Current Status of the OpenMX Code

- History of OpenMX
- Basic theoretical framework
- Parallelization of OpenMX
- Some new functionalities
- Future developments

https://www.openmx-square.org/

Taisuke Ozaki

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The 4th OpenMX developer's meeting on Nov. 9-10, 2023

OpenMX Open source package for Material eXplorer

- Software package for density functional calculations of molecules and bulks
- Norm-conserving pseudopotentials (PPs)
- Variationally optimized numerical atomic basis functions

Basic functionalities

- SCF calc. by LDA, GGA, DFT+U
- Total energy and forces on atoms
- Band dispersion and density of states
- Geometry optimization by BFGS, RF, EF
- Charge analysis by Mullken, Voronoi, ESP
- Molecular dynamics with NEV and NVT ensembles
- Charge doping
- Fermi surface
- Analysis of charge, spin, potentials by cube files
- Database of optimized PPs and basis funcitons

Extensions

- O(N) and low-order scaling diagonalization
- Non-collinear DFT for non-collinear magnetism
- Spin-orbit coupling included self-consistently
- Electronic transport by non-equilibrium Green function
- Electronic polarization by the Berry phase formalism
- Maximally localized Wannier functions
- Effective screening medium method for biased system
- Reaction path search by the NEB method
- Band unfolding method
- STM image by the Tersoff-Hamann method
- etc.

History of OpenMX

2000 Start of development

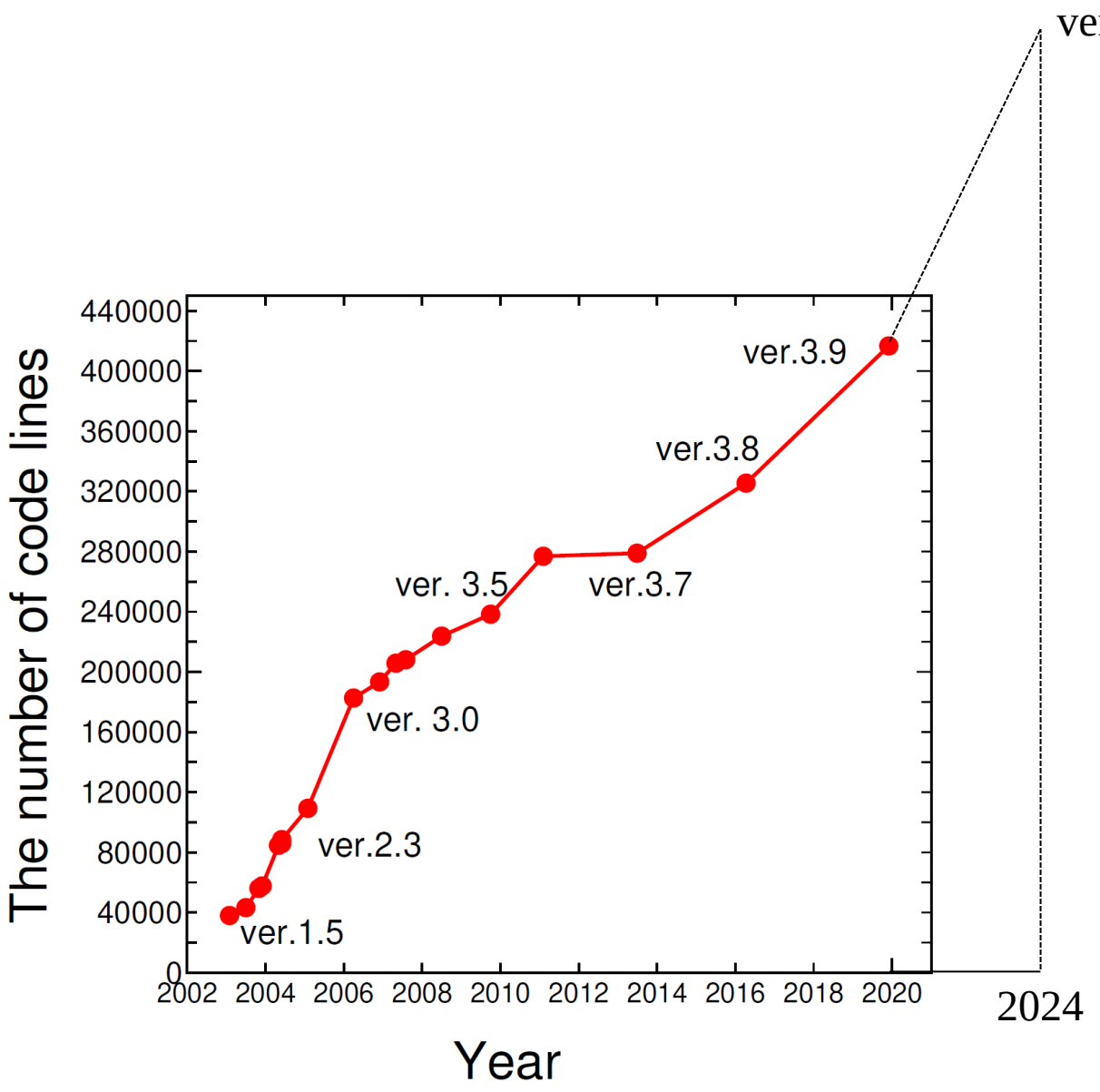
2003 Public release (GNU-GPL)

2003 Collaboration:
AIST, NIMS, SNU
KAIST, JAIST,
Kanazawa Univ.
CAS, UAM
NISSAN, Fujitsu Labs.
etc.

2019 19 public releases
Latest version: 3.9

http://www.openmx-square.org

Development of OpenMX code



- NPT molecular dynamics by Dr. Iitaka of RIKEN
- Closest Wannier functions
- COHP and COOP analysis
- Finite electric field method based on the Berry phase by Dr. Yamaguchi
- Contracted diagonalization
- Tensor regression model for on-the-fly machine learning potential by Mr. Li.
- Rational function method of variable cell optimization with arbitrary constraint

Contributors to OpenMX development

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Materials studied by OpenMX

First characterization of silicene on ZrB_2 in collaboration with experimental groups

A. Fleurence et al., Phys. Rev. Lett. 108, 245501 (2012).

First identification of Jeff=1/2 Mott state of Ir oxides

B.J. Kim et al., Phys. Rev. Lett. 101, 076402 (2008).

Theoretical proposal of topological insulators

C.-H. Kim et al., Phys. Rev. Lett. 108, 106401 (2012).
H. Weng et al., Phy. Rev. X 4, 011002 (2014).

First-principles molecular dynamics simulations for Li ion battery

T. Ohwaki et al., J. Chem. Phys. 136, 134101 (2012).
T. Ohwaki et al., J. Chem. Phys. 140, 244105 (2014).

Magnetic anisotropy energy of magnets

Z. Torbatian et al., Appl. Phys. Lett. 104, 242403 (2014).
I. Kitagawa et al., Phys. Rev. B 81, 214408 (2010).

Electronic transport of graphene nanoribbon on surface oxidized Si

H. Jippo et al., Appl. Phys. Express 7, 025101 (2014).
M. Ohfuchi et al., Appl. Phys. Express 4, 095101 (2011).

Interface structures of carbide precipitate in bcc-Fe

H. Sawada et al., Modelling Simul. Mater. Sci. Eng. 21, 045012 (2013).

Universality of medium range ordered structure in amorphous metal oxides

K. Nishio et al., Phys. Rev. Lett. 340, 155502 (2013).

Materials treated so far

Silicene, graphene
Carbon nanotubes
Transition metal oxides
Topological insulators
Intermetallic compounds
Molecular magnets
Rare earth magnets
Lithium ion related materials
Structural materials
etc.

