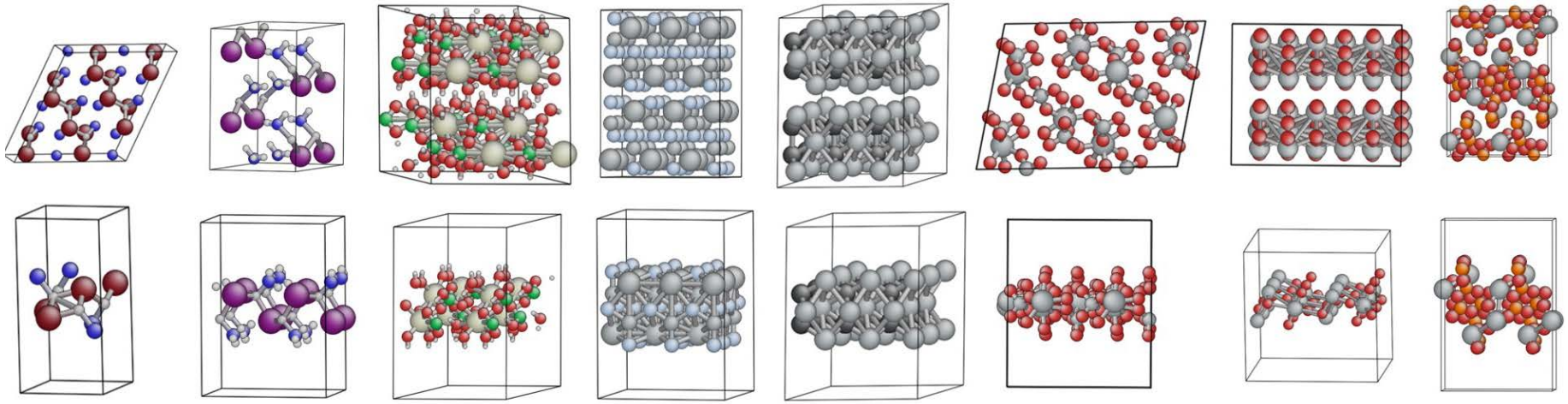
MARVEL
NATIONAL CENTRE OF COMPETENCE IN RESEARCH

MAX DRIVING THE EXASCALE TRANSITION

THEOS
THEORY AND SIMULATION OF MATERIALS

EPFL
ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

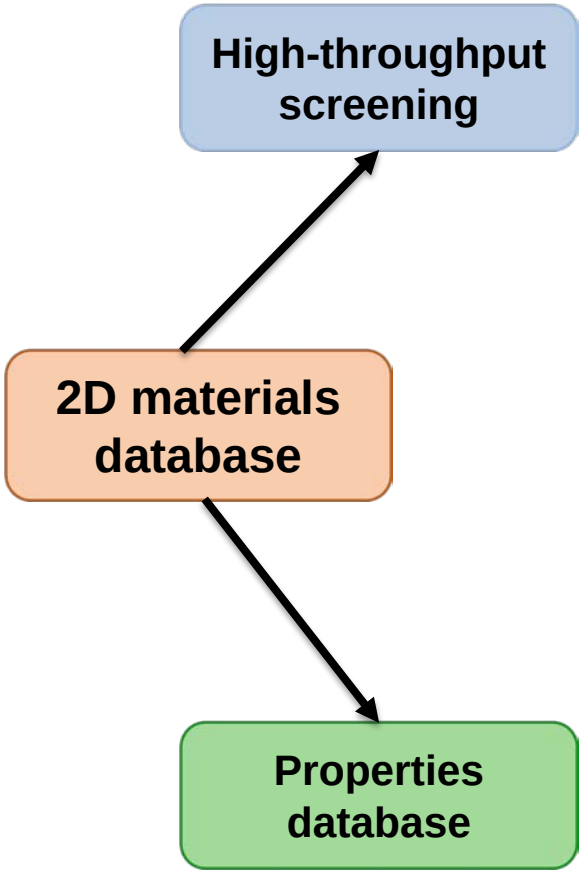
Two-dimensional materials from high-throughput computational exfoliation of experimentally known compounds



Nicolas Mounet, Marco Gibertini, Philippe Schwaller, Davide Campi, Andrius Merkys, Antimo Marrazzo, Thibault Sohier, Ivano Eligio Castelli, Andrea Cepellotti, Giovanni Pizzi, Nicola Marzari

Theory and Simulation of Materials (THEOS), and National Centre for Computational Design and Discovery of Novel Materials (MARVEL), EPFL

Motivation



- **High-mobility** materials
- Good **ionic conductors**
- **2D catalysts**
- **Topological insulators**
- **Piezoelectric /ferroelectric** materials
- **Superconductors**
- Materials for **spintronics**
- **Porous** membranes

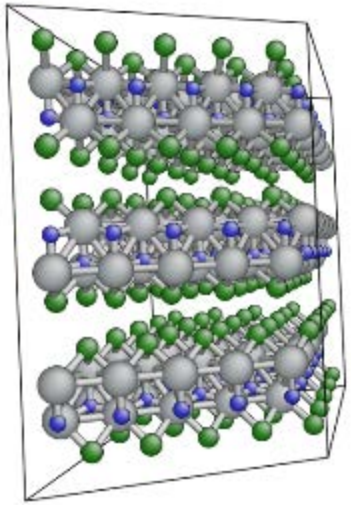
- **Thermomechanical** properties
- Mechanical / dynamical / chemical **stability**
- **Electronic** / **magnetic** properties

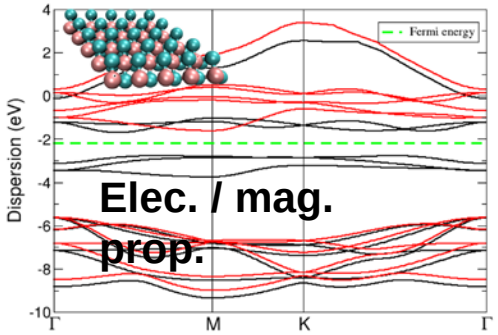
Aim: computational exfoliation of novel 2D materials from known 3D structures

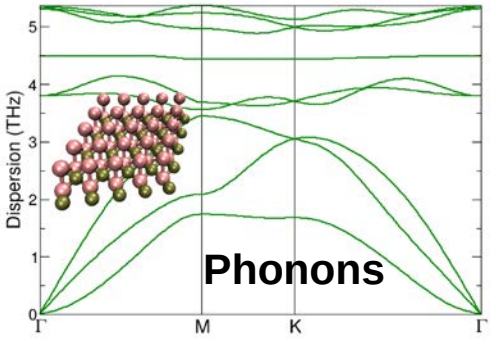
External databases (ICSD, COD)

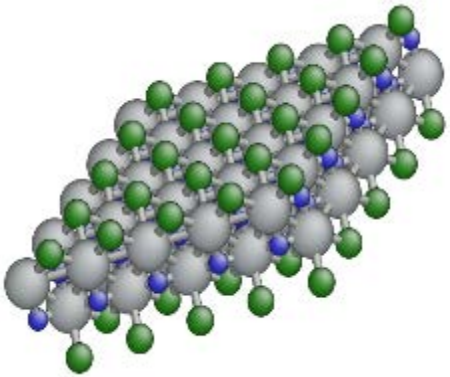
Screening

Layered structures



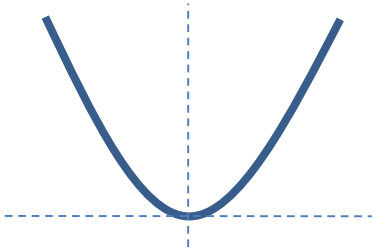


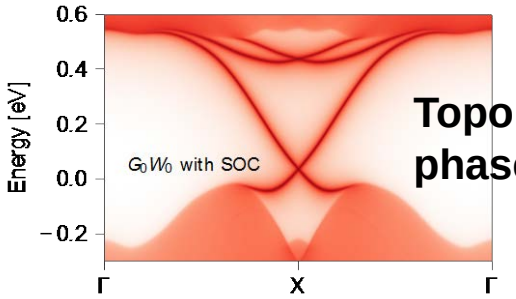




Relaxation

Binding energies





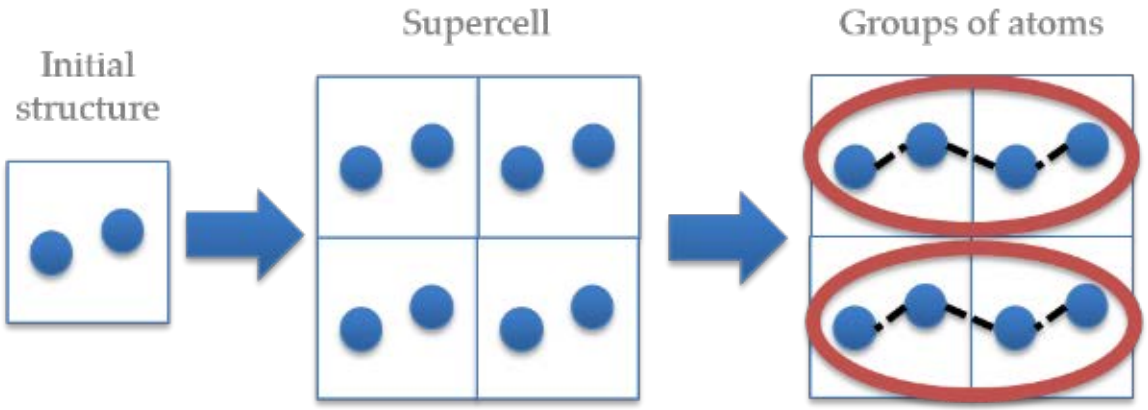
Topological phases

Filtering

2D database & properties

Similar study identifying **92** two-dimensional compounds, by ***S. Lebègue et al., PRX (2013)***

