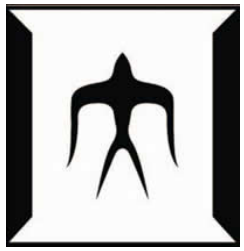
iDFT @ Riken 23. Jun. 2017

Computational materials database toward discovering novel semiconductors

Yu Kumagai

MCES, Tokyo Institute of Technology

PREST, JST



Why computational materials database?

① Screening of materials at an early stage

entry #1
- electronic structure
- point defects
- dielectric const.
- effective mass
- optical absorption
- structure
- energy
- band gap
- IP

Continue?

change?

abort?

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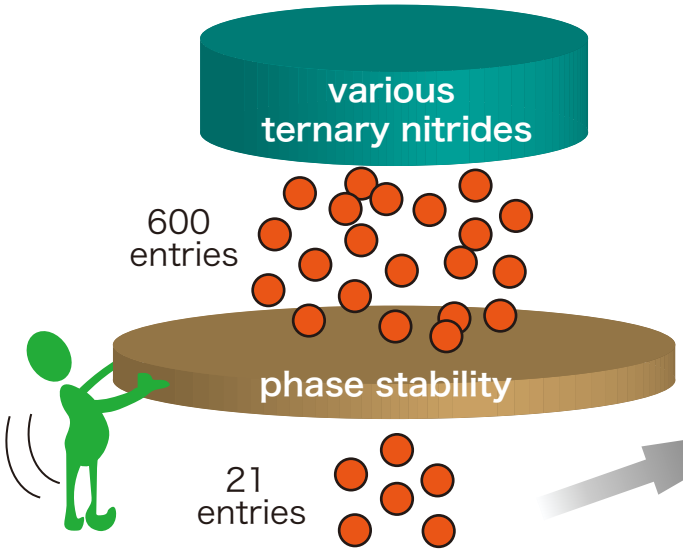
② Rediscovery of reported materials



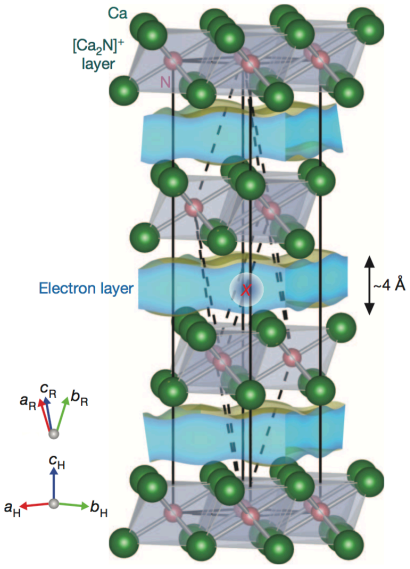
Promising semiconductors

- 11 new compounds
  We synthesized
  a new semiconductor $CaZn_2N_2$
- 2 theoretically reported
- 8 experimentally reported

Out of 4 had not been
considered as semiconductors

Extract full potential
of reported materials

Ca_2N:2-D electride



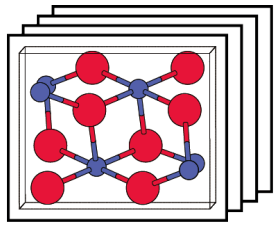
Lee *et al.*, Nature (2013)

Hinuma, Hatakeyama, YK *et al.*, Nat. Comm. (2016)

Why computational materials database?

③ Dataset for machine learning

Massive and homogeneous dataset



strcutre database (ICSD)

DFT calc.

E_f, E_g, $\boldsymbol{P}$, $\boldsymbol{m}^*$, IP dataset

$\boldsymbol{x} = (x_1, x_2, x_3, \ldots)$ feature quantity

machine learning

modeling

$E_f(\boldsymbol{x})$, $E_g(\boldsymbol{x})$, $\boldsymbol{P}(\boldsymbol{x})$, $\boldsymbol{m}^*(\boldsymbol{x})$, IP($\boldsymbol{x}$)

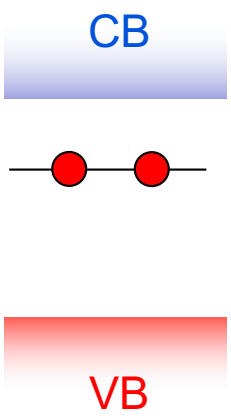
④ Construction of theories

Ex：Oxygen vacancy

Deep level

- ZnO
- SnO_2
- In_2O_3
- CdO
- HfO_2
- ……

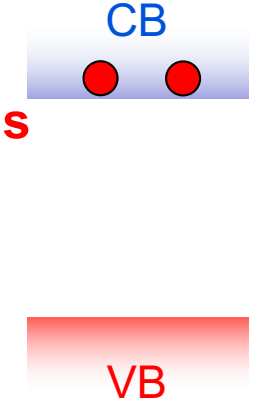
universal



Shallow level

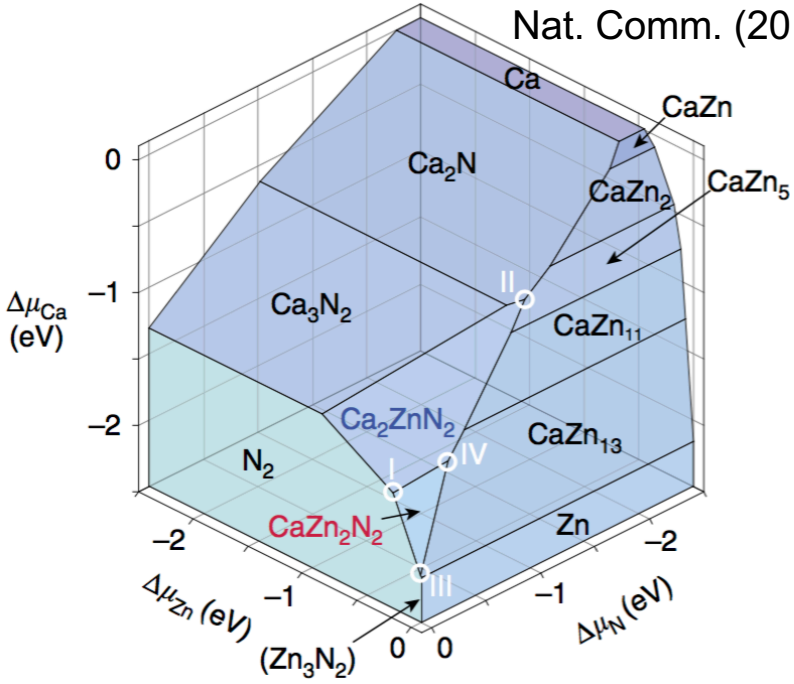
- $SrTiO_3$

anomalous



⑤ Energies for discovering new materials

Nat. Comm. (2016)

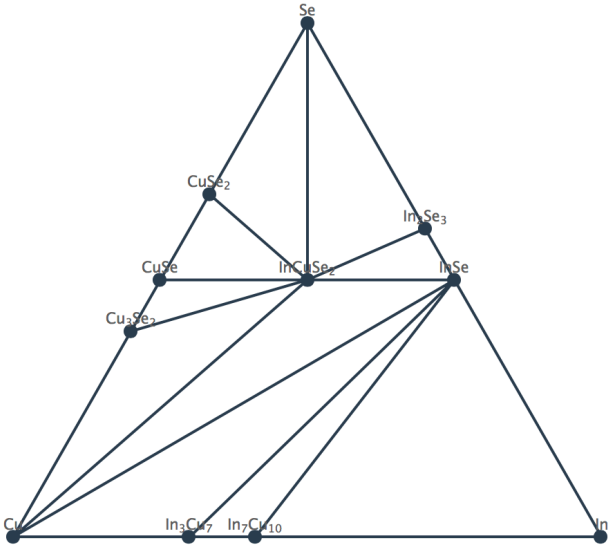


Available databases

MATERIALS PROJECT

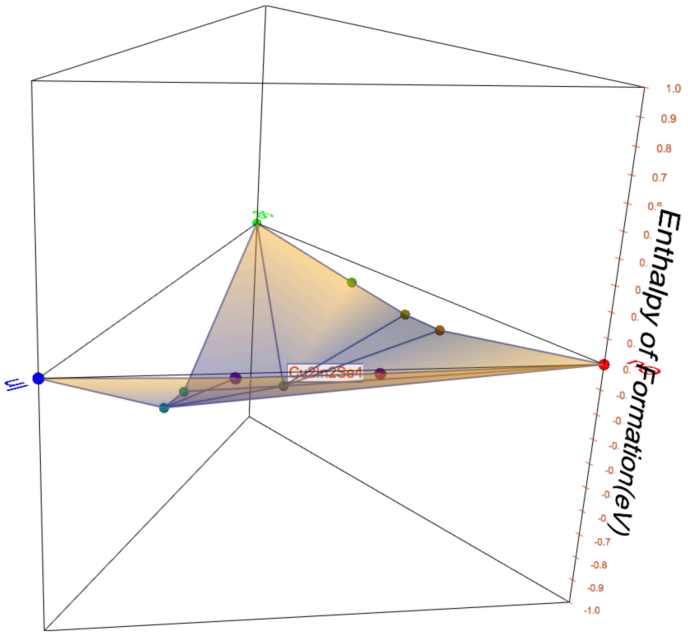
69,623	54,058
INORGANIC COMPOUNDS	BANDSTRUCTURES

The Materials Project: A materials genome approach to accelerating materials innovation, Jain, Ceder *et al.*, APL Mat. 1, 011002 (2013).



AFLOW
Automatic - FLOW for Materials Discovery

Aflow is a globally available database of **1,668,252** material compounc



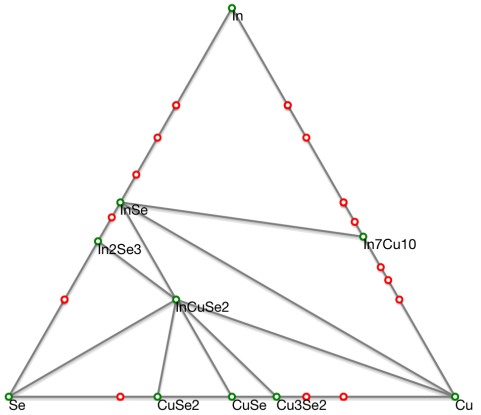
AFLOW: An automatic framework for high-throughput materials discovery,
Curtarolo *et al.*, Comp. Mat. Sci., 58, 218 (2012).

