LAPW FOR LOW-DIMENSIONAL MATERIALS

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Dresden, DPG2026

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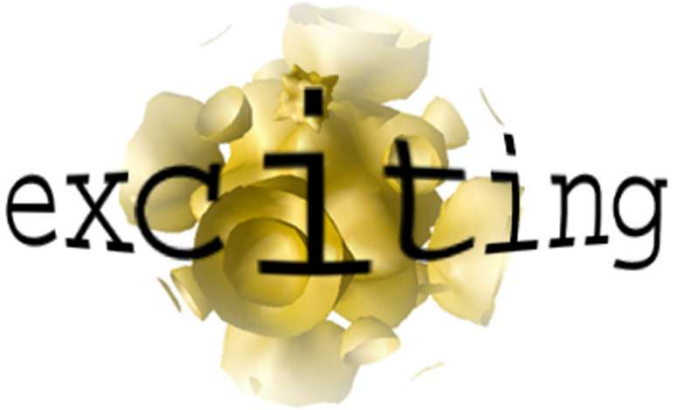
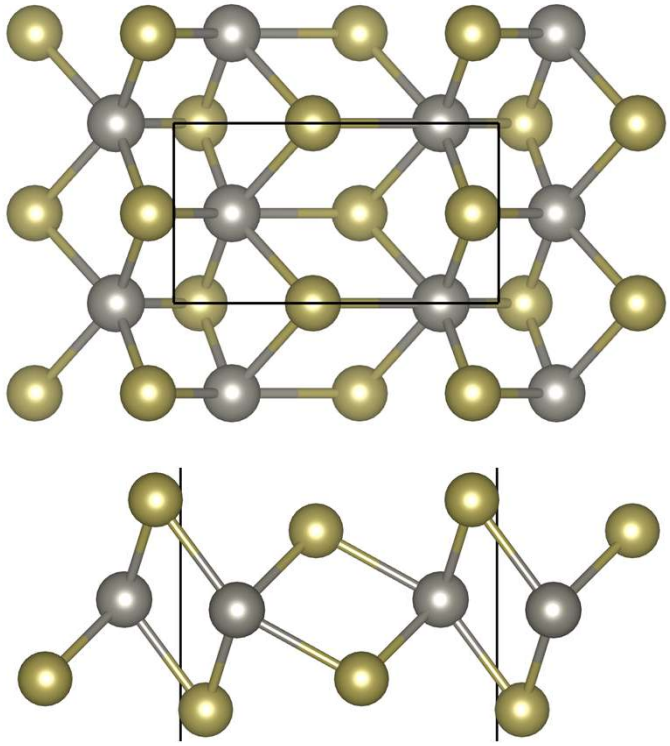
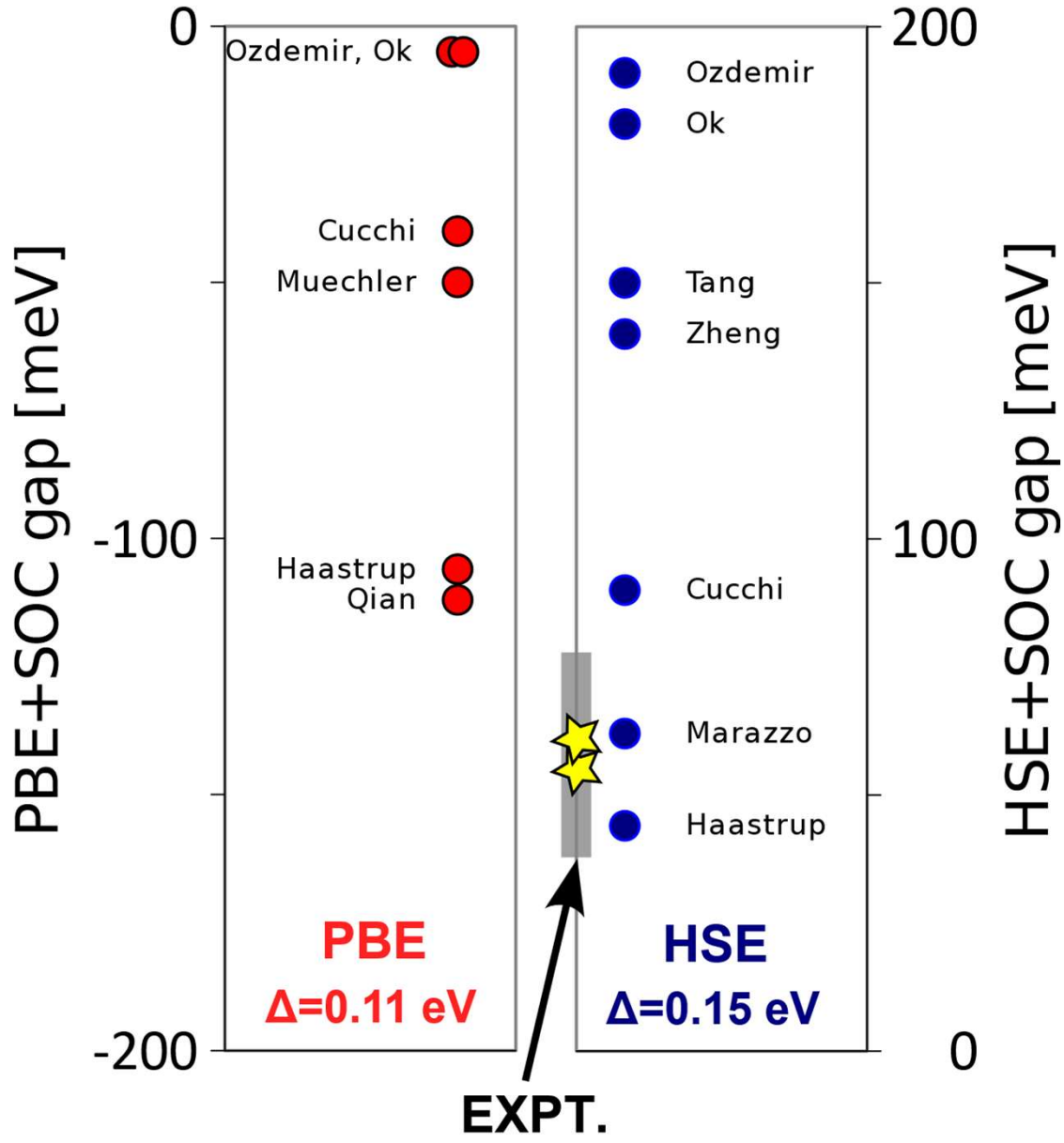