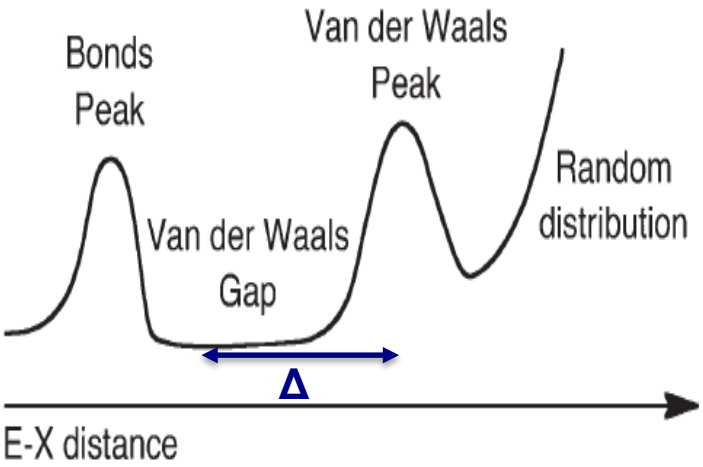
Low dimensionality screening



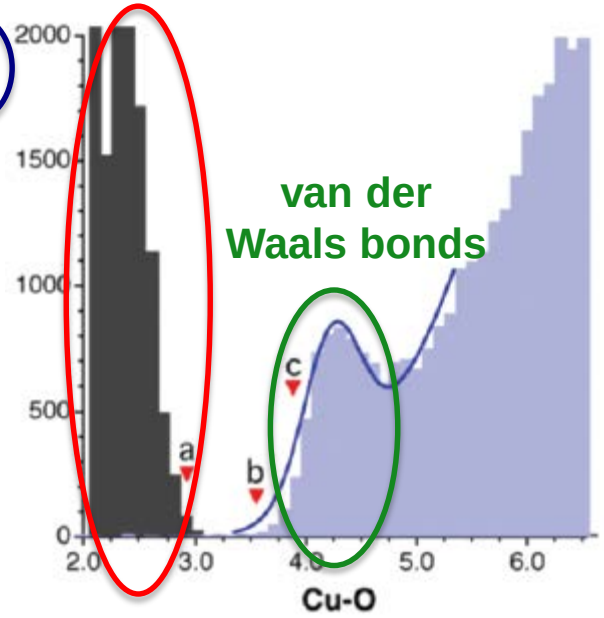
We group together chemically bonded atoms, defined as those separated by distance $d_{i,j}$ such that

$$d_{i,j} < r_i^{vdW} + r_j^{vdW} - \Delta$$

van der Waals radii of atoms *i*, *j*



S. Alvarez, Dalton Trans. 42, 8617–8636 (2013)

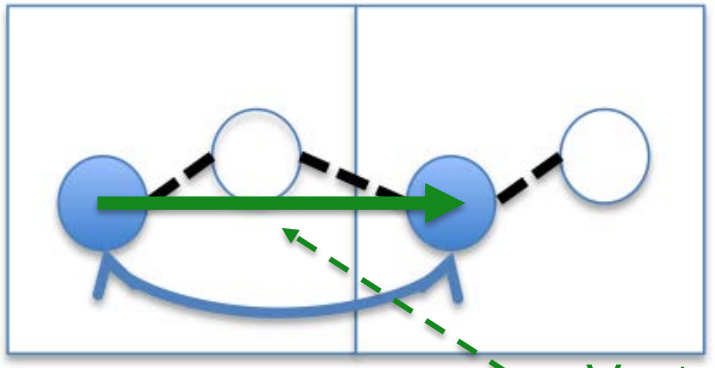


van der Waals bonds

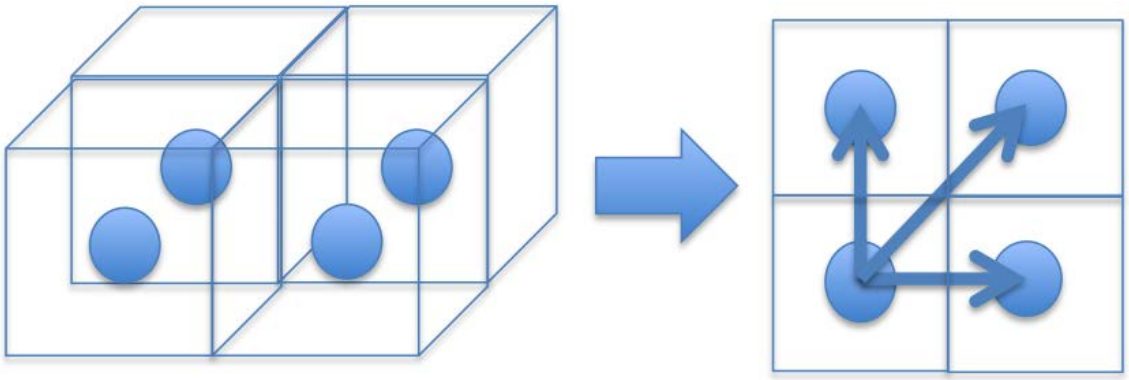
chemical bonds

2D, or not 2D?

Connected periodic copies of a given atom:

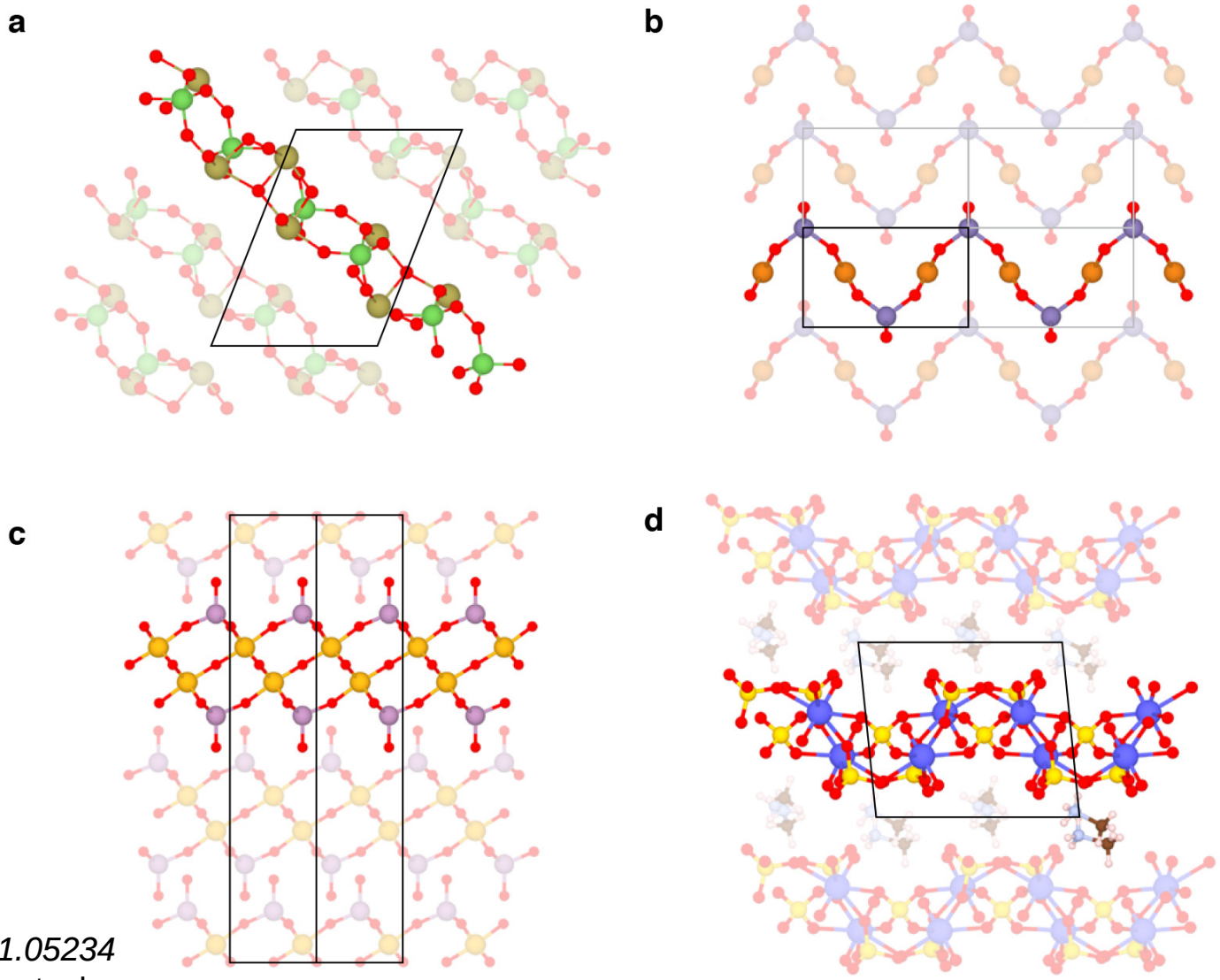


From the full supercell, get all the vectors connecting periodic copies:



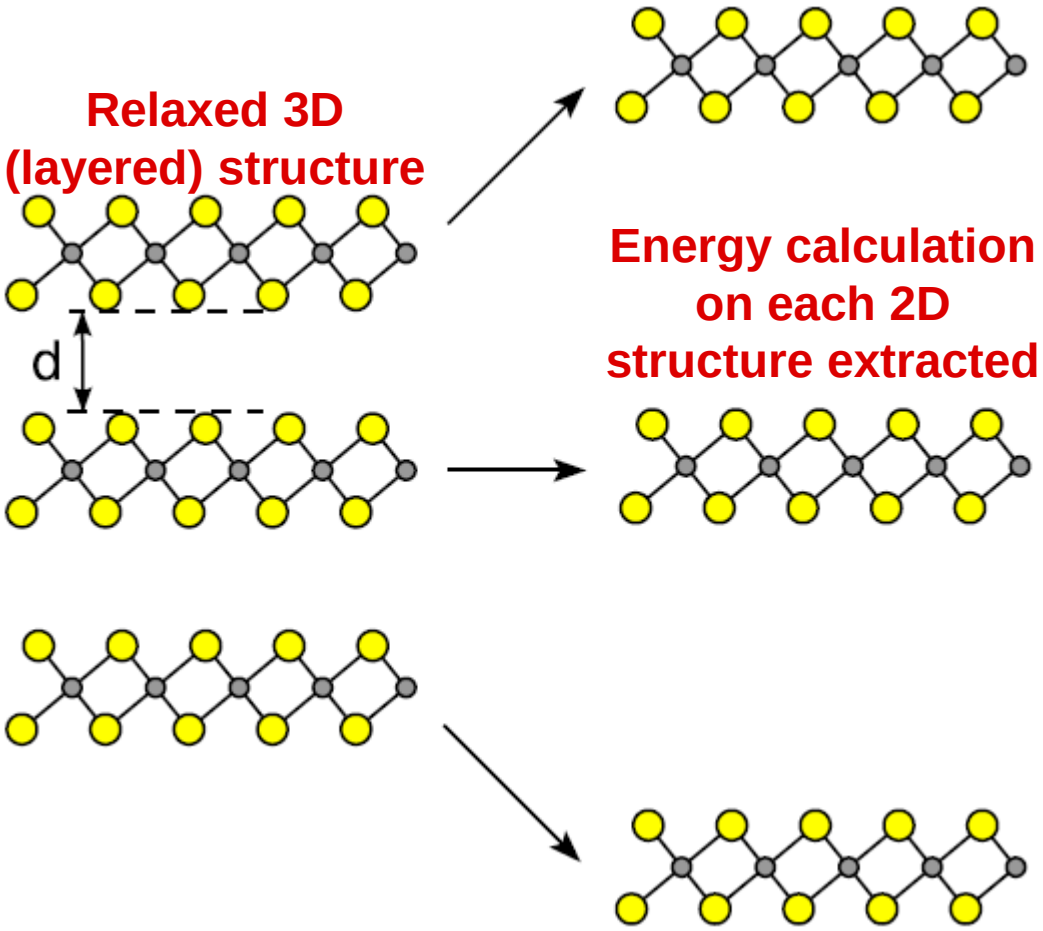
→ the rank of the ensemble of vectors found from periodic copies of the atom, gives the dimensionality of the group.

A few complex examples



NM et al, *arXiv:1611.05234* (2016), Nature Nanotech., in press (2018).

Binding energy computation



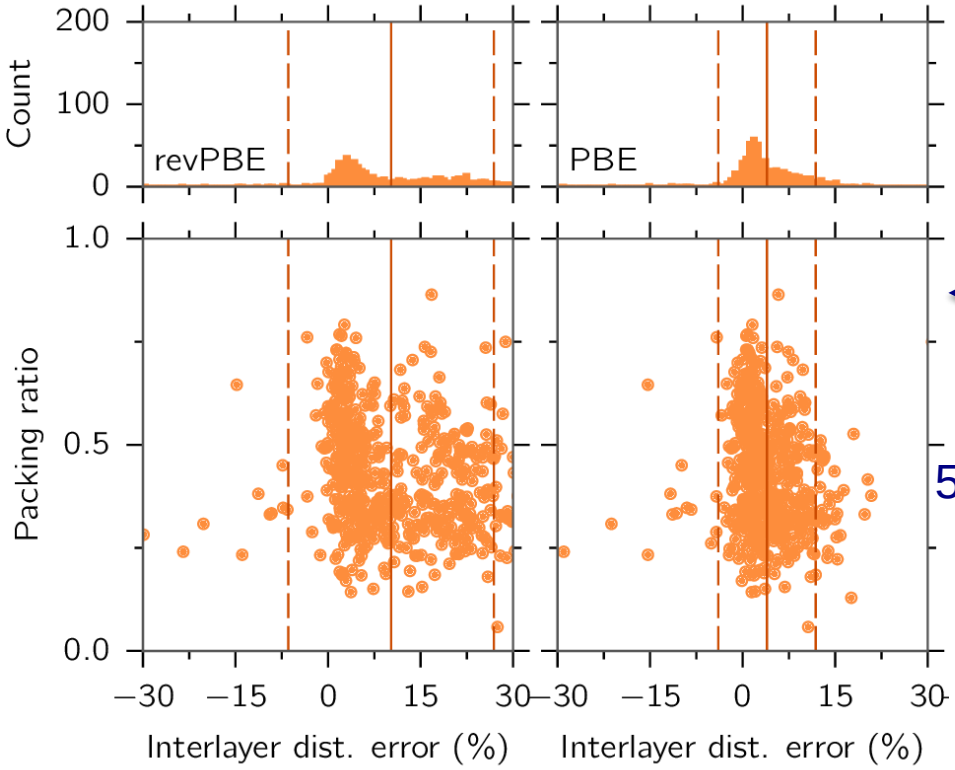
Pseudopotentials: **SSSP** library (*I. E. Castelli et al.*, *http://materialscloud.org*), **most accurate pseudo library** so far, w.r.t. all-electron calculations *http://molmod.ugent.be/deltacodesdft*.

Following up a study on **72** layered materials by *T. Björkman et al., PRL 108, 235502 (2012)*

Computations handled by **Quantum ESPRESSO** **using PBE and vdW functionals:**

- DF2 with C09 exchange - *K. Lee et al., PRB 82, 081101 (2010); V. R. Cooper, PRB 81, 161104 (2010*),
- rVV10 - *O. A. Vydrov and T. Van Voorhis, JCP 133, 244103 (2010); R. Sabatini et al., PRB 87, 041108 (2013*).

How reliable are the functionals?



Relative change in out-of-plane lattice parameter w.r.t. experimental structure

non-vdW functionals ←

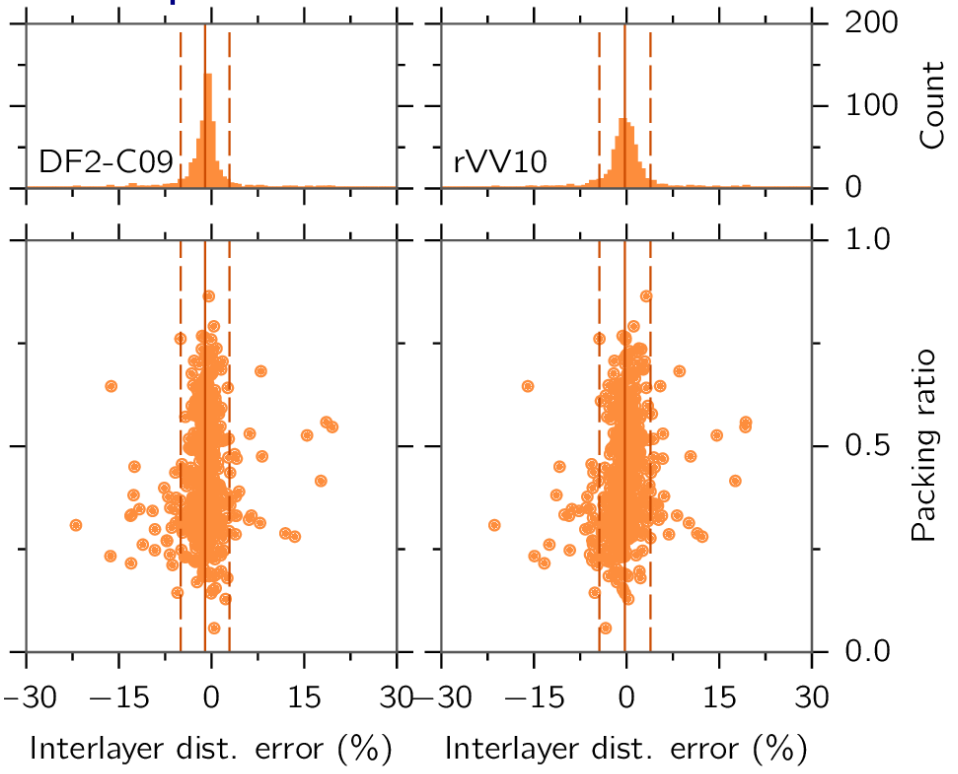
→ Large average error, large spread

573 samples

vdW functionals →

→ Small average error (-1% for DF2-C09, -0.3% for rVV10)

