

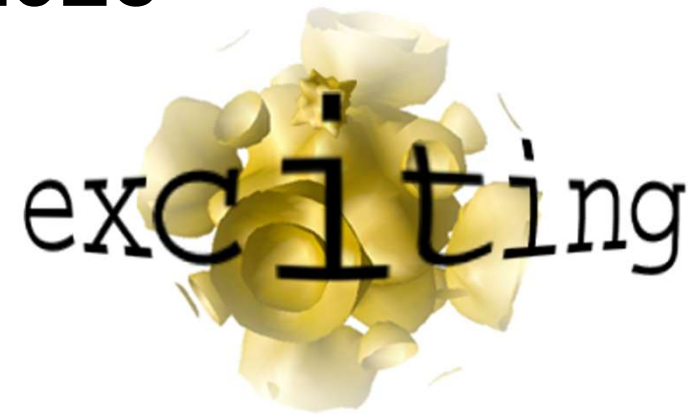
Fast and robust hybrid-functional calculations in LAPW enabled by ACE

Andris Gulans

Berlin, HoW exciting! 2026



**LATVIJAS
UNIVERSITĀTE**



Towards accurate gaps from hybrid functionals

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

Band-gap errors (eV) w.r.t. expt. – J. Yang *et al.*, [npj Computational Materials 9, 108 \(2023\)](#)

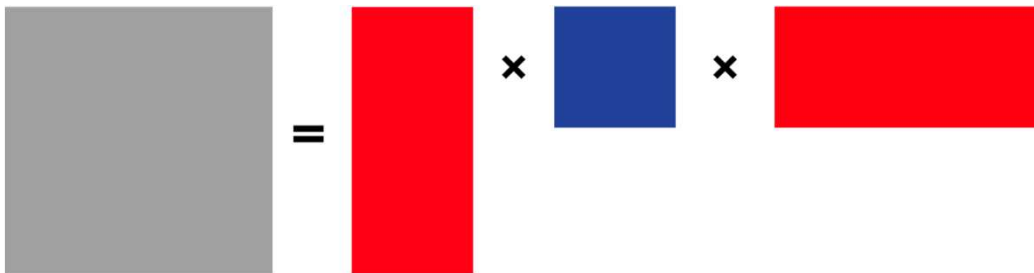
	PBE0	DD-PBE0	HSE06	TF	$\mu_{\text{fix}}^{a_s=0.25}$	DSH	DD-CAM	$\mu_{\text{fix}}^{a_s=1}$
26 materials								
MAE	0.73	0.44	0.98	0.41	0.41	0.24	0.23	0.23
MARE	18.0%	17.2%	17.9%	14.3%	14.6%	6.8%	6.2%	6.1%

- Target accuracy: 0.1 eV
- Best achieved accuracy: 0.23 eV
- Precision: ???

Hybrid functionals in LAPW (standard approach)

$$\hat{V}_{HF}\psi_{nk}(\mathbf{r}) = -\frac{1}{N_k}\sum_{k'}\sum_m\psi_{mk'}(\mathbf{r})\int\frac{\psi_{mk'}^*(\mathbf{r}')\psi_{nk}(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}d\mathbf{r}'$$

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



- Dependence on empty bands
- N^4 computational effort
- Well integrated with features in **exciting**
- Available in **exciting** Mg

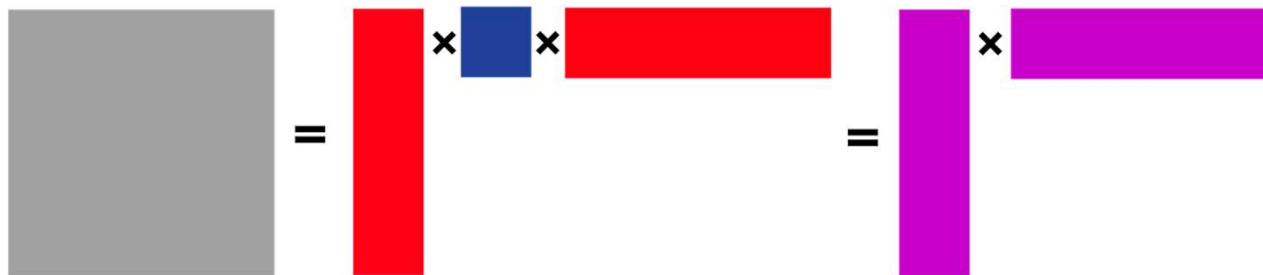
Adaptively compressed exchange

$$\hat{V}_{HF}\psi_{nk}(\mathbf{r}) = -\frac{1}{N_{k'}} \sum_{k'} \sum_m \psi_{mk'}(\mathbf{r}) \int \frac{\psi_{mk'}^*(\mathbf{r}')\psi_{nk}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}'$$

