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/

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**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 Mulliken, 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 functions

### 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.
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

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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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First characterization of silicene on ZrB$_2$ in collaboration with experimental groups

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

Theoretical proposal of topological insulators

First-principles molecular dynamics simulations for Li ion battery

Magnetic anisotropy energy of magnets

Electronic transport of graphene nanoribbon on surface oxidized Si

Interface structures of carbide precipitate in bcc-Fe

Universality of medium range ordered structure in amorphous metal oxides

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
One-particle KS orbital

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

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.
Variational optimization of basis functions

One-particle wave functions

\[ \psi_{\mu}(r) = \sum_{i\alpha} c_{\mu,i\alpha} \phi_{i\alpha}(r - r_i) \]

Contracted orbitals

\[ \phi_{i\alpha}(r) = \sum_{q} a_{i\alpha q} \chi_{i\eta}(r) \]

The variation of \( E \) with respect to \( c \) with fixed \( a \) gives

\[ \frac{\partial E_{\text{tot}}}{\partial c_{\mu,i\alpha}} = 0 \quad \rightarrow \quad \sum_{j\beta} \langle \phi_{i\alpha} | \hat{H} | \phi_{j\beta} \rangle c_{\mu,j\beta} = \varepsilon_{\mu} \sum_{j\beta} \langle \phi_{i\alpha} | \phi_{j\beta} \rangle c_{\mu,j\beta} \]

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

\[ \frac{\partial E_{\text{tot}}}{\partial a_{i\alpha q}} = \frac{\delta E_{\text{tot}}}{\delta \rho(r)} \frac{\delta \rho(r)}{\delta a_{i\alpha q}} \]

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

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

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

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

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

\[ B_{ij} = \langle \phi_i | \chi_j \rangle \]

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

If \( Q=0 \), then \( B-B^*=0 \)

This is the norm-conserving PP used in OpenMX.
Optimization of pseudopotentials

1. Choice of valence electrons (semi-core included?)
2. Adjustment of cutoff radii by monitoring shape of pseudopotentials
3. Adjustment of the local potential
4. Generation of PCC charge

(i) Choice of parameters

(ii) Comparison of logarithm derivatives
If the logarithmic derivatives for PP agree well with those of the all electron potential, go to the step (iii), or return to the step (i).

(iii) Validation of quality of PP by performing a series of benchmark calculations.

Optimization of PP typically takes a half week per a week.
Reproducibility in density functional theory calculations of solids

Δ-gauge
A way of comparing accuracy of codes

\[ \Delta_i(a, b) = \sqrt{\frac{\int_{0.94V_{0,i}}^{1.06V_{0,i}} (E_{b,i}(V) - E_{a,i}(V))^2 dV}{0.12V_{0,i}}} \]

Energy of element \(i\) (meV/atom)

Volume (Å³/atom)
Comparison of codes by $\Delta$-gauge

<table>
<thead>
<tr>
<th>NCPP</th>
<th>USPP</th>
<th>PAW</th>
<th>AE</th>
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<tbody>
<tr>
<td>MBK2013/OpenMX</td>
<td>VASP</td>
<td>VASP</td>
<td>WEN2k,acc</td>
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<td>63</td>
<td>05</td>
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The mean $\Delta$-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

Recursive atomic partitioning

The method guarantees

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

Allocation of atoms to processes

Diamond 16384 atoms, 19 processes

Multiply connected CNT, 16 processes
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.
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.

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**.

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


<table>
<thead>
<tr>
<th>Sphere size</th>
<th>Elements</th>
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<tbody>
<tr>
<td>Small Neutrons</td>
<td><strong>H</strong> E, H, Li, Be, Na, Mg, Al, K, Ca, Sc, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn</td>
</tr>
<tr>
<td>Semismall</td>
<td>From Li to Hg (except for B to Ne, Si to Ar, Br, Kr, Xe, Ce to Lu, Pt)</td>
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<tr>
<td>Medium</td>
<td>From K to Hg (except for Br, Kr, Xe, Ce to Lu, Pt)</td>
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<tr>
<td>Large</td>
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By minimizing the distance measure function, one can obtain the closest Wannier functions to a given set of localized orbitals such as atomic orbitals.

\[
F[B] = \sum_p \langle R_{0p} | R_{0p} \rangle,
\]
\[
= \frac{1}{N_{BC}} \sum_k X[B, k]
\]

Tight-binding hopping integrals as a function of distance

arXiv:2306.15296
https://www.openmx-square.org/cwf/
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