Data-driven Materials Design and Applications to Two-Dimensional Nano-systems

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HZDR Georgian-German Science Bridge 2025

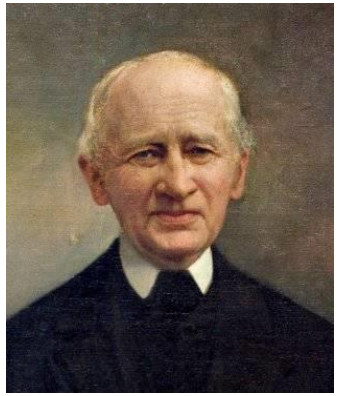
Predictive Theory

Discovery of Neptune

universal law of gravitation

$$\mathbf{F}_1 = \gamma \frac{m_1 m_2}{r_{12}^2} \frac{\mathbf{r}_{12}}{r_{12}} = m_1 \ddot{\mathbf{r}}_1$$

Urbain Le Verrier
(1811-1877)



Johann Gottfried Galle
(1812-1910)

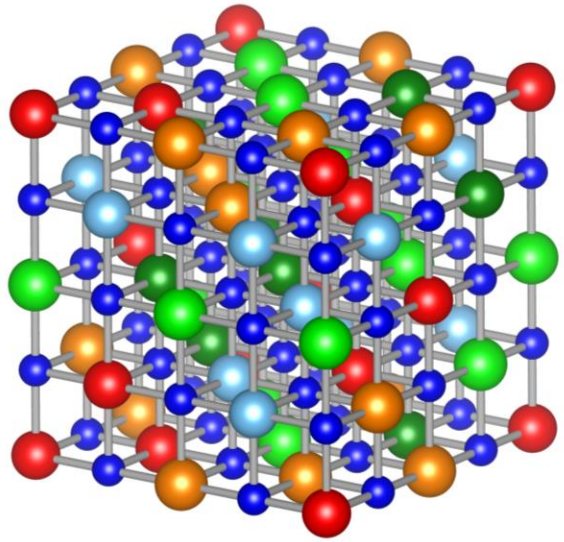
Predictive Solid-state Theory

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

→ problem of ~10^{23} interacting particles

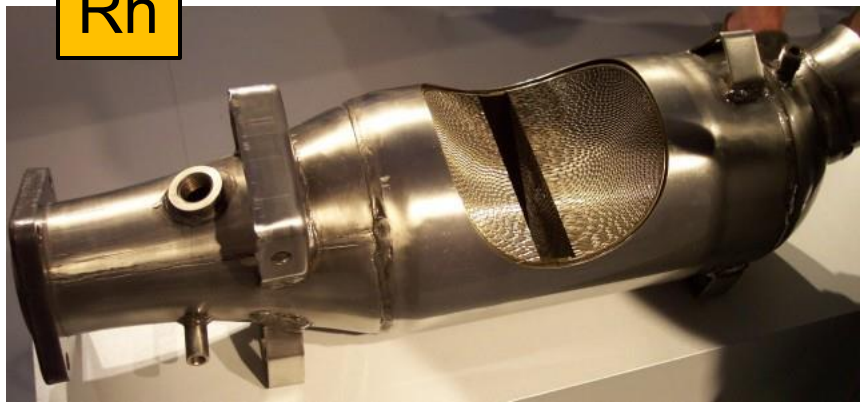
$$E_{\mathrm{KS}}[n] = T_{\mathrm{s}}[\{\phi_i[n]\}] + V_{\mathrm{eK}}[n] + U_{\mathrm{H}}[n] + E_{\mathrm{xc}}[n]$$

→ DFT as ultimate work horse for predictive materials discovery

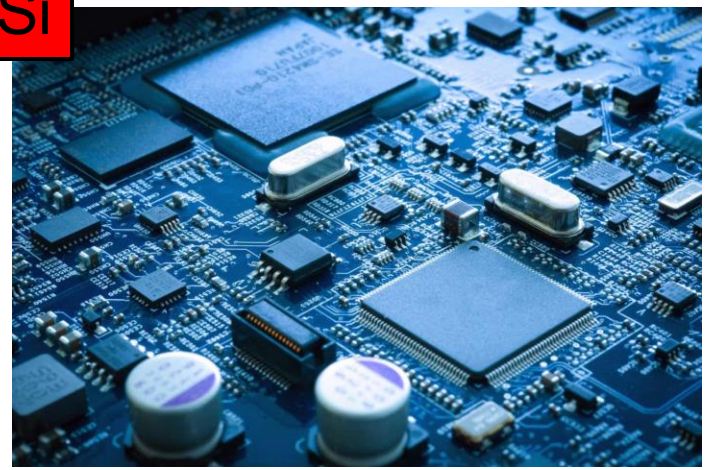


Materials, Science & Technology

Rh



Si

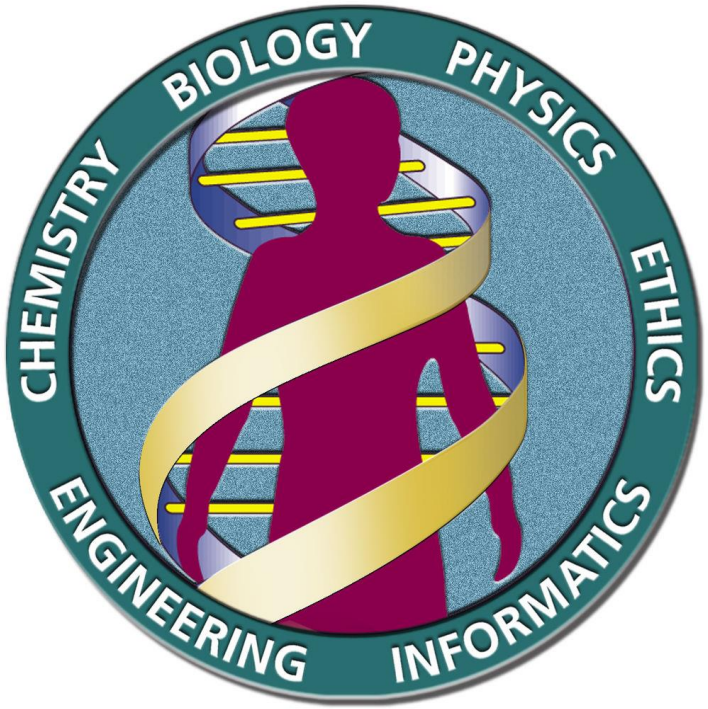


Li

CoPt

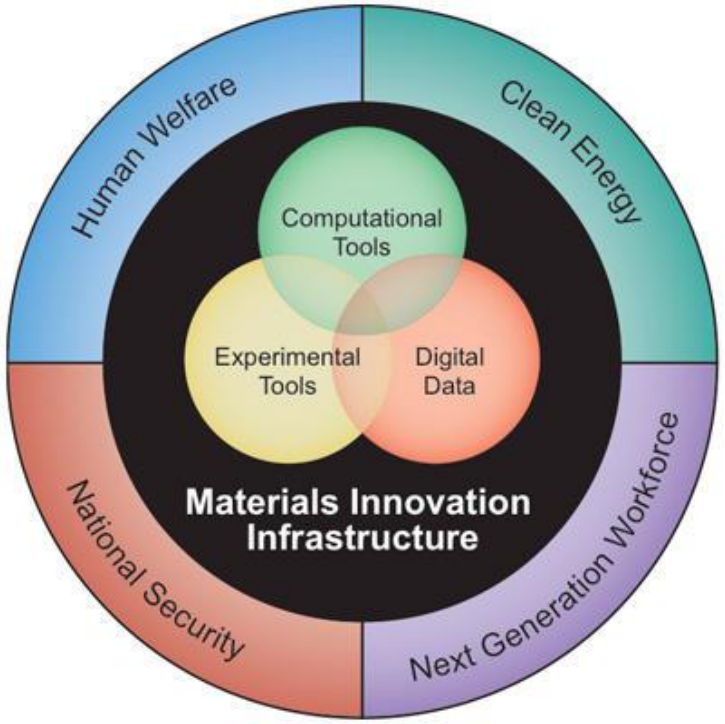
Materials Genome Initiative

Human Genome Project (1990-2003)



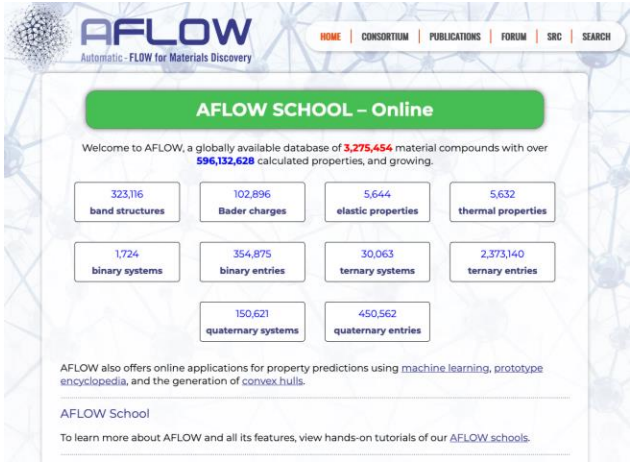
By U.S. Department of Energy, Human Genome Project - http://www.ornl.gov/hgmis, Public Domain, https://commons.wikimedia.org/w/index.php?curid=2485616

Materials Genome Initiative (2011)

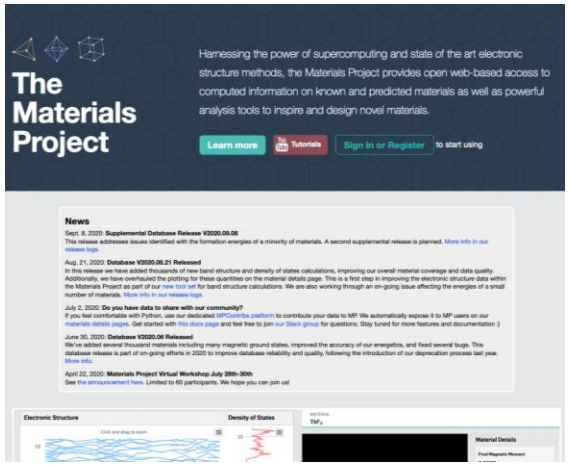


https://www.mgi.gov/

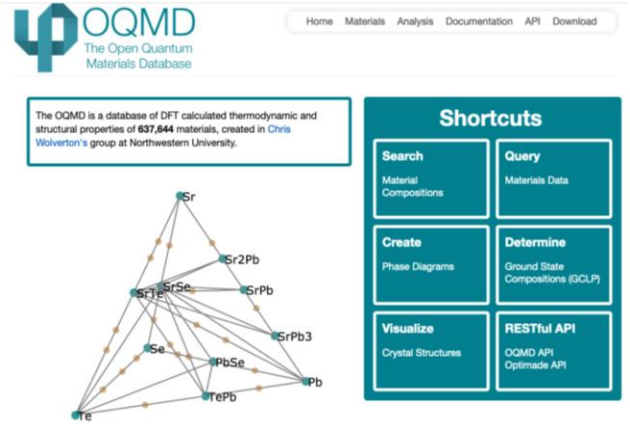
Materials Genome Initiative – Databases



http://aflow.org/

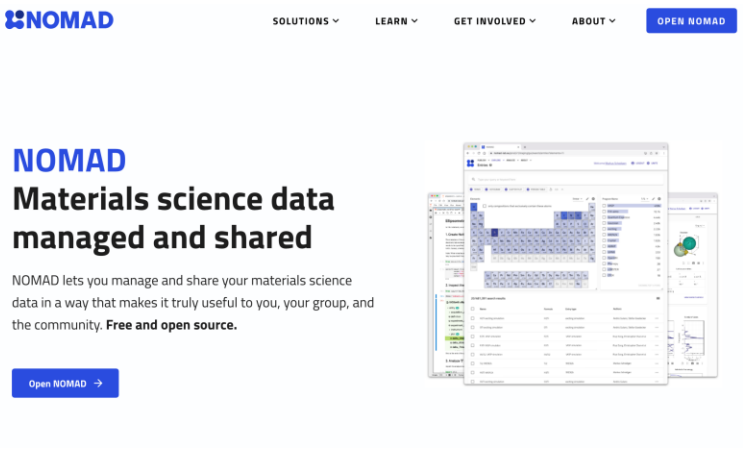


https://materialsproject.org/



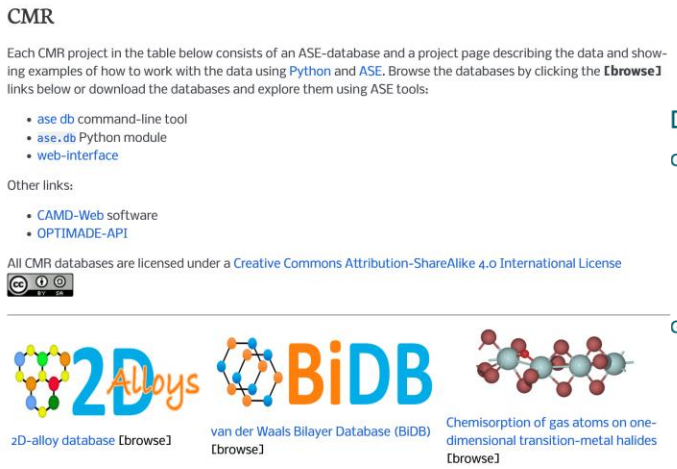
http://oqmd.org/

...

