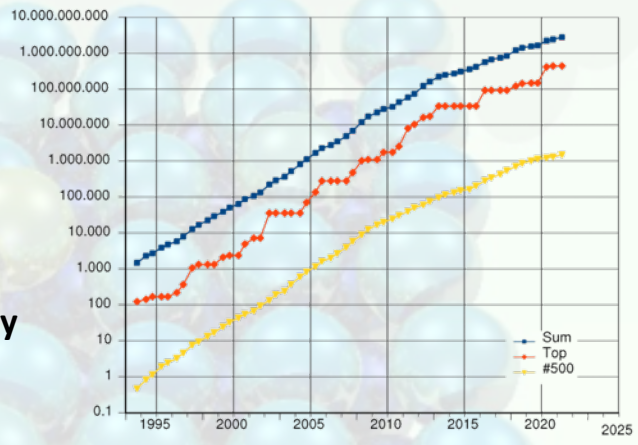
DIGITAL INFRASTRUCTURES EMPOWERING MATERIALS DISCOVERY

Nicola Marzari, EPFL & PSI

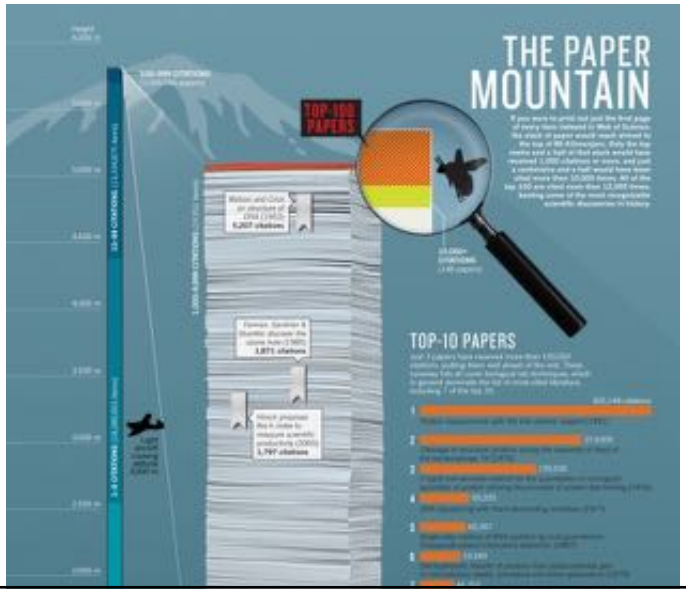
THE RISE OF COMPUTATIONAL SCIENCE

A calculation that took **one year in 1993** takes **30 seconds in 2022** (1-million-fold increase).

And this is just with bits: neurons are in, and qubits on the horizon. **21st-century science and discovery will be driven by computational science.**



IMPACT OF COMPUTATIONAL QUANTUM MECHANICS



THE TOP 100 PAPERS:

12 papers on density-functional theory in the top-100 most cited papers in the entire scientific-medical-engineering literature, ever.

NATURE, OCT 2014

MOST CITED PAPERS IN THE HISTORY OF APS

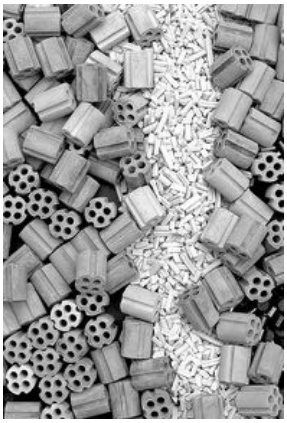
	Journal	# cites	Title	Author(s)
1	PRL (1996)	78085	Generalized Gradient Approximation Made Simple	Perdew, Burke, Ernzerhof
2	PRB (1988)	67303	Development of the Colle-Salvetti Correlation-Energy ...	Lee, Yang, Parr
3	PRB (1996)	41683	Efficient Iterative Schemes for Ab Initio Total-Energy ...	Kresse and Furthmuller
4	PR (1965)	36841	Self-Consistent Equations Including Exchange and Correlation ...	Kohn and Sham
5	PRA (1988)	36659	Density-Functional Exchange-Energy Approximation ...	Becke
6	PRB (1976)	31865	Special Points for Brillouin-Zone Integrations	Monkhorst and Pack
7	PRB (1999)	30940	From Ultrasoft Pseudopotentials to the Projector Augmented ...	Kresse and Joubert
8	PRB (1994)	30801	Projector Augmented-Wave Method	Blochl
9	PR (1964)	30563	Inhomogeneous Electron Gas	Hohenberg and Kohn
10	PRB (1993)	19903	Ab initio Molecular Dynamics for Liquid Metals	Kresse and Hafner
11	PRB (1992)	17286	Accurate and Simple Analytic Representation of the Electron ...	Perdew and Wang
12	PRB (1990)	15618	Soft Self-Consistent Pseudopotentials in a Generalized ...	Vanderbilt
13	PRB (1992)	15142	Atoms, Molecules, Solids, and Surfaces - Applications of the ...	Perdew, Chevary, ...
14	PRB (1981)	14673	Self-Interaction Correction to Density-Functional Approx. ...	Perdew and Zunger
15	PRB (1986)	13907	Density-Functional Approx. for the Correlation-Energy ...	Perdew
16	RMP (2009)	13513	The Electronic Properties of Graphene	Castro Neto et al.
17	PR (1934)	12353	Note on an Approximation Treatment for Many-Electron Systems	Moller and Plesset
18	PRB (1972)	11840	Optical Constants on Noble Metals	Johnson and Christy
19	PRB (1991)	11580	Efficient Pseudopotentials for Plane-Wave Calculations	Troullier and Martins
20	PRL (1980)	10784	Ground-State of the Electron-Gas by a Stochastic Method	Ceperley and Alder

Marzari
(11 Apr 2019)

THE RISE OF MATERIALS SCIENCE

IF WE FLY AGAIN…

THE MOST IMPORTANT MATERIAL EVER?



MATERIALS ARE KEY TO SOCIETAL WELL BEING

We need novel materials for:

- **Energy harvesting, conversion, storage, efficiency**
- **Environmental protection and reparation**
- **High-tech and high-value industries**
- **Information and communication technologies**
- **Health care and biomedical engineering**
- **Pharmaceuticals** (crystallization, stability, polytypes)
- **Monitoring, provenance, and safety of foods**
- **Fundamental science** (graphene and 2D materials, topological insulators, entangled spins for quantum computing, high-T_c)
- **Experimental science** (detectors, sensors, magnets)

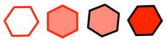
COMPUTATIONAL MATERIALS DESIGN AND DISCOVERY

3 Technologies That Could Create Trillion-Dollar Markets Over the Next Decade

By Greg Satell Updated April 21, 2019 9:00 a.m. ET

Yet today, we're in the midst of a materials revolution. Powerful simulation techniques, combined with increased computing power and machine learning, are enabling researchers to automate much of the discovery process, vastly accelerating the development of new materials

BARRON'S (April 2019)

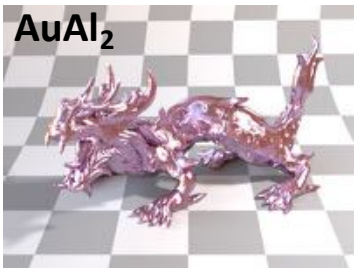


THE COLOUR OF MATERIALS: REFLECTIVITY IN THE OPTICAL LIMIT

Os

Cs

$AuAl_2$



G. Prandini, G.M. Rignanese, and N. Marzari, npj Computational Materials 5, 129 (2019)

PHONON-LIMITED RESISTIVITY IN DOPED GRAPHENE

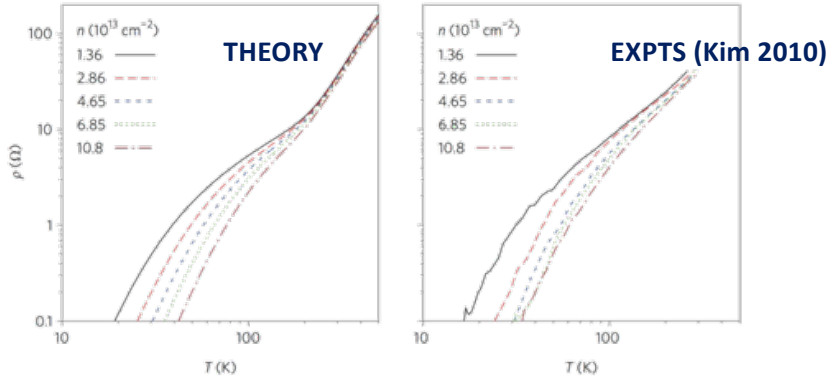
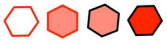
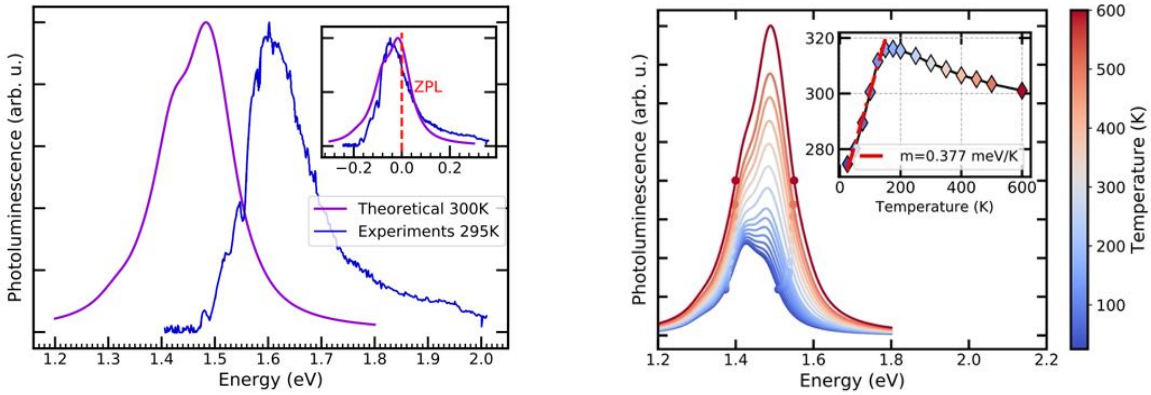


Figure 1 | Electrical resistivity of graphene as a function of temperature and doping (ρ, electrical resistivity; T, temperature; n, carrier density). Left panel: first-principles results obtained using a combination of density-functional perturbation theory, many-body perturbation theory and Wannier interpolations to solve the Boltzmann transport equation. Right panel: experimental data. Adapted from ref. 4, American Chemical Society.

