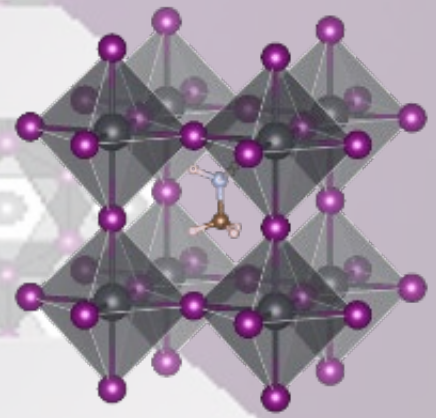
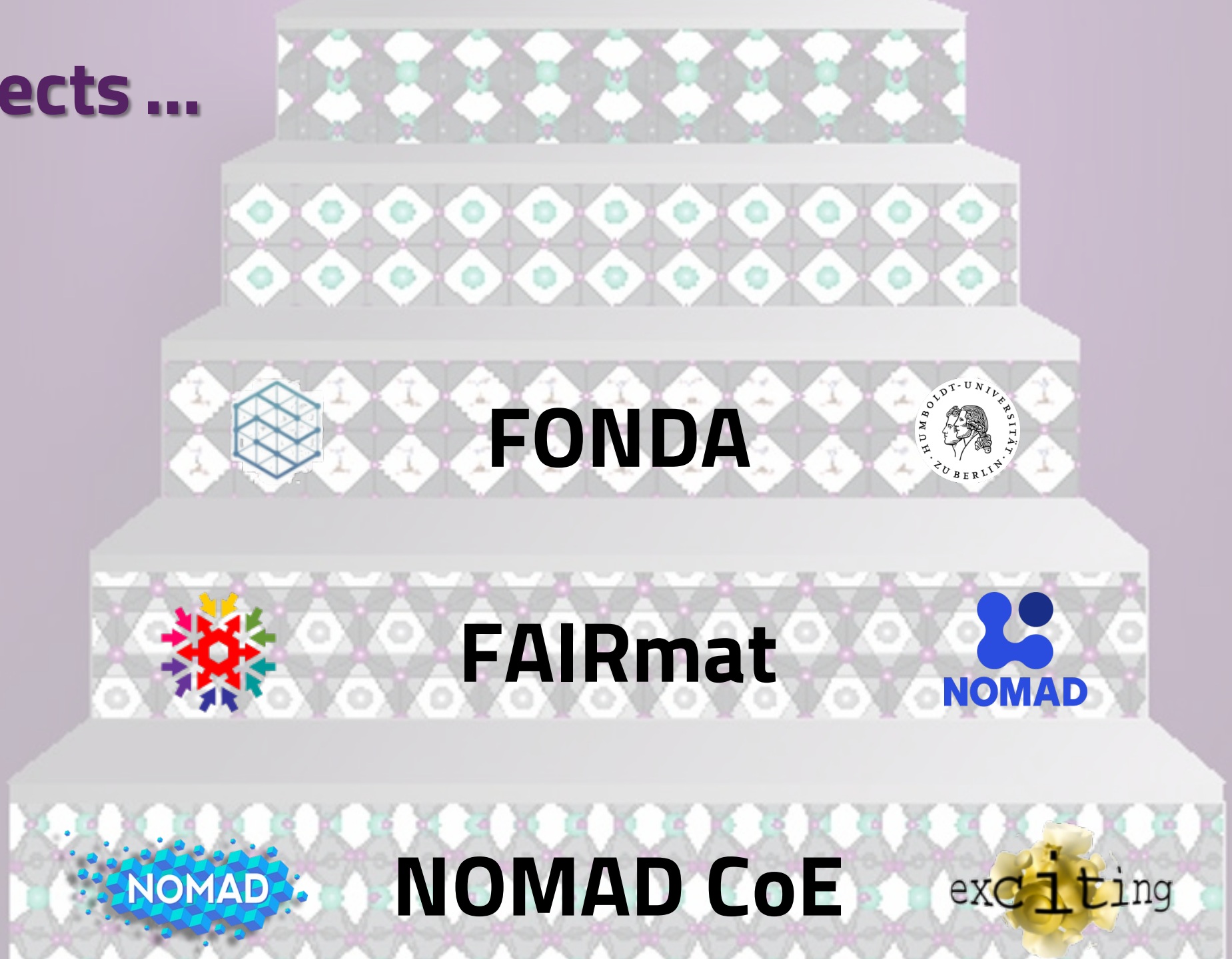
Claudia Draxl

High-throughput spectroscopy and materials discovery by beyond-DFT workflows and data-analysis frameworks



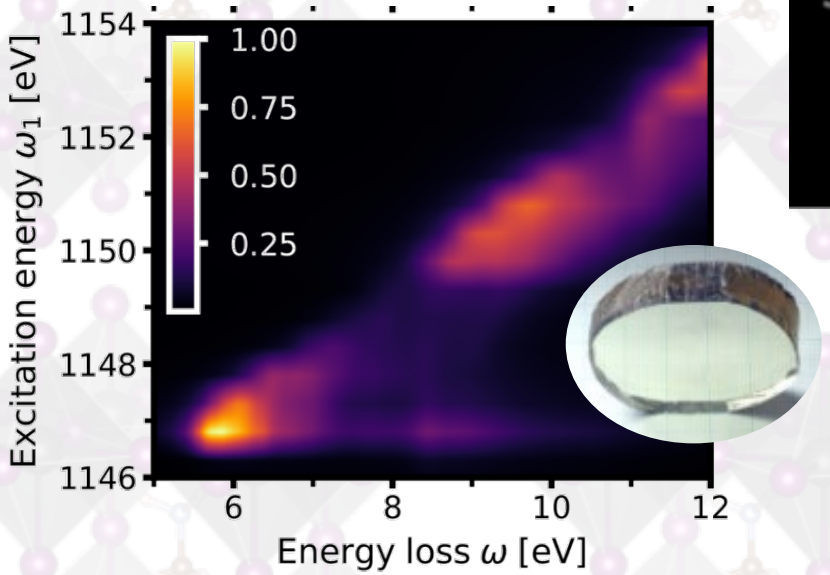
3 projects ...