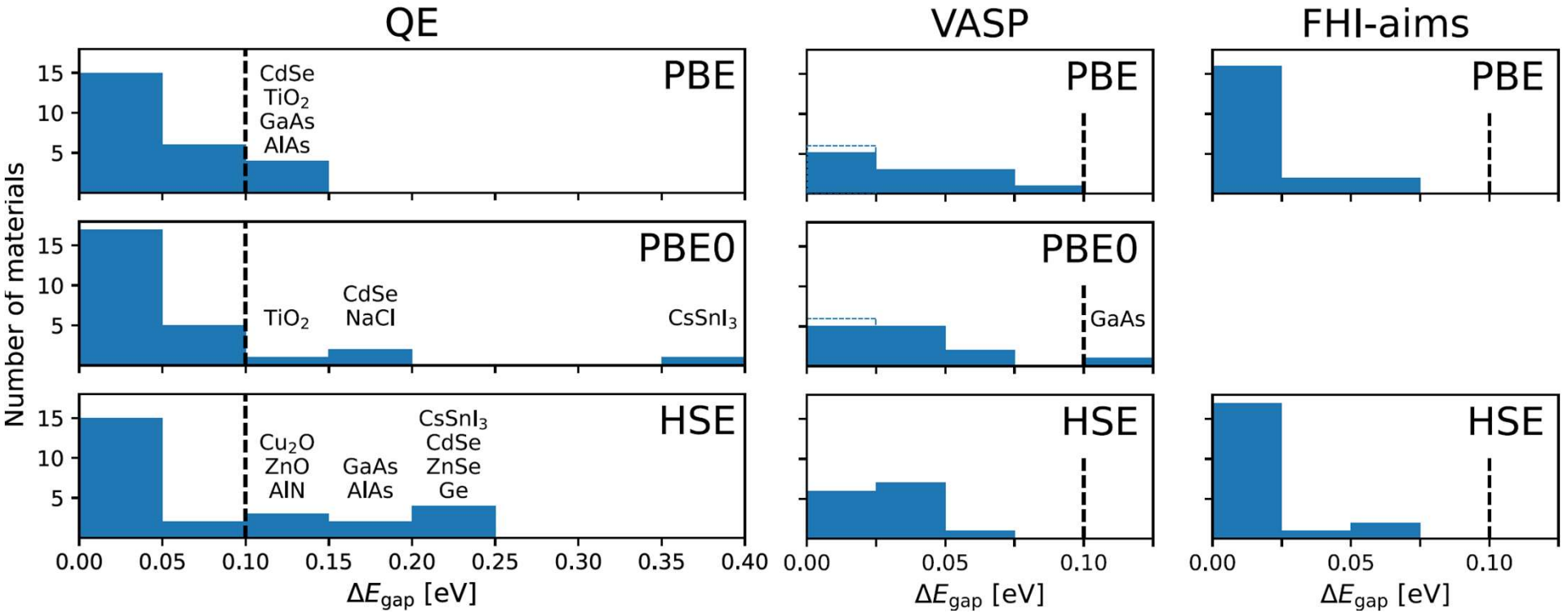
Motivation

Single layer 1T´-WTe_2





More examples: 3D materials



J. Užulis, A. Sorokin, A. Gulans, npj Computational Materials 11, 257 (2025)

LAPW as reference for 2D materials

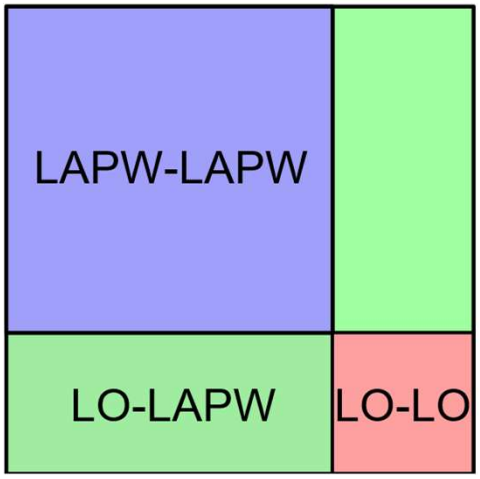
Challenges with standard tools

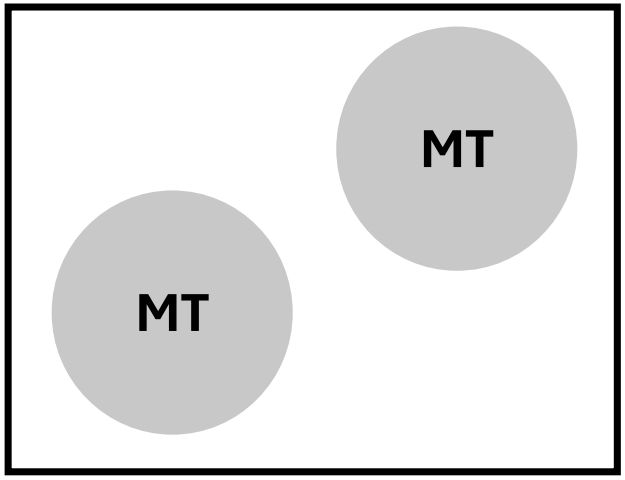
- Thick vacuum layer – ***slow* and memory-hungry eigensolvers**, with $O(V^3)$ and $O(V^2)$ time- and memory-complexities, respectively
- Hybrid functionals $O(V^2 N_{\mathrm{at}}^2)$ – even **slower than eigensolvers** and dependence on empty bands
- **Coulomb singularity** in hybrid functionals if you want to go beyond HSE functionals

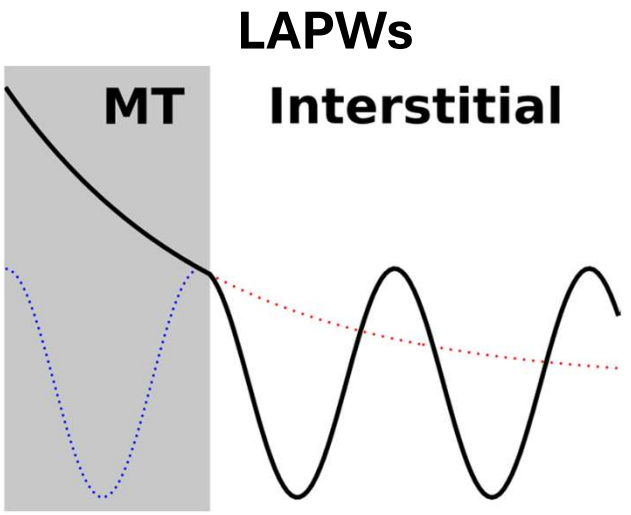
$$E_{xc} = aE_x^{HF,SR}(\omega) + (1-a)E_x^{PBE,SR}(\omega) + E_x^{HF,LR}(\omega) + E_c^{PBE}$$

