Introduction to FHI-aims

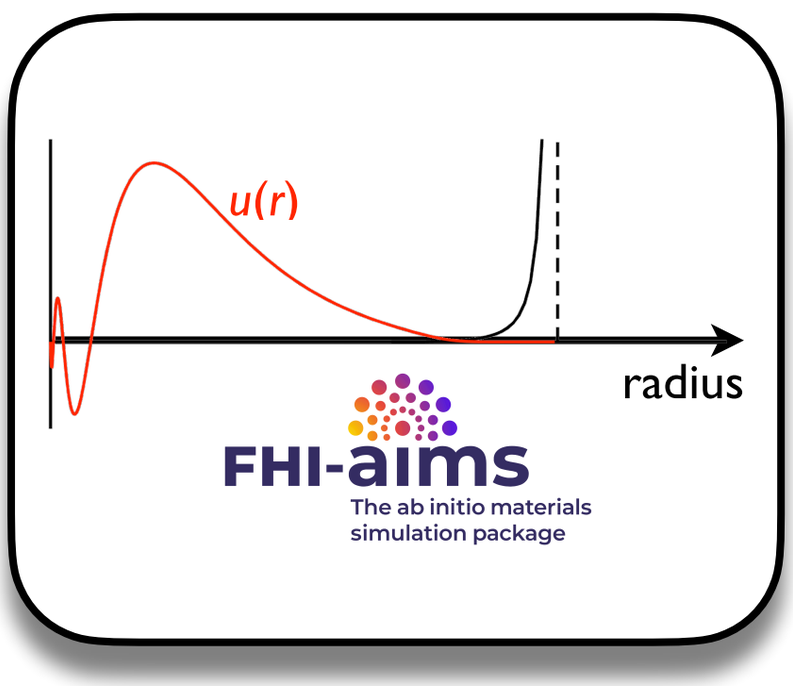
Efficient, Accurate Electronic Structure Theory for Molecules and Solids

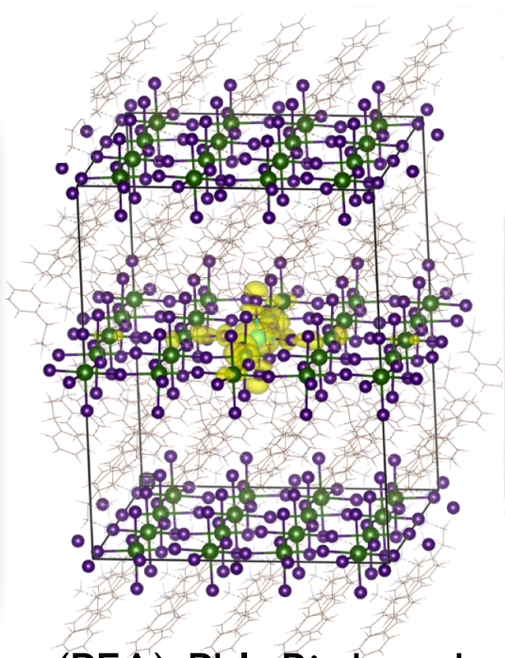
Volker Blum

MEMS & Chemistry Departments, Duke University, Durham, NC

http://aims.pratt.duke.edu

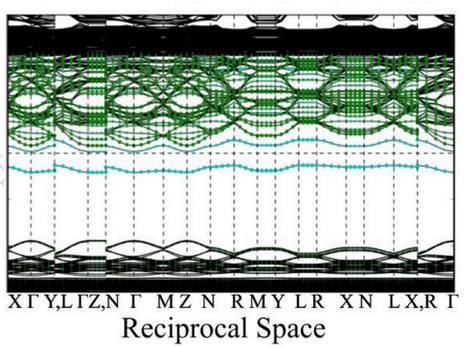
ACI-1450280
DMR-1729297

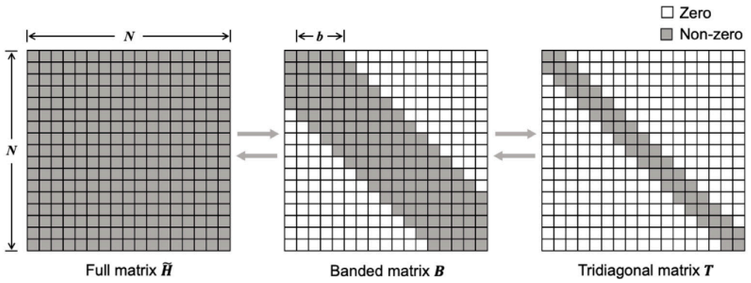




$(PEA)_2PbI_4$, Bi-doped

HSE06+SOC





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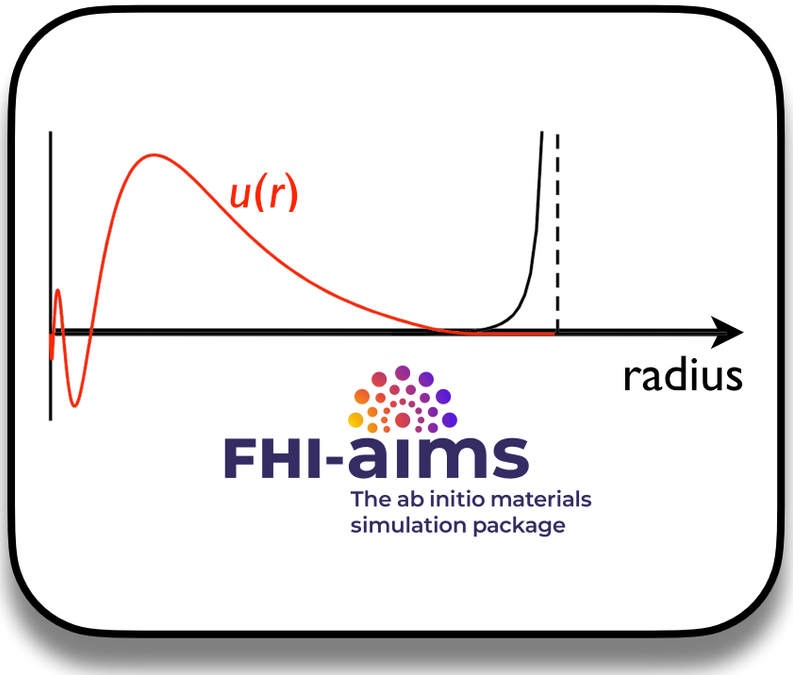
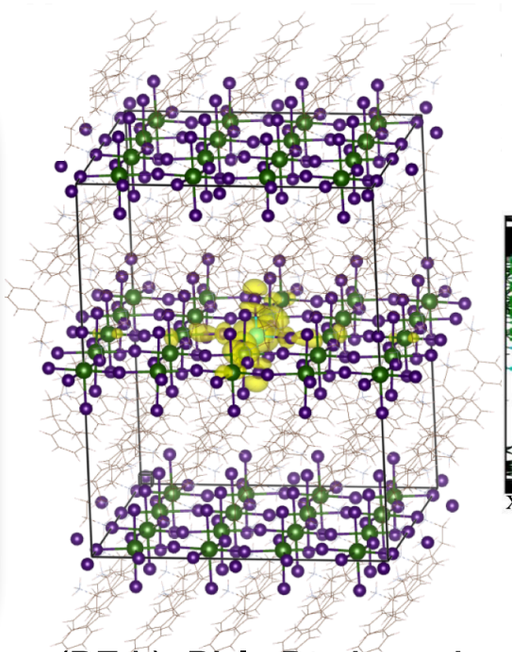
All-Electron Theory in Numeric Atom-Centered Basis Sets



Large, Complex Systems: Feasible.



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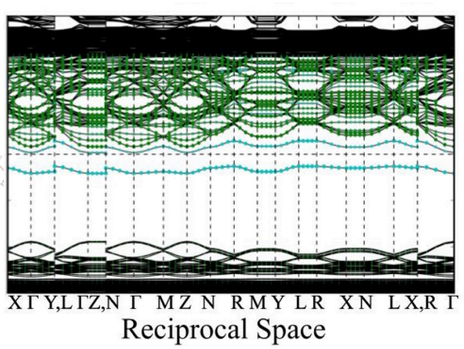
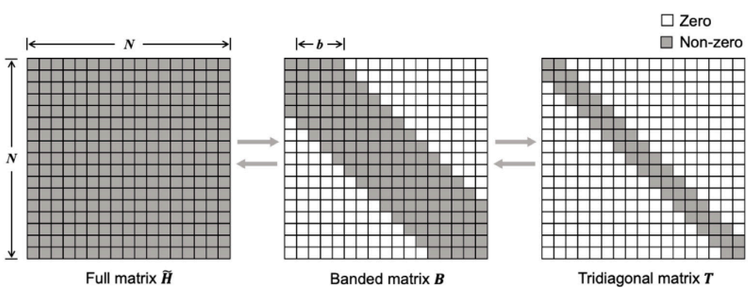


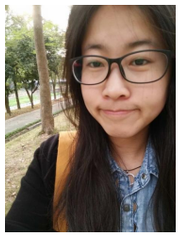
Software Ecosystem: ELPA, ELSI, GIMS, CECAM ESL



Acknowledgments

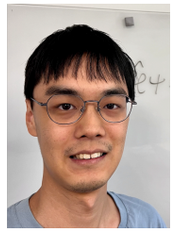
Ab Initio Materials Simulations Group (Duke Univ.)

Present



Xixi Qin

Gabrielle Koknat

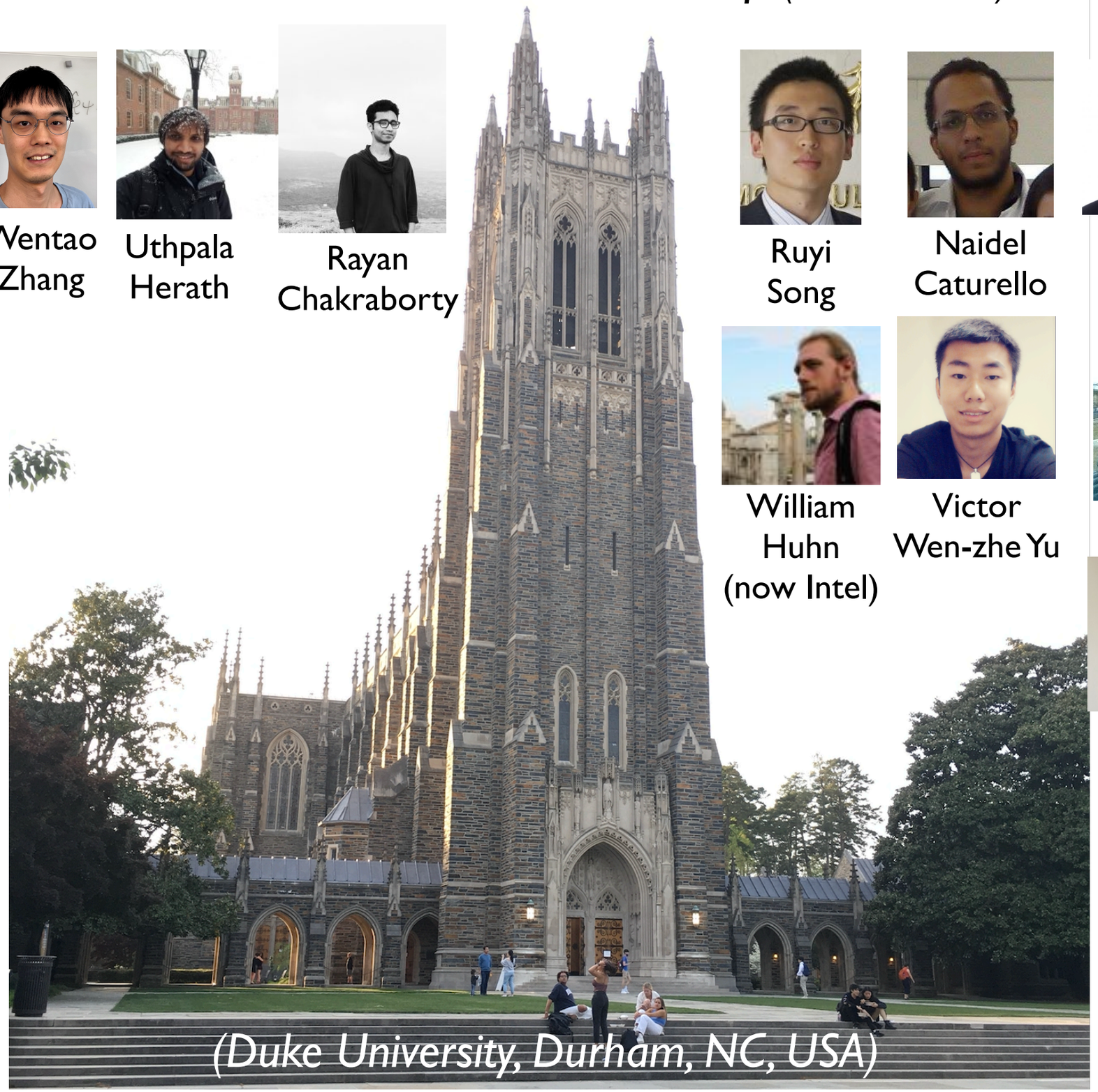


Wentao Zhang

Uthpala Herath

Rayan Chakraborty

Jack Morgenstein
Ignacio Valdez
Jannik Eisenlohr
Reyna Vrbensky
Douglas Heine
Kelly Ma



(Duke University, Durham, NC, USA)

Ruyi Song

Naidel Caturello

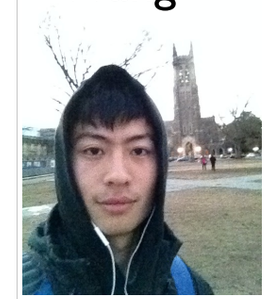
William Huhn (now Intel)

Victor Wen-zhe Yu

Past

Tianlin Wang

Yi Yao



Chi Liu

Raul Laasner

Rundong Zhao

… and many more!

Duke
PRATT SCHOOL of ENGINEERING

FHI-aims Code: Large Community

https://fhi-aims.org

FHI-aims
The ab initio materials simulation package

FHI-aims team and collaborators: Matthias Scheffler (Berlin), Sebastian Kokott (Berlin), Mariana Rossi (Hamburg), Xinguo Ren (Beijing), Karsten Reuter (Berlin), over 150 individuals with contributions to the project. Development in Berlin, Duke, UNC, Beijing, Helsinki, Warwick, Cardiff, Dresden, Dalhousie, etc.

Coordinators: Blum, Kokott, Rossi, Scheffler

Alaa Akkoush, Heiko Appel, Victor Atalla, Kurt Baarmann, Carsten Baldauf, Alexej Bagrets, Jörg Behler, Daniel Berger, Josh Berryman, Sheng Bi, Benedikt Biedermann, Bjoern Bieniek, Volker Blum, Saeed Bohloul, Tiago Botari, Connor Box, Danilo Brambila, Gabriel Bramley, Daniel Bultrini, Christian Carbogno, Fabio Caruso, Marco Casadei, Michele Ceriotti, Wael Chibani, Sucismita Chutia, Francisco Antonio Delesma, Fabio Della Sala, Maria Dragoumi, Andreas Dolfen, Marc Dvorak, Simon Erker, Ferdinand Evers, Eduardo Fabiano, Matt Farrow, Nicola Ferri, Karen Fidanyan, Jakob Filser, Lukas Gallandi, Ralf Gehrke, Luca Ghiringhelli, Mark Glass, Vivekanand Gobre, Dorothea Golze, Matthias Gramzow, Patrick Guetlein, Stefan Gutzeit, Volker Haigis, Felix Hanke, Paula Havu, Ville Havu, Joscha Hekele, Olle Hellman, Jan Hermann, Oliver Hofmann, Johannes Hoja, Xiaojuan Hu, William Huhn, Lukas Hörmann, Arvid Ihrig, Timo Jacob, Adam Jackson, Svenja Janke, Ran Jia, Rainer Johanni, Erin Johnson, Werner Jürgens, Matthias Kahk, Yosuke Kanai, Levi Keller, Matthias Kick, Woo Youn Kim, Jan Kloppenburg, Alexander Knoll, Florian Knoop, Franz Knuth, Simone Koecher, Gabrielle Koknat, Sebastian Kokott, Raul Laasner, Lucas Lang, Bjoern Lange, Marvin Lechner, Susi Lehtola, Maja-Olivia Lenz, Sergey Levchenko, Alan Lewis, Jiachen Li, Xinzheng Li, Kailai Lin, Xinyi Lin, Konstantin Lion, Yair Litman, Chi Liu, Andrew Logsdail, Andreas Marek, Thomas Markovich, Reinhard Maurer, Florian Merz, Joerg Meyer, Wenhui Mi, Evgeny Moerman, Christoph Muschielok, Mohammad Nakhaee, Lydia Nemec, Norbert Nemec, Kane O'Donnell, Harald Oberhofer, Berk Onat, Alberto Otero de la Rosa, Ramon L. Panades-Barrueta, Eszter Pos, Alastair Price, Thomas Purcell, Nathaniel Raimbault, Karsten Rasim, Xinguo Ren, Karsten Reuter, Norina Richter, Stefan Ringe, Patrick Rinke, Herzain Rivera, Matti Ropo, Mariana Rossi, Tuomas Rossi, Adrienn Ruzsinszky, Nikita Rybin, Georg Michelitsch, Andrea Sanfilippo, Matthias Scheffler, Markus Schneider, Christoph Schober, Franziska Schubert, Honghui Shang, Tonghao Shen, Markus Sinstein, Justin Clifford Smith, Ari-Pekka Soikkeli, Ruyi Song, Aloysius Soon, Pavel Stishenko, Muhammad Tahir, Alexandre Tkatchenko, Thomas Theis, Alvaro Vazquez Mayagoitia, Suzy Wallace, Tianlin Wang, Yanyong Wang, Jürgen Wieferink, Scott Woodley, Jianhang Xu,Yong Xu,Yi Yao, Mina Yoon,Ted Yu, Victor Yu, Zhenkun Yuan, Marios Zacharias, Guo-Xu Zhang, Igor Ying Zhang, Wenxing Zhang, Rundong Zhao, Ruiyi Zhou, Yuanyuan Zhou, Tong Zhu

MS1P
Molecular Simulations from First Principles e.V.

... and, with absolute certainty, more! Thank you!

How Are “We” Organized

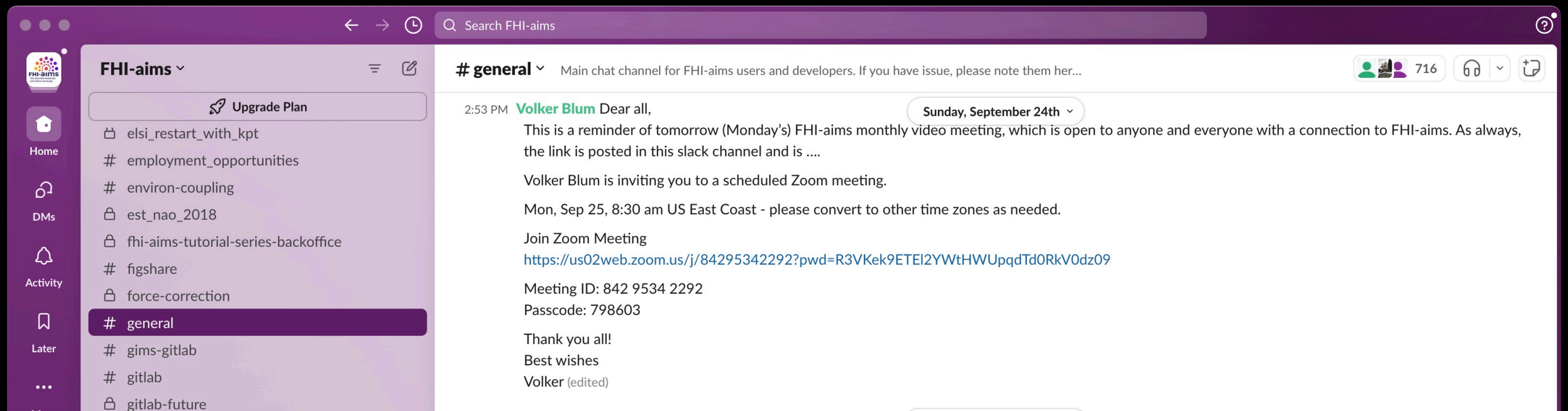
FHI-aims is licensed through MS1P e.V. - registered non-profit organization, objective is support of basic science

“fhi-aims.org” used for registration, distribution of point versions

Slack Channel - fhi-aims.slack.com, 716 users (Oct. 1, 2023)

Community Server - https://aims-git.rz-berlin.mpg.de, 672 users (Oct. 1, 2023)
Development, stable versions, issue tracker, wiki & build instructions, …

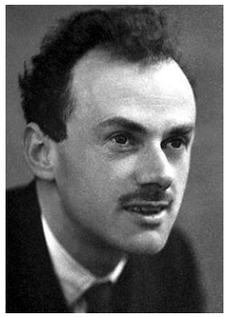
Broad ecosystem of connected infrastructure:
ASE, i-Pi, phonopy, FHI-vibes, GIMS, MDAnalysis, GAtor/Genarris, CECAM ESL, many more.



We Have a Predictive Theory for (Almost) Everything!

$$\hat{\mathcal{H}}\Psi = E\Psi$$

P.A.M. Dirac



<u>*Dirac 1929:*</u>

"The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known ..."

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$$\hat{\mathcal{H}}\Psi = E\Psi$$

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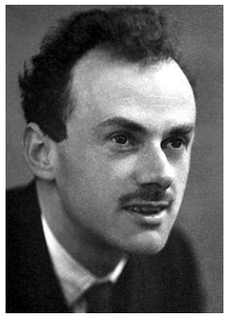

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"..., and the difficulty is only that the exact application of these laws leads to equations much too complicated to be soluble."

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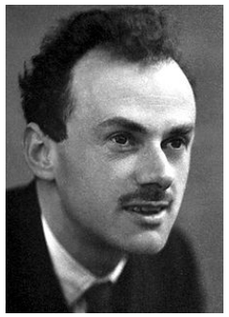


<u>*Dirac 1929:*</u>

"The underlying physical laws necessary for the mathematical theory of a large part of physics and the whole of chemistry are thus completely known ..."

"... , and the difficulty is only that the exact application of these laws leads to equations much too complicated to be soluble."

"... it therefore becomes desirable that approximate practical methods of applying quantum mechanics should be developed, which can lead to an explanation of the main features of complex atomic systems without too much computation."

Our Approach to Electronic Structure Theory: FHI-aims

FHI-aims
The ab initio materials simulation package

https://fhi-aims.org

Vision:
Quantum mechanics based simulations of real, complex molecules, materials and their properties without a priori precision and accuracy limitations.

→ Algorithmic choices and priorities:

- All-electron
- Non-periodic and periodic systems on equal footing
- Scalability to large systems (thousands of atoms) without precision limitations
- Seamless scalability from laptop to massively parallel and/or new HPC architectures
- Density functional theory and correlated methods (RPA, GW, CC via CC4S…)

Critical choice

- Numeric atom-centered basis functions (accurate representation of occupied orbitals and densities)

MS1P
Molecular Simulations from First Principles e.V.

Stewardship: MS1P e.V. (non-profit organization, purpose: basic science)
Advisory board, large & very active community

What Can We Do?

DFT …. Basically, Bread and Butter

Structure, MD, Molecules and Solids

nature chemistry

Article

https://doi.org/10.1038/s41557-023-01311-0

Thickness control of organic semiconductor-incorporated perovskites

Received: 4 January 2022

Accepted: 31 July 2023

Published online: 31 August 2023

Check for updates

Jee Yung Park[1,12], Ruyi Song[2,12], Jie Liang[3,12], Linrui Jin[4,12], Kang Wan
Shunran Li[5,6], Enzheng Shi[7], Yao Gao[8], Matthias Zeller[4], Simon J. Teat
Peijun Guo[5,6], Libai Huang[4], Yong Sheng Zhao[3], Volker Blum[2
& Letian Dou[1,4,11]

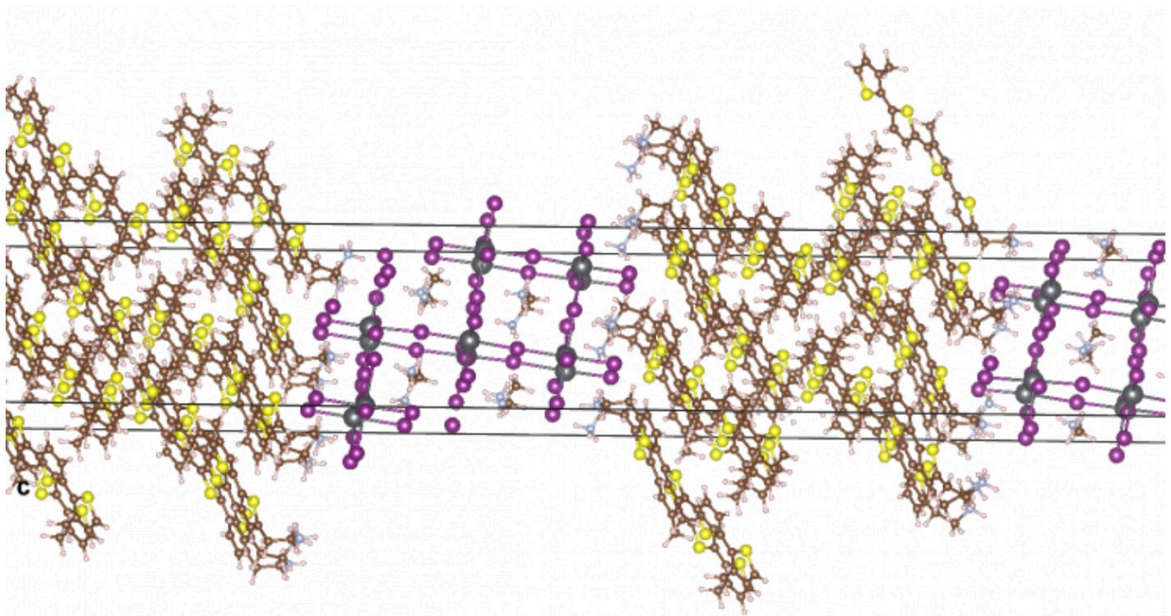
Two-dimensional organic semiconductor-incorporated perovskites are
promising family of hybrid materials for optoelectronic applications, o
in part to their inherent quantum well architecture. Tuning their structu
and properties for specific properties, however, has remained challeng
Here we report a general method to tune the dimensionality of phase-p
organic semiconductor-incorporated perovskite single crystals during
their synthesis, by judicious choice of solvent. The length of the conjug

Home » About » News & Media »

Growing Triple-Decker Hybrid Crystals for Lasers

AUGUST 31, 2023

Collaboration between Duke and Purdue explores the fabrication and optoelectric characterization of multilayered perovskites



Spin-orbit coupled DFT-HSE06 band structures + energy level alignments: 9 single-crystal compounds, up to 952 atoms / unit cell, all electron. Experimentally realized + theory.

What Can We Do?

GW *et al.* for Molecules - Early Success

New Journal of Physics
The open-access journal for physics

Resolution-of-identity approach to Hartree–Fock, hybrid density functionals, RPA, MP2 and *GW* with numeric atom-centered orbital basis functions

Xinguo Ren[1], Patrick Rinke, Volker Blum, Jürgen Wieferink, Alexandre Tkatchenko, Andrea Sanfilippo, Karsten Reuter[2] and Matthias Scheffler

What Can We Do?

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All-electron *ab initio* Bethe-Salpeter equation approach to neutral excitations in molecules with numeric atom-centered orbitals

Cite as: J. Chem. Phys. **152**, 044105 (2020); https://doi.org/10.1063/1.5123290
Submitted: 04 August 2019 . Accepted: 12 November 2019 . Published Online: 24 January 2020

Chi Liu, Jan Kloppenburg, Yi Yao, Xinguo Ren, Heiko Appel, Yosuke Kanai, and Volker Blum

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JCTC Journal of Chemical Theory and Computation

pubs.acs.org/JCTC Article

All-Electron BSE@*GW* Method for *K*-Edge Core Electron Excitation Energies

Yi Yao, Dorothea Golze,* Patrick Rinke,* Volker Blum,* and Yosuke Kanai*

Cite This: *J. Chem. Theory Comput.* 2022, 18, 1569–1583 Read Online

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https://pubs.acs.org/doi/10.1021/acs.jctc.1c01180?fig=fig4&ref=pdf

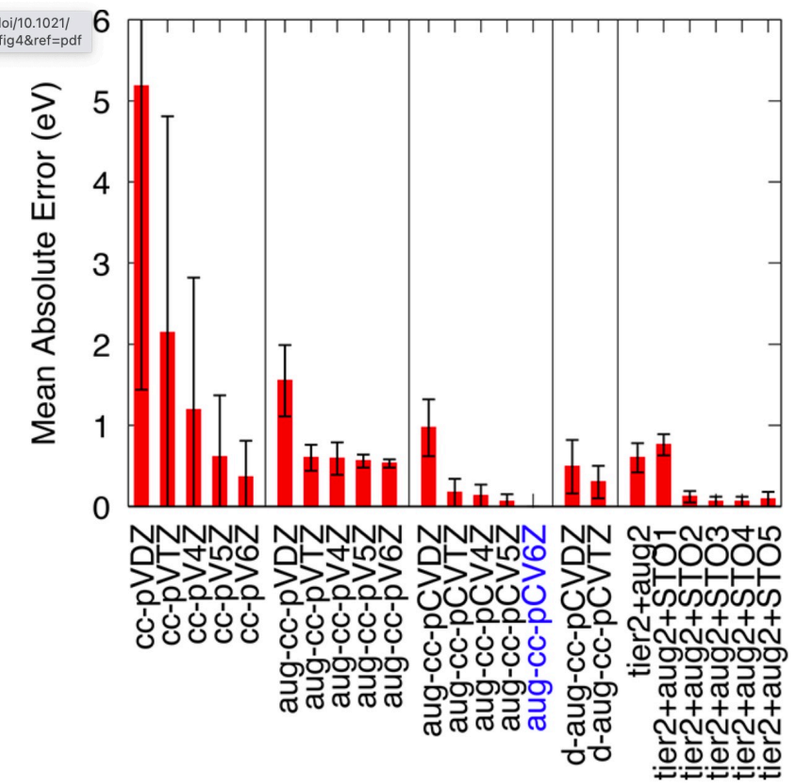

Figure 4. Basis set convergence of the mean absolute error (MAE) in eV for the BSE@*GW* method (with the aZORA+RC relativistic correction) on the 31 *K*-edge benchmark test set. The aug-cc-pCV6Z GTO basis set was used as the reference and is displayed in blue. The black error bars indicate the standard deviation.