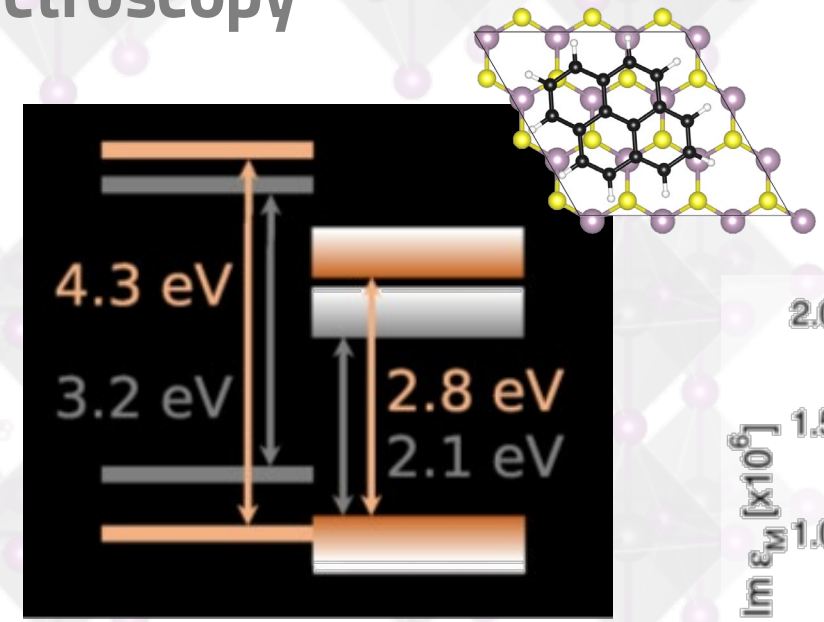


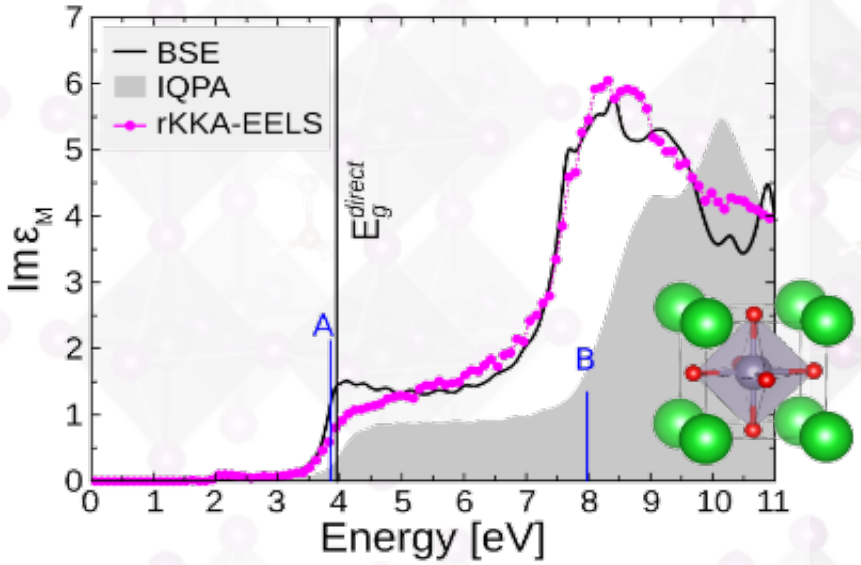
My research field

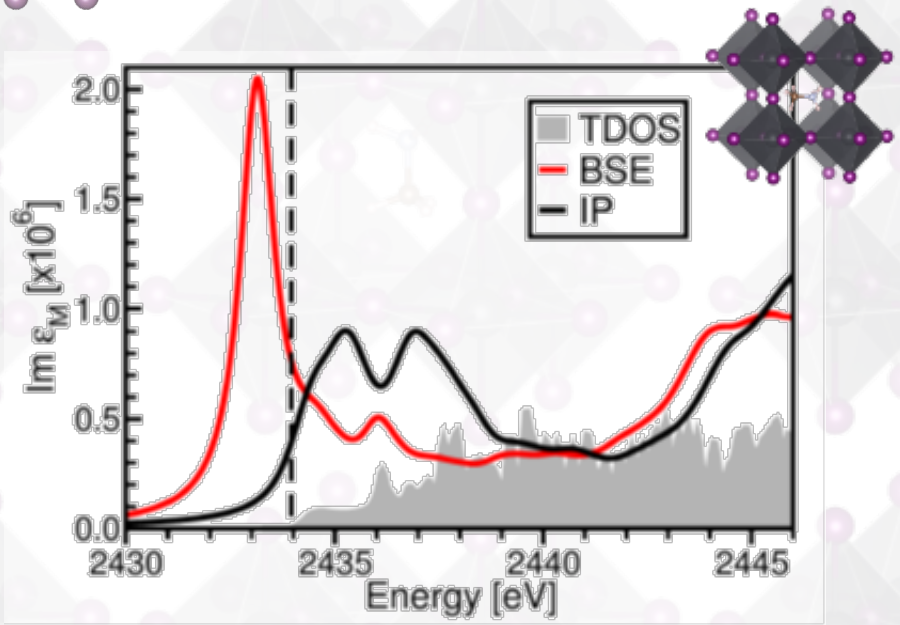
Computational materials science

Focus on theoretical spectroscopy









... and why do I care about workflows?

3 projects ...

NOMAD

NOMAD CoE

exciting

What limits us?

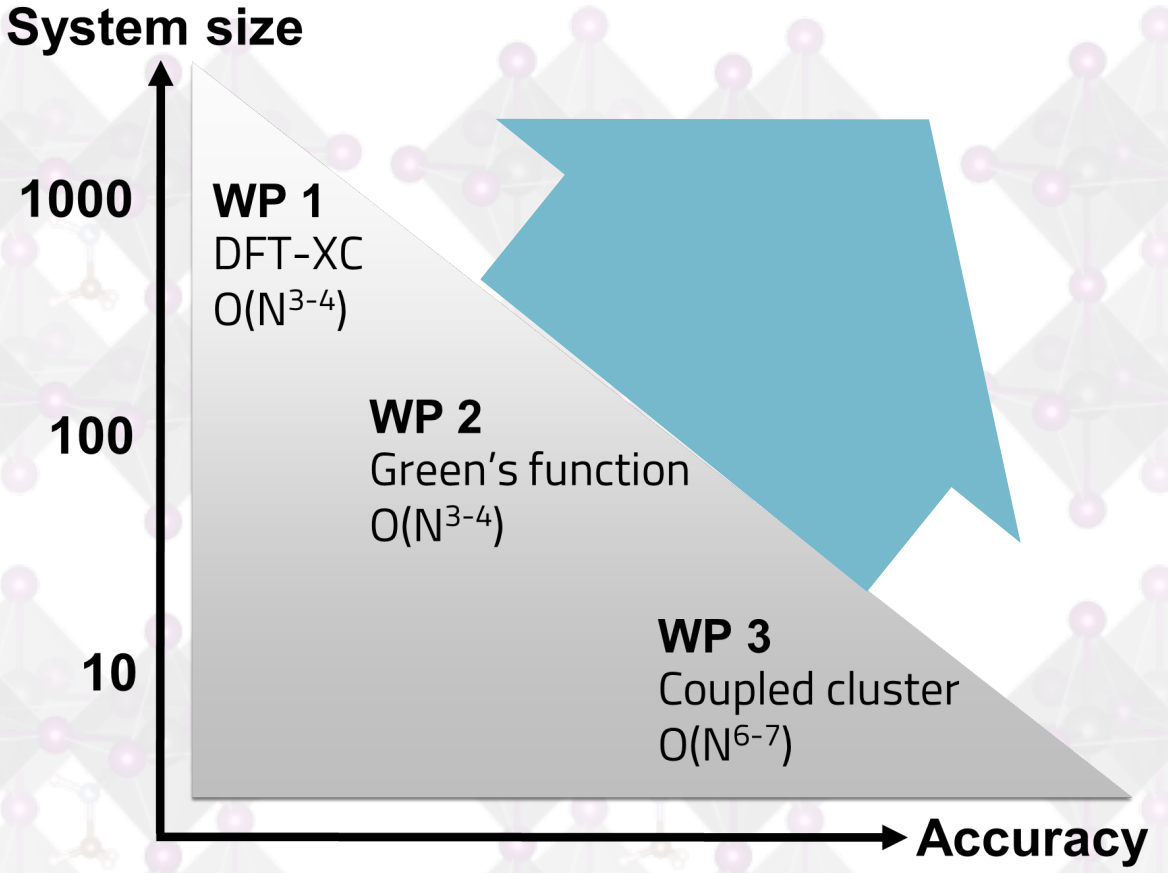
Many materials / properties require methods better than semi-local DFT

Basically everything relevant for e.g. energy research

Complex systems require larger simulation cells

1 atom out of thousand(s) may determine the color

... and/or better methodology



Exascale codes

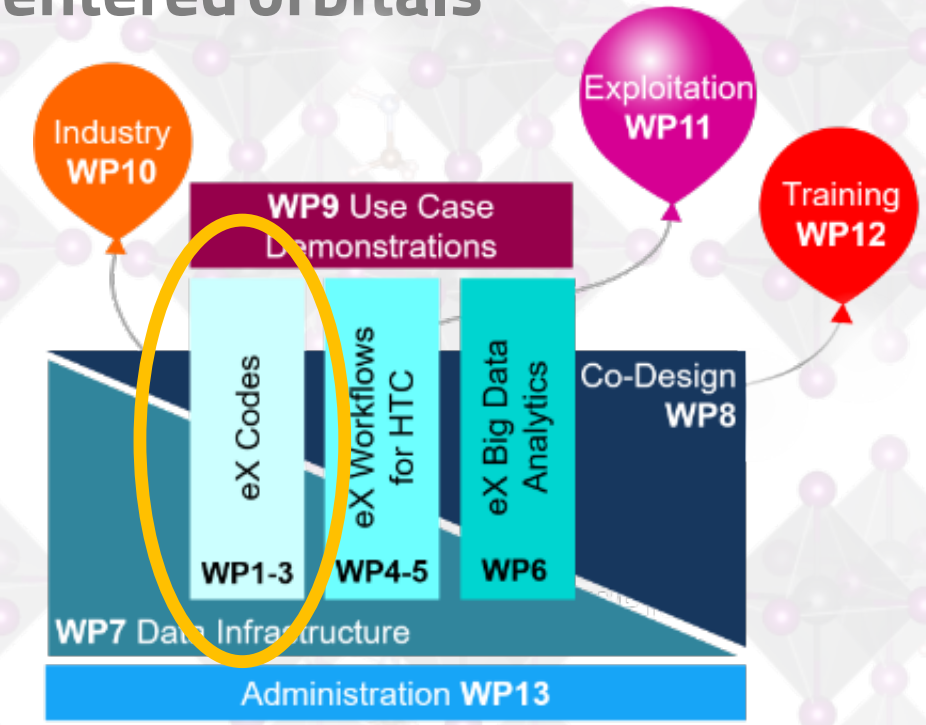
Bring DFT, Green-function based methods, and coupled-cluster theory to exascale