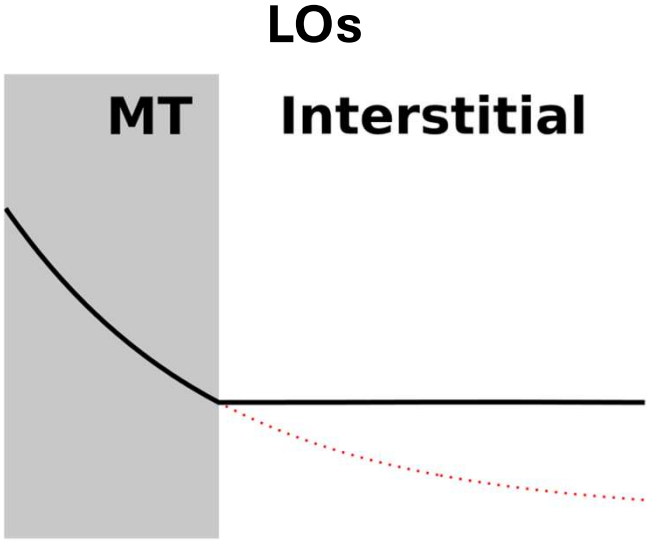
Eigensolver

$$H\psi = ES\psi$$



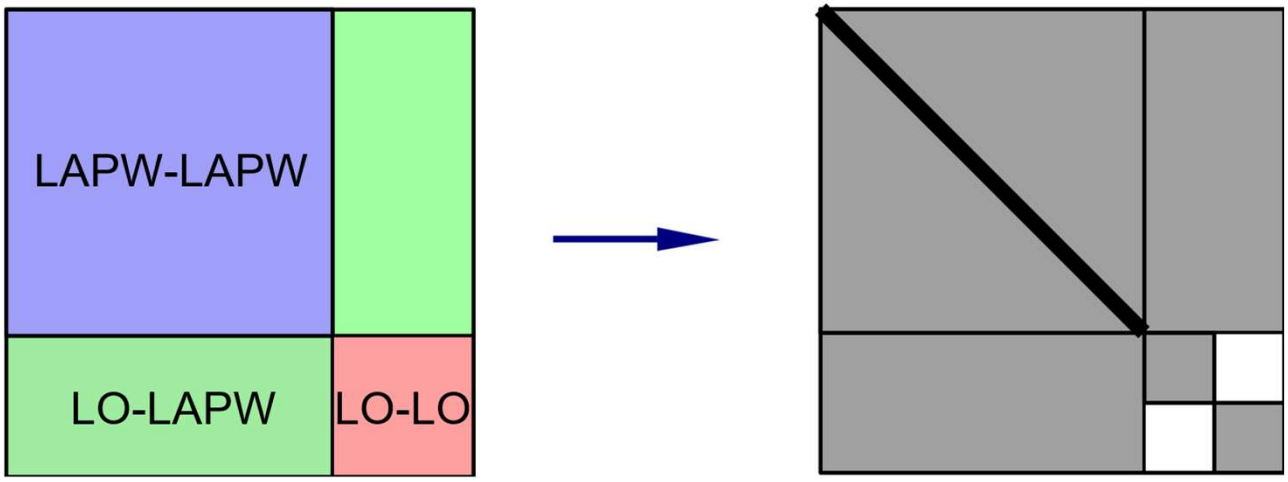






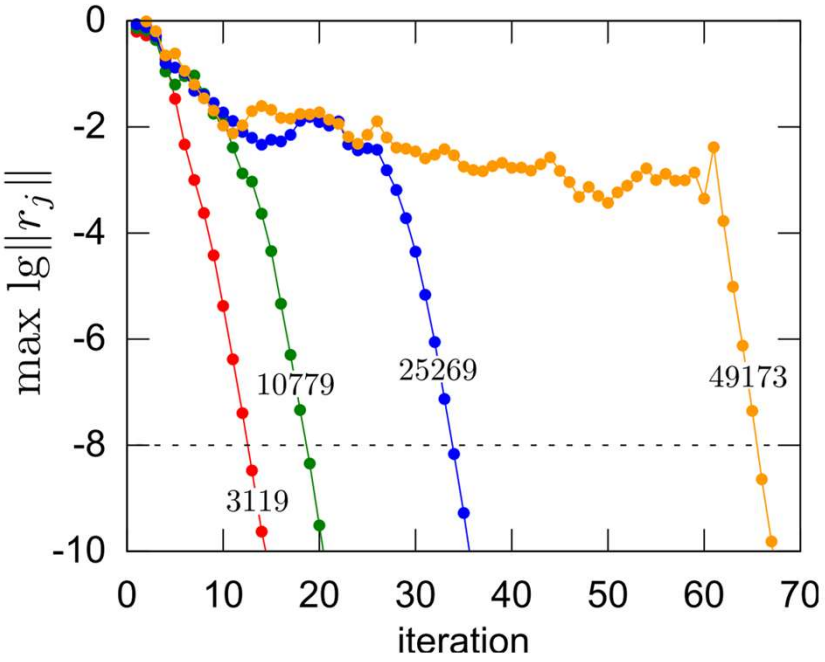
Eigensolver

$$H\psi = ES\psi$$

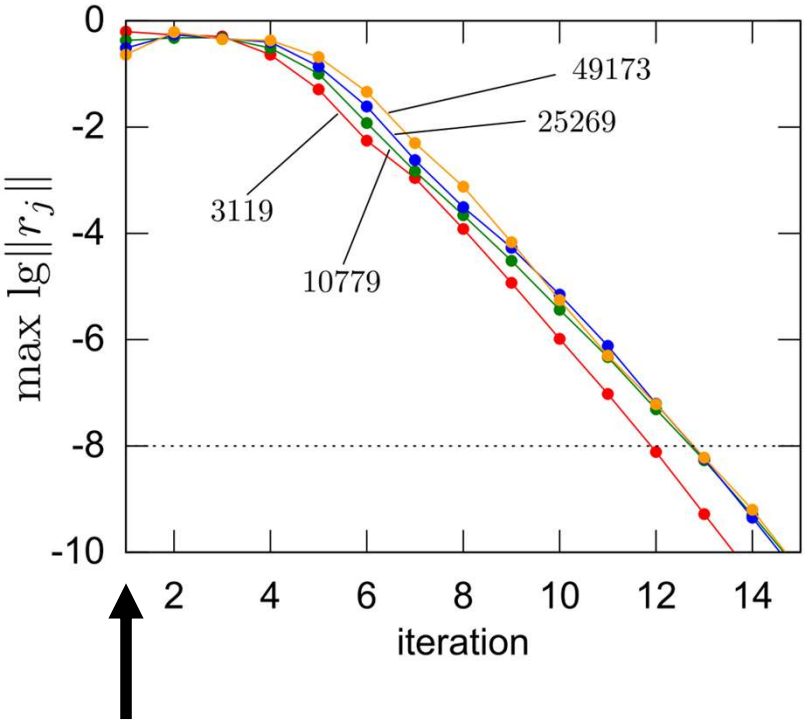


- Diagonally dominant block
- Potentially high condition number – the key difference wrt plane-wave codes
- A good initial guess is available most of the time

H2O in a box: iterative eigensolver

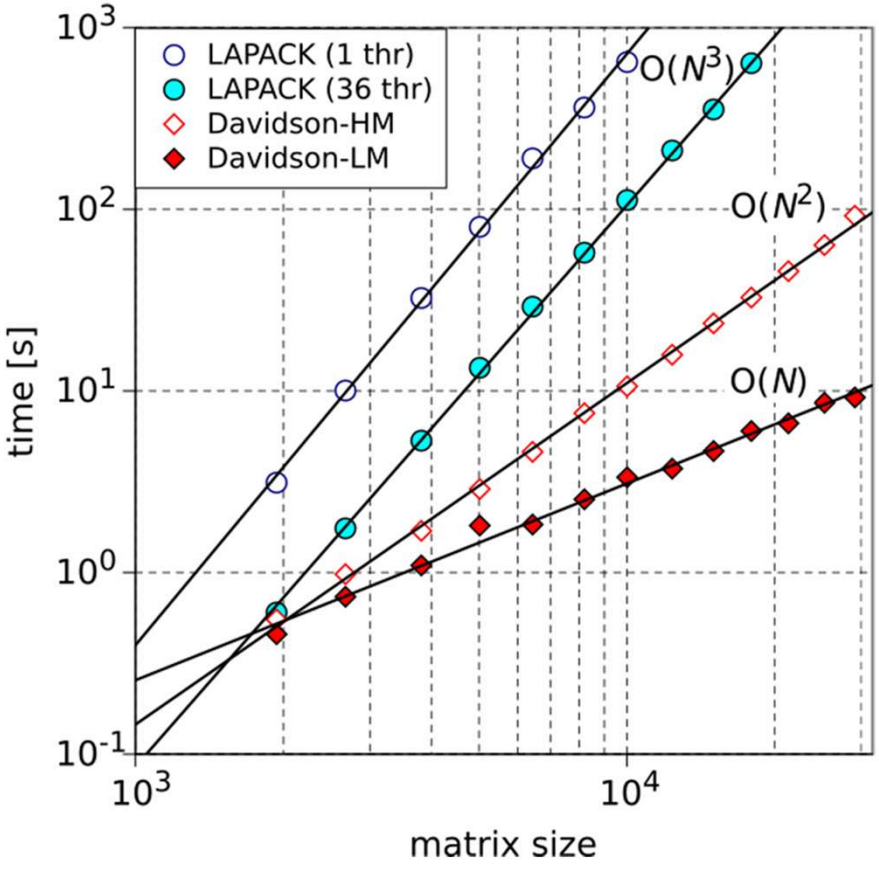


Davidson algorithm,
similar to Singh, Phys. Rev. B **40**, 5428 (1989)



Preparatory work: dump all difficult degrees
of freedom into the diagonalisation subspace

Computational complexity



Gavini et al.,
Modelling Simul. Mater. Sci. Eng. **31**, 063301 (2023)

LAPACK: $O\left(N_{LAPW}^3\right) = O\left(V^3\right)$

High memory Davidson:
$O\left(N_{\mathrm{at}} N_{\mathrm{LAPW}}^2\right) = O\left(N_{\mathrm{at}} V^2\right)$

Matrix-free Davidson:
$O\left(N_{\mathrm{at}}^2 N_{\mathrm{LAPW}}\right) = O\left(N_{\mathrm{at}}^2 V\right)$

A key ingredient in microHartree-level verification

- AG, Kozhevnikov, Draxl, Phys. Rev. B **97**, 161105R (2018)
- Zavickis, Kacars, Cīmurs, **AG**, Phys. Rev. B 106, 165101 (2022)
- Brakestad et al., J. Chem. Theory Comput. 20, 728 (2024)
- Užulis, O 47.3, DPG 2026