About 1200 published papers

Basic theoretical framework

- Density functional theory
- LCPAO method
- Basis functions
- Pseudopotentials
- Total energy

LCPAO method
(Linear-Combination of Pseudo Atomic Orbital Method)

One-particle KS orbital

$$\psi_{\sigma\mu}^{(\mathbf{k})}(\mathbf{r}) = \frac{1}{\sqrt{N}} \sum_{\mathrm{n}}^{N} \mathrm{e}^{\mathrm{i}\mathbf{R}_{\mathrm{n}}\cdot\mathbf{k}} \sum_{i\alpha} c_{\sigma\mu,i\alpha}^{(\mathbf{k})} \phi_{i\alpha}(\mathbf{r} - \tau_i - \mathbf{R}_{\mathrm{n}}),$$

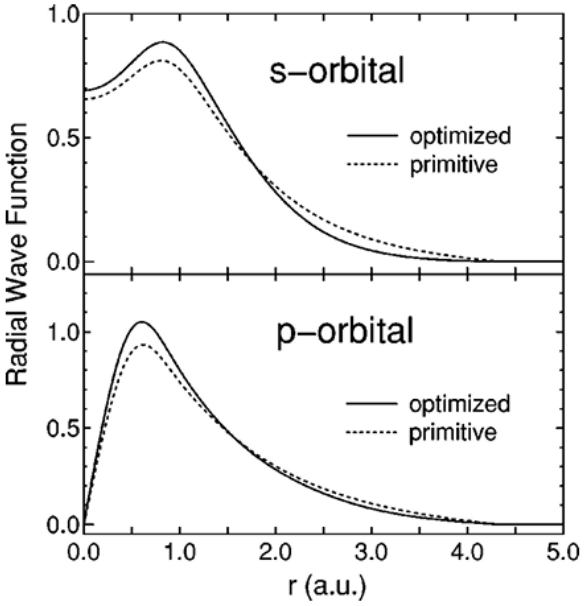
is expressed by a linear combination of atomic like orbitals in the method.

$$\phi(\mathbf{r}) = Y_l^m(\hat{\mathbf{r}})R(r)$$

Features:

- It is easy to interpret physical and chemical meanings, since the KS orbitals are expressed by the atomic like basis functions.
- It gives rapid convergent results with respect to basis functions due to physical origin. (however, it is not a complete basis set, leading to difficulty in getting full convergence.)
- The memory and computational effort for calculation of matrix elements are O(N).
- It well matches the idea of linear scaling methods.

Radial orbitals of carbon atom



Variational optimization of basis functions

One-particle wave functions

$$\psi_\mu(\mathbf{r}) = \sum_{i\alpha} c_{\mu,i\alpha}\phi_{i\alpha}(\mathbf{r}-\mathbf{r}_i)$$

Contracted orbitals

$$\phi_{i\alpha}(\mathbf{r}) = \sum_q a_{i\alpha q}\chi_{i\eta}(\mathbf{r})$$

The variation of E with respect to *c* with fixed *a* gives

$$\partial E_{\mathrm{tot}}/\partial c_{\mu,i\alpha} = 0 \quad \rightarrow \quad \sum_{j\beta} \langle \phi_{i\alpha}|\hat{H}|\phi_{j\beta}\rangle c_{\mu,j\beta} = \varepsilon_\mu \sum_{j\beta} \langle \phi_{i\alpha}|\phi_{j\beta}\rangle c_{\mu,j\beta}$$

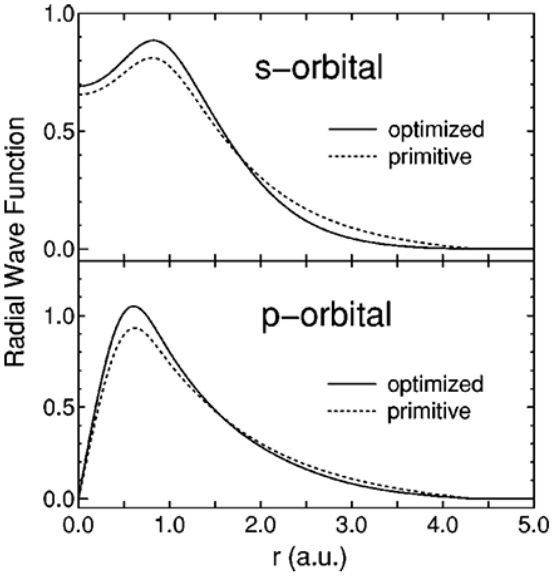
Regarding c as dependent variables on *a* and assuming KS eq. is solved self-consistently with respect to *c*, we have

$$\frac{\partial E_{\mathrm{tot}}}{\partial a_{i\alpha q}} = \frac{\delta E_{\mathrm{tot}}}{\delta \rho(\mathbf{r})}\frac{\delta \rho(\mathbf{r})}{\delta a_{i\alpha q}}$$

$$= 2\sum_{j\beta} \left(\Theta_{i\alpha,j\beta}\langle \chi_{i\eta}|\hat{H}|\phi_{j\beta}\rangle - E_{i\alpha,j\beta}\langle \chi_{i\eta}|\phi_{j\beta}\rangle\right)$$

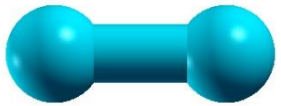
Ozaki, PRB 67, 155108 (2003)

Radial orbitals of carbon atom

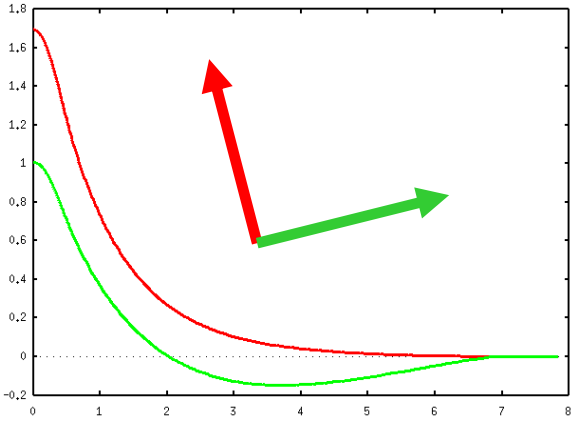
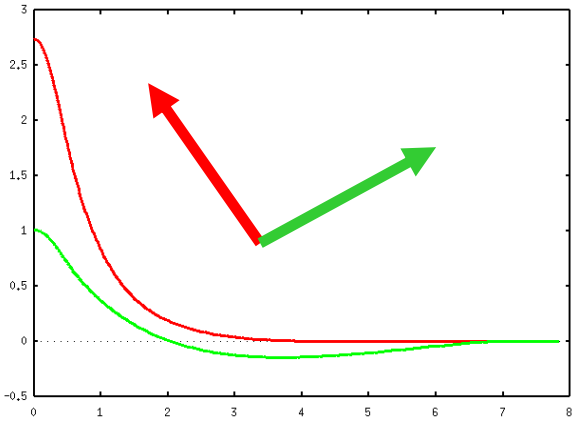
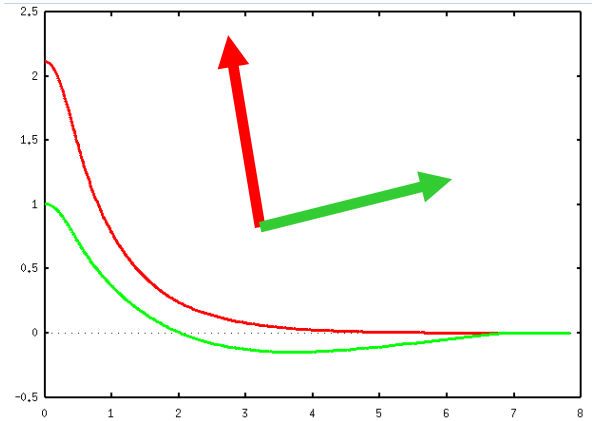


Optimization of basis functions

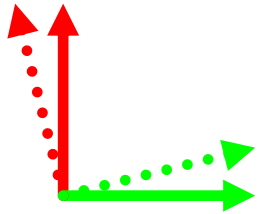
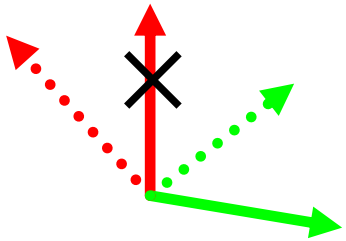
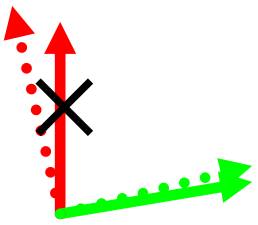
1. Choose typical chemical environments


2. Optimize variationally the radial functions

3. Rotate a set of optimized orbitals within the subspace, and discard the redundant funtions

Norm-conserving Vanderbilt pseudopotential

I. Morrion, D.M. Bylander, and L. Kleinman, PRB 47, 6728 (1993).

The following non-local operator proposed by Vanderbilt guarantees that scattering properties are reproduced around multiple reference energies.