Develop libraries for code families

Covering planewaves, LAPW's, numerical atom-centered orbitals

All-electron & pseudopotentials

Demonstrator codes

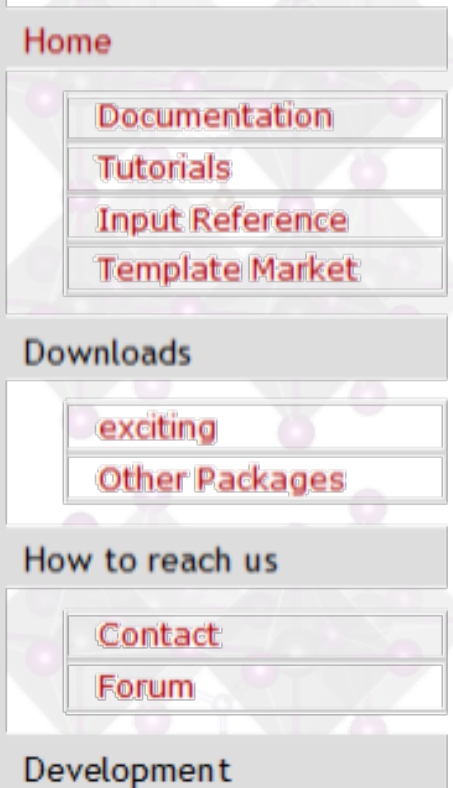


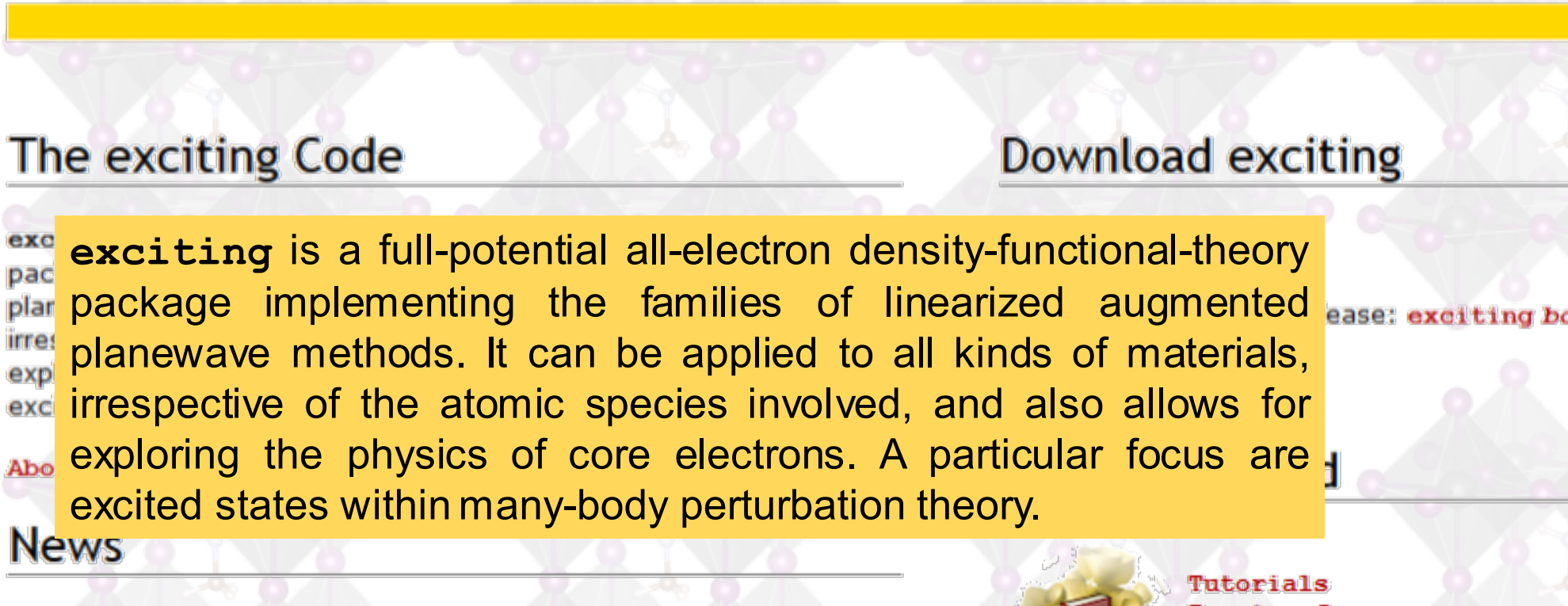
Our instrument

http://exciting-code.org

A. Gulans, S. Kontur, C. Meisenbichler, D. Nabok, P. Pavone, S. Rigamonti, S. Sagmeister, U. Werner, and CD, J. Phys: Condens. Matter 26, 363202 (2014).

C. Vorwerk, B. Aurich, C. Cocchi, and C. Draxl, Electronic Structure, 1, 037001 (2019).

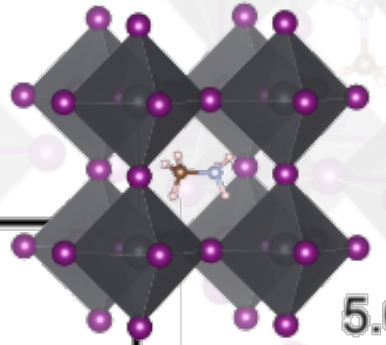




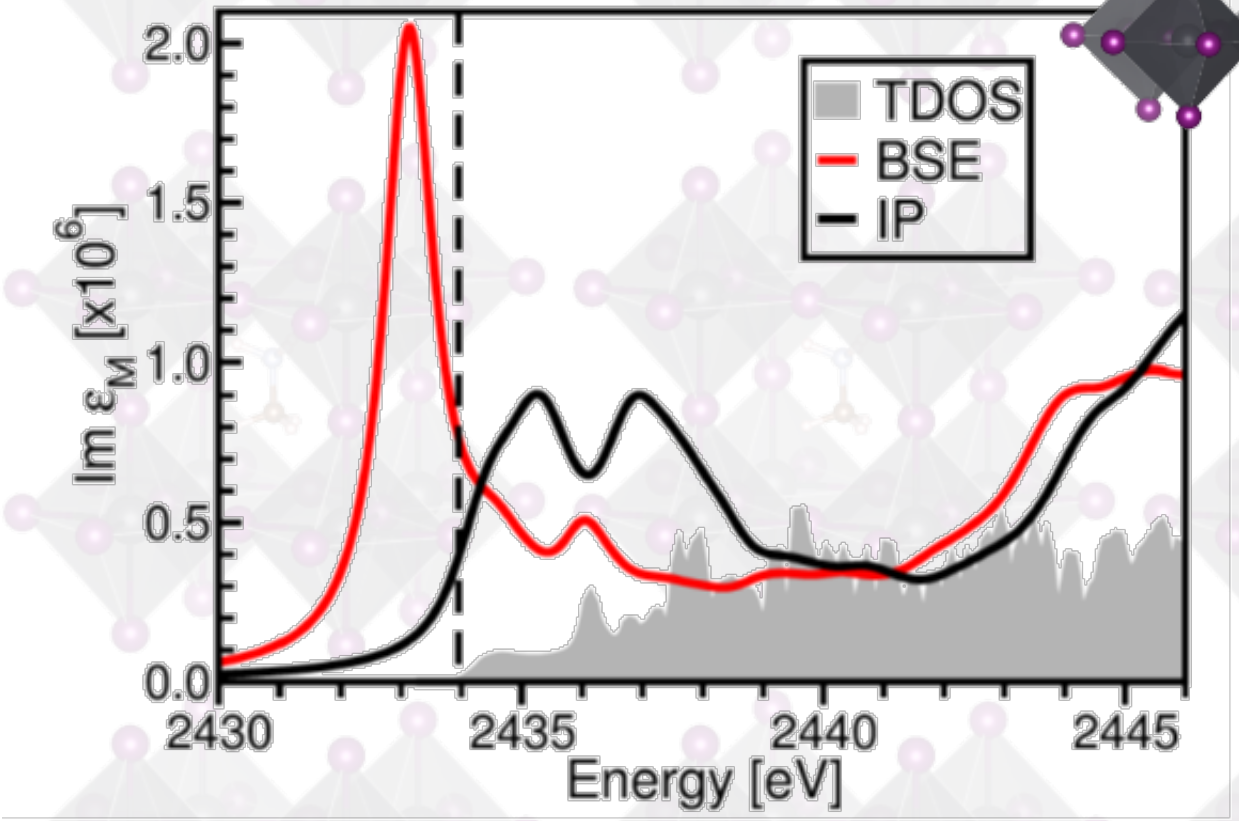
exciting is a full-potential all-electron density-functional-theory package implementing the families of linearized augmented planewave methods. It can be applied to all kinds of materials, irrespective of the atomic species involved, and also allows for exploring the physics of core electrons. A particular focus are excited states within many-body perturbation theory.

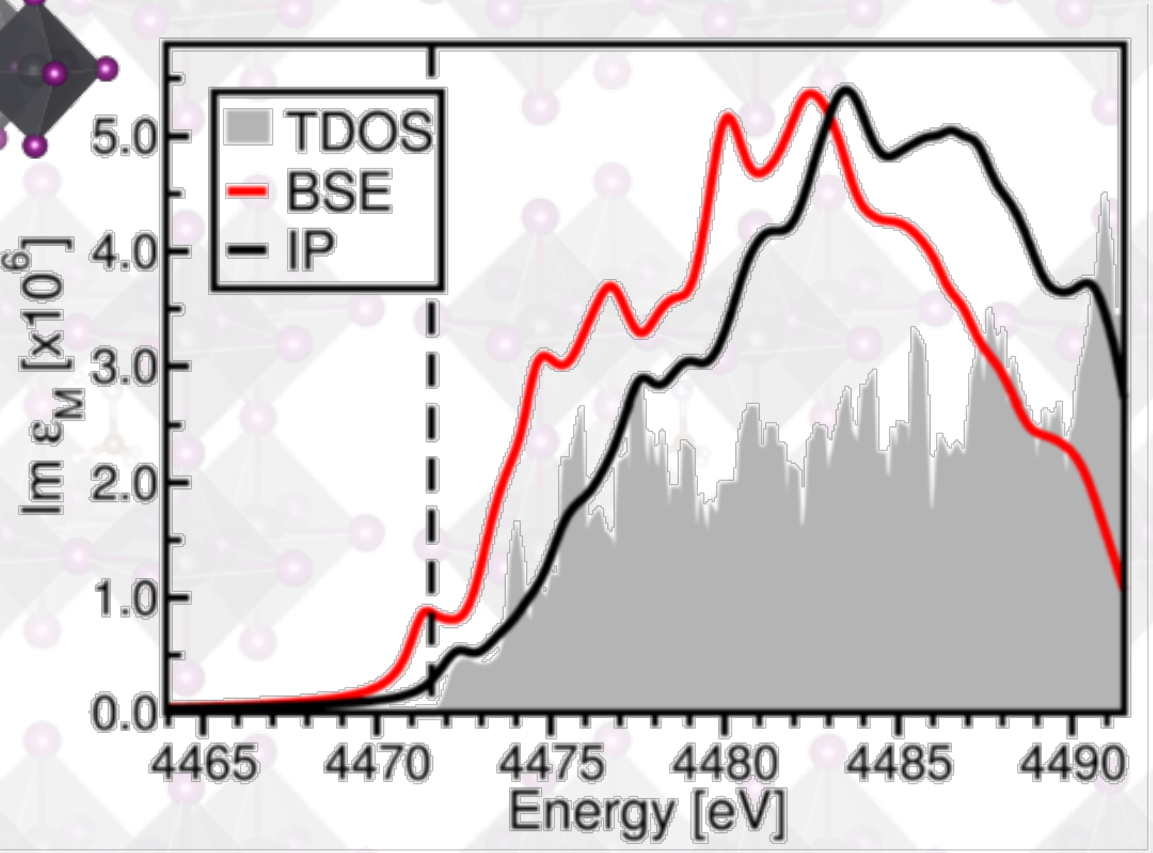
Example: core-level spectra in $MAPbI_3$

Pb M_5 edge



I L_3 edge





C. Vorwerk, C. Hartmann, C. Cocchi, G. Sadoughi, S. Habisreutinger, R. Felix, R. Wilks, H.J. Snaith, M. Bär & CD, JCPL 9, 1852 (2018).