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

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

- No empty bands dependence
- New feature in **exciting**
- Available in future releases



Lin, J. Chem. Theory Comput. **12**, 2242 (2016)

Zavickis, Kacars, Cīmurs, Gulans, Phys. Rev. B **106**, 165101 (2022)

Towards cubic scaling for hybrids

$$\hat{V}_{HF}\psi_{nk}(\mathbf{r}) = -\frac{1}{N_{k'}} \sum_{k'} \sum_m \psi_{mk'}(\mathbf{r}) \int \frac{\psi_{mk'}^*(\mathbf{r}')\psi_{nk}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d\mathbf{r}'$$

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

LAPW-specific solver relies heavily on the Rayleigh formula

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

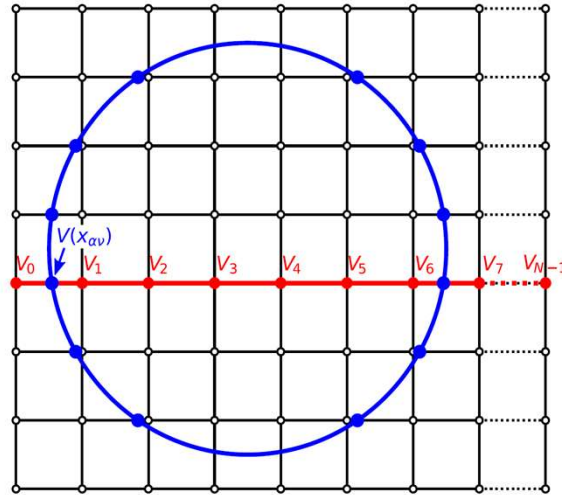
$O(N_{\text{at}}\Omega)$ FLOPs per Poisson solve – $O(N_{\text{at}}^3\Omega)$ FLOPs in total for hybrids

Approximate your way around it!

Exact calculation in G-space

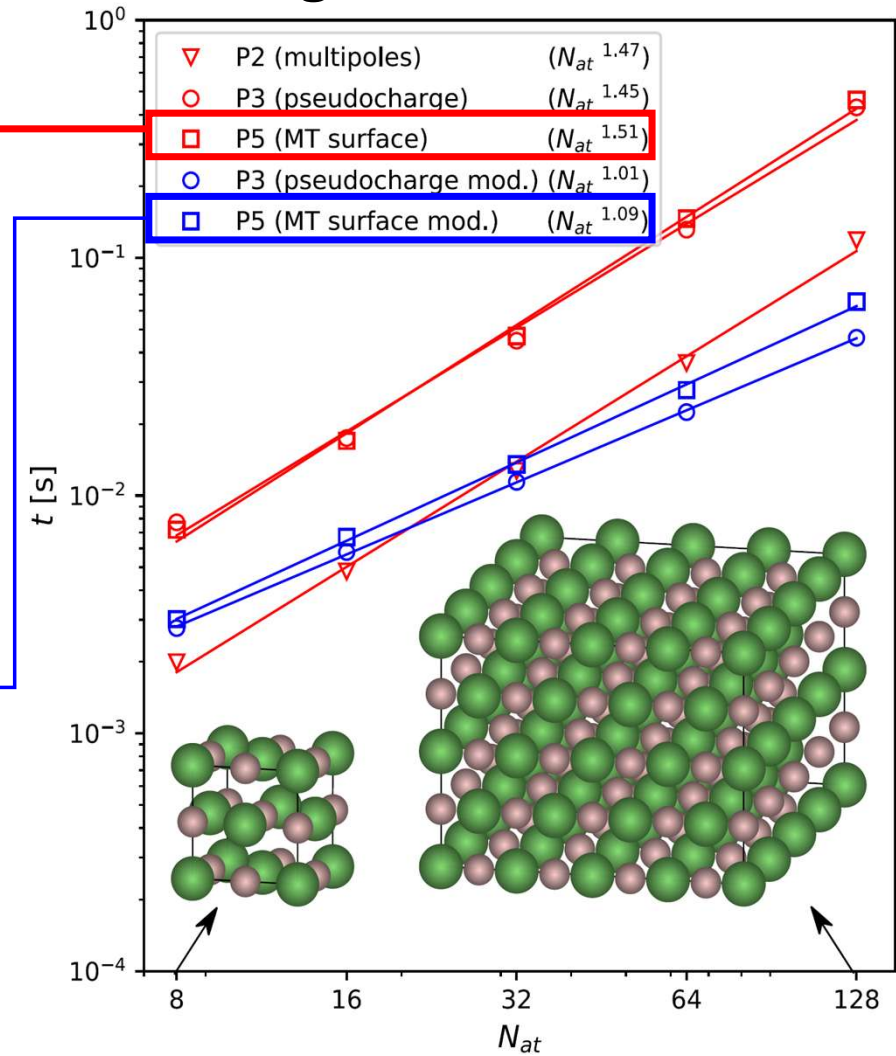
$$V_{\alpha;\ell m}^{\text{surf}} = 4\pi i^\ell \sum_{\mathbf{G}} j_\ell(GR_\alpha) V^{\text{I}}(\mathbf{G}) e^{i\mathbf{G}\mathbf{R}_\alpha} Y_{\ell m}^*(\hat{\mathbf{G}})$$