D. Vanderbilt, PRB 41, 7892 (1990).

$$V_{\mathrm{NL}} = \sum_{i,j} B_{ij} |\beta_i\rangle\langle\beta_j|$$

$$|\chi_i\rangle = V_{\mathrm{NL}}^{(i)} |\phi_i\rangle = (\varepsilon_i - T - V_{\mathrm{loc}})|\phi_i\rangle$$

$$B_{ij} = \langle\phi_i|\chi_j\rangle$$

$$|\beta_i\rangle = \sum_j (B^{-1})_{ji} |\chi_j\rangle$$

If the following generalized norm-conserving condition is fulfilled, the matrix B is Hermitian, resulting in that V_{NL} is also Hermitian.

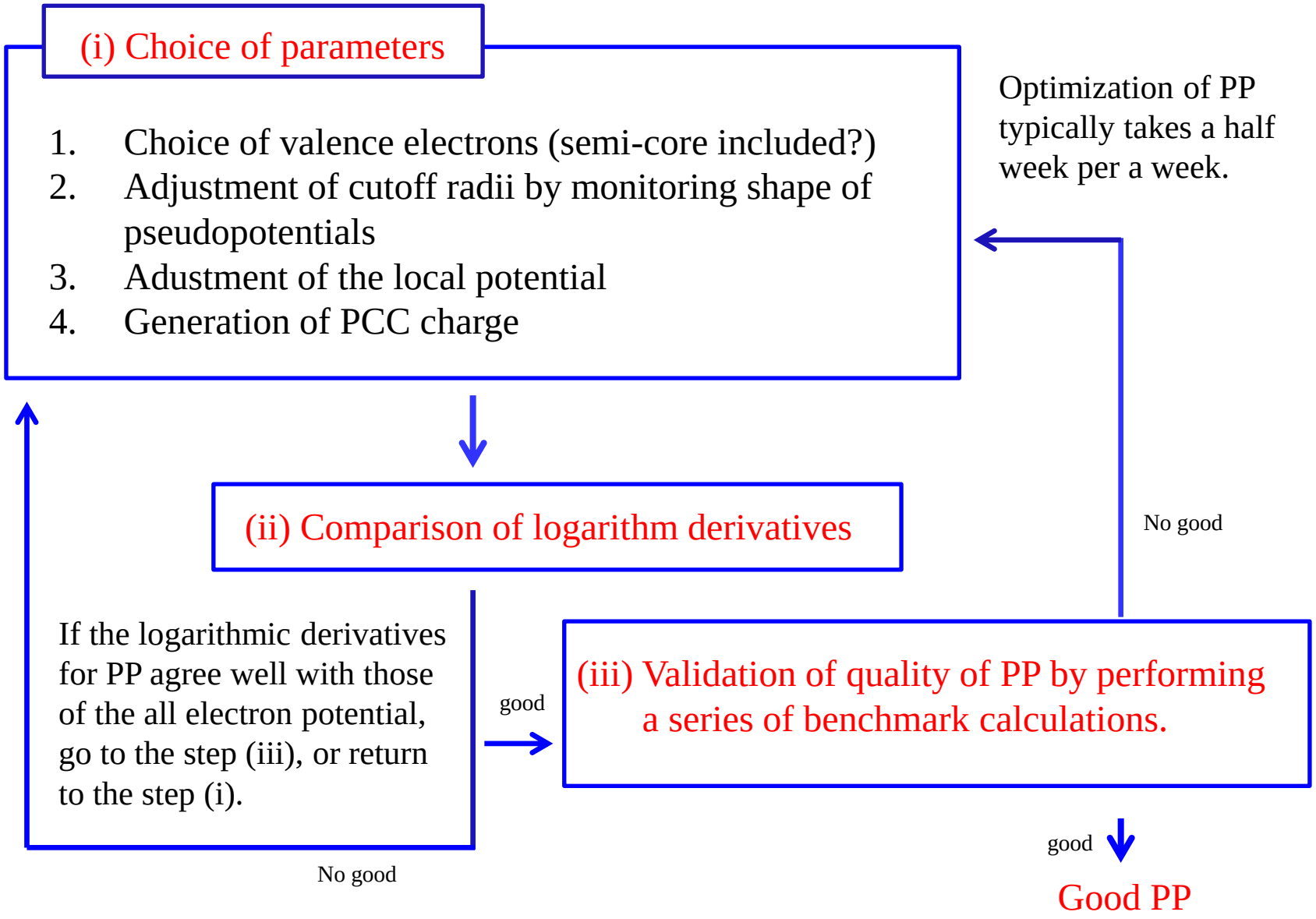
$$Q_{ij} = \langle\psi_i|\psi_j\rangle_R - \langle\phi_i|\phi_j\rangle_R$$