Available databases

OQMD: The database now contains **471857** entries.

The Open Quantum Materials Database

Materials Design and Discovery with High-Throughput Density Functional Theory: The Open Quantum Materials Database (OQMD), Saal, Wolverton, *et al.*, JOM 65, 1501 (2013).



NOMAD

Computational data repository

High Performance Computing Center
Materials Database

CMDB from NREL group

Problems of existing databases for researching semiconductors

① Lack of physical properties (e.g., ionization potential, point-defect properties)

② Lack of accuracy of some properties (e.g., crystal structures, band gaps)

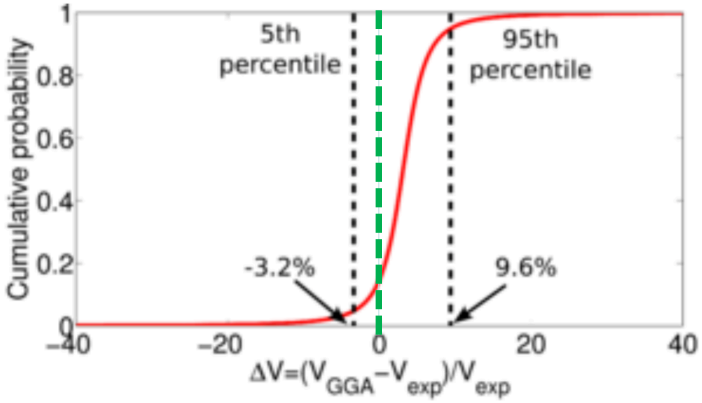
We need a new computational materials database toward investigating/discovering novel semiconductors

Crystal structures in available databases

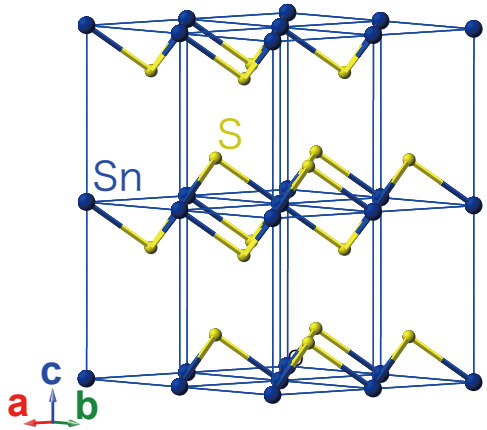
ex. 1) Materials Project

Cumulative distribution of the volume

Sun *et al.*,
Sci. Adv. (2016)



Mean error: 3.2 %
(# of structures : 29,902)
by PBE functional

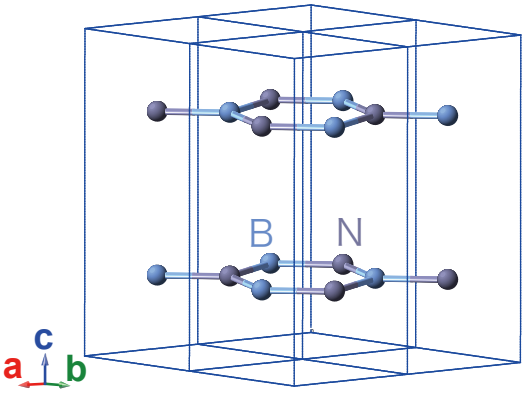


ex. 2) Layered materials: c-axis in SnS_2

exp.	Materials Project	aflow	OQMD	NoMad	NREL MatDB
5.90 Å	6.98	6.67	6.23	NA	6.3

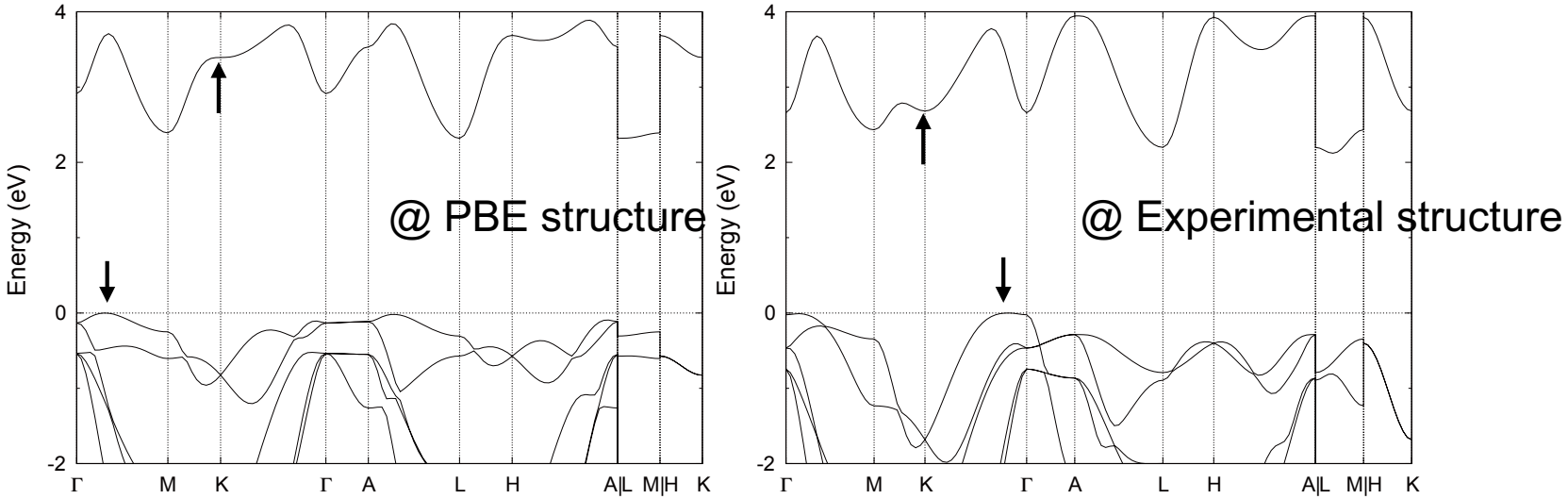
ex. 3)) Layered materials: c-axis in *h*-BN

exp.	Materials Project	aflow	OQMD	NoMad	NREL MatDB
6.661 Å	7.71	7.88	6.43 (LDA)	NA	7.6

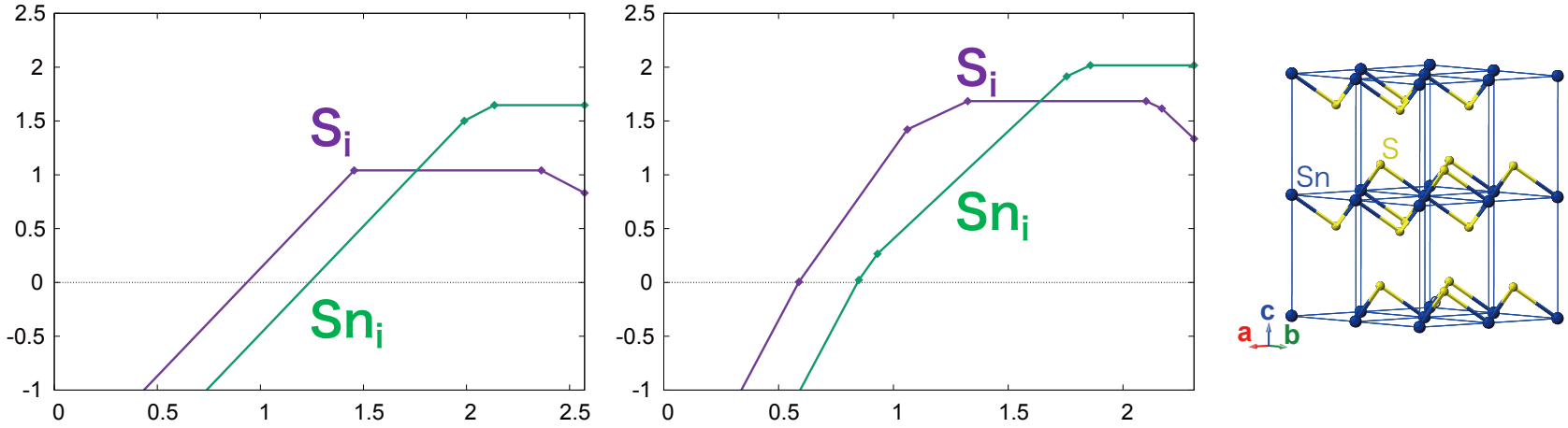


Importance of reproducibility of structures

ex. 1) Electronic structure of SnS_2

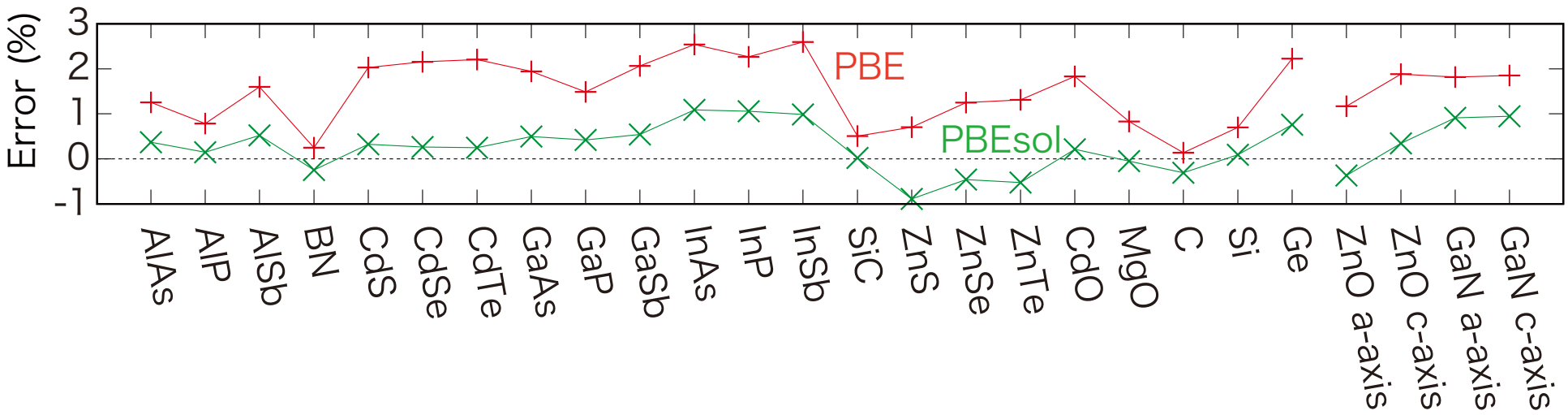


ex. 2) Formation energy of interstitials in SnS_2



Structures are of crucially importance for semiconductor researches

PBEsol+U



Errors of interlayer distances in layered oxides

	PBE+U	PBEsol+U
ME	5.9%	0.4%
RMSE	5.5%	3.0%

Hinuma, Hayashi, YK, Tanaka, Oba., submitted.