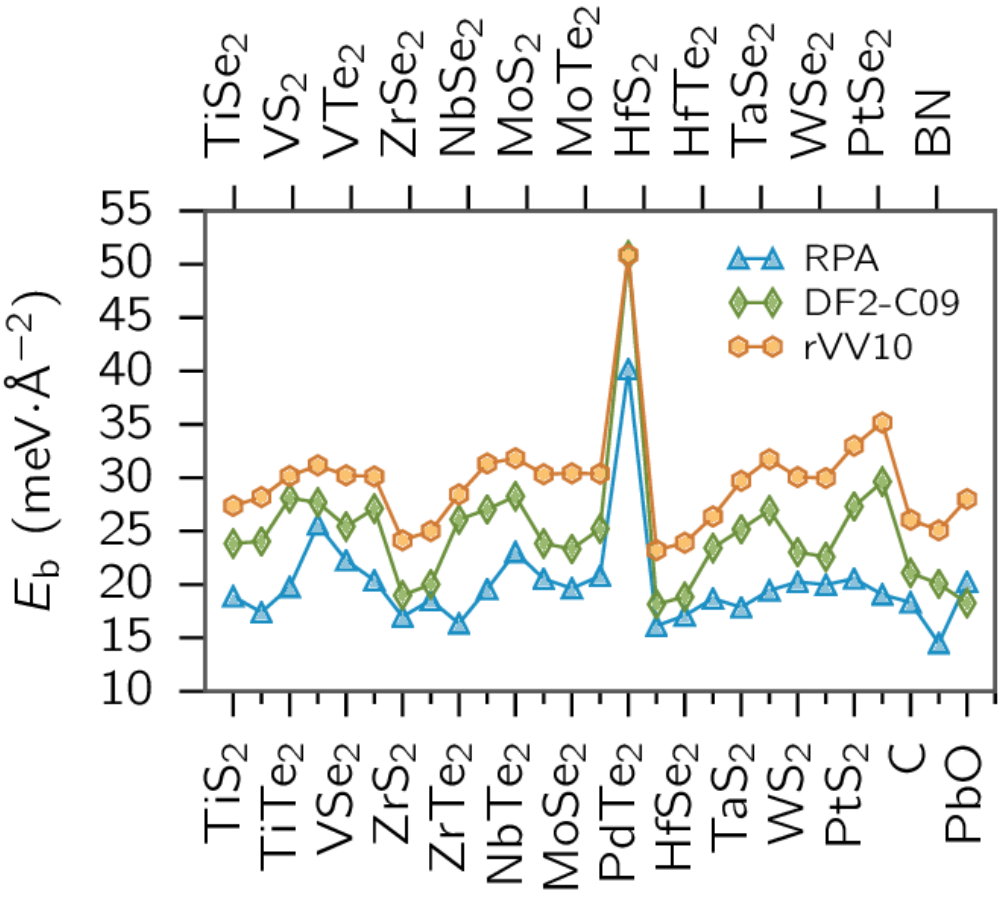
→ Small MAPE (1.5% for both)



NM et al, *arXiv:1611.05234* (2016), Nature Nanotech., in press (2018).

How reliable are the functionals?

Binding energies: RPA vs. DF2-C09 and rVV10



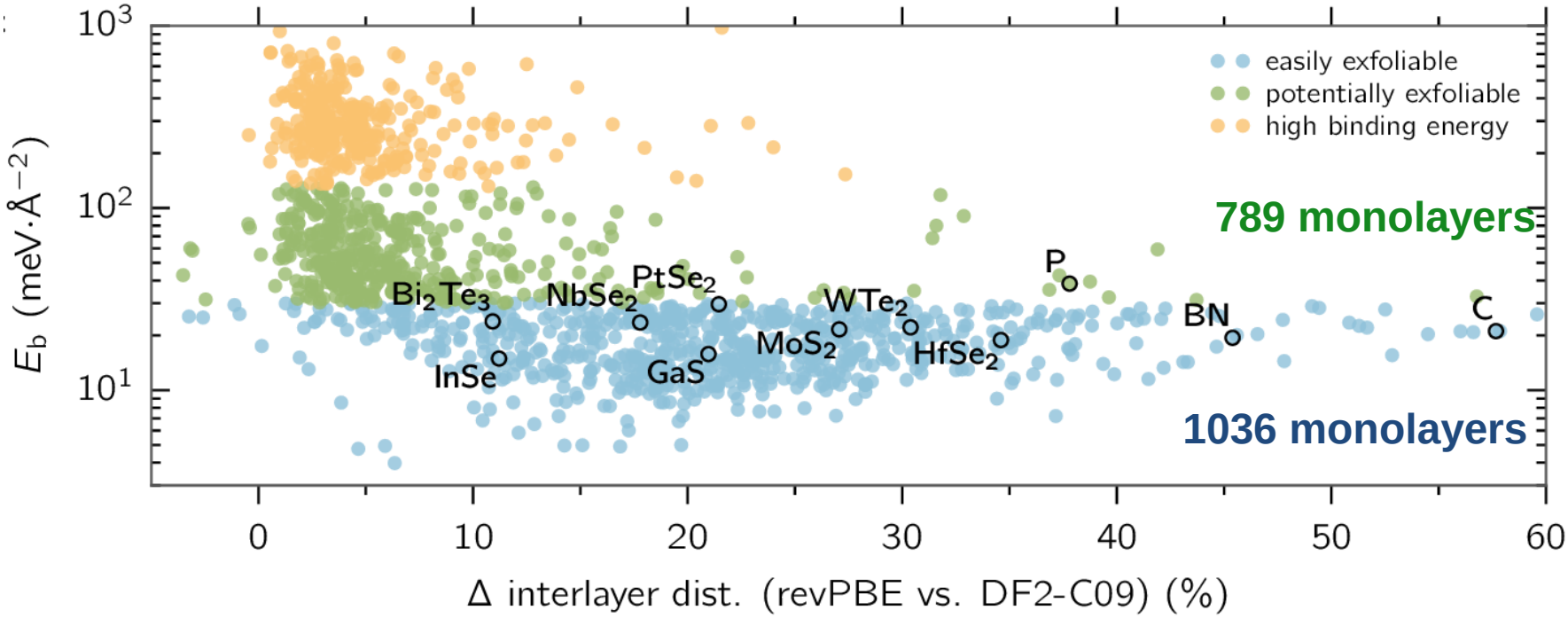
RPA calculations from *T. Björkman et al., PRL 108, 235502 (2012)*

→ Overall good agreement (in particular for the variation from compound to compound)

→ Both vdW functionals slightly overbind (rVV10 more than DF2-C09)

Refining the screening of layered materials

Binding energy E_b vs difference in interlayer distance when computed with / without vdW functionals:



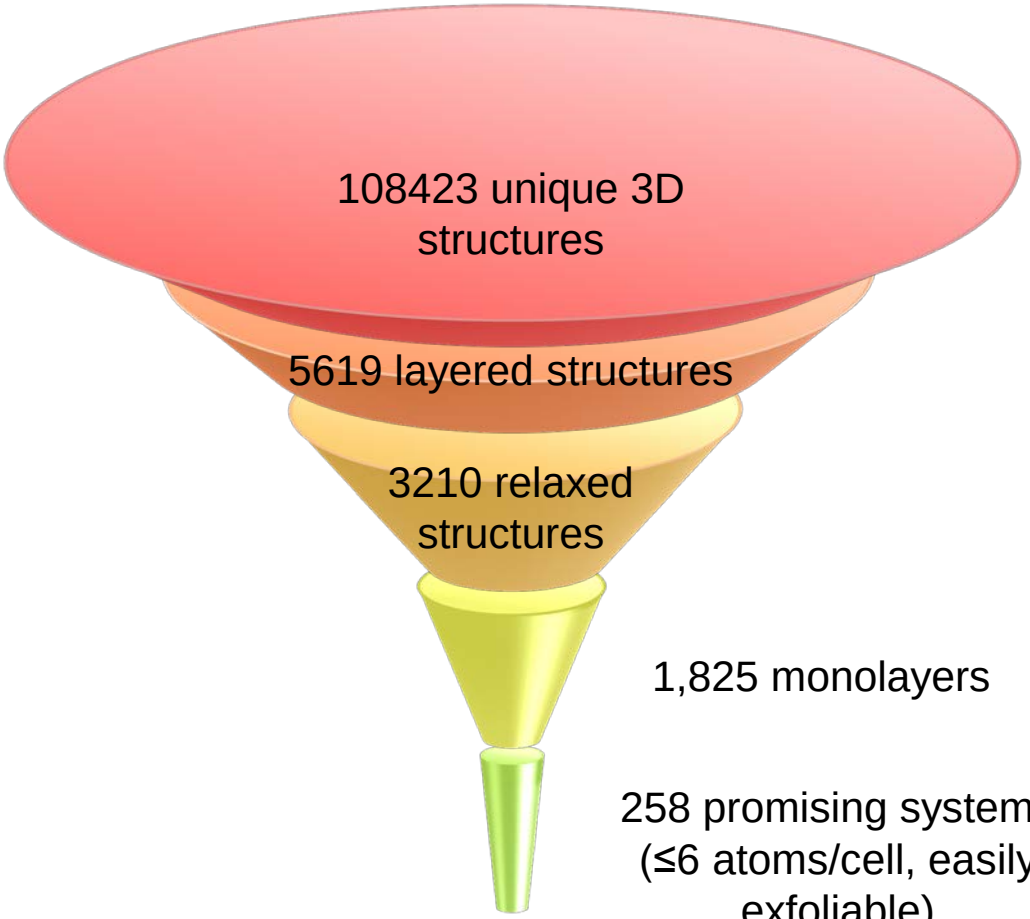
Three groups:

- $E_b < 30$ meV/Å^2 (DF2-C09) or $E_b < 35$ meV/Å^2 (rVV10) → 2D, easily exfoliable
- $E_b > 130$ meV/Å^2 → not 2D (discarded)
- In-between → 2D, potentially exfoliable

NM et al, *arXiv:1611.05234* (2016), Nature Nanotech., in press (2018).