1D Fourier interpolation



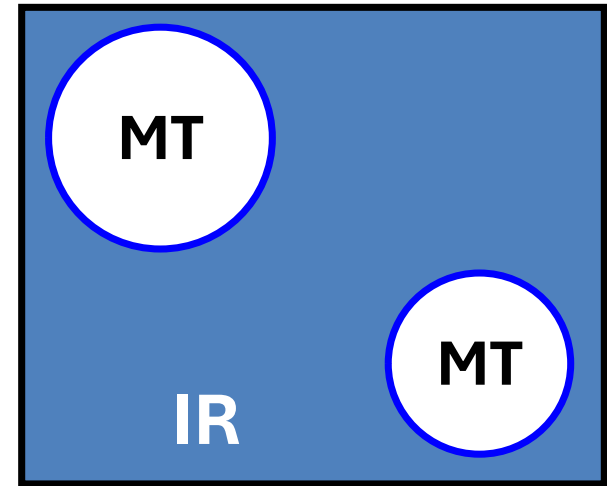
Least-squares fit $V(\hat{\mathbf{r}}_{\alpha\nu}) = \sum_{\ell m} V_{\alpha;\ell m}^{\text{surf}} Y_{\ell m}(\hat{\mathbf{r}}_{\alpha\nu})$

Single Poisson solve



Precision issues: basis

$$\phi_{\mathbf{G}+\mathbf{k}}^{\text{LAPW}}(\mathbf{r}) = \begin{cases} \sum_{\xi lm} A_{\xi lm}^{\mathbf{G}+\mathbf{k}} u_{\xi l}(r) Y_{lm}(\hat{\mathbf{r}}) & \mathbf{r} \in \text{MT} \\ \frac{1}{\sqrt{\Omega}} e^{i(\mathbf{G}+\mathbf{k})\mathbf{r}} & \mathbf{r} \in \text{IR} \end{cases}$$



We obtain the radial functions from the radial Schrödinger equation:

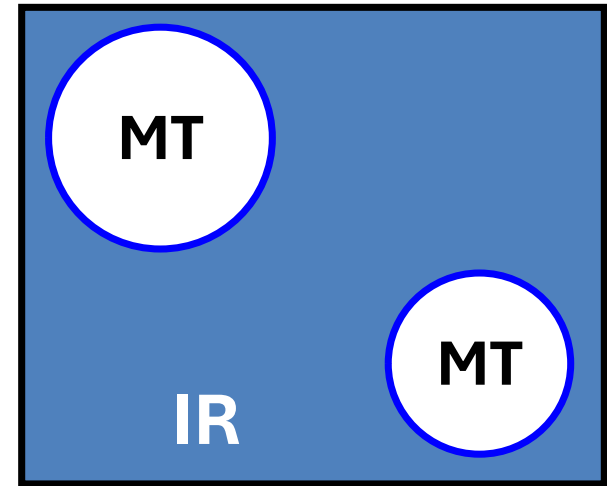
$$\left(-\frac{1}{2} \frac{\partial^2}{\partial r^2} + \frac{\ell(\ell+1)}{2r^2} + v_0(r) - E_\ell \right) r u_\ell(r; E_\ell) = 0$$