JCTC Journal of Chemical Theory and Computation
pubs.acs.org/JCTC Article

All-Electron BSE@*GW* Method for *K*-Edge Core Electron Excitation Energies

Yi Yao, Dorothea Golze,* Patrick Rinke,* Volker Blum,* and Yosuke Kanai*

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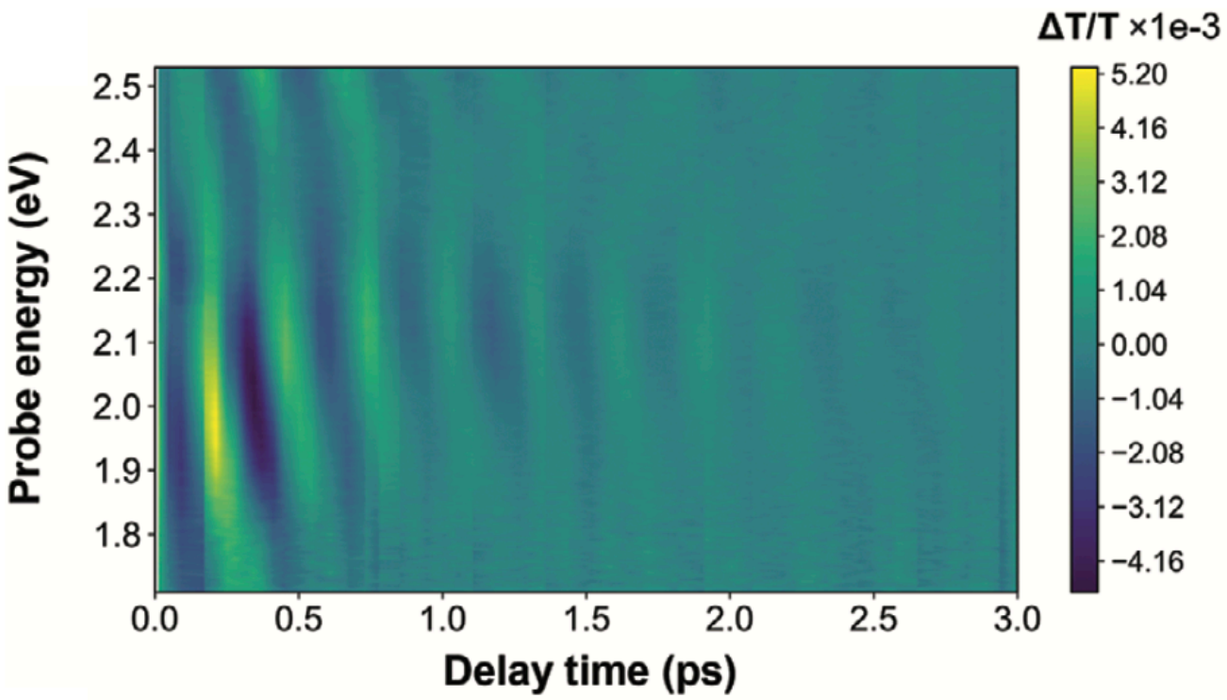
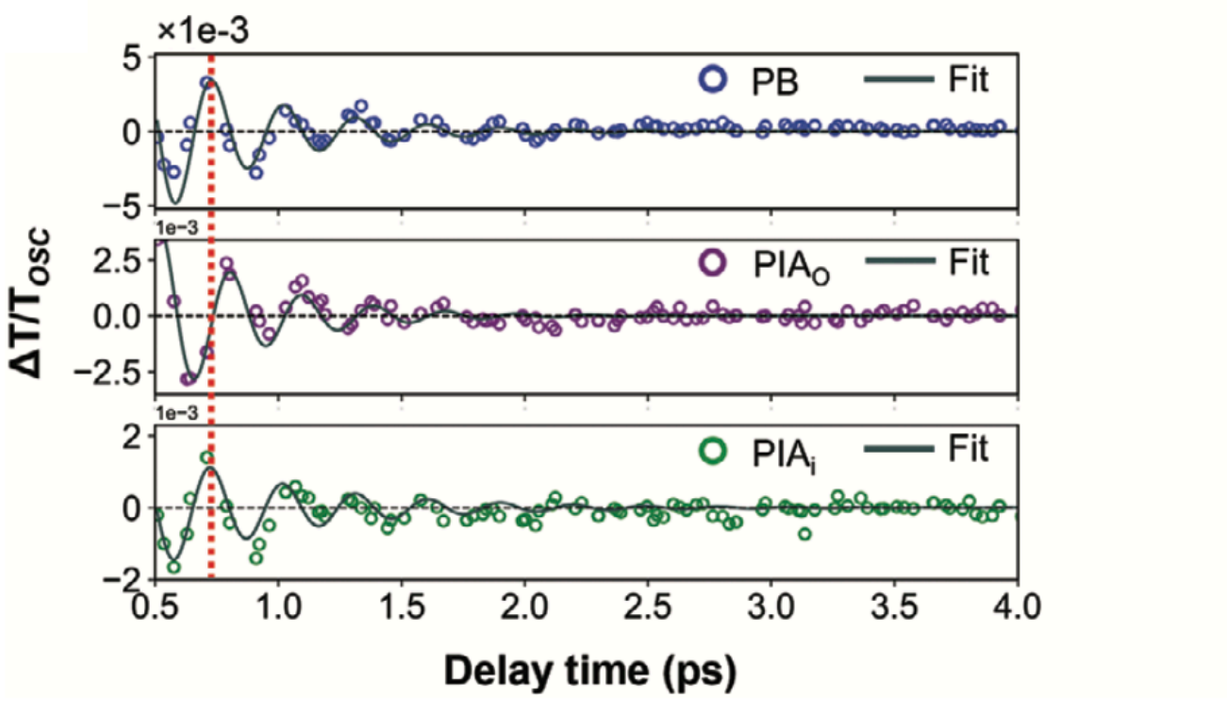
Electron-Phonon Mediated Coherent Charge Oscillations

Seyitleyev, Qin et al., Adv. Funct. Mater. 2023, 33, 2213021.





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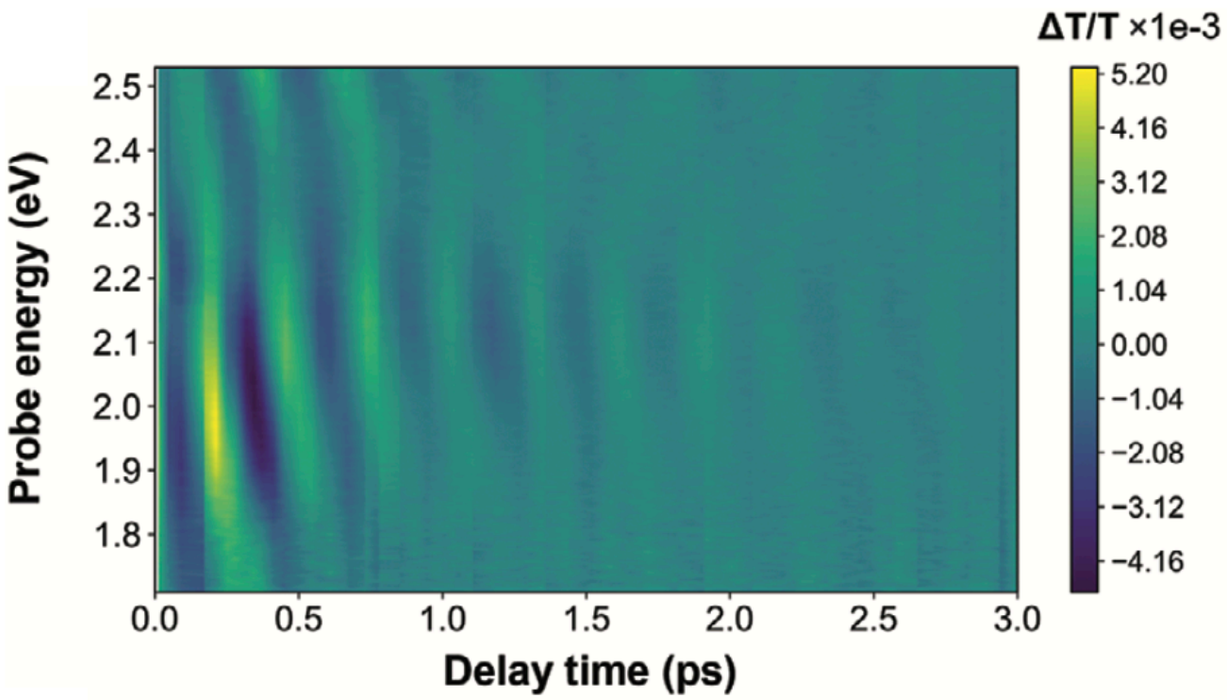

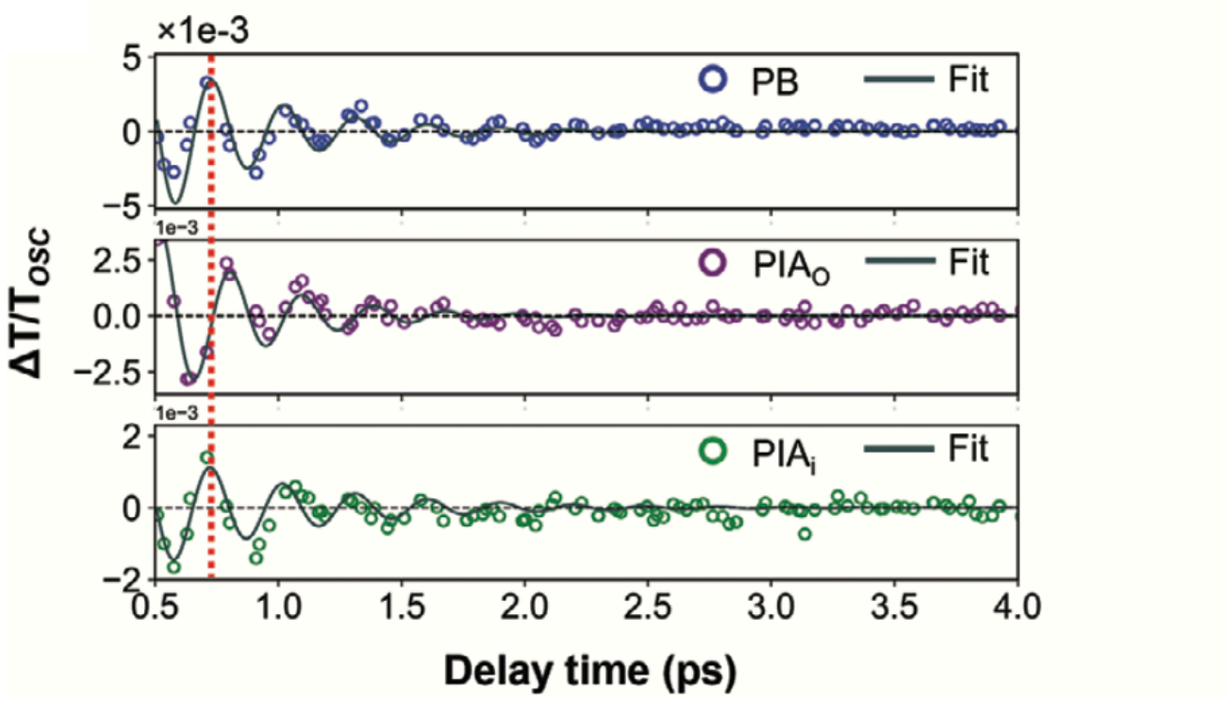

<u>Long Story Short</u>:

Identified a phonon mode that coherently transfers charge between organic, inorganic upon initial laser stimulation

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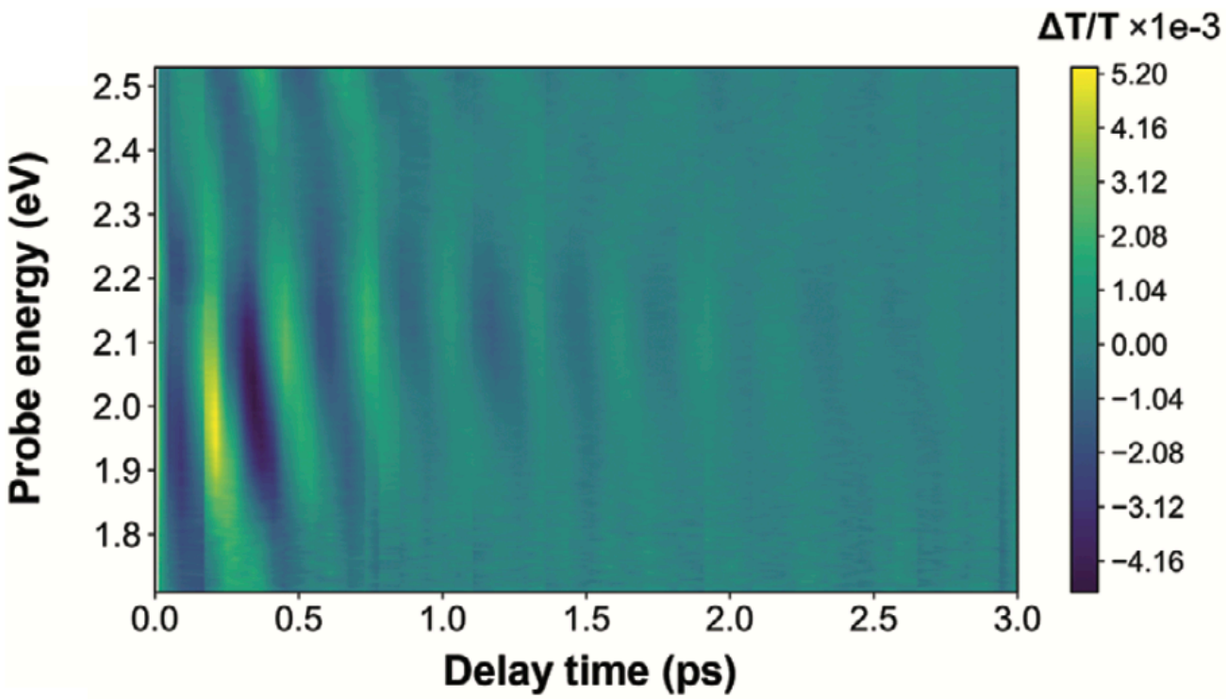

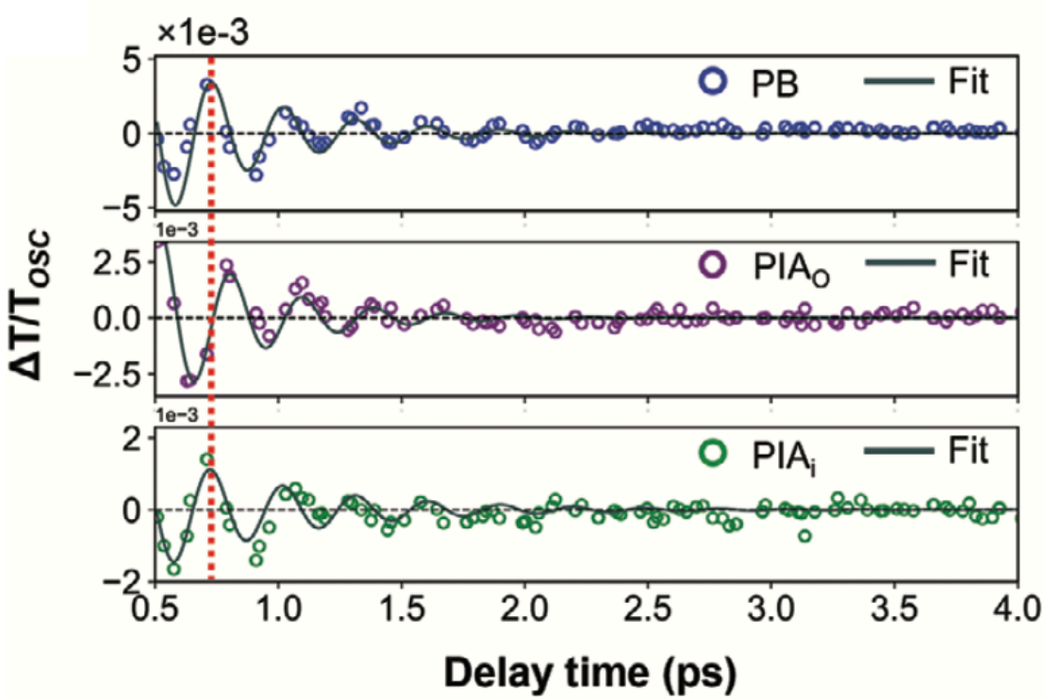

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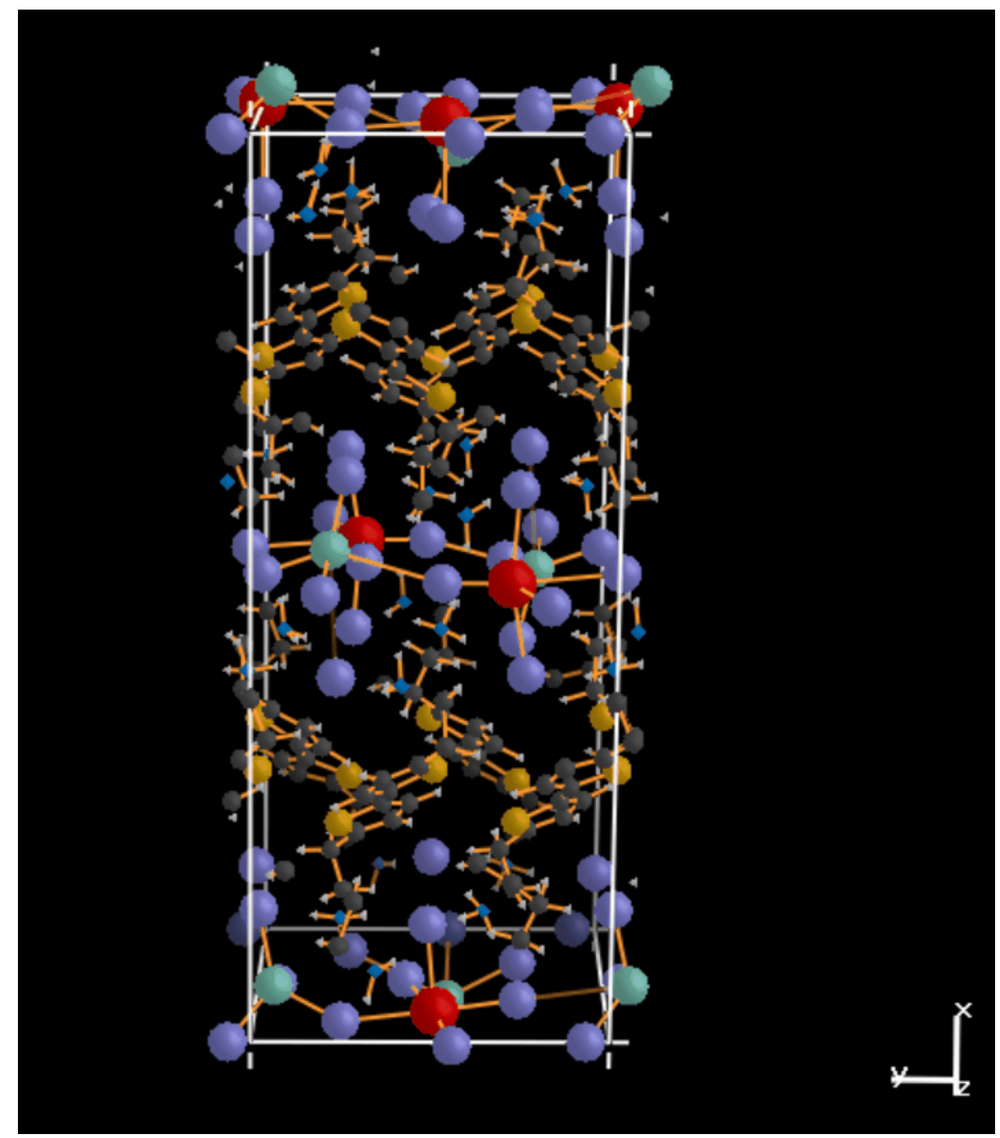
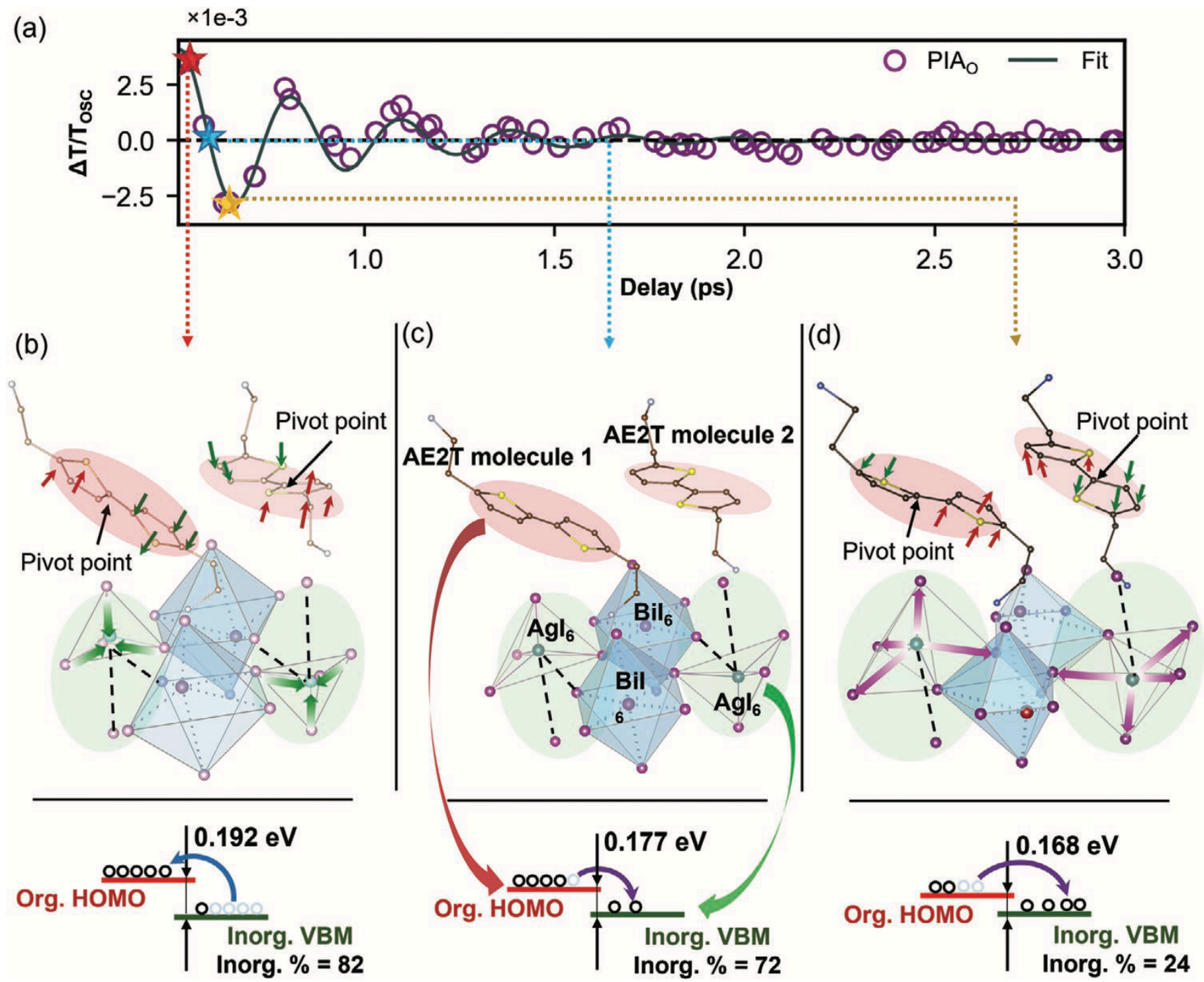


Coherent, Laser-Controlled Charge Oscillations in $(AE2T)_2AgBiI_8$

Seyitleyev, Qin et al., Adv. Funct. Mater. 2023, 33, 2213021.



Example - Bi-Doping of Layered HOIS, Hybrid DFT

Predicted Observable	High-precision atomic structure, energies and energy differences	Energy band structures for large, complex systems

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Predicted Observable	High-precision atomic structure, energies and energy differences	Energy band structures for large, complex systems
Level of Theory	Semilocal density functional theory with van der Waals corrections	Hybrid density functional theory including spin-orbit coupling

U.S. DEPARTMENT OF ENERGY | Office of Science

CHOISE Center for Hybrid Organic Inorganic Semiconductors for Energy

MAX-PLANCK-GESELLSCHAFT

NSF

XSEDE Extreme Science and Engineering Discovery Environment

Example - Bi-Doping of Layered HOIS, Hybrid DFT

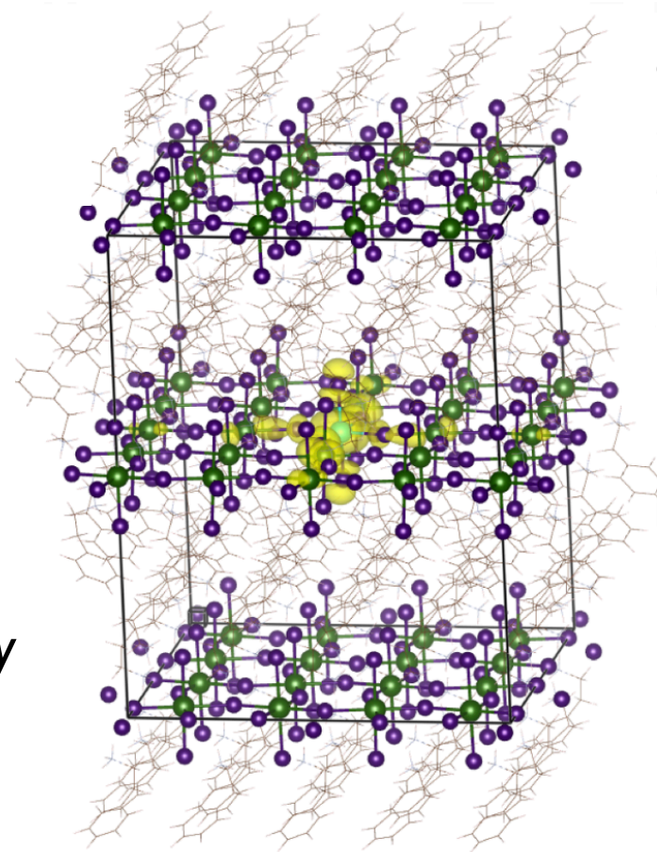
Predicted Observable	High-precision atomic structure, energies and energy differences	Energy band structures for large, complex systems
Level of Theory	Semilocal density functional theory with van der Waals corrections	Hybrid density functional theory including spin-orbit coupling

Dr. Yi Yao

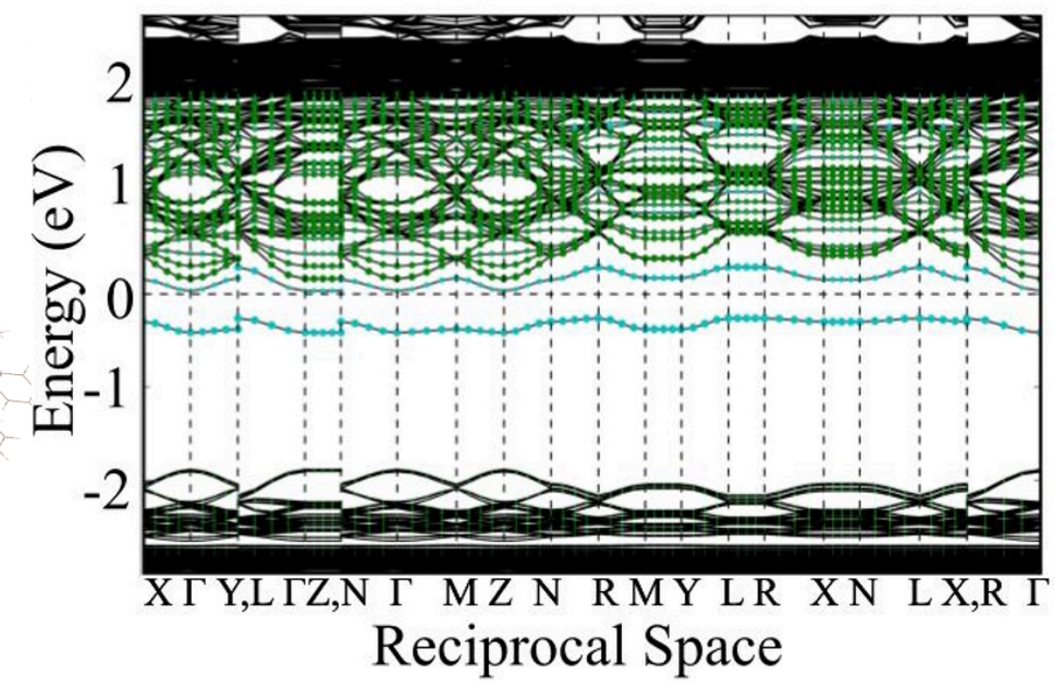
Gabrielle Koknat

Bi-doped $(PEA)_2PbI_4$
1,504 atoms
HSE06+SOC,
all-electron
High precision -
no tricks!

Lu et al., PRX Energy 2, 023010 (2023).



DFT-HSE06+SOC Band Structure



U.S. DEPARTMENT OF ENERGY | Office of Science

CHOISE
Center for Hybrid Organic Inorganic Semiconductors for Energy

MAX-PLANCK-GESELLSCHAFT

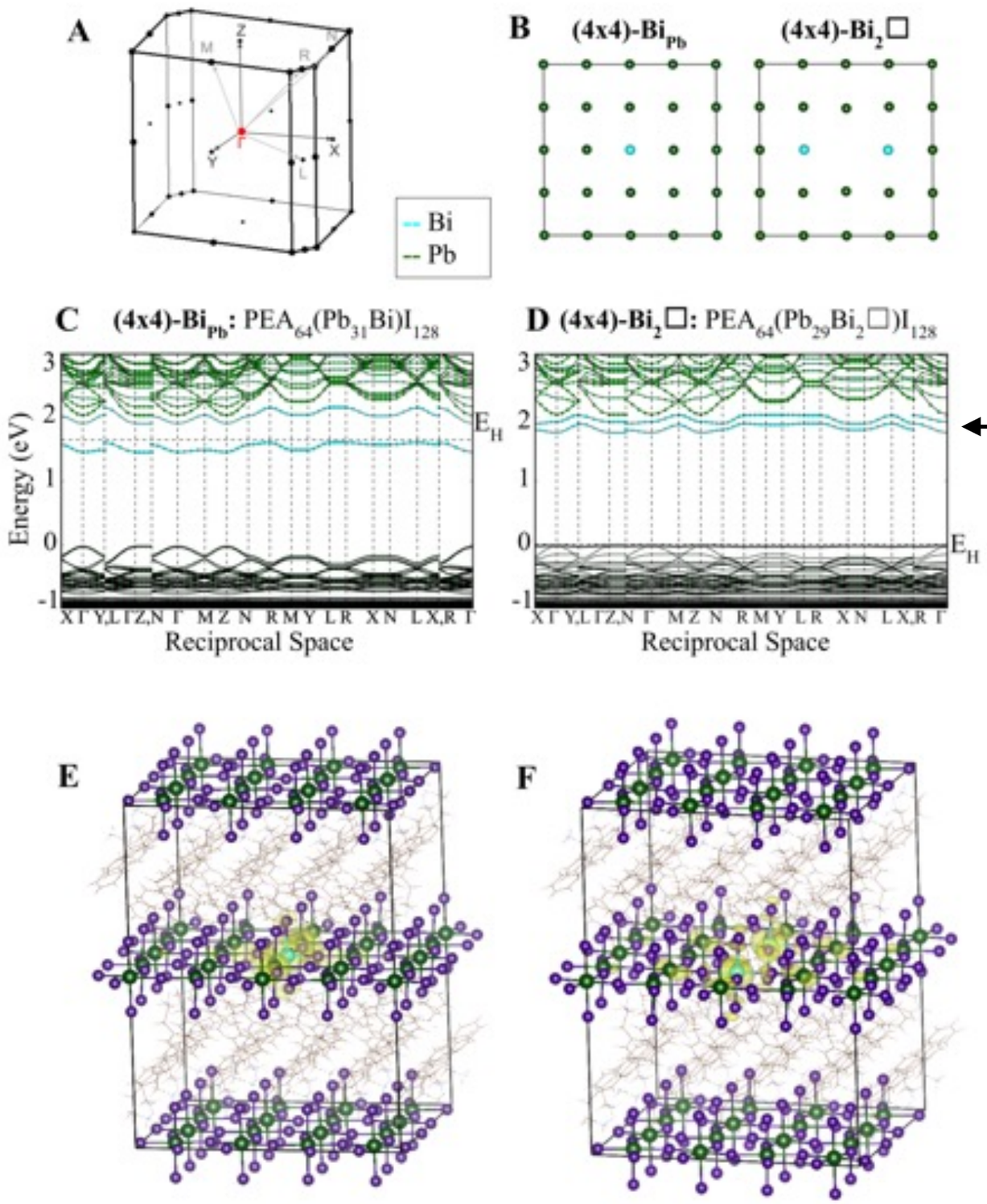
NSF

XSEDE
Extreme Science and Engineering Discovery Environment

So Are We There Yet?