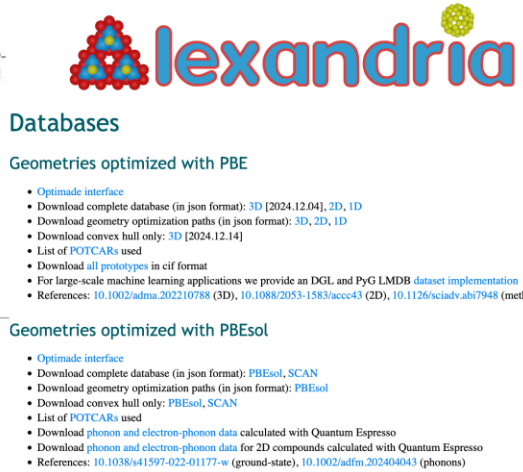


https://nomad-lab.eu/nomad-lab/

https://www.materialscloud.org/



https://cmr.fysik.dtu.dk/



https://alexandria.icams.rub.de/

Outline

1. The AFLOW Database and Software

- AFLOW Apps and search functionality

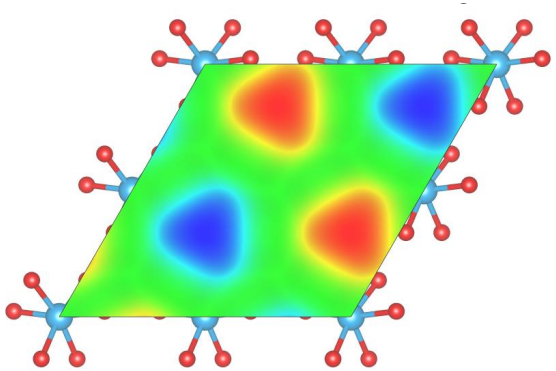
2. Data-driven Quest for Non-vdW 2D Materials

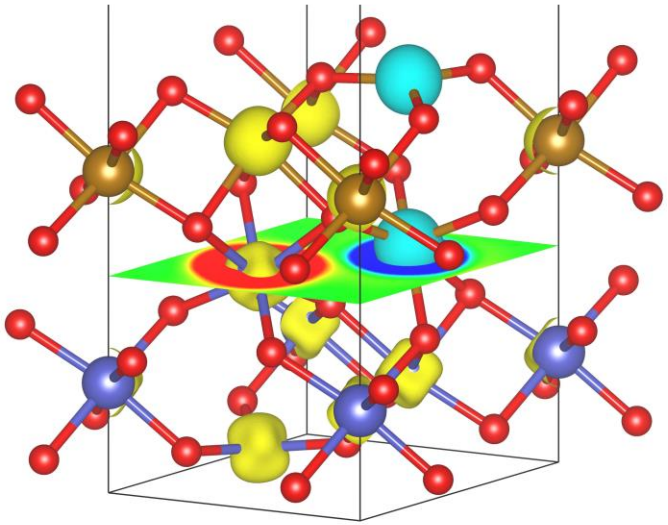
- Search criterion
- Exfoliation energies and properties

3. Magnetic State Control via Hydrogenation

- Surface Passivation
- Magnetic state switching

AFLOW
Automatic - FLOW for Materials Discovery





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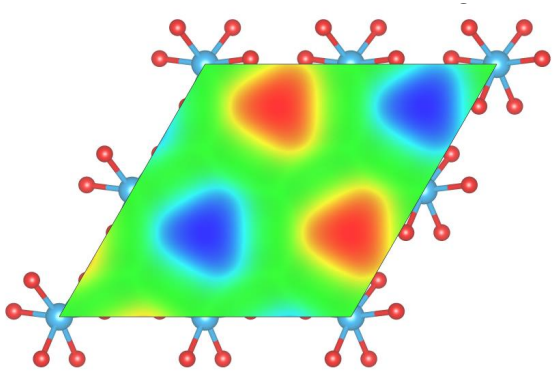
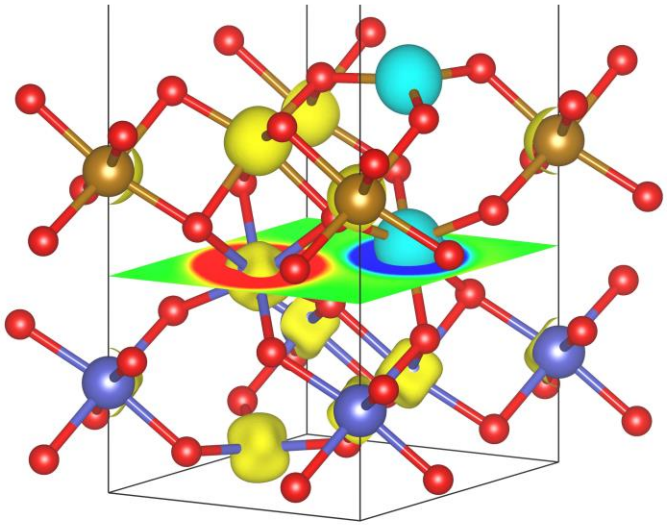
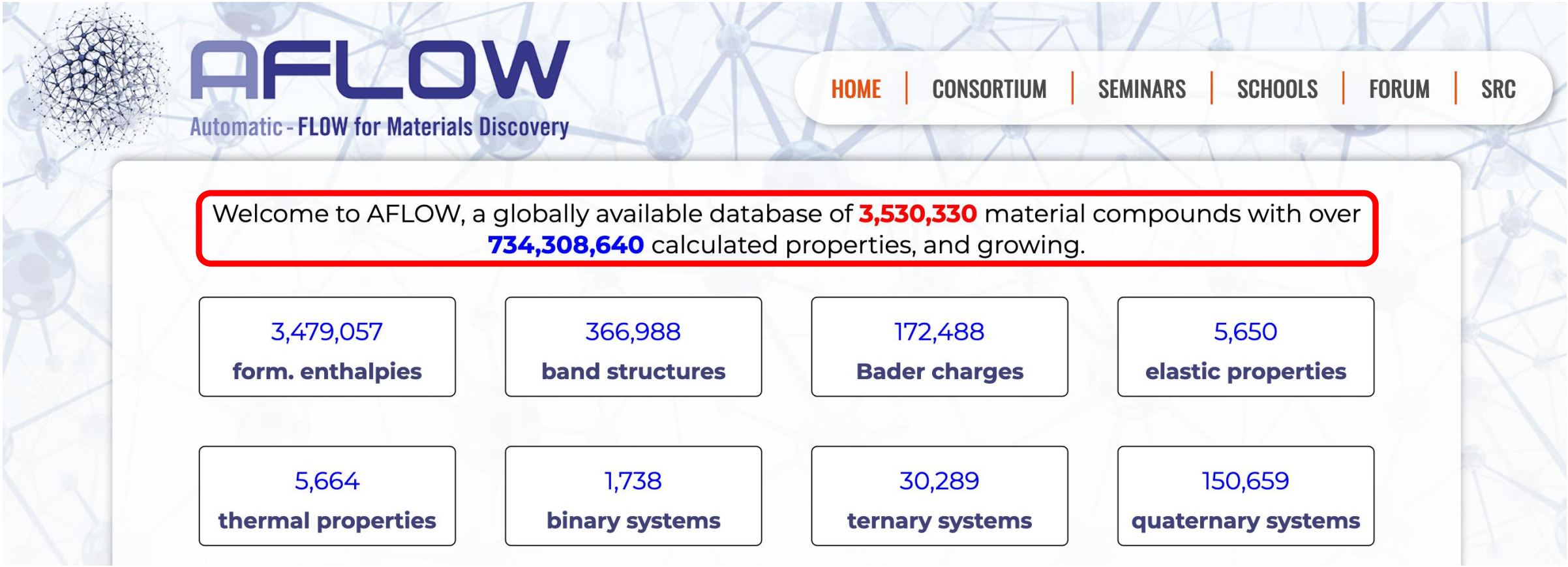




AFLOW Database & Software: aflow.org



Duke UNIVERSITY

S. Curtarolo *et al.*, Comput. Mater. Sci. **58**, 218 (2012); S. Curtarolo *et al.*, Comput. Mater. Sci. **58** 227 (2012);
M. Esters *et al.*, Comput. Mater. Sci. **216**, 111808 (2023);
C. Oses *et al.*, Comput. Mater. Sci. **217**, 111889 (2023);
S. Divilov *et al.*, High Entropy Alloys & Materials **3**, 178 (2025)

UNTERSTÜTZT VON / SUPPORTED BY
Alexander von HUMBOLDT STIFTUNG

AFLOW Software & Apps at aflow.org

AFLOW

Automatic - FLOW for Materials Discovery

S. Curtarolo *et al.*, Comput. Mater. Sci. **58**, 218 (2012)

- written in C++ utilizing VASP as density functional theory program
- AFLOW standard for:
  - *k*-point sets
  - plane wave cutoffs
  - PAW data sets ...

C. E. Calderon *et al.*, Comput. Mater. Sci. **108**, 233 (2015)

b-initio VASP Vienna Package Simulation

G. Kresse and J. Hafner, Phys. Rev. B **49**,14251 (1994)
G. Kresse and J. Furthmüller, Phys. Rev. B **54**,11169 (1996)
G. Kresse and J. Furthmüller, Comput. Mater. Sci. **6**,15 (1996)

MendeLIB search

AFLOW database search application

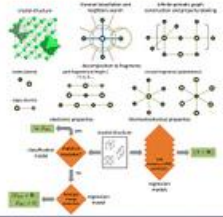
AFLOW-online

Online interface for AFLOW's symmetry, structure comparison, CCE, POCC, and other functionality

Prototype encyclopedia

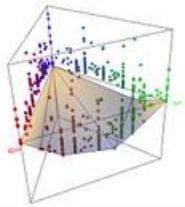
The AFLOW Prototype Encyclopedia with over 1,100 prototypes

AFLOW-ML



Machine Learning application for the PLMF, MDF, and ASC model

AFLOW-CHULL



Convex HULL application for thermodynamic stability and synthesizability

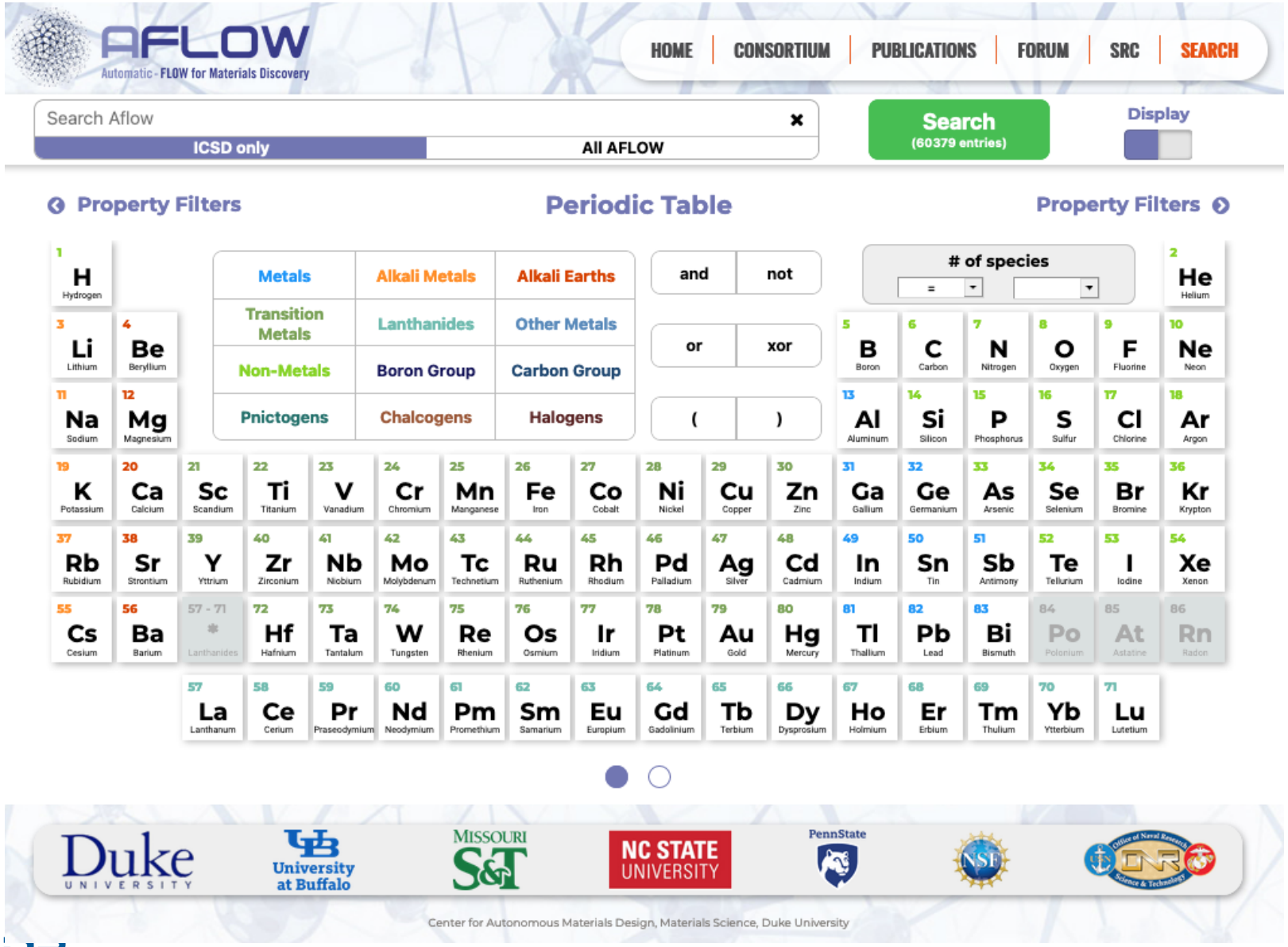
REST-API Docs

AFLOW REST-API WIKI

Documentation for the AFLOW REST-API

- the AFLOW software is structured into purpose dedicated, fully interoperable modules