Building the 2D database

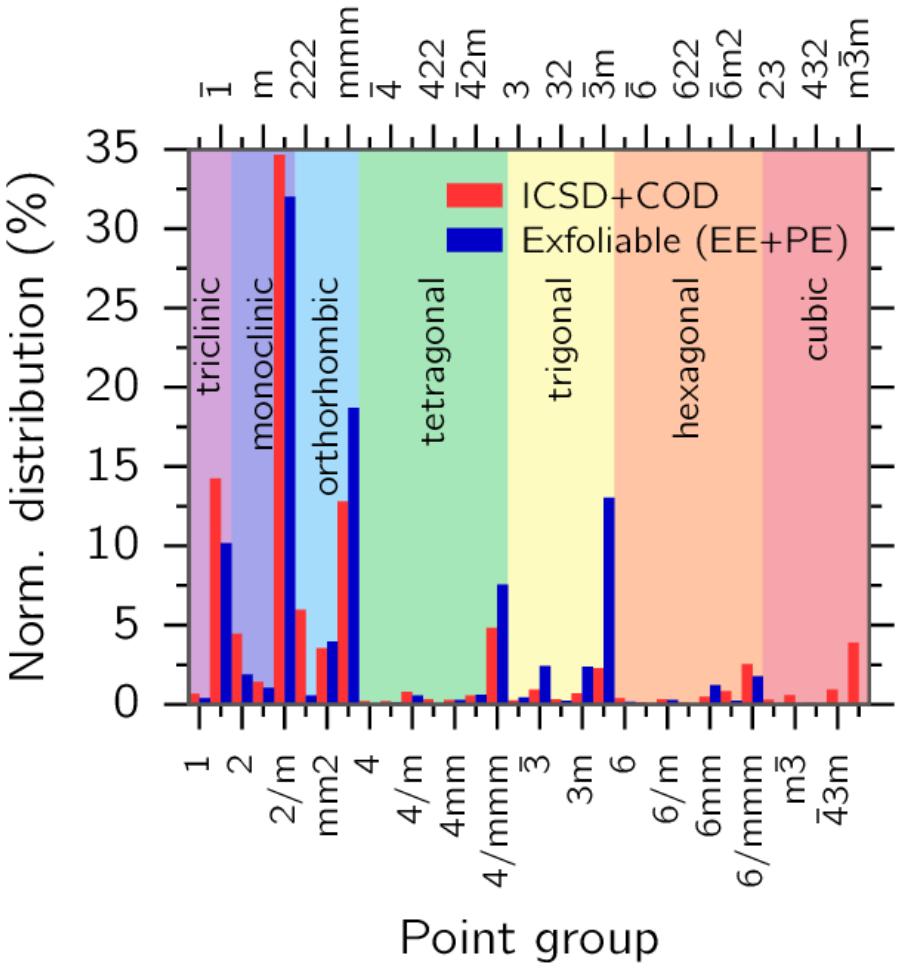
Starting from the ICSD (www.fiz-karlsruhe.com/icsd.html) and COD (www.crystallography.net) databases:

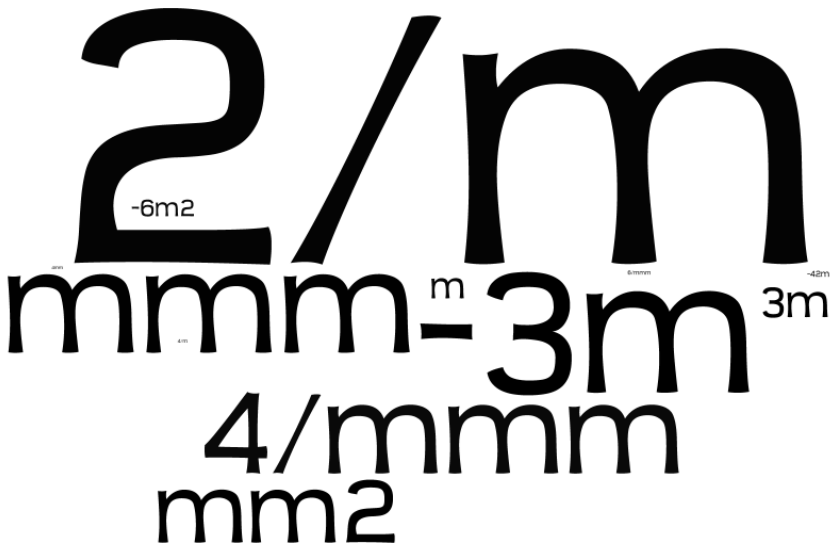


- Geometrical selection
- Structural relaxation
- Binding energy
- Exfoliation
- Identification of prototypes
- Structural relaxation
- Phonons & structure refinement
- Magnetic / electronic properties
- Topological phases

Layered materials statistics

- Distribution of point groups of layered materials, vs. ICSD+COD:



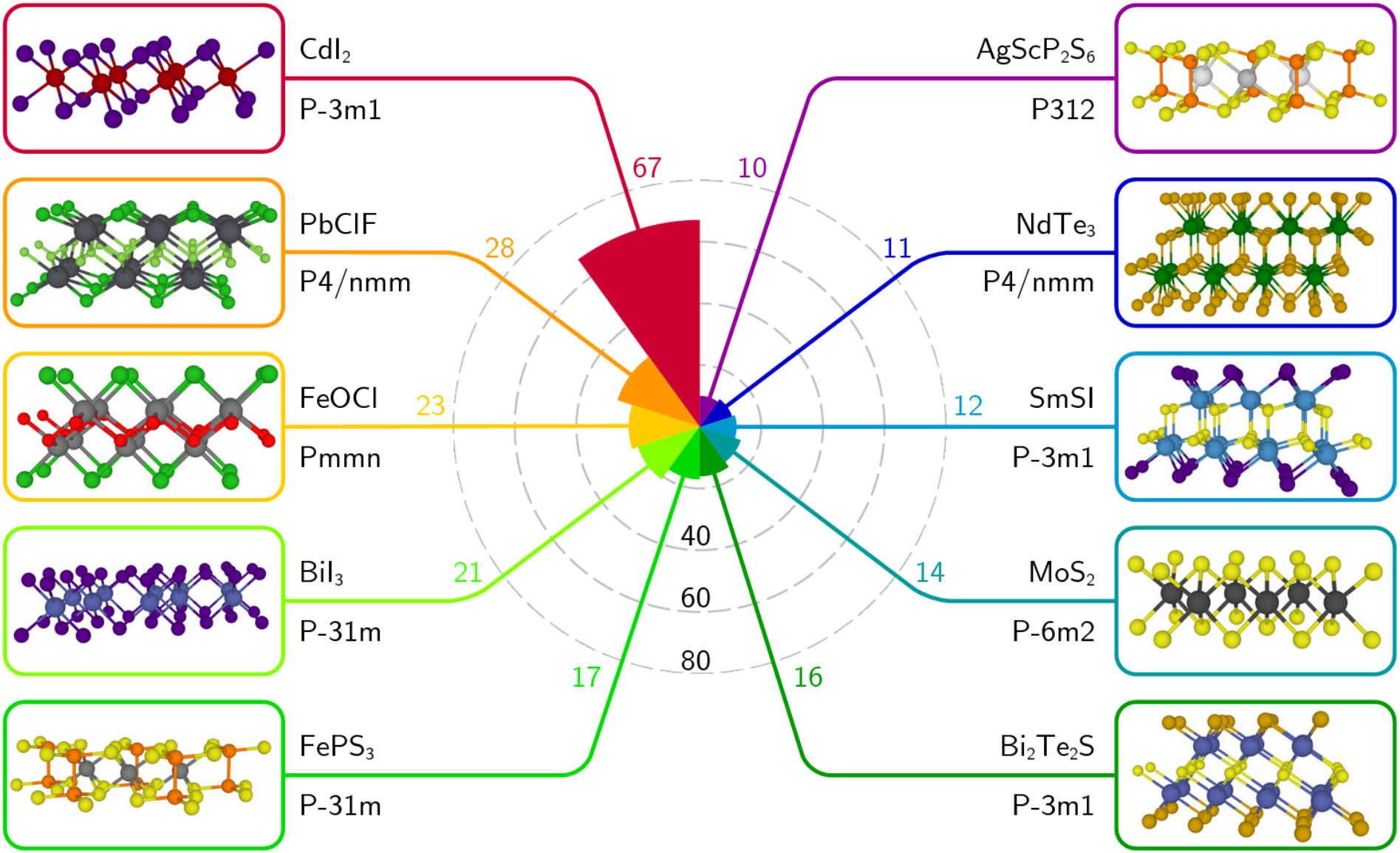


NM et al, *arXiv:1611.05234* (2016), Nature Nanotech., in press (2018).

- -3, 3m, -3m & 6mm point groups are more frequent in layered structures
- 222 is much less present; cubic groups obviously absent from layered materials

2D structural prototypes

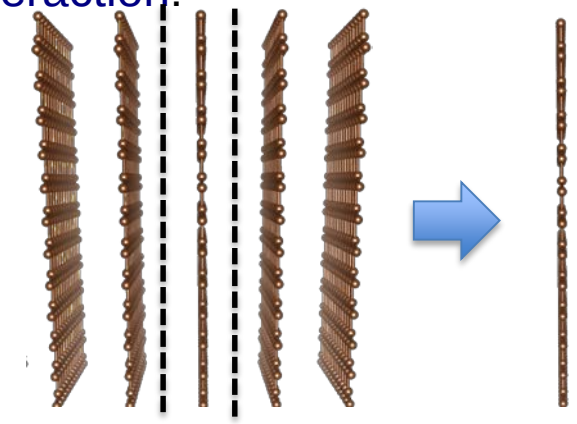
Most common prototypes:



NM et al, *arXiv:1611.05234* (2016), Nature Nanotech., in press (2018).