local potential

Precision issues: basis

$$\phi_{\mathbf{G}+\mathbf{k}}^{\text{LAPW}}(\mathbf{r}) = \begin{cases} \sum_{\xi l} A_{\xi lm}^{\mathbf{G}+\mathbf{k}} u_{\xi l}(r) Y_{lm}(\hat{\mathbf{r}}) & \mathbf{r} \in \text{MT} \\ \frac{1}{\sqrt{\Omega}} e^{i(\mathbf{G}+\mathbf{k})\mathbf{r}} & \mathbf{r} \in \text{IR} \end{cases}$$



We obtain the radial functions from the radial Schrödinger equation:

$$\left(-\frac{1}{2} \frac{\partial^2}{\partial r^2} + \frac{\ell(\ell+1)}{2r^2} + v_0(r) - E_\ell \right) r u_\ell(r; E_\ell) = -r h_\ell(r)$$


 local potential

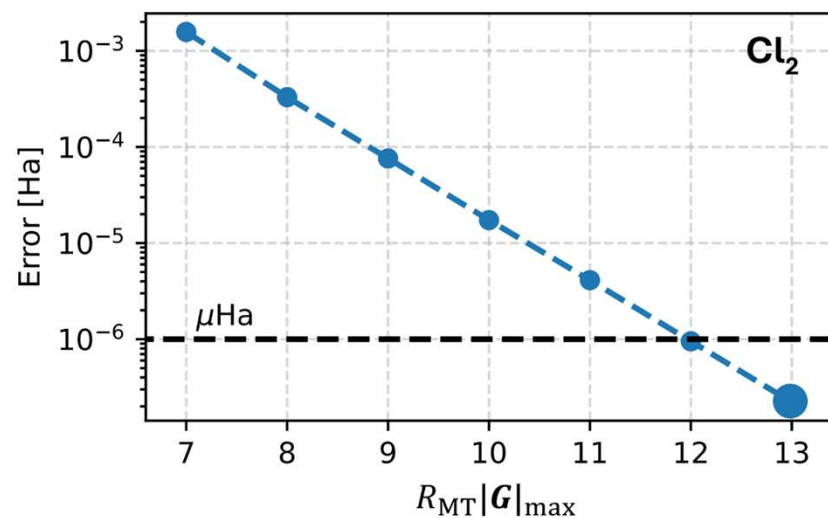

 nonlocal
contribution

$$\hat{K}[u_\ell(r; E_\ell) Y_{\ell m}(\hat{r})] = h_\ell(r) Y_{\ell m}(\hat{r})$$