AFLOW.org Search Page



R. H. Taylor *et al.*, Comput. Mater. Sci. **93**, 178-192 (2014); M. Esters *et al.*, Comput. Mater. Sci. **216**, 111808 (2023)

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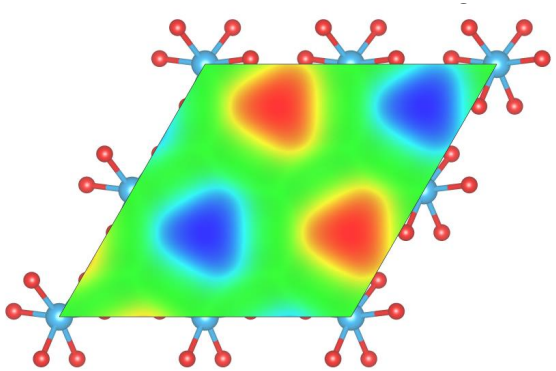
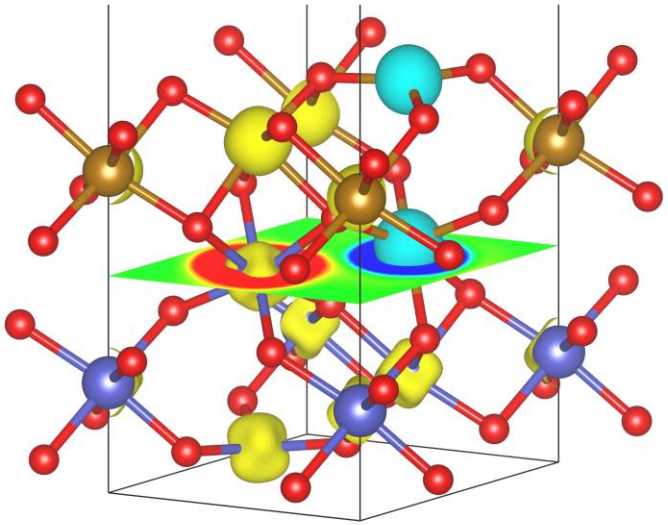
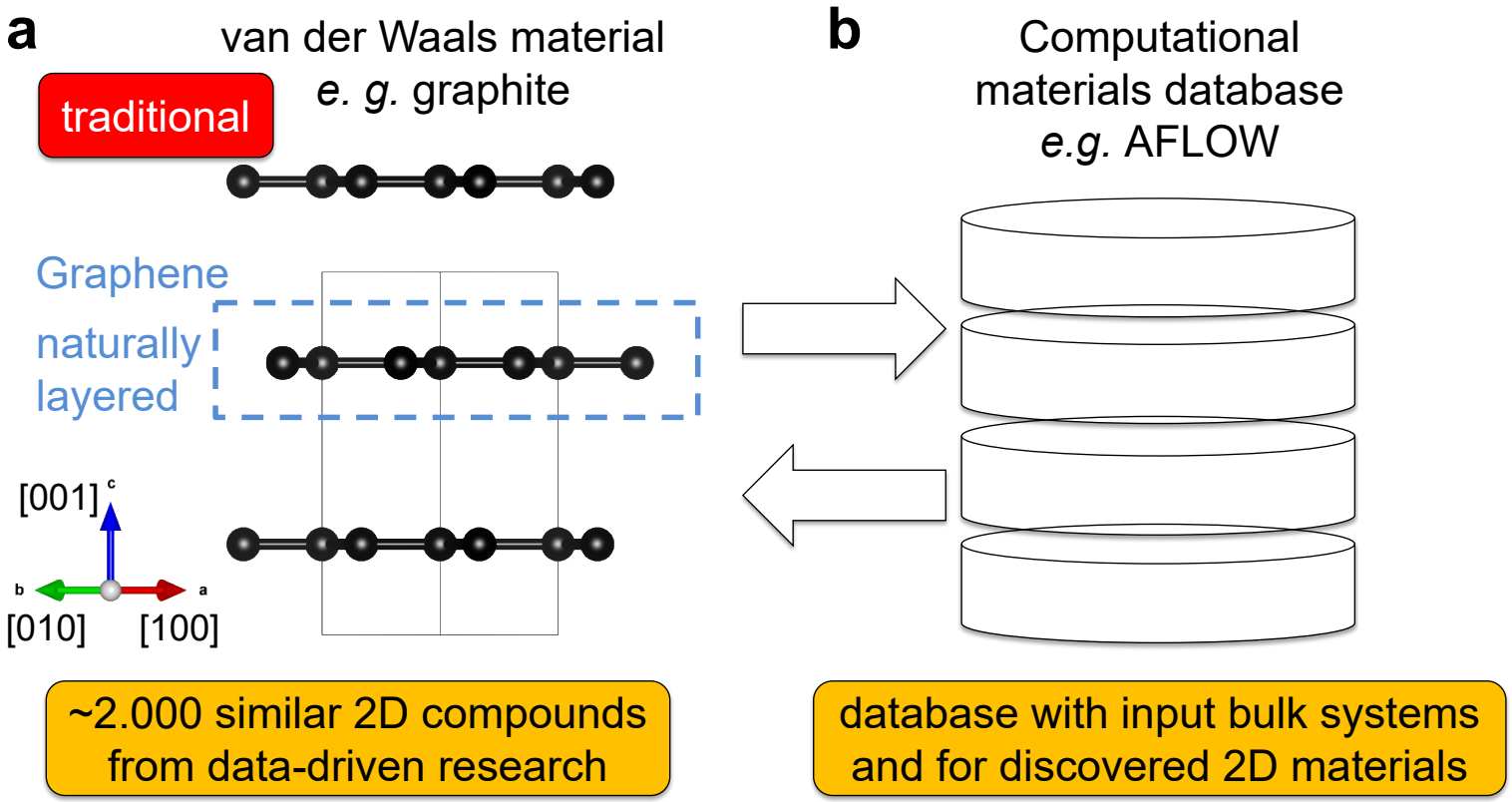




Traditional vs Computational 2D Materials



~2.000 similar 2D compounds from data-driven research

database with input bulk systems and for discovered 2D materials

- data-driven research predicted several thousands of vdW 2D systems

N. Mounet *et al.*, Nat. Nanotechnol. **13**, 246 (2018)
See also: S. Lebegue *et al.*, Phys. Rev. X **3**, 031002 (2013); G. Cheon *et al.*, Nano. Lett. **17**, 1915 (2017)

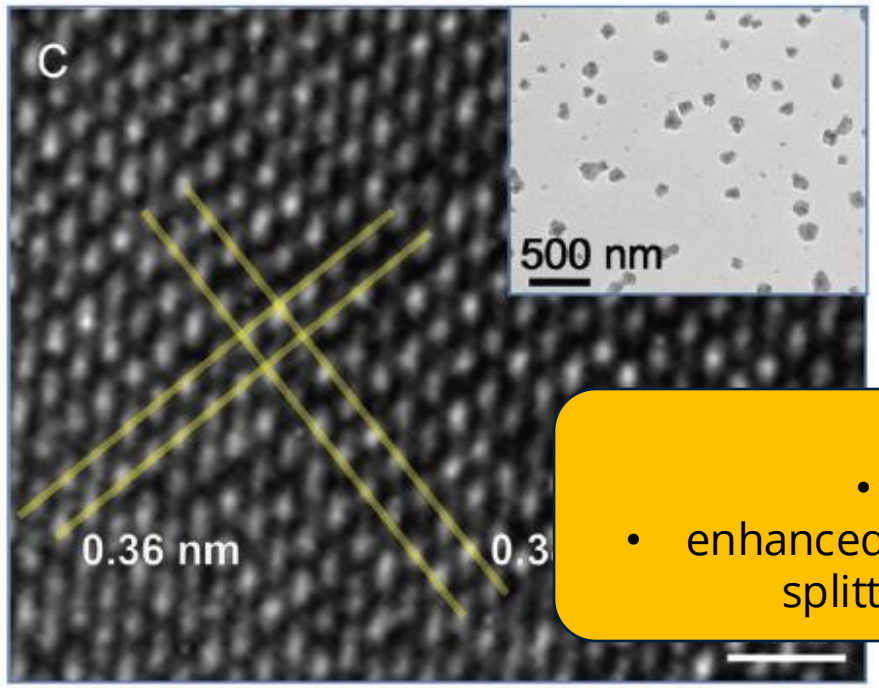
→ Can we obtain 2D sheets from non-vdW materials?

COMMUNICATION
2D Nanomaterials
ADVANCED MATERIALS
www.advmat.de

Electrostatic-Driven Exfoliation and Hybridization of 2D Nanomaterials

Guijian Guan, Jing Xia, Shuhua Liu, Yuan Cheng, Shiqiang Bai, Si Yin Tee, Yong-Wei Zhang, and Ming-Yong Han**

WO_3 exfoliation

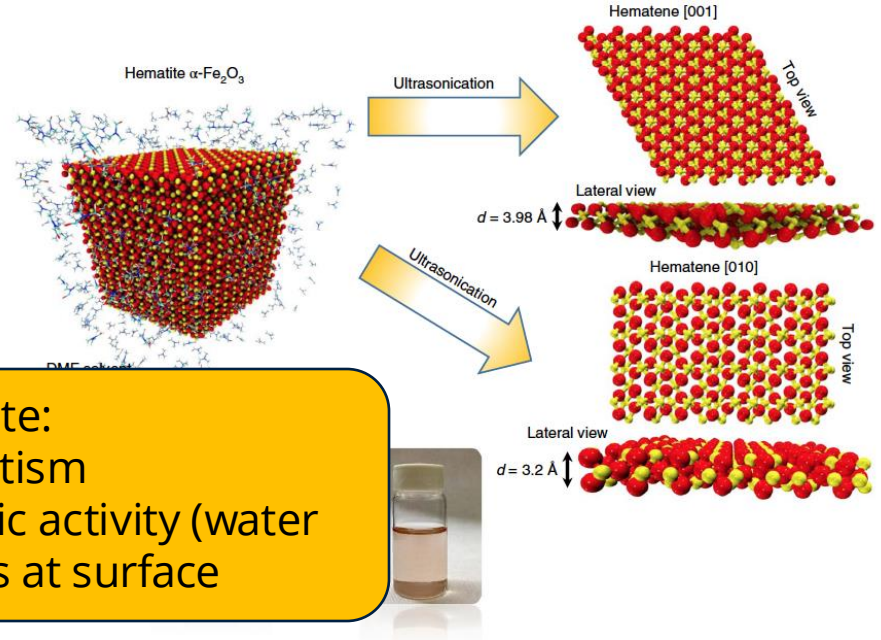


G. Guan *et al.*, Adv. Mater. **29**, 1700326 (2017)

ARTICLES
https://doi.org/10.1038/s41565-018-0134-y
nature nanotechnology

Exfoliation of a non-van der Waals material from iron ore hematite

Aravind Puthirath Balan[1,2,11], Sruthi Radhakrishnan[1,11], Cristiano F. Woellner[3], Shyam K. Sinha[4], Liangzi Deng[5], Carlos de los Reyes[6], Banki Manmadha Rao[7], Maggie Paulose[7], Ram Neupane[7], Amey Apte[1], Vidya Kochat[1], Robert Vajtai[1], Avetik R. Harutyunyan[8], Ching-Wu Chu[5,9], Gelu Costin[10], Douglas S. Galvao[3], Angel A. Martí[6], Peter A. van Aken[4], Oomman K. Varghese[7], Chandra Sekhar Tiwary[1]*, Anantharaman Malie Madom Ramaswamy Iyer[1,2]* and Pulickel M. Ajayan[1]*