Hybrids – adaptively compressed exchange

$$\hat{V}_{HF}\psi_{n\boldsymbol{k}}(\boldsymbol{r}) = -\frac{1}{N_{\boldsymbol{k}}}\sum_{\boldsymbol{k'}}\sum_{m}\psi_{m\boldsymbol{k'}}(\boldsymbol{r})\int\frac{\psi^{*}_{m\boldsymbol{k'}}(\boldsymbol{r'})\psi_{n\boldsymbol{k}}(\boldsymbol{r'})}{|\boldsymbol{r}-\boldsymbol{r'}|}d\boldsymbol{r'}$$

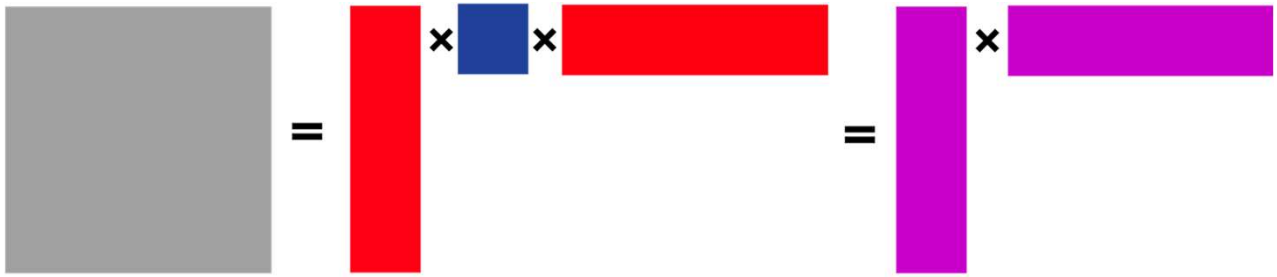
$$\hat{V}_{HF} \approx \sum_{mn}\hat{V}_{HF}|\psi_m\rangle\langle\psi_m|\hat{V}_{HF}^{-1}|\psi_n\rangle\langle\psi_n|\hat{V}_{HF}$$

$$\downarrow$$

$$\langle\psi_m|\hat{V}_{HF}|\psi_n\rangle^{-1}$$

$$\hat{V}_{HF} \approx \sum_{mn}|\psi_m\rangle\langle\psi_m|\hat{V}_{HF}|\psi_n\rangle\langle\psi_n|$$

Alternative approach



Lin, J. Chem. Theory Comput. **12**, 2242 (2016)
Zavickis, Kacars, Cīmurs, AG, Phys. Rev. B **106**, 165101 (2022)

Towards cubic scaling for hybrids

$$\hat{V}_{HF}\psi_{n\boldsymbol{k}}(\boldsymbol{r}) = -\frac{1}{N_{\boldsymbol{k}}}\sum_{\boldsymbol{k}'}\sum_{m}\psi_{m\boldsymbol{k}'}(\boldsymbol{r})\int\frac{\psi^{*}_{m\boldsymbol{k}'}(\boldsymbol{r}')\psi_{n\boldsymbol{k}}(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|}d\boldsymbol{r}'$$

$N_{\mathrm{occ}} \cdot N_{\mathrm{bands}}$ Poisson solves

LAPW-specific solver relies heavily on the Rayleigh formula

$$e^{i\mathbf{k}\cdot\mathbf{r}} = 4\pi\sum_{\ell=0}^{\infty}\sum_{m=-\ell}^{\ell} i^{\ell} j_{\ell}(kr) Y_{\ell}^{m}(\hat{\mathbf{k}}) Y_{\ell}^{m*}(\hat{\mathbf{r}})$$

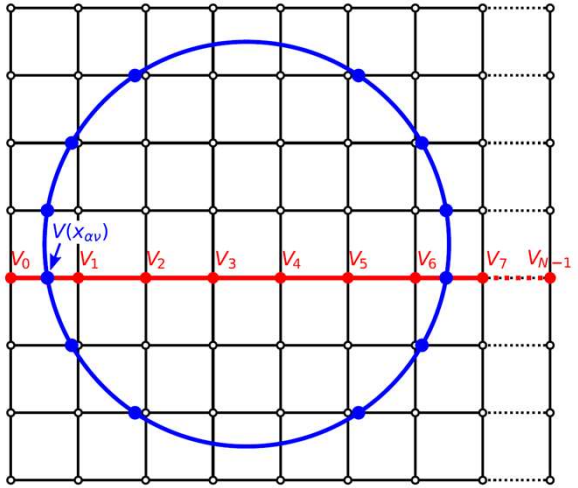
$O(N_{\mathrm{at}}V)$ FLOPs per Poisson solve – $O\left(N_{\mathrm{at}}^{3}V\right)$ FLOPs in total for hybrids

Towards cubic scaling for hybrids

$$\hat{V}_{HF}\psi_{n\boldsymbol{k}}(\boldsymbol{r}) = -\frac{1}{N_{\boldsymbol{k}}}\sum_{\boldsymbol{k}\prime}\sum_{m}\psi_{m\boldsymbol{k}\prime}(\boldsymbol{r})\int\frac{\psi^{*}_{m\boldsymbol{k}\prime}(\boldsymbol{r}')\psi_{n\boldsymbol{k}}(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|}d\boldsymbol{r}'$$

$N_{\mathrm{occ}} \cdot N_{\mathrm{bands}}$ Poisson solves

LAPW-specific solver relies heavily on the Rayleigh formula
Approximate your way around it!