Are these structures stable?

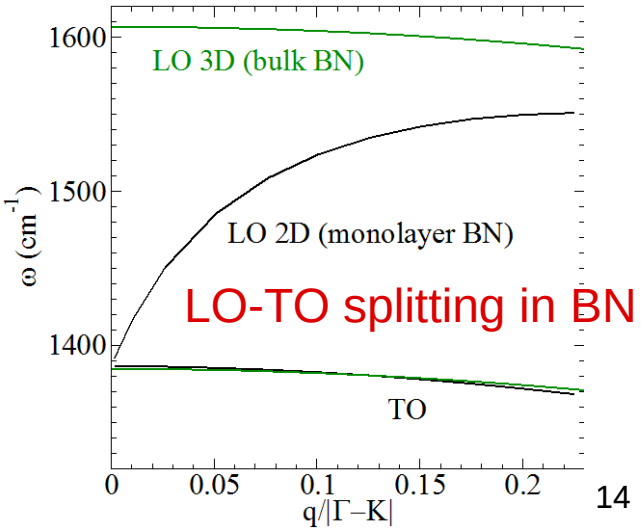
- We assess mechanical stability by computing phonons, using Density-Functional Perturbation Theory (DFPT) as implemented in the Quantum ESPRESSO code.
- For 2D monolayers, 3D periodic boundary conditions may not work well: long-wavelength perturbations induce long-ranged Coulomb interactions → periodic images interact.
- We use a 2D version of the DFT and DFPT code, with a truncated Coulomb interaction:



T. Sohier, M. Calandra, F. Mauri, Phys. Rev. B 96, 075448 (2017)

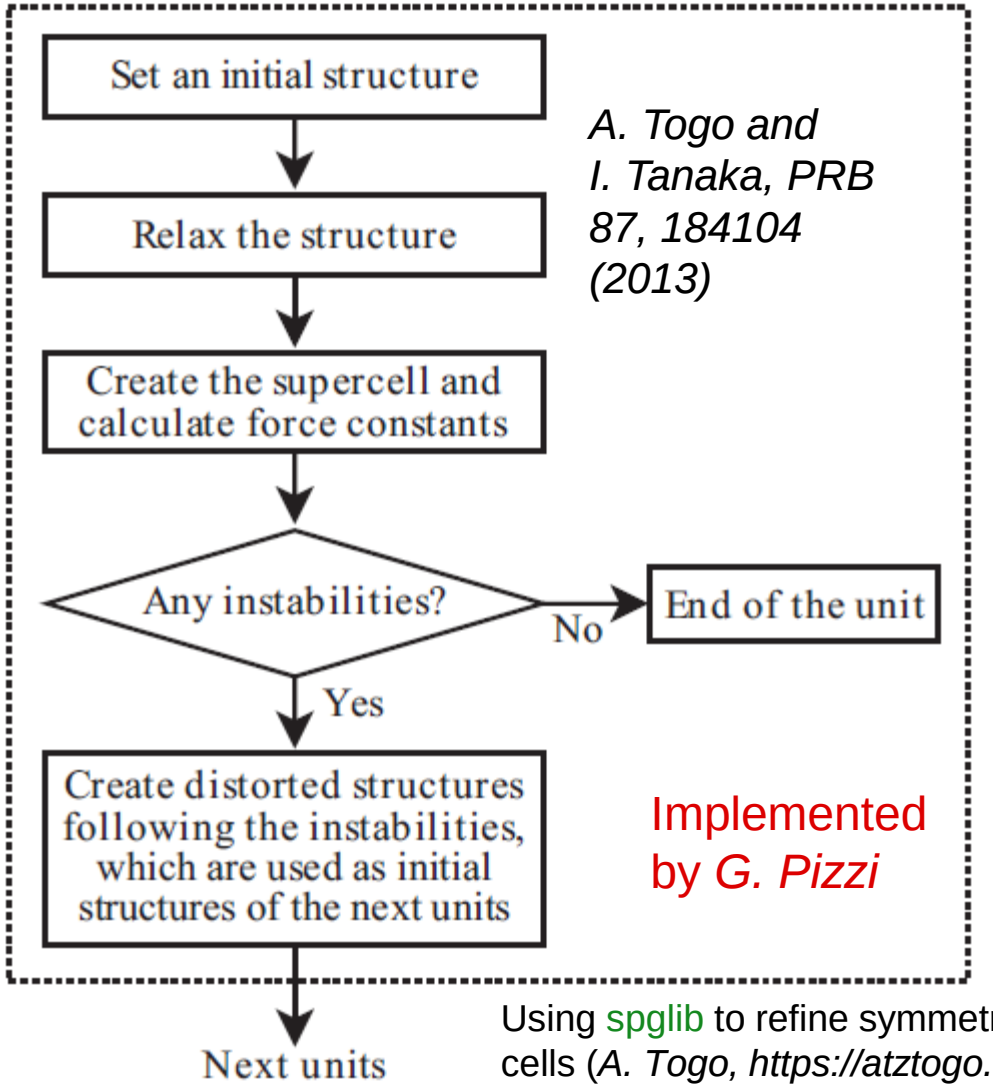
- This allows to compute properly the LO-TO splitting in 2D insulators:

*Sohier, Gibertini, Calandra, Mauri, Marzari, Nano Lett., **2017**, 17 (6), pp 3758–3763*



Dealing with unstable structures

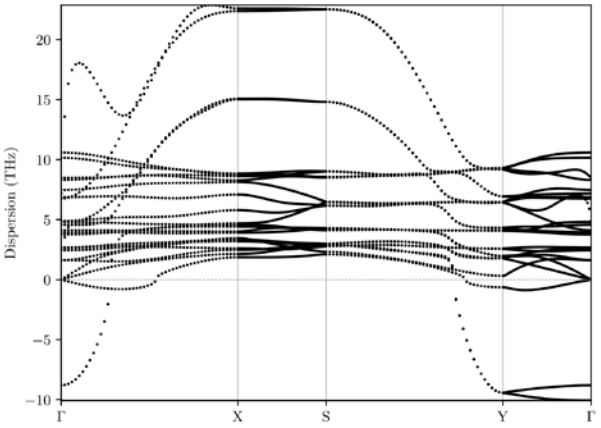
Computing phonons at Γ, we can check the unstable ones and "follow" them to get a stabilized structure:



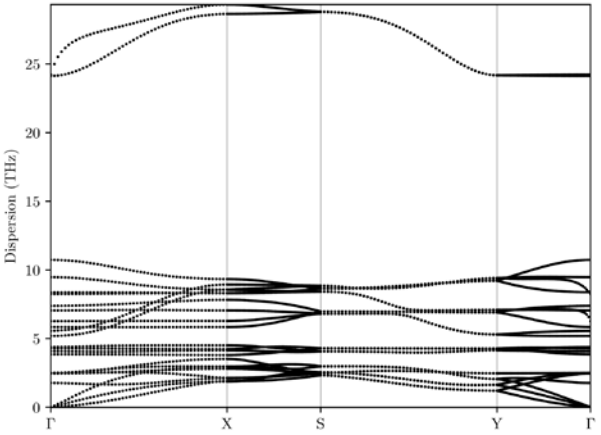
A. Togo and I. Tanaka, PRB 87, 184104 (2013)

Implemented by *G. Pizzi*

Example of initial phonon dispersion:



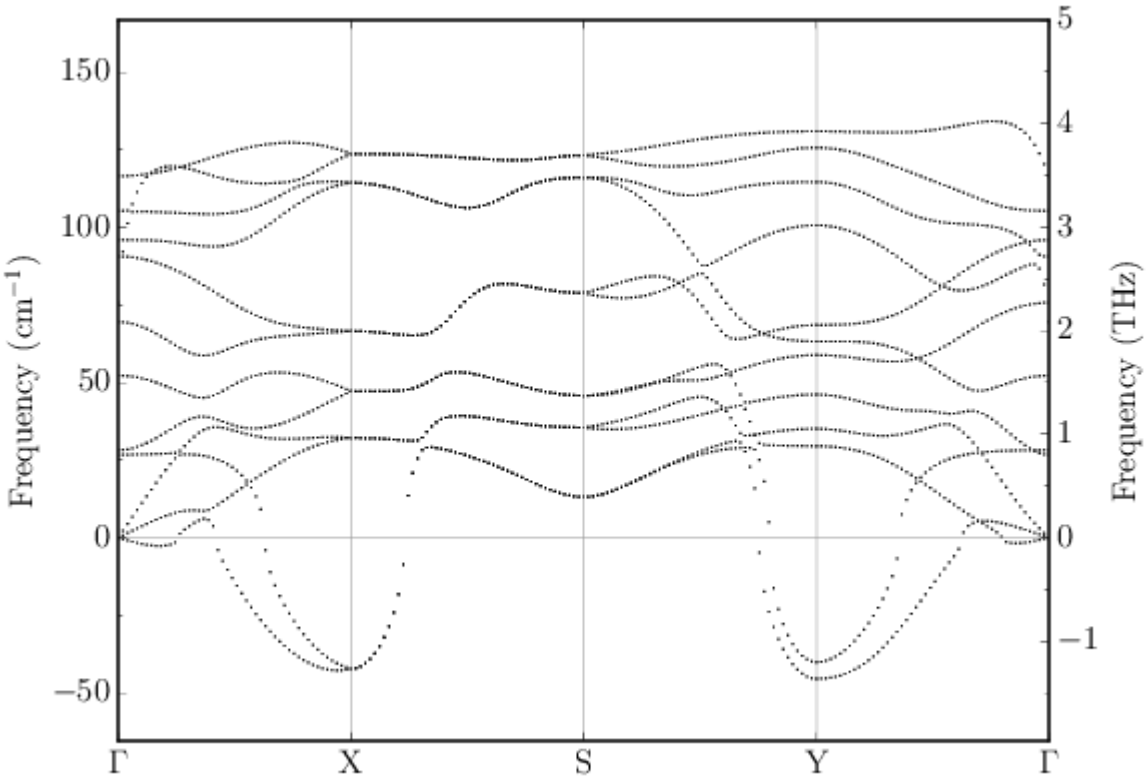
After stabilization:



Using spglib to refine symmetries and find primitive cells (*A. Togo, https://atztogo.github.io/spglib*)

Phonon dispersions

Vibrational properties of 245 monolayers:



NM et al, *arXiv:1611.05234* (2016), Nature Nanotech., in press (2018).