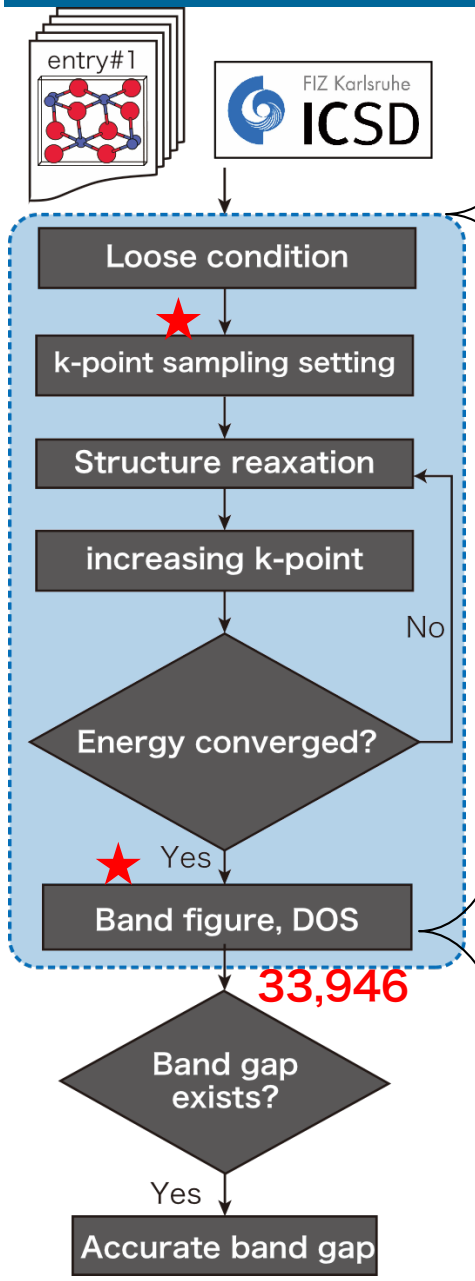
c-axis in SnS_2

exp.	Materials Project	aflow	my DB
5.90 Å	6.98	6.67	5.91

c-axis in *h*-BN

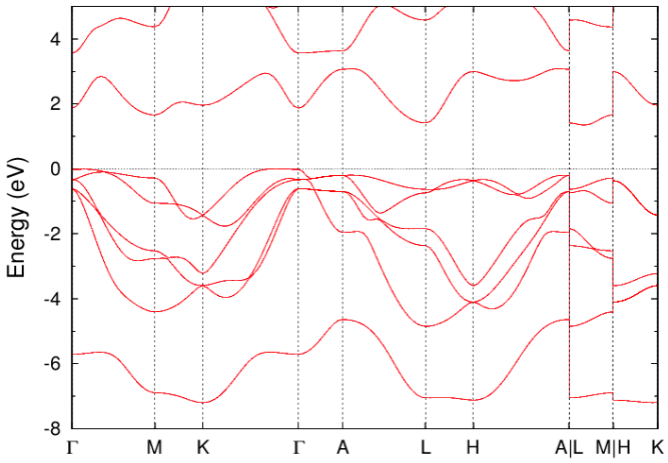
exp.	Materials Project	aflow	my DB
6.661 Å	7.71	7.88	6.69

Flowchart

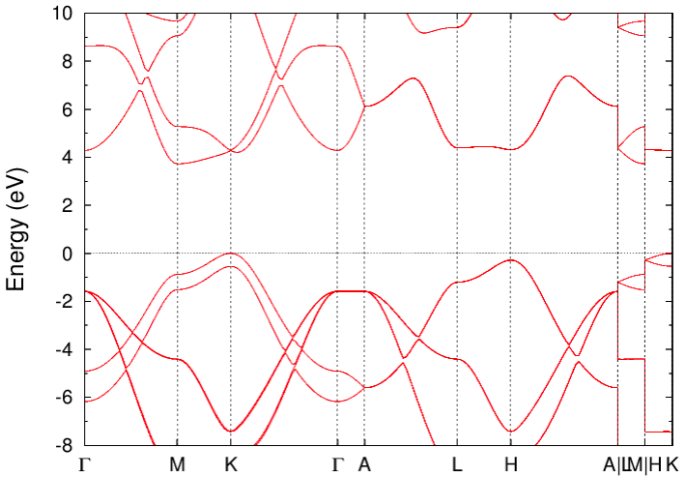


1. Remove entries with partial occupancies
2. Search for symmetries and transform the unit cell based on crystallography★
3. Remove redundant structures ★
4. Remove entries with lanthanide and/or actinide elements
5. Remove entries containing more than 100 atoms
   →# of remaining entries: 46,969
6. Classify entries by space groups ★

SnS_2



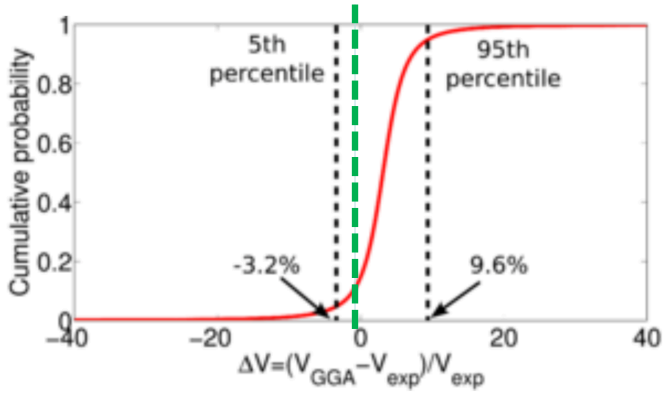
h-BN



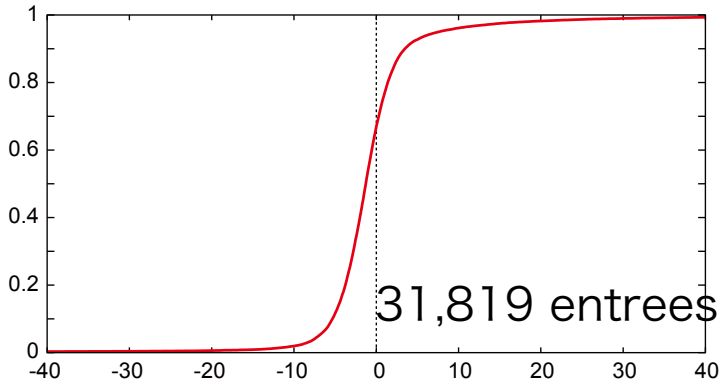
Automation of band paths: Hinuma, Pizzi, YK, Oba, Tanaka, Comput. Mater. Sci. (2017).

Analysis of the structures

Cumulative distribution of the volume



PBE-sol



ME=-0.82% compared to experiments including high-temperature phases

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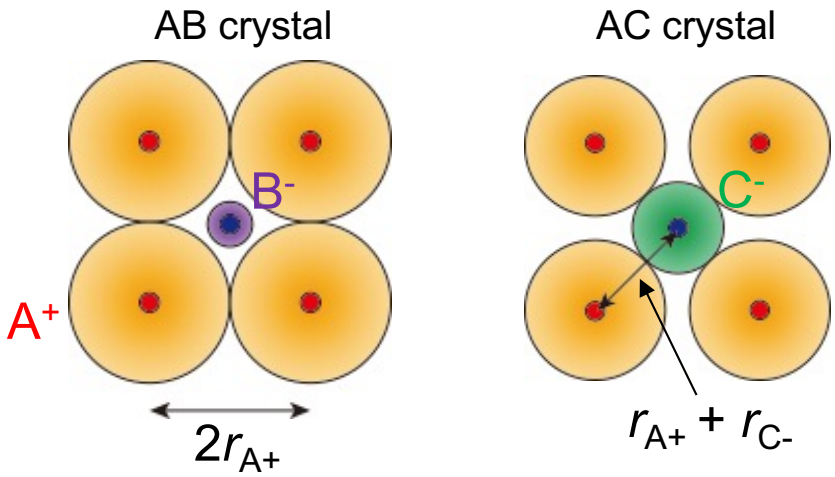
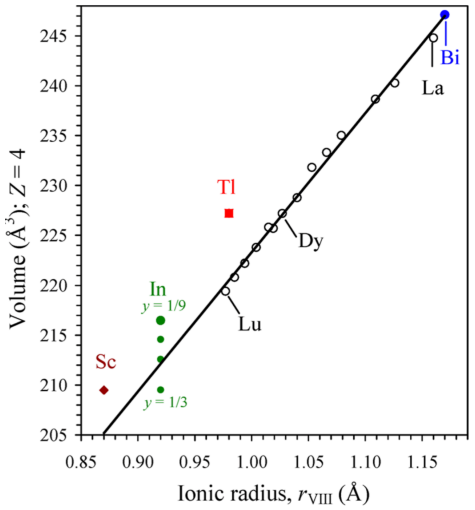
Acta Cryst. (1976). A **32**, 751

Revised Effective Ionic Radii and Systematic Studies of Interatomic Distances in Halides and Chalcogenides