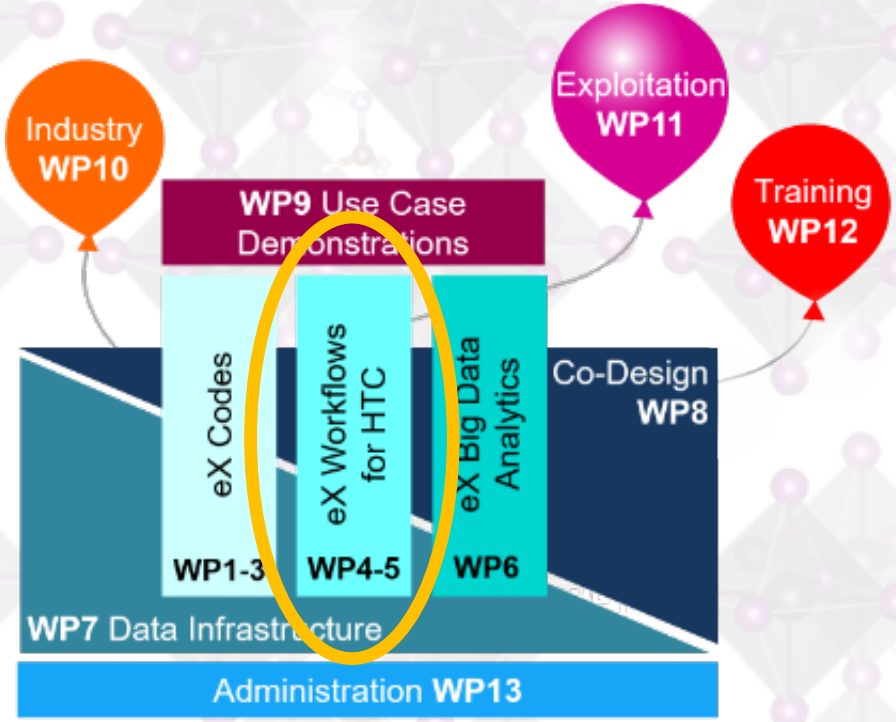
Exascale workflows

Serve as a glue

- Controling computational parameters
  - Way more parameters in advanced methodology
- Combining tasks / ‘recipes‘
  - To be executed either in parallel or consecutively
- Handling jobs
  - To guarantee error-free or error-corrected execution
- Collecting results

Are

- Method-specific
- Largely code-specific



Example: high-throughput absorption spectra

Bethe-Salpeter equation

$$\sum_{v'c'\mathbf{k}'} H^{e-h}_{vc\mathbf{k},v'c'\mathbf{k}'} A^{\lambda}_{v'c'\mathbf{k}'} = E_\lambda A^{\lambda}_{vc\mathbf{k}}$$

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Bethe-Salpeter equation

$$\sum_{v'c'\mathbf{k}'} \hat{H}_{vc\mathbf{k},v'c'\mathbf{k}'} A^{\lambda}_{v'c'\mathbf{k}'} = E^{\lambda} A^{\lambda}_{vc\mathbf{k}}$$

Exciton wavefunction

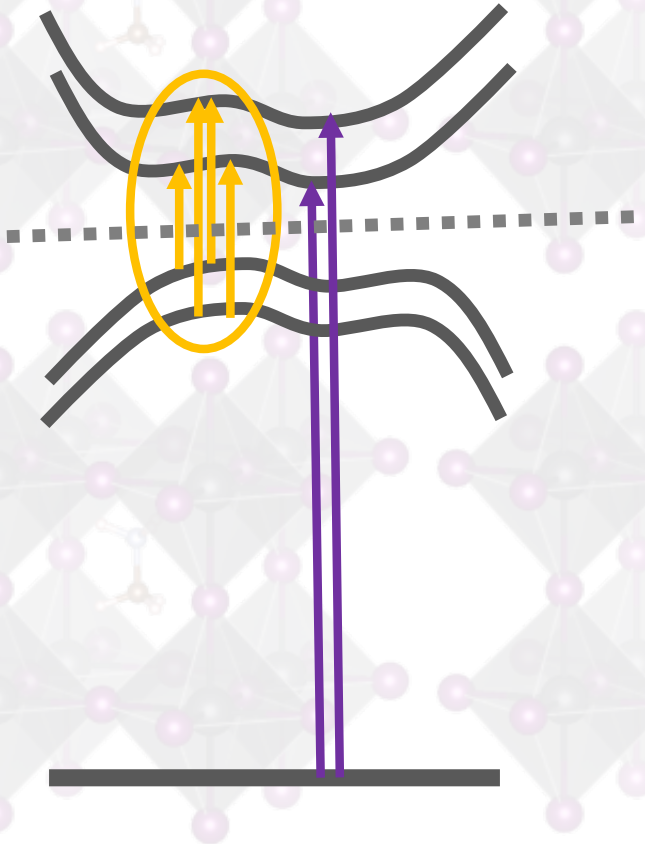
Optical excitations

valence conduction

$$\phi^{\lambda}(\mathbf{r}_e, \mathbf{r}_h) = \sum_{vc\mathbf{k}} A^{\lambda}_{vc\mathbf{k}} \psi^{*}_{v\mathbf{k}}(\mathbf{r}_h) \psi_{c\mathbf{k}}(\mathbf{r}_e)$$

Core excitations

core conduction



Spectra

$$\mathrm{Im}\,\varepsilon_M(\omega) = \frac{8\pi^2}{\Omega} \sum_{\lambda} \left| \sum_{vc\mathbf{k}} A^{\lambda}_{vc\mathbf{k}} \frac{\langle v\mathbf{k}|\hat{\mathbf{p}}|c\mathbf{k}\rangle}{\epsilon_{c\mathbf{k}} - \epsilon_{v\mathbf{k}}} \right|^2 \delta(\omega - E_{\lambda})$$

High-throughput absorption spectra ...

Bethe-Salpeter equation

$$\sum_{v'c'\mathbf{k}'} H^{e-h}_{vc\mathbf{k},v'c'\mathbf{k}'} A^{\lambda}_{v'c'\mathbf{k}'} = E_\lambda A^{\lambda}_{vc\mathbf{k}}$$

$$H^{\text{diag}}_{vc\mathbf{k},v'c'\mathbf{k}'} = (\varepsilon_{c\mathbf{k}} - \varepsilon_{v\mathbf{k}})\delta_{vv'}\delta_{cc'}\delta_{\mathbf{k}\mathbf{k}'}$$

$$H^{\text{dir}}_{cv\mathbf{k},c'v'\mathbf{k}'} = \int \mathrm{d}^3r\,\mathrm{d}^3r' \frac{\psi_{v\mathbf{k}}(\mathbf{r})\psi^*_{c\mathbf{k}}(\mathbf{r}')\epsilon^{-1}(\mathbf{r},\mathbf{r}')\psi^*_{v'\mathbf{k}'}(\mathbf{r})\psi_{c'\mathbf{k}'}(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}$$

$$H^{\times}_{vc\mathbf{k},v'c'\mathbf{k}'} = \int \mathrm{d}^3r\,\mathrm{d}^3r'\,\psi_{v\mathbf{k}}(\mathbf{r})\psi^*_{c\mathbf{k}}(\mathbf{r})\,\bar{v}(\mathbf{r},\mathbf{r}')\,\psi^*_{v'\mathbf{k}'}(\mathbf{r}')\psi_{c'\mathbf{k}'}(\mathbf{r}')$$

Bethe-Salpeter equation

$$\sum_{v'c'\mathbf{k}'} \hat{H}_{vc\mathbf{k},v'c'\mathbf{k}'} A^{\lambda}_{v'c'\mathbf{k}'} = E^{\lambda} A^{\lambda}_{vc\mathbf{k}}$$

Ex

O

C

Sp

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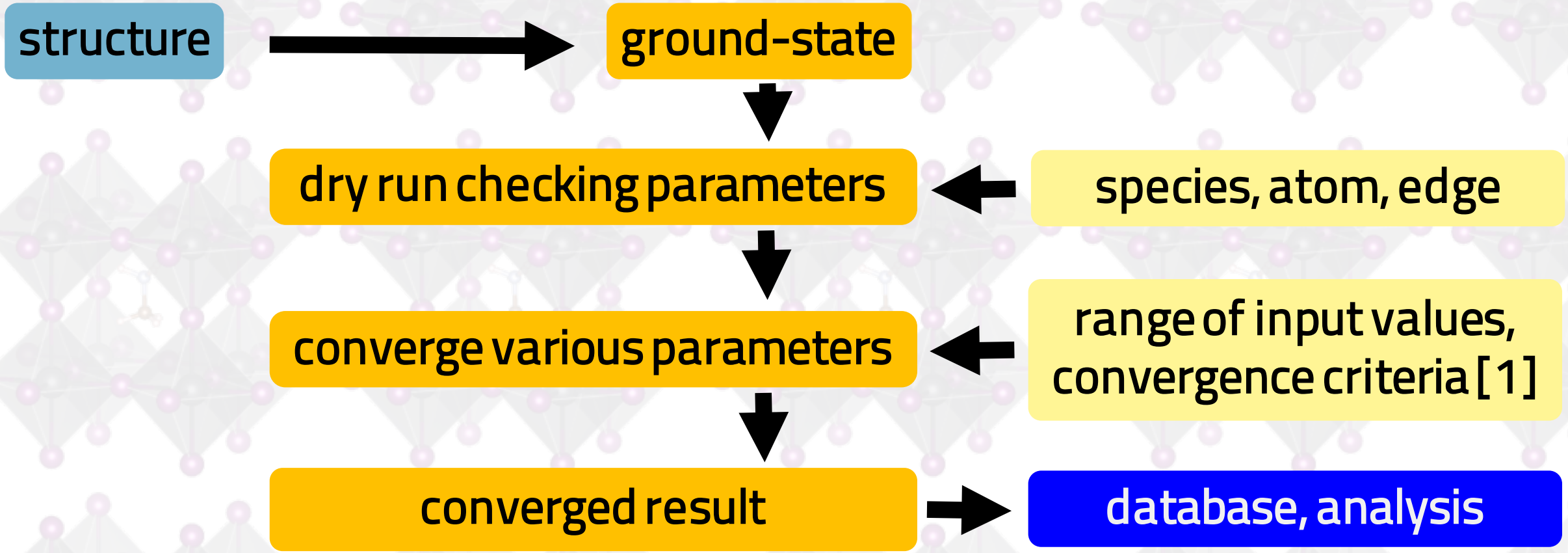
Workflow for converging (core) spectra