Replace $O(N_{\mathrm{at}}V)$ steps with $O\left(N_{\mathrm{at}}^{1/3}V\right)$ and $O(V\log V)$ and move towards $O\left(N_{\mathrm{at}}^{2}V\log V\right)$ FLOPs in total

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Coulomb singularity

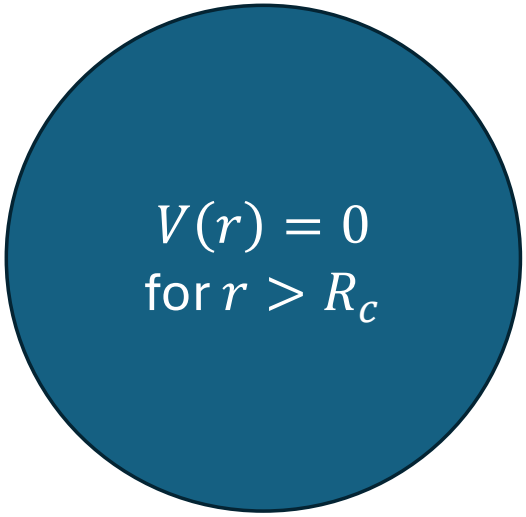
$$\hat{V}_{HF}\psi_{n\boldsymbol{k}}(\boldsymbol{r}) = -\frac{1}{N_{\boldsymbol{k}}}\sum_{\boldsymbol{k'}}\sum_{m}\psi_{m\boldsymbol{k'}}(\boldsymbol{r})\int\frac{\psi^{*}_{m\boldsymbol{k'}}(\boldsymbol{r'})\psi_{n\boldsymbol{k}}(\boldsymbol{r'})}{|\boldsymbol{r}-\boldsymbol{r'}|}d\boldsymbol{r'}$$

singularity for $n = m$ and $\boldsymbol{k} = \boldsymbol{k}'$

3D case:
introduce a cutoff in the Coulomb kernel!

$$V(G) = \frac{4\pi}{G^2}(1-\cos GR_c)$$

$$\frac{4}{3}\pi R_c^3 = VN_{\boldsymbol{k}}$$



Spencer and Alavi, Phys. Rev. B **77**, 193110 (2008)

Coulomb singularity

$$\hat{V}_{HF}\psi_{n\boldsymbol{k}}(\boldsymbol{r}) = -\frac{1}{N_{\boldsymbol{k}}}\sum_{\boldsymbol{k'}}\sum_{m}\psi_{m\boldsymbol{k'}}(\boldsymbol{r})\int\frac{\psi^{*}_{m\boldsymbol{k'}}(\boldsymbol{r'})\psi_{n\boldsymbol{k}}(\boldsymbol{r'})}{|\boldsymbol{r}-\boldsymbol{r'}|}d\boldsymbol{r'}$$

singularity for $n = m$ and $\boldsymbol{k} = \boldsymbol{k}'$

2D case:
introduce a cylindrical cutoff (highball glass)!

$$v(G) = \iiint_{\mathcal{D}} v(r)e^{i\boldsymbol{G}\boldsymbol{r}}d\boldsymbol{r}$$

Coulomb singularity

$$\pi\rho^2 = VN_{\boldsymbol{k}}/c$$

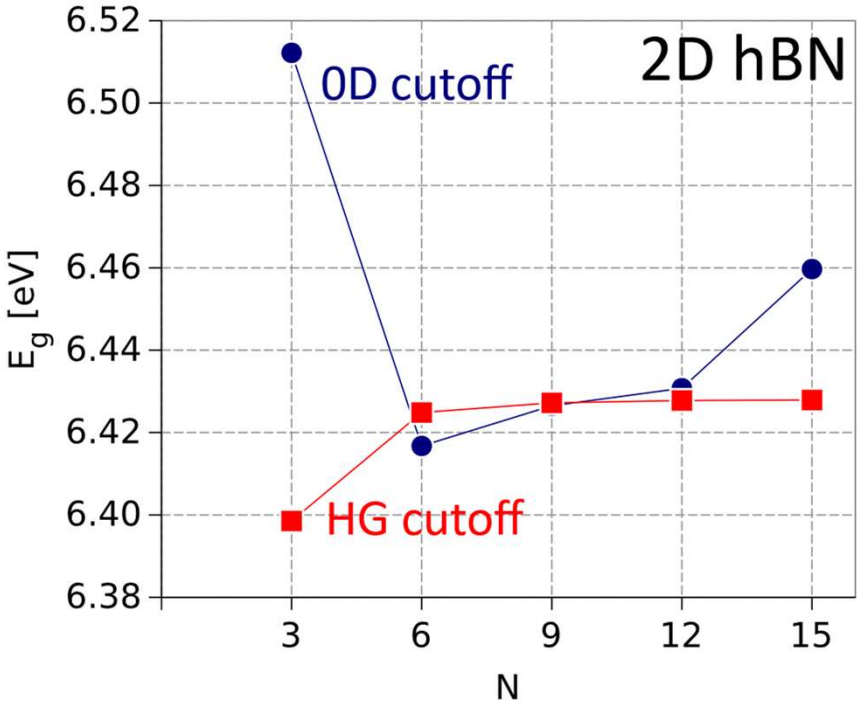
$$\hat{V}_{HF}\psi_{n\boldsymbol{k}}(\boldsymbol{r}) = -\frac{1}{N_{\boldsymbol{k}}}\sum_{\boldsymbol{k}'}\sum_{m}\psi_{m\boldsymbol{k}'}(\boldsymbol{r})\int\frac{\psi^*_{m\boldsymbol{k}'}(\boldsymbol{r}')\psi_{n\boldsymbol{k}}(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|}d\boldsymbol{r}'$$