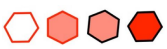
C.-H. Park *et al.,* Nano Letters 14, 1113 (2014)
T. Y. Kim, C.-H. Park, and N. Marzari, Nano Letters 16, 2439 (2016)



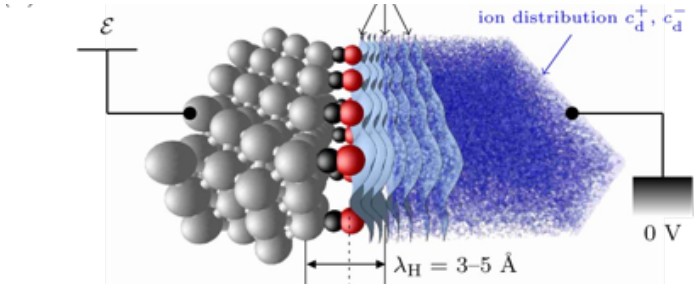
READ-OUT OF A QUBIT: PHOTOLUMINESCENCE OF A DEFECT



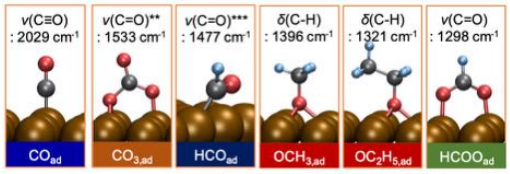
F. Libbi, P.M. de Melo, Z. Zanolli, M. Verstraete, and N. Marzari, Phys. Rev. Lett. 128, 167401 (2022)

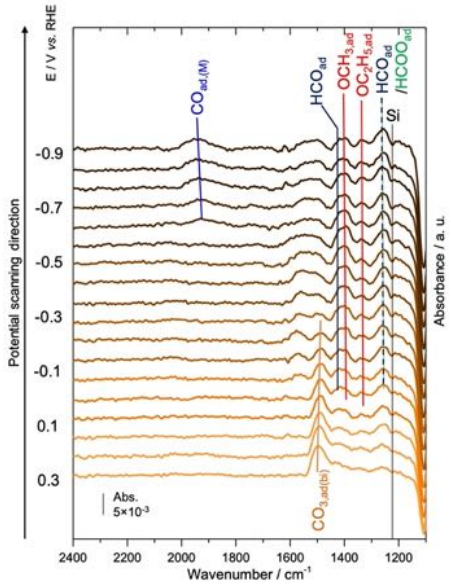


OPERANDO SPECTROSCOPIES



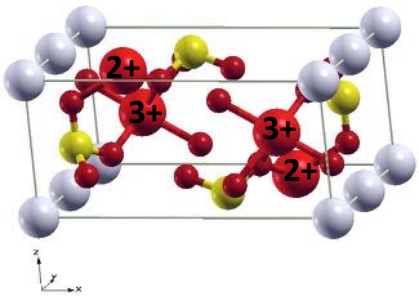
O. Andreussi *et al.*, J. Chem. Phys. 136, 064102 (2012)
F. Nattino *et al.*, J. Chem. Phys. 150, 041722 (2019)

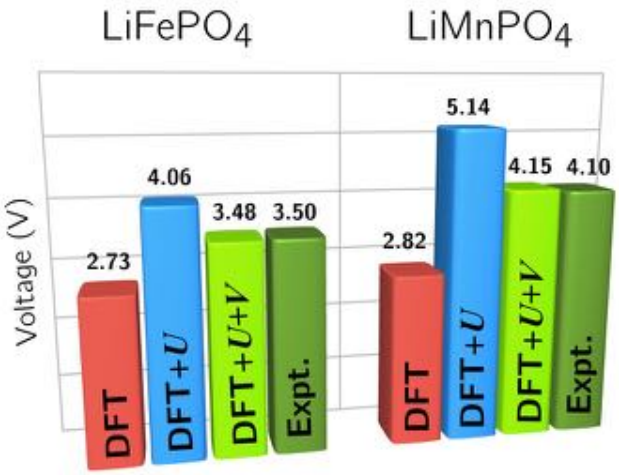




LI-ION CATHODES

$Li_{0.5}FePO_4$





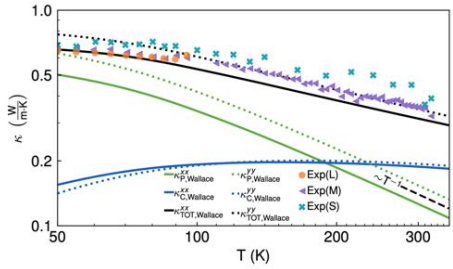
M. Cococcioni and N. Marzari, Phys. Rev. Materials 3, 033801 (2019)
I.Timrov *et al.*, arXiv (2022)

SOME NEW PHYSICS DRIVEN BY THERMOELECTRICS

nature physics ARTICLES
https://doi.org/10.1038/s41567-019-0520-x

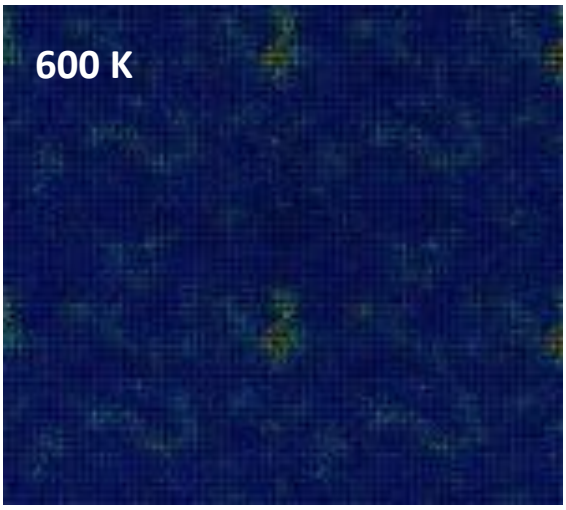
Unified theory of thermal transport in crystals and glasses

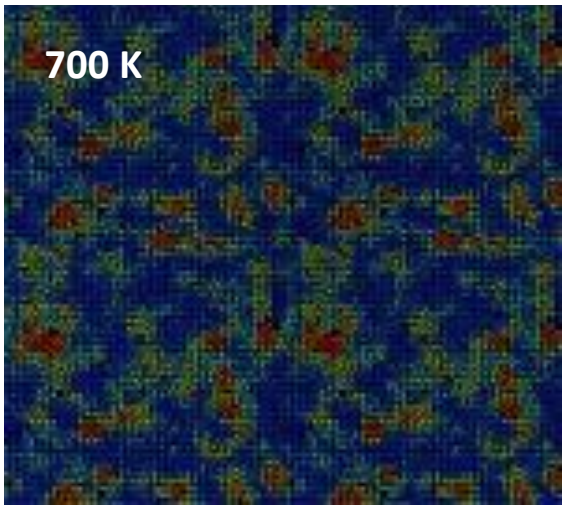
Michele Simoncelli[1], Nicola Marzari[1] and Francesco Mauri[2*]



$$\kappa^{\alpha\beta} = \kappa_{\mathrm{P}}^{\alpha\beta} + \frac{\hbar^2}{k_B T^2}\frac{1}{\mathcal{V}N_c}\sum_{\boldsymbol{q}}\sum_{s\neq s'}\frac{\omega(\boldsymbol{q})_s + \omega(\boldsymbol{q})_{s'}}{2}V^{\alpha}(\boldsymbol{q})_{s,s'}V^{\beta}(\boldsymbol{q})_{s',s}\times$$
$$\times\frac{\omega(\boldsymbol{q})_s\bar{N}^T(\boldsymbol{q})_s[\bar{N}^T(\boldsymbol{q})_s+1]+\omega(\boldsymbol{q})_{s'}\bar{N}^T(\boldsymbol{q})_{s'}[\bar{N}^T(\boldsymbol{q})_{s'}+1]}{4[\omega(\boldsymbol{q})_{s'}-\omega(\boldsymbol{q})_s]^2+[\Gamma(\boldsymbol{q})_s+\Gamma(\boldsymbol{q})_{s'}]^2}[\Gamma(\boldsymbol{q})_s+\Gamma(\boldsymbol{q})_{s'}]$$

BEYOND EXPERIMENTAL RESOLUTION: 2D MELTING OF AL(100)

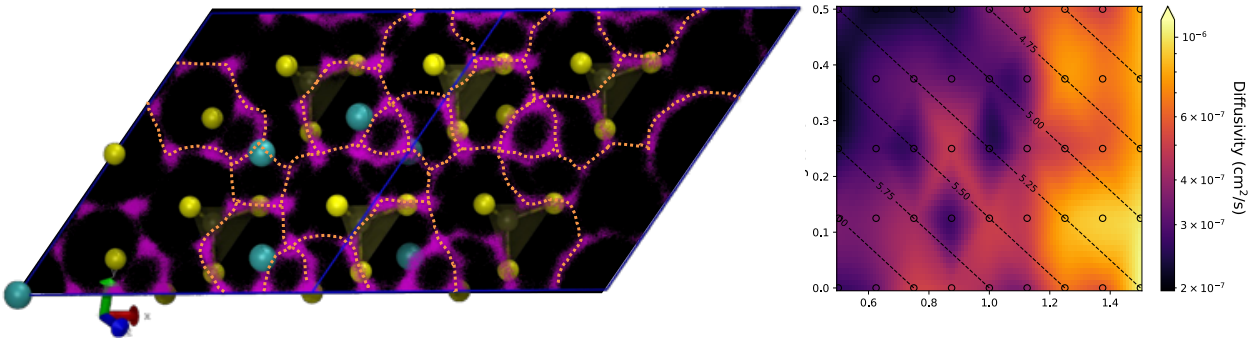




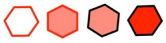
N. L. Nguyen, L. Kahle, F. Baletto, and N. Marzari (in preparation, 2022)

ENGINEERING SOLID-STATE ELECTROLYTES

Cl-doped argyrodite (Li_6PS_5Cl)



S. Muy, L. Ercole, N. Marzari, 2 patents filed with SOLVAY (2022)



RELIABLY, REPRODUCIBLY, HIGH-THROUGHPUT

VOLUME 88, NUMBER 25 PHYSICAL REVIEW LETTERS 24 JUNE 2002

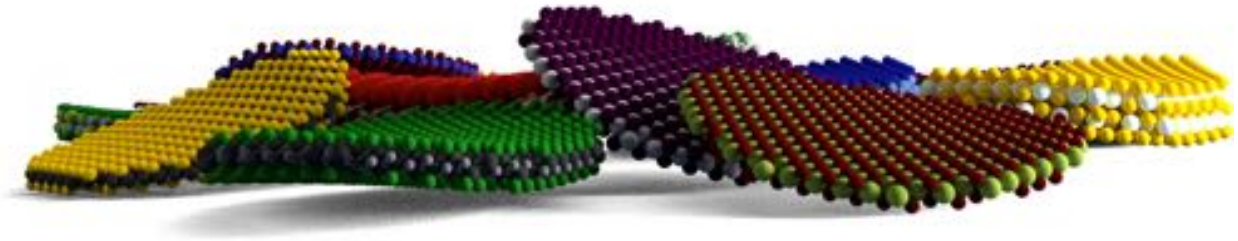
Combined Electronic Structure and Evolutionary Search Approach to Materials Design

G. H. Jóhannesson, T. Bligaard, A. V. Ruban, H. L. Skriver, K. W. Jacobsen, and J. K. Nørskov
Center for Atomic-Scale Materials Physics, Department of Physics, Technical University of Denmark, DK-2800, Lyngby, Denmark
(Received 20 February 2002; published 10 June 2002)

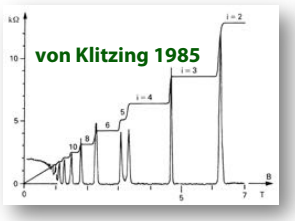
We show that density functional theory calculations have reached an accuracy and speed making it possible to use them in conjunction with an evolutionary algorithm to search for materials with specific properties. The approach is illustrated by finding the most stable four component alloys out of the 192 016 possible fcc and bcc alloys that can be constructed out of 32 different metals. A number of well known and new "super alloys" are identified in this way.