[1] M. Kuban et al., Sci Data 9, 646 (2022).

excitingtools

Parser for over 50 `exciting` output files

+ tools for post-processing

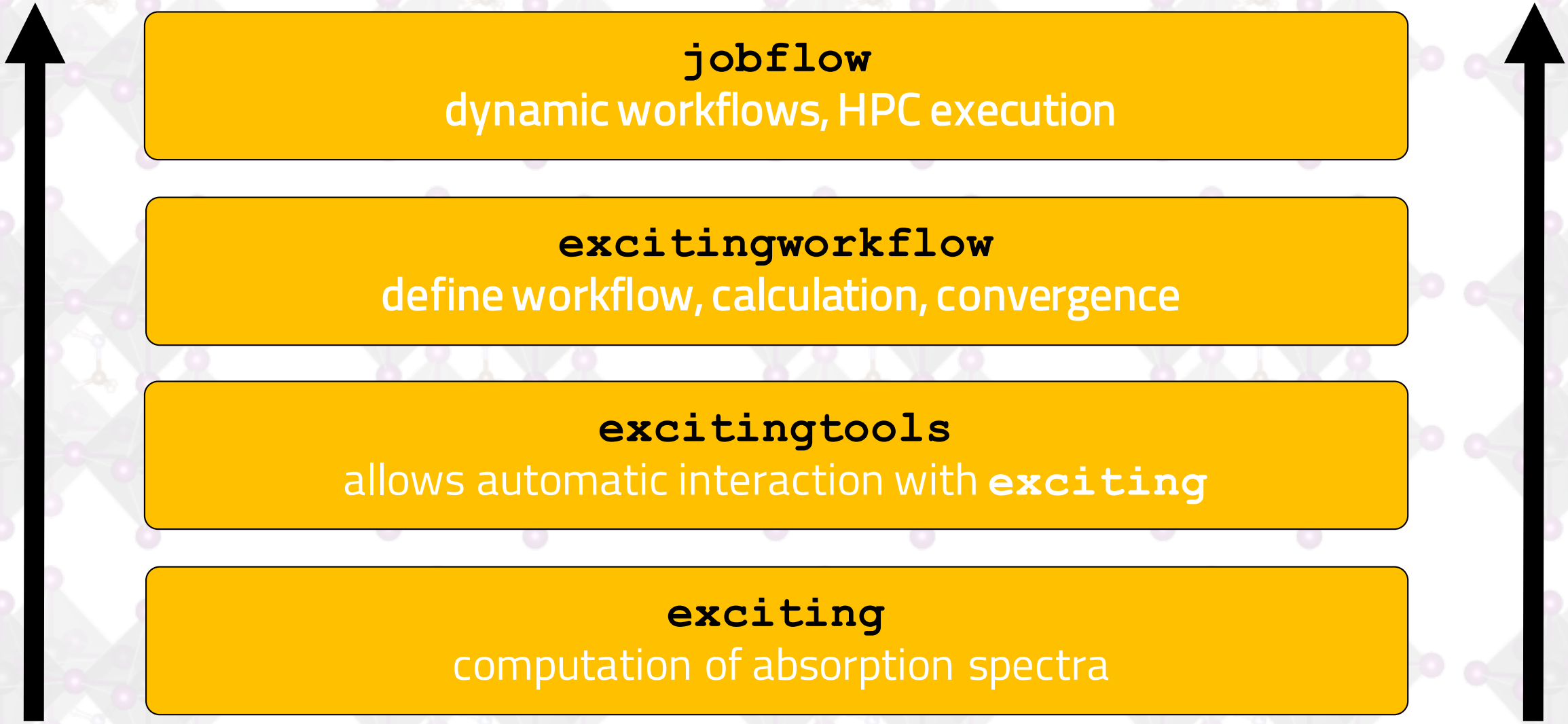
Utilities for constructing `exciting` input files

Easy integration with workflow managers

https://pypi.org/project/excitingtools/

https://github.com/exciting/excitingtools/

Code pieces


jobflow
dynamic workflows, HPC execution
excitingworkflow
define workflow, calculation, convergence
excitingtools
allows automatic interaction with exciting
exciting
computation of absorption spectra

XANES benchmark project

Probing 3 different codes

Multi-code Benchmark on Simulated Ti K-edge X-ray Absorption Spectra of Ti-O Compounds

Fanchen Meng,[1] Benedikt Maurer,[2] Fabian Peschel,[2] Sencer Selcuk,[1] Mark Hybertsen,[1] Xiaohui Qu,[1] Christian Vorwerk,[3,*] Claudia Draxl,[2,†] John Vinson,[4,‡] and Deyu Lu[1,§]

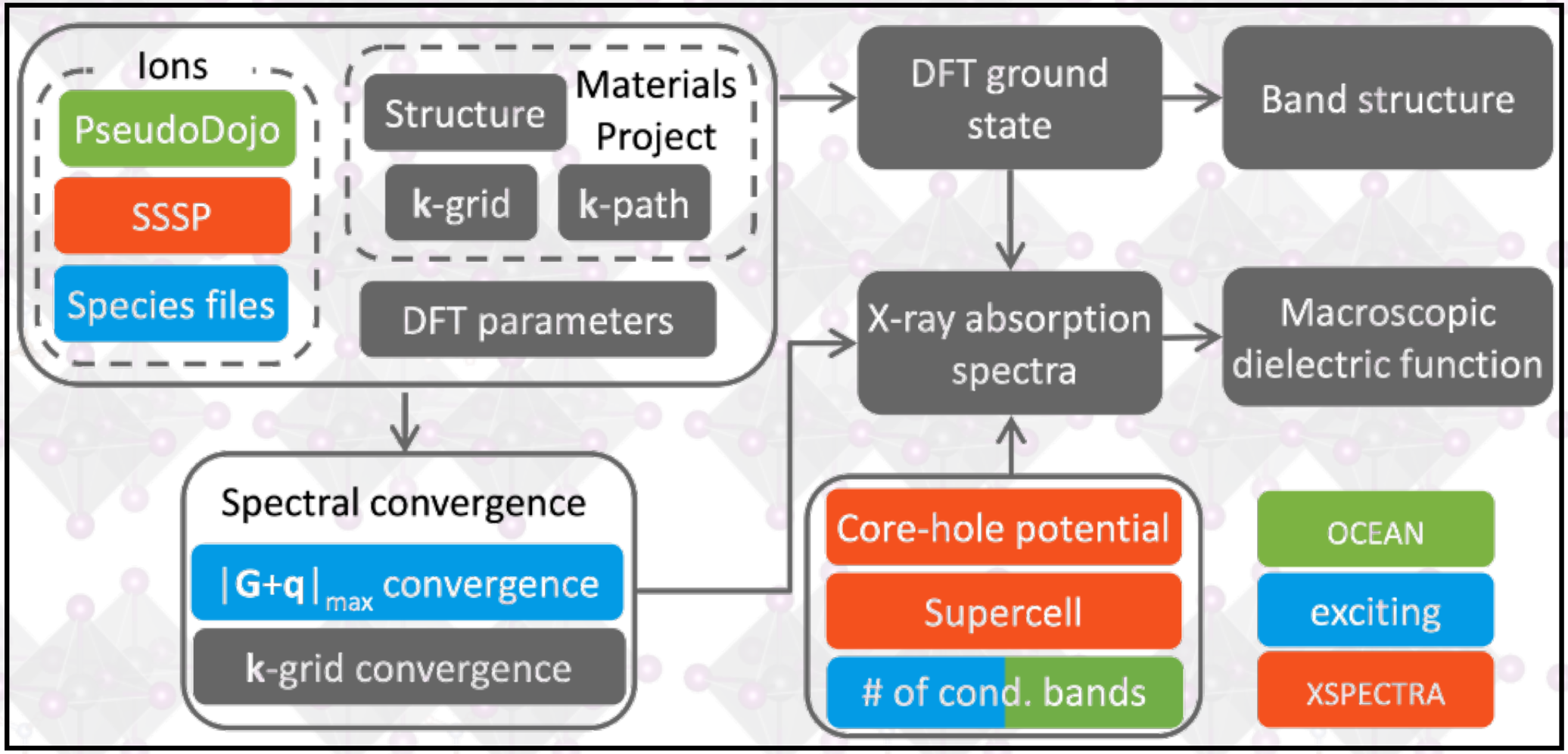
[1] *Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, New York 11973, United States*
[2] *Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, Berlin Germany*
[3] *Pritzker School of Molecular Engineering, University of Chicago, Chicago, Illinois 60637, United States*
[4] *Material Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, United States*