J. Užulis, A. Gulans,
[arXiv:2606.20116](https://arxiv.org/abs/2606.20116) (2026)

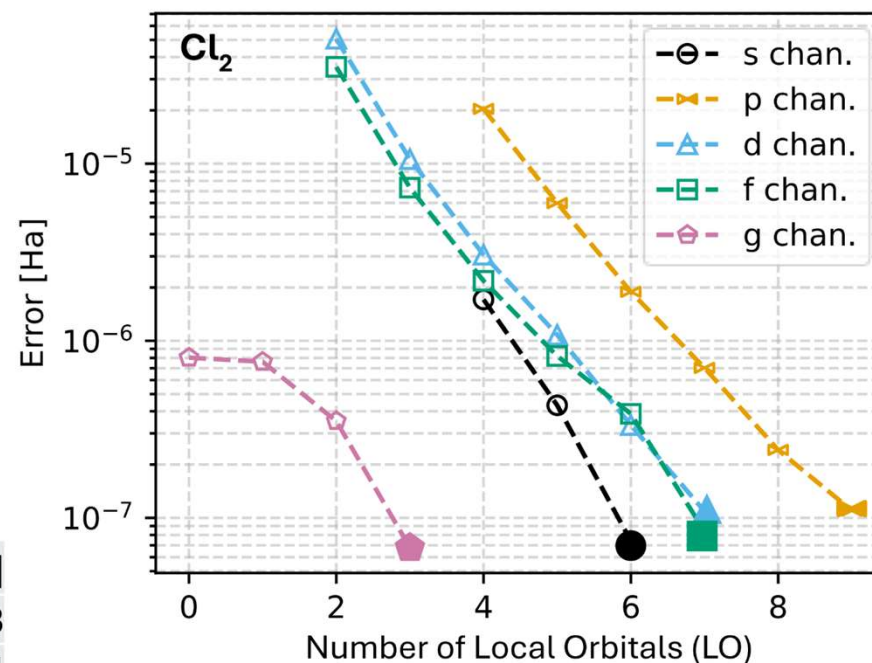
Basis completeness – total HF energy

Interstitial region basis convergence.



Molecule	exciting [Ha]	FDM or FEM [Ha]	ΔE [μHa]
H ₂	-1.133624	-1.133624*	-0.3
FH	-100.070801	-100.070803 ¹	1.4
F ₂	-198.773442	-198.773445 ¹	2.3
ClF	-558.917625	-558.917626 ¹	0.9
Cl ₂	-919.008932	-919.008935 ¹	2.6

MT region basis convergence.



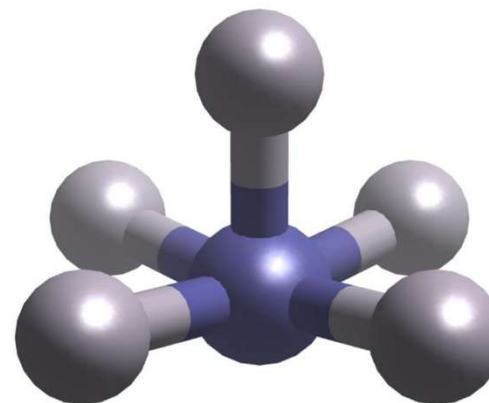
* – calculated with HELFEM

¹ – Jensen, Theoretical Chemistry
Accounts 113, 187–190 (2005)

J. Użulis, A. Gulans, [arXiv:2606.20116](https://arxiv.org/abs/2606.20116) (2026)

Basis completeness – total HF energy

$$E_{\text{reaction}} = E_{\text{products}} - E_{\text{reactants}}$$



exciting (LAPW)

R_{MT}	G_{max}	Core type	Basis type	error [kcal/mol]		
				FH*	ClF	ClF ₅
13		HF	HF	-	-	-
8		HF	HF	0.01	0.00	0.00
8		PBE	PBE	0.00	0.00	0.04

NWChem (LCAO)

Basis						
	aug-cc-pV6Z	0.00	0.03	0.60		
	aug-cc-pV5Z	0.00	0.08	1.72		
	aug-cc-pVQZ	-0.04	0.24	6.98		
	aug-cc-pVTZ	0.06	0.74	13.10		
	aug-cc-pVDZ	-1.51	-0.54	32.47		