Automation	Data	Environment	Sharing
Automation	Database	Research environment	Social
Remote management	Provenance	Scientific workflows	Sharing
High-throughput	Storage	Data analytics	Standards

AiiDA

http://www.aiida.net
(MIT BSD, jointly developed with Robert Bosch)
G. Pizzi et al., Comp. Mat. Sci. 111, 218 (2016)

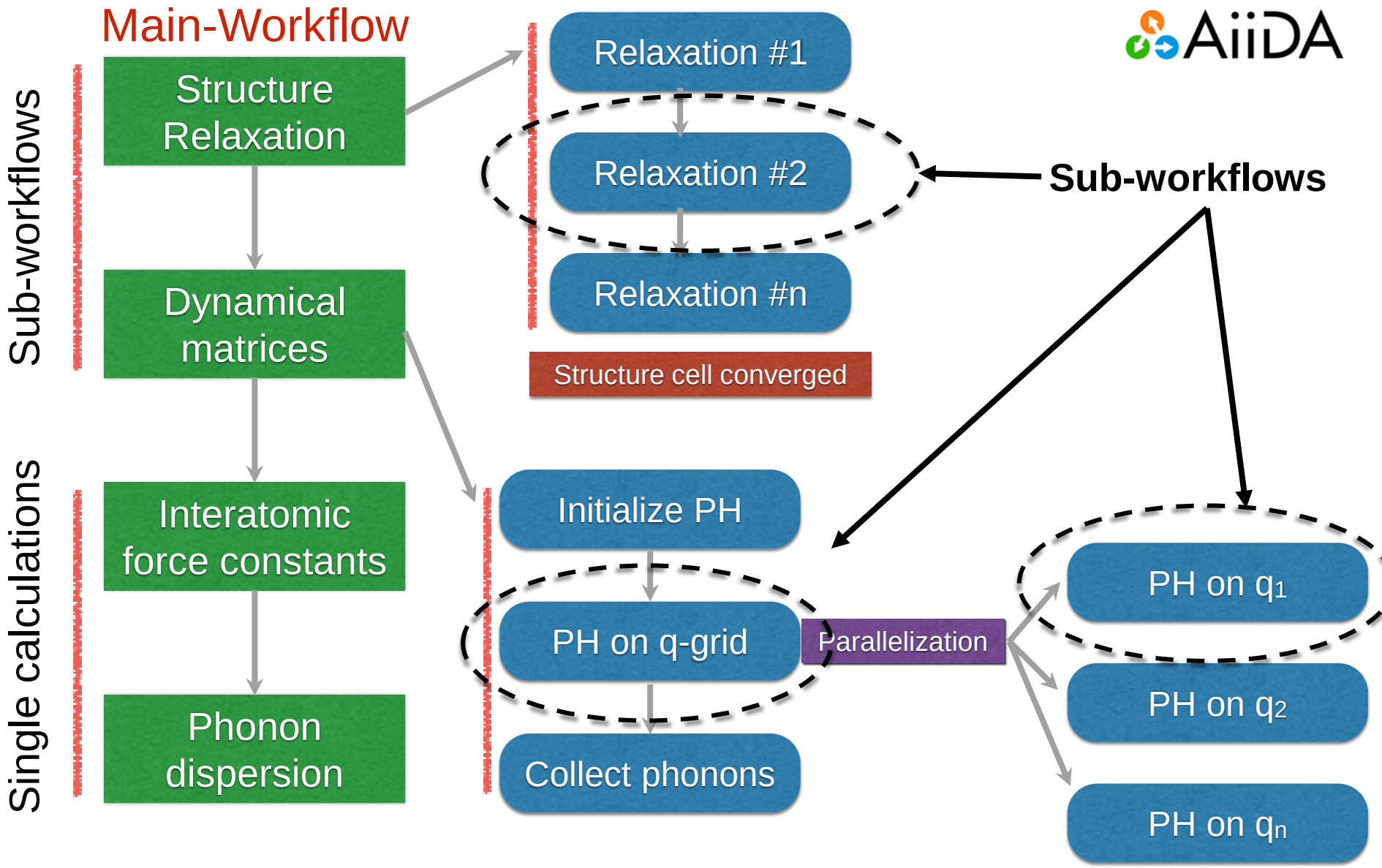
A factory

A library

A scholar

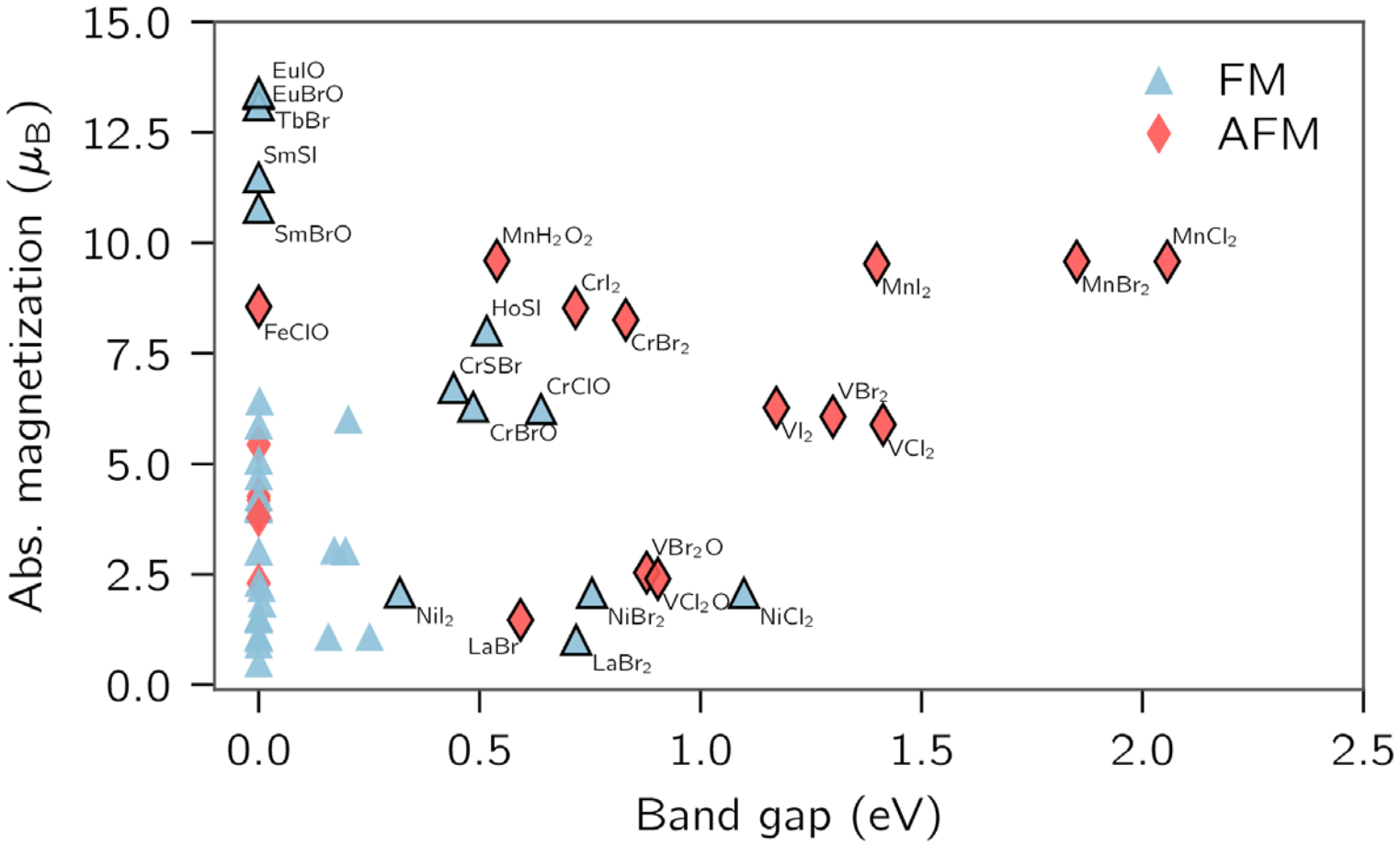
A community

An AiiDA workflow: phonon dispersions



Magnetic / electronic properties

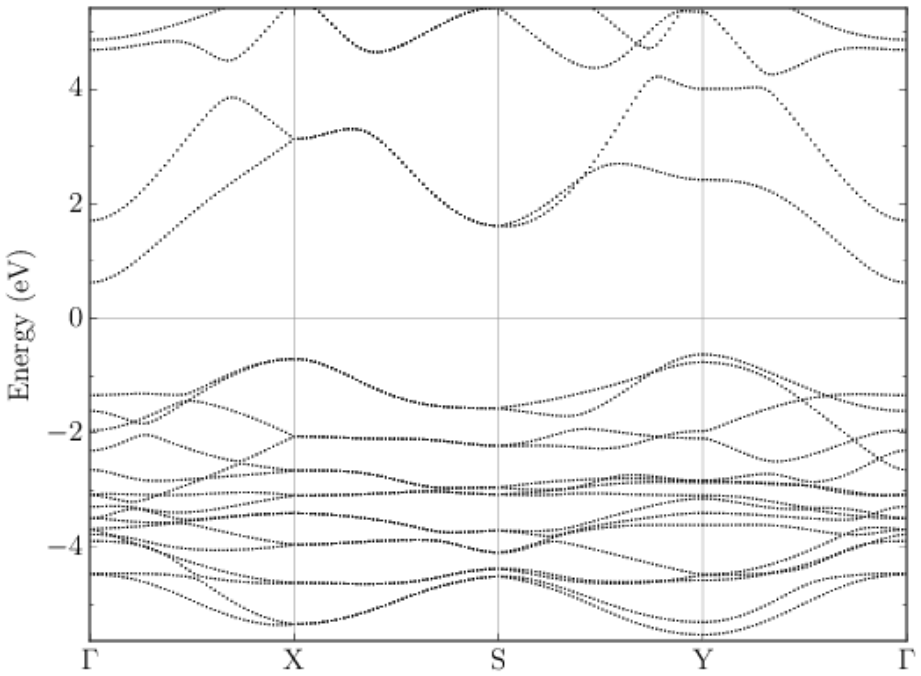
- Magnetic ground state found after exploration of possible ferro- and antiferro-magnetic configurations (DFT-PBE level), using supercells.
- Mapping band-gaps and magnetic properties for the **258** most promising monolayers:

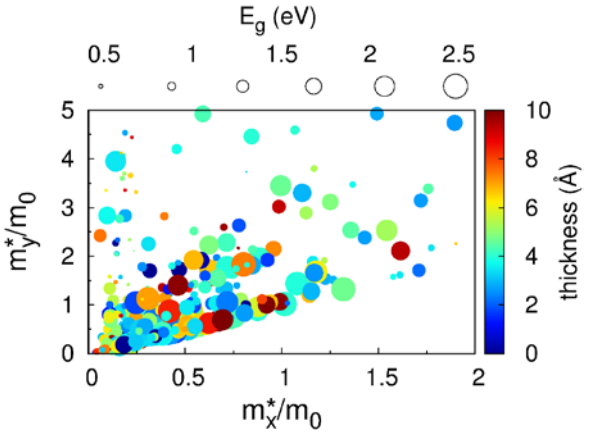


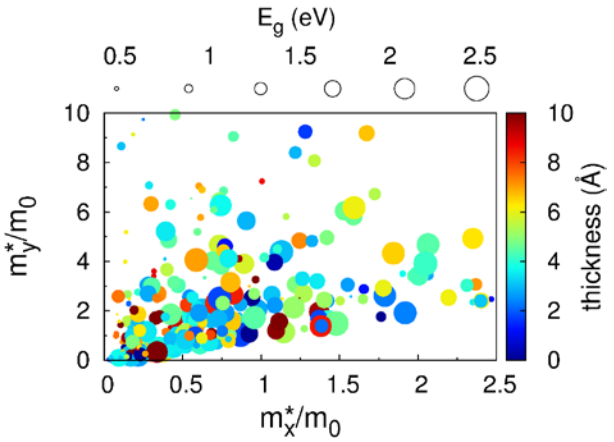
NM et al, *arXiv:1611.05234* (2016), Nature Nanotech., in press (2018).

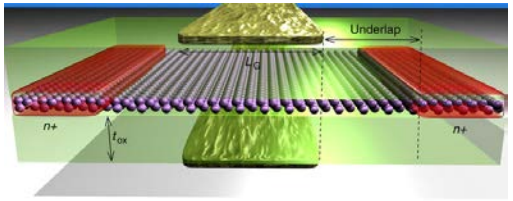
Optimal 2D materials for electronic applications

- Computing electronic band structures → band gap & effective masses (at the DFT-PBE level)







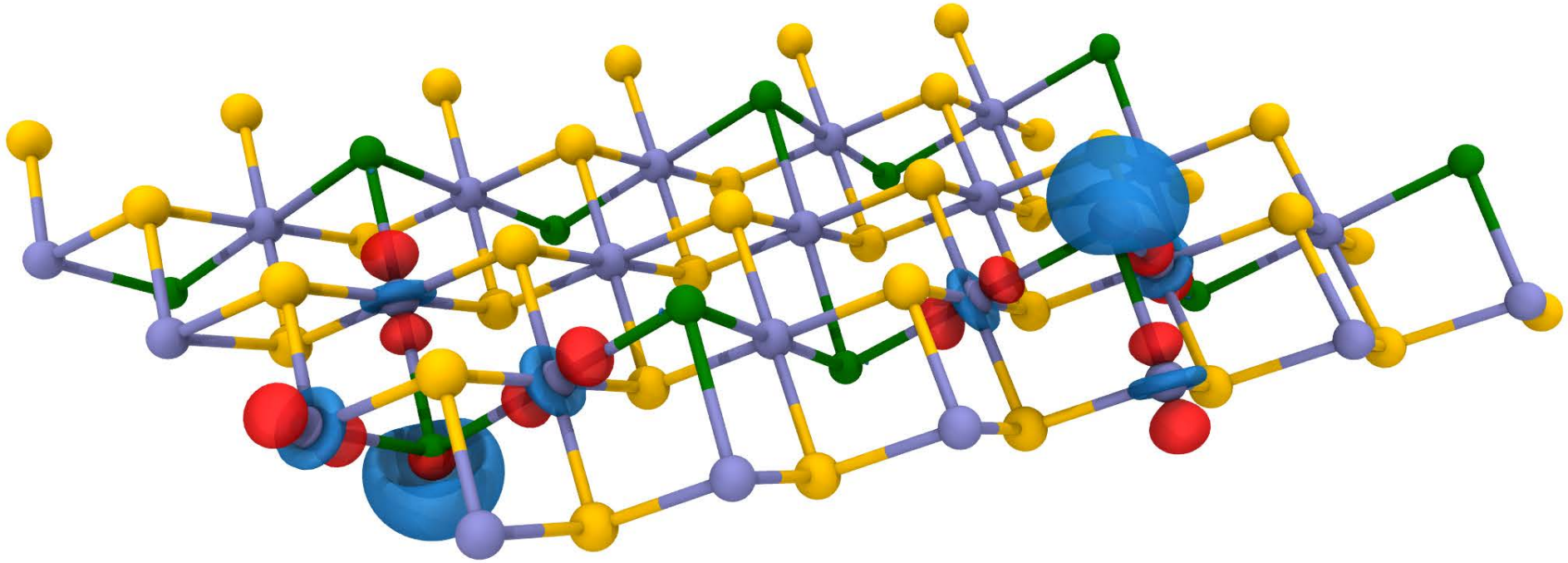


D. Campi, in preparation

Search for topological insulators

- Novel 2D topological insulators candidates found, the optimal being Jacutingaite (Pt_2HgSe_3 - 3D bulk form discovered in 2008, in Brazil)

→ see Antimo Marrazzo's poster, "*Prediction of a large-gap and switchable Kane-Mele quantum spin Hall insulator*"