If Q=0, then B-B*=0

$$B_{ij} - B_{ji}^* = (\varepsilon_i - \varepsilon_j) Q_{ij}$$

This is the norm-conserving PP used in OpenMX

Optimization of pseudopotentials



Reproducibility in DFT calcs

RESEARCH ARTICLE

DFT METHODS

Reproducibility in density functional theory calculations of solids

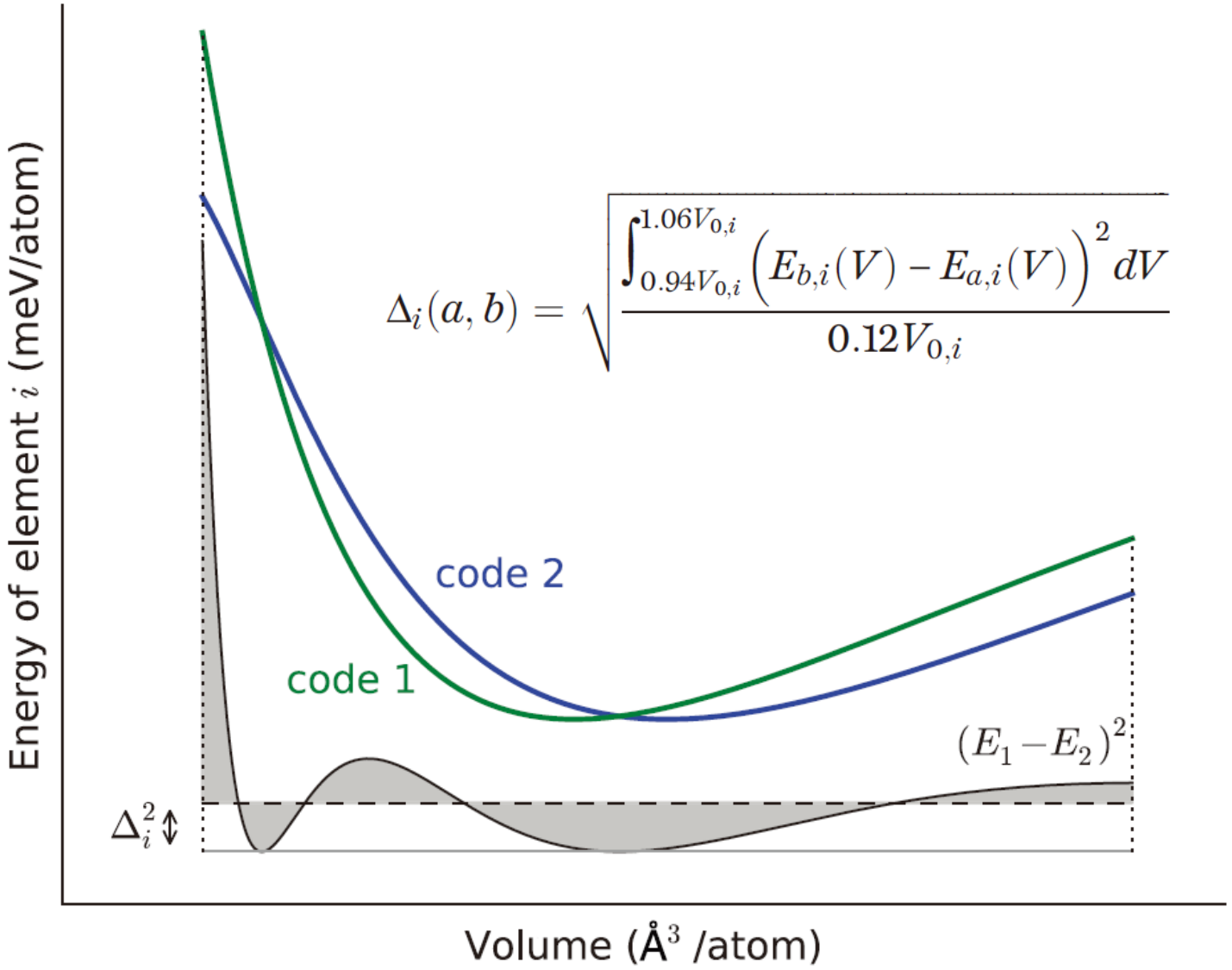
Kurt Lejaeghere,[1*] Gustav Bihlmayer,[2] Torbjörn Björkman,[3,4] Peter Blaha,[5] Stefan Blügel,[2] Volker Blum,[6] Damien Caliste,[7,8] Ivano E. Castelli,[9] Stewart J. Clark,[10] Andrea Dal Corso,[11] Stefano de Gironcoli,[11] Thierry Deutsch,[7,8] John Kay Dewhurst,[12] Igor Di Marco,[13] Claudia Draxl,[14,15] Marcin Dułak,[16] Olle Eriksson,[13] José A. Flores-Livas,[12] Kevin F. Garrity,[17] Luigi Genovese,[7,8] Paolo Giannozzi,[18] Matteo Giantomassi,[19] Stefan Goedecker,[20] Xavier Gonze,[19] Oscar Grånäs,[13,21] E. K. U. Gross,[12] Andris Gulans,[14,15] François Gygi,[22] D. R. Hamann,[23,24] Phil J. Hasnip,[25] N. A. W. Holzwarth,[26] Diana Iuşan,[13] Dominik B. Jochym,[27] François Jollet,[28] Daniel Jones,[29] Georg Kresse,[30] Klaus Koepernik,[31,32] Emine Küçükbenli,[9,11] Yaroslav O. Kvashnin,[13] Inka L. M. Locht,[13,33] Sven Lubeck,[14] Martijn Marsman,[30] Nicola Marzari,[9] Ulrike Nitzsche,[31] Lars Nordström,[13] Taisuke Ozaki,[34] Lorenzo Paulatto,[35] Chris J. Pickard,[36] Ward Poelmans,[1,37] Matt I. J. Probert,[25] Keith Refson,[38,39] Manuel Richter,[31,32] Gian-Marco Rignanese,[19] Santanu Saha,[20] Matthias Scheffler,[15,40] Martin Schlipf,[22] Karlheinz Schwarz,[5] Sangeeta Sharma,[12] Francesca Tavazza,[17] Patrik Thunström,[41] Alexandre Tkatchenko,[15,42] Marc Torrent,[28] David Vanderbilt,[23] Michiel J. van Setten,[19] Veronique Van Speybroeck,[1] John M. Wills,[43] Jonathan R. Yates,[29] Guo-Xu Zhang,[44] Stefaan Cottenier[1,45*]

Science 351, aad3000 (2016)

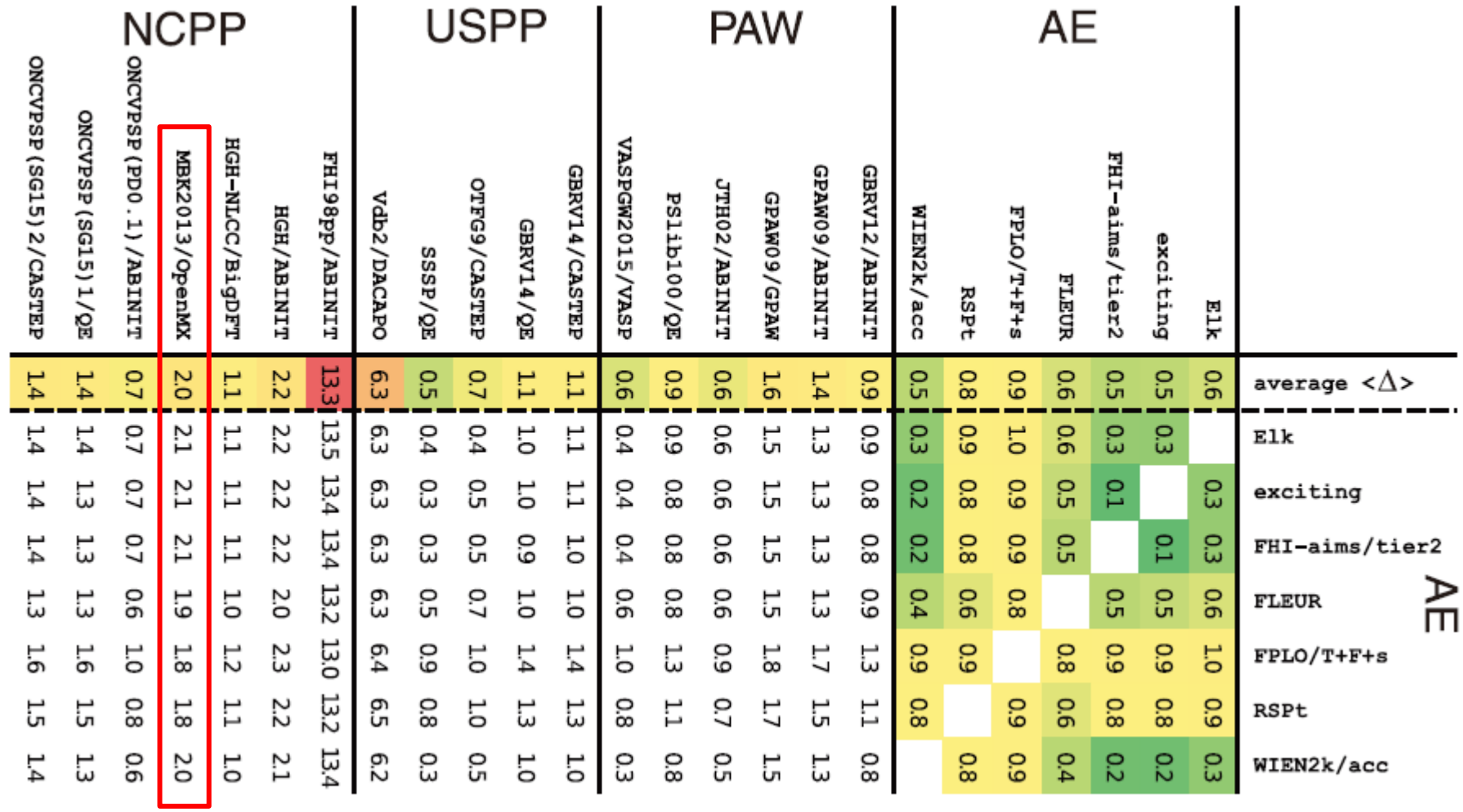
15 codes
69 researchers
71 elemental bulks
GGA-PBE
Scalar relativistic