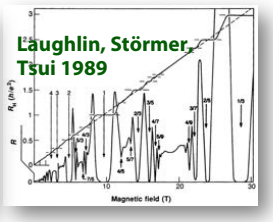
DOI: 10.1103/PhysRevLett.88.255506 PACS numbers: 81.05.Bx, 61.66.Dk, 71.15.Mb

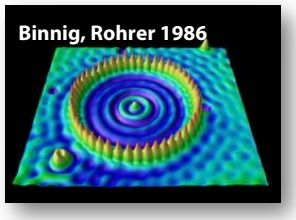
EXAMPLE: COMPUTATIONAL EXFOLIATION OF ALL KNOWN INORGANIC MATERIALS

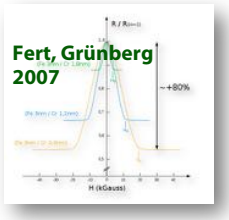


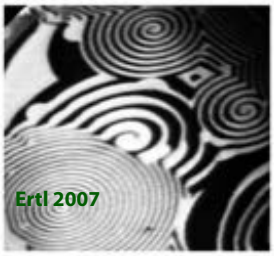
PHYSICS AND CHEMISTRY IN LOW DIMENSIONS

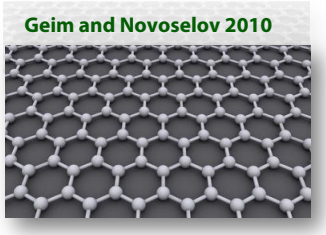






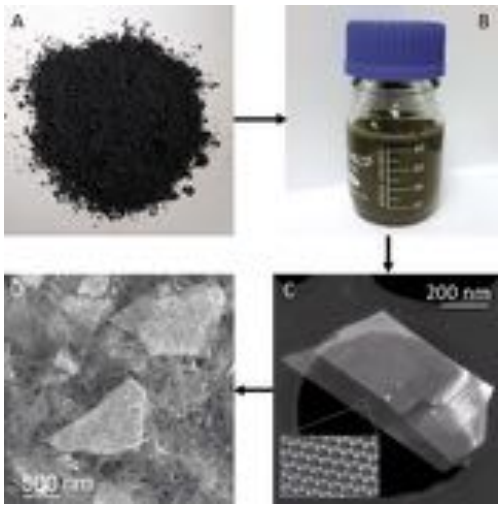




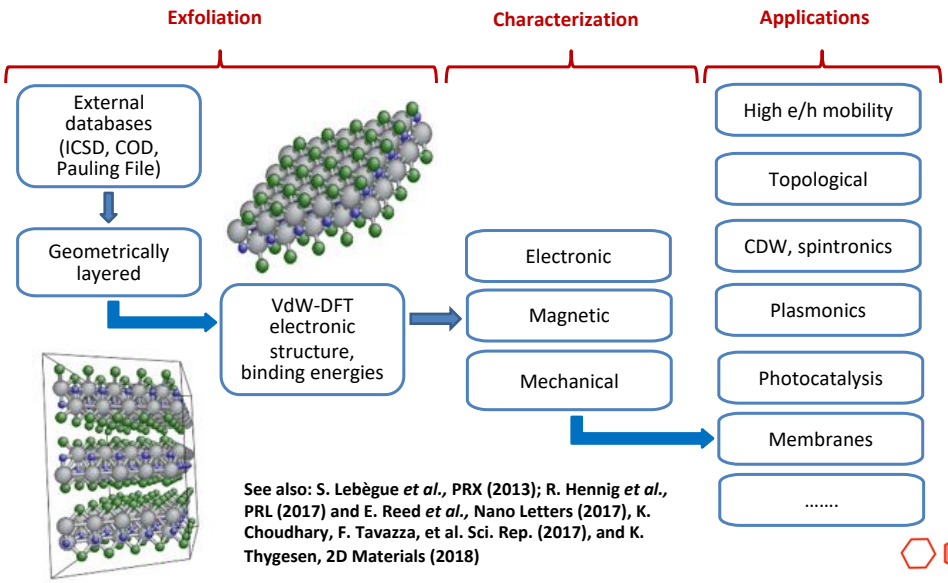


HOW DO WE PRODUCE 2D MATERIALS?

Mechanical (e.g. Geim/Novoselov, fig. from Nature/NUS) **or liquid exfoliation** (e.g. Nicolosi/Coleman, fig. from Science), **electrochemical intercalation**. Also, bottom-up: **CVD and wet chemical synthesis.**

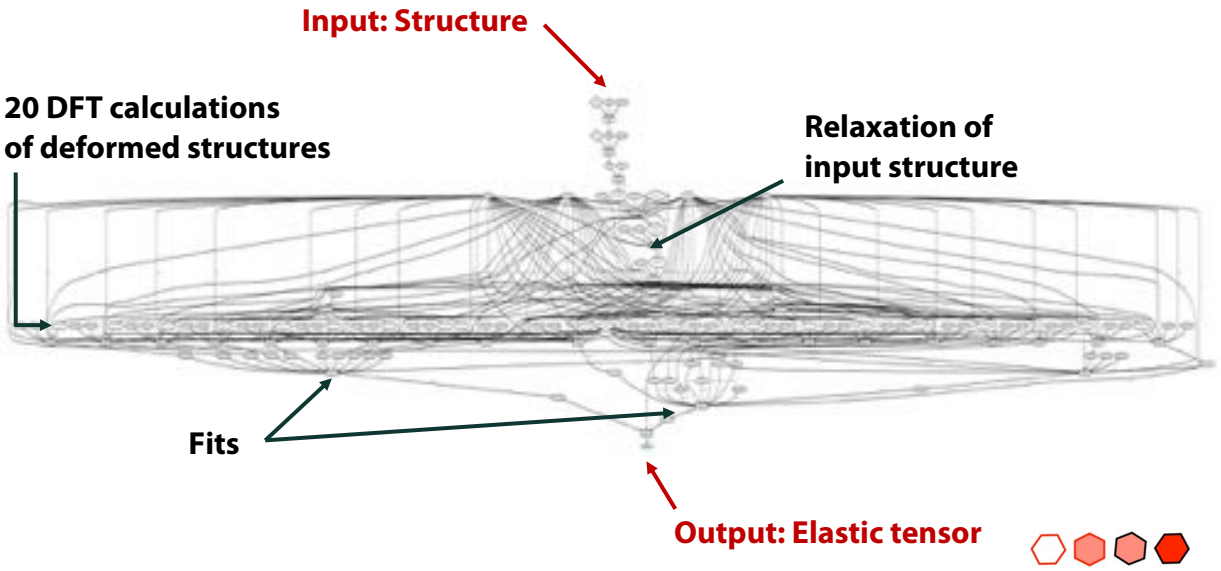


HIGH-THROUGHPUT COMPUTATIONAL EXFOLIATION

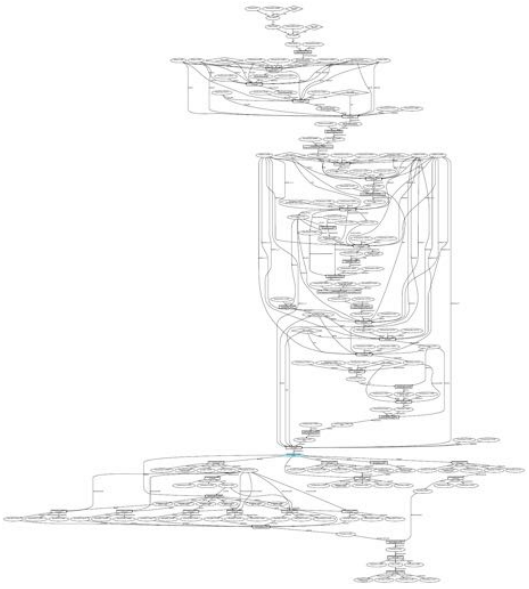


See also: S. Lebègue *et al.*, PRX (2013); R. Hennig *et al.*, PRL (2017) and E. Reed *et al.*, Nano Letters (2017), K. Choudhary, F. Tavazza, et al. Sci. Rep. (2017), and K. Thygesen, 2D Materials (2018)

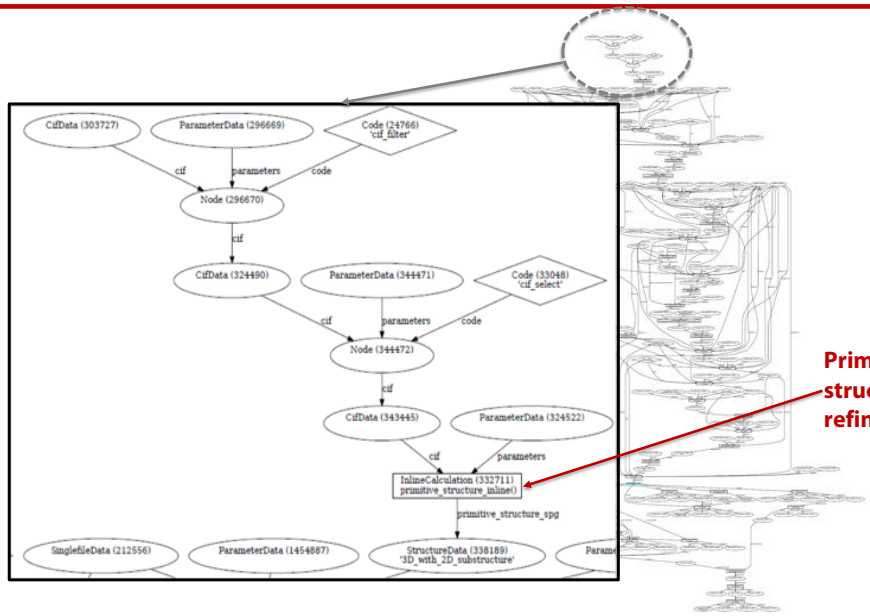
AUTOMATIC WORKFLOWS: FROM STRUCTURE TO PROPERTY



LET'S START FROM A MATERIAL ($VOBr_2$)