A. Marrazzo, M. Gibertini, D. Campi, NM, N. Marzari, arXiv:1712.03873 (2017)

2D THANKS

Marco Gibertini

Philippe Schwaller

Davide Campi

Andrius Merkys

Antimo Marrazzo

Thibault Sohier

Ivano E. Castelli

Andrea Cepellotti

Giovanni Pizzi

Nicola Marzari

MARVEL

NATIONAL CENTRE OF COMPETENCE IN RESEARCH

CSCS

Centro Svizzero di Calcolo Scientifico
Swiss National Supercomputing Centre

MAX DRIVING THE EXASCALE TRANSITION

Summary

- Around 5600 layered materials were extracted from close to 480000 non unique, classified 3D structures.
- Among them, at least 1800 structures exhibit weak interlayer bonding and 1000 of them are very good candidates for easy exfoliation.
- 2600 binding energies were computed, all within the AiiDA platform (*G. Pizzi et al., Comp. Mat. Sci. 111, 218 - 2016)* which allows sharing, reproducibility, automatization, and efficient querying.
- Phonons / magnetic / electronic / topological properties were computed for 258 of them (easily exfoliable, small unit cell).
- All computed data with its provenance is available on Materials Cloud (https://beta.materialscloud.org), as well as under the doi https://doi.org/10.24435/materialscloud:2017.0008/v1

NM, M. Gibertini, P. Schwaller, D. Campi, A. Merkys, A. Marrazzo, T. Sohier, I. E. Castelli, A. Cepellotti, G. Pizzi and N. Marzari, *arXiv:1611.05234* (2016), **Nature Nanotechnology**, in press (2018).