Δ-gauge

A way of comparing accuracy of codes



Comparison of codes by Δ-gauge

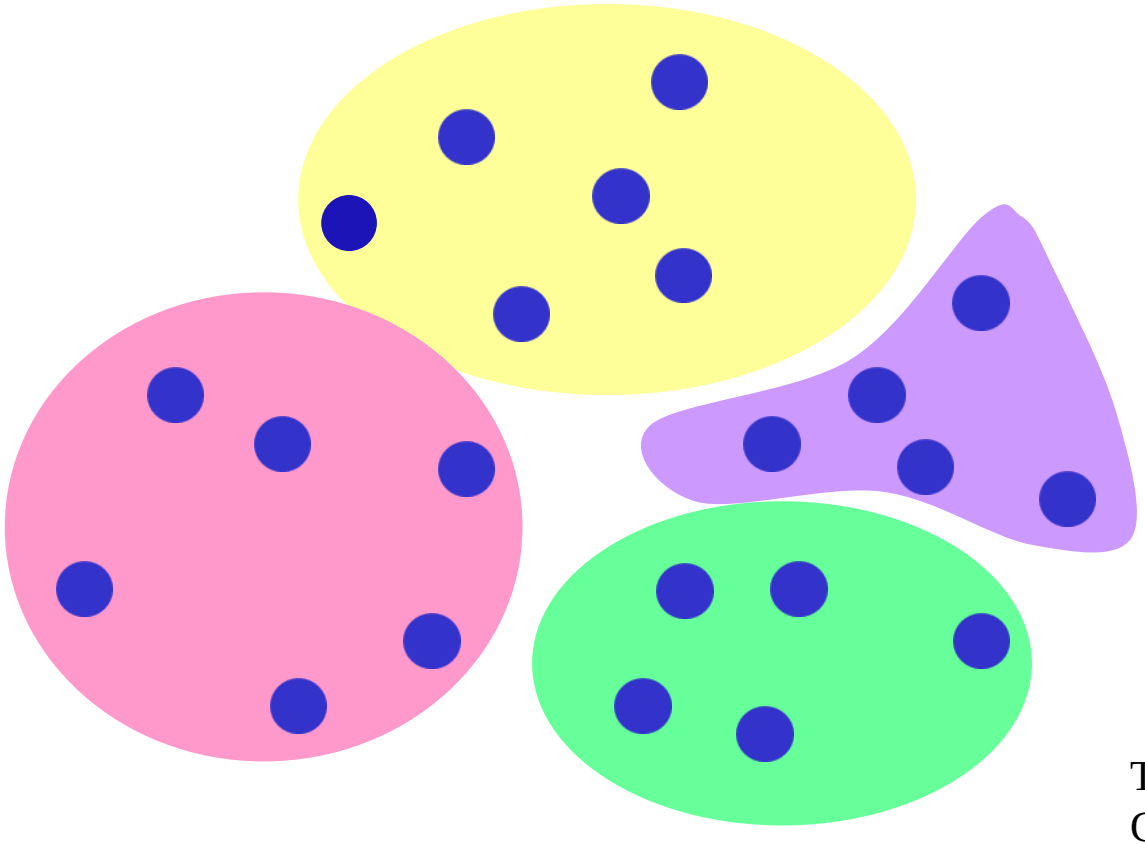


	average <Δ>	Elk	exciting	FHI-aims/tier2	FLEUR	FPLO/T+F+s	RSPt	WIEN2k/acc
Elk	0.6		0.3	0.3	0.6	1.0	0.9	0.3
exciting	0.5	0.3		0.1	0.5	0.9	0.8	0.2
FHI-aims/tier2	0.5	0.3	0.1		0.5	0.9	0.8	0.2
FLEUR	0.6	0.6	0.5	0.5		0.8	0.6	0.4
FPLO/T+F+s	0.9	1.0	0.9	0.9	0.8		0.9	0.9
RSPt	0.8	0.9	0.8	0.8	0.6	0.9		0.8
WIEN2k/acc	0.5	0.3	0.2	0.2	0.4	0.9	0.8	
GBRV12/ABINIT	0.9	0.9	0.8	0.8	0.9	1.3	1.1	0.8
GPAW09/ABINIT	1.4	1.3	1.3	1.3	1.3	1.7	1.5	1.3
GPAW09/GPAW	1.6	1.5	1.5	1.5	1.5	1.8	1.7	1.5
JTH02/ABINIT	0.6	0.6	0.6	0.6	0.6	0.9	0.7	0.5
PSlib100/QE	0.9	0.9	0.8	0.8	0.8	1.3	1.1	0.8
VASPGW2015/VASP	0.6	0.4	0.4	0.4	0.6	1.0	0.8	0.3
GBRV14/CASTEP	1.1	1.1	1.1	1.0	1.0	1.4	1.3	1.0
GBRV14/QE	1.1	1.0	1.0	0.9	1.0	1.4	1.3	1.0
OTFG9/CASTEP	0.7	0.4	0.5	0.5	0.7	1.0	1.0	0.5
SSSP/QE	0.5	0.4	0.3	0.3	0.5	0.9	0.8	0.3
Vdb2/DACAPO	6.3	6.3	6.3	6.3	6.3	6.4	6.5	6.2
FHI98pp/ABINIT	13.3	13.5	13.4	13.4	13.2	13.0	13.2	13.4
HGH/ABINIT	2.2	2.2	2.2	2.2	2.0	2.3	2.2	2.1
HGH-NLCC/BigDFT	1.1	1.1	1.1	1.1	1.0	1.2	1.1	1.0
MBK2013/OpenMX	2.0	2.1	2.1	2.1	1.9	1.8	1.8	2.0
ONCVPSP(PD0.1)/ABINIT	0.7	0.7	0.7	0.7	0.6	1.0	0.8	0.6
ONCVPSP(SG15)1/QE	1.4	1.4	1.3	1.3	1.3	1.6	1.5	1.3
ONCVPSP(SG15)2/CASTEP	1.4	1.4	1.4	1.4	1.3	1.6	1.5	1.4

The mean Δ-gauge of OpenMX is 2.0meV/atom.

Atomic 3D atomic partitioning

How one can partition atoms to minimize communication and memory usage in the parallel calculations ?



Requirement:

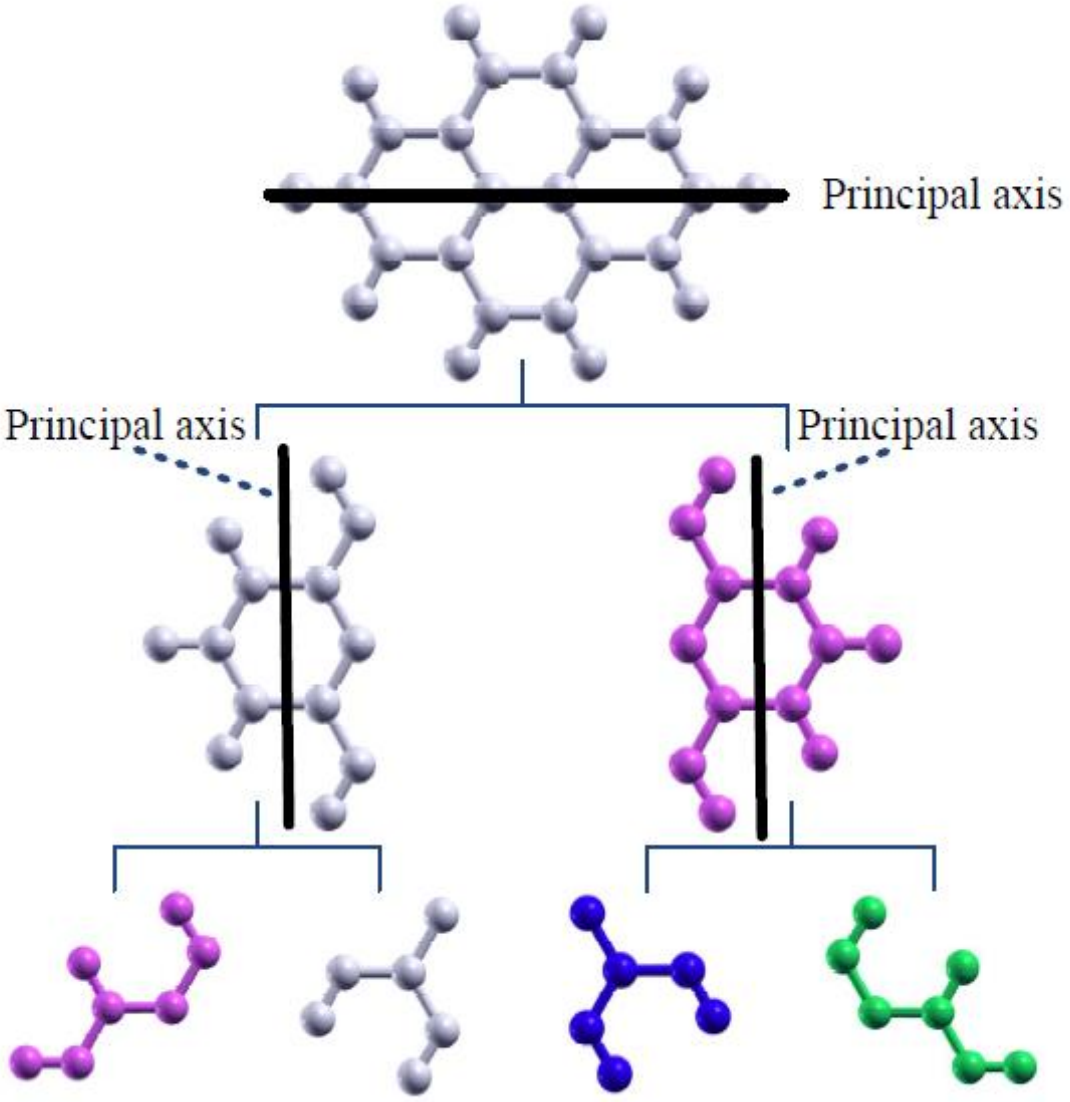
- Locality
- Same computational cost
- Applicable to any systems
- Small computational overhead