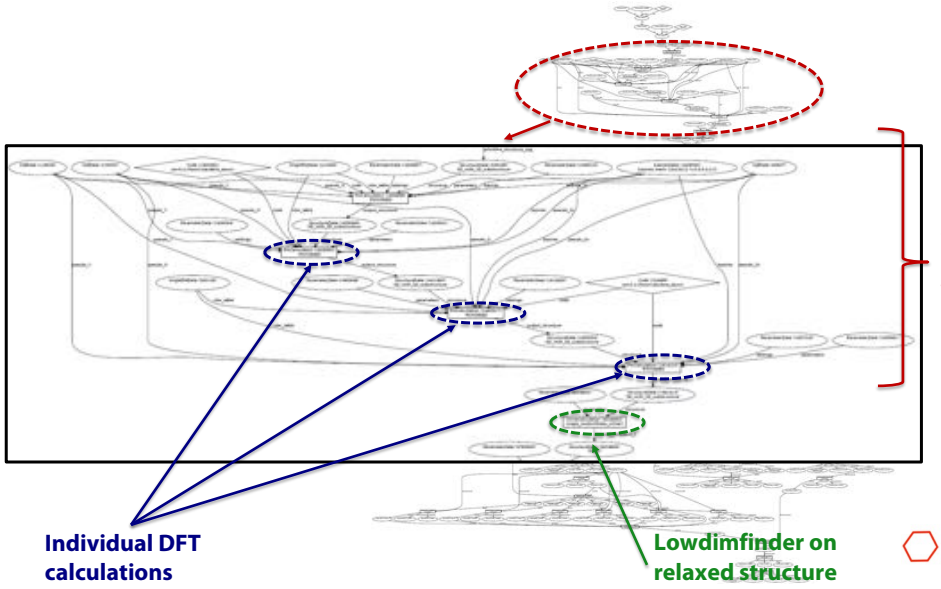


FROM DATABASE ENTRY TO A WORKING STRUCTURE



Primitive cell & structure symmetry refinement

3D RELAXATION

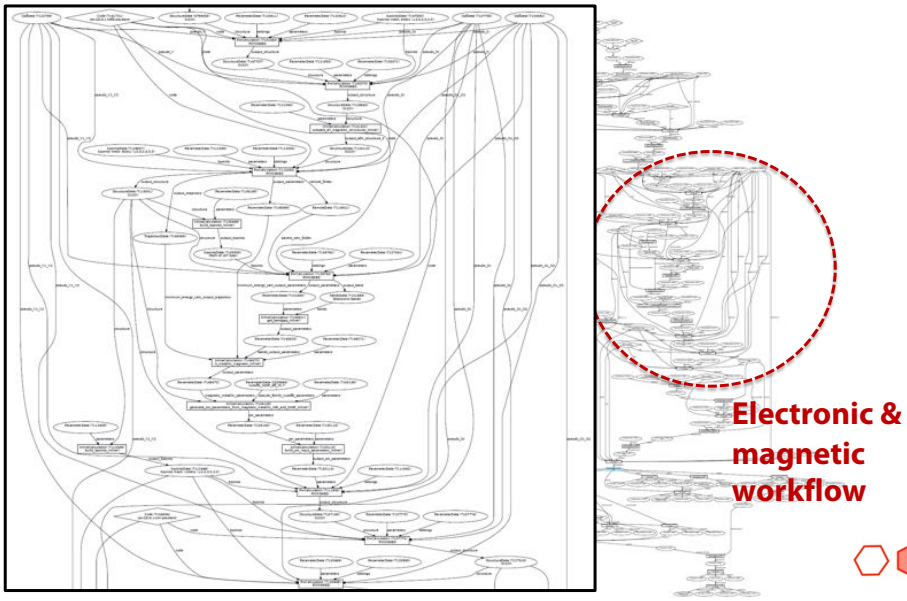


Quantum ESPRESSO Workflow

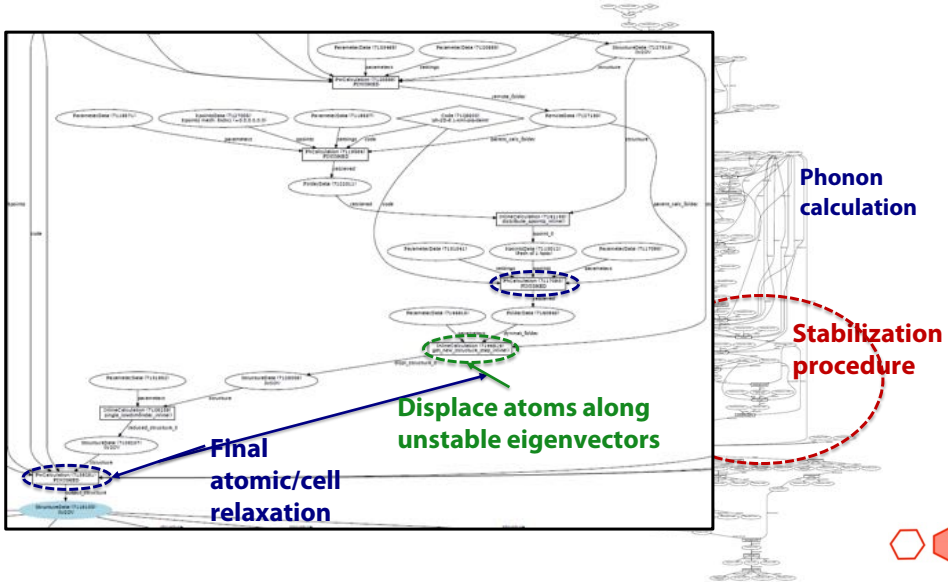
Individual DFT calculations

Lowdimfinder on relaxed structure

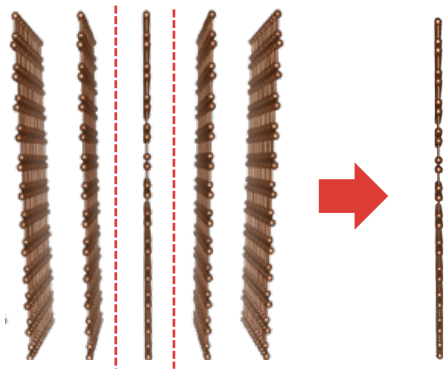
MAGNETIC SCREENING OF THE 2D MONOLAYER



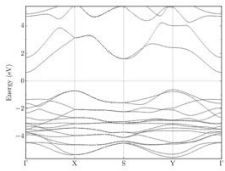
REMOVING MECHANICAL INSTABILITIES



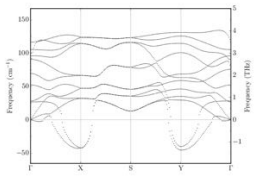
ALL AUTOMATED…



Band structures



Phonon dispersions



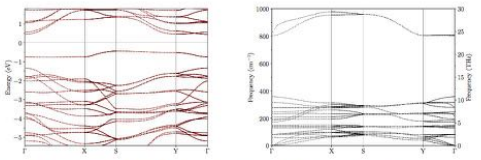
FINALLY…

215 VBr_2O (Pmm2)

Info and properties (for more details and definitions see page 2)

Formula	VBr_2O	**DF2-C09 Binding energy [meV/Å²]**	14.4
Spacegroup	Pmm2	**rVV10 Binding energy [meV/Å²]**	21.6
Prototype	VCl_2O (Pmm2)	**Band gap [eV]**	0.9
Parent 3D	VBr_2O	**Magnetic State**	AFM
Source DB	ICSD	**Tot. Magnetization [μ_B/cell]**	0.0
DB ID	24381	**Abs. Magnetization [μ_B/cell]**	2.54

Band structure and phonon dispersions



Band structure: energy bands of VBr_2O (66 electrons) in a window around the chemical potential and along a high-symmetry path. The number of bands included in the calculation is 80.

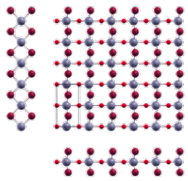
Phonon dispersions: phonon frequencies of VBr_2O (8 atoms/cell) along a high-symmetry path.

Crystal structure

Structural parameters: cell (top) and atomic positions (bottom) of VBr_2O in cartesian coordinates.

	x [Å]	y [Å]	z [Å]
a_1	3.80622044	0.00000000	0.00000000
a_2	0.00000000	7.17029927	0.00000000
a_3	0.00000000	0.00000000	19.47346306

		x [Å]	y [Å]	z [Å]
•	Br	2.00107500	5.37772439	−1.78545446
•	Br	2.00107500	1.79257489	−1.78545446
•	V_1	1.70214333	3.58514964	0.00000000
•	V_2	1.70214341	0.00000000	0.00000000
•	Br	2.00107500	5.37772439	1.78545446
•	Br	2.00107500	1.79257489	1.78545446
•	O_1	0.06788642	3.58514964	0.00000000
•	O_2	0.06788661	0.00000000	0.00000000

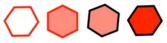


Orthographic projections: different views of VBr_2O from the x axis (left), the y axis (bottom) and the z axis (center).

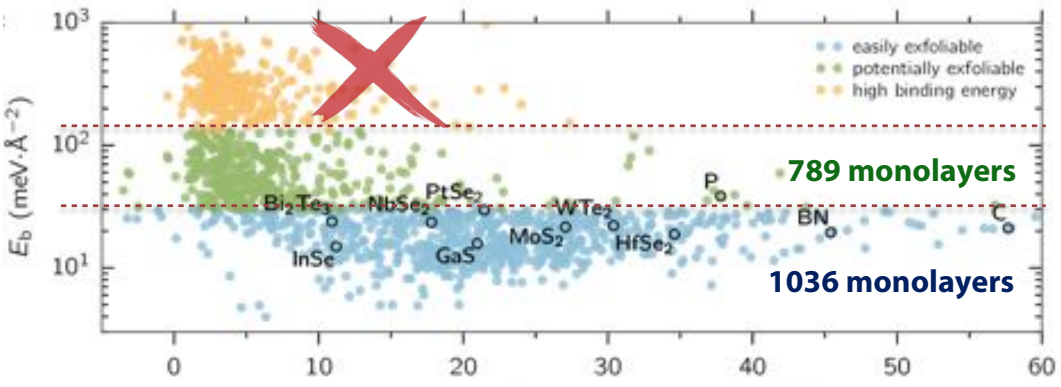
HOW MANY CANDIDATES? GEOMETRIC SCREENING

	Unique to COD	Unique to ICSD	Common to both	Total
Entries analyzed	307616	172370		479986*
CIF inputs	99212	87070		186282*
Unique 3D structures	60354	34548	13521	108423
Layered 3D structures	1180	3257	1182	5619

*At this level unicity is not tested



HOW MANY CANDIDATES? QUANTUM SCREENING

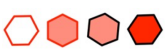


Not exfoliable

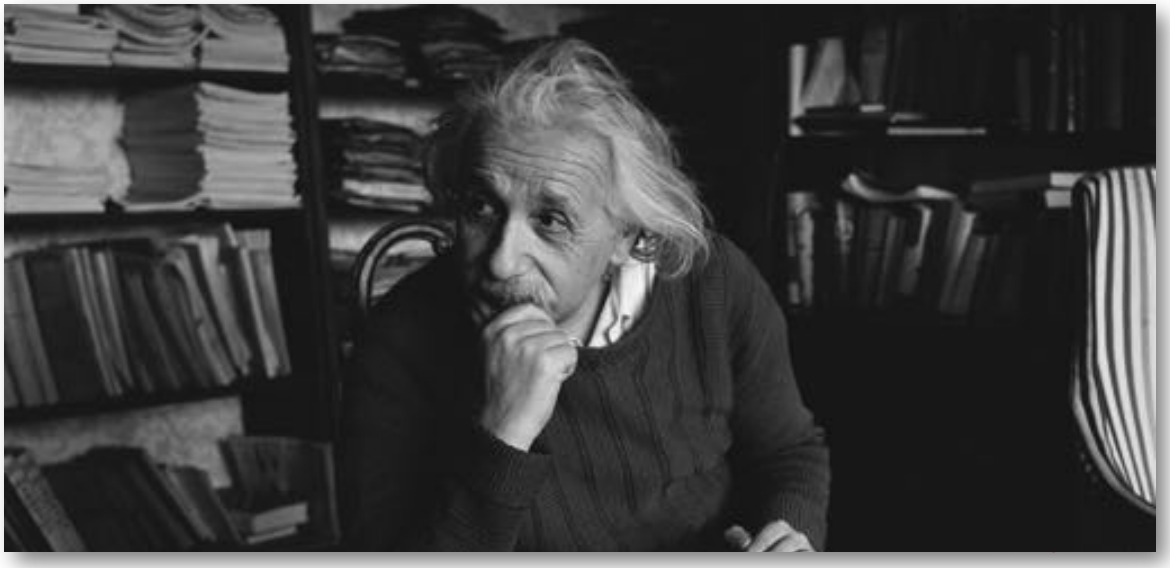
Potentially exfoliable

Easily exfoliable

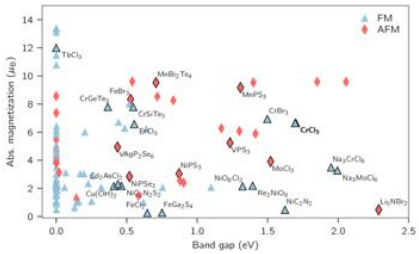
- *E_b < 30 meV/Å^2 (DF2-C09) or E_b < 35 meV/Å^2 (rVV10) → 2D, easily exfoliable*
- *In-between → 2D, potentially exfoliable*
- *E_b > 130 meV/Å^2 → not 2D (discarded)*



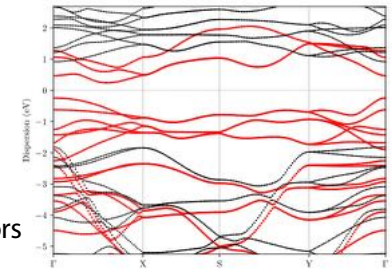
WHAT TO DO NEXT?