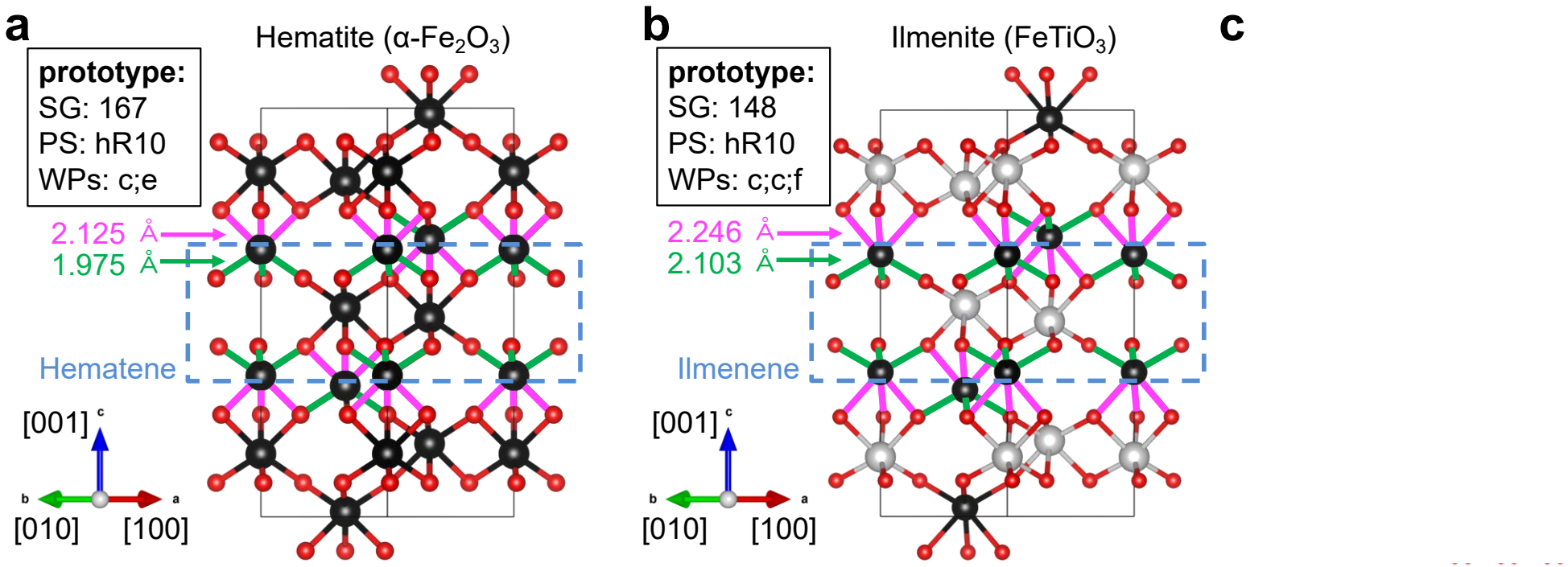
results indicate:
- ferromagnetism
- enhanced photocatalytic activity (water splitting) → cations at surface

A. Puthirath Balan *et al.*, Nat. Nanotechnol. **13**, 602 (2018)
A. Puthirath Balan *et al.*, Chem. Mater. **30**, 5923 (2018)
A. Puthirath *et al.*, J. Phys. Chem. C **125**, 18927 (2021)

→ Results shatter intuitive understanding of exfoliability (and cleavage)

See also: K. Jiang *et al.*, Nat. Synth. **2**, 58 (2023)

Discovering Non-vdW 2D Materials



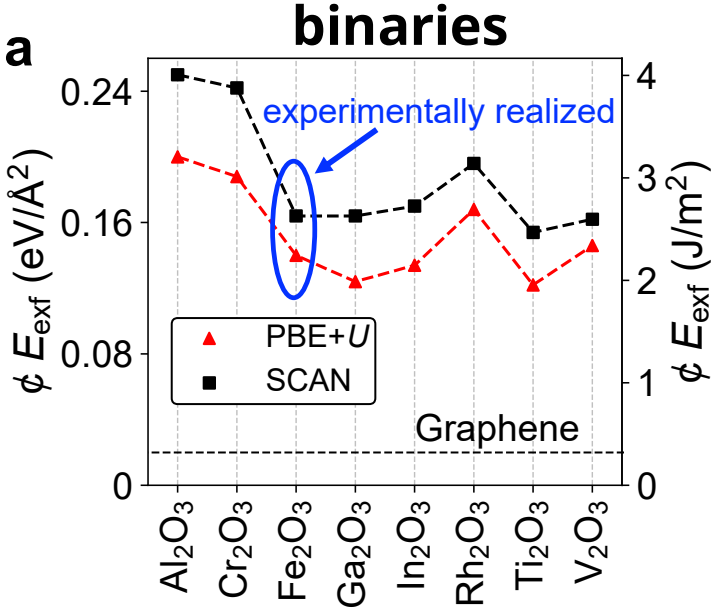
R. Friedrich, M. Ghorbani-Asl, S. Curtarolo and A. V. Krasheninnikov, Nano Lett. **22**, 989 (2022)

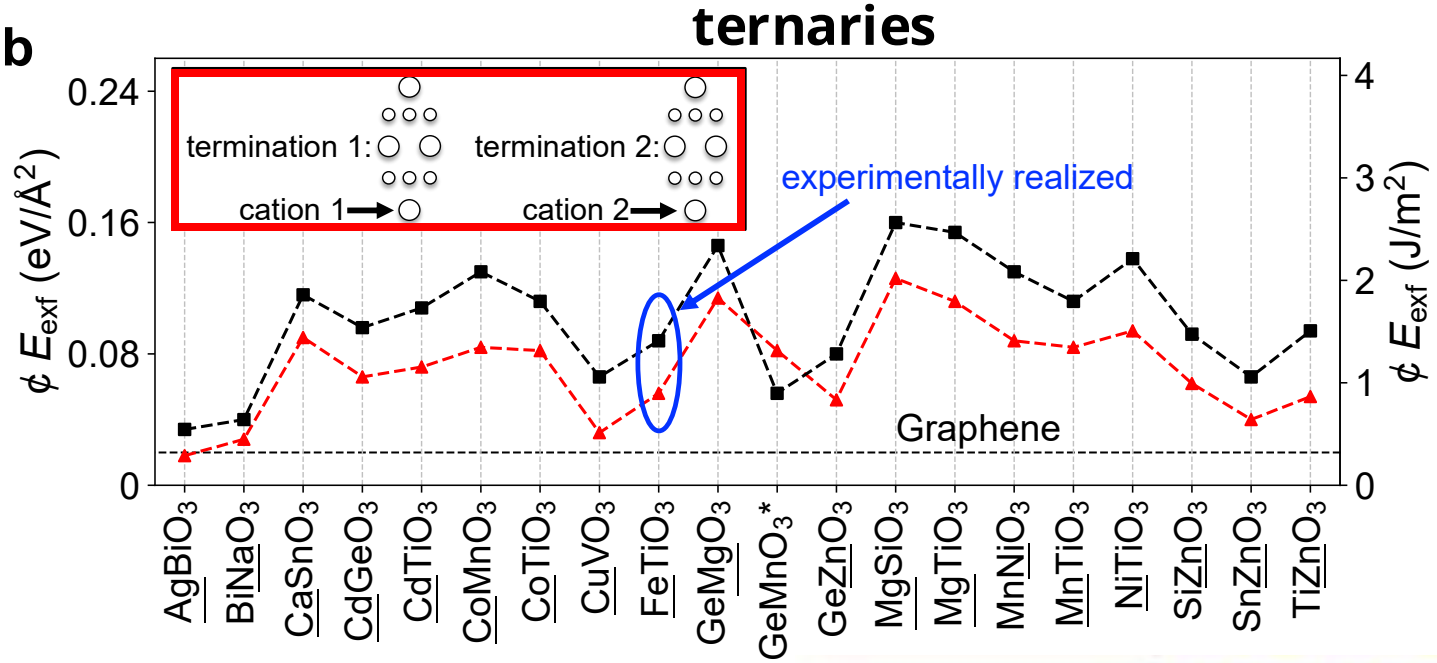
T. Barnowsky, A. V. Krasheninnikov, and R. Friedrich, Adv. El. Mats. **9**, 2201112 (2023)

- use structural prototype of first experimentally realized materials to filter AFLOW database