HSE06+SOC. No tricks.

Lu, Koknat et al., PRX Energy 2, 023010 (2023).



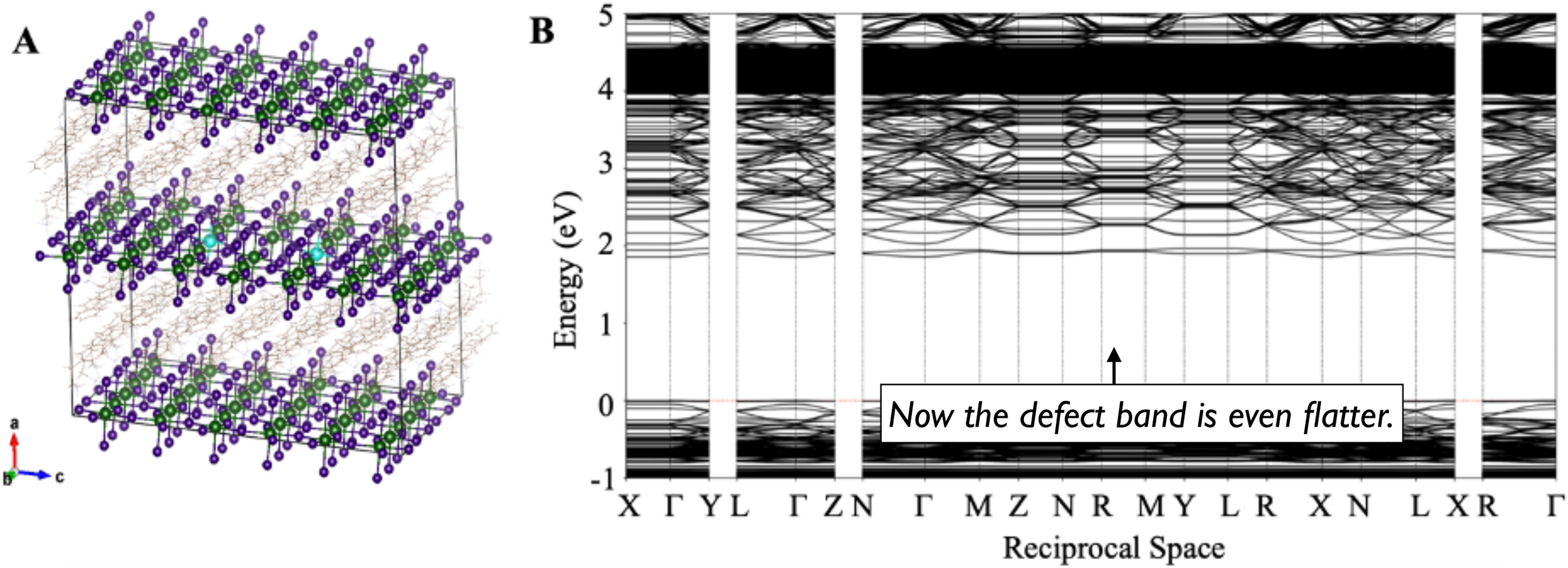
Anonymous reviewer:

"Finally, I actually worry that in some cases the supercell is not large enough despite its large number of atoms. In particular, for the Bi2 case shown in Fig. 3B, within the utilized supercell the Bi atoms are actually second-nearest neighbors with each other, likely indicating that the supercell is too small. A hint of this can be seen in Fig. 3D, which shows that the defect orbitals are not flat as expected but show some dispersion."

So How About 3,383 Atoms.

Lu, Koknat et al., PRX Energy 2, 023010 (2023).

HSE06+SOC. Still no tricks.



Electronic structure of the (6x6)-Bi_2□ structure.

3,383 atoms, 30,736 electrons, 75,100 basis functions, 24,076 KS orbitals (closed shell system).
3x3x3 s.c.f. k-space grid, i.e., Born-von Karman cell.
12,096 Intel IceLake cores. SCF: 1 EXX matrix evaluation - 490s, 14 Eigenvalue problems - 225s

Central Decision: Basis Set

$$\left[-\frac{\nabla^2}{2} + v_{\text{ext}}(\boldsymbol{r}) + v_{\text{es}}(\boldsymbol{r}) + v_{\text{xc}}(\boldsymbol{r})\right]\psi_k(\boldsymbol{r}) = \epsilon_k \psi_k(\boldsymbol{r})$$

Kohn, Sham 1965

"Basis set expansion":

$$\psi_k(\boldsymbol{r}) = \sum_i c_{ki}\varphi_i(\boldsymbol{r})$$

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Generalized eigenvalue problem:

$$\boxed{\underline{\underline{h}}\,\underline{c}_k = \epsilon_k\,\underline{\underline{s}}\,\underline{c}_k}$$

$$h_{ij} = \langle\varphi_i|\hat{h}_{\text{KS}}|\varphi_j\rangle$$

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<u>Many good options:</u>

- Plane waves (*VASP*, *abinit*, ...)

$$\varphi_{\underline{k}}(\boldsymbol{r}) = \frac{1}{N}e^{i\underline{k}\underline{r}}$$

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$$\varphi_{\underline{k}}(\boldsymbol{r}) = \frac{1}{N}e^{i\underline{k}\underline{r}}$$

- Gaussian-type orbitals

$$\varphi_{lmn}(\boldsymbol{r}) = \frac{1}{N}x^l y^m z^n e^{-\alpha r^2}$$

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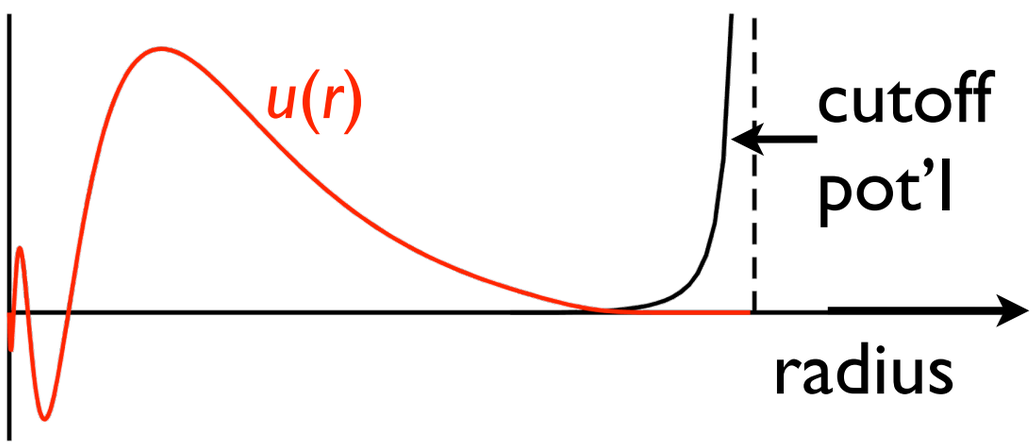
- <u>*Many*</u> others: "Augmented plane waves", wavelets, finite elements, numeric atom-centered functions, ...

Our choice!

$$\varphi_{i[lm]}(\boldsymbol{r}) = \frac{u_i(r)}{r}\cdot Y_{lm}(\Omega)$$

Numeric Atom-Centered Basis Sets

$$\varphi_{i[lm]}(\boldsymbol{r}) = \frac{u_i(r)}{r} \cdot Y_{lm}(\Omega)$$



- Flexible shape
- "Naturally" all-electron
- Strictly localizable - *O*(*N*) computational scaling for most expensive steps
- Rather compact basis sets for converged results

<u>Many popular implementations:</u>
DMol³ (Delley), FPLO (Eschrig *et al.*) PLATO (Horsfield *et al.*), ADF, BDF, PAOs (Siesta, Conquest, OpenMX[2], Fireball, ...)

We have a basis set library for all elements (1-102), from fast qualitative to meV-converged calculations. (total energy, DFT).

Blum, Gehrke, Hanke, Havu, Havu, Ren, Reuter, Scheffler, Computer Physics Communications ***180****, 2175 (2009)*

Constructing a Basis Set Library for DFT

Goal: Element-dependent, *transferable* basis sets
from fast qualitative to meV-converged total energy accuracy (ground-state DFT)

Can't we have the computer pick
good basis sets for us?

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Occupied free
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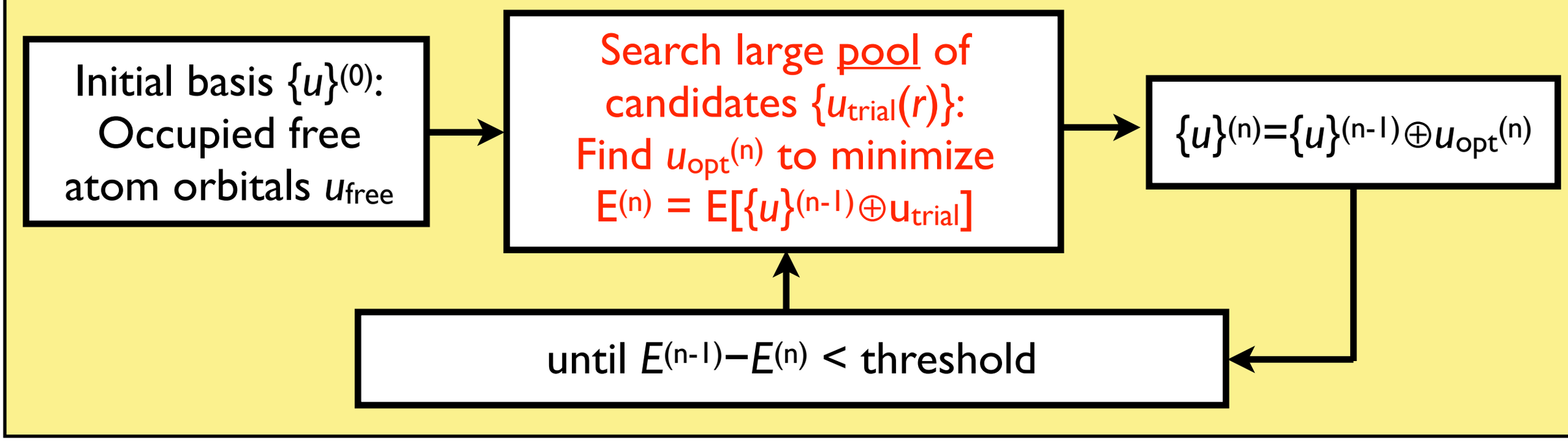

Blum et al., Comp. Phys. Commun. ***180****, 2175-2196 (2009)*

Iterative Selection of NAO Basis Functions

"Pool" of trial basis functions:
2+ ionic *u(r)*
Hydrogen-like *u(r)* for *z*=0.1-20

Optimization target:
Non-selfconsistent symmetric dimers, averaged for different *d*

Pick basis functions one by one, up to complete *total energy* convergence

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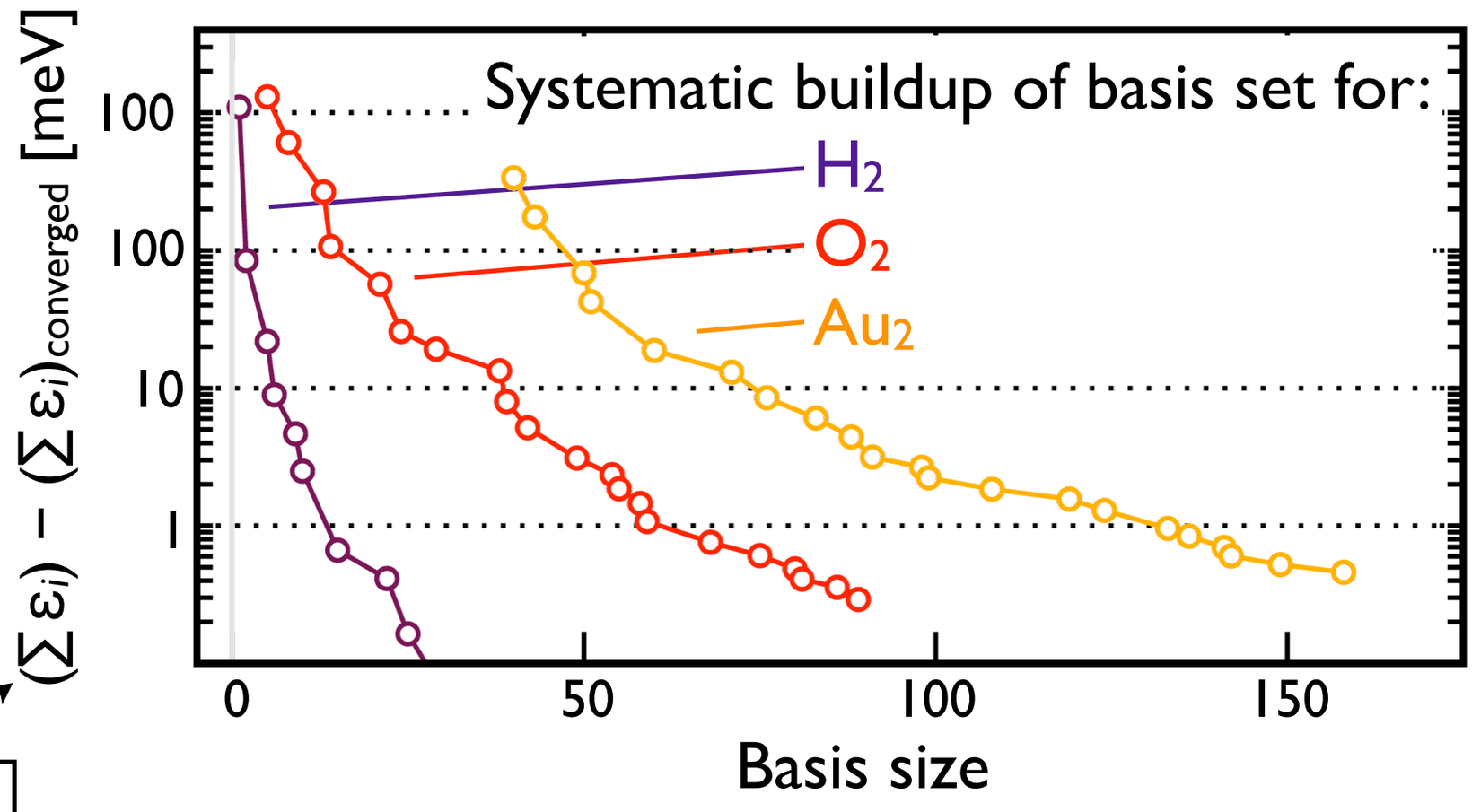
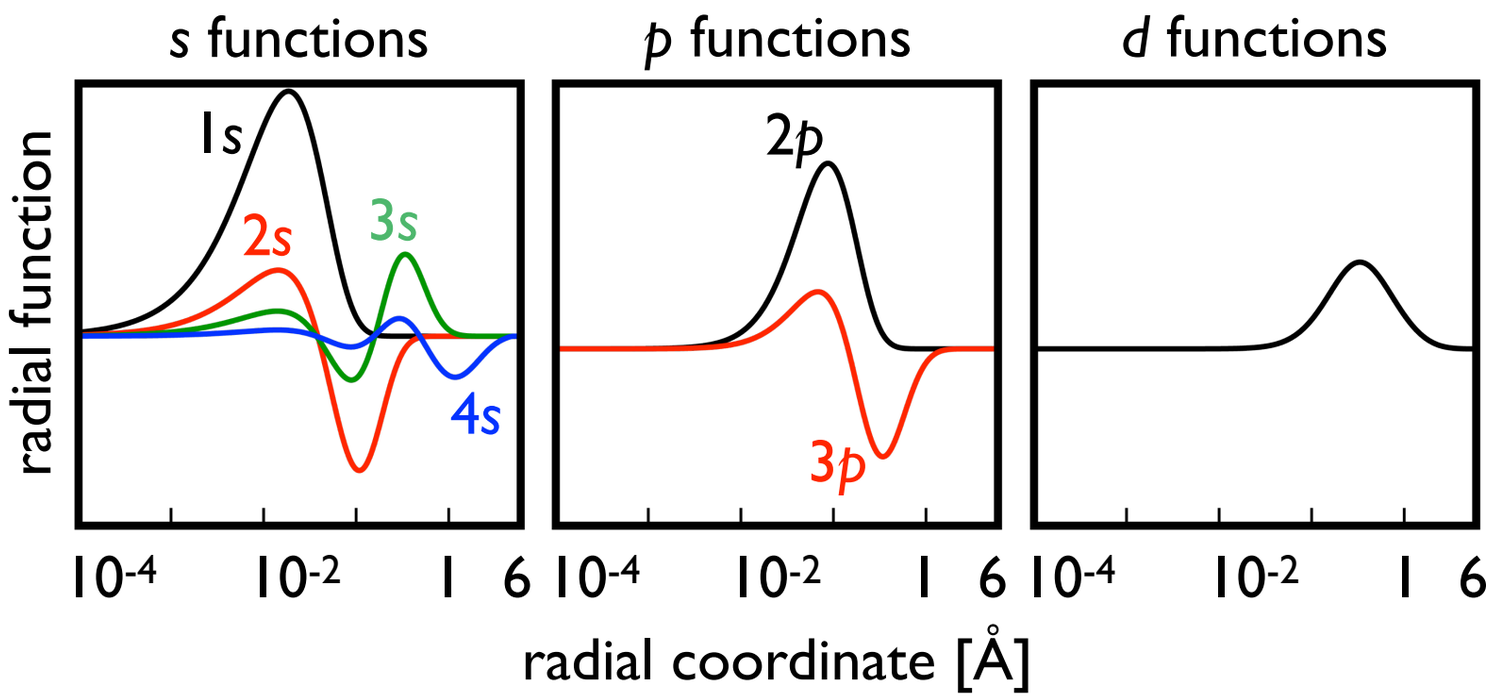


Remaining basis set error

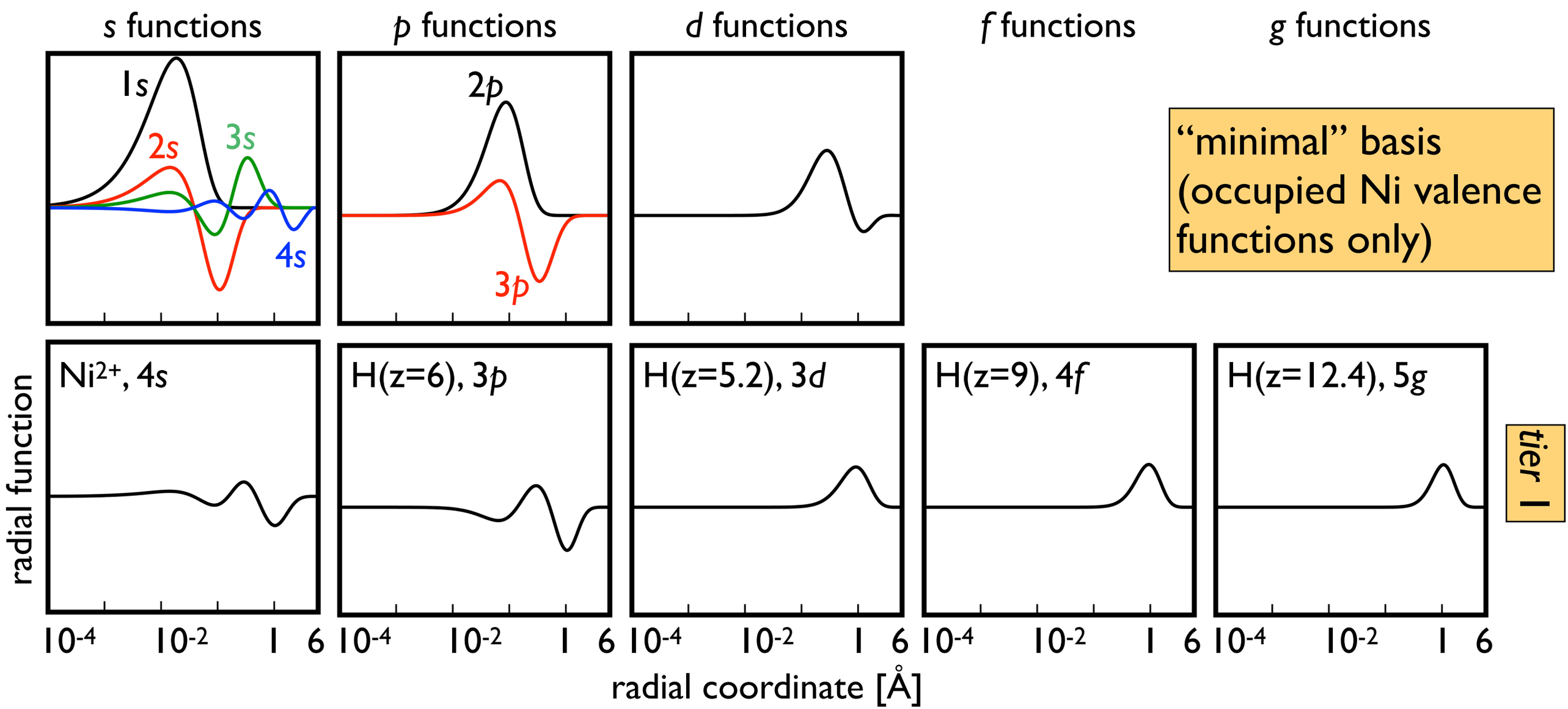
Blum et al., Comp. Phys. Commun. ***180****, 2175-2196 (2009)*

Ni radial functions in practice - "*tier*" by "*tier*"



"minimal" basis (occupied Ni valence functions only)

Ni radial functions in practice - "*tier*" by "*tier*"



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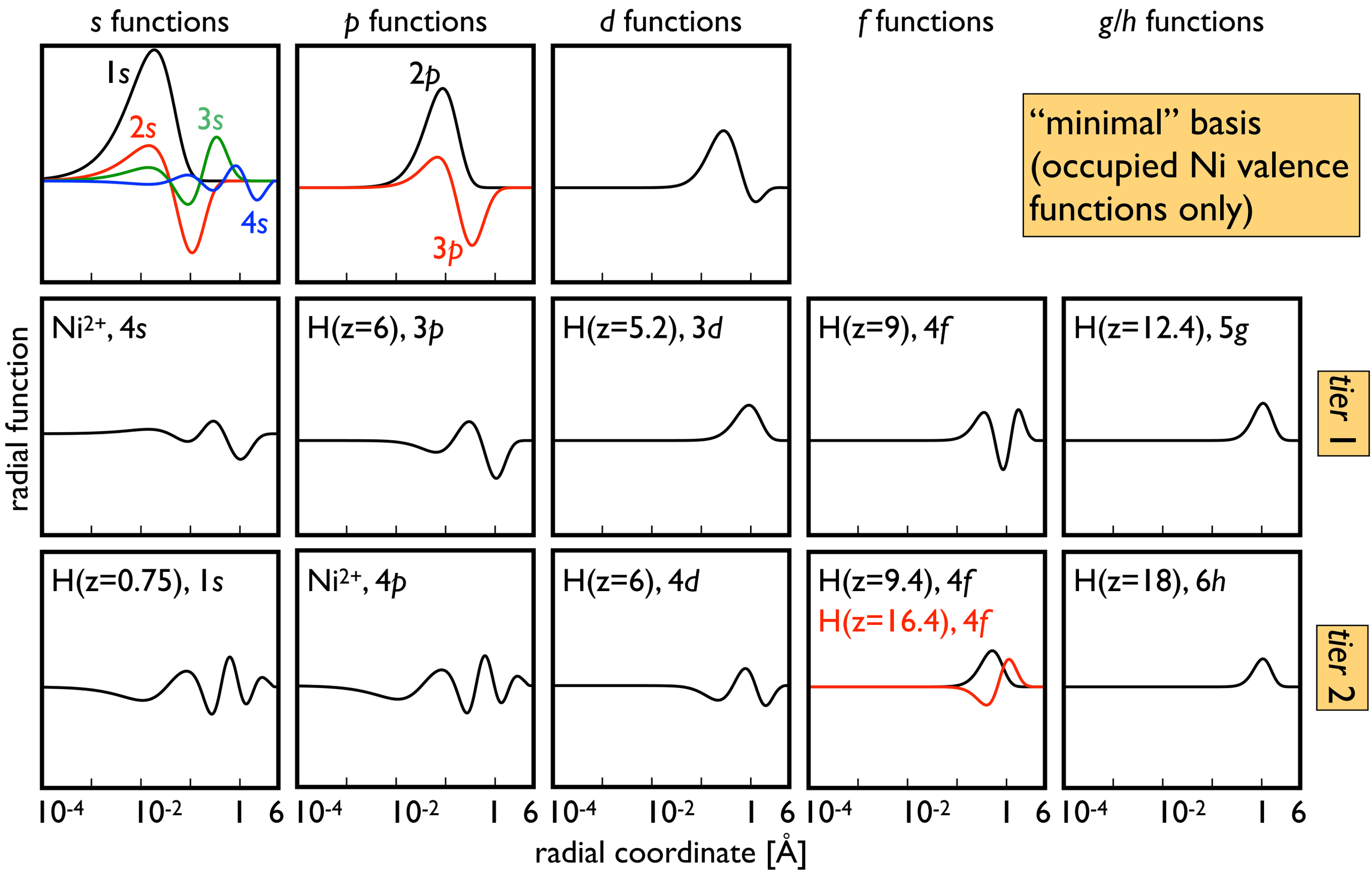

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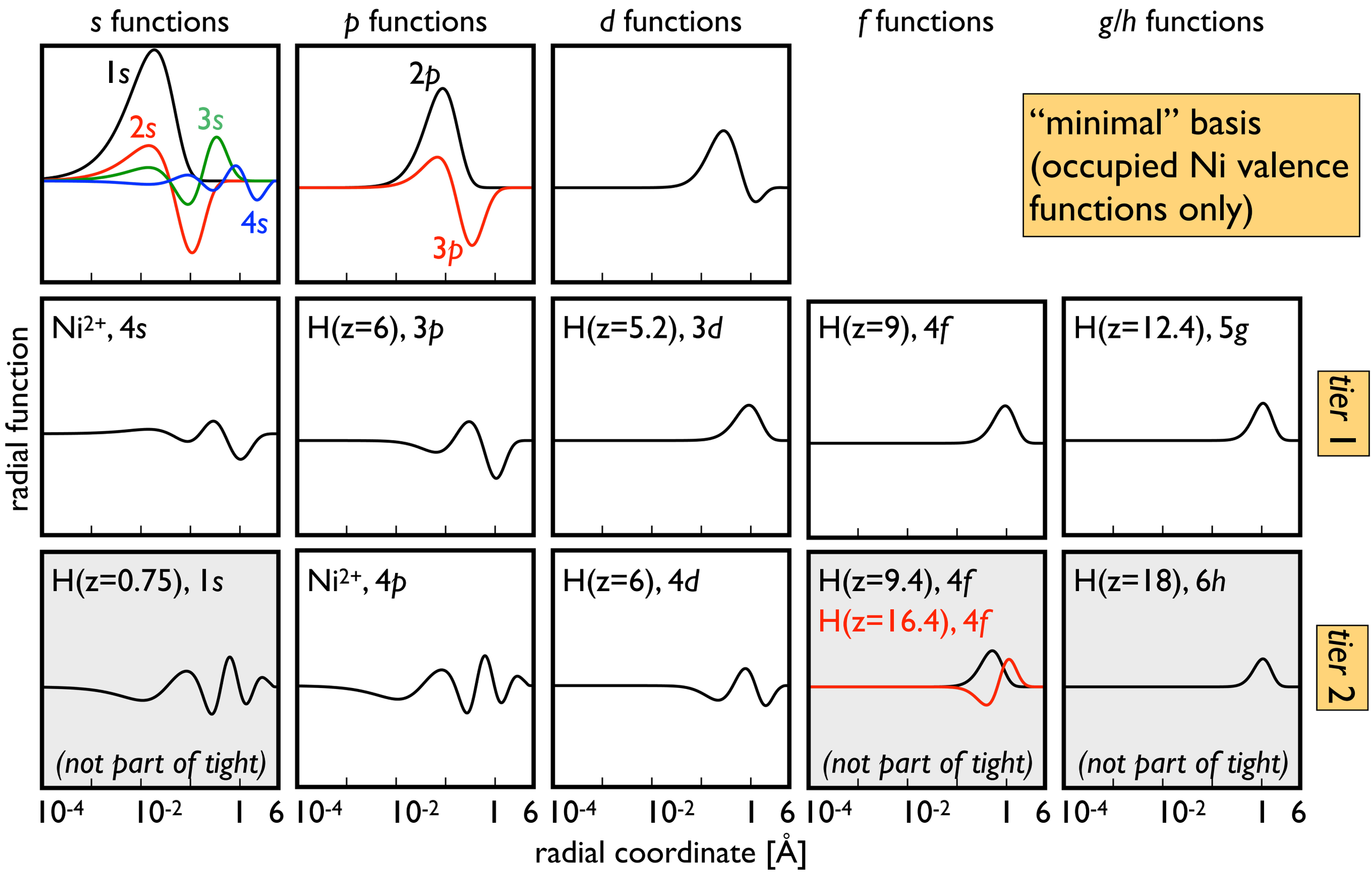

Outcome: Hierarchical Basis Set Library for Elements 1-102

	H	C	O	Au
minimal	$1s$	[He]+$2s2p$	[He]+$2s2p$	[Xe]+$6s5d4f$
Tier 1	H($2s$,2.1)	H($2p$,1.7)	H($2p$,1.8)	Au^{2+}(6p)
	H($2p$,3.5)	H($3d$,6.0)	H($3d$,7.6)	H($4f$,7.4)
		H($2s$,4.9)	H($3s$,6.4)	Au^{2+}(6s)
				H($5g$,10)
				H($6h$,12.8)
				H($3d$,2.5)
Tier 2	H($1s$,0.85)	H($4f$,9.8)	H($4f$,11.6)	H($5f$,14.8)
	H($2p$,3.7)	H($3p$,5.2)	H($3p$,6.2)	H($4d$,3.9)
	H($2s$,1.2)	H($3s$,4.3)	H($3d$,5.6)	H($3p$,3.3)
	H($3d$,7.0)	H($5g$,14.4)	H($5g$,17.6)	H($1s$,0.45)
		H($3d$,6.2)	H($1s$,0.75)	H($5g$,16.4)
				H($6h$,13.6)
Tier 3	H($4f$,11.2)	H($2p$,5.6)	O^{2+}($2p$)	H($4f$,5.2)*
	H($3p$,4.8)	H($2s$,1.4)	H($4f$,10.8)	H($4d$,5.0)
	...	...	...	...

Systematic hierarchy of basis (sub)sets, iterative *automated* construction based on *dimers*

"First *tier* (level)"

"Second *tier*"

"Third *tier*"

...

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		H($2s$,		
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		H($3d$,		
Tier 3	H($4f$,11.2)	H($2p$,		
	H($3p$,4.8)	H($2s$,		
	...	...		