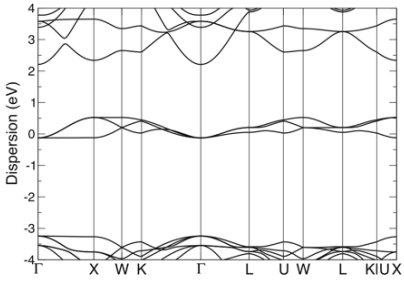
FROM ELECTRONICS…



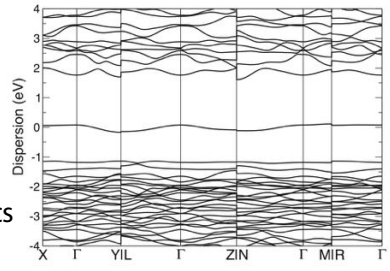
Magnetic metals and insulators



Half-semiconductors

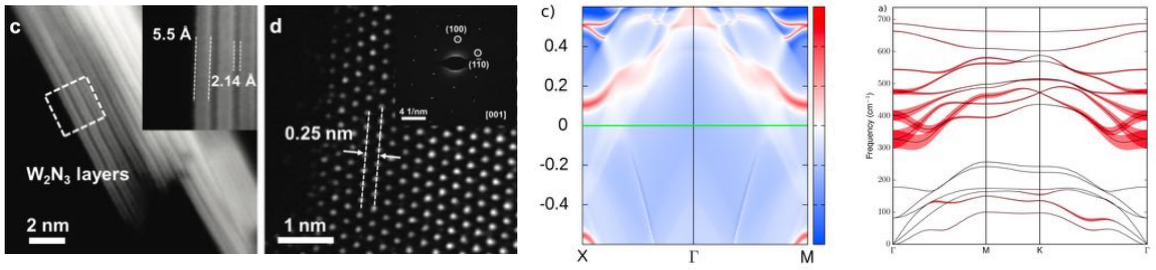


Transparent conductors



Plasmonics

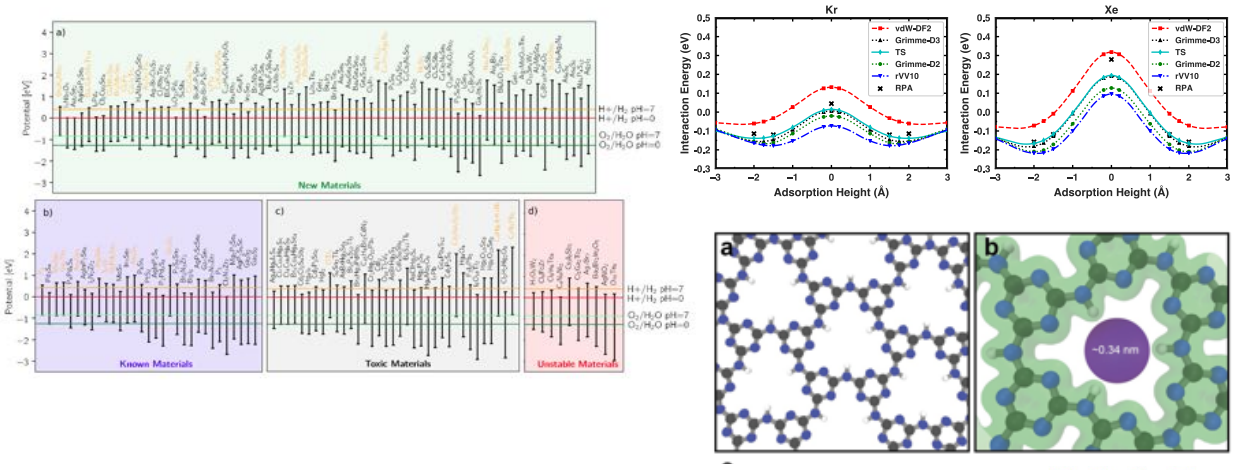
…TO THE LARGEST SUPERCONDUCTING Tc IN 2D…



H. Jun *et al.*, Advanced Materials 31, 1902709 (2019)

D. Campi, S. Kumari, and N. Marzari, Nano Letters 21, 3435 (2021)

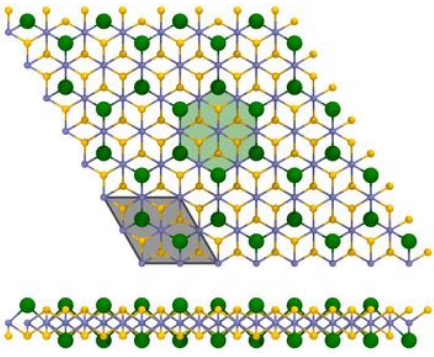
…TO MATERIALS: PHOTOCATALYSIS, MEMBRANES



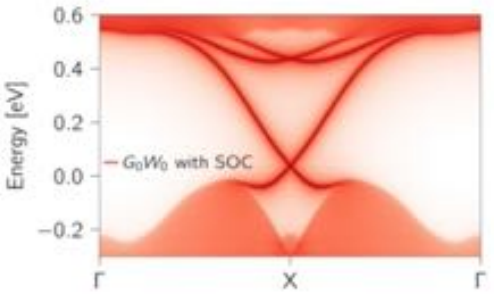
Science Advances (2019), and under review (2022)

THE DISCOVERY OF JACUTINGAITE

THE DISCOVERY OF JACUTINGAITE

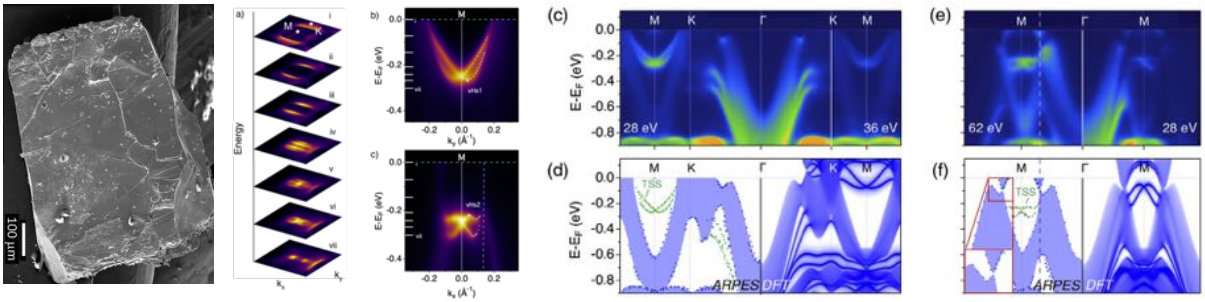


Classified as potentially exfoliable
(binding energy of 60 meV Å^{-2})

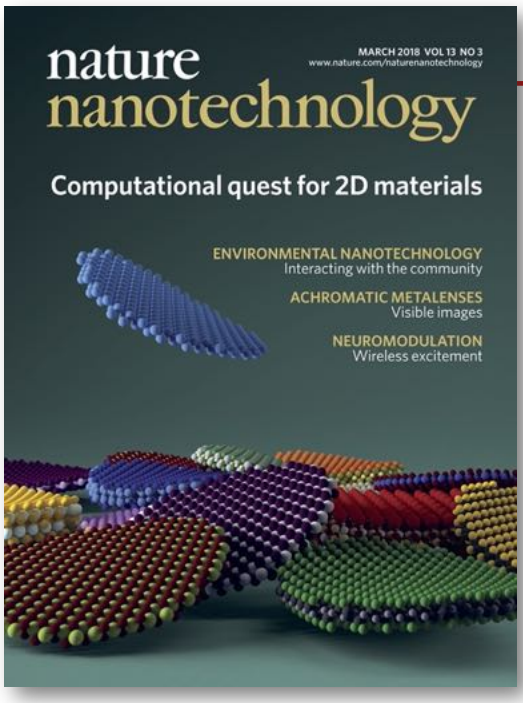


A. Marrazzo *et al.*, Phys. Rev. Lett. 120, 117701 (2018)

ROOM-TEMPERATURE KANE-MELE QSHI



A. Marrazzo *et al.*, Phys. Rev. Lett. 120, 117701 (2018)
I. Cucchi, *et al.*, Phys. Rev. Lett. 124, 106402 (2020)
A. Marrazzo, N. Marzari, and M. Gibertini, Phys. Rev. Res. 2, 012063(R) (2020)



THERE IS PLENTY OF ROOM AT THE TOP

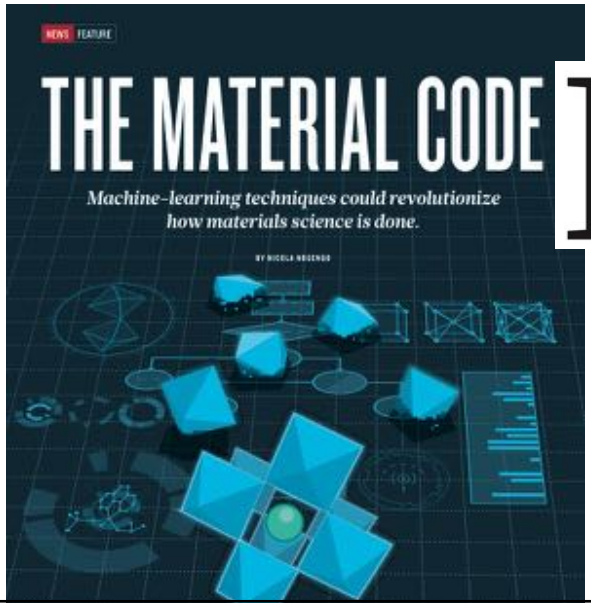
- High electron/hole mobility devices
- Topological insulators, quantum computing
- Ferromagnetic/spintronics in 2D
- Charge-density waves and superconductors
- Plasmonics, transparent conductors

3D layered parents:

- Solid-state ionic conductors
- Hydrogen or oxygen evolution catalysts
- Membranes for filtration/separation
- Piezo, ferro, and thermoelectrics

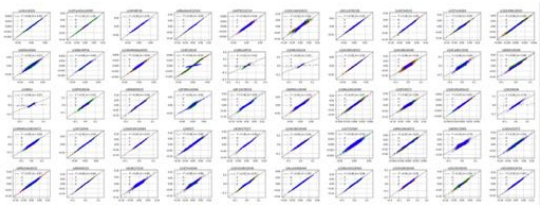
N. Mounet, M. Gibertini, P. Schwaller, D. Campi, A. Merkys, A. Marrazzo, T. Sohier, I. E. Castelli, A. Cepellotti, G. Pizzi and N. Marzari, Nature Nanotechnology 13, 246 (2018)