T.V.T. Duy and T. Ozaki, Comput. Phys. Commun. 185, 777-789 (2014).

Recursive atomic partitioning

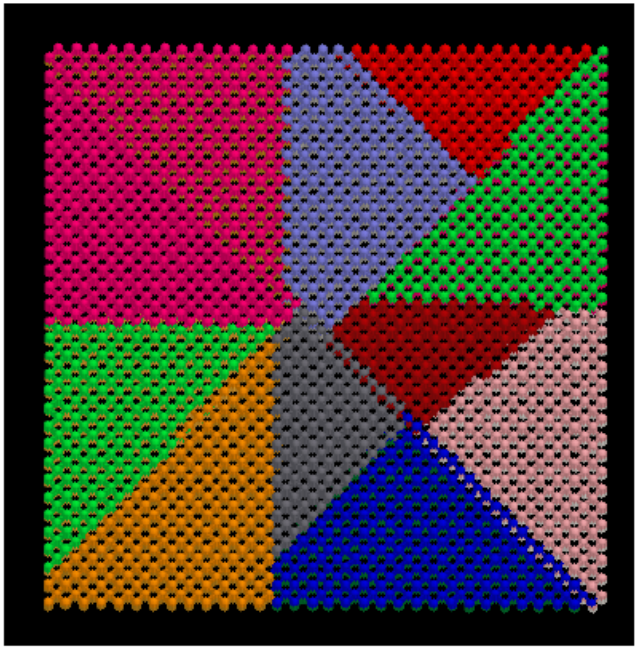
The method guarantees

- Locality of atomic partitioning
- Balanced computational cost
- Applicability to any systems
- Small computational cost

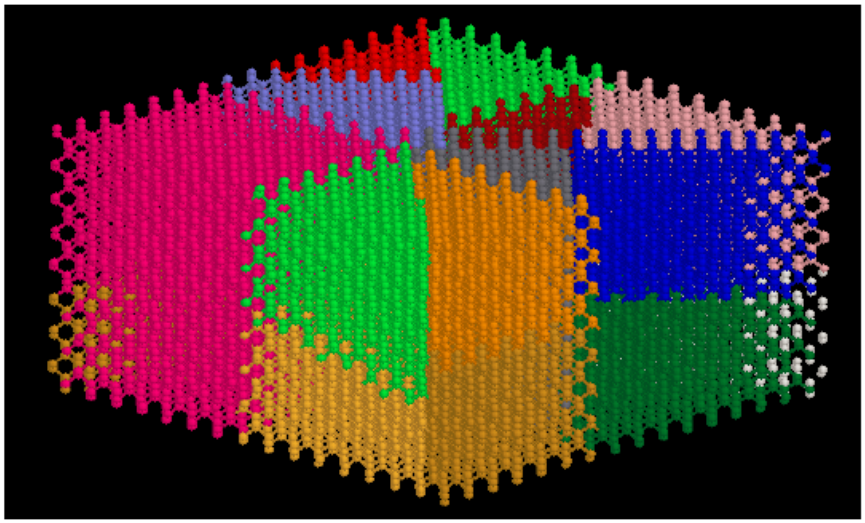


T.V.T. Duy and T. Ozaki, CPC 185, 777 (2014).