Systematic hierarchy of basis (sub)sets, iterative *automated* construction

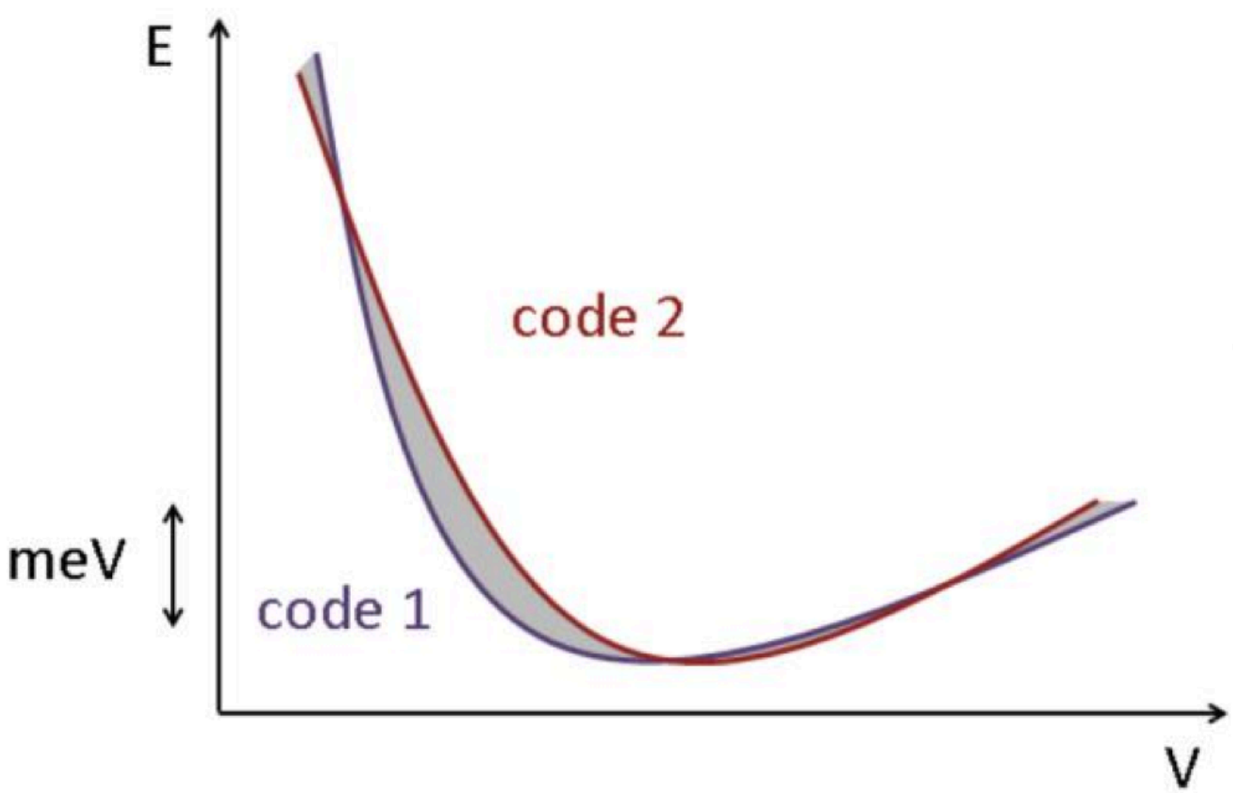
In FHI-aims input (example: Hydrogen, tight settings):

```
##########################################################################
#
#  Definition of "minimal" basis
#
##########################################################################
#     valence basis states
    valence      1  s   1.
#     ion occupancy
    ion_occ      1  s   0.5
##########################################################################
#
#  Suggested additional basis functions. For production calculations,
#  uncomment them one after another (the most important basis functions are
#  listed first).
#
#  Basis constructed for dimers: 0.5 A, 0.7 A, 1.0 A, 1.5 A, 2.5 A
#
##########################################################################
#  "First tier" - improvements: -1014.90 meV to -62.69 meV
     hydro 2 s 2.1
     hydro 2 p 3.5
#  "Second tier" - improvements: -12.89 meV to -1.83 meV
     hydro 1 s 0.85
     hydro 2 p 3.7
     hydro 2 s 1.2
     hydro 3 d 7
#  "Third tier" - improvements: -0.25 meV to -0.12 meV
#     hydro 4 f 11.2
#     hydro 3 p 4.8
#     hydro 4 d 9
#     hydro 3 s 3.2
##########################################################################
```

Precision in Community Wide Benchmark - "Delta Test"

Reproducibility in Density-Functional Calculations of Solids,
K. Lejaeghere, ... **68 coauthors!** ..., S. Cottenier,
Science **351**, aad3000 (2016).
https://molmod.ugent.be/deltacodesdft

E(V) for 71 elemental solids - 15 codes, all-electron & 40 pseudopot'l sets



→ $$\Delta_i(a,b) = \sqrt{\int_{0.94V_{0,i}}^{1.06V_{0,i}} \frac{\left(E_{b,i}(V) - E_{a,i}(V)\right)^2}{0.12V_{0,i}} \mathrm{d}V}$$

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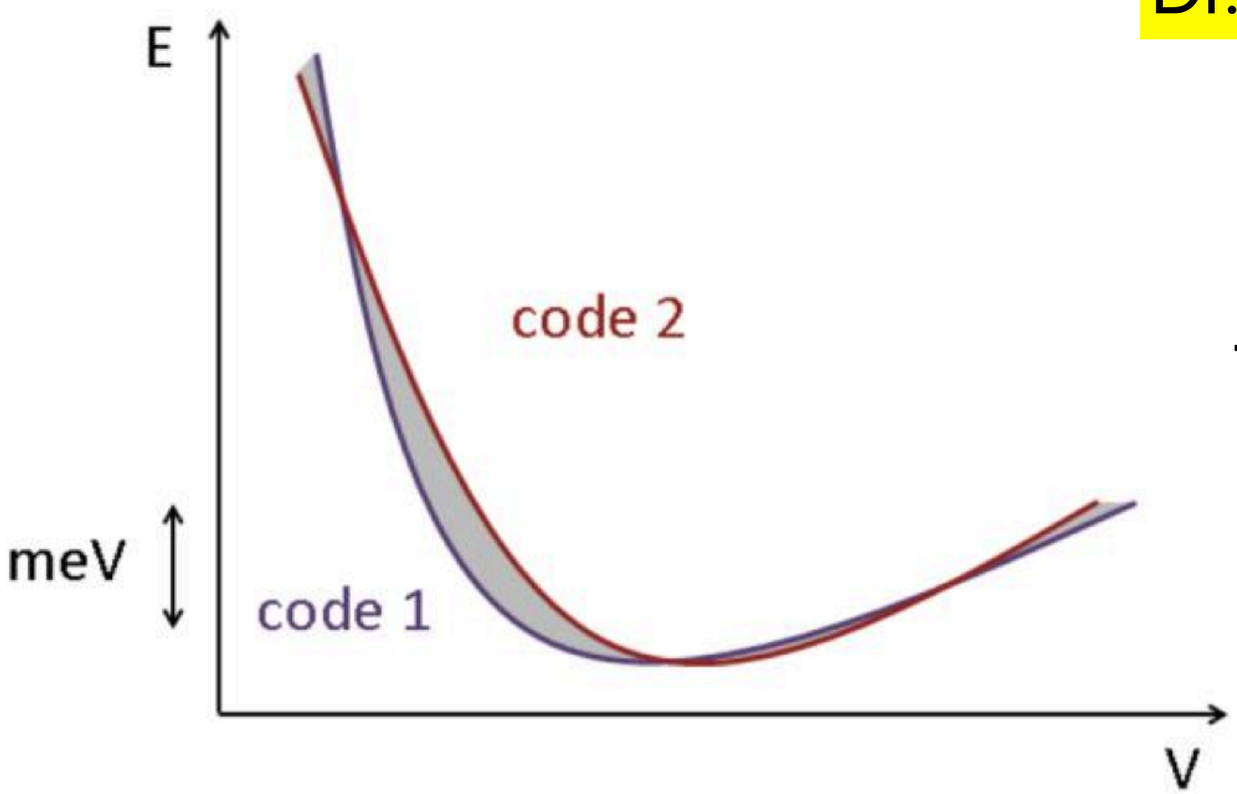
FHI-aims: Test carried out independently by Dr. Marcin Dulak, DTU (Copenhagen)



$$\rightarrow \quad \Delta_i(a,b) = \sqrt{\int_{0.94V_{0,i}}^{1.06V_{0,i}} \frac{\left(E_{b,i}(V) - E_{a,i}(V)\right)^2}{0.12V_{0,i}} \, \mathrm{d}V}$$

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Code	Basis	Electron treatment	Delta (meV)
Wien2k 13.1	LAPW/APW+lo	All-electron	0
FHI-aims 081213*	NAO, tier2	All-electron (scalar rel. atomic ZORA)	0.2
Exciting (dev.)	LAPW+xlo	All-electron	0.2
Quantum Espresso 5.1	plane waves	SSSP accuracy (mixed NC/US/PAW library)	0.3
VASP 5.2.12	plane waves	PAW 2015	0.3
FHI-aims 081213*	NAO, tier2	All-electron (scalar rel., scaled ZORA)	0.3
ELK 3.1.5	APW+lo	All-electron	0.3

...

**Results: Marcin Dulak, DTU (Copenhagen)*

(N)AO Basis Set Alternatives & Beyond Semilocal, Hybrid DFT

- Random-Phase Approximation, *GW* (Molecules, H-Ar)

  Valence Correlation Consistent NAO Basis Sets "NAO-VCC-nZ"

  Igor Ying Zhang, Xinguo Ren, Patrick Rinke, Volker Blum, and Matthias Scheffler, New Journal of Physics 15, 123033 (2013).

- Neutral Excitations: *GW*+BSE, LR-TDDFT, RT-TDDFT

  FHI-aims' DFT basis sets plus Dunning augmentation, "tier2+aug2"

  Chi Liu, Jan Kloppenburg, Yi Yao, Xinguo Ren, Heiko Appel, Yosuke Kanai, Volker Blum The Journal of Chemical Physics, 152, 044105 (2020).

  J Hekele, Y Yao, Y Kanai, V Blum, P Kratzer The Journal of Chemical Physics 155 (15), 154801

- Core-level Excitations: *GW*+BSE

  FHI-aims' DFT basis sets plus Dunning augmentation plus tight core, "tier2+aug2+STO3"

  Y Yao, D Golze, P Rinke, V Blum, Y Kanai Journal of Chemical Theory and Computation (2022), https://doi.org/10.1021/acs.jctc.1c01180

- Any existing Gaussian-type, Slater-type basis sets will work as well.

  E.g., MolSSI Basis Set Exchange - Susi Lehtola

Using Numeric Atom-Centered Basis Functions: Pieces

- *Numerical* Integration $$h_{ij} = \int d^3r \varphi_i(\boldsymbol{r}) \hat{h}_{\mathrm{KS}} \varphi_j(\boldsymbol{r})$$
- Electron density update $$n(\boldsymbol{r}) = \sum_k f_k |\psi_k(\boldsymbol{r})|^2$$
- All-electron electrostatics $$v_{\mathrm{es}}(\boldsymbol{r}) = \int d^3r' \frac{n(\boldsymbol{r}')}{|\boldsymbol{r} - \boldsymbol{r}'|}$$
- Relativity *needed for heavy elements*
- Eigenvalue solver $$\underline{\underline{h}}\, \underline{c}_k = \epsilon_k\, \underline{\underline{s}}\, \underline{c}_k$$
- Periodic systems $$\Psi_{n,\mathbf{k}} = \exp(i\mathbf{k}\mathbf{r}) \cdot u_{n,\mathbf{k}}(\mathbf{r})$$
- Coulomb operator $$(ij|kl) = \int d^3r d^3r' \frac{\varphi_i(\boldsymbol{r})\varphi_j(\boldsymbol{r}')\varphi_k(\boldsymbol{r})\varphi_l(\boldsymbol{r}')}{|\boldsymbol{r} - \boldsymbol{r}'|}$$

Numeric Atom-Centered Basis Functions: Integration

$$h_{ij} = \int d^3r \varphi_i(\boldsymbol{r}) \hat{h}_{\mathrm{KS}} \varphi_j(\boldsymbol{r})$$

- *Discretize* to integration grid: $\int d^3r f(\boldsymbol{r}) \rightarrow \sum_{\boldsymbol{r}} w(\boldsymbol{r}) f(\boldsymbol{r})$

... but even-spaced integration grids are *out*:
f(*r*) strongly peaked near all nuclei!

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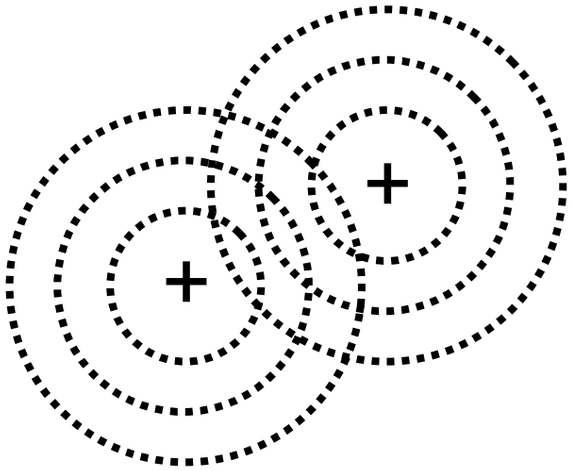
f(*r*) strongly peaked near all nuclei!

- Overlapping atom-centered integration grids:
  - Radial *shells (e.g., H, light: 24; Au, tight: 147)*
  - *Specific* angular point distribution ("Lebedev") exact up to given integration order *l* *(50, 110, 194, 302, ... points per shell)*



<u>Pioneered by</u>
Becke JCP 88, 2547 (1988), Delley, JCP 92, 508 (1990), MANY others!

Numeric Atom-Centered Basis Functions: Integration

$$h_{ij} = \int d^3r \varphi_i(\boldsymbol{r}) \hat{h}_{\mathrm{K}}$$

In FHI-aims input (example: Hydrogen, tight settings):

```
#
    radial_base          24 7.0
    radial_multiplier    2
    angular_grids        specified
      division   0.1930   50
      division   0.3175  110
      division   0.4293  194
      division   0.5066  302
      division   0.5626  434
#      division   0.5922  590
#      division   0.6227  974
#      division   0.6868 1202
#      outer_grid  770
      outer_grid  434
```

- *Discretize* to integration grid: $\int d^3r f$

... but even-spaced integratio
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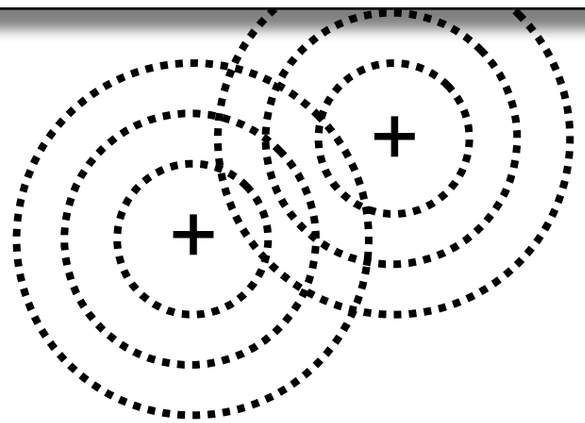

Pioneered by

Becke JCP 88, 2547 (1988), Delley, JCP 92, 508 (1990), MANY others!

Overlapping Atom-Centered Grids: "Partitioning of Unity"

Becke, 1988

$$h_{ij} = \int d^3r \varphi_i(\boldsymbol{r}) \hat{h}_{\mathrm{KS}} \varphi_j(\boldsymbol{r})$$

- Rewrite to atom-centered integrands:

$$\int d^3r f(\boldsymbol{r}) = \sum_{\mathrm{atoms}} \int d^3r p_{\mathrm{atom}}(\boldsymbol{r}) f(\boldsymbol{r})$$

exact: $\sum_{\mathrm{atoms}} p_{\mathrm{atom}}(\boldsymbol{r}) = 1$

through $p_{\mathrm{atom}}(\boldsymbol{r}) = \dfrac{g_{\mathrm{atom}}(\boldsymbol{r})}{\sum_{\mathrm{atom}'} g_{\mathrm{atom}'}(\boldsymbol{r})}$

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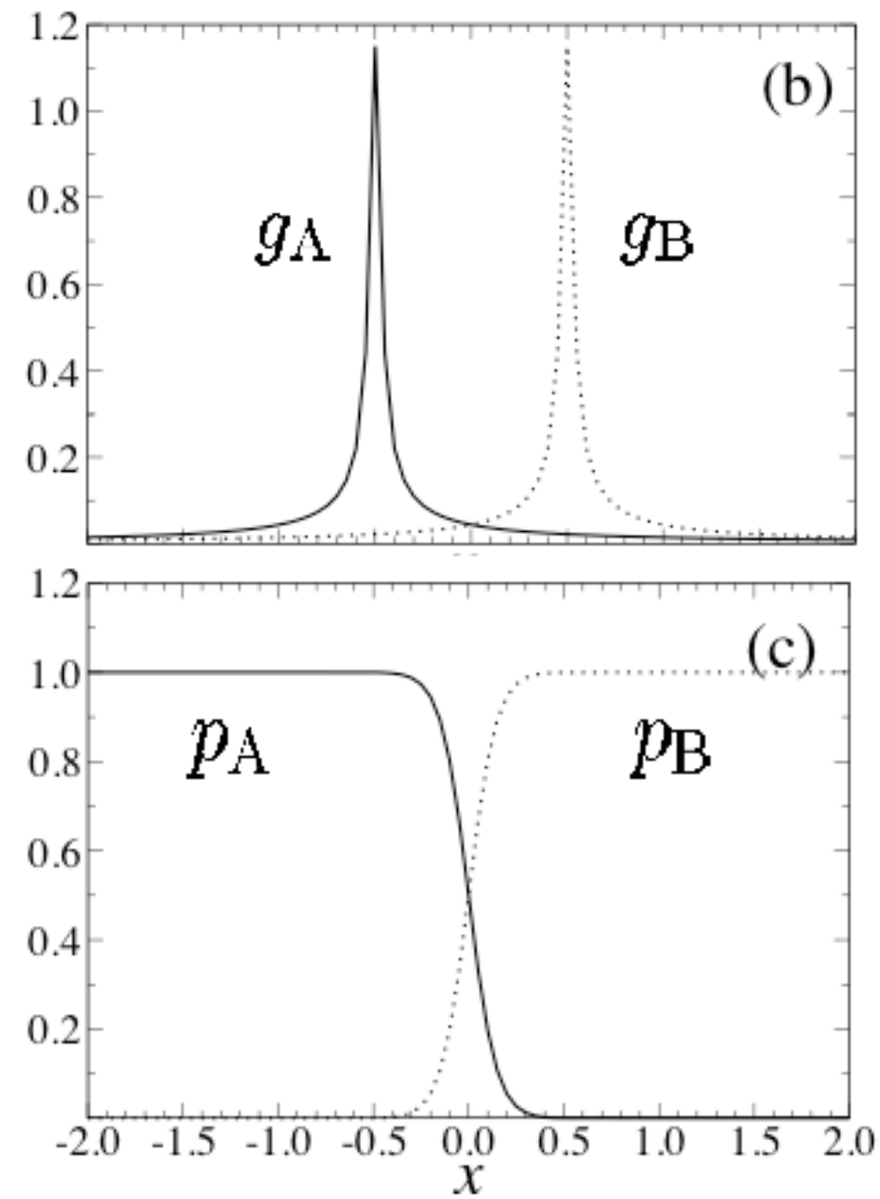
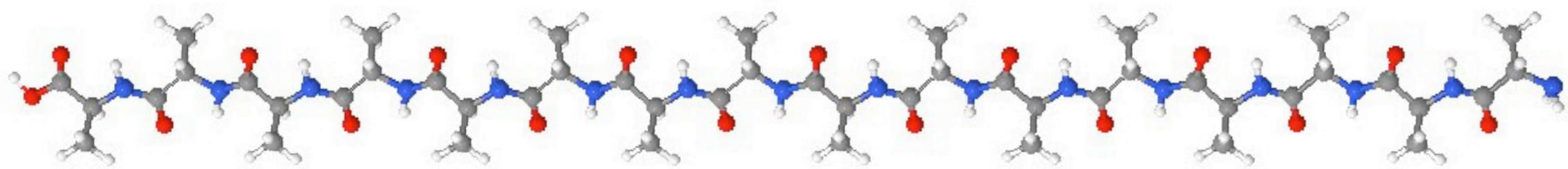
- e.g.: $g_{\text{atom}} = \dfrac{\rho_{\text{atom}}(r)}{r^2}$ *(Delley 1990)*

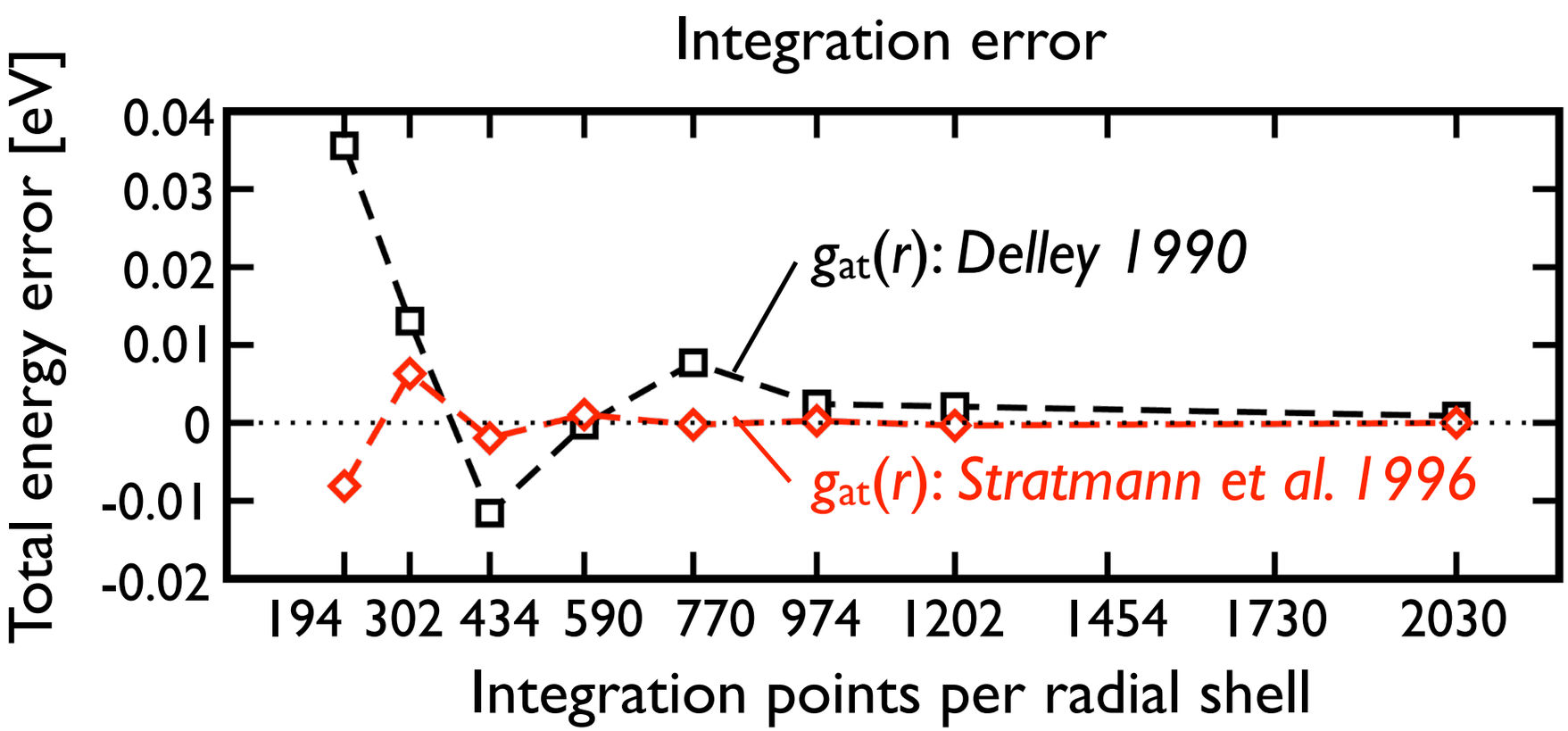
many alternatives:
Becke 1988, Stratmann 1996, Koepernik 1999, ...



Integration in Practice: Large Systems, Small Errors!



Fully extended Polyalanine peptide molecule Ala_{20}, DFT-PBE (203 atoms)



Hartree Potential (Electrostatics): Overlapping Multipoles

$$v_{\mathrm{es}}(\boldsymbol{r}) = \int d^3r' \frac{n(\boldsymbol{r}')}{|\boldsymbol{r} - \boldsymbol{r}'|}$$

Becke 1988
Delley 1990

- Partitioning of Unity:
  (same trick as used for integrals)

$$n(\boldsymbol{r}) = \sum_{\mathrm{atoms}} p_{\mathrm{atom}}(\boldsymbol{r}) n(\boldsymbol{r})$$

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- Multipole expansion:

$$n_{\mathrm{atom},lm}(r) = \int_{s=|r'-R_{\mathrm{atom}}|} p_{\mathrm{atom}}(\boldsymbol{r}') n(\boldsymbol{r}') Y_{lm}(\Omega)$$

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$$n_{\mathrm{atom},lm}(r) = \int_{s=|r'-R_{\mathrm{atom}}|} p_{\mathrm{atom}}(\boldsymbol{r}') n(\boldsymbol{r}') Y_{lm}(\Omega)$$

- Classical electrostatics:

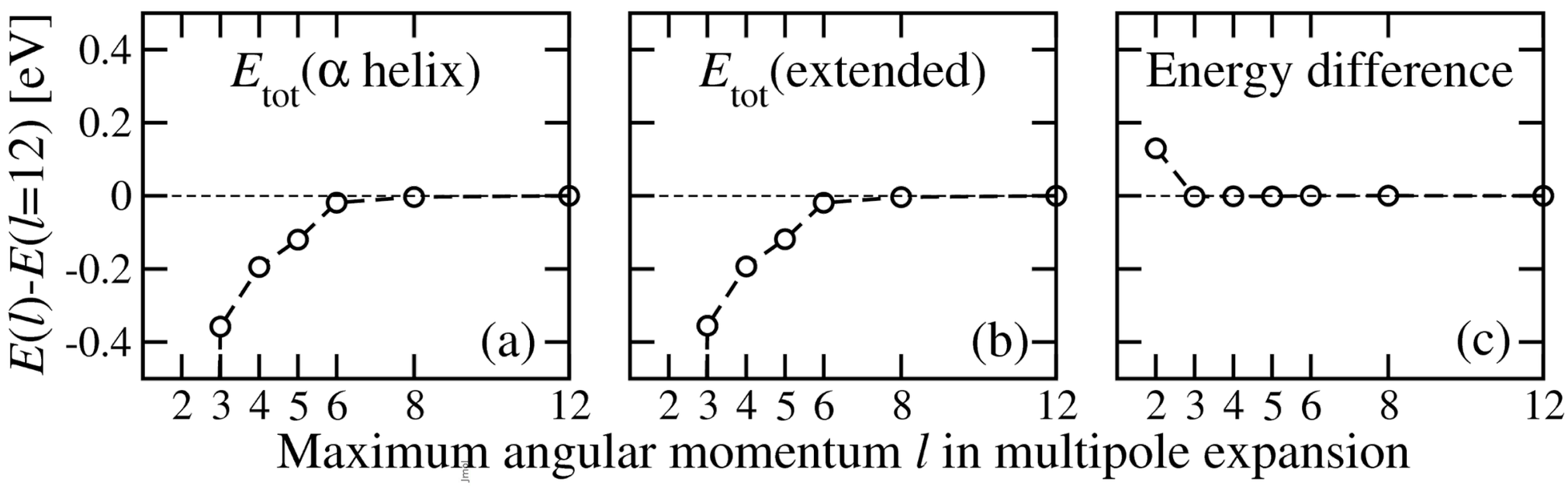
$$v_{\mathrm{es}}(\boldsymbol{r}) = \sum_{\mathrm{atoms}} \sum_{lm}^{l_{\mathrm{max}}} v_{\mathrm{atom},lm}(|\boldsymbol{r} - \boldsymbol{R}_{\mathrm{atom}}|) Y_{lm}(\Omega_{\mathrm{atom}})$$

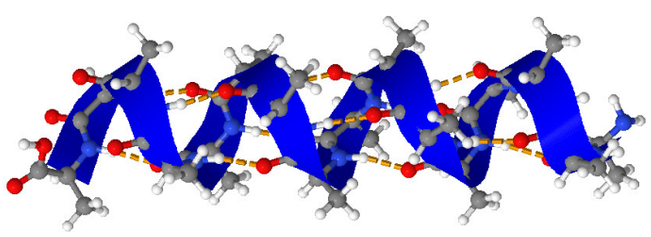
Electrostatics: Multipole expansion

$$v_{\mathrm{es}}(\boldsymbol{r}) = \sum_{\mathrm{atoms}} \sum_{lm}^{l_{\mathrm{max}}} v_{\mathrm{atom},lm}(|\boldsymbol{r} - \boldsymbol{R}_{\mathrm{atom}}|) Y_{lm}(\Omega_{\mathrm{atom}})$$

Polyalanine Ala_{20}, DFT-PBE (203 atoms)