MACHINE LEARNING AS THE GREAT ACCELERATOR

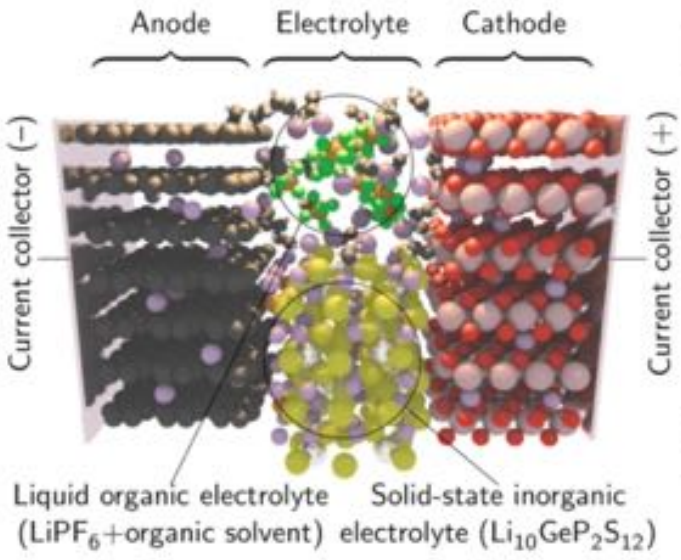


Nature, May 2016

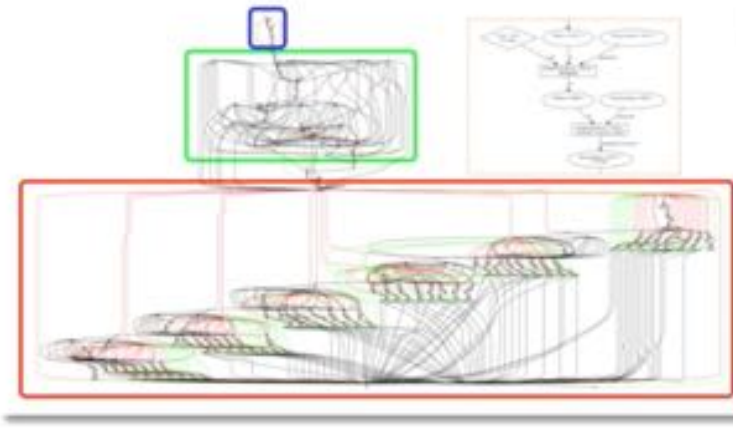
It's a strong contender for the geekiest video ever made: a close-up of a smartphone with line upon line of numbers and symbols scrolling down the screen. But when visitors stop by Nicola Marzari's office, which overlooks Lake Geneva, he can hardly wait to show it off. "It's from 2010," he says, "and this is my cellphone calculating the electronic structure of silicon in real time!"

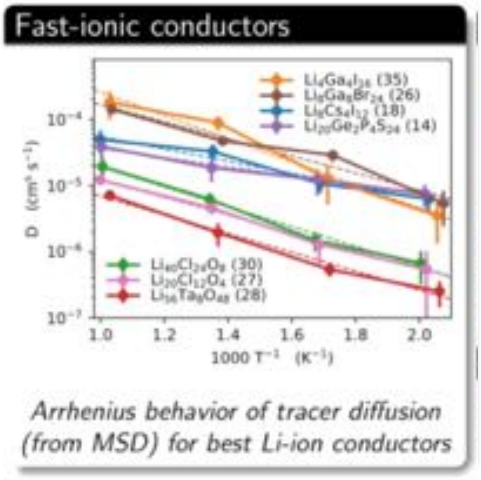


ALL SOLID-STATE BATTERIES

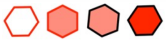


AUTOMATED SCREENING





L. Kahle, A. Marcolongo, and N. Marzari, Energy & Environmental Science 13, 928 (2020)



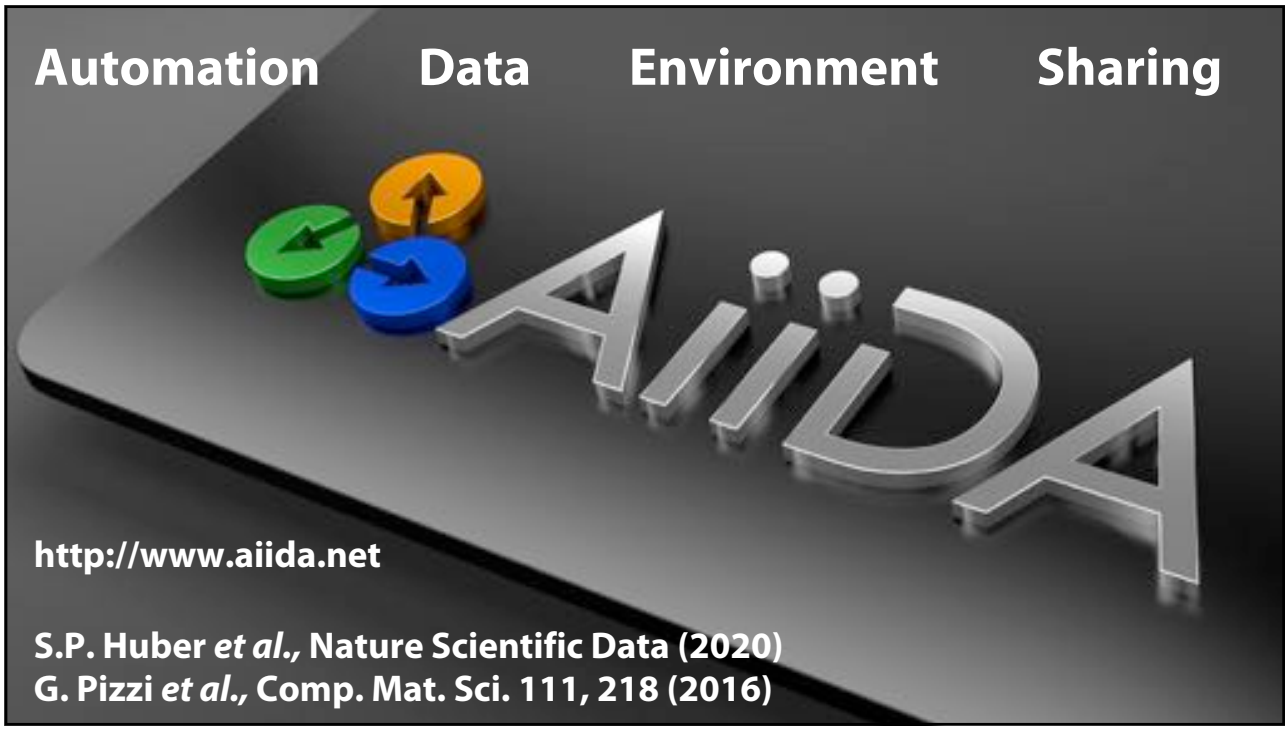
OPEN SCIENCE TECHNOLOGY STACK

1. Widely used, **open-source community codes**

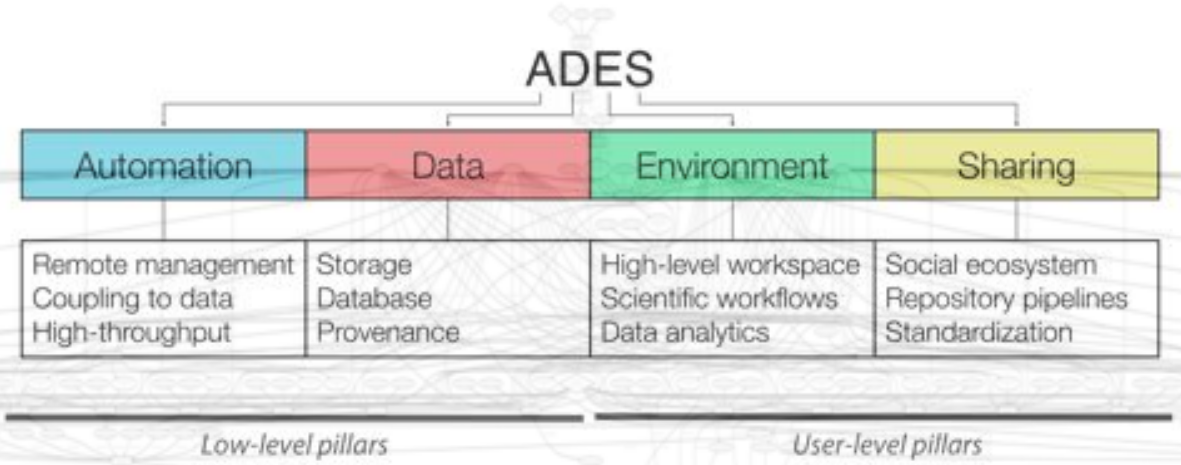
2. An **operating system** for high-throughput computational science, data provenance and reproducibility – http://aiida.net

3. A **work environment** for non specialist where to run simulations

4. A **dissemination platform** for raw and curated data, simulation services, educational tools



ADES MODEL FOR COMPUTATIONAL SCIENCE



S.P. Huber *et al.*, Nature Scientific Data (2020)
G. Pizzi *et al.*, Comp. Mat. Sci. 111, 218 (2016)

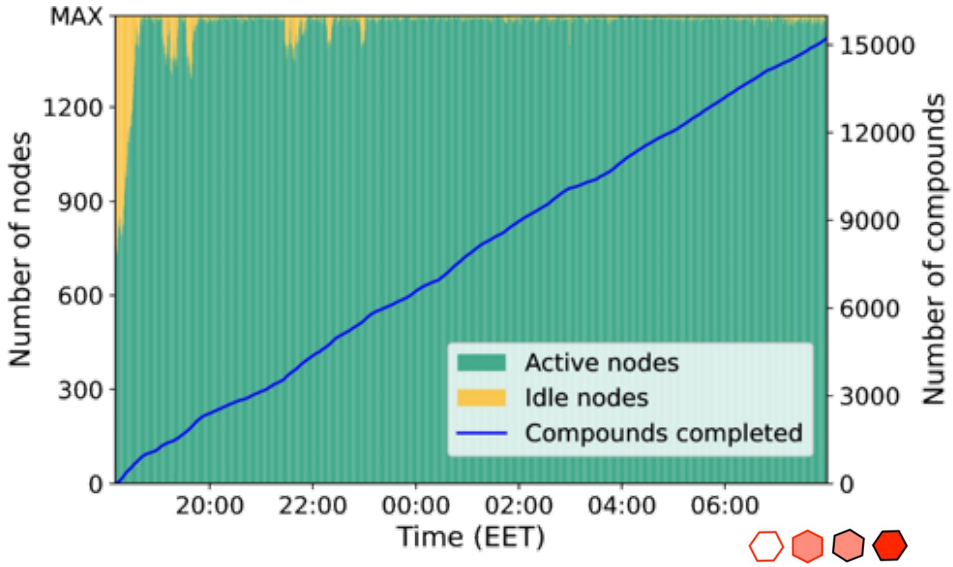
LUMI-C HERO RUN

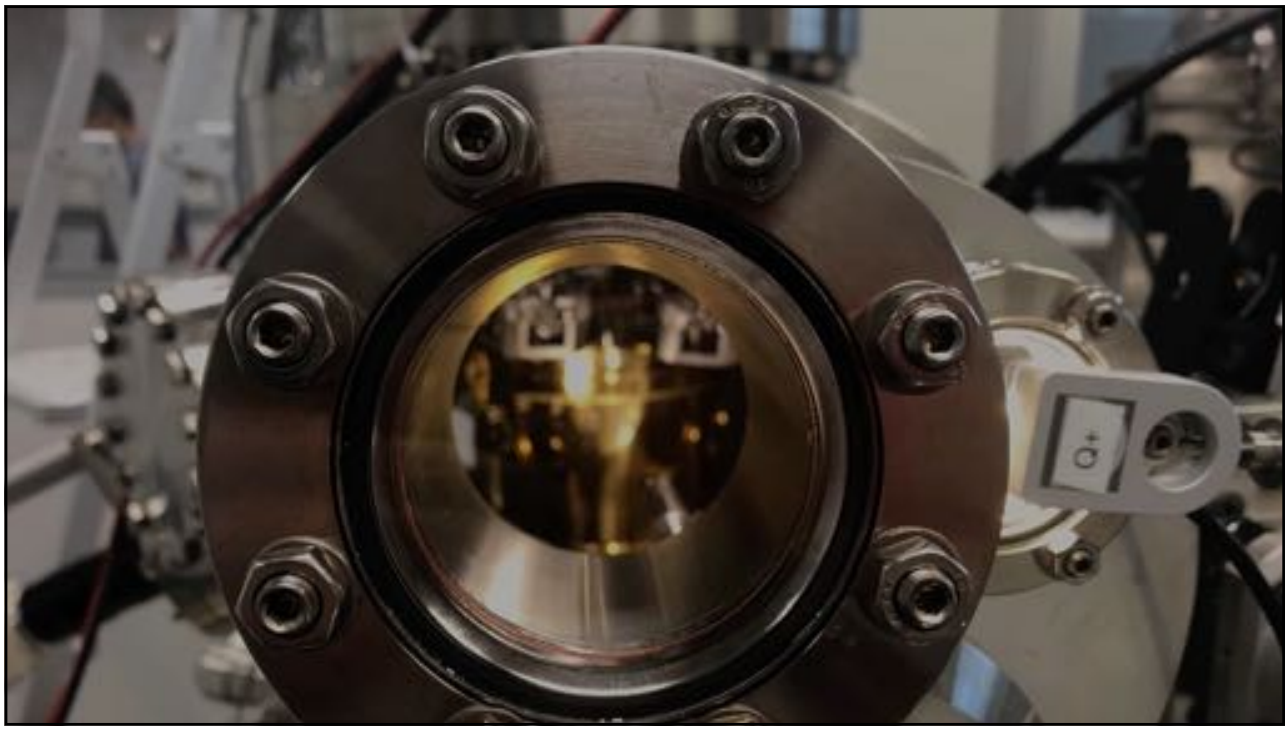
12 hours and 196,608 cores:

ran **55704 QE calculations**

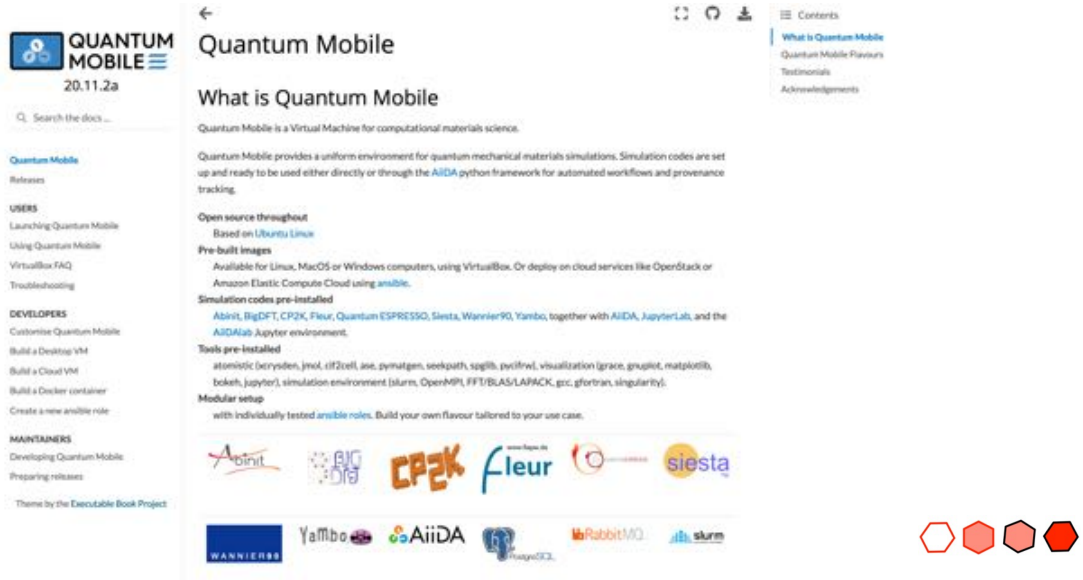
handled **7887 errors on the fly**

fully optimized geometries of 15324 different compounds





READY TO GO IN THE QUANTUM MOBILE



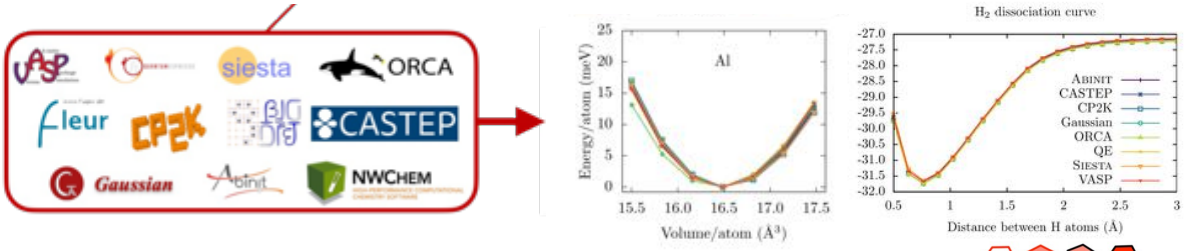
COMMON WORKFLOWS

ARTICLE OPEN

Common workflows for computing material properties using different quantum engines

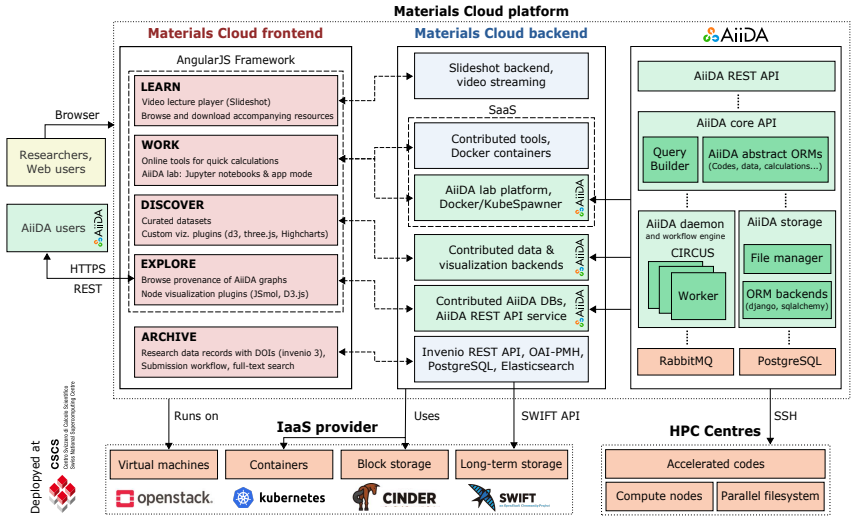
Sebastiaan P. Huber[1✉], Emanuele Bosoni[2], Marnik Bercx[1], Jens Bröder[3,4], Augustin Degomme[5], Vladimir Dikan[2], Kristjan Eimre[6], Espen Flage-Larsen[7,8], Alberto Garcia[2], Luigi Genovese[5], Dominik Gresch[9], Conrad Johnston[10], Guido Petretto[11], Samuel Poncé[1], Gian-Marco Rignanese[11], Christopher J. Sewell[1], Berend Smit[12], Vasily Tseplyaev[3,4], Martin Uhrin[1], Daniel Wortmann[3], Aliaksandr V. Yakutovich[1,12], Austin Zadoks[1], Pezhman Zarabadi-Poor[13,14], Bonan Zhu[14,15], Nicola Marzari[1] and Giovanni Pizzi[1✉]

```
$ aiida-common-workflows launch eos siesta --structure=Al --protocol=precise
```



S. Huber *et al.*, npj Computational Materials 7, 136 (2021)

MATERIALS CLOUD



Indexed by

SCIENTIFIC DATA

re3data.org

FAIRsharing.org

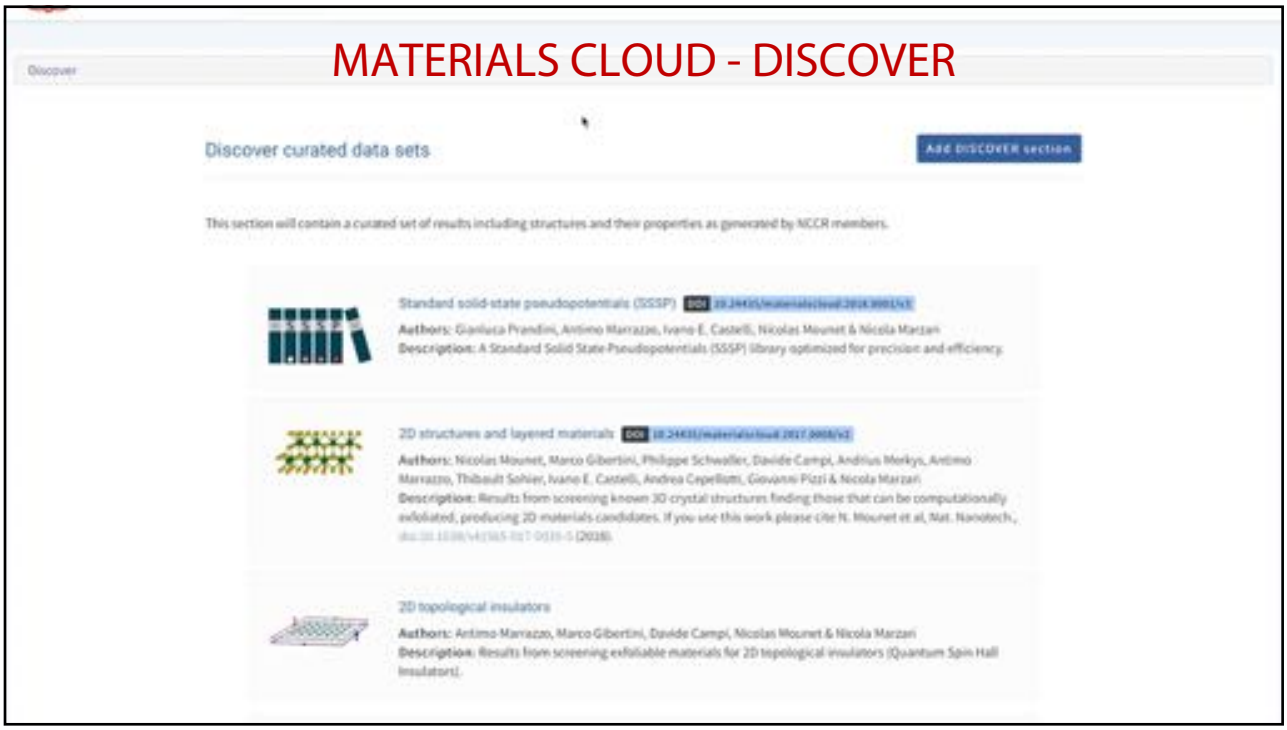
GO FAIR

Google Dataset Search

B2FIND EUDAT

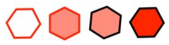
L. Talirz *et al.*, Scientific Data 7, 299 (2020)