(Dated: March 15, 2023)

https://arxiv.org/abs/2303.17089

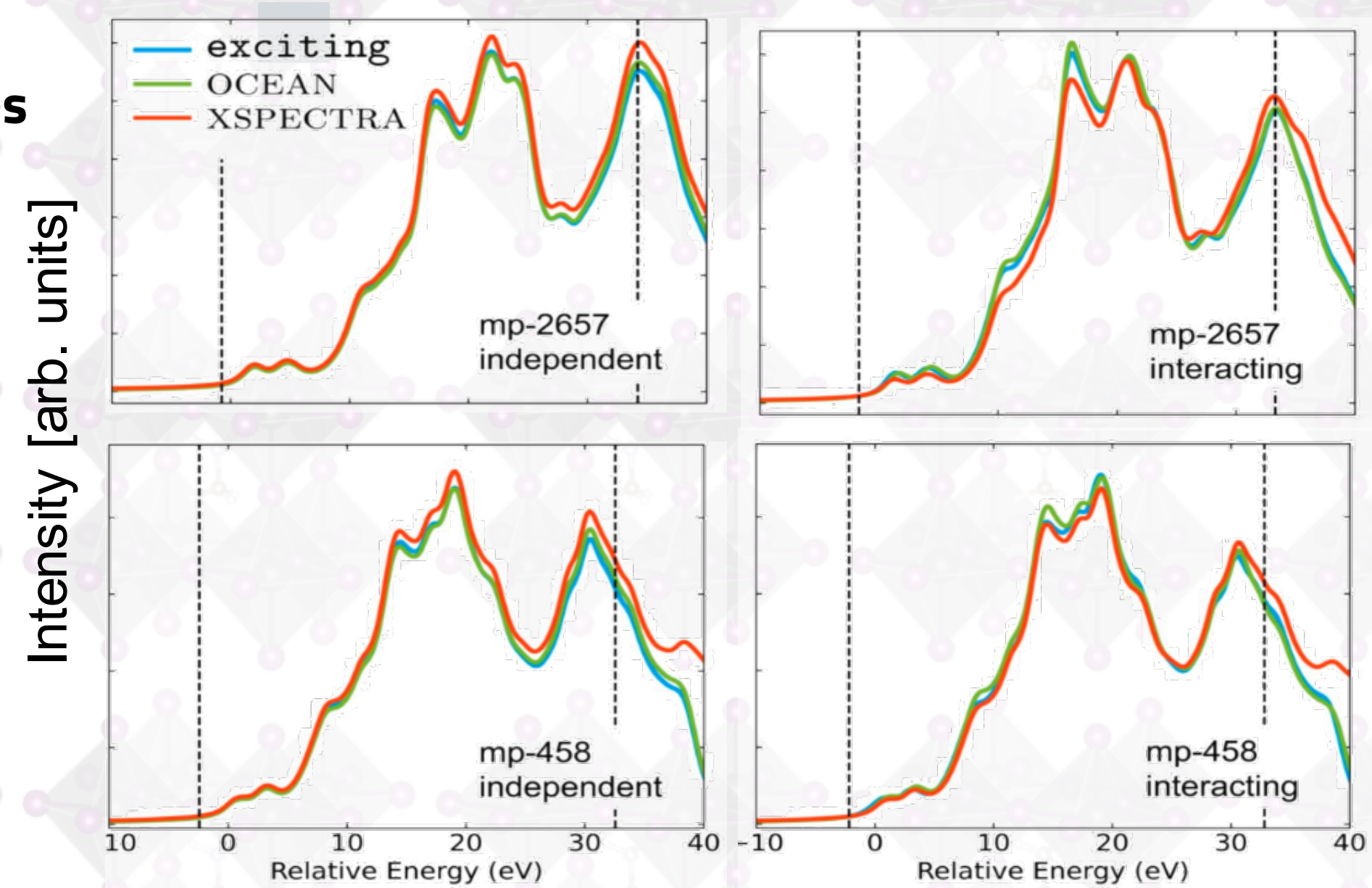
XANES benchmark workflow

Workflow combining 3 different codes



Ti K edge

Ti-O structures



XANES code: Lightshow

Tool serving 3 different codes

Lightshow: a Python package for generating computational x-ray absorption spectroscopy input files

Matthew R. Carbone,[1,*] Fanchen Meng,[2,†] Christian Vorwerk,[3] Benedikt Maurer,[4] Fabian Peschel,[4] Xiaohui Qu,[2] Eli Stavitski,[5] Claudia Draxl,[4] John Vinson,[6] and Deyu Lu[2]

[1] *Computational Science Initiative, Brookhaven National Laboratory, Upton, New York 11973, United States*
[2] *Center for Functional Nanomaterials, Brookhaven National Laboratory, Upton, New York 11973, United States*
[3] *Pritzker School of Molecular Engineering, University of Chicago, Chicago, Illinois 60637, United States*
[4] *Physics Department and IRIS Adlershof, Humboldt-Universität zu Berlin, D-12489 Berlin, Germany*
[5] *National Synchrotron Light Source II, Brookhaven National Laboratory, Upton, New York 11973, United States*
[6] *Material Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, United States*

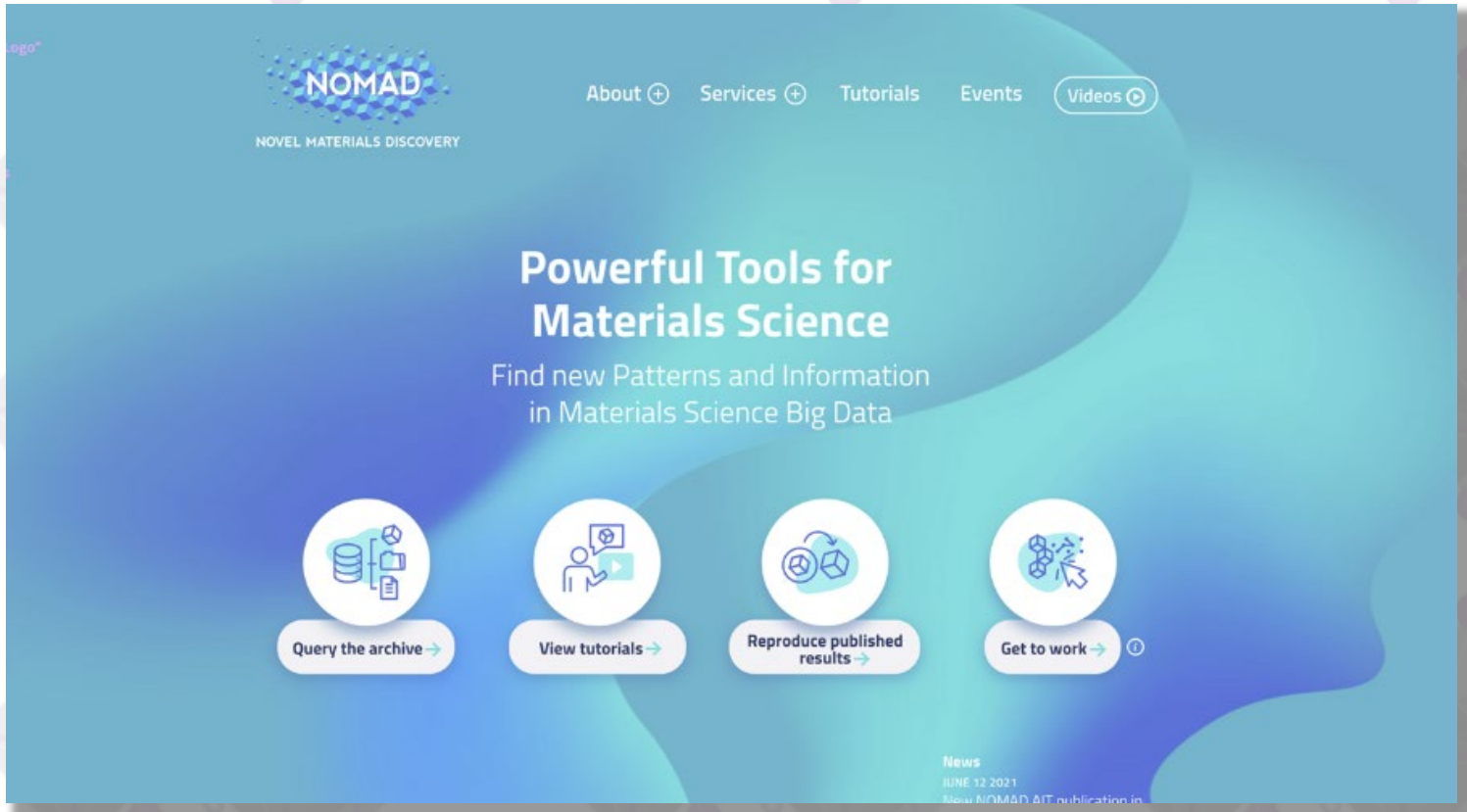
(Dated: November 9, 2022)