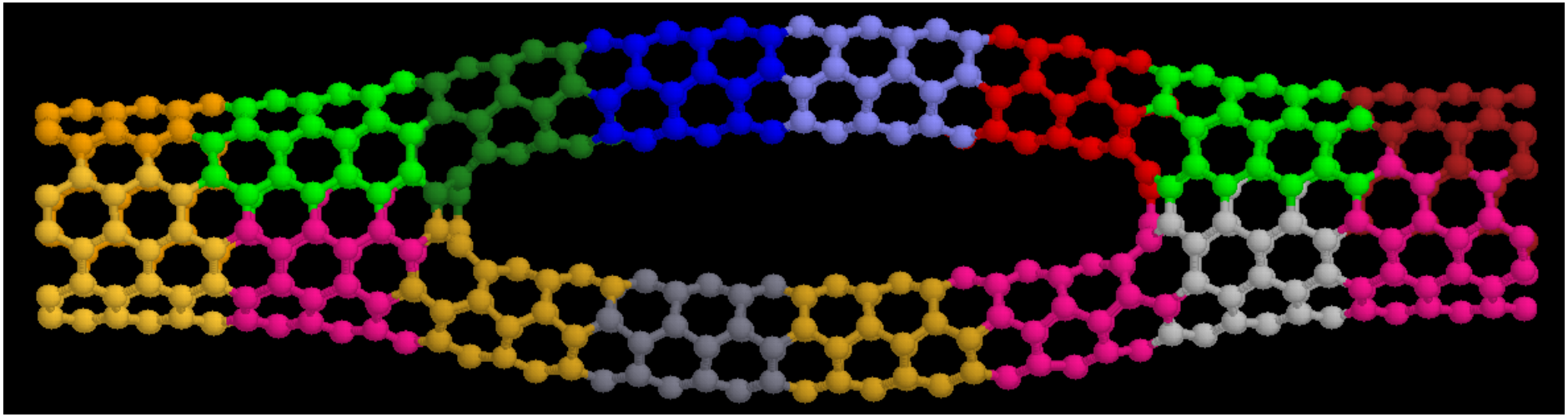
Allocation of atoms to processess



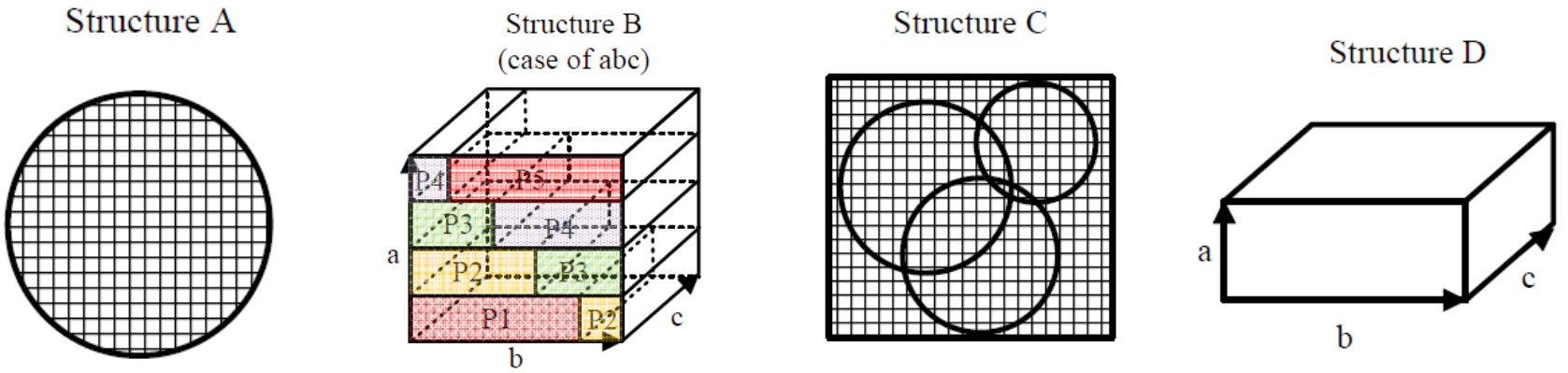
Diamond 16384 atoms, 19 processes

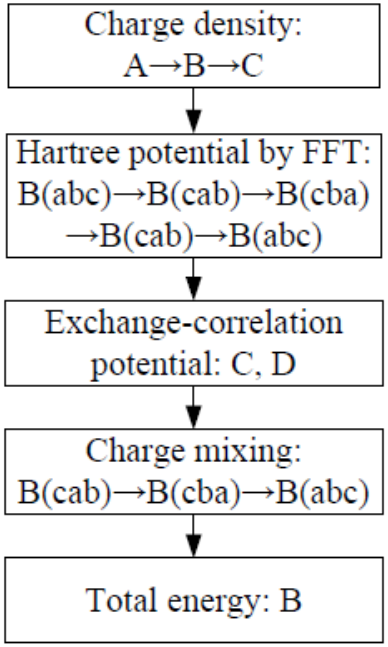


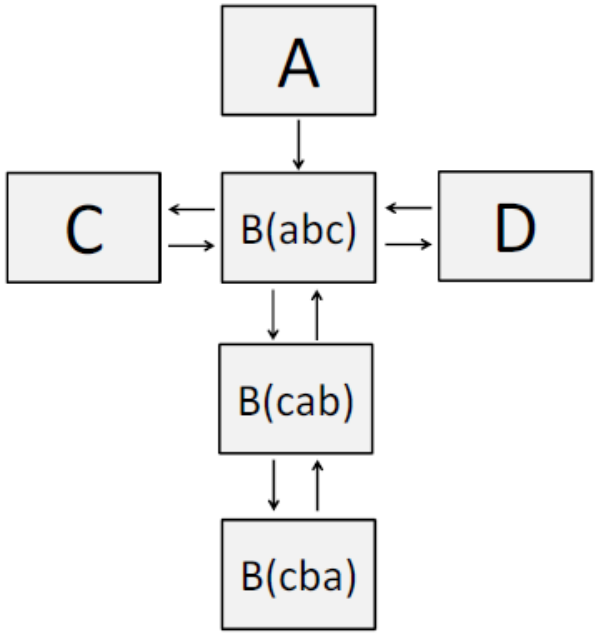
Multiply connected CNT, 16 processes



Partitioning of grids







Uniform grid is used to calculate matrix elements and solve Poisson's equation. A hundred million grid points for a few dozen thousand atoms.

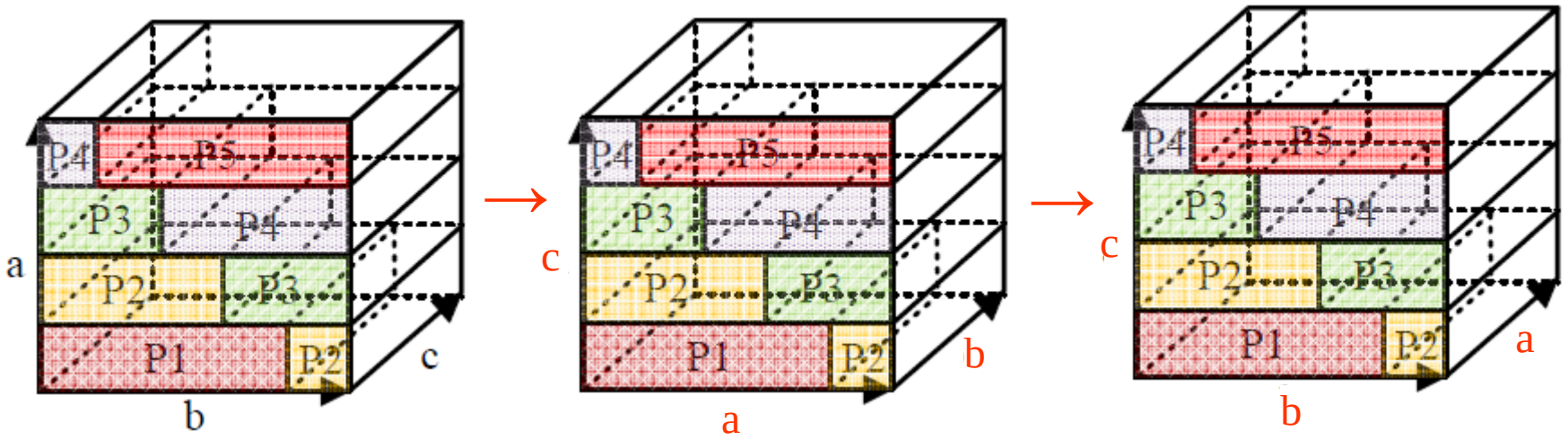
A proper one of four data structures for grid is used for each calculation.

They are designed to minimize the MPI communication.

These data structure are all constructed in Construct_MPI_Data_Structure_Grid() and Set_Inf_SndRcv() of truncation.c.

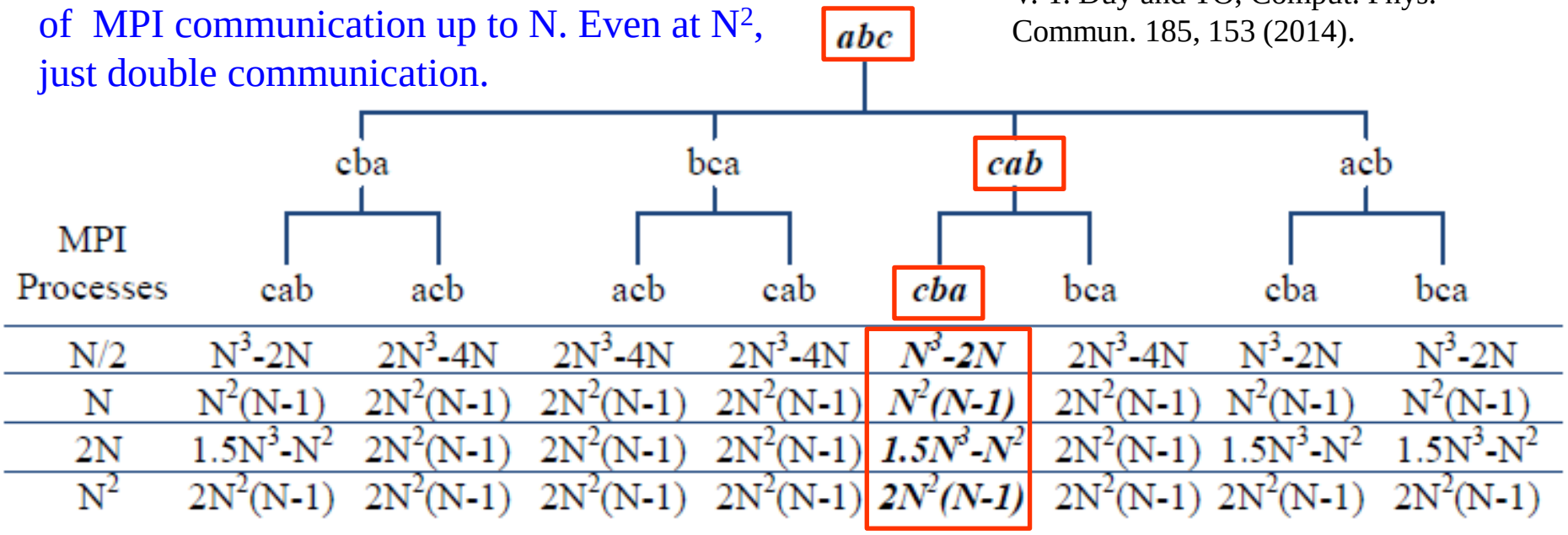
T.V.T. Duy and T. Ozaki, CPC 185, 777 (2014).