BY R. D. SHANNON

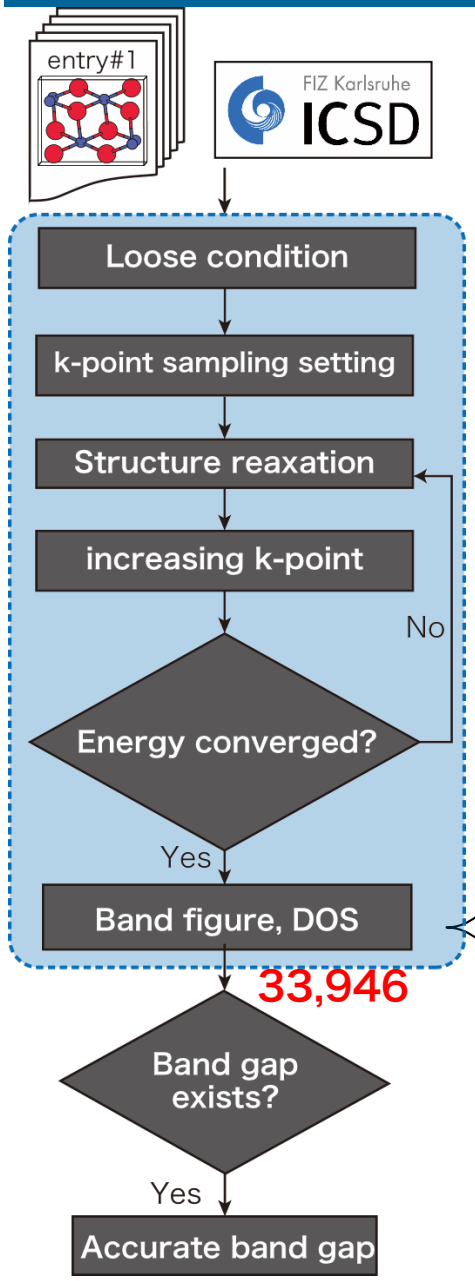


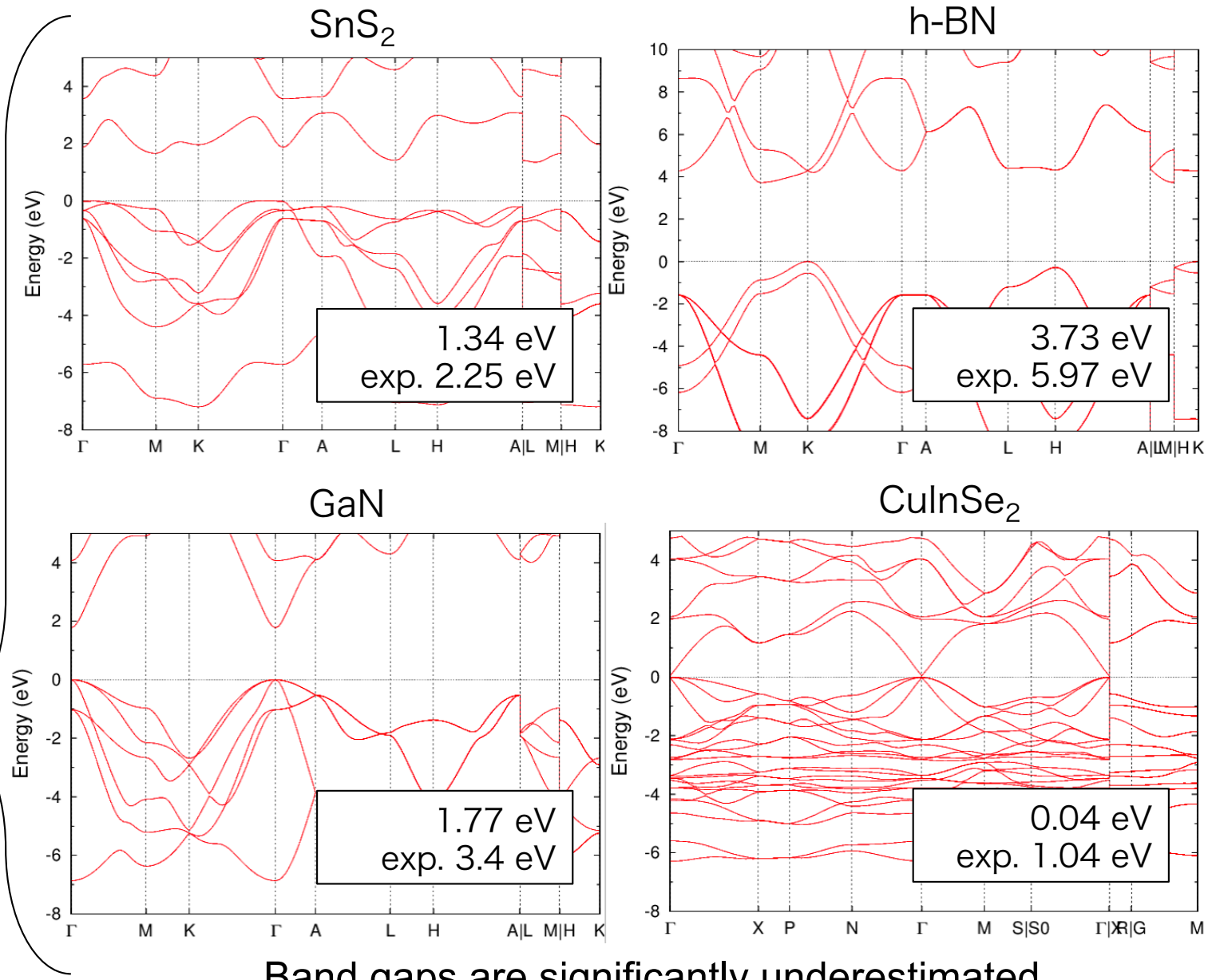


including inaccurate data

Yi, YK, *et al.*, Inorg. Chem. (2014).

Flowchart

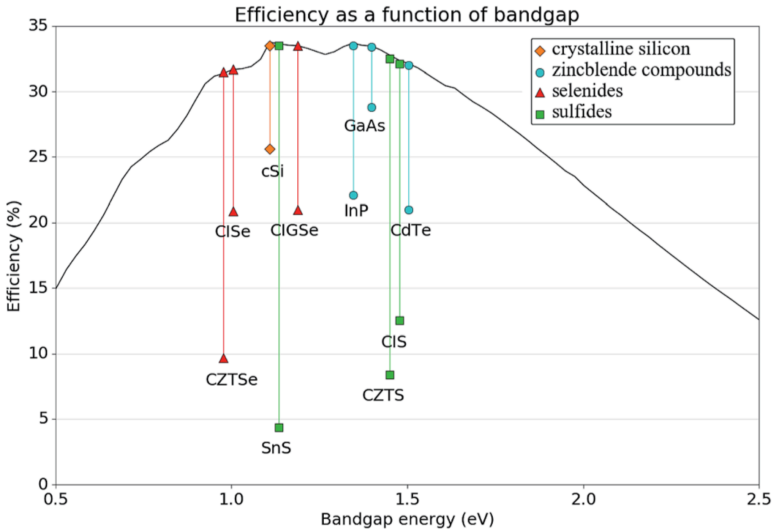




Band gaps are significantly underestimated.

Applications related to band gaps

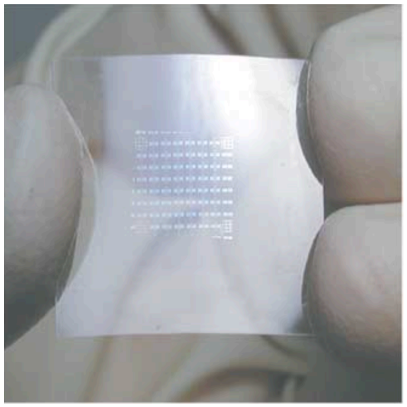
Solar cell



Lee, YK, Walsh, Oba, J. Mat. Chem. A (2017).