α-helical vs. extended: Total energy convergence with l_{max}



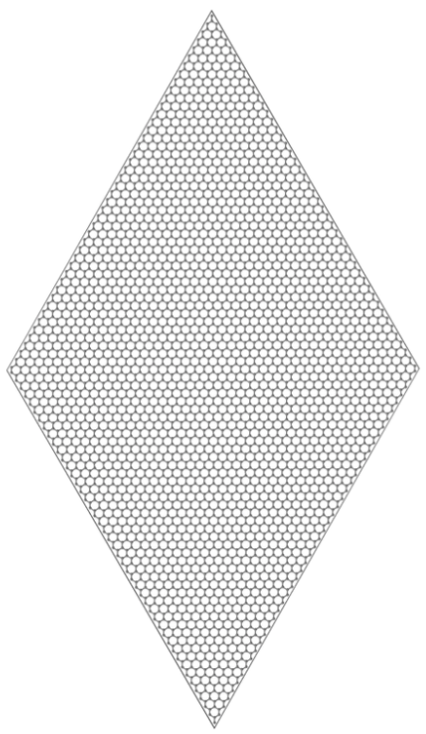


α

extended

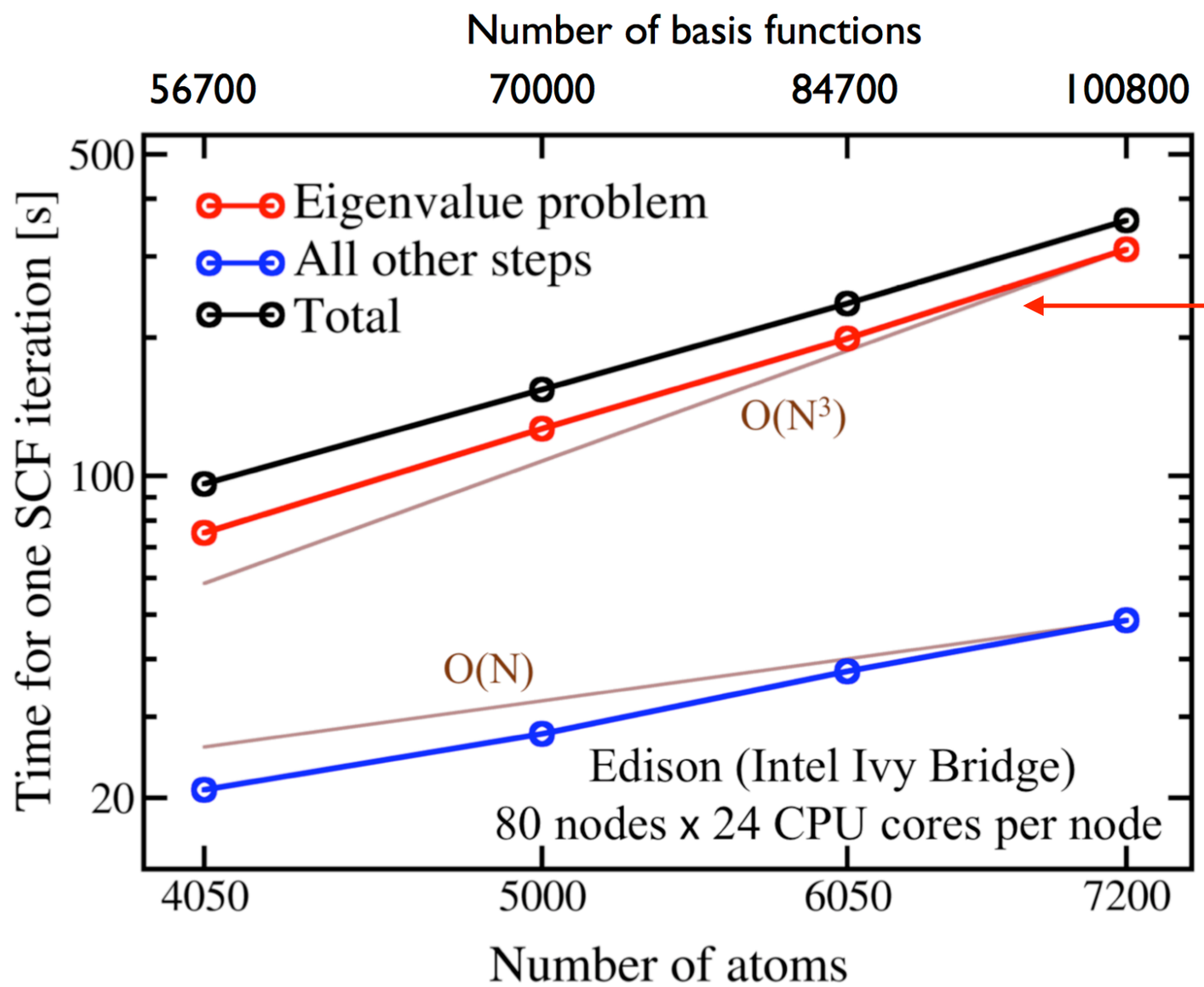
Typical Scaling - $O(N^3)$ Wall

$$H\varphi = \epsilon S\varphi$$



Graphene monolayer,
4050 atoms

FHI-aims, PBE, "light" settings



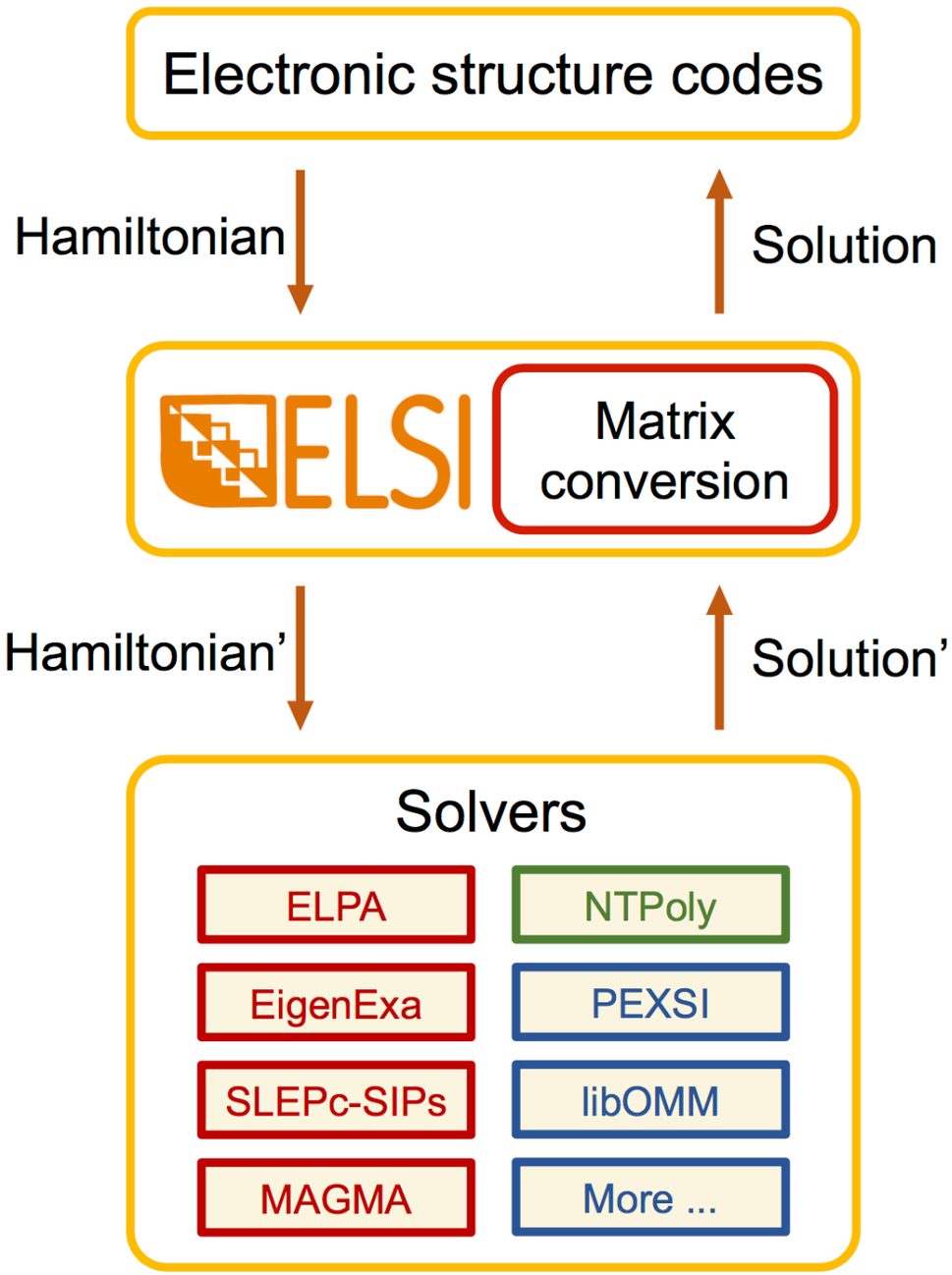
ELPA Library
http://elpa.rzg.mpg.de

Generic problem for any Kohn-Sham DFT code ... solution strategies?

ELSI: Connecting Electronic Structure Codes and Solvers

Yu et al., "ELSI – An Open Infrastructure for Electronic Structure Solvers" Comp. Phys. Commun. 256, 107459 (2020).

Yu et al., Comput. Phys. Commun. 2018
http://elsi-interchange.org
http://git.elsi-interchange.org/elsi-devel/elsi-interface

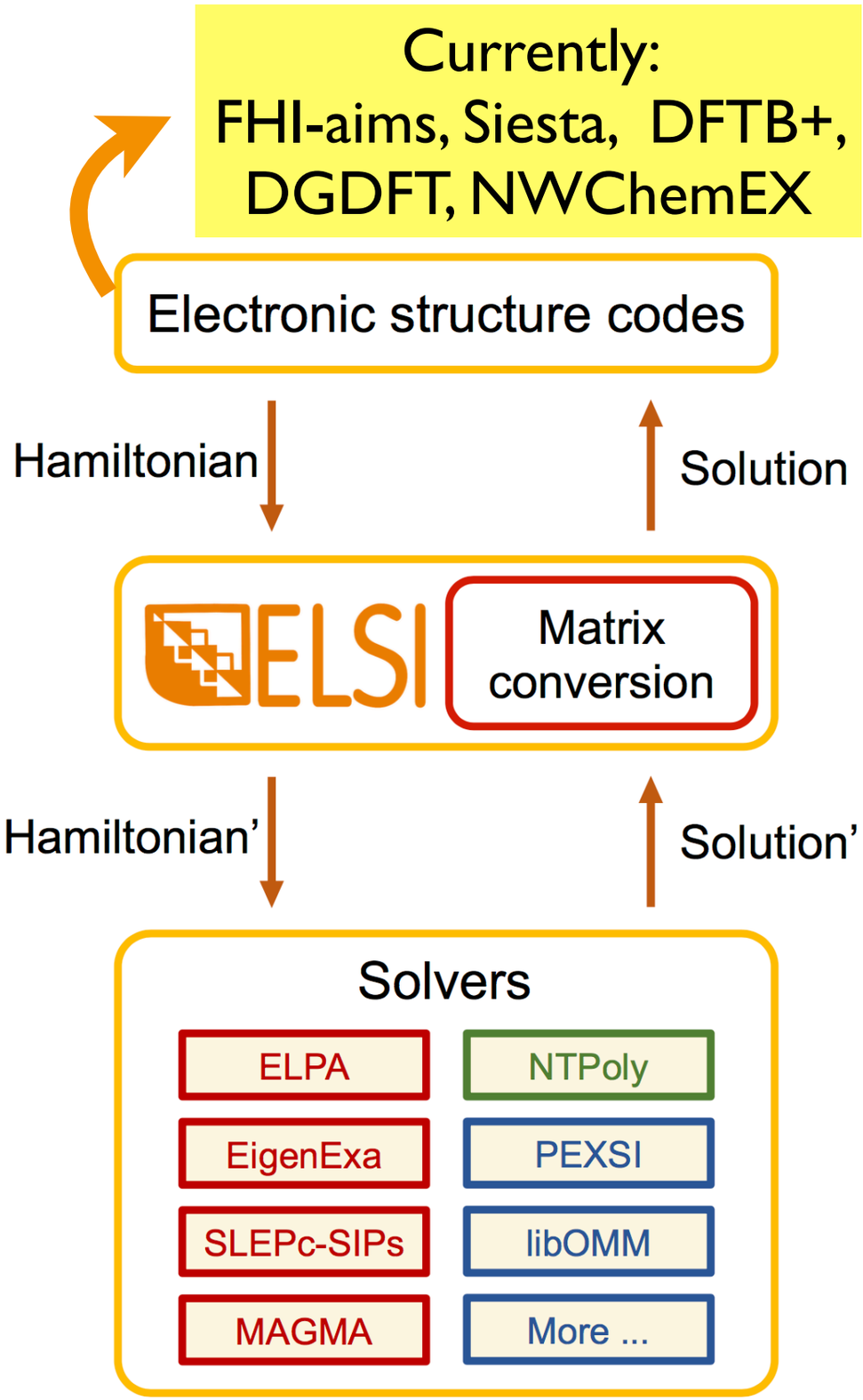


Unified interface connecting KS-DFT codes and solvers

Fast, automatic matrix format conversion and redistribution

Recommendation of optimal solver based on benchmarks

ELSI: Connecting Electronic Structure Codes and Solvers



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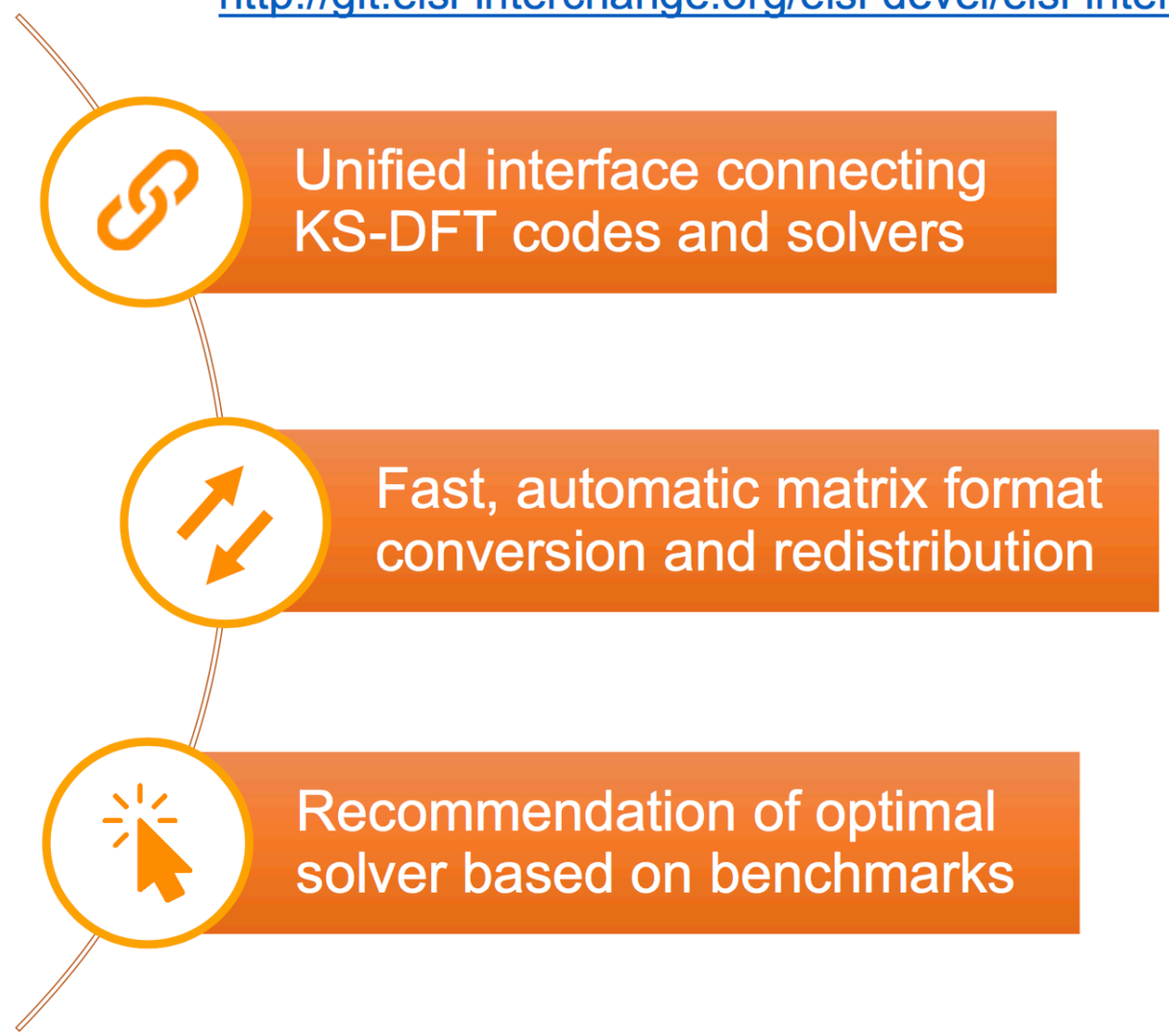


ELSI: Current Functionality

- Designed for rapid integration into a variety of electronic structure codes
- Compatible with common workflows
  - Single self-consistent field (SCF)
  - Multiple SCF cycles (geometry relaxation or molecular dynamics)
- Supports density matrix solvers and eigensolvers on equal footing
- Technical settings adjustable for experienced users
- Object-oriented: Concurrent instances

- **python interface (elsipy)**

<u>Currently supported "solvers":</u>

- Lapack
- ELPA (incl. GPU)
- EigenExa
- MAGMA
- PEXSI
- NTPoly
- libOMM
- SLEPc-SIPS
- BSEPACK
- **ChaSE - E. de Napoli et al.**

FHI-aims+ELSI Coupled with Google TPUs

JCTC Journal of Chemical Theory and Computation

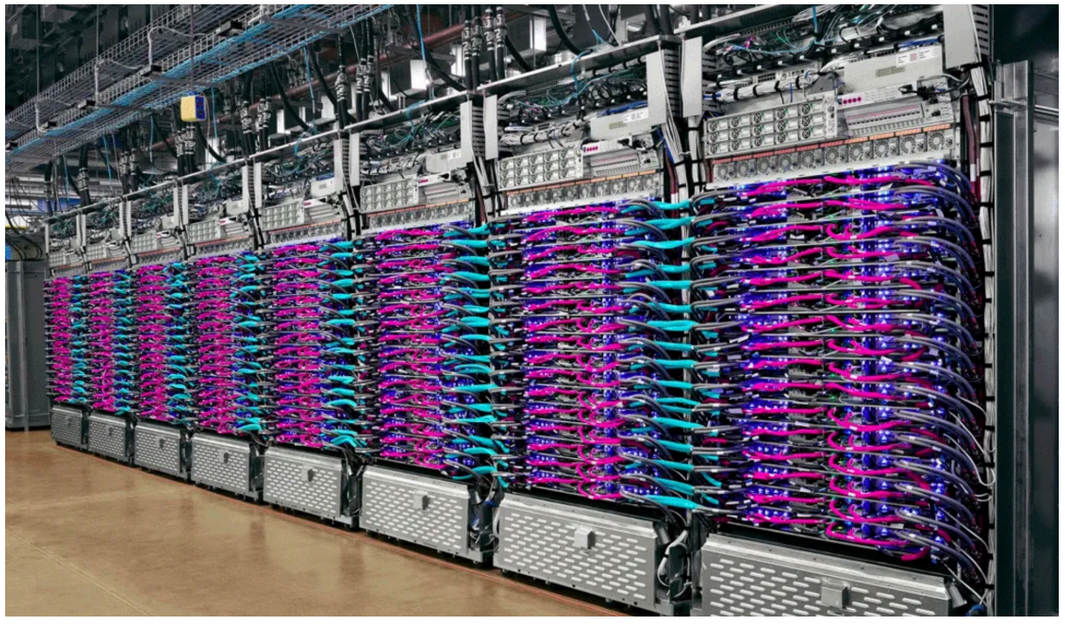
J. Chem. Theory Comput. 2023, 19, 1, 25–32

pubs.acs.org/JCTC Letter

Large Scale Quantum Chemistry with Tensor Processing Units

Ryan Pederson,* John Kozlowski, Ruyi Song, Jackson Beall, Martin Ganahl, Markus Hauru, Adam G. M. Lewis, Yi Yao, Shrestha Basu Mallick, Volker Blum, and Guifre Vidal

Cite This: https://doi.org/10.1021/acs.jctc.2c00876 Read Online



Google TPU pod v3, > 100 PFlops

Hamiltonian build

cost $O(N)$-$O(N^2)$

↔

Density matrix purification

cost $O(N^3)$

run on **CPU**s

FHI-aims & ELSI

run on **TPU**s

JAX & distributed linear algebra library

Prototype: Disk transfer of ELSI CSC stored matrices

FHI-aims+ELSI Coupled with Google TPUs

J. Chem. Theory Comput. 2023, 19, 1, 25–32

H_2O clusters up to 30,981 atoms

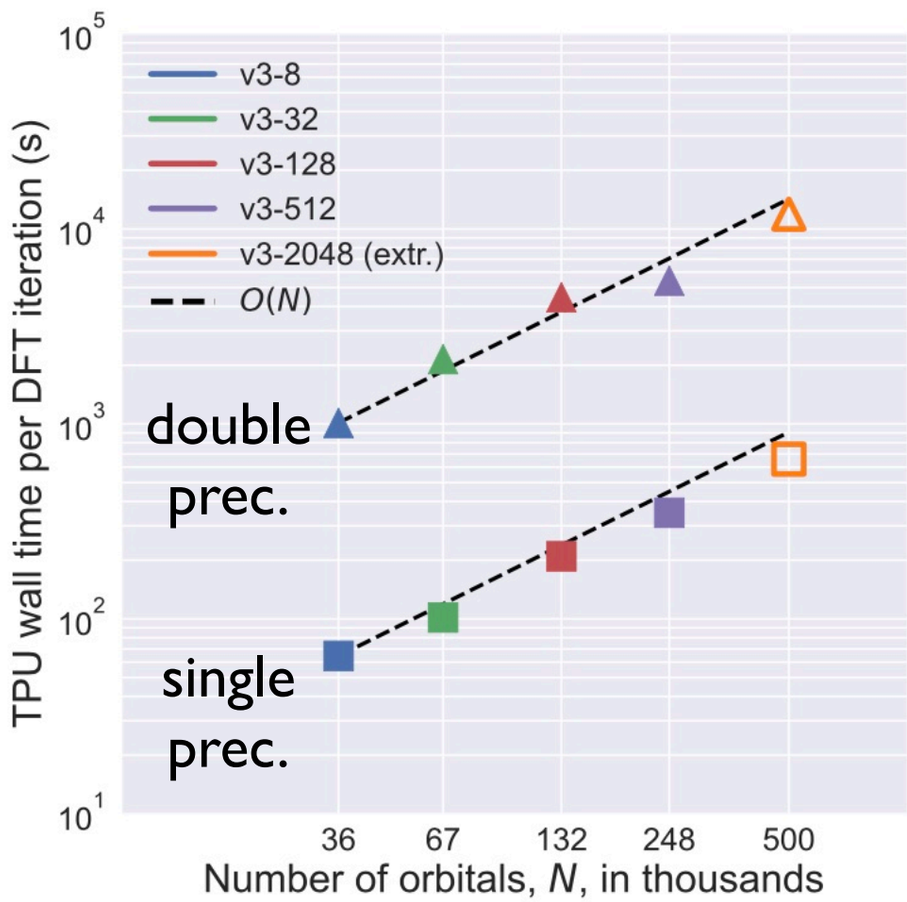


FIG. 1. TPU v3 wall time for $O(N^3)$ density matrix purification, Eqs. (5)-(7), as a function of the number N of orbitals, for clusters of water molecules, both in single (squares) and double (triangles) precision. A full TPU v3 pod with 2048 cores can handle $N \sim 500\,000$ orbitals (extrapolated results).

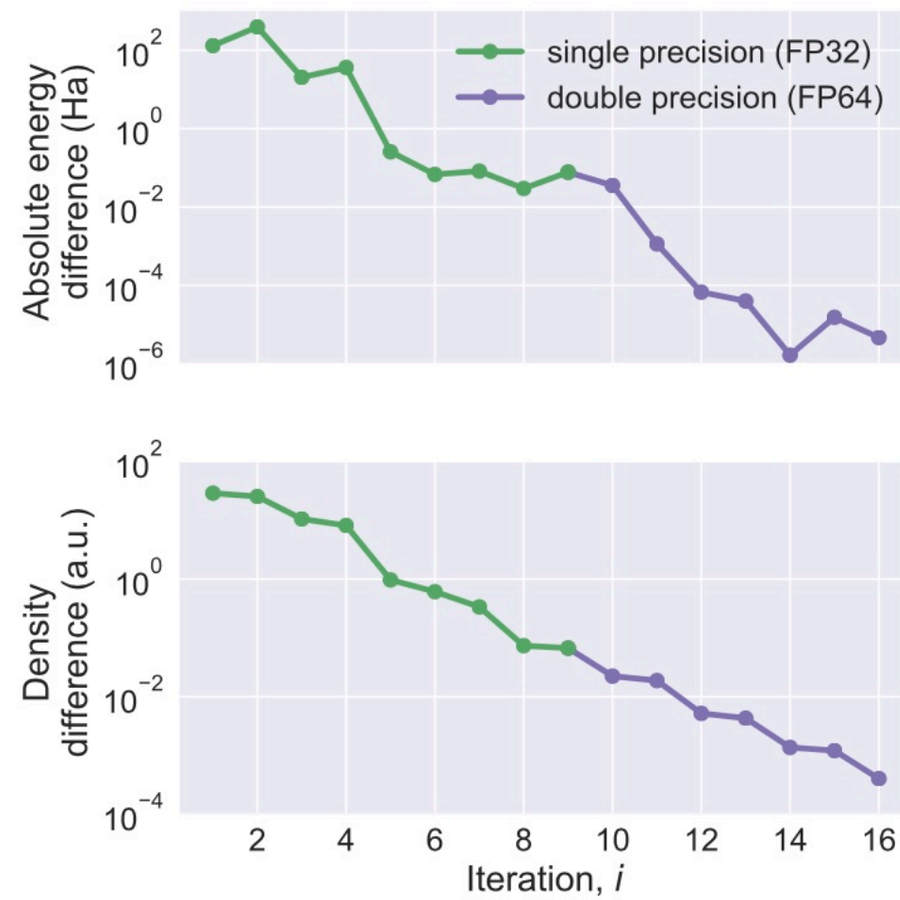
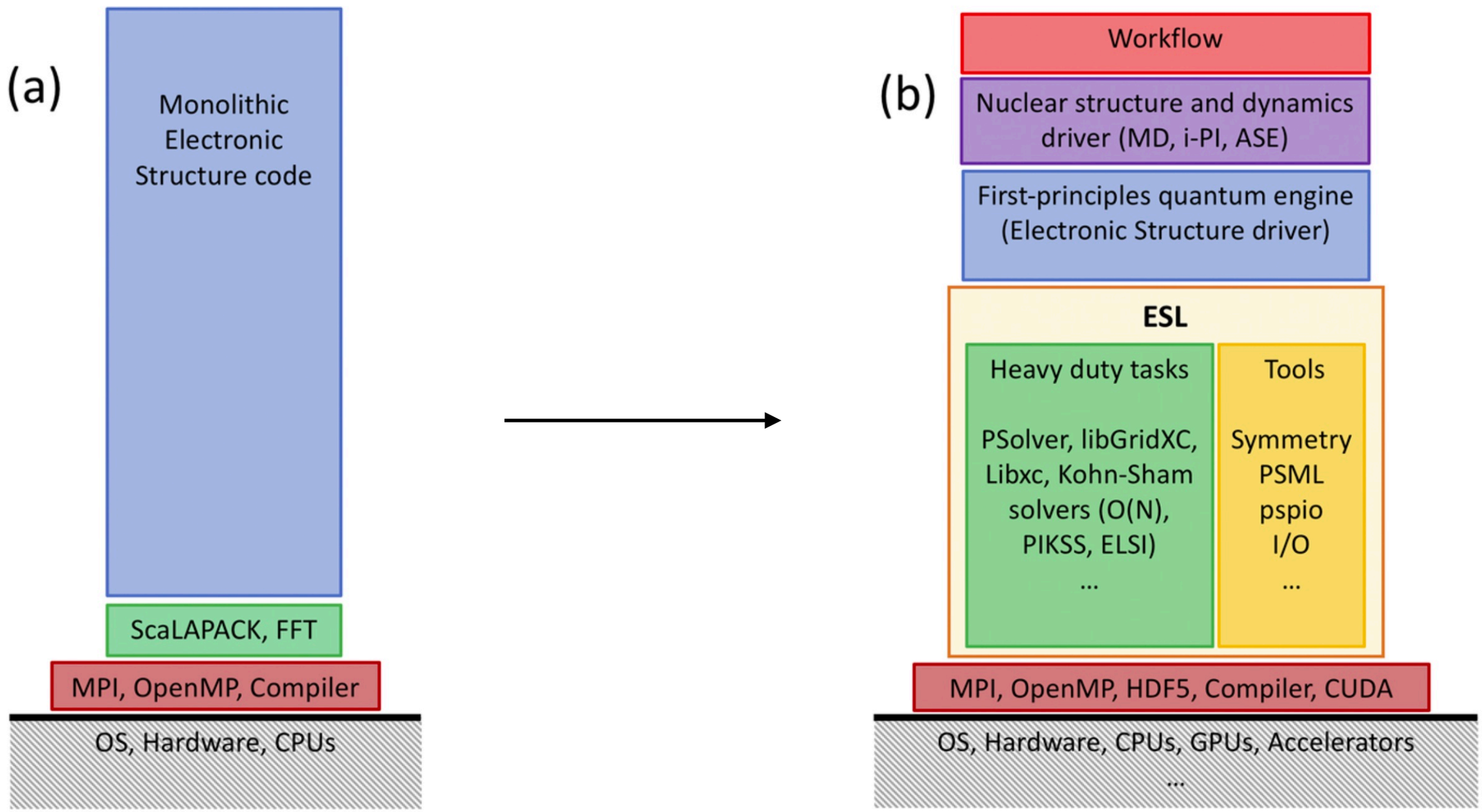


FIG. 4. Convergence trajectory of an end-to-end dynamic precision DFT calculation on a $(H_2O)_{10327}$ cluster. The absolute total energy differences between subsequent DFT iterations, i and $i-1$, are plotted (top). The corresponding difference in real-space densities within the L^1 norm is plotted (bottom).

CECAM Electronic Structure Library

Electronic Structure Theory underpins a broad swath of science,
with "engines" that live for decades: Much of our infrastructure is shared.



Oliveira et al., J. Chem. Phys. 153, 024117 (2020).

Next ESL Coding Workshop

Electronic Structure Software Development: Advancing the Modular Paradigm

https://www.cecam.org/workshop-details/1194

February 19 - March 1, 2024

CECAM-HQ, Lausanne, Switzerland

cecam
Centre Européen de Calcul Atomique et Moléculaire

<u>Save the Date!</u>

2 days of strategy / high-level discussions, 10 days of actual coding in Lausanne.

Beyond DFT-LDA/GGA: Two-Electron Integrals

Common bottleneck: Two-electron interactions, e.g.:

$$\int d^3r d^3r' \phi_i(r)\phi_j(r') \frac{1}{|r-r'|} \phi_k(r')\phi_l(r)$$

Ren, Rinke, Blum, Wieferink, Tkatchenko, Sanfilippo, Reuter, Scheffler, New J. Phys. ***14****, 053020 (2012)*

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$$\int d^3r d^3r' \phi_i(r)\phi_j(r') \frac{1}{|r-r'|} \phi_k(r')\phi_l(r)$$

Early Solution: "Resolution of the identity"

[Boys and Shavitt (1950s), Whitten (1974), Dunlap et al. (1979), Vahtras et al. (1993), many others]

1. Expand pair products in smaller auxiliary basis set $\{P_\mu\}$

$$\varphi_i(\boldsymbol{r})\varphi_j(\boldsymbol{r}) = \sum_\mu C_{ij}^\mu P_\mu(\boldsymbol{r})$$

2. Use "Coulomb metric" *V* to cancel linear error terms

$$C_{ij}^\mu = \sum_\nu (ij|\nu) V_{\nu\mu}^{-1}$$

$$(ij|\nu) = \int d^3r d^3r' \frac{\varphi_i(\boldsymbol{r})\varphi_j(\boldsymbol{r})P_\nu(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|}$$

→ The rest is matrix algebra - works with NAO basis sets!
Hartree-Fock, hybrid functionals, MP2, *GW*, RPA, LR-TDLDA, ...