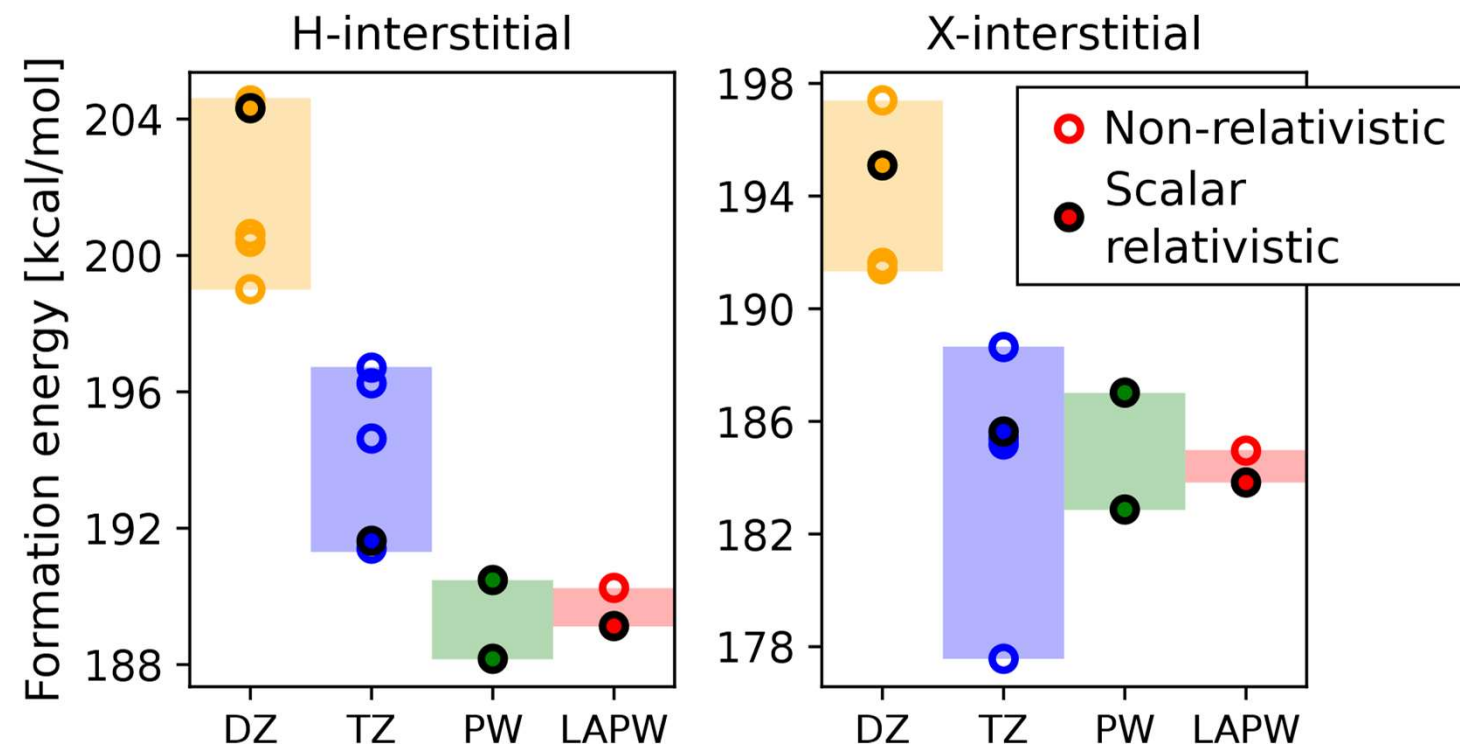
* - hydrogen fluoride denoted as FH, to avoid confusion with Hartee-Fock.

- corresponds to CBL
 - does not correspond to CBL

- error above chemical accuracy (1 kcal/mol)

J. Užulis, A. Gulans, [arXiv:2606.20116](https://arxiv.org/abs/2606.20116) (2026)

Basis completeness – self-interstitials in Si



LAPW computational details:

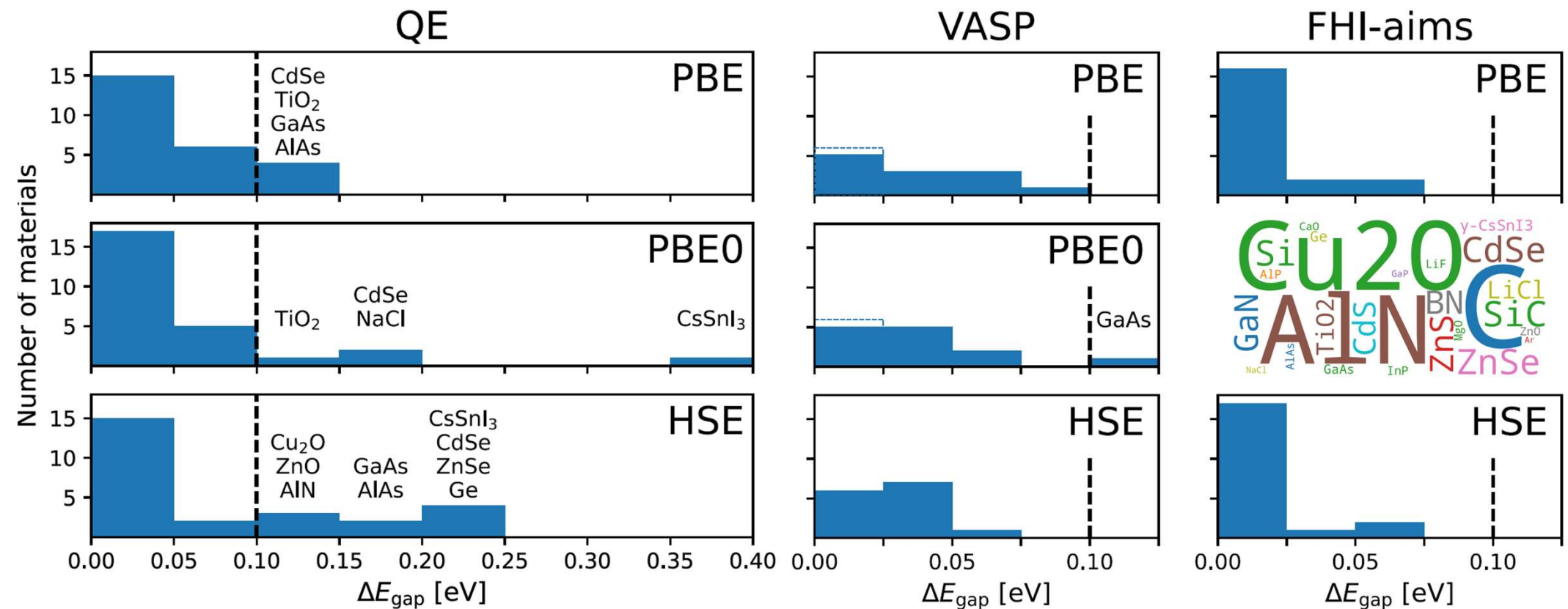
- 17-atom supercell
- Hartree-Fock
- 4x4x4 **k**-grid
- Spherical Coulomb cutoff

F. Salihbegović, A. Gallo, A. Grüneis, Phys. Rev. B 108, 115125 (2023)

K. Simula et al., Phys. Rev. B 113, 1951 (2026)

J. Užulis, A. Gulans, [arXiv:2606.20116](https://arxiv.org/abs/2606.20116) (2026)

Band gaps comparison: 3D materials



J. Užulis, A. Sorokin, A. Gulans, [npj Computational Materials 11, 257 \(2025\)](#)

Acknowledgments



Jānis Užulis



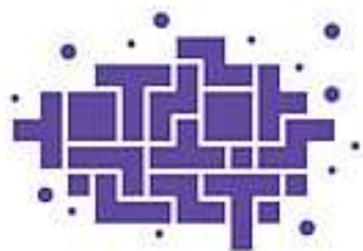
Ernests Lazdāns



Kristaps Feldmans

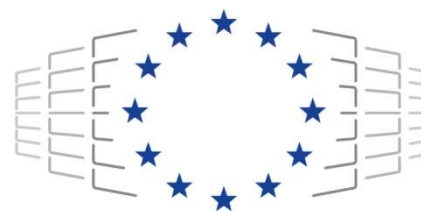


<https://quantumtheory.lu.lv>



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PROJEKTI

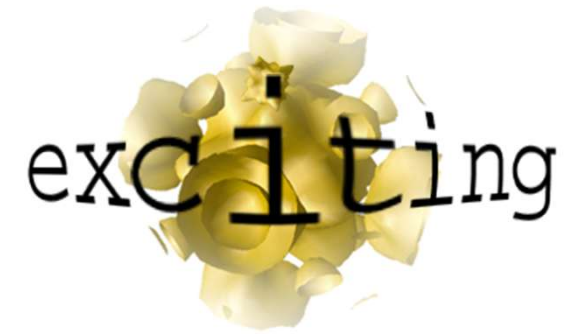
LZP-2024/1-0202
Precise methods for modelling
quantum materials



EuroHPC
Joint Undertaking

EuroHPC development access project
at BSC / MareNostrum V

Summary and outlook



Hybrids via adaptively compressed exchange

- no additional empty bands required
- cubic complexity in sight
- microhartree precision
- access to core electrons via hybrids
- various cutoffs (ask me if interested)
- recommended method in LAPW
- available in future **exciting** releases or on github