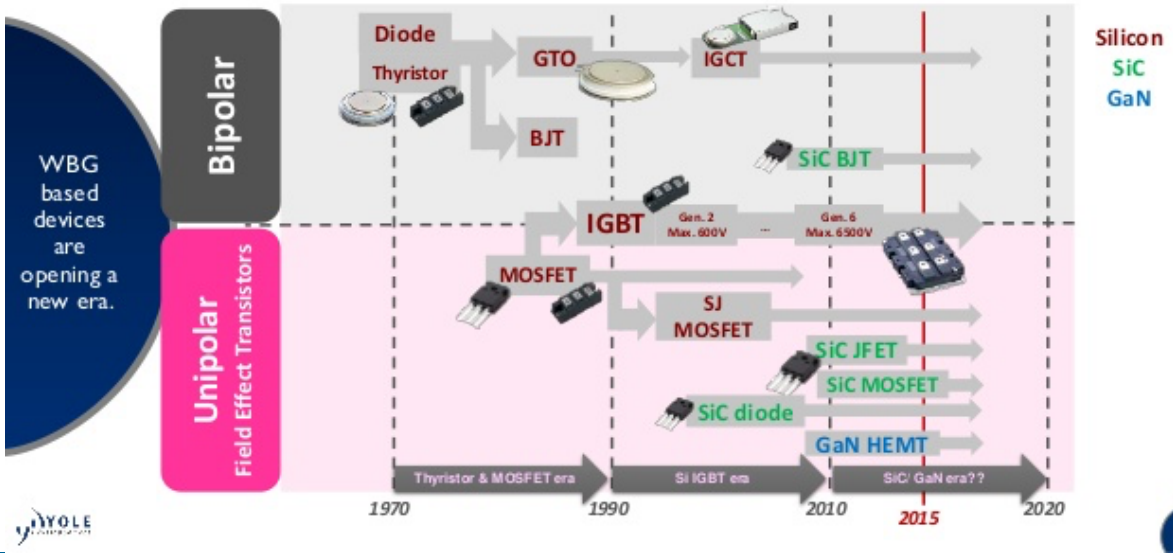
LED

Transparent conductors

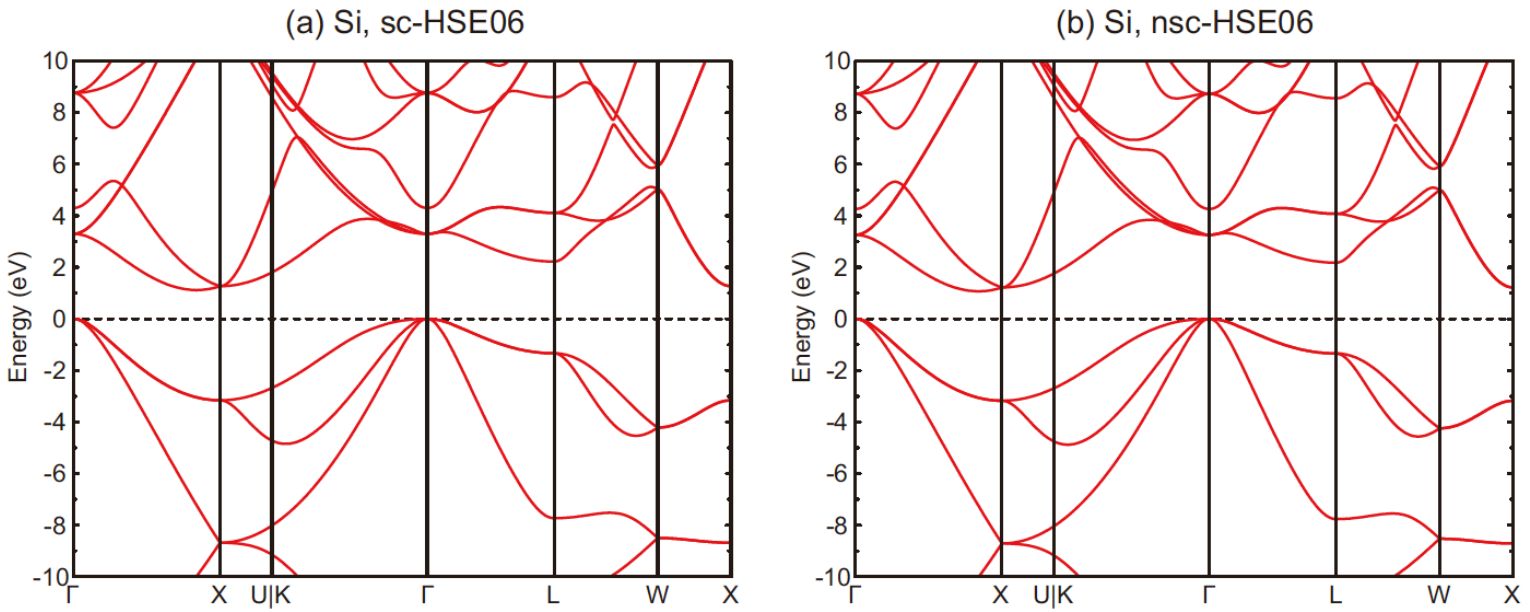


Nomura, *et al.*, Nature (2004).

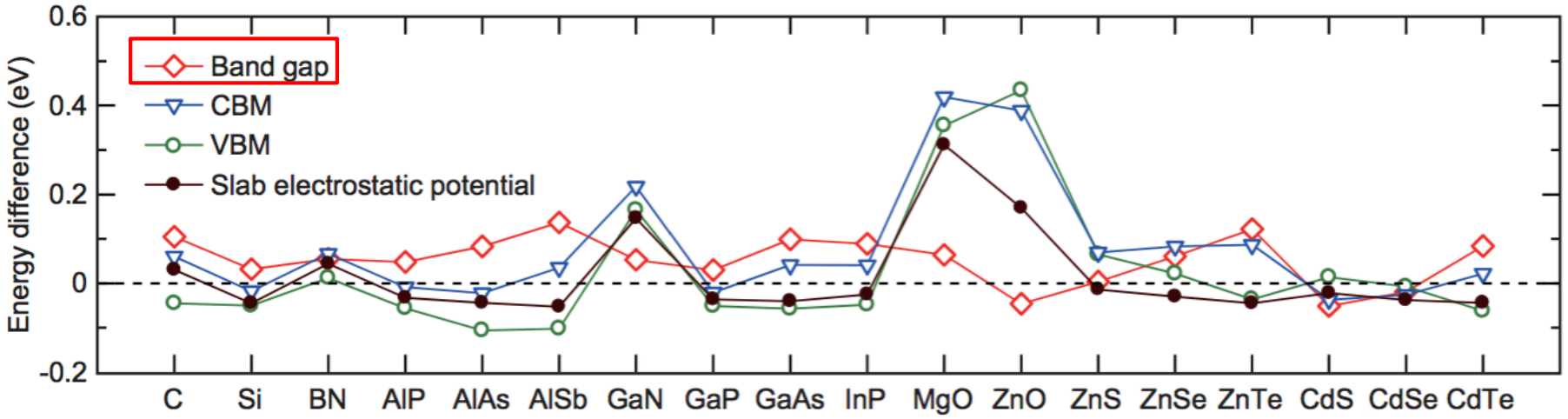
Power devices



non-selfconsistent (nsc) HSE06

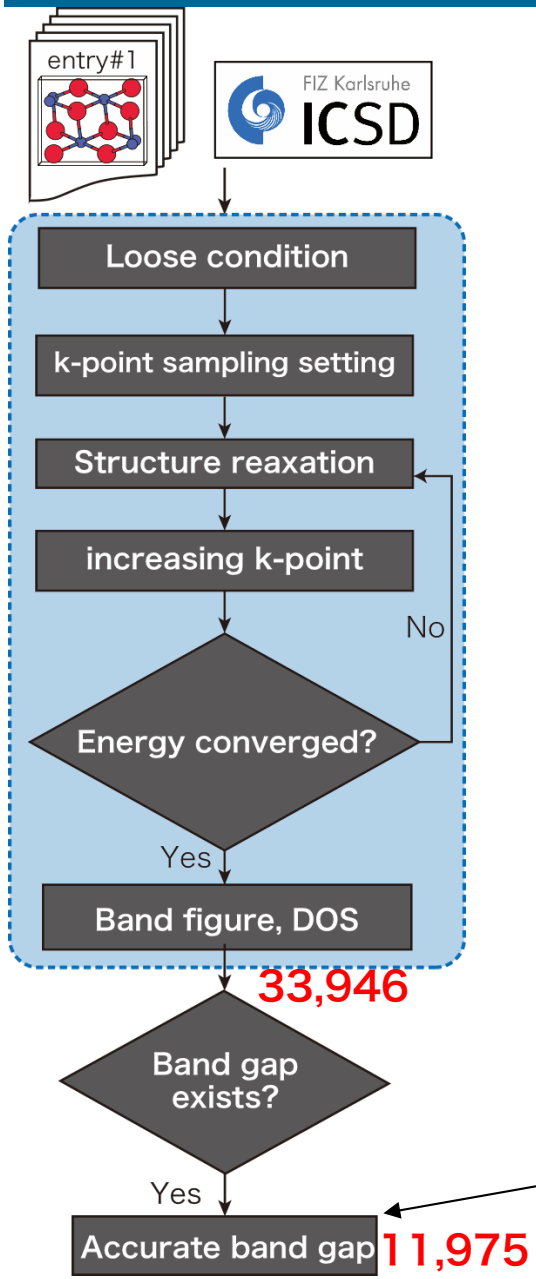


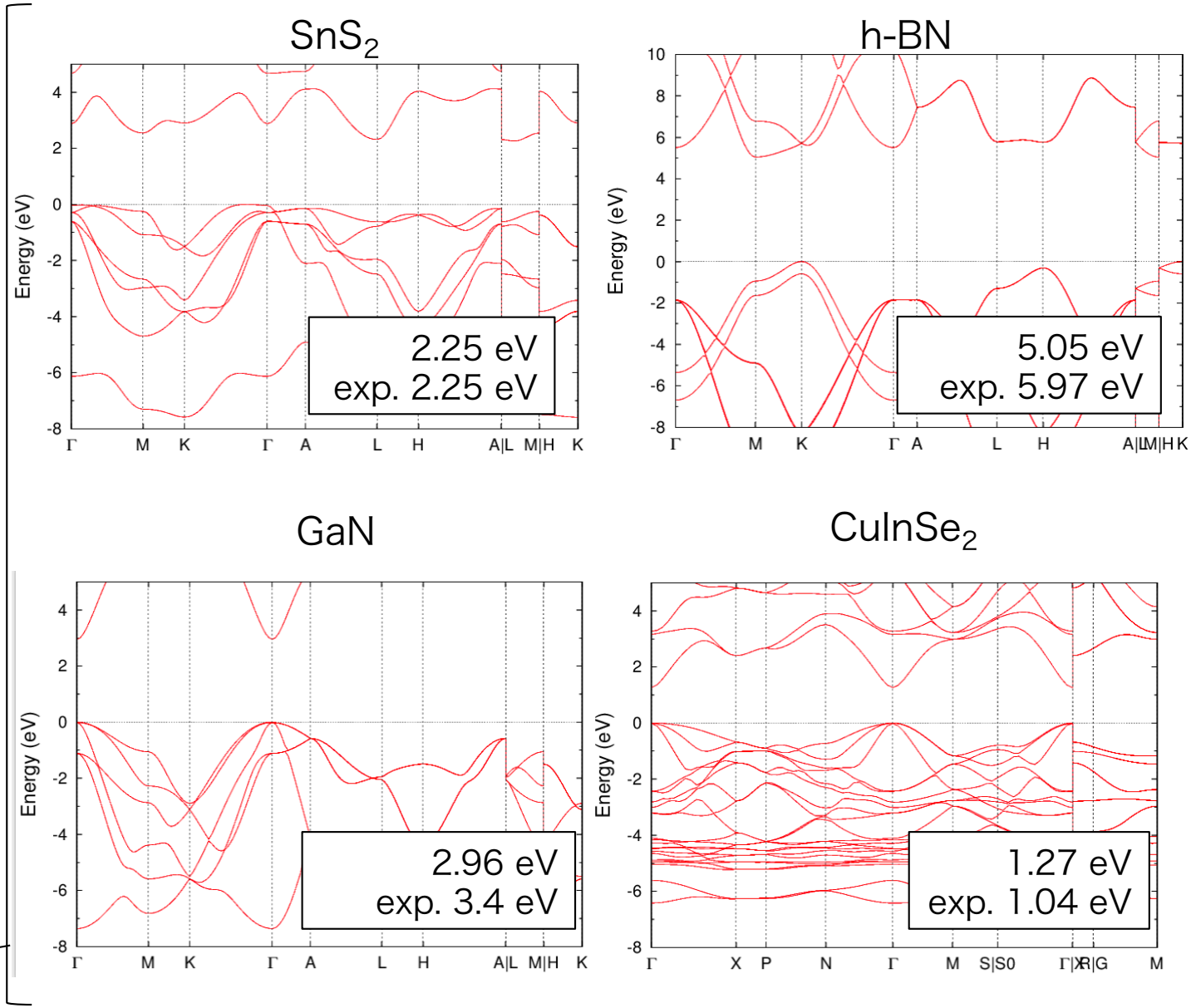
Band gap: 1.12 eV (exp. 1.17 eV)