Ren, Rinke, Blum, Wieferink, Tkatchenko, Sanfilippo, Reuter, Scheffler, New J. Phys. ***14****, 053020 (2012)*

Localized "Resolution of Identity" (RI) for Two-Electron Terms

$$(ij|kl) = \int d^3r d^3r' \frac{\varphi_i(\boldsymbol{r})\varphi_j(\boldsymbol{r}')\varphi_k(\boldsymbol{r})\varphi_l(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|}$$

$$\varphi_i(\boldsymbol{r})\varphi_j(\boldsymbol{r}) = \sum_\mu C_{ij}^\mu P_\mu(\boldsymbol{r})$$

$$C_{ij}^\mu = \sum_\nu (ij|\nu) V_{\nu\mu}^{-1}$$

Problem: Full RI-V delocalizes *C* across entire system

In FHI-aims: *Ihrig, Wieferink, Zhang, Ropo, Ren, Rinke, Scheffler, Blum, New J. Phys.* ***17****, 093020 (2015)*

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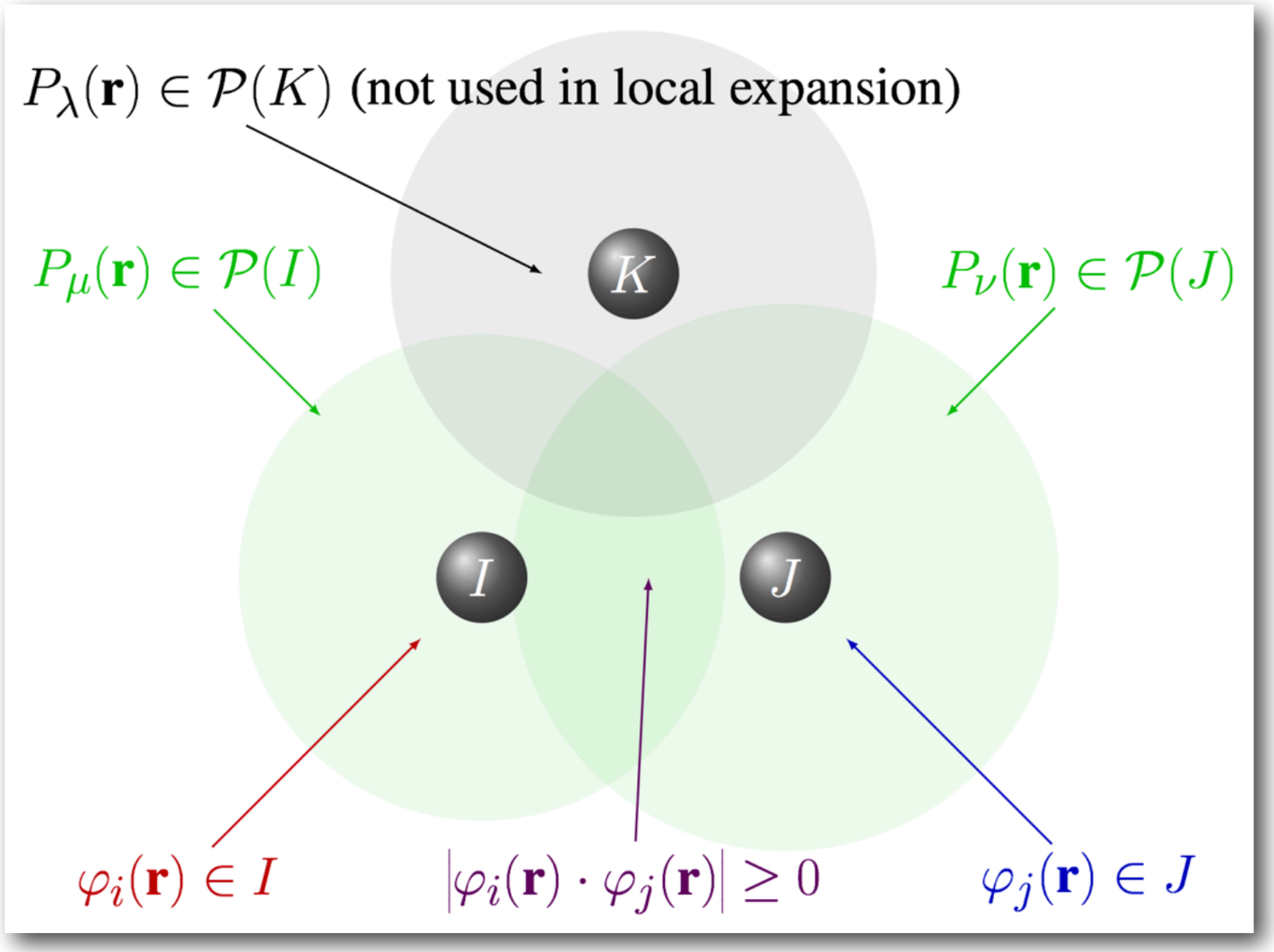


Problem: Full RI-V delocalizes *C* across entire system

Solution: For each C_{ij}^μ, restrict μ only to atoms I and J at which i and j are centered.

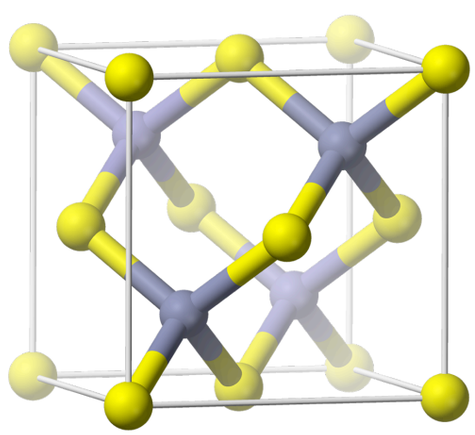
In FHI-aims: *Ihrig, Wieferink, Zhang, Ropo, Ren, Rinke, Scheffler, Blum, New J. Phys.* ***17****, 093020 (2015)*

Hybrid DFT in FHI-aims 2015

Ihrig, Wieferink, Zhang, Ropo, Ren, Rinke, Scheffler, Blum, New J. Phys. ***17****, 093020 (2015)*
Levchenko, Ren, Wieferink, Rinke, Johanni, Blum, Scheffler, Comp. Phys. Commun. ***192****, 60-69 (2015).*

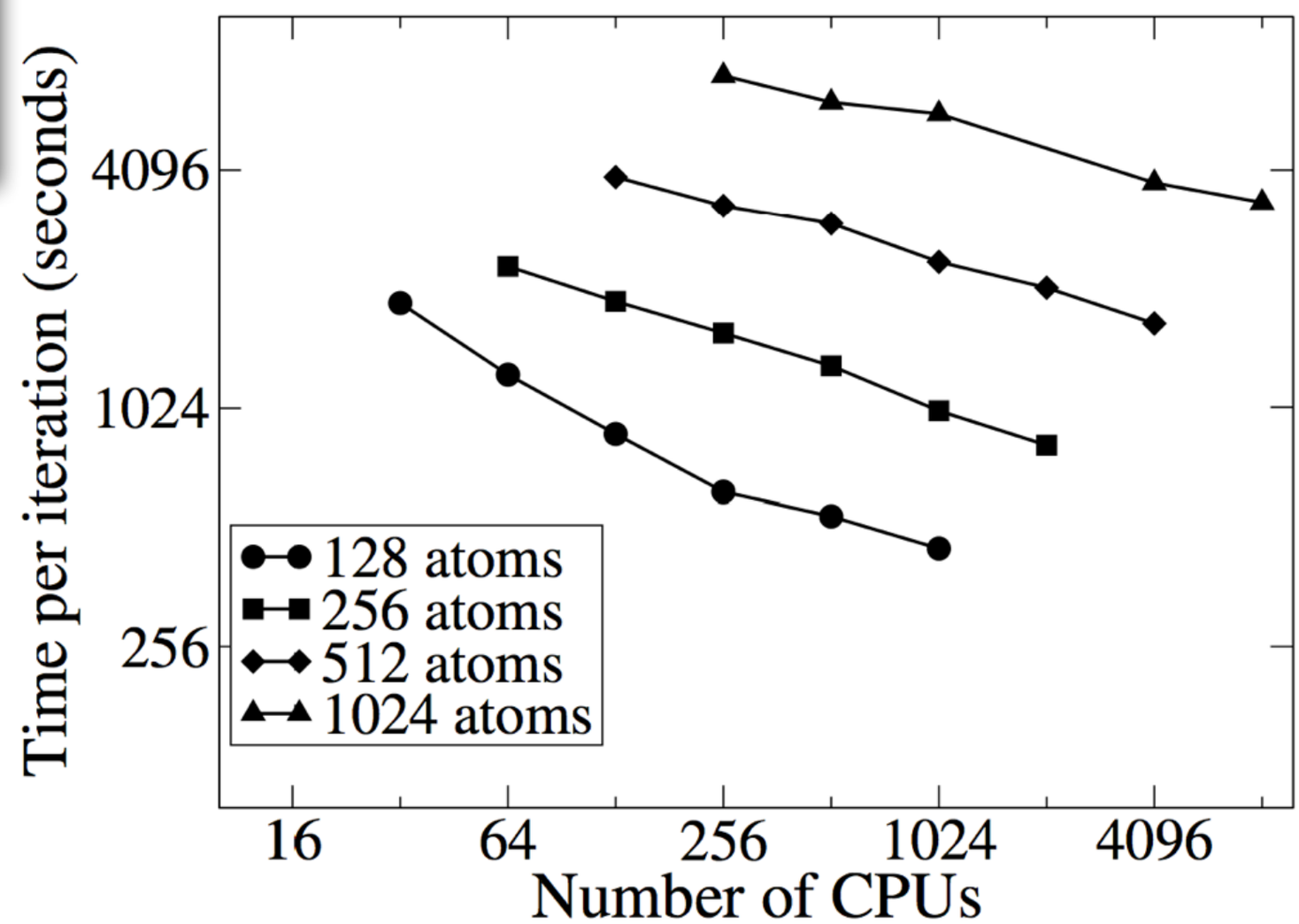
O(N) scaling implementation, localized resolution of identity.

Note: Localized RI also works for MP2, RPA, GW, etc. (but not O(N))



Zincblende GaAs

Computational Scaling of Periodic GaAs, "tight" HSE06 Hybrid Density-Functional Theory



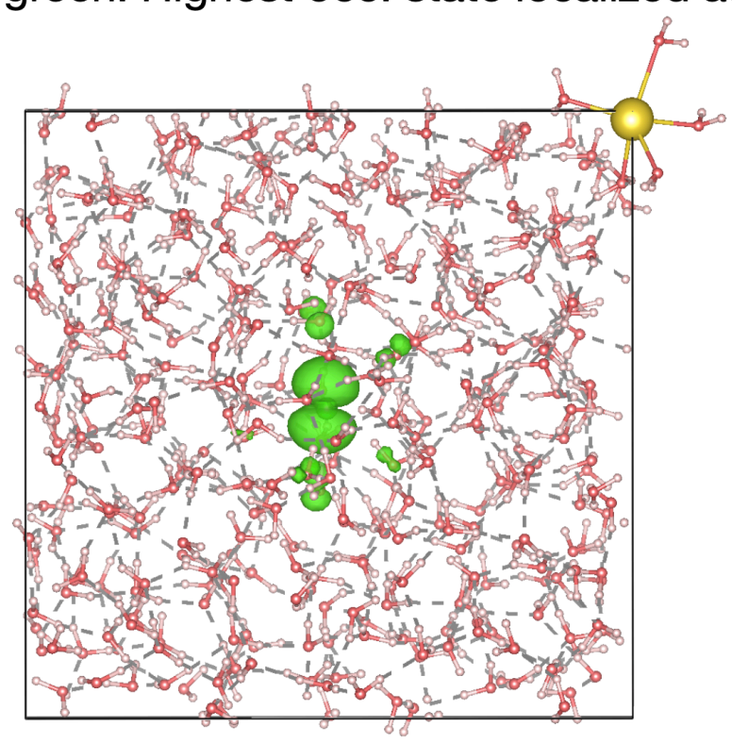
… proved incredibly useful, but not enough. Can we be faster, larger?

Hybrid DFT in FHI-aims 2023

Florian Merz[1], Andreas Marek[2], **Sebastian Kokott**[3], Christian Carbogno[3], Yi Yao[3,5], Mariana Rossi[4], Markus Rampp[2], Matthias Scheffler[3], and Volker Blum[5]

Charge Localization

Saline Liquide = $Na^+ + Cl^- + (H_2O)_{360}$
PBE0(α=0.5), T=300K
In green: Highest occ. state localized at Cl



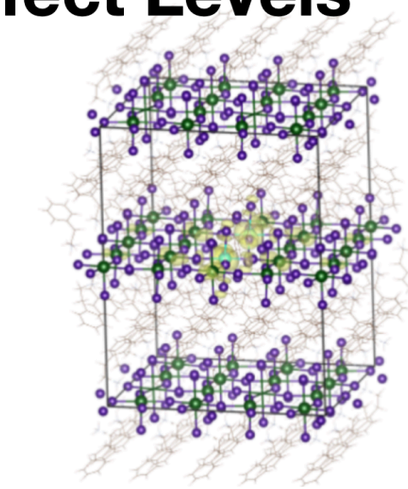
Band Gaps/Defect Levels

phenylethylammonium lead iodide

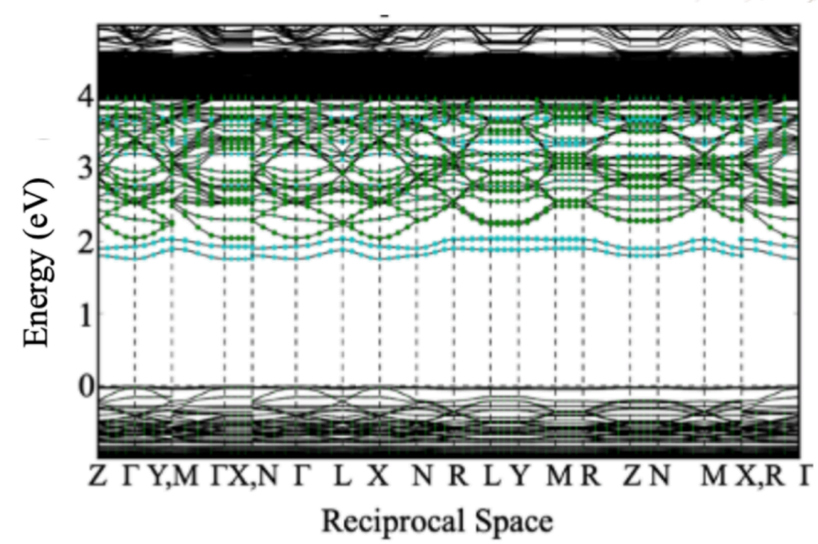
$PEA_{64}(Pb_{29}Bi_2\square)I_{128}$

=1503 atoms

HSE06 + pSOC



Haipeng Lu, Gabrielle Koknat et al., submitted (2022)



[1]*Lenovo HPC Innovation Center, Stuttgart*
[2]*Max Planck Computing and Data Facility, Garching*
[3]*The NOMAD Laboratory at the FHI-MPG and IRIS-HU, Berlin*
[4]*MPI for the Structure and Dynamics of Matter, Hamburg*
[5]*Duke University, Durham, North Carolina, USA*

MS1P
Molecular Simulations from First Principles e.V.

NOMAD

Code Improvements (Hard Work But Effective)

$$(ij|kl) = \int d^3r d^3r' \frac{\varphi_i(\boldsymbol{r})\varphi_j(\boldsymbol{r}')\varphi_k(\boldsymbol{r})\varphi_l(\boldsymbol{r}')}{|\boldsymbol{r}-\boldsymbol{r}'|}$$

$$\varphi_i(\boldsymbol{r})\varphi_j(\boldsymbol{r}) = \sum_\mu C^\mu_{ij} P_\mu(\boldsymbol{r})$$

$$C^\mu_{ij} = \sum_\nu (ij|\nu) V^{-1}_{\nu\mu}$$

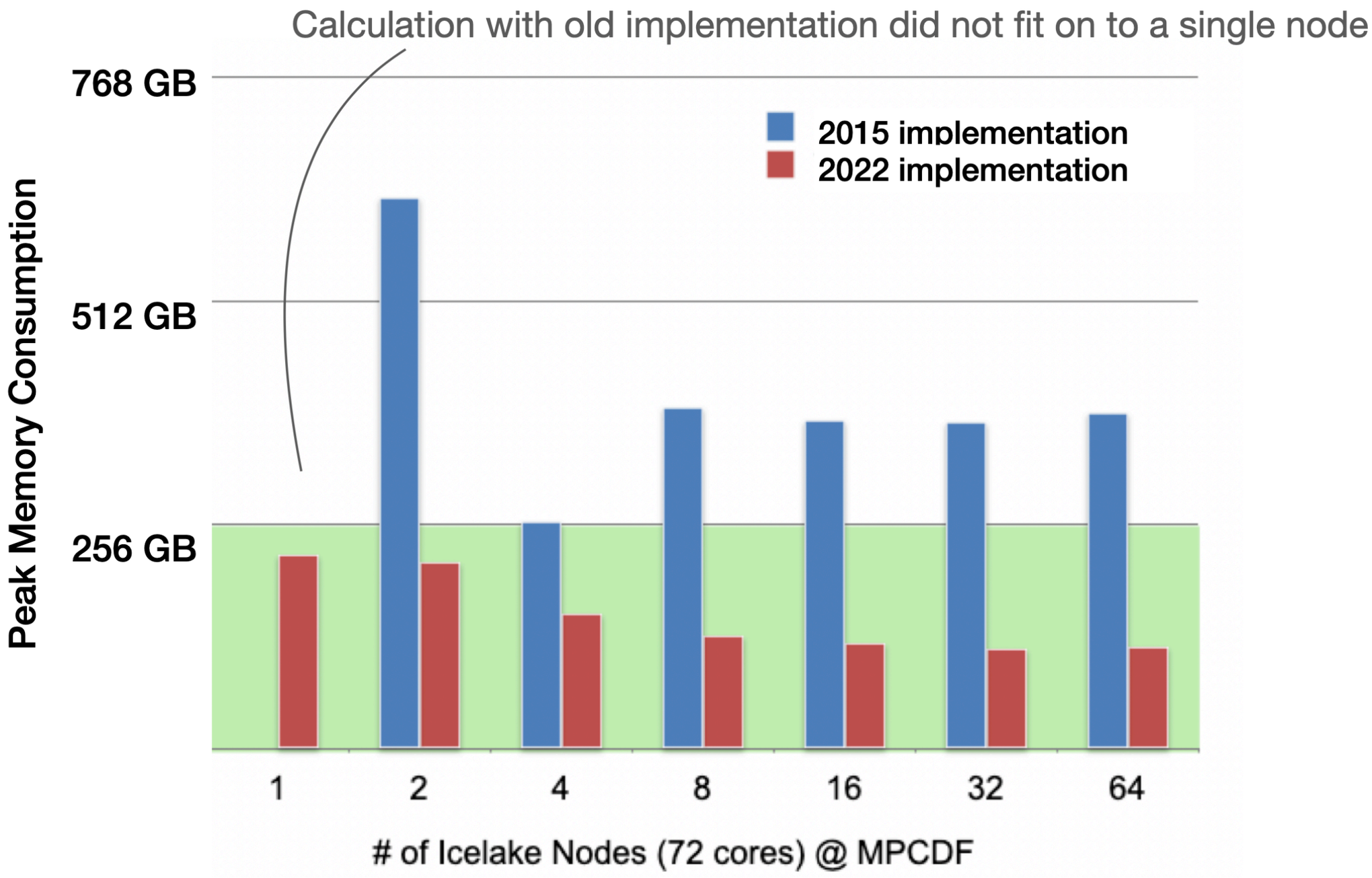
CPUs only!

FHI-aims
The ab initio materials simulation package

- MPI-3 intra-node shared memory arrays
  - Facilitate data redistribution via one-sided MPI routines; e.g. Coulomb matrix
- Compression of the RI-coefficients; exploiting sparsity in real-space
  - Same screening mechanism as for Coulomb matrix (2015)
- Auto-tuning mechanism for blocking the data
  - Minimize communication, maximize memory/node usage
- Additional parallelization layers
  - Finer granulation on the level of basis functions (previously: on the level of atoms)
- Auto-tuning for an optimal workload balance

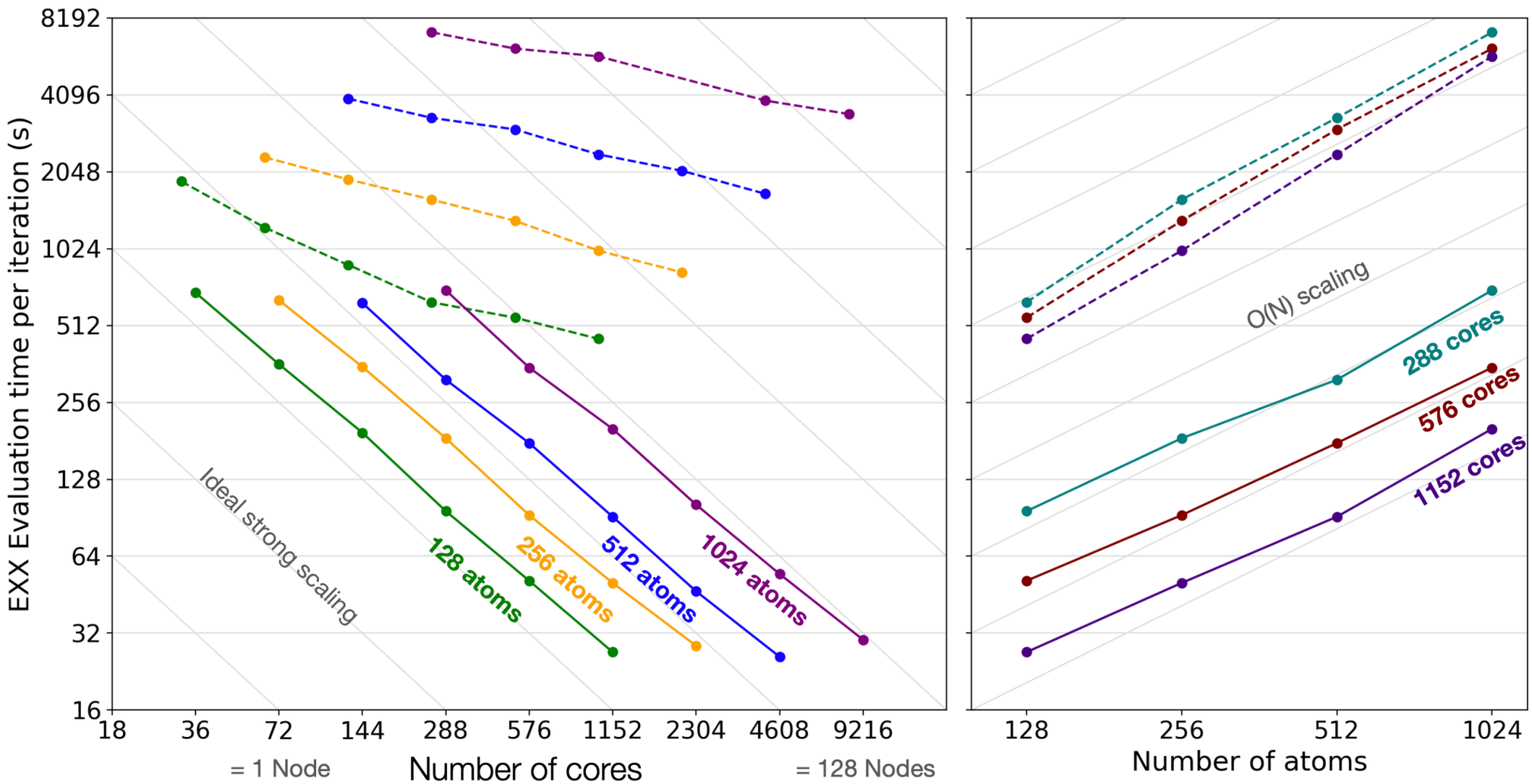
Memory Optimizations

GaAs supercell, 512 atoms: HSE06, tight settings



Drastic Scalability Improvements*

GaAs supercells: HSE06, tight settings



Hydra @ MPCDF
Intel Sandy Bridge
32 cores / node

Raven @ MPCDF
Intel Xeon IceLake
(Platinum 8360Y)
72 cores / node
256 GB / node

MPCDF
MAX PLANCK
COMPUTING & DATA FACILITY

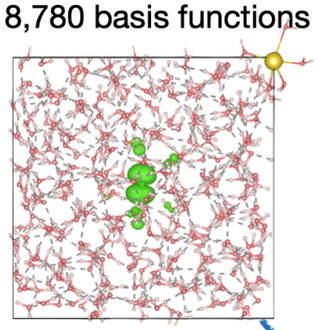
*Not so simple. Our 2015 implementation had already seen some years of work by several very smart and focused individuals.

Scalability: Hybrid DFT for Very Large Systems

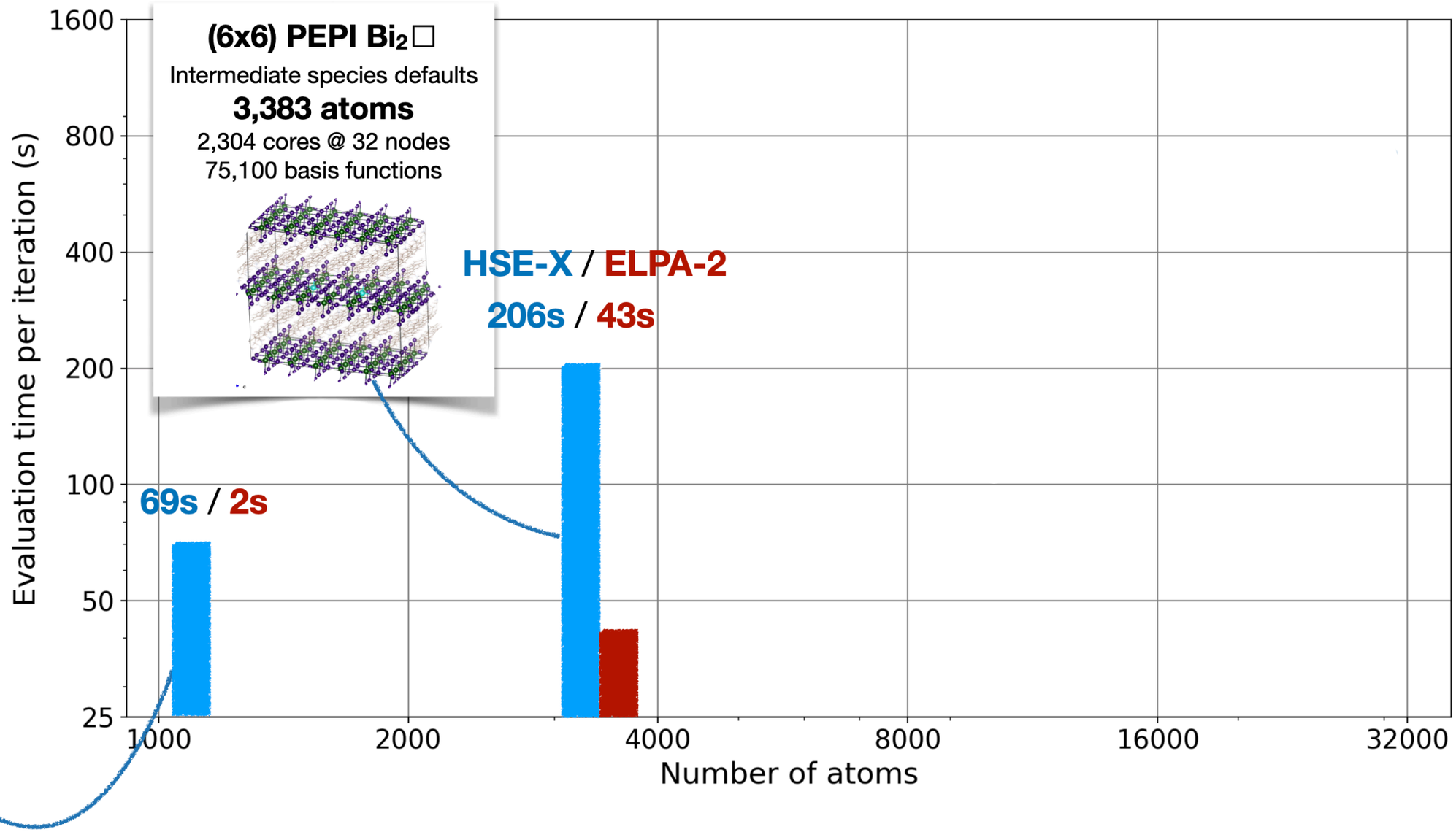
Raven @ MPCDF
Intel Xeon IceLake
(Platinum 8360Y)
72 cores / node
256 GB / node

MPCDF
MAX PLANCK
COMPUTING & DATA FACILITY

Saline Liquide (PBE0)
Intermediate species defaults
1,082 atoms
576 cores @ 8 nodes
18,780 basis functions



(6x6) PEPI $Bi_2\square$
Intermediate species defaults
3,383 atoms
2,304 cores @ 32 nodes
75,100 basis functions



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MAX PLANCK
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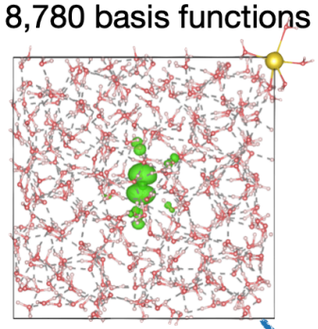
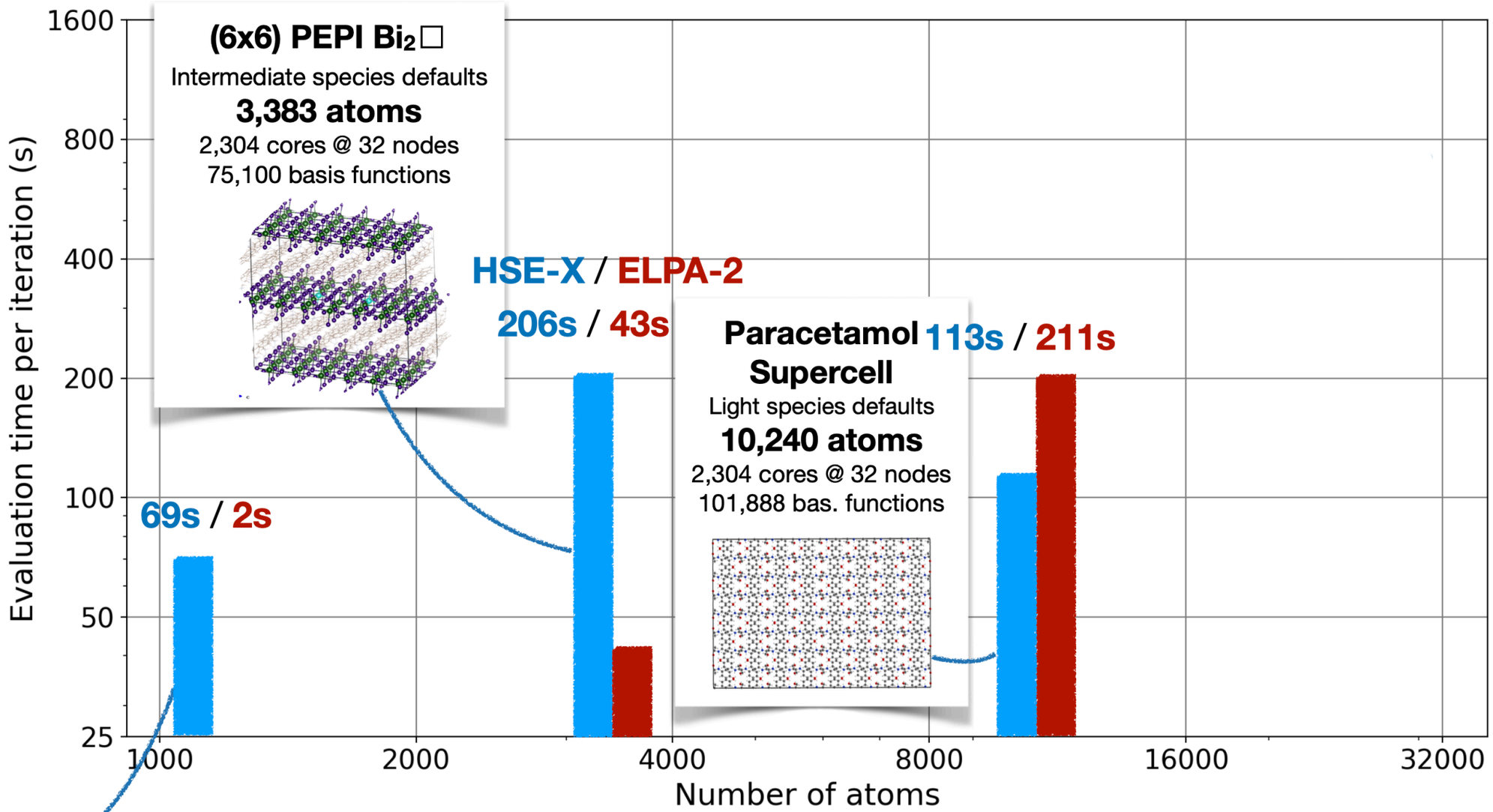
18,780 basis functions