2D-parallelization of 3D-FFT with the smallest communication



Compared to 1D-parallelization, no increase of MPI communication up to N. Even at N^2, just double communication.

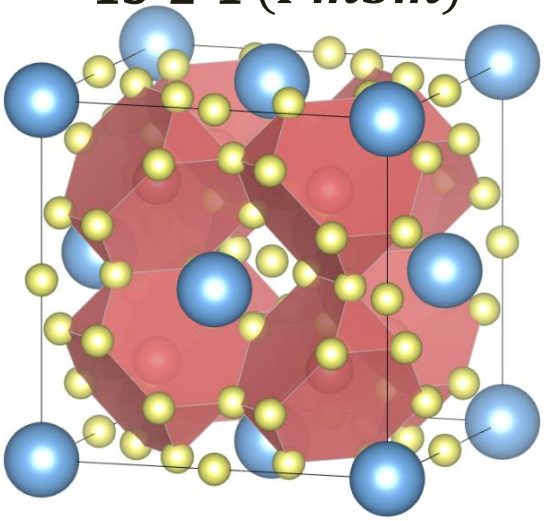
V. T. Duy and TO, Comput. Phys. Commun. 185, 153 (2014).



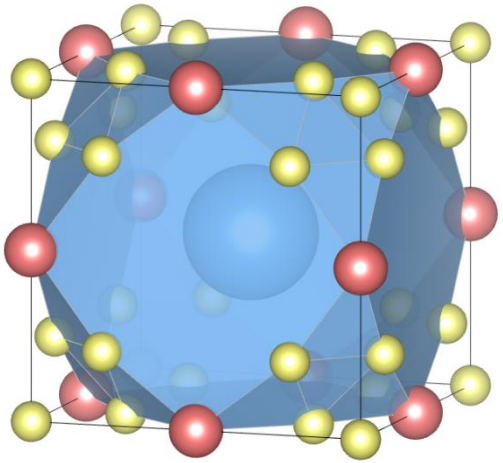
Prediction of quaternary hydrides

Based on the two densest packing structures, we performed exhaustive exploration of hydrides under 10 GPa by the NPT-MD developed by Dr. Iitaka.

13-2-1 ($Fm\bar{3}m$)



$13-3-1$ ($Pm\bar{3}m$)



Among 73,304 candidate hydrides, we identified 23 hydrides with static and dynamic stability, and two of them exhibits SC at about 6 K.

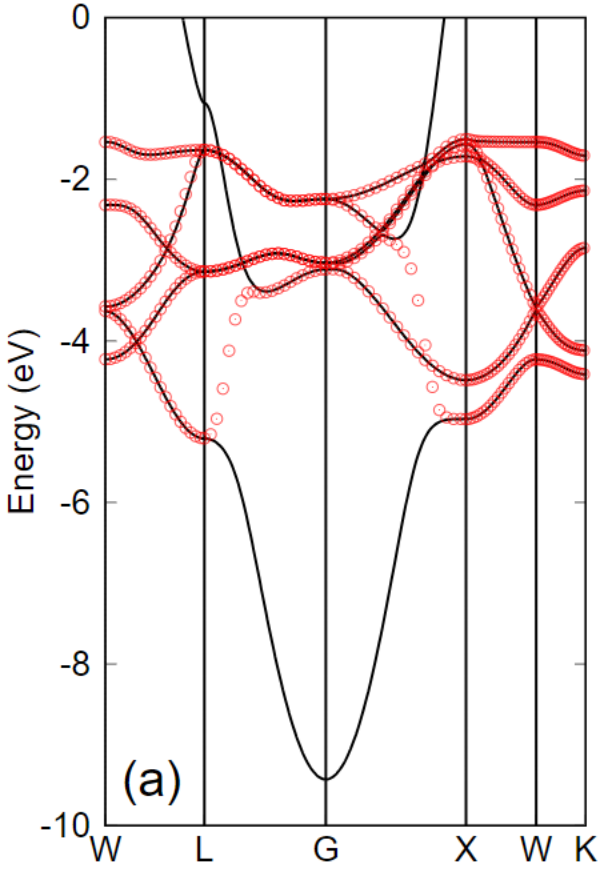
R. Koshoji and TO et al.,
Phys. Rev. Mat. 6, 114802 (2022).

Sphere size	Elements
Small	H
Semismall	E, H, Li, Be, Na, Mg, Al, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn
Medium	From Li to Hg (except for B to Ne, Si to Ar, Br, Kr, Xe, Ce to Lu, Pt)
Large	From K to Hg (except for Br, Kr, Xe, Ce to Lu, Pt)

Closest Wannier functions

FCC copper

Wannier interpolated bands
Solid: conventional
Red circle: Wannier interpolated



By minimizing the distance measure function, one can obtain the closest Wannier functions to a given set of localized orbitals such as atomic orbitals.

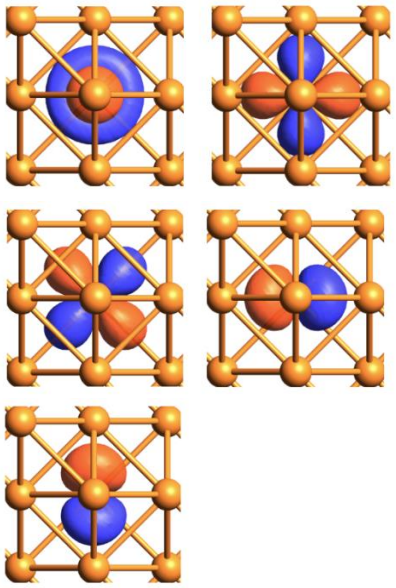
$$F[B] = \sum_p \langle R\mathbf{0}_p | R\mathbf{0}_p \rangle,$$

$$= \frac{1}{N_{\mathrm{BC}}} \sum_{\mathbf{k}} X[B, \mathbf{k}]$$

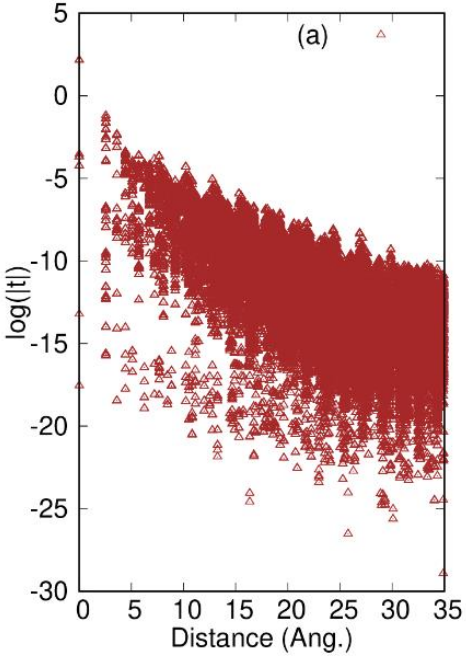
arXiv:2306.15296

https://www.openmx-square.org/cwf/

CWFs



Tight-binding hopping integrals as a function of distance



Future developments

To make OpenMX a further versatile and useful tool for materials science researches, we have been discussing the following developments.

Efficiency

- On-the-fly machine learning potentials
- Contracted and selective diagonalizations
- GPGPU parallelization

Accuracy

- PPs and basis functions for lanthanide and core level spectroscopies
- metaGGA, Hybrid functional, GW, BSE, vdW-DFT

Functionality

- Electronic excitation spectra
- Berry phase related properties
- Analysis of magnetic systems calculate exchange coupling and Dzyaloshinskii-Moriya interaction
- Phonon related properties

Community

- Database of basic computational results
- Data repository service
- Publication of an overview paper