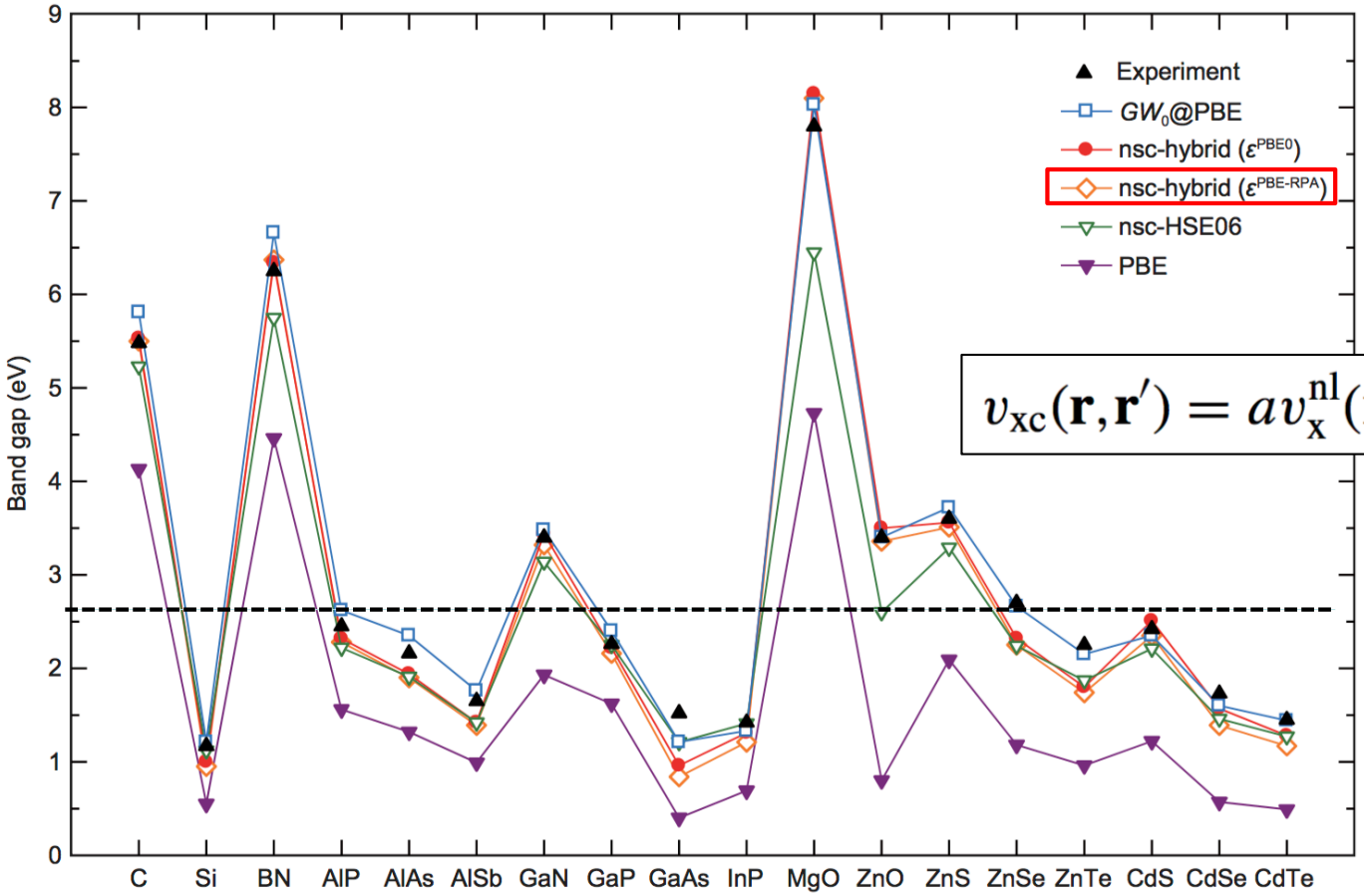
Hinuma, YK, Tanaka, and Oba, Phys. Rev. B (2017)

nsc HSE06


entry#1
FIZ Karlsruhe
ICSD
Loose condition
k-point sampling setting
Structure reaxation
increasing k-point
No
Energy converged?
Yes
Band figure, DOS
33,946
Band gap exists?
Yes
Accurate band gap
11,975


SnS2
Energy (eV)
Γ M K Γ A L H A|L M|H K
2.25 eV
exp. 2.25 eV
h-BN
Energy (eV)
Γ M K Γ A L H A|L M|H K
5.05 eV
exp. 5.97 eV
GaN
Energy (eV)
Γ M K Γ A L H A|L M|H K
2.96 eV
exp. 3.4 eV
CuInSe2
Energy (eV)
Γ X P N Γ M S|S0 Γ|Y R|G M
1.27 eV
exp. 1.04 eV

Band gaps for wide gap materials



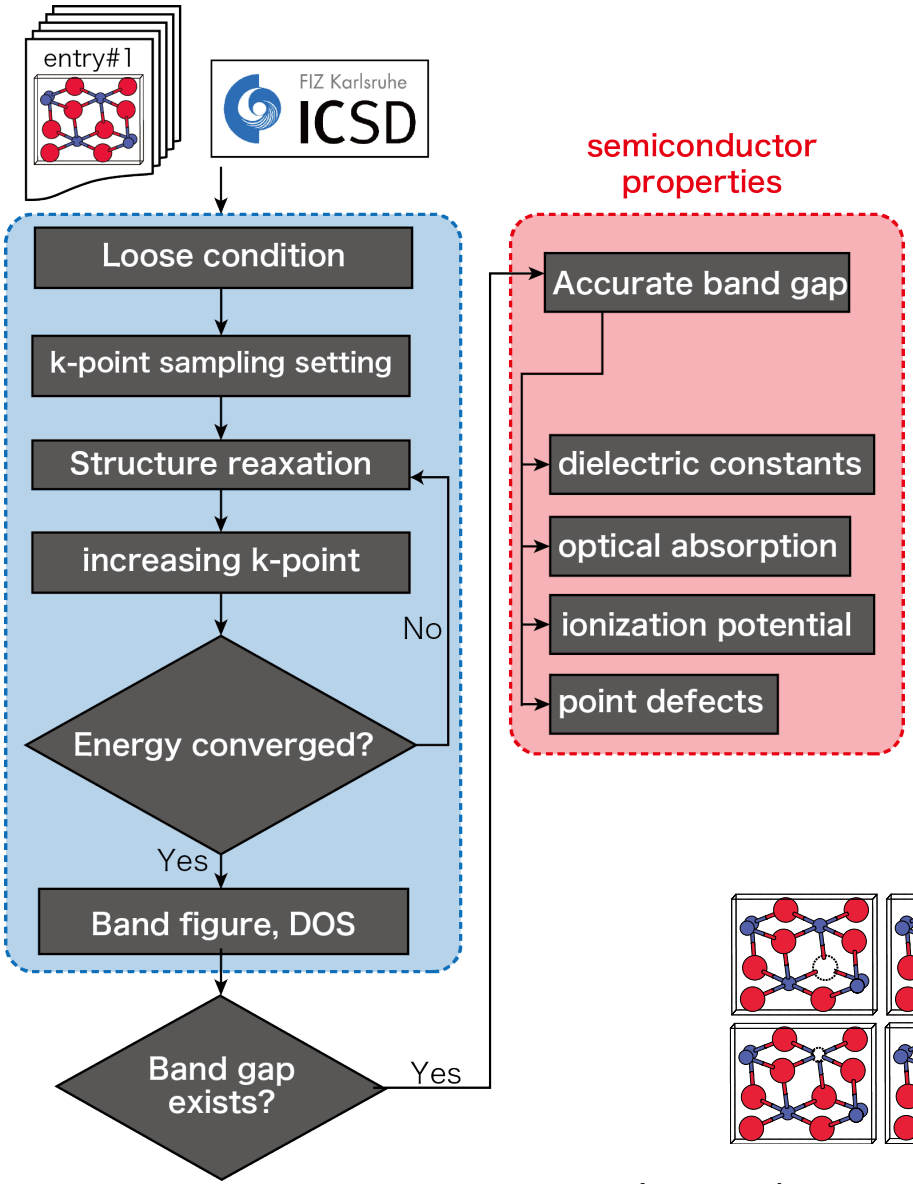
$$v_{\mathrm{xc}}(\mathbf{r},\mathbf{r}') = a v_{\mathrm{x}}^{\mathrm{nl}}(\mathbf{r},\mathbf{r}') + (1-a) v_{\mathrm{x}}^{\mathrm{l}}(\mathbf{r}) + v_{\mathrm{c}}(\mathbf{r})$$

$$a = \varepsilon_{\infty}^{-1}$$

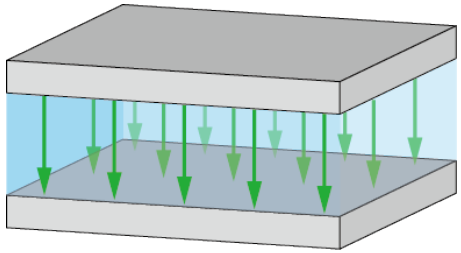
Hinuma, YK, Tanaka, and Oba, Phys. Rev. B (2017)

	PBE	nsc-HSE06	nsc-hybrid($\varepsilon_{\infty}^{\mathrm{PBE-RPA}}$)	nsc-hybrid($\varepsilon_{\infty}^{\mathrm{PBE0}}$)	GW_0@PBE	Experiment
⋮						
CdS	1.22	2.21	2.35	2.51	2.35	2.42
CdSe	0.57	1.46	1.39	1.57	1.60	1.73
CdTe	0.49	1.27	1.17	1.28	1.44	1.45
MAE	1.30	0.34	0.23	0.19	0.14	