(6x6) PEPI $Bi_2\square$
Intermediate species defaults
3,383 atoms
2,304 cores @ 32 nodes
75,100 basis functions

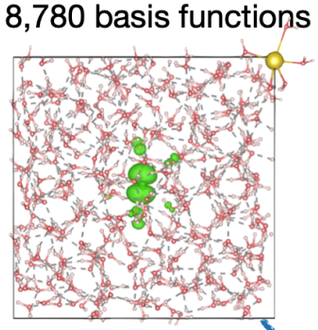
Paracetamol Supercell
Light species defaults
10,240 atoms
2,304 cores @ 32 nodes
101,888 bas. functions

Scalability: Hybrid DFT for Very Large Systems

Raven @ MPCDF
Intel Xeon IceLake
(Platinum 8360Y)
72 cores / node
256 GB / node

MAX PLANCK
COMPUTING & DATA FACILITY

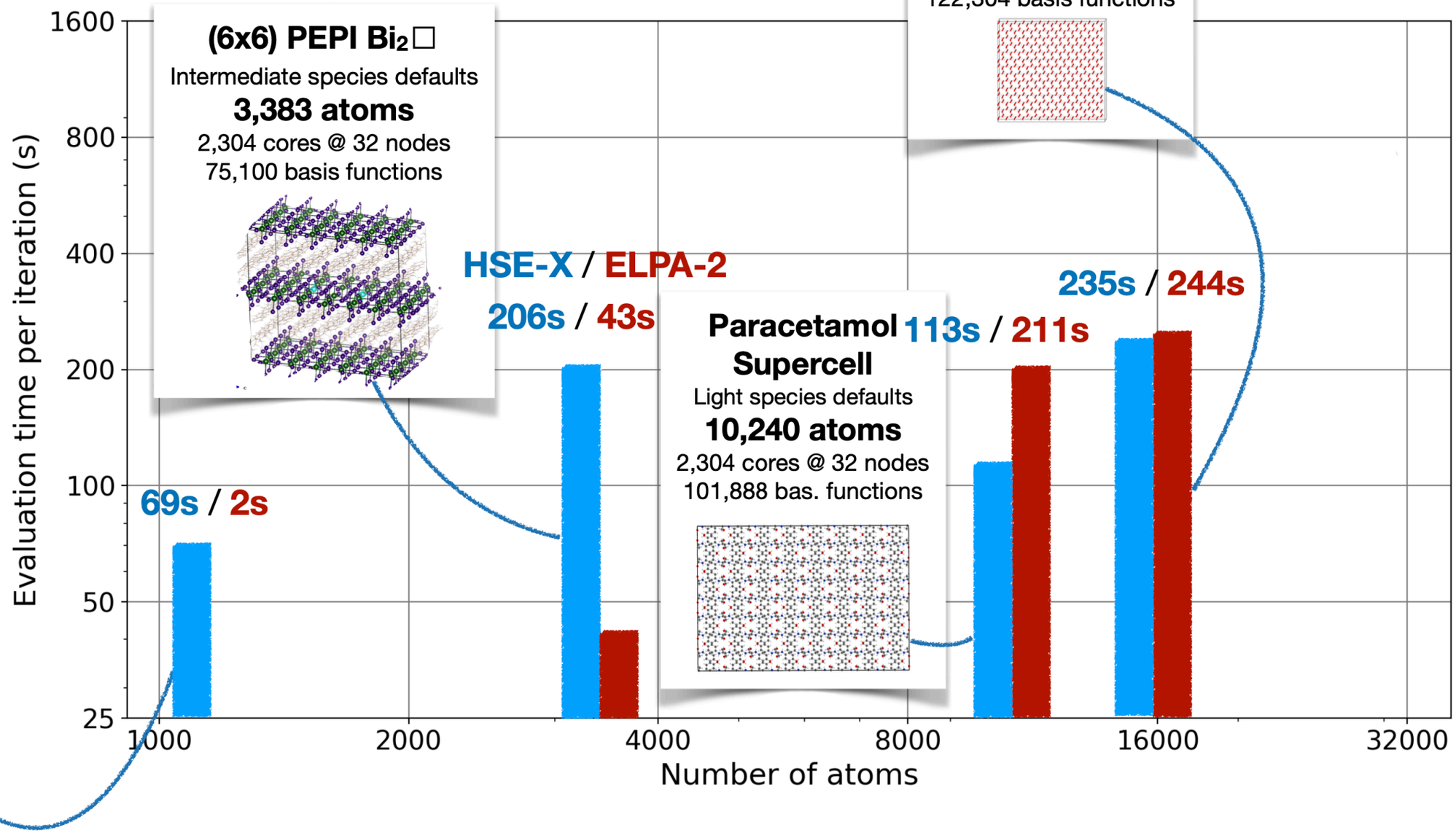
Saline Liquide (PBE0)
Intermediate species defaults
1,082 atoms
576 cores @ 8 nodes
18,780 basis functions



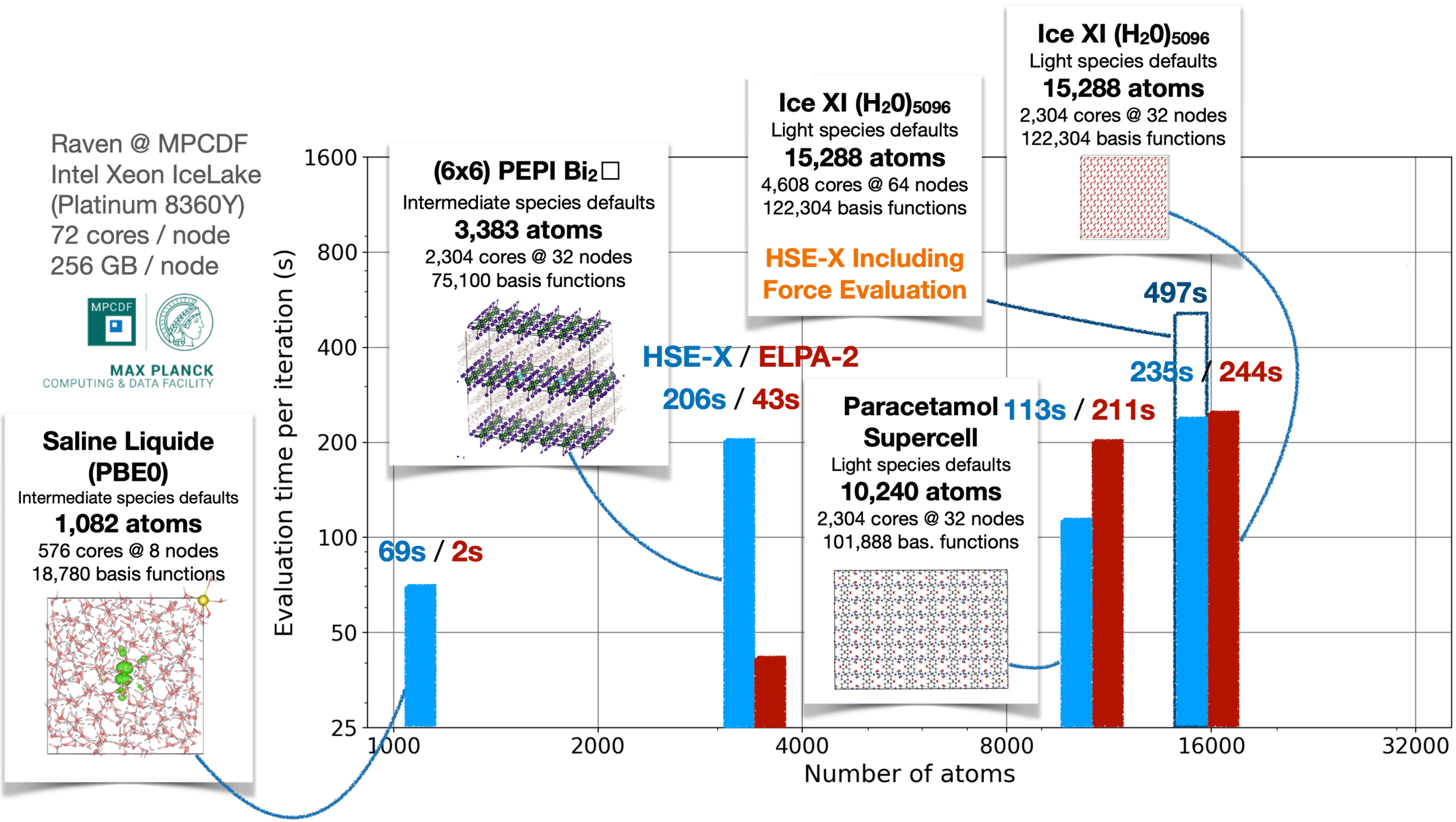
(6x6) PEPI $Bi_2\square$
Intermediate species defaults
3,383 atoms
2,304 cores @ 32 nodes
75,100 basis functions

Ice XI $(H_20)_{5096}$
Light species defaults
15,288 atoms
2,304 cores @ 32 nodes
122,304 basis functions

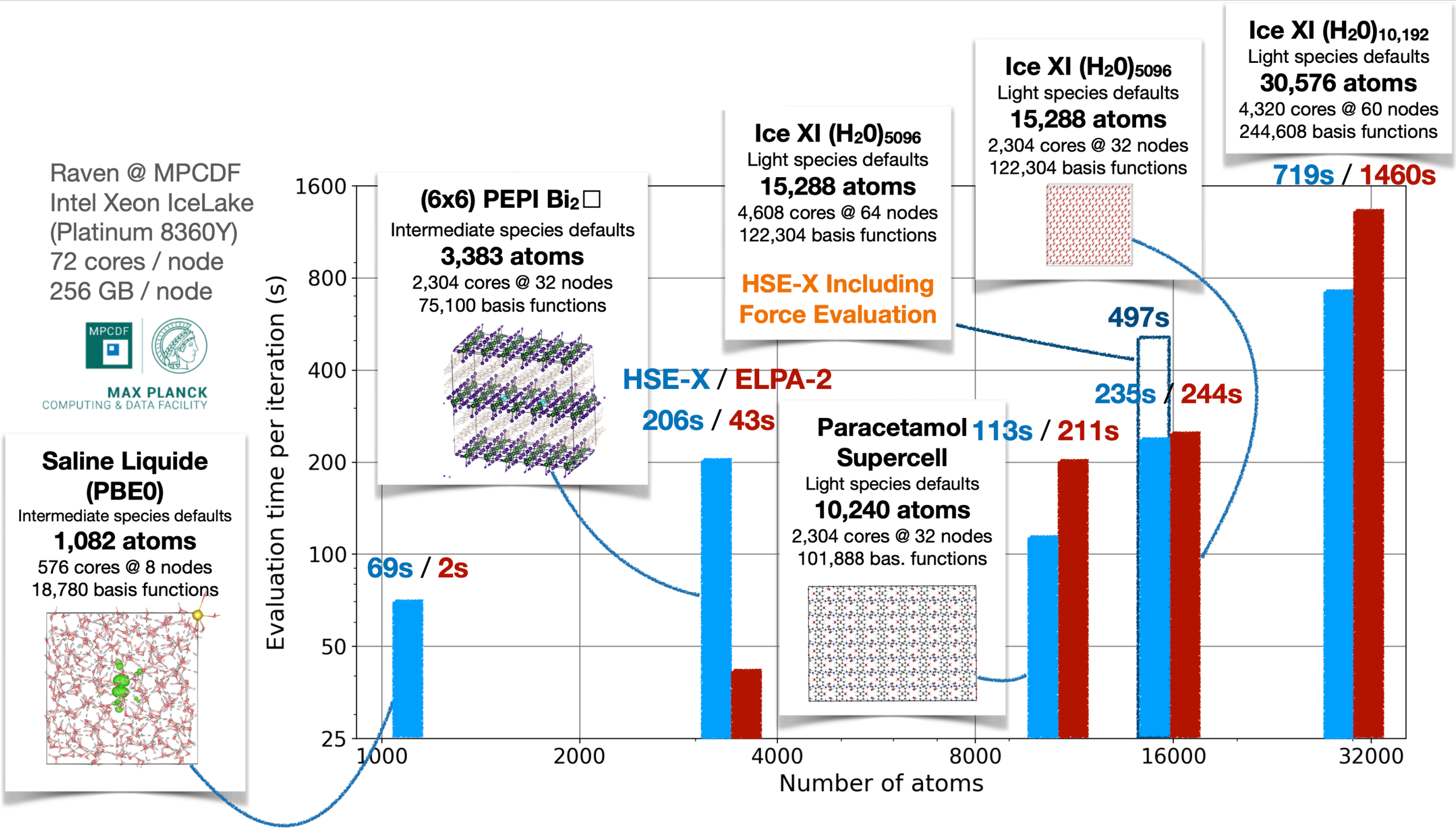
Paracetamol Supercell
Light species defaults
10,240 atoms
2,304 cores @ 32 nodes
101,888 bas. functions



Scalability: Hybrid DFT for Very Large Systems



Scalability: Hybrid DFT for Very Large Systems



Relativity in FHI-aims

Non-relativistic QM: Schrödinger Equation

$$V\phi + \frac{\boldsymbol{p}^2}{2m}\phi = \epsilon\phi$$

- one component (two with spin)
- one Hamiltonian for all states

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Relativistic: Dirac-Coulomb Hamiltonian

$$\begin{pmatrix} V & c\sigma \cdot p \\ c\sigma \cdot p & -2c^2 + V \end{pmatrix} \begin{pmatrix} \phi \\ \chi \end{pmatrix} = \epsilon \begin{pmatrix} \phi \\ \chi \end{pmatrix}$$

Relativity in FHI-aims

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... simply rewrite:

$$V\phi + \boldsymbol{\sigma}\cdot\boldsymbol{p}\frac{c^2}{2c^2+\epsilon-V}\boldsymbol{\sigma}\cdot\boldsymbol{p}\phi = \epsilon\phi$$

- ε-dependent Hamiltonian
- Not negligible for $\epsilon - v(\boldsymbol{r}) \approx 2c^2$

⇔ affects near-nuclear part of *any* wave function

FHI-aims: Scalar Relativity plus Spin-Orbit Coupling

$$V\phi + \boldsymbol{\sigma}\cdot\boldsymbol{p}\frac{c^2}{2c^2+\epsilon-V}\boldsymbol{\sigma}\cdot\boldsymbol{p}\phi = \epsilon\phi$$

$$\sigma = (\sigma_1, \sigma_2, \sigma_3)$$

Pauli matrices

William Huhn

Huhn, Blum, Phys. Rev. Materials ***1****, 033803 (2017).*

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Pauli matrices

William Huhn

<u>Rewrite:</u>

$$V\phi + (\boldsymbol{p}\frac{c^2}{2c^2+\epsilon-V}\boldsymbol{p} + i\boldsymbol{p}\frac{c^2}{2c^2+\epsilon-V} \times \boldsymbol{p}\cdot\boldsymbol{\sigma})\phi = \epsilon\phi$$

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Approximate:

$$V\phi + \boldsymbol{p}\frac{c^2}{2c^2 - V_{\text{free atom}}}\boldsymbol{p}\phi = \epsilon\phi$$

e.g. (FHI-aims): atomic ZORA

- No gauge-invariance problem
- Simple total-energy gradients

$$H_{SOC} = \frac{i}{4c^2}\boldsymbol{p}V \times \boldsymbol{p}\cdot\boldsymbol{\sigma}$$

[lowest order in $(\varepsilon - V)/2c^2$]

- Can be applied (cheaply) as non-selfconsistent correction to DFT eigenvalues

Huhn, Blum, Phys. Rev. Materials ***1****, 033803 (2017).*

Quasi-Four-Component (Q4C) Approach in FHI-aims

Zhao, Yu, Zhang, Xiao, Zhang, Blum, Phys. Rev. B ***103****, 245144 (2021).*

Rundong Zhao

Dirac-Coulomb Equation

$$\begin{pmatrix} V & c\sigma\cdot p \\ c\sigma\cdot p & -2c^2+V \end{pmatrix}\begin{pmatrix}\phi\\ \chi\end{pmatrix} = \epsilon\begin{pmatrix}\phi\\ \chi\end{pmatrix}$$

Q4C: *Liu and Peng, J. Chem. Phys. 125, 044102 (2006).*

1) $$\begin{pmatrix}\phi_i\\ \chi_i\end{pmatrix} = C_{i\mu}\cdot\begin{pmatrix}\varphi_\mu^L\\ \varphi_\mu^S\end{pmatrix}$$

2) $$n^{SCF}(\mathbf{r}) = n^{L,SCF}(\mathbf{r}) + \sum_{\text{atoms A}}\left[n_A(\mathbf{r}) - n_A^L(\mathbf{r})\right]$$

Each four-component basis function: Fixed relation $\varphi_\mu^S = \hat{K}\varphi_\mu^L$

Quasi-Four-Component (Q4C) Approach in FHI-aims

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Each four-component basis function: Fixed relation $\varphi_\mu^S = \hat{K}\varphi_\mu^L$

Atomic basis functions and atomic densities:
DFTATOM code *Certik, Pask, Vackar, Comp. Phys. Commun. 184, 1777 (2013).*

$$\hat{K} = \left[\frac{c}{2c^2+\epsilon_{A,\mu}-V_A}\right](\sigma\cdot p)$$

Other basis functions (analogous to at. ZORA):

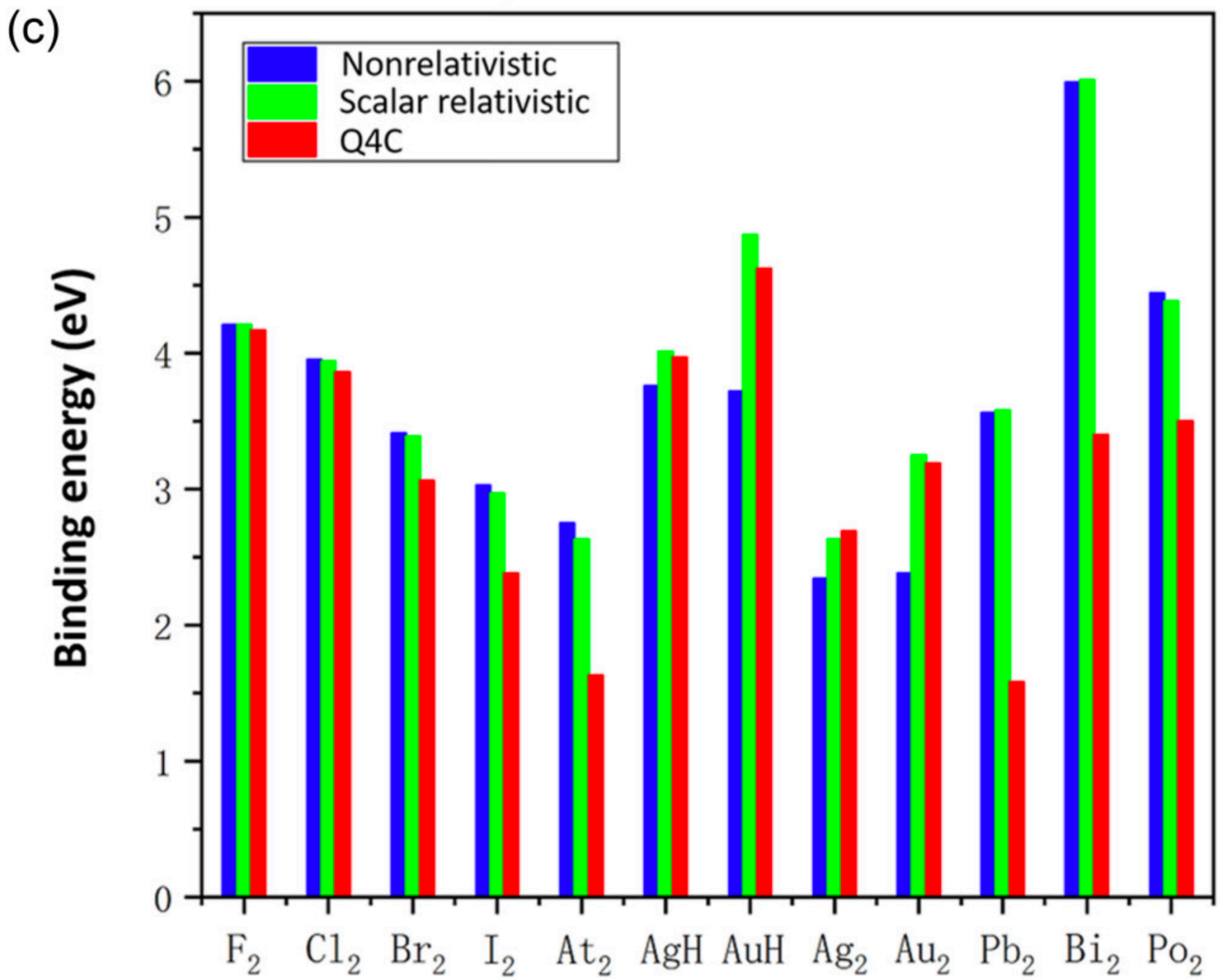
$$\hat{K} = \left[\frac{c}{2c^2-V_A}\right](\sigma\cdot p)$$

Benchmarking Lower Levels of Relativity vs. Q4C

Zhao, Yu, Zhang, Xiao, Zhang, Blum, Phys. Rev. B ***103****, 245144 (2021).*

Rundong Zhao

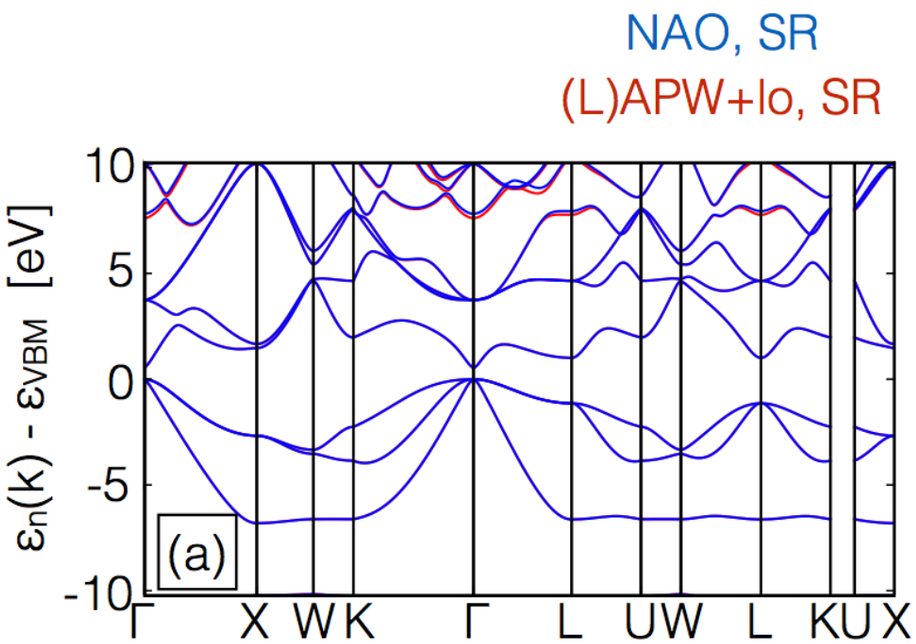
Binding Energies - Symmetric Dimers
(closed shell, FHI-aims tight/tier2, DFT-LDA)



Unsurprising but instructive:

- s-bonded dimers need scalar relativity
- p-bonded dimers need spin-orbit coupling
- *NB: Test of impact of relativity only - closed-shell / DFT-LDA not sufficient for physically satisfactory results*

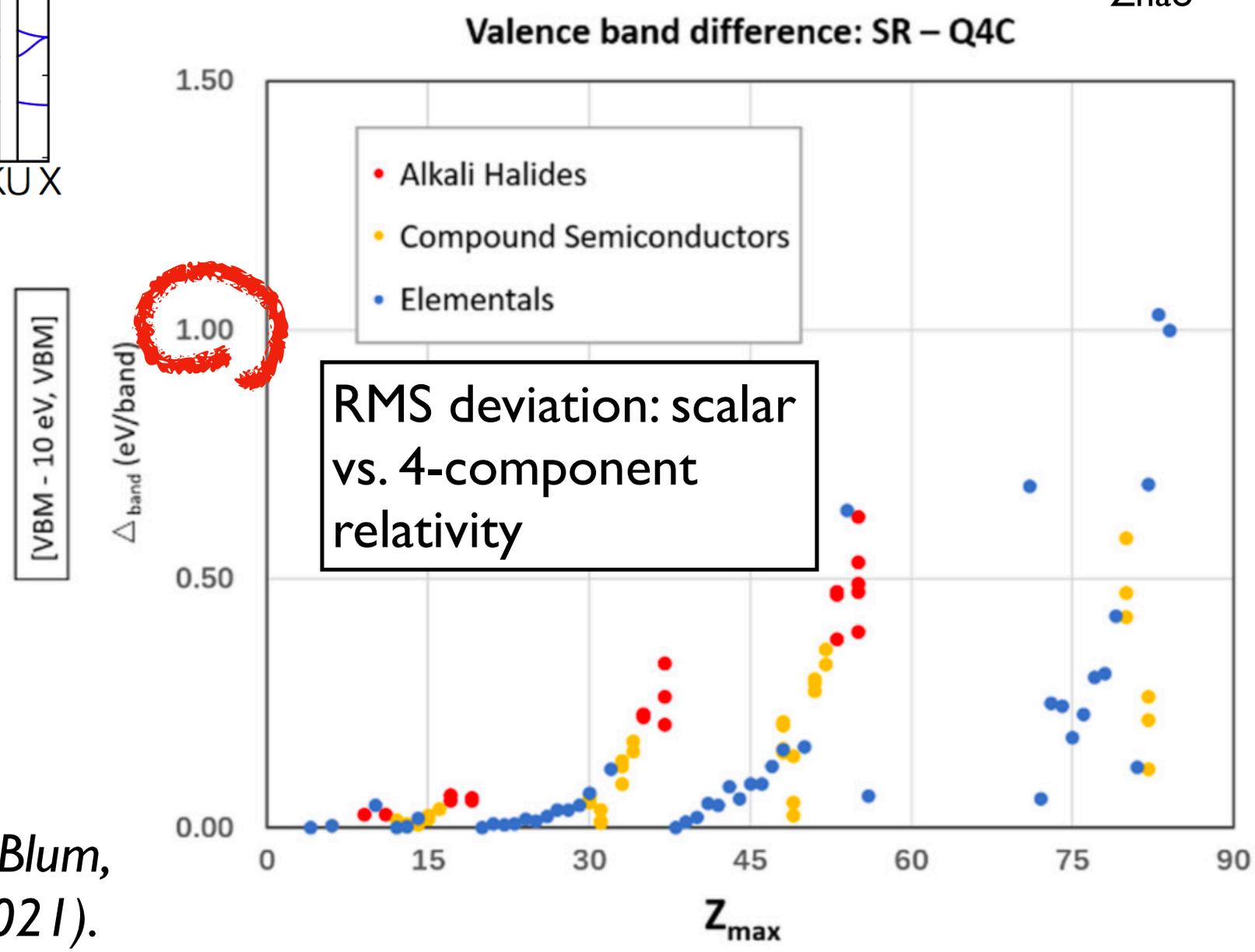
Benchmark: Scalar vs. 4-Component Relativistic Band Structures



Clearly, scalar relativistic energy band structures are not acceptable for heavy elements.

