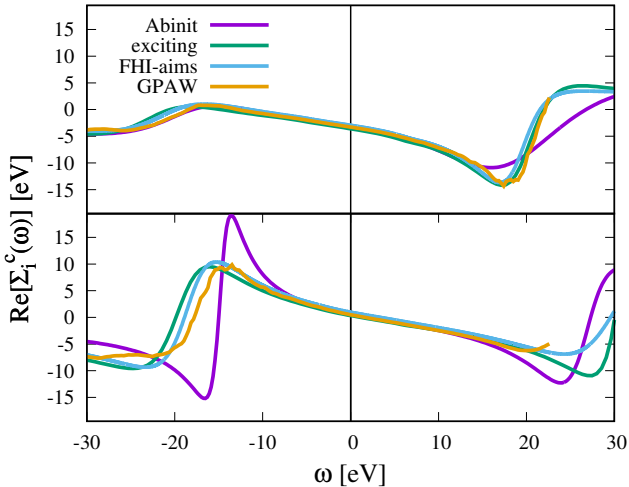


GPAW uses full-frequency

G_0W_0 band-gap

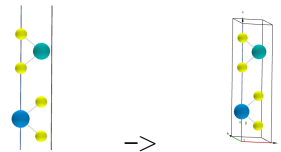
System	k-point	Abinit	exciting	FHI-aims	GPAW
ZrO_3 (A1)	6x6x6	5.144	5.319	5.366	5.41
$Zr_2Y_2O_7$ (C1)	4x4x4	4.136	4.346	4.127	4.42
Si	8x8x8	1.164	1.252	1.064	1.25
TiO_2	6x6x10	3.653	3.364	3.347	3.59
ZnO	8x8x5	2.764	2.834	2.431	2.46

MaxAD 0.4

Band-gap extrapolation $\Delta E(E_c^{\chi}) = \Delta E_g(\infty) + B_3 E_c^{\chi(-3/2)} + B_5 E_c^{\chi(-5/2)}$

Problems to be solved

- k-point sampling
- complete basis set limit
  - LAPW: adding high-energy localized orbitals improves ZnO
    (D. Nabok et al, Phys. Rev. B 94, 035118 (2016)).
  - NAOs: adding STOs (X. Ren et al. Phys. Rev. Mat. 5, 013807 (2021))
    e.g. Si (1.13 eV) and ZnO (2.70 eV) –> (20x expensive)
- Γ-point singularity ($q = 0$)
  - Bilayer $MoWS_4$



- **Computational cost**

Outline

Preliminary conclusions

We must take into account

- The quality of the basis set (Auxiliary basis)
- Number of imaginary frequency points
- k-point sampling
- Analytic continuation

Kohn-Sham band-gaps agree within 0.1 eV

G_0W_0 band-gaps agree within 0.4 eV

Acknowledgements

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