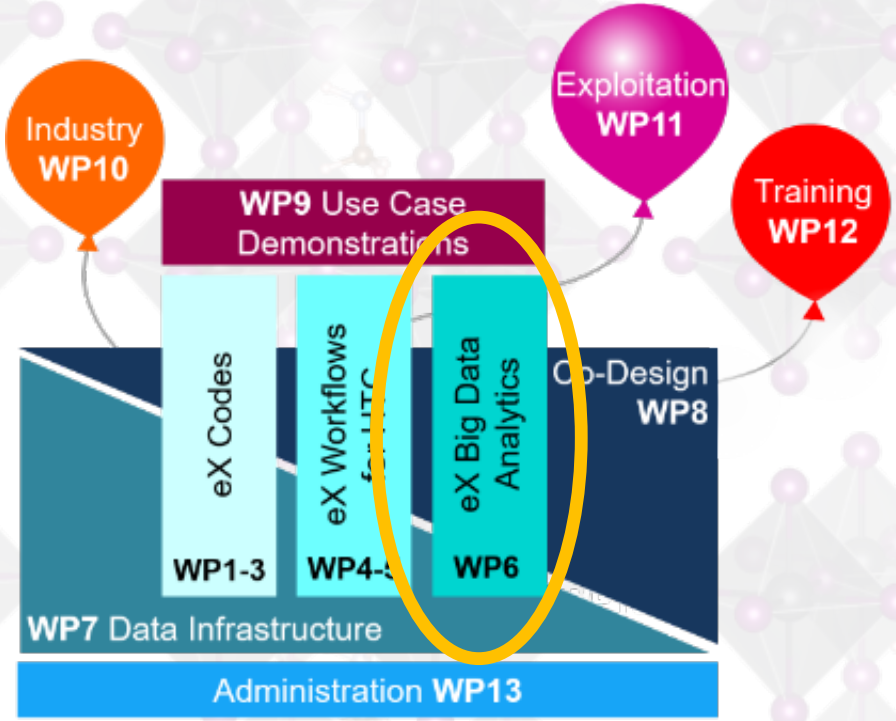
https://arxiv.org/abs/2211.04452

Extreme-scale data

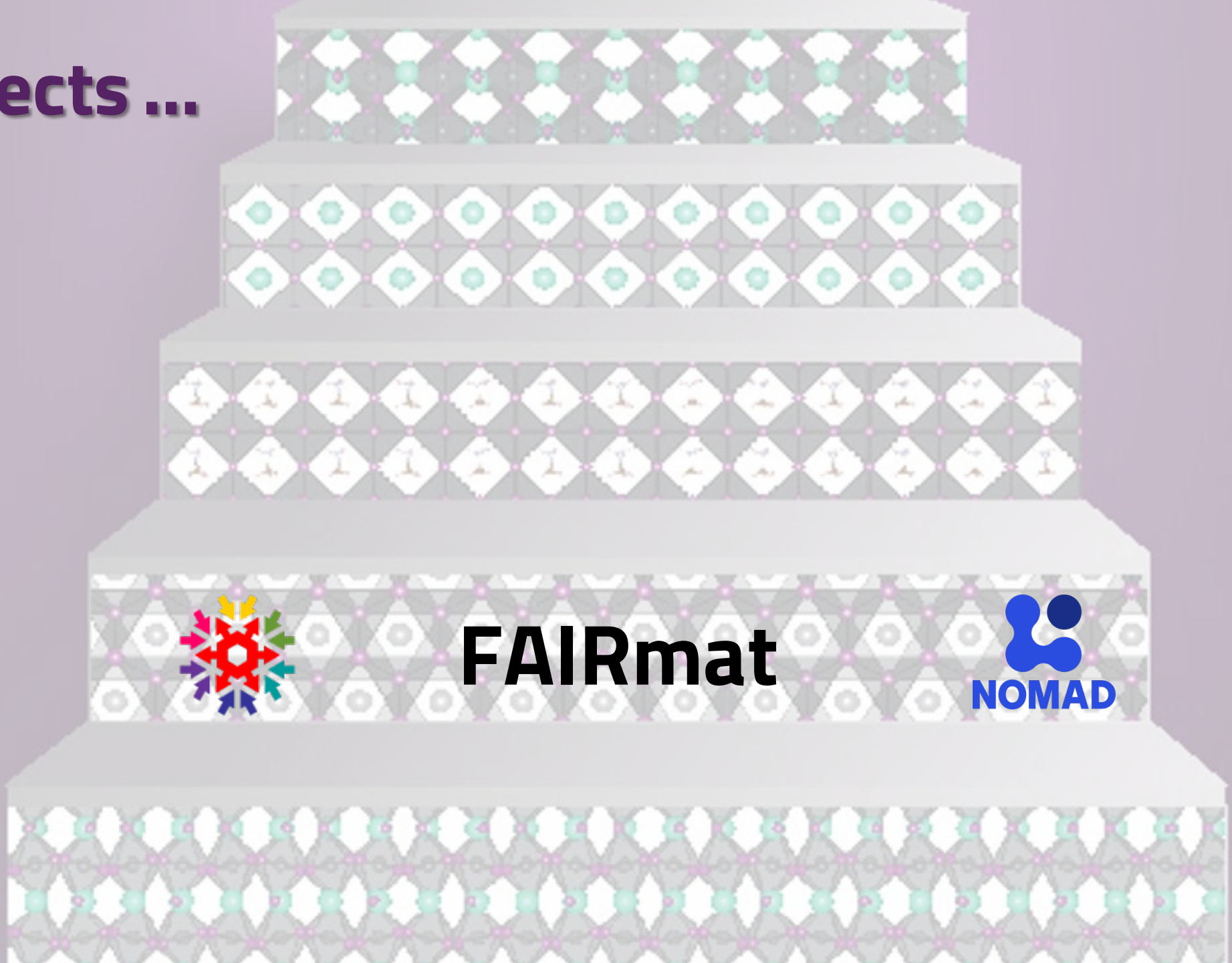
Artificial-Intelligence Toolkit

Brought to "real-time" performance





3 projects ...



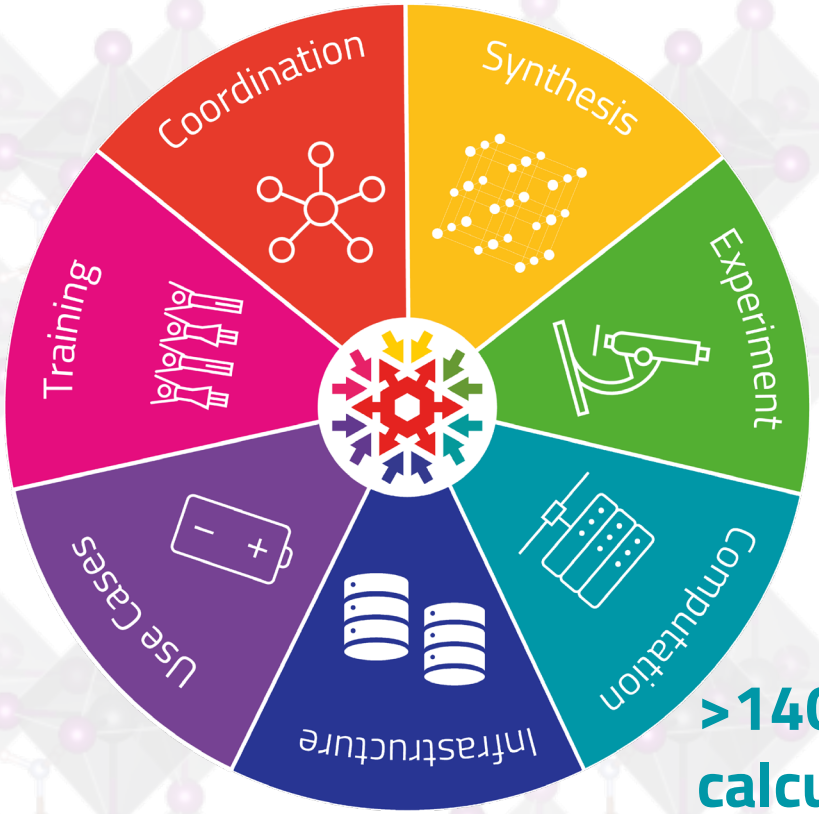
FAIRmat

NOMAD

FAIRmat / NOMAD data infrastructure

FAIRmat is the NFDI consortium for solid state physics and the chemical physics of solids