Dr. Rundong Zhao

45 elemental structures,
37 compound semiconductors,
21 strongly ionic compounds;
66 elements included



Zhao, Yu, Zhang, Xiao, Zhang, Blum, Phys. Rev. B ***103****, 245144 (2021).*

Benchmark: Q4C vs. Wien2k Relativistic Band Structures

Q4C: Band structures ~equal to Wien2k, selfconsistent SOC - in fact likely slightly better for 5*d* elements

Dr. Rundong Zhao

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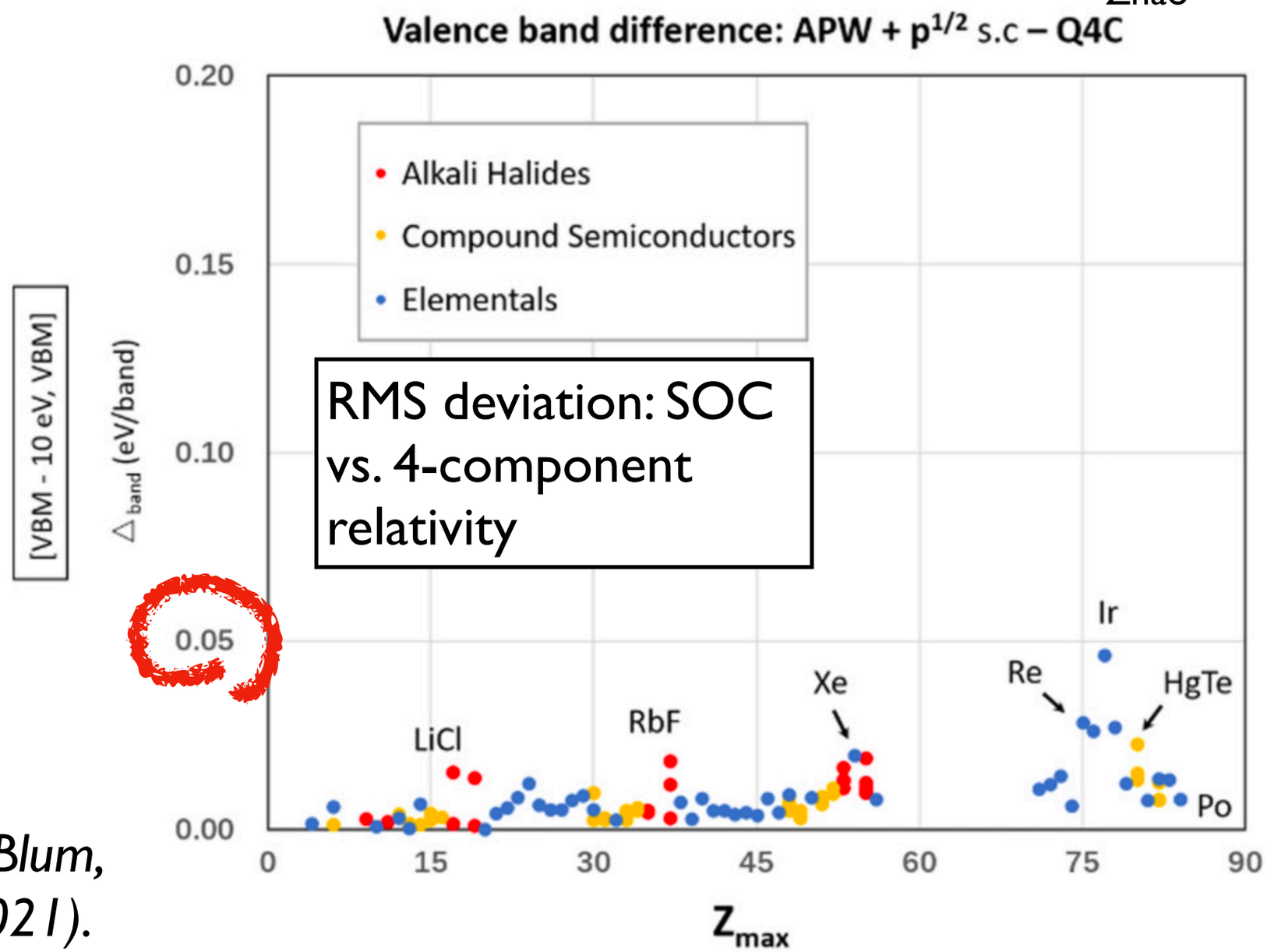

Zhao, Yu, Zhang, Xiao, Zhang, Blum, Phys. Rev. B ***103****, 245144 (2021).*

Benchmark: SOC vs. 4-Component Relativistic Band Structures

Non-selfconsistent SOC shows acceptable results (error less than typical error of hybrid DFT or better)

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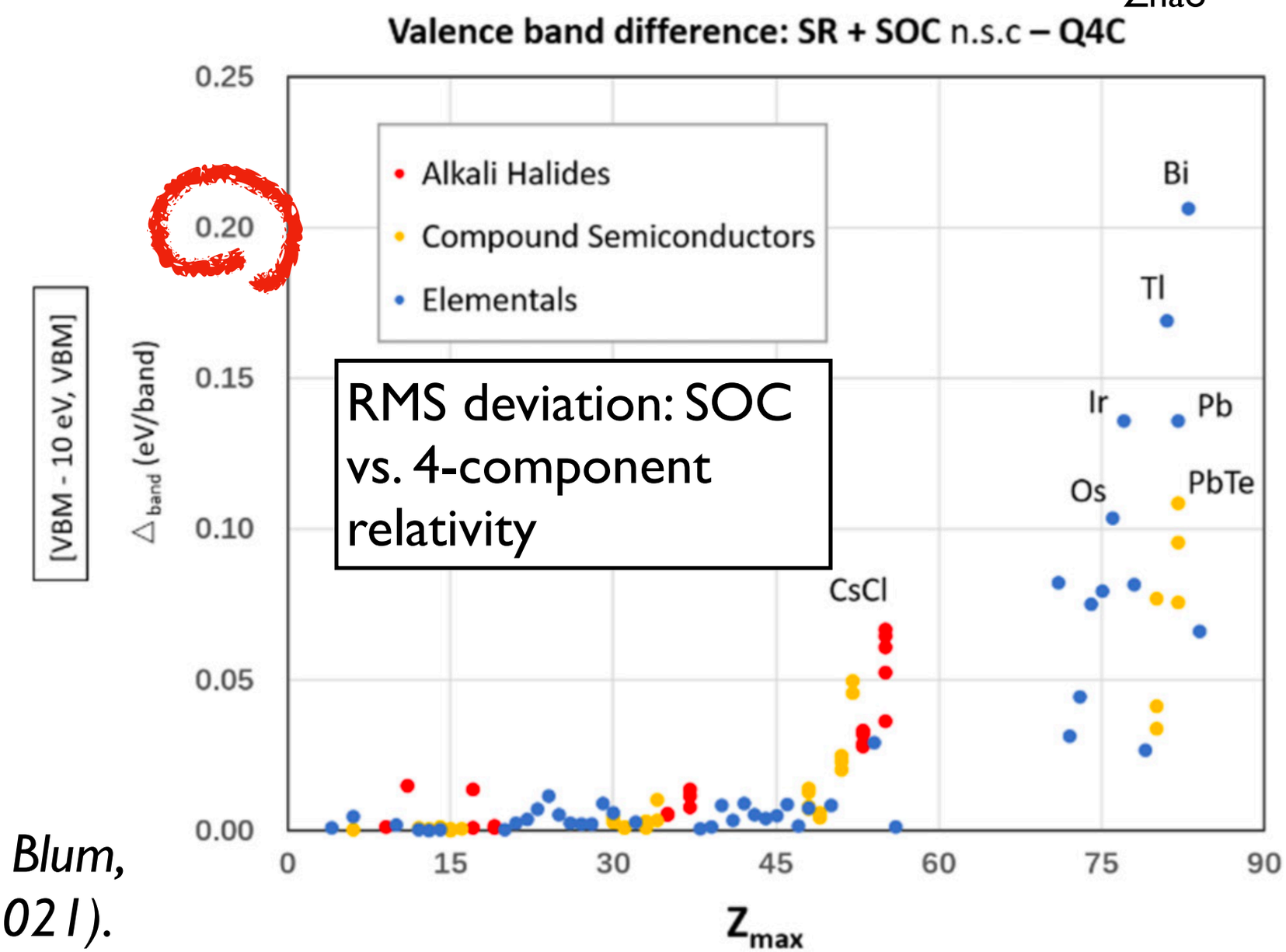
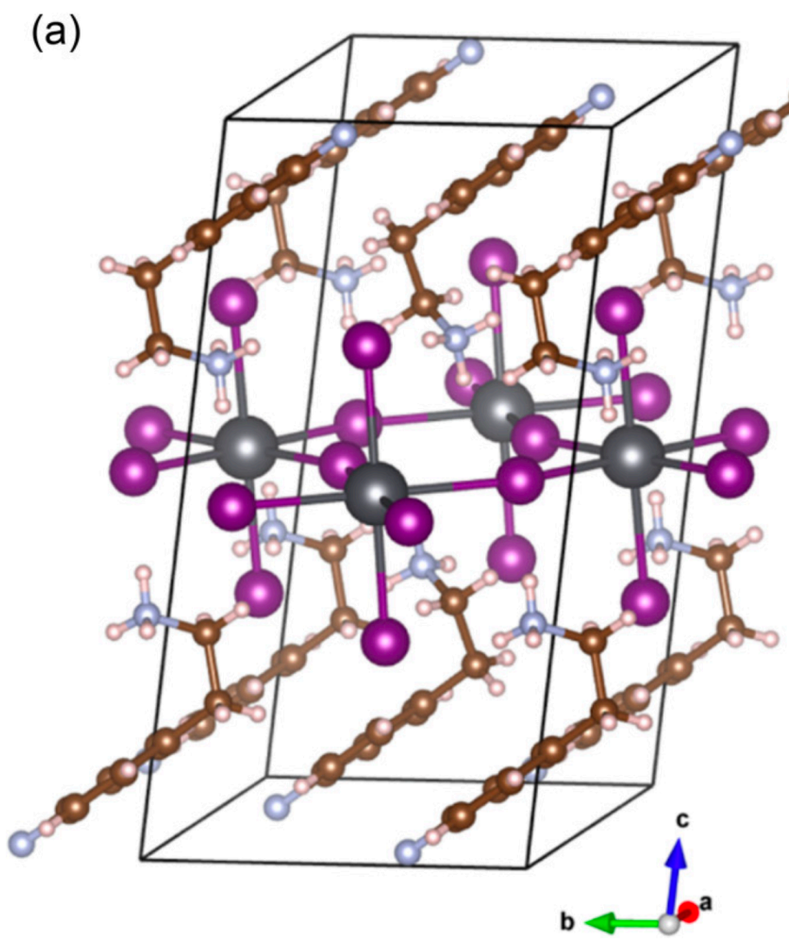
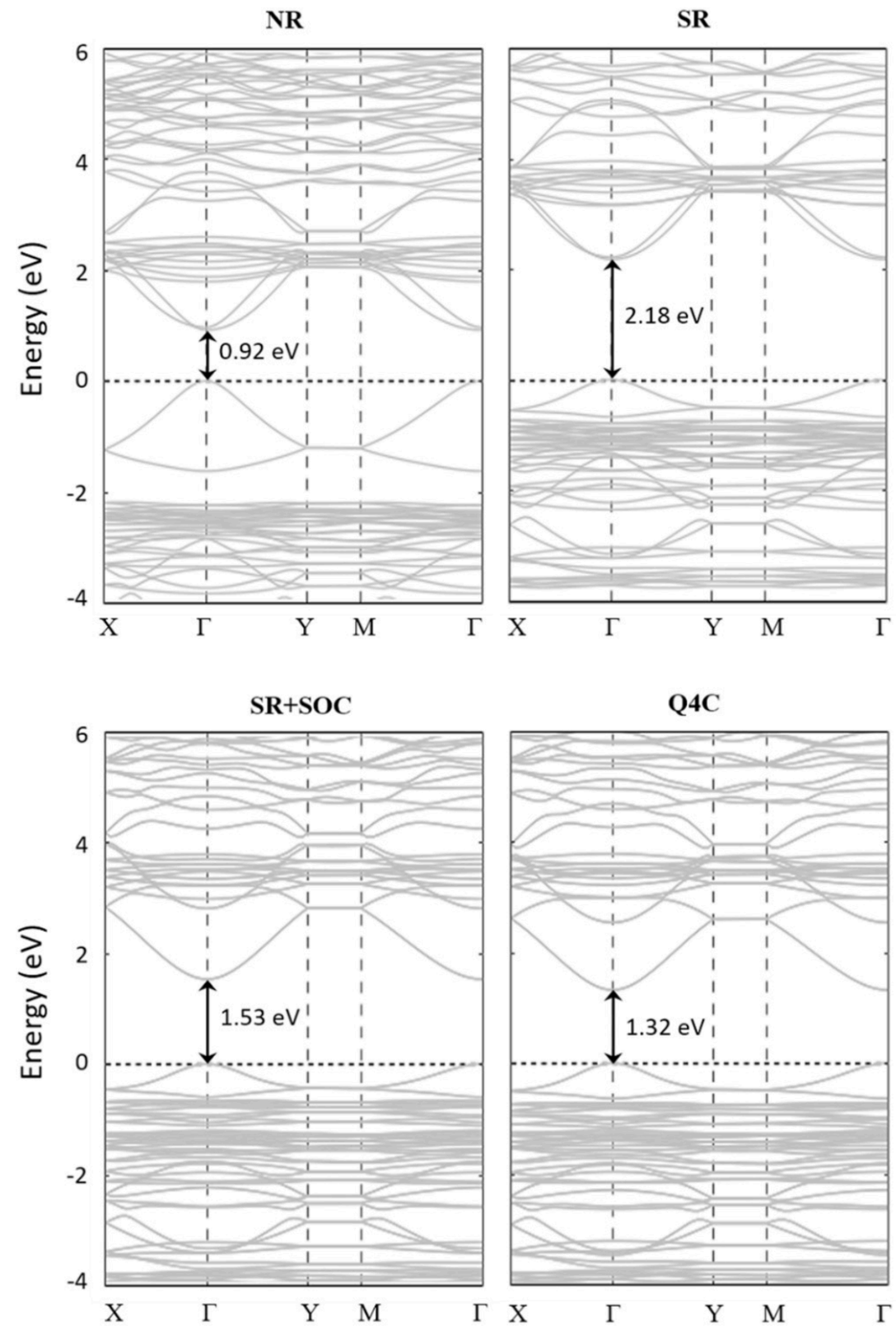


Zhao, Yu, Zhang, Xiao, Zhang, Blum, Phys. Rev. B ***103****, 245144 (2021).*

Q4C - Straightforward Pathway to Large Systems

Energy band structure,
$(FPEA)_2PbI_4$, 96 atoms/unit cell
FHI-aims, tight settings, DFT-PBE

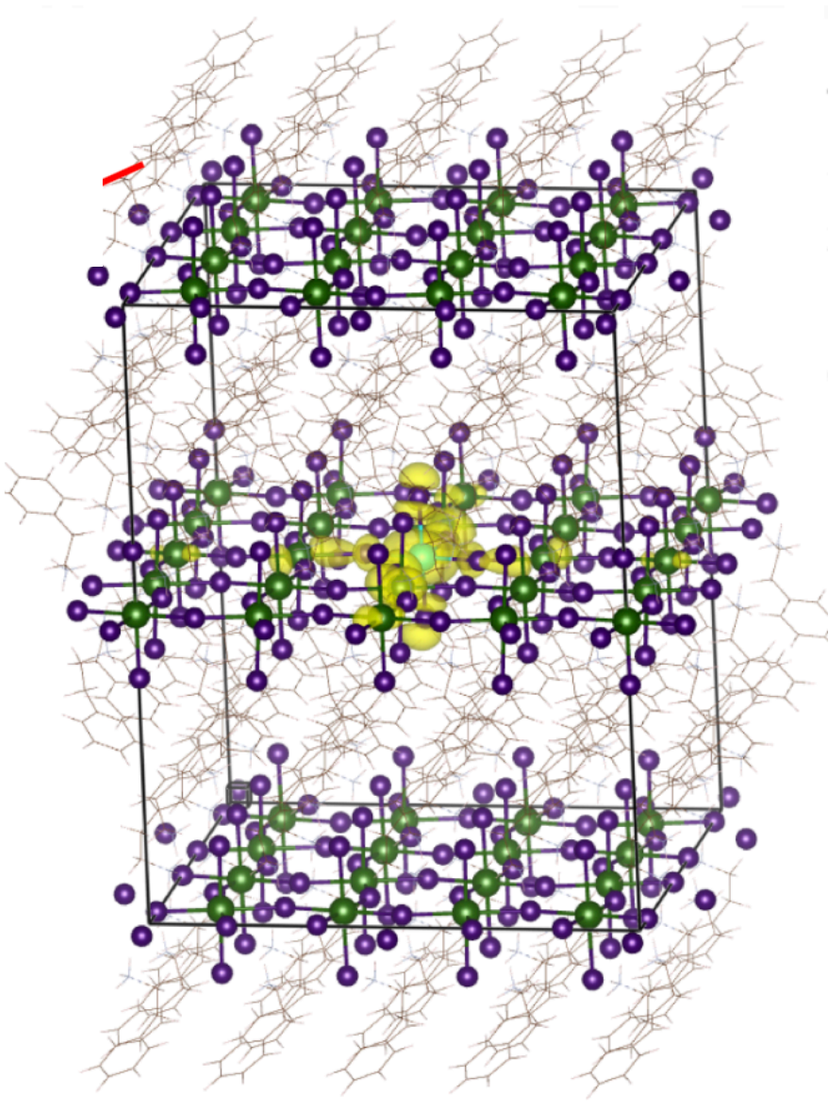




Zhao, Yu, Zhang, Xiao, Zhang, Blum, Phys. Rev. B ***103****, 245144 (2021).*

GIMS - Graphical Interface for Materials Science

Journal of Open Source Software, 6(57), 2767 (2021). https://doi.org/10.21105/joss.02767



Bi/$(PEA)_2PbI_4$
1,504 atom supercell

Recurring problem in complex materials simulations:
Too many moving pieces.

- Many atoms to keep track of
- Algorithmic choices must match problem
- Easy to skip cross-checks (e.g., structure validation) when working remotely & with text editors
- High cost if mistakes are found during production simulation because they were buried in text files.

Jupyter notebooks & workflows help, but need programming skills and still leave moving pieces.

Well thought out graphical interfaces that guide users solve this problem - but when we checked (2019), we did not find an open-source, cross-code, easy-to-use interface. (Sorry if we missed one.)

GIMS - Graphical Interface for Materials Science

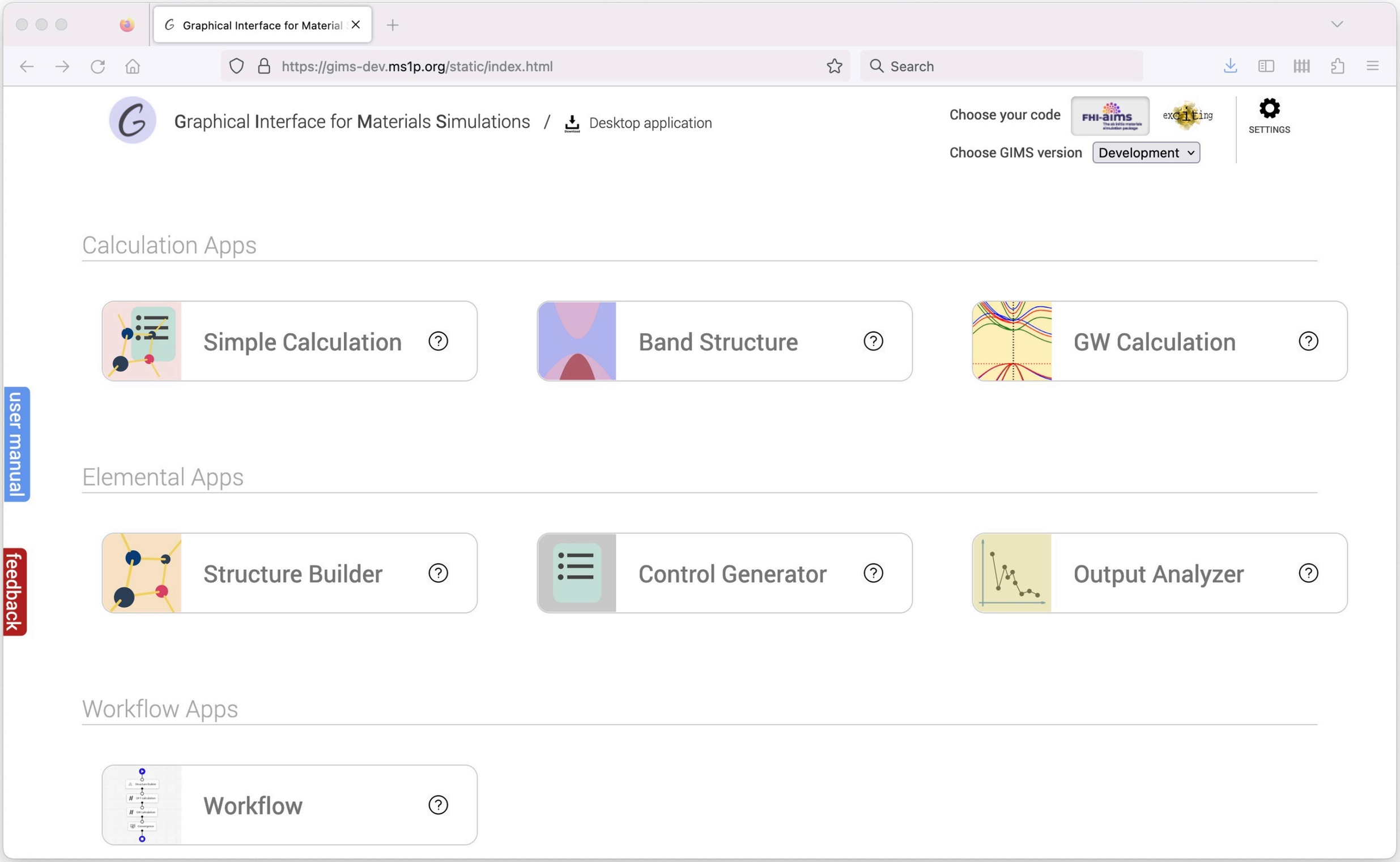
Journal of Open Source Software, 6(57), 2767 (2021). https://doi.org/10.21105/joss.02767

https://gims.ms1p.org

<u>Our Solution Approach:</u>

- Browser-based graphical interface (no installation requirements, "just works")
- Support for multiple codes planned at the outset (currently FHI-aims, exciting, more would be great)
- Javascript atop python layer, widely used and code-agnostic open-source software underneath (ASE, spglib)
- Focus on simulation setup, data evaluation (not trying to drive computers)
- Local desktop version without web server via flask.

GIMS Core Functionality: Separated into Apps



https://gims.ms1p.org

Generic, Editable Structure Builder

Graphical Interface for Material

https://gims-dev.ms1p.org/static/index.html#StructureBuilder

Search

Graphical Interface for Materials Simulations / StructureBuilder

Choose your code FHI-aims exciting

Choose GIMS version Development

SETTINGS

CdSrTa.in ×

+

Import Export

View options

OFF Edit structure

0/0

Lattice Vectors

All numbers in units of Å

a:	7.550000	0.000000	0.000000
b:	0.000000	7.550000	0.000000
c:	0.000000	0.000000	7.550000

Scale atom positions with lattice vectors

Remove Lattice Vectors

Structure Info

Supercell

Standardized Cells

Surface (Slab) Construction

Basis Atoms

1/2

New atom

Fractional coordinates

Constrain all atoms

Click on the species color to specify more atomic properties (e.g. initial moment).

All numbers in units of Å

1.	0.000000	3.775000	3.775000	Cd
2.	5.662500	5.662500	1.887500	Sr

Cd

Sr

Ta

b

a

c

α

β

γ

user manual

feedback

Undo/Redo Ctrl+Z / Ctrl+Y

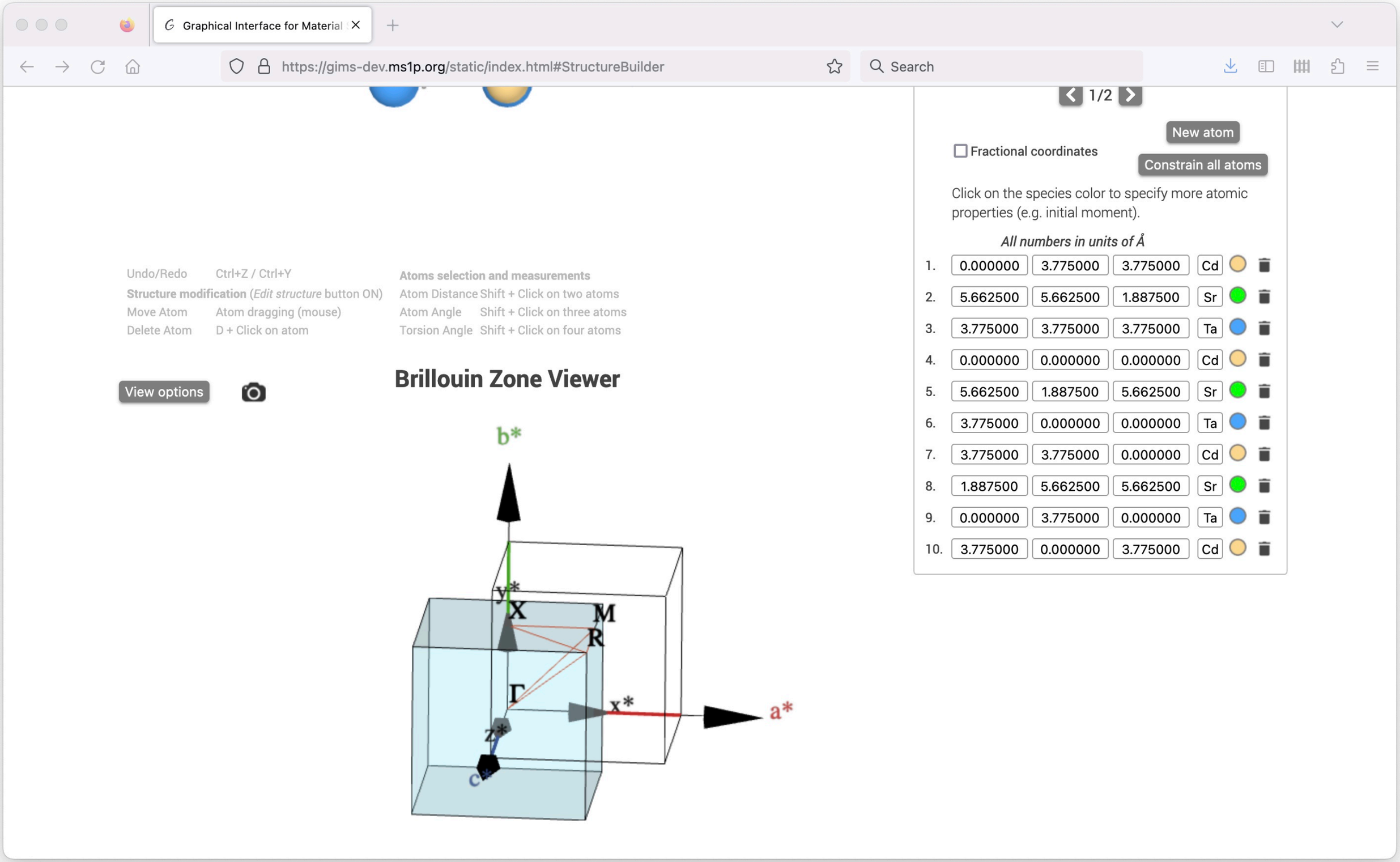
Structure modification (*Edit structure* button ON)

Atoms selection and measurements

Atom Distance Shift + Click on two atoms

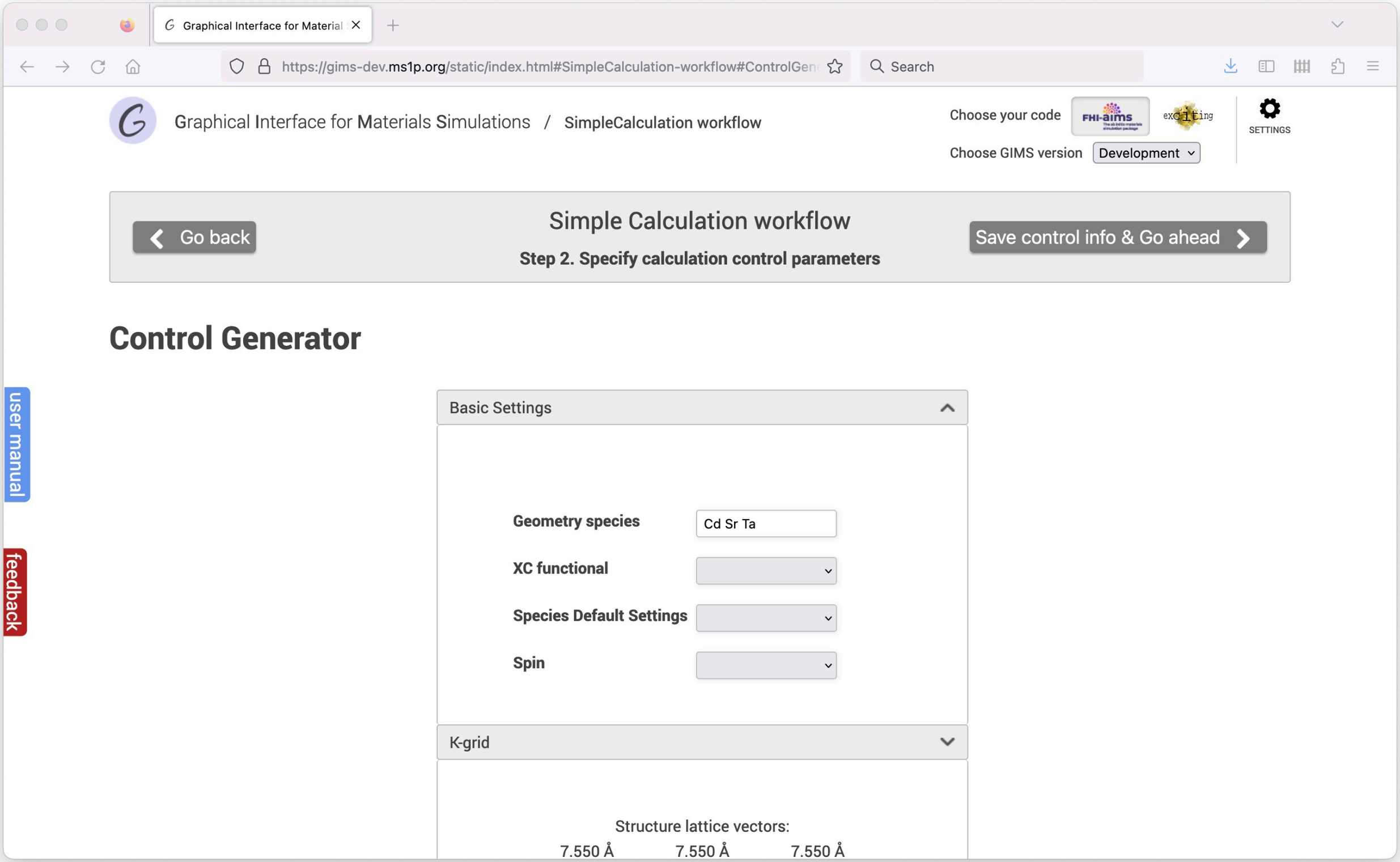
https://gims.ms1p.org

For Periodic Systems, Brillouin Zone Included



https://gims.ms1p.org

Code Specific Setup - Easy to Replicate for Other Codes



https://gims.ms1p.org

Simulation Analysis: Load Output When Complete

Graphical Interface for Material

https://gims-dev.ms1p.org/static/index.html#OutputAnalyzer

Search

Graphical Interface for Materials Simulations / OutputAnalyzer

Choose your code FHI-aims exciting

SETTINGS

Choose GIMS version Development

Output Analyzer

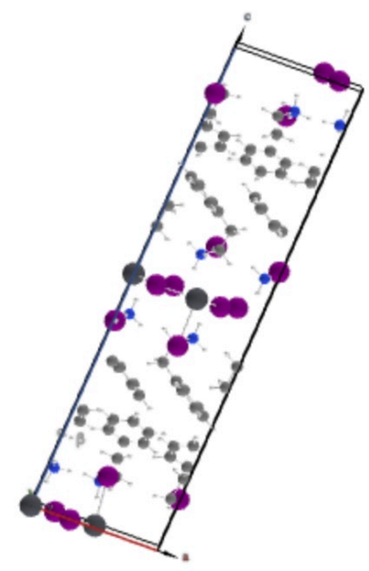
Import output files: Browse... aims.out Select a folder containing the files

Chemical Formula	Pb4I16N8C64H96
Number of atoms	188

View options

52/52

Pb
I
N
C
H



Results

Total Energy (eV)	-5585978.18794
Fermi Energy (eV)	-5.61014
Highest occupied state (eV)	-6.20064
Lowest unoccupied state (eV)	-4.12467
Estimated HOMO-LUMO gap (eV)	2.07597
Cell Volume (Å^3)	2497.93671

Download final geometry

Calculation Summary

user manual

feedback

https://gims.ms1p.org

FHI-aims: Current Ongoing Directions

- GW for periodic structures

  Xinguo Ren, Minye Zhang, Uthpala Herath, Florian Merz et al.

  works in a *k*-space implementation - next steps: computational cost, spin-orbit coupling, GPU, accurate basis sets

  Xinguo Ren et al., Phys. Rev. Materials 5, 013807 (2021).

- BSE@GW for periodic structures

  Ruiyi Zhao, Yosuke Kanai et al.

  works (outside mainline) - similar next steps to periodic *GW*

- Nuclear-electronic orbitals (NEO) for quantum nuclei

  Jianhang Xu, Ruiyi Zhao, Sharon Hammes-Schiffer, Yosuke Kanai et al.

  works - next steps: RT-NEO-TDDFT

  Jianhang Xu et al., J. Chem. Phys. 156, 224111 (2022).

- Four-component relativity (Q4C)

  Rundong Zhao, Wentao Zhang, Dorothea Golze, Qing-Long Liu et al.

  Single-point, PBE benchmarked - next steps: large systems, forces, open shell)

  Rundong Zhao et al., Phys. Rev. B 103, 255144 (2021).

- Many more! (CC4S, solvation, QM/MM, GIMS, SEAMM@MolSSI, …)

Summary

FHI-aims
The ab initio materials simulation package

Large systems, accurate density functionals, very active "beyond electronic DFT" platform in FHI-aims. Key scalability improvements for exact exchange. Robust, easy to use, efficient.

ELSI
ELPA
Electronic Structure Library
cecam

Common libraries (ELPA, ELSI & supported libraries, libxc etc.) furnish essential parts of the numerics, bridge to other projects.

G

Make electronic structure theory accessible to students who are scientists but not strong programmers: GIMS.

MS1P
NSF
CHOISE
Center for Hybrid Organic Inorganic Semiconductors for Energy
U.S. DEPARTMENT OF ENERGY
Office of Science
U.S. DEPARTMENT OF ENERGY
INCITE
LEADERSHIP COMPUTING
MIRA
Argonne NATIONAL LABORATORY
ALCF
XSEDE
Extreme Science and Engineering Discovery Environment
NERSC
NREL
Transforming ENERGY