→ oxides, sulfides, chlorides

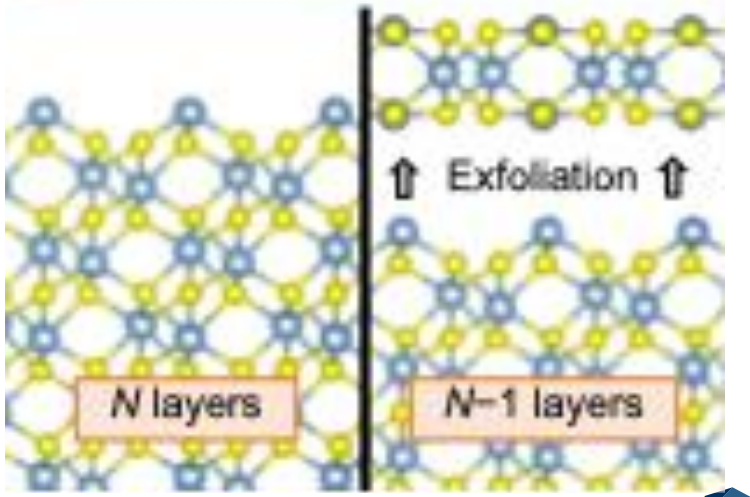
Exfoliation Energies



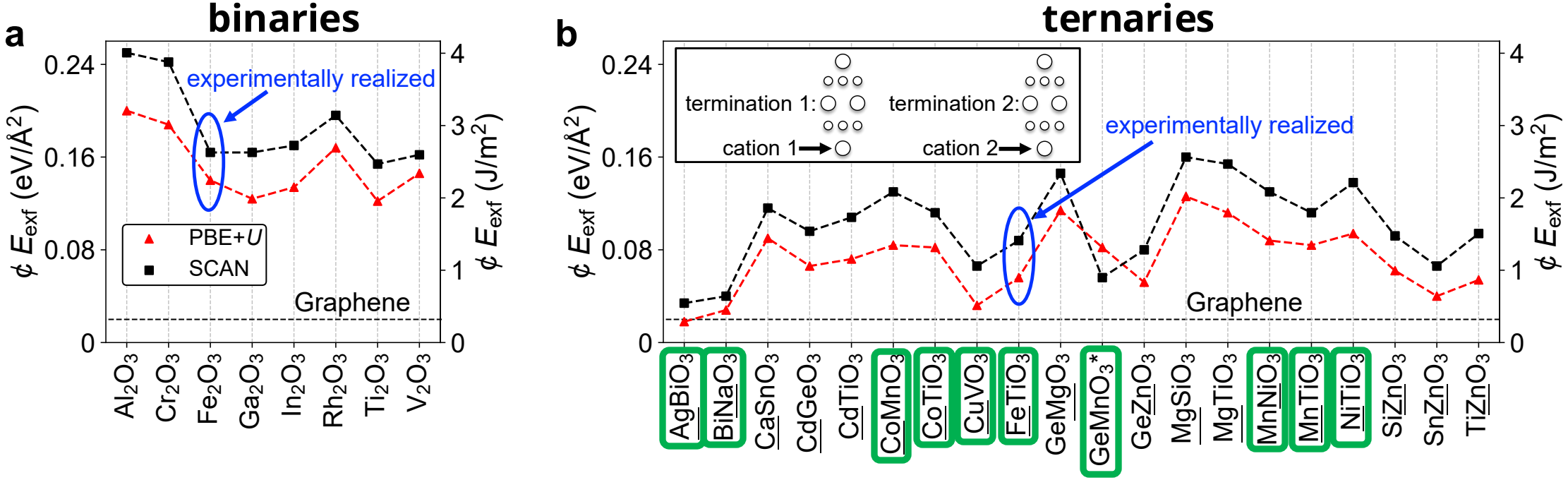


$$\Delta E_{exf} = \frac{E_{slab} - E_{bulk}}{A}$$

J. H. Jung *et al*., Nano Lett. **18**, 2759 (2018)

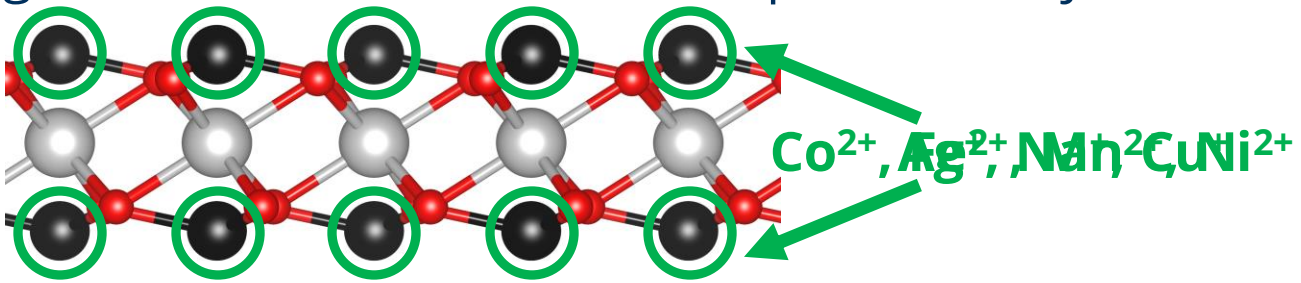


Exfoliation Energies



- all exfoliation energies are close to the ones of exp. realized systems

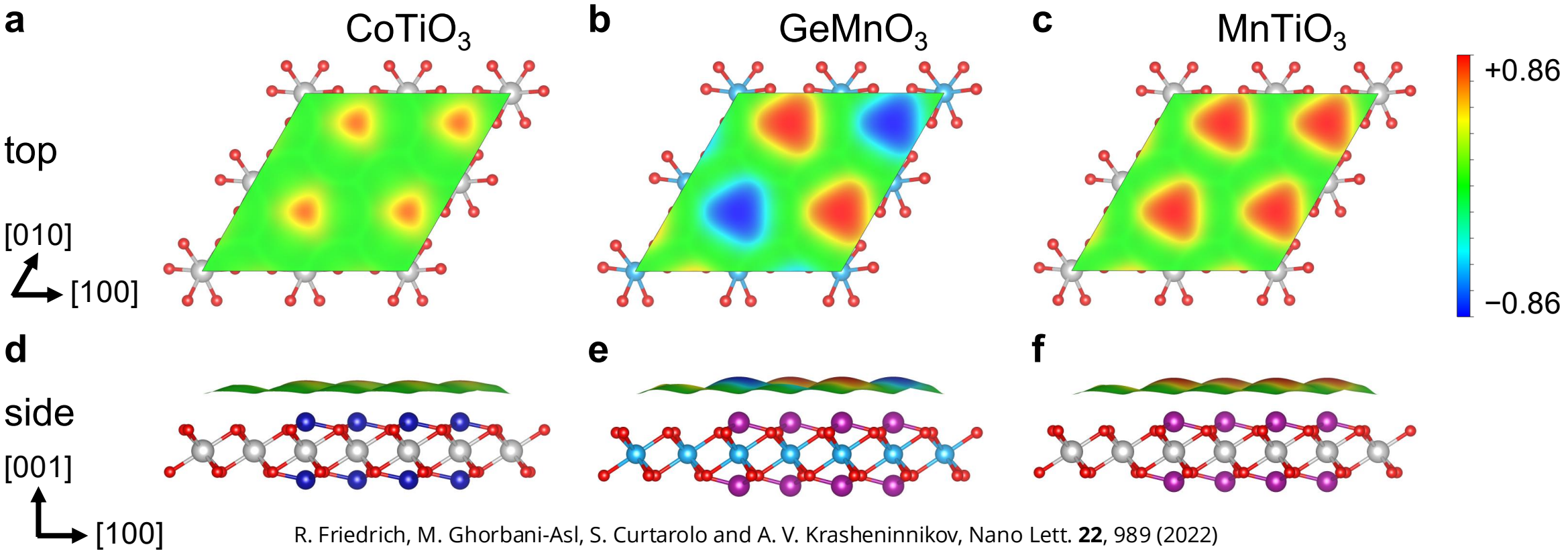
side view



→ **easy exfoliation if cation at surface is in low oxidation state**

→ predictive descriptor for future 2D materials discovery

Magnetism



R. Friedrich, M. Ghorbani-Asl, S. Curtarolo and A. V. Krasheninnikov, Nano Lett. **22**, 989 (2022)

- very diverse surface spin polarization for non-vdW 2D materials

$$P = \frac{n_\uparrow - n_\downarrow}{n_\uparrow + n_\downarrow}$$

measurable by spin-polarized scanning tunneling microscopy

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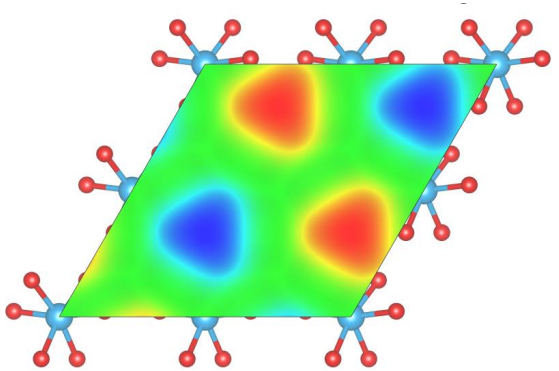
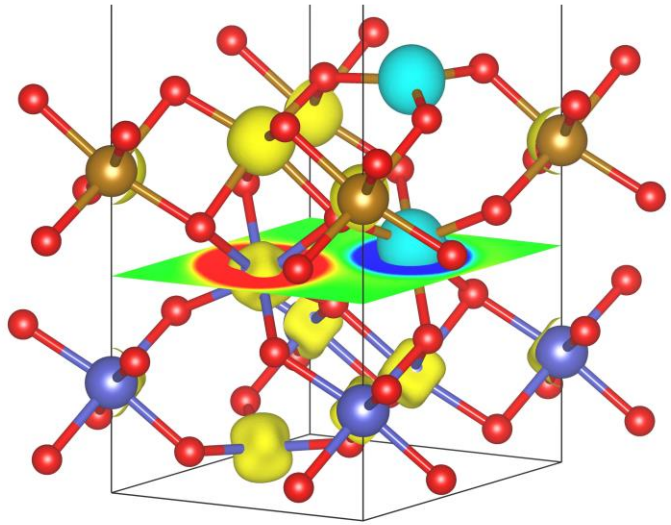
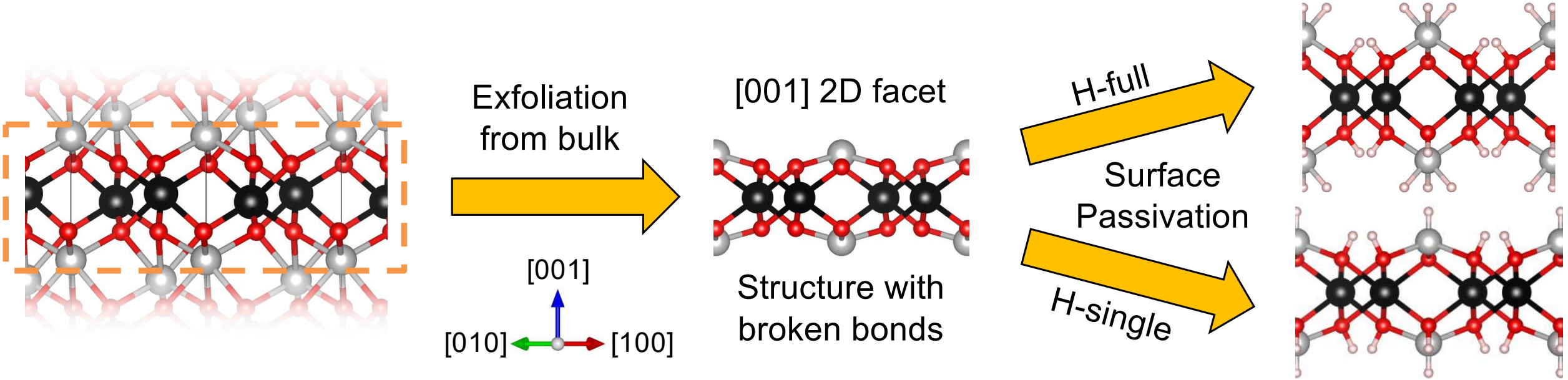




Surface Passivation of Dangling Bonds

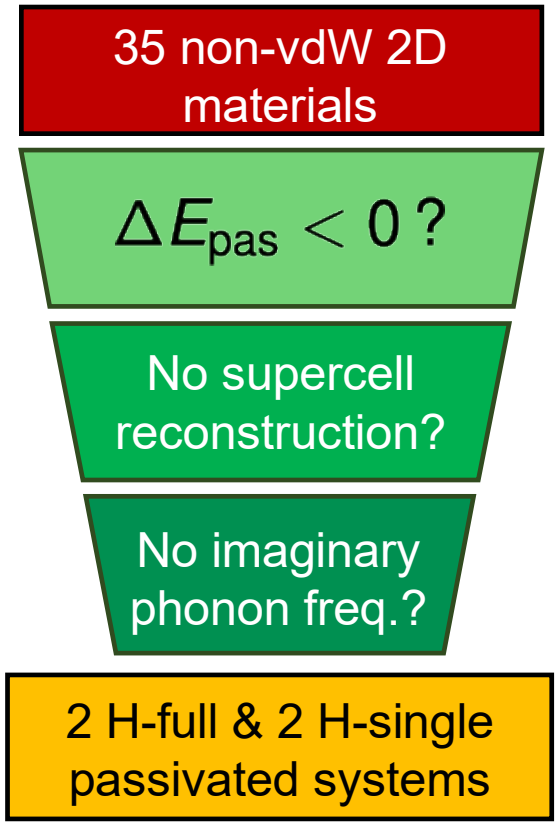


- different surface passivation types mimicking different H chemical potentials

Tom Barnowsky

Filtering Scheme

a



T. Barnowsky, S. Curtarolo, A. V. Krasheninnikov, T. Heine, and R. Friedrich, Nano Letters **24**, 3874 (2024)

- only 4 out of 35 systems pass all stability tests
- special geometry for 2D $CdTiO_3$:H-single → validated from different starting geometries

Filtering Scheme

a

35 non-vdW 2D materials

$\Delta E_{pas} < 0$?

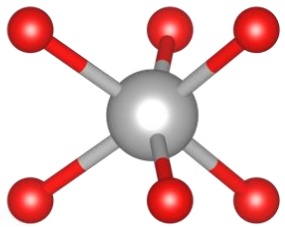
No supercell reconstruction?

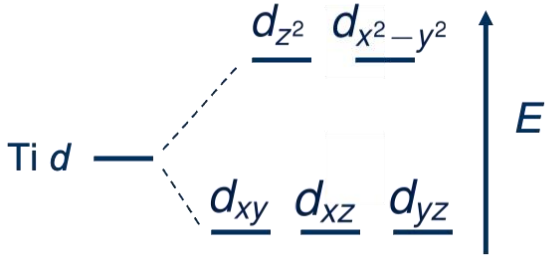
No imaginary phonon freq.?

2 H-full & 2 H-single passivated systems

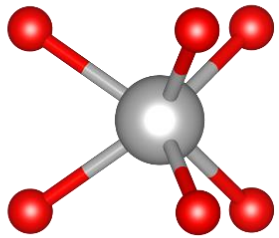
Coordination change during relax.