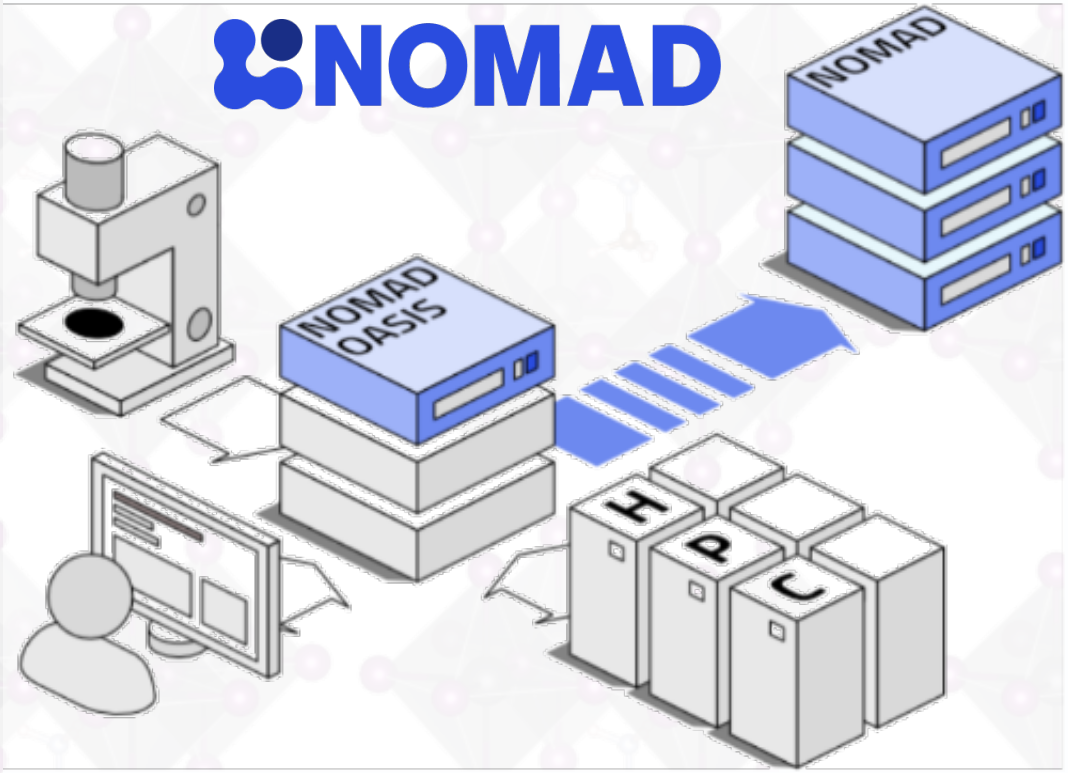
https://fairmat-nfdi.eu



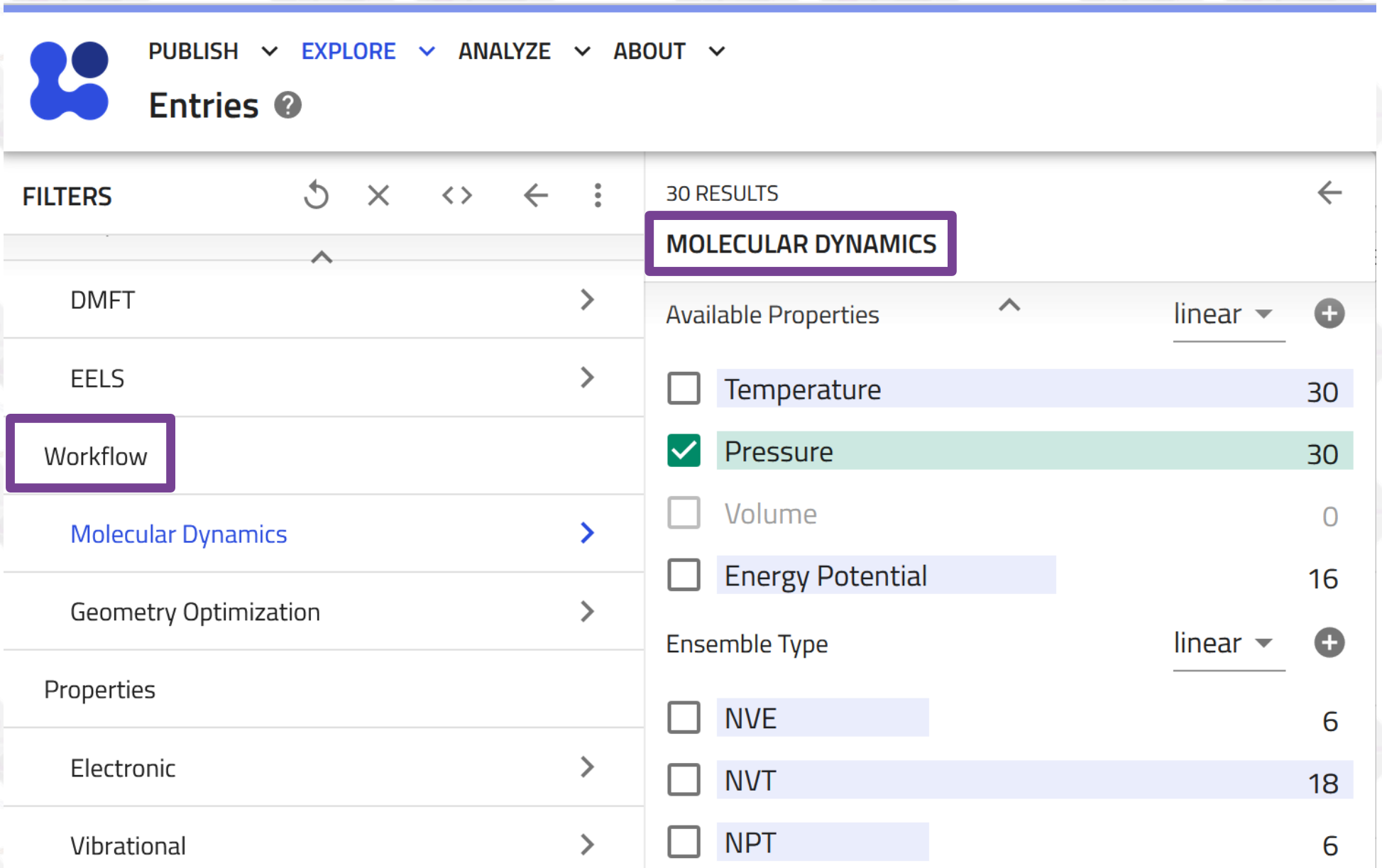
>140 mio calculations

NOMAD is a web-based software for FAIR research data management in materials science

https://nomad-lab.eu



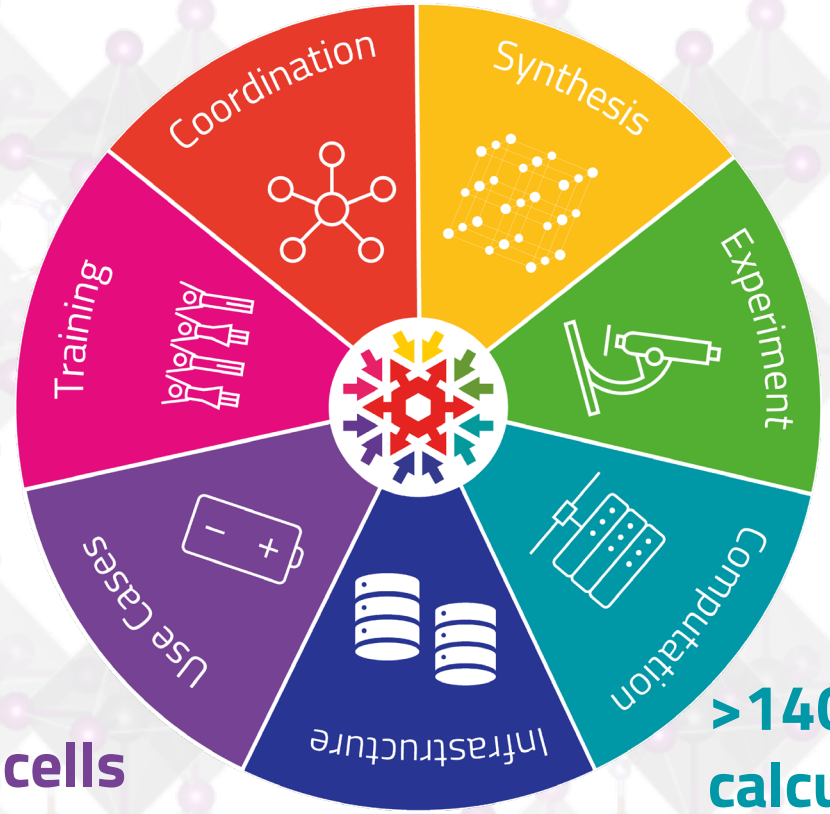
Workflows in NOMAD



FAIRmat / NOMAD data infrastructure

FAIRmat is the NFDI consortium for solid state physics and the chemical physics of solids

https://fairmat-nfdi.eu

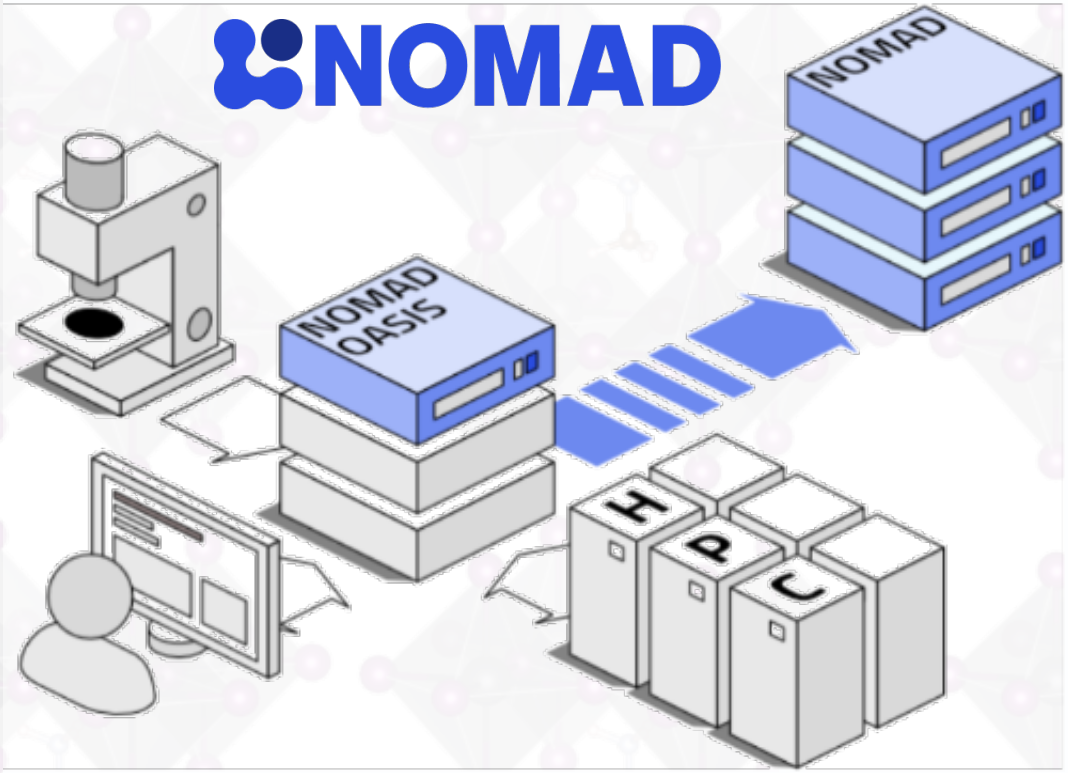


Solar cells