MATERIALS CLOUD - DISCOVER



MATERIALS CLOUD - ARCHIVE

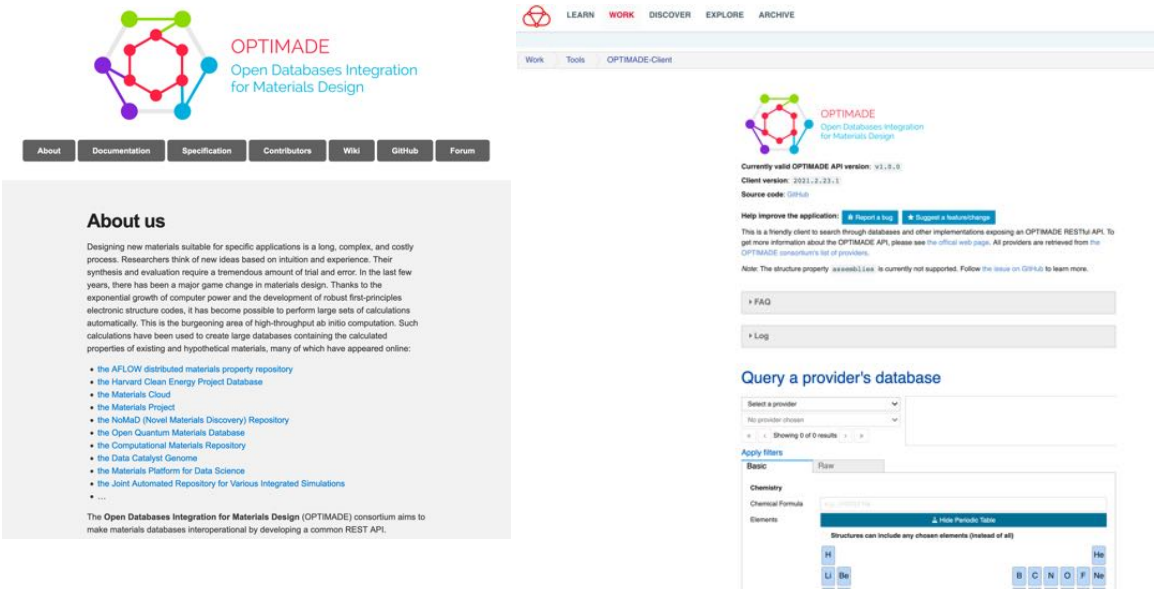
LET'S BROADEN THE HORIZON



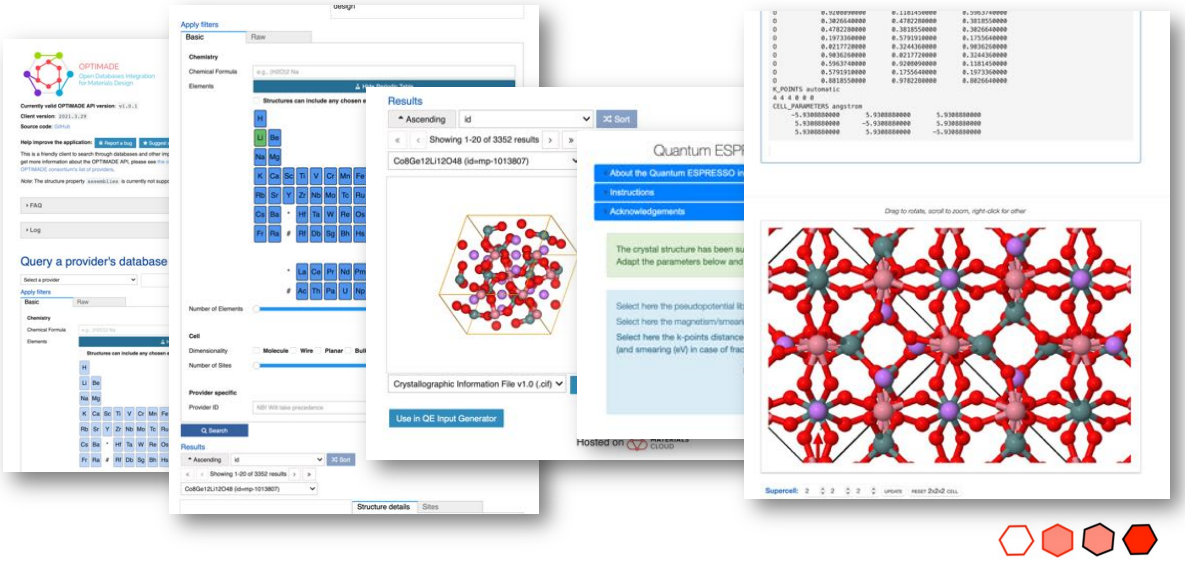
THE BEZOS MANDATE: EXTERNALIZABLE SERVICE INTERFACES

1) All teams will henceforth **expose their data and functionality through service interfaces.**
2) Teams must communicate with each other through these interfaces.
3) There will be no other form of interprocess communication allowed.
4) All service interfaces, without exception, must be designed **from the ground up to be externalizable**.

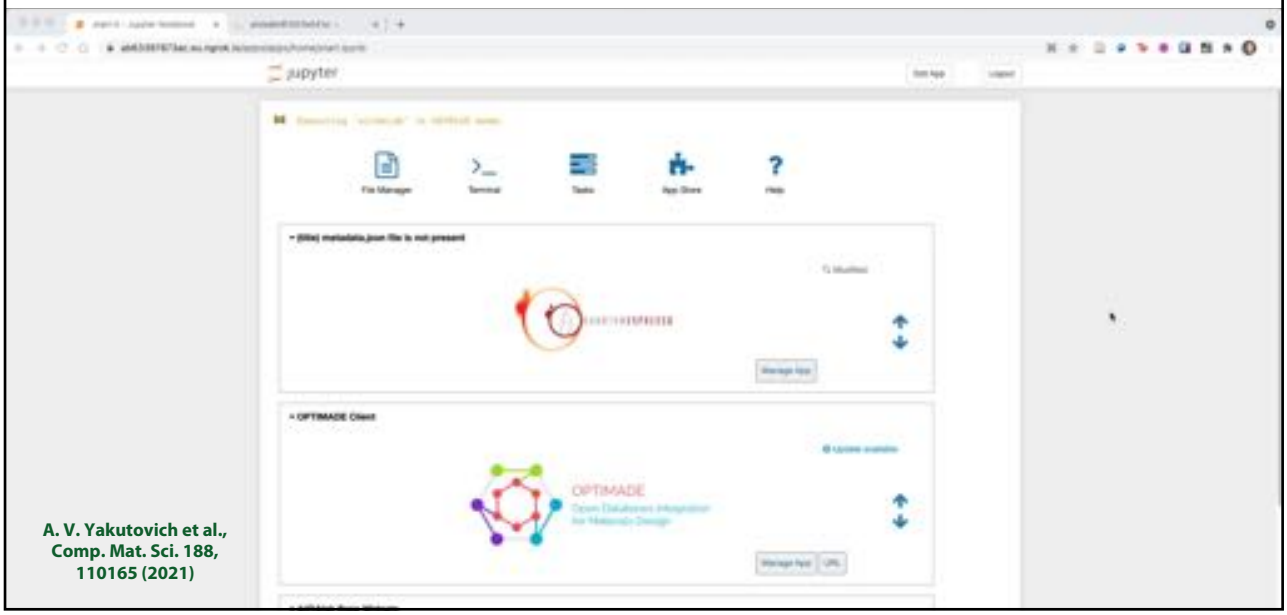
IN ACTION: OPTIMADE UNIVERSAL REST API



IN ACTION: OPTIMADE UNIVERSAL REST API

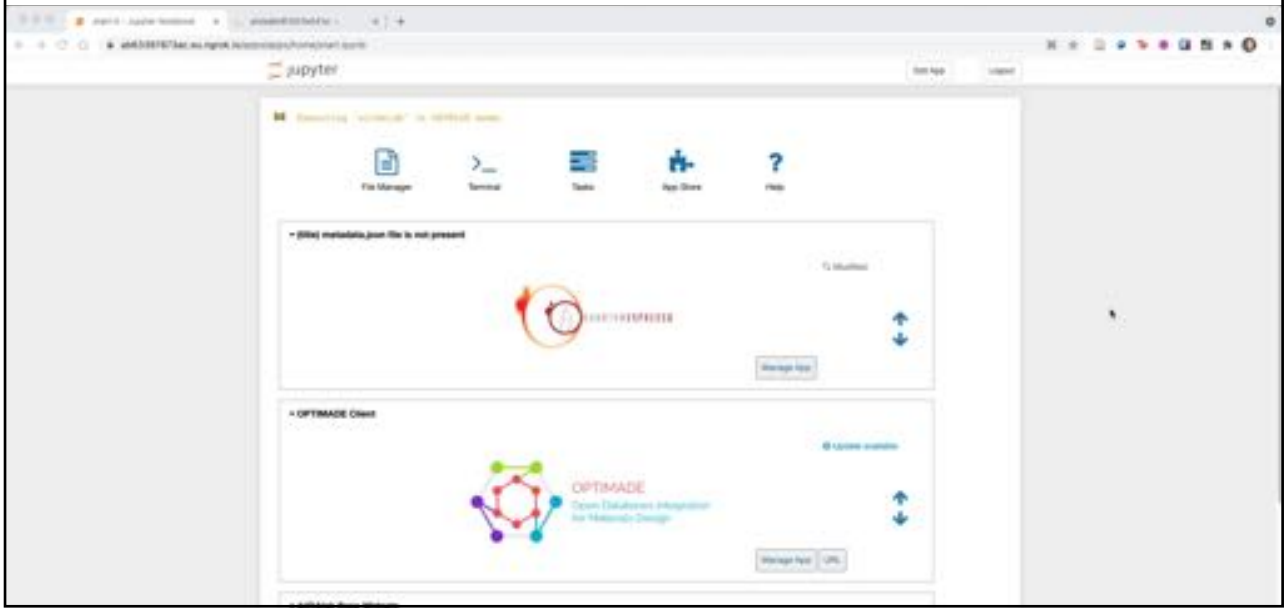


QUANTUM-AS-A-SERVICE – AiiDAlab deployment



A. V. Yakutovich et al., Comp. Mat. Sci. 188, 110165 (2021)

QUANTUM-AS-A-SERVICE – AiiDAlab deployment



SCIENCE CONCLUSIONS

There is a push **to accelerate invention and discovery in science and technology**, and especially to transform and accelerate the design and discovery of novel materials.

Key enablers:

- **the predictive accuracy of quantum simulations**
- **HPC/HTC capacity scaling**
- **the synergy of modeling and simulation with the ideas and tools of computer science**

DATA CONCLUSIONS

- I see three kind of data:
  - **Social (harvested, mostly uncontrolled conditions)**
  - **Experimental (harvested, controlled conditions)**
  - **Computational (generated, controlled conditions)**
- **Importance of data-on-demand** through reliable, robust workflows.
- **Value of data is in their reuse – curation is key, not the storage.**

FURTHER THOUGHTS ON COMPUTATIONAL SCIENCE

- **Computational science is central to the entire scientific and technological effort in the 21st century** – no other enabler can compete in terms of speed and acceleration
- **Very few have understood its structural needs, and long-term opportunity**
- As a side note, it is **a powerfully democratic force**, since it can be shared freely

ACKNOWLEDGEMENTS

ACKNOWLEDGEMENTS

MARVEL

http://nccr-marvel.ch
Swiss National Centre for Computational Design and Discovery of Novel Materials (2014-18, 2018-22, 2022-26)

MAX DRIVING THE EXASCALE TRANSITION

http://max-centre.eu
H2020 Centre of Excellence MaX: Materials Design at the Exascale (2015-18, 2018-22, …)

BIG-MAP

https://www.big-map.eu
H2020 Battery Interface Genome – Materials Acceleration Platform (Battery 2030+) (2020-23, …)

Universität Bremen

https://www.uni-bremen.de/mapex
U Bremen Excellence Chair (2018-25, …)

Related projects:
H2020 Nanoscience Foundries and Fine Analysis
H2020 European Materials Modelling Council
H2020 Marketplace
H2020 Intersect
H2020 DOME 4.0
H2020 OpenModel
H2020 NEP
H2020 EPFL Fellows
H2020 EPFL Innovators
H2020 Marie Curie
EPFL Open Science
PASC
PRACE
IBM
Constellium
Innosuisse
Solvay
Samsung
Richemont Varinor

"Things were done very differently on the farm when I was your age, Kenny."