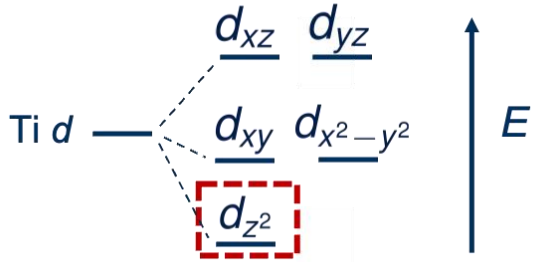
Octahedral





Trigonal-prismatic

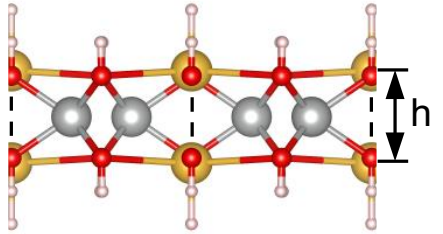


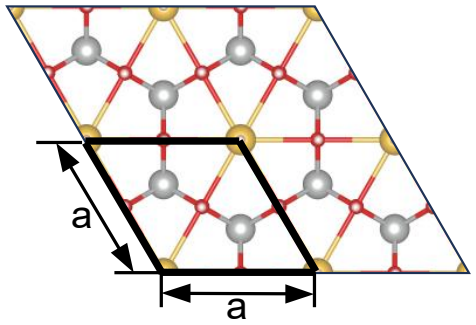


e

2D $\underline{Cd}TiO_3$:H-single

$\Delta E_{pas} = -0.14\,eV/H_2$

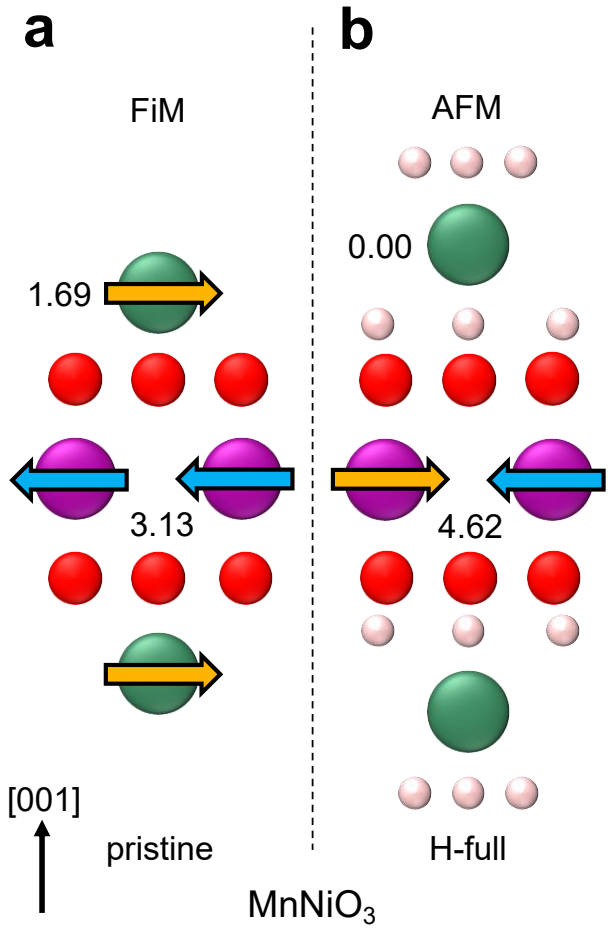




T. Barnowsky, S. Curtarolo, A. V. Krasheninnikov, T. Heine, and R. Friedrich, Nano Letters **24**, 3874 (2024)

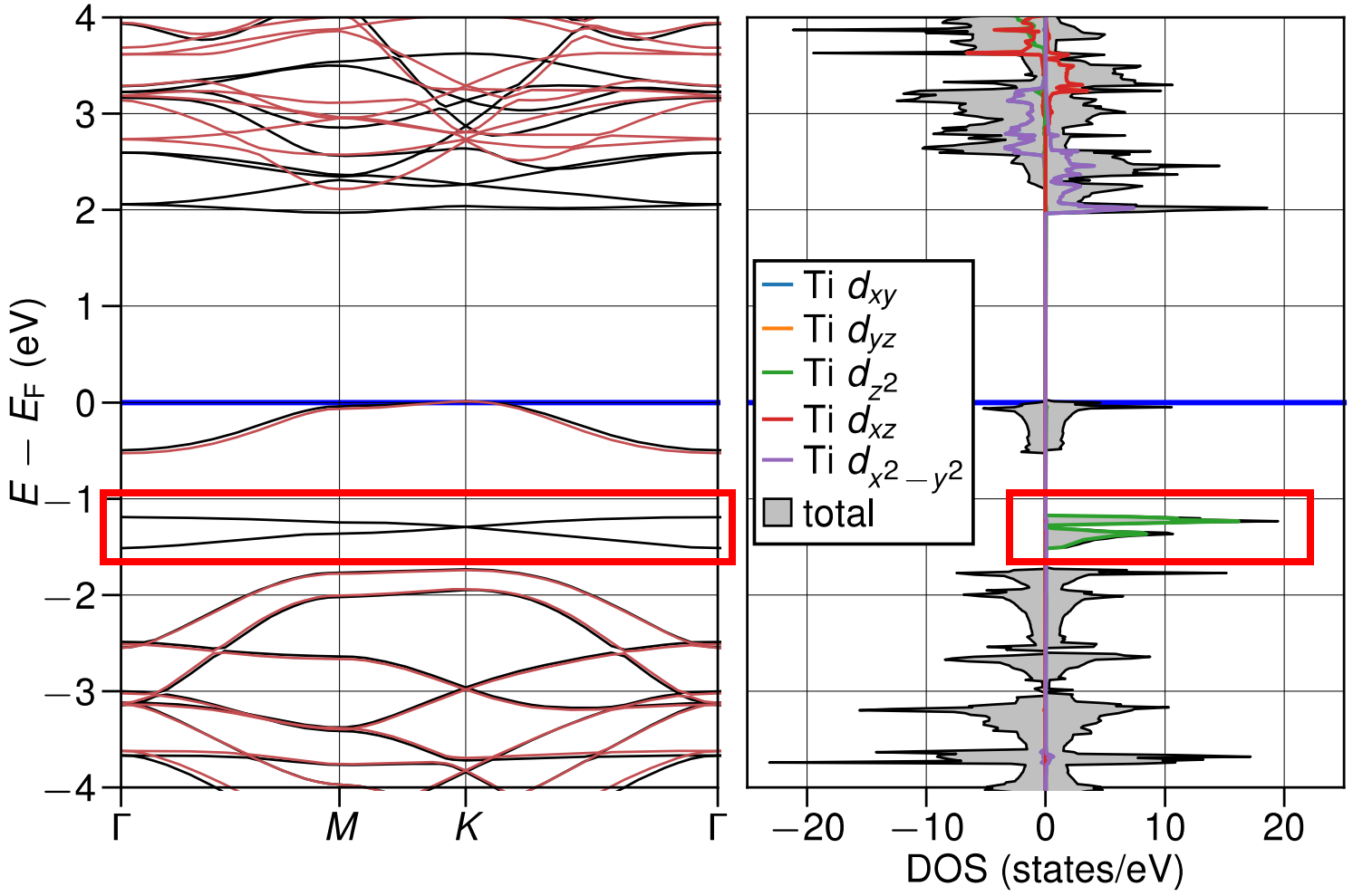
- only 4 out of 35 systems pass all stability tests
- special geometry for 2D $CdTiO_3$:H-single → validated from different starting geometries

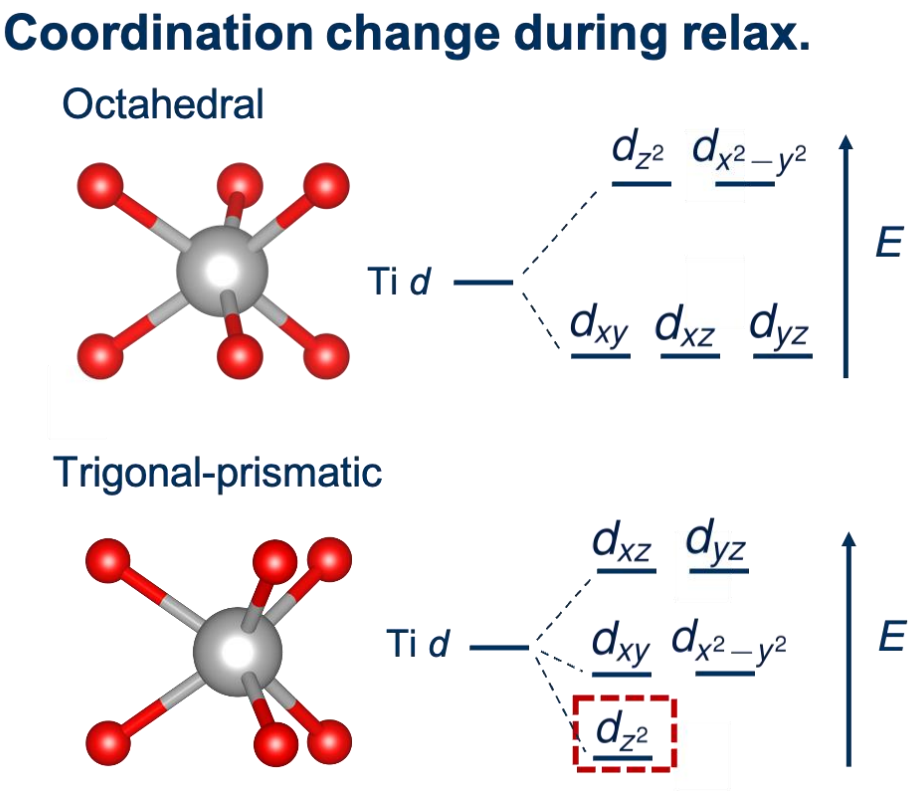
Magnetic Configurations



- switching to states with flipped and enhanced moments due to passivation
- onset of ferromagnetism for $CdTiO_3$:H-single → mean field Curie temperature T_C^{MF}~ 10K

$CdTiO_3$:H-single Density of States





- occupation of $3d_{z^2}$ majority spin bands connected to structural change

Magnetization Density Difference

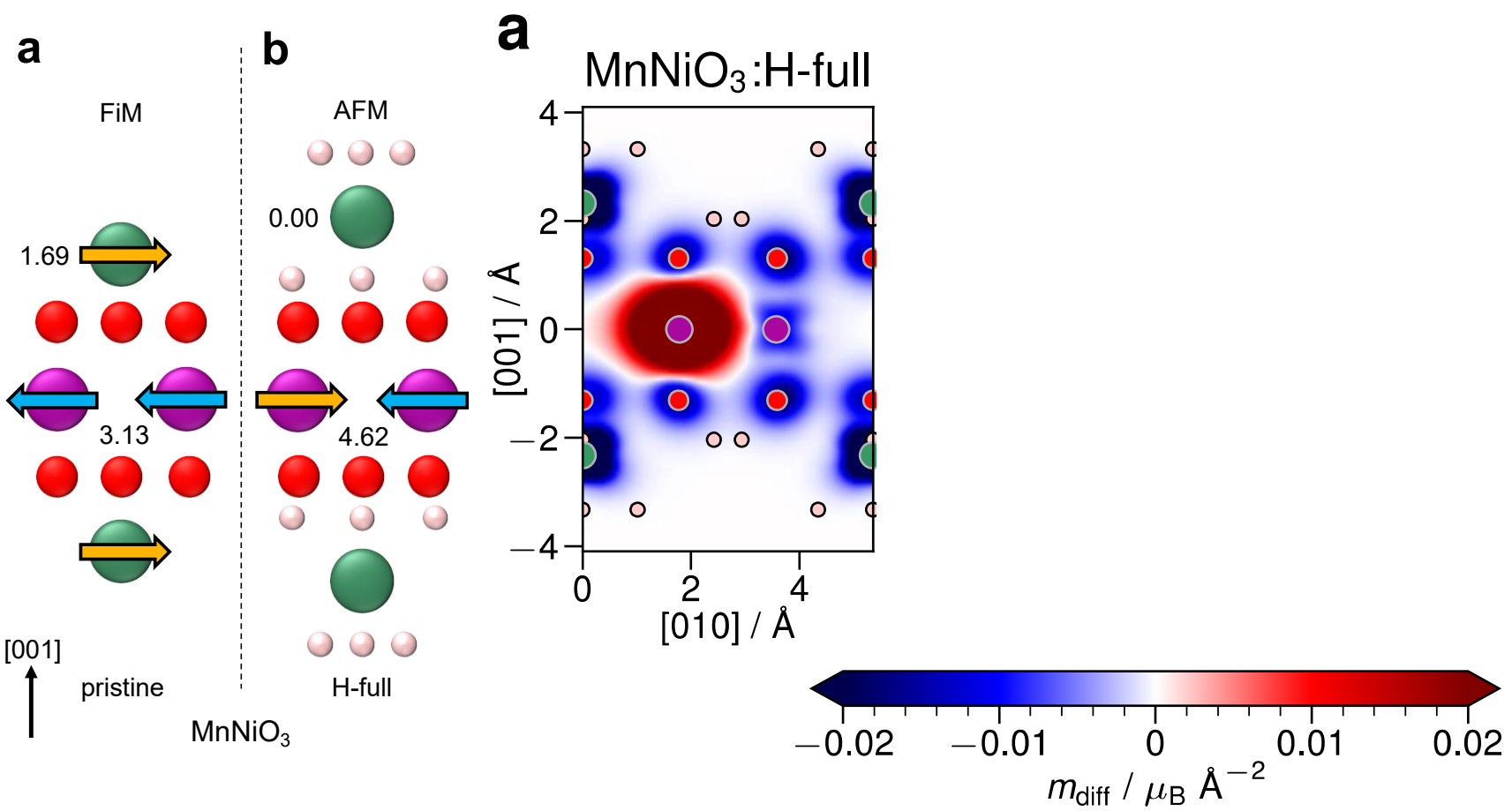
- magnetization density as difference between spin up and spin down electron densities:

$$m(\mathbf{r}) = \mu_{\mathrm{B}}(n_{\uparrow}(\mathbf{r}) - n_{\downarrow}(\mathbf{r}))$$