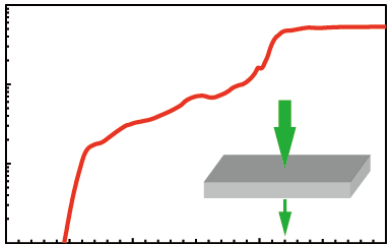
Further properties…



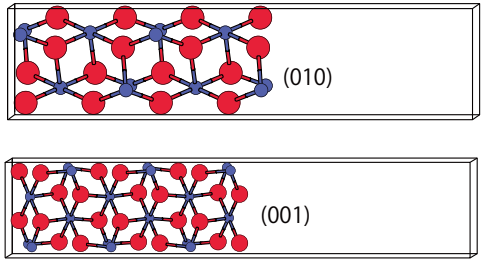
- Dielectric constant



- Optical absorption

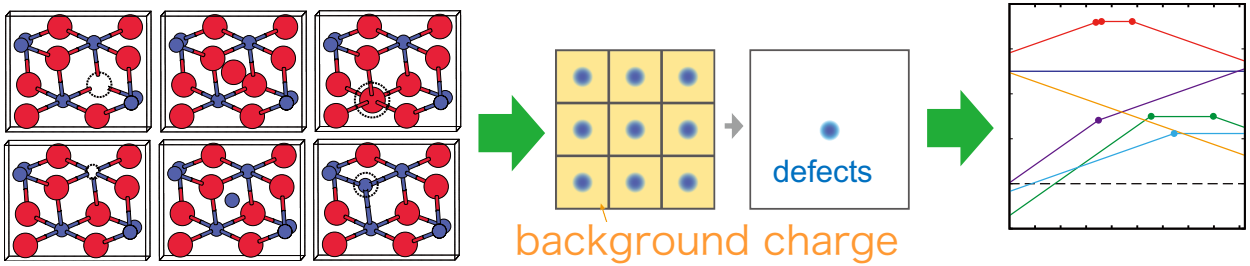


- Ionization potential



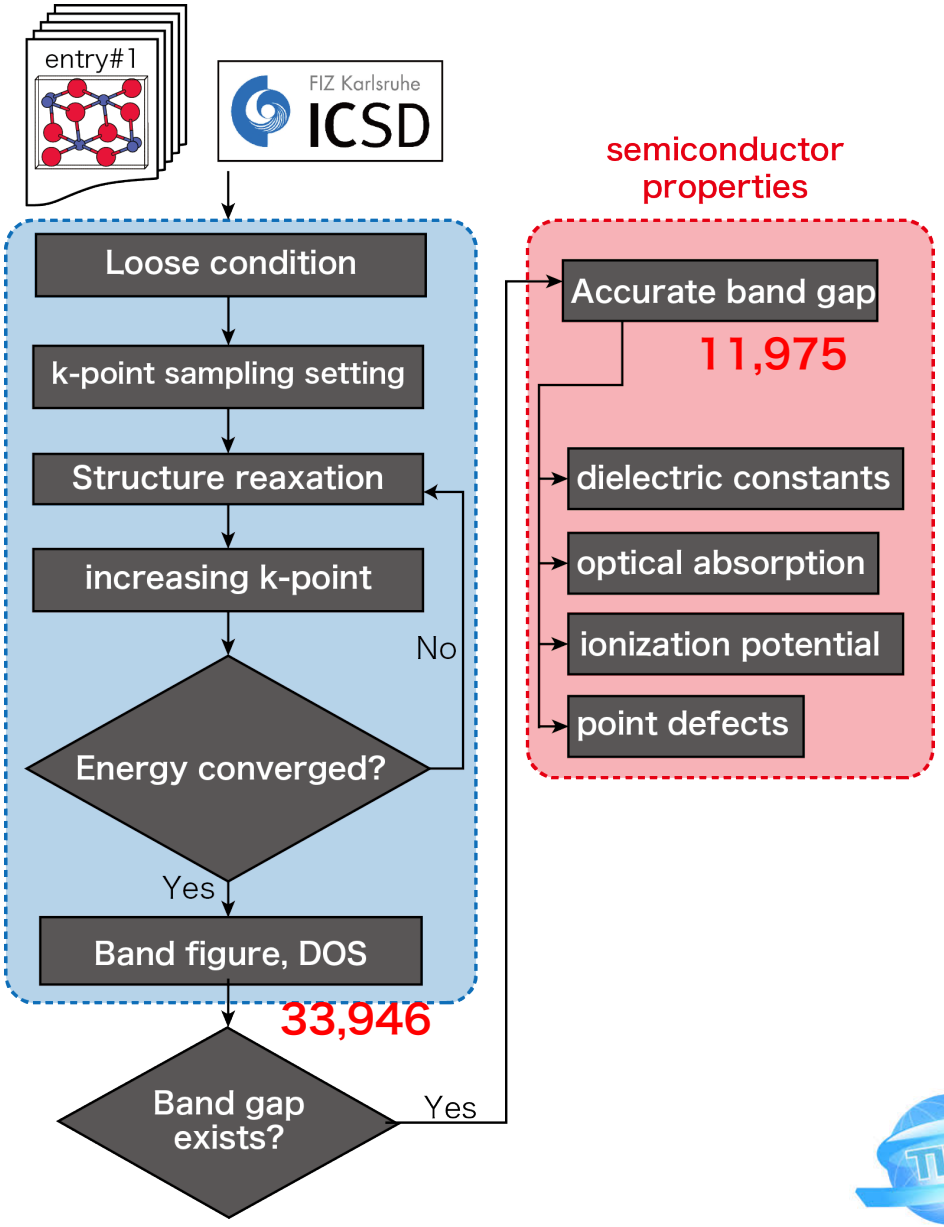
Automatic construction of surface models
Hinuma, YK, Oba, Tanaka, Comput. Mater. Sci. (2016).

- Point defects



Automatic construction of point defect calculations, YK, and Oba, in preparation

Summary

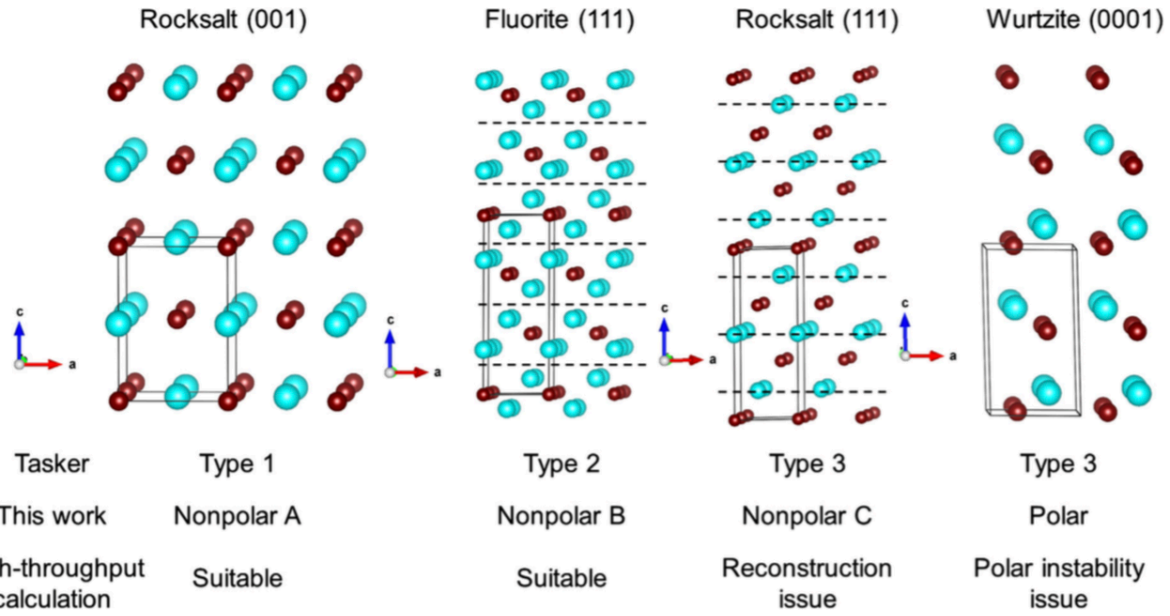


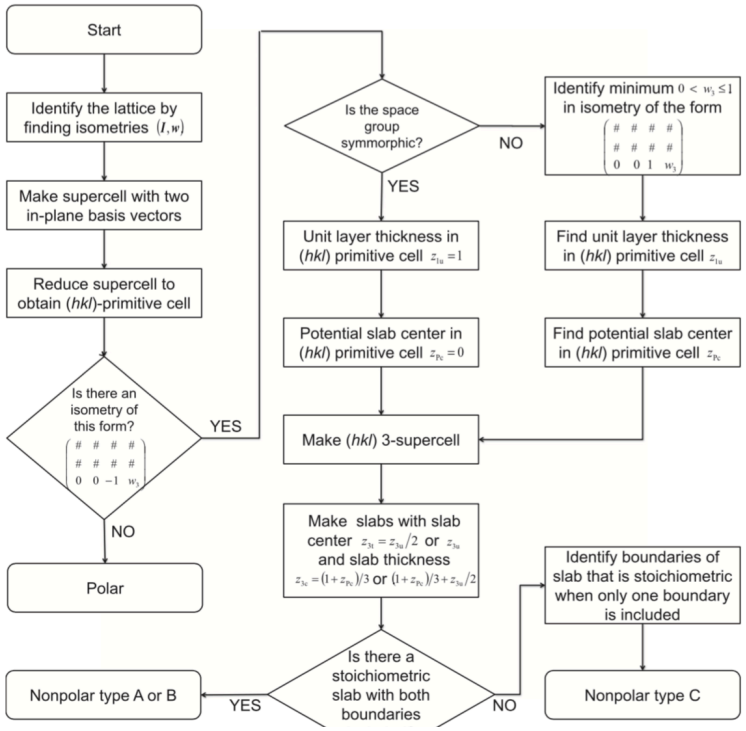
- Construction of computational materials database
- PBEsol+U functional works well for reproducing the experimental structures
- non-selfconsistent HSE06 works well for predicting the band gaps
- Further semiconductor properties will be calculated in the future

- Collaborators
  - Prof. Fumiyasu Oba
  - Prof. Yoyo Hinuma

- Financial support
  - MEXT Elements Strategy Initiative
  - Grants-in-Aid for Young Scientists A
  - PRESTO