singularity for $n = m$ and $\boldsymbol{k} = \boldsymbol{k}'$

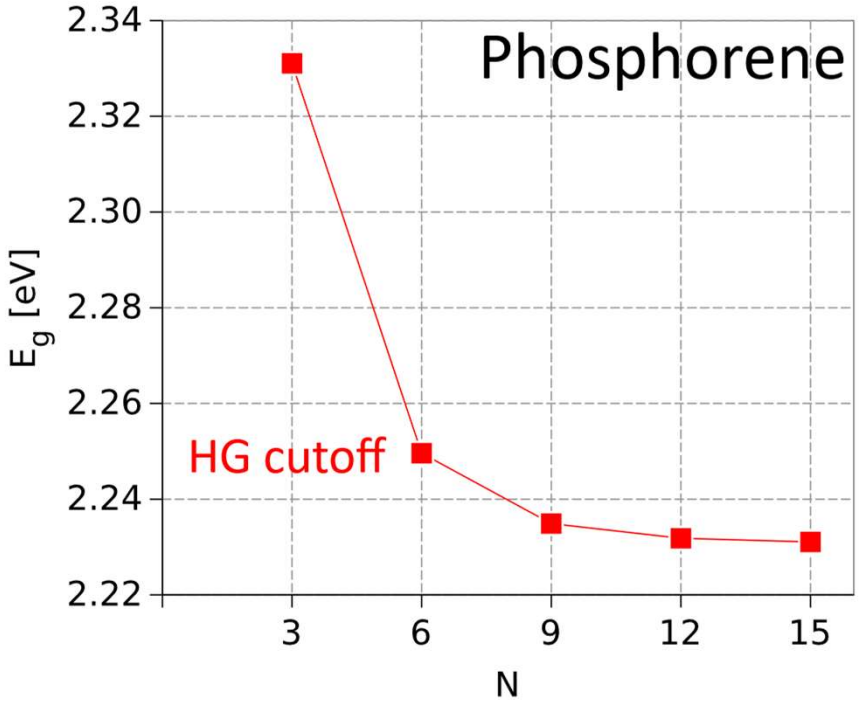
2D case:
introduce a cylindrical cutoff (highball glass)!

$$v(G) = 2\pi\int_{-c/2}^{c/2}dz\int_{0}^{\rho}dr\,\frac{rJ_0(G_{\parallel}r)}{\sqrt{r^2+z^2}}$$

Small prototype application: PBE0 band gaps



Ref. 6.35 eV
Expt. 6.0 eV



Ref. 2.22 eV
Expt. 2.05 eV

Ref: Wiktor and Pasquarello, Phys. Rev. B **94**, 245411 (2016)

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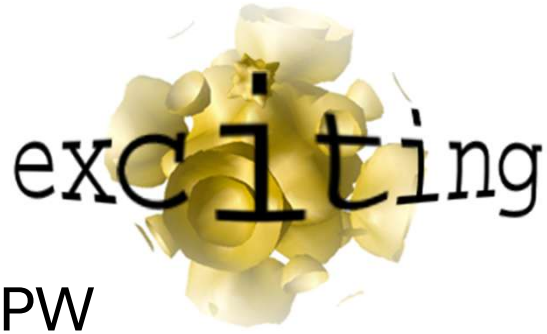
FUNDAMENTĀLO UN LIETIŠĶO PĒTĪJUMU PROJEKTI

LZP-2024/1-0202
Precise methods for modelling quantum materials

EuroHPC
Joint Undertaking

EuroHPC development access projects at BSC / MareNostrum V

Summary



We addressed efficiency and functionality needs for LAPW applied for low-dimensional systems.

- Iterative eigensolver (17x speedup over LAPACK for 2D-hBN)
- Hybrids via adaptively compressed exchange
  - no empty bands required
  - cubic complexity in sight
  - recommended method in LAPW
- Highball-glass Coulomb cutoff (efficient BZ integration for Fock exchange, can be useful also in 0D, 1D and 3D)