- magnetization density difference between pristine and passivated systems:

$$m_{\mathrm{diff}}(\mathbf{r}) = m_{\mathrm{passivated}}(\mathbf{r}) - m^{\mathrm{SCF}}_{\mathrm{pristine}}(\mathbf{r})$$

Magnetization Density Difference

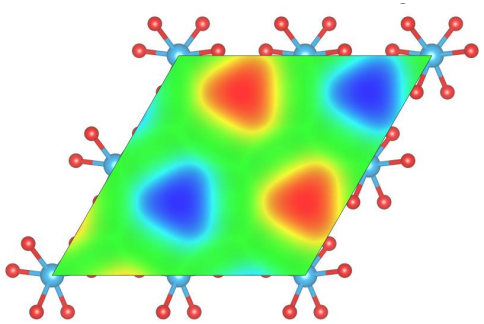


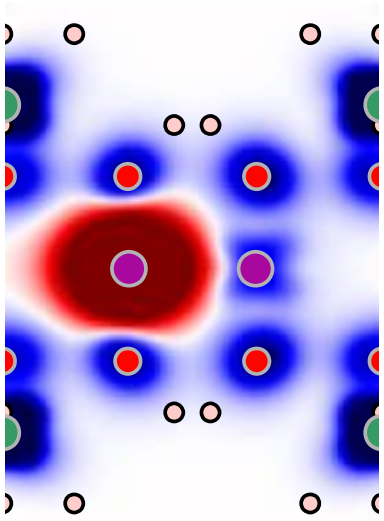
T. Barnowsky, S. Curtarolo, A. V. Krasheninnikov, T. Heine, and R. Friedrich, Nano Letters **24**, 3874 (2024)

- local spin symmetry change for $MnNiO_3$:H-full
- $CdTiO_3$:H-single: spin transfer to Ti in $3d_{z^2}$ orbital shape

Summary and Conclusions

- data-driven materials design is a powerful tool enabling the discovery of novel compounds
- non-van der Waals 2D materials represent an emerging new class of low-dimensional compounds
- their active cation-terminated surfaces give rise to strong surface spin polarization and magnetic state control through passivation





R. Friedrich, M. Ghorbani-Asl, S. Curtarolo and A. V. Krasheninnikov, Nano Lett. **22**, 989 (2022)

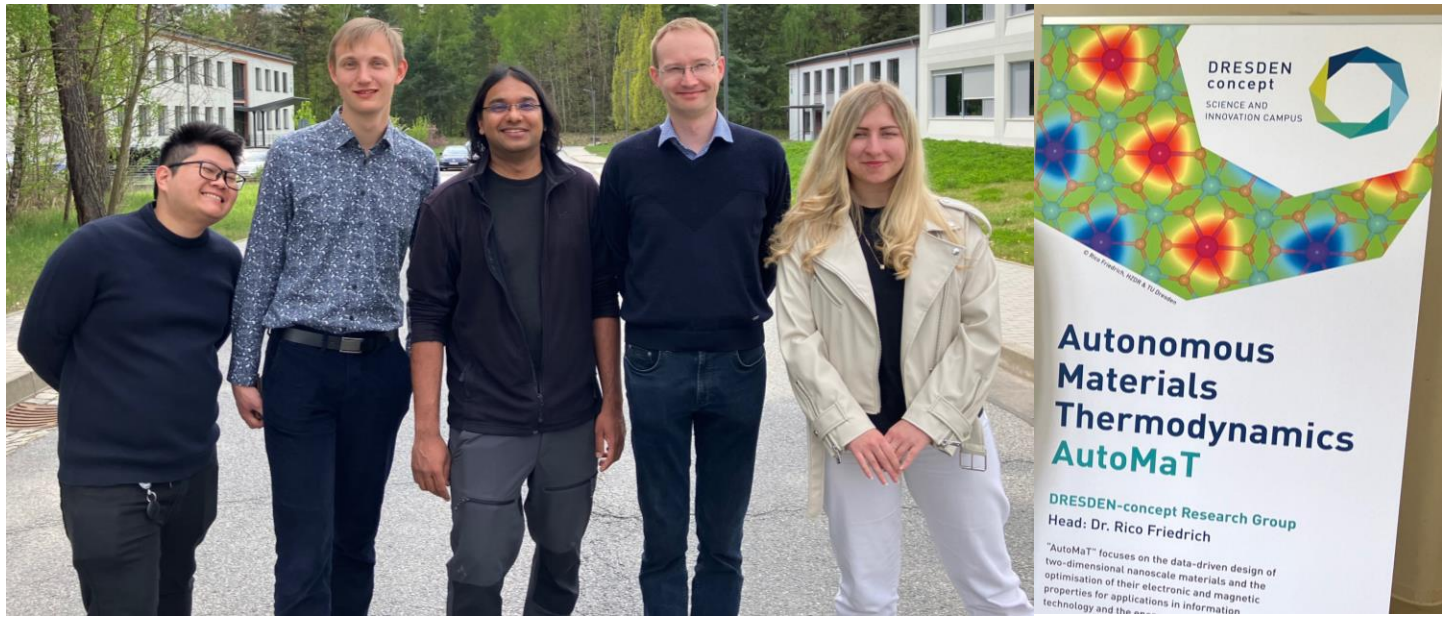
T. Barnowsky, A. V. Krasheninnikov, and R. Friedrich, Adv. El. Mats. **9**, 2201112 (2023)

T. Barnowsky, S. Curtarolo, A. V. Krasheninnikov, T. Heine, and R. Friedrich, Nano Letters **24**, 3874 (2024)

A. Nihei, T. Barnowsky, and R. Friedrich, submitted, arXiv:2503.12209 (2025)

A. P. Balan *et al*., Materials Today **58**, 164 (2022)

Thank You for Your Attention



+

Balaram Thakur

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IFW Dresden:	Axel Lubk, Kornelius Nielsch
Duke University:	Hagen Eckert, Simon Divilov, Stefano Curtarolo
UT Dallas:	Cormac Toher

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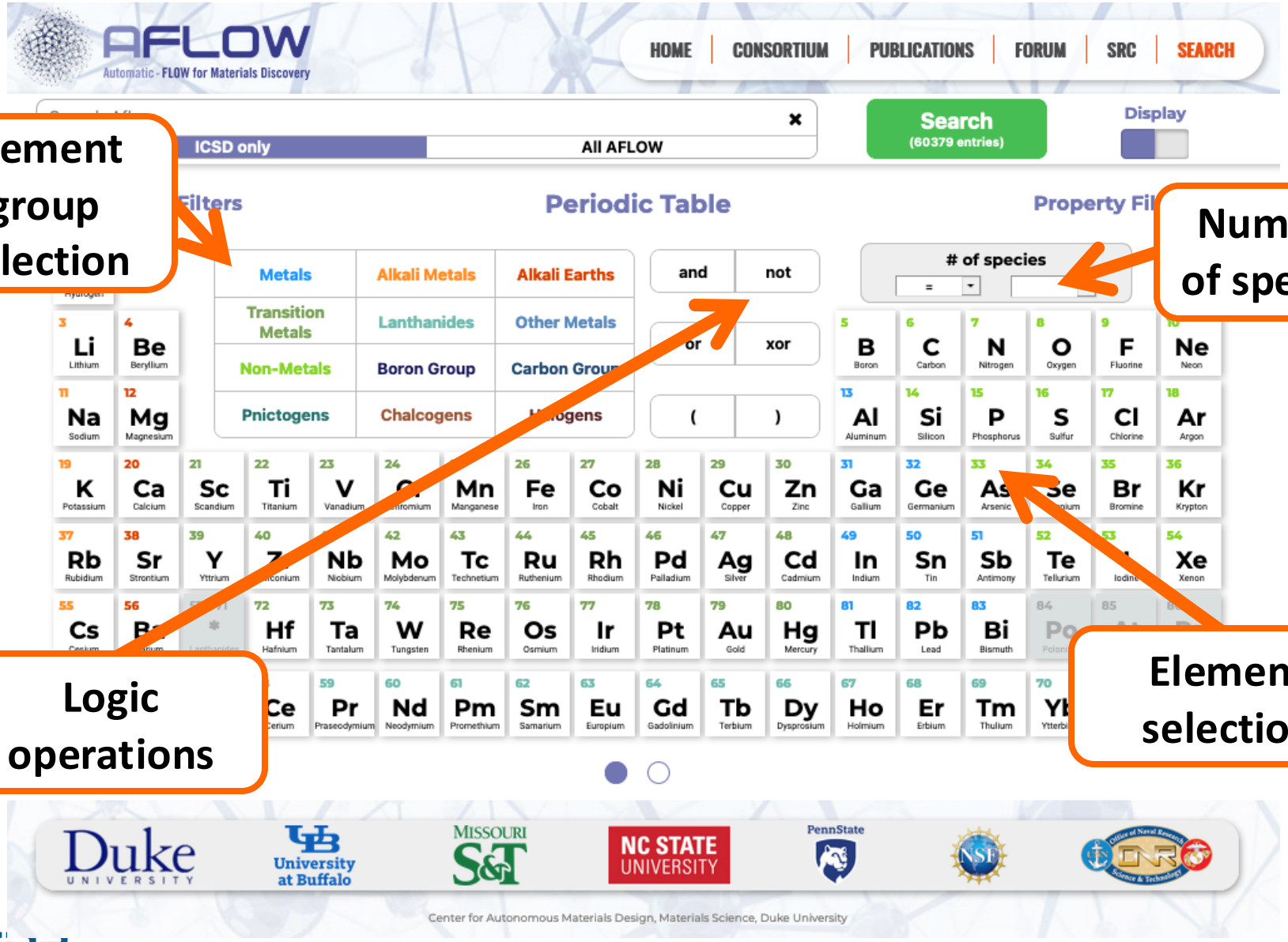

R. H. Taylor *et al.*, Comput. Mater. Sci. **93**, 178-192 (2014); M. Esters *et al.*, Comput. Mater. Sci. **216**, 111808 (2023)

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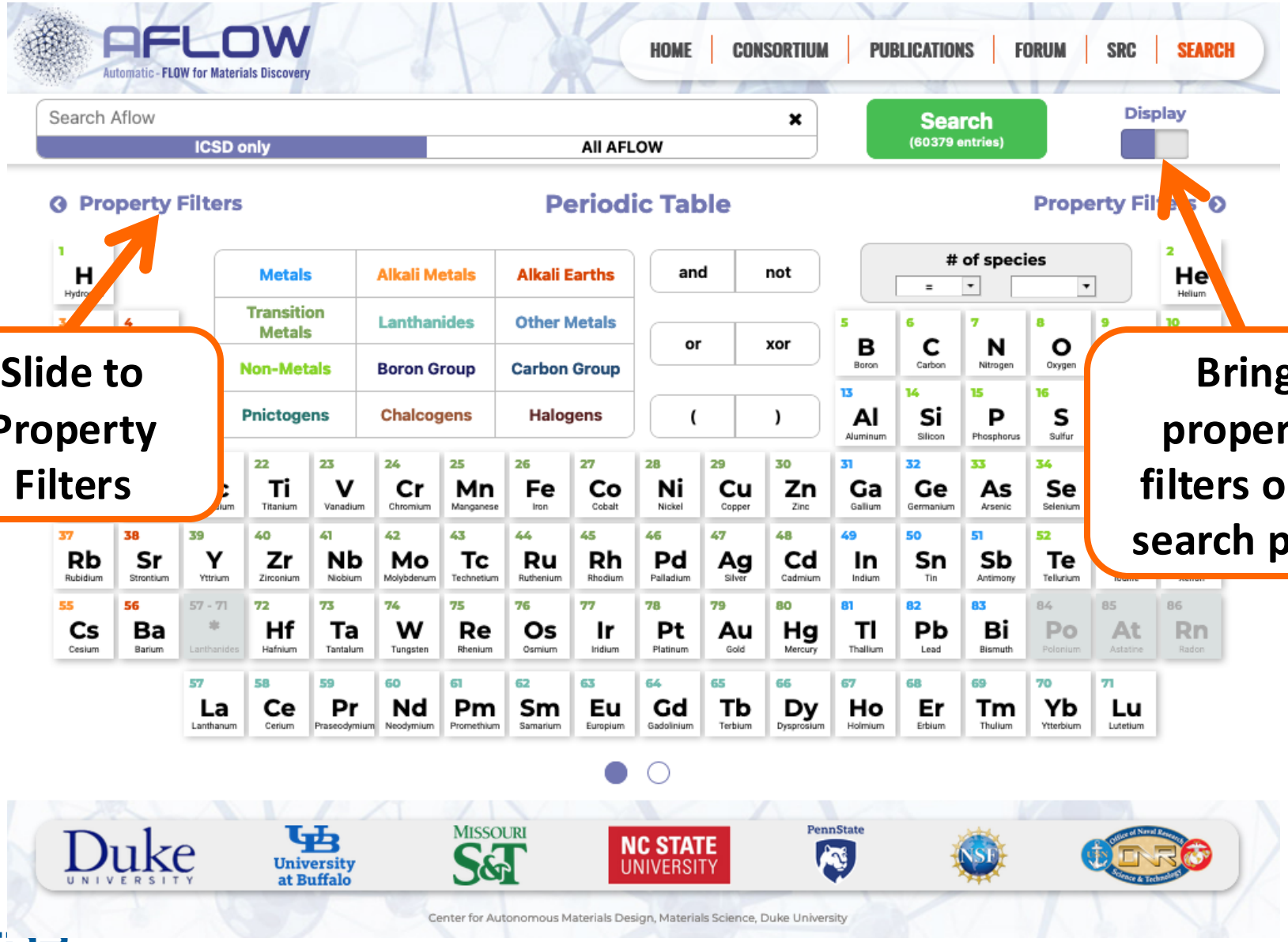

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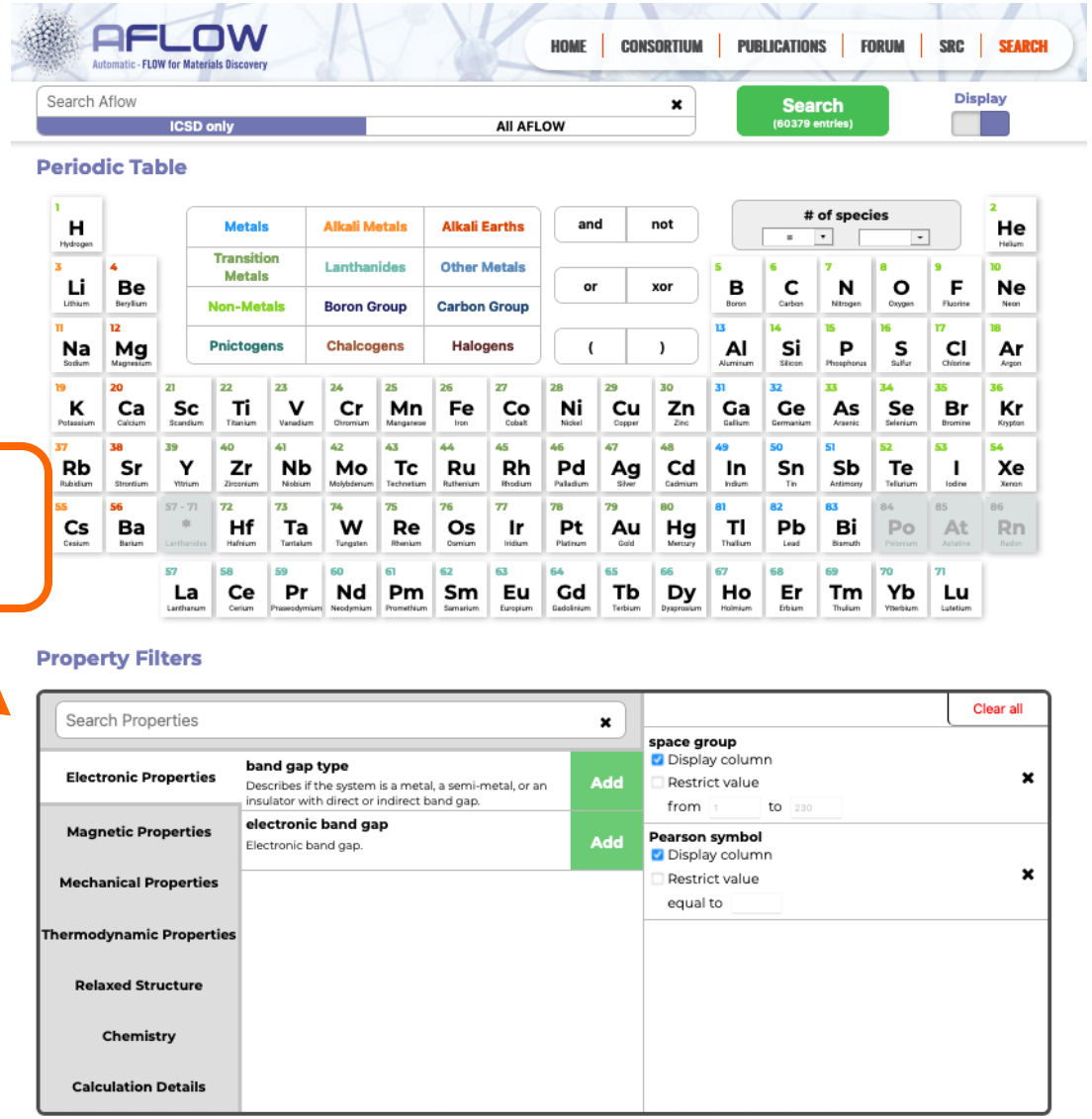

Property Filters

R. H. Taylor *et al.*, Comput. Mater. Sci. **93**, 178-192 (2014); M. Esters *et al.*, Comput. Mater. Sci. **216**, 111808 (2023)

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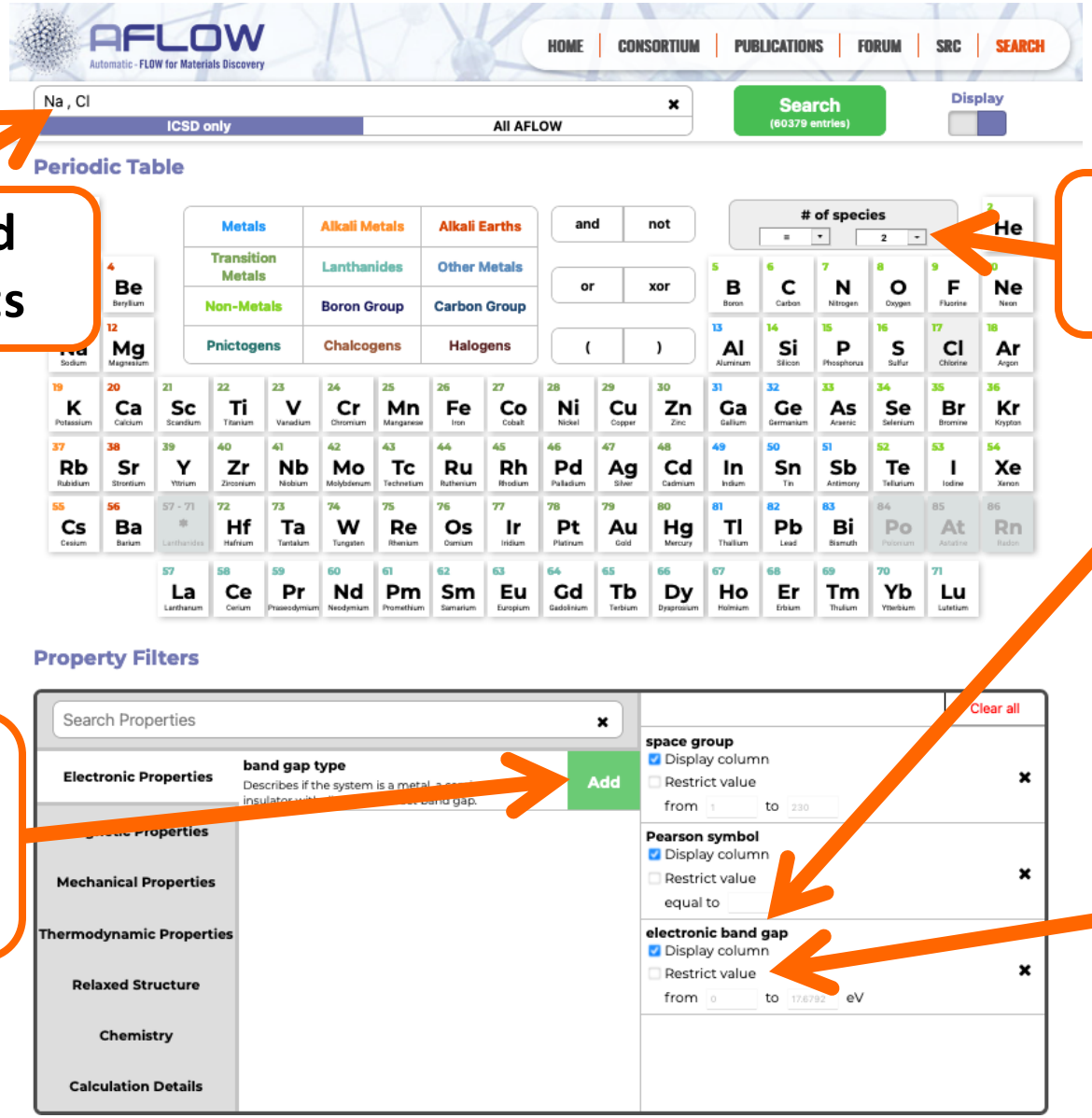

Selected Elements

Number of species=2

Added filter for Egap: electronic band bap

Select "Add" to add filter

Check box to restrict search results to specific value range

R. H. Taylor *et al.*, Comput. Mater. Sci. **93**, 178-192 (2014); M. Esters *et al.*, Comput. Mater. Sci. **216**, 111808 (2023)

AFLOW.org Search Page

Live demo examples:

1. Let us use the advanced search functionality to find the band gap for SiC in the zincblende structure (space group number 216).

2. Let us use the advanced search functionality to find the materials containing Sn but not Pb with band gaps between 1 eV and 3 eV in the ICSD catalog of the AFLOW database. How many results are returned?

Alternative server: aflowlib.duke.edu/search/ui/

R. H. Taylor *et al.*, Comput. Mater. Sci. **93**, 178-192 (2014), F. Rose et al., Comput. Mater. Sci. **137**, 362 (2017) ; M. Esters *et al.*, Comput. Mater. Sci. **216**, 111808 (2023)

AFLOW.org Search Page

Live demo examples:

1. Let us use the advanced search functionality to find the band gap for SiC in the zincblende structure (space group number 216). → **1.37 eV**

2. Let us use the advanced search functionality to find the materials containing Sn but not Pb with band gaps between 1 eV and 3 eV in the ICSD catalog of the AFLOW database. How many results are returned? → **369 results**

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AFLOW.org Search Page

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Entry page for each item of the database

R. H. Taylor *et al.*, Comput. Mater. Sci. **93**, 178-192 (2014), F. Rose et al., Comput. Mater. Sci. **137**, 362 (2017) ; M. Esters *et al.*, Comput. Mater. Sci. **216**, 111808 (2023)

AFLOW Software & Apps at aflow.org

AFLOW

Automatic - FLOW for Materials Discovery

S. Curtarolo *et al.*, Comput. Mater. Sci. **58**, 218 (2012)

- written in C++ utilizing VASP as density functional theory program
- AFLOW standard for:
  - *k*-point sets
  - plane wave cutoffs
  - PAW data sets ...

C. E. Calderon *et al.*, Comput. Mater. Sci. **108**, 233 (2015)

b-initio VASP Vienna Package Simulation

G. Kresse and J. Hafner, Phys. Rev. B **49**,14251 (1994)
G. Kresse and J. Furthmüller, Phys. Rev. B **54**,11169 (1996)
G. Kresse and J. Furthmüller, Comput. Mater. Sci. **6**,15 (1996)

MendeLIB search

AFLOW database search application

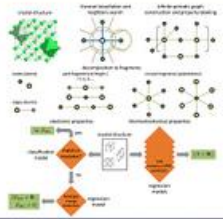
AFLOW-online

Online interface for AFLOW's symmetry, structure comparison, CCE, POCC, and other functionality

Prototype encyclopedia

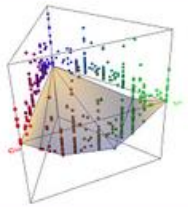
The AFLOW Prototype Encyclopedia with over 1,100 prototypes

AFLOW-ML



Machine Learning application for the PLMF, MDF, and ASC model

AFLOW-CHULL



Convex HULL application for thermodynamic stability and synthesizability

REST-API Docs

AFLOW REST-API WIKI

Documentation for the AFLOW REST-API

- the AFLOW software is structured into purpose dedicated, fully interoperable modules