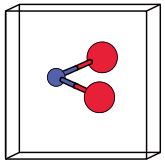
Automatic construction of surface models



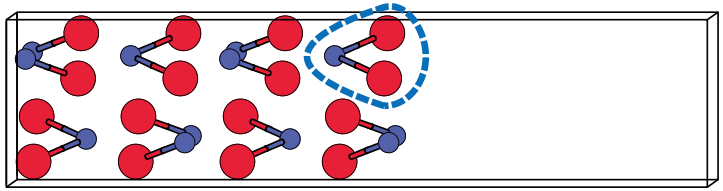


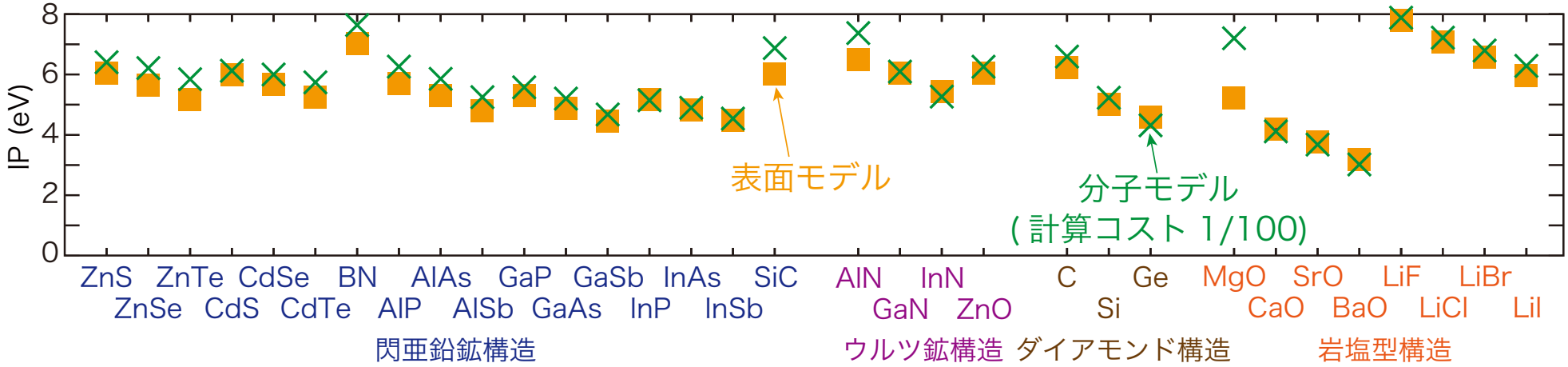
Molecular model approximation

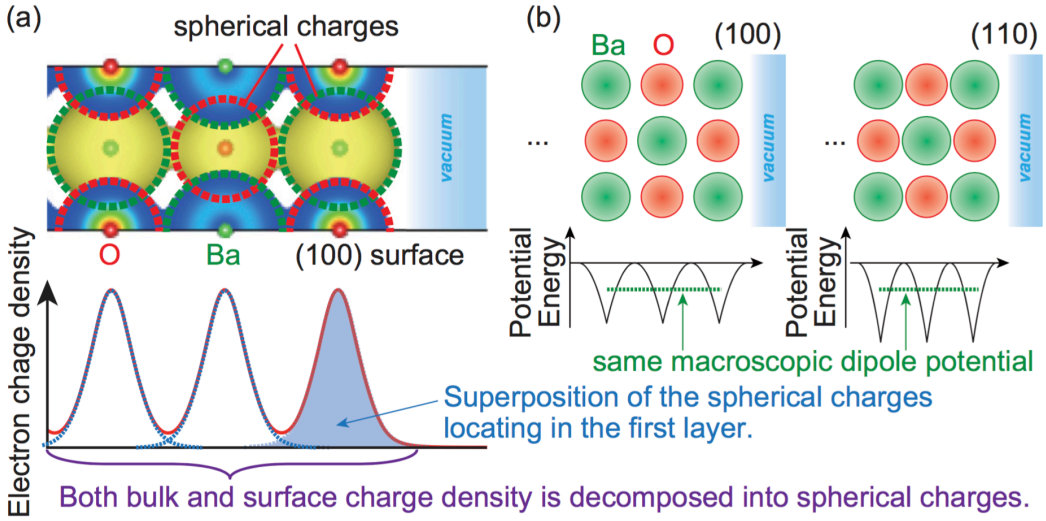
原子・分子モデル
からのIP近似計算

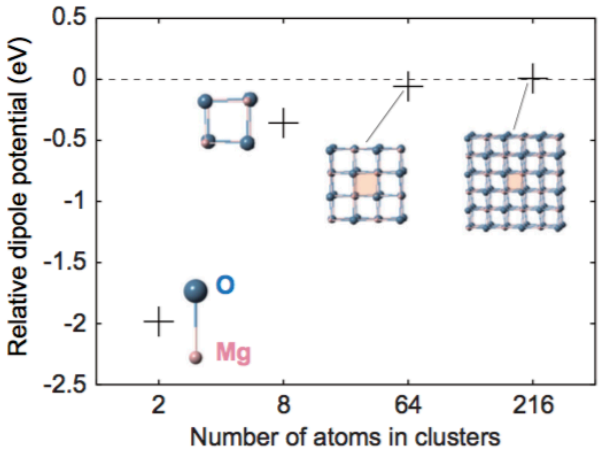


分子モデル





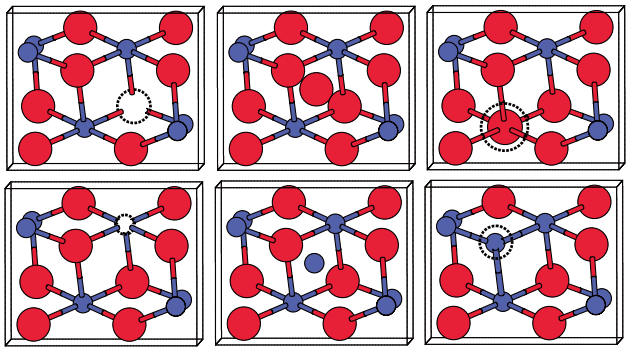




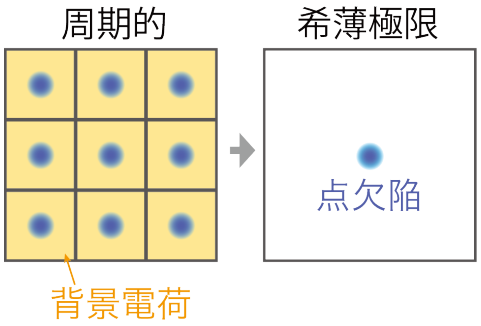
YK, Butler, Walsh, Oba, Phys. Rev. B (2017)

Automation of point defect calculations

様々な欠陥モデルの計算

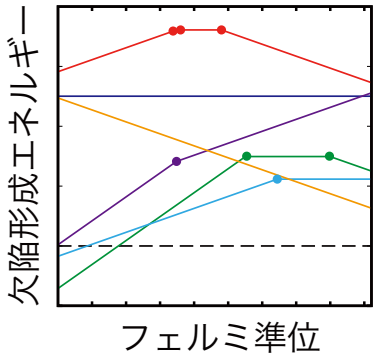


欠陥形成エネルギーの補正



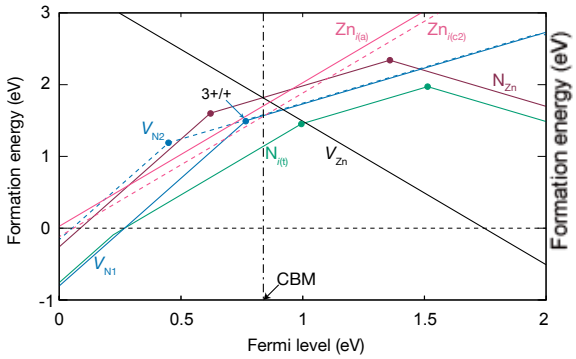
YK *et al.*, Phys. Rev. B (2014)
ibid (2014), Phys. Rev. Appl. (2016)

欠陥形成エネルギーの図



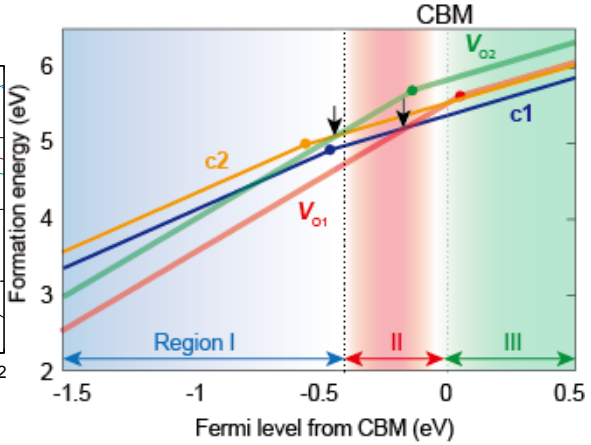
一連の計算プログラムを開発

応用例



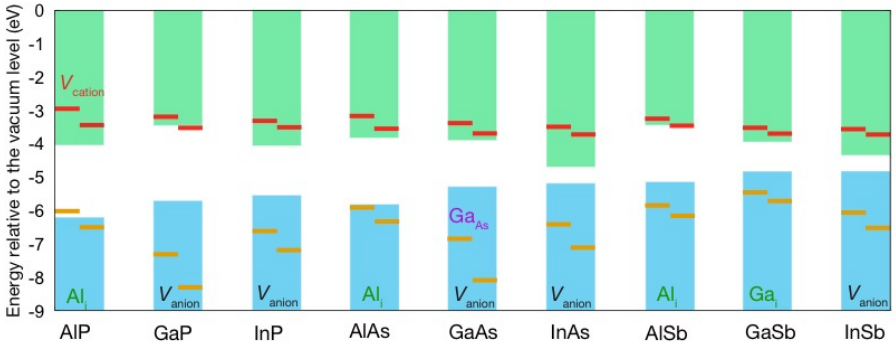
Zn_3N_2の点欠陥

YK, *et al.*,
Phys. Rev. Appl. in press



$SrTiO_3$中の酸素クラスタ

YK and Oba, in preparation



24個の典型的半導体を網羅的に計算

YK, Nishiya, and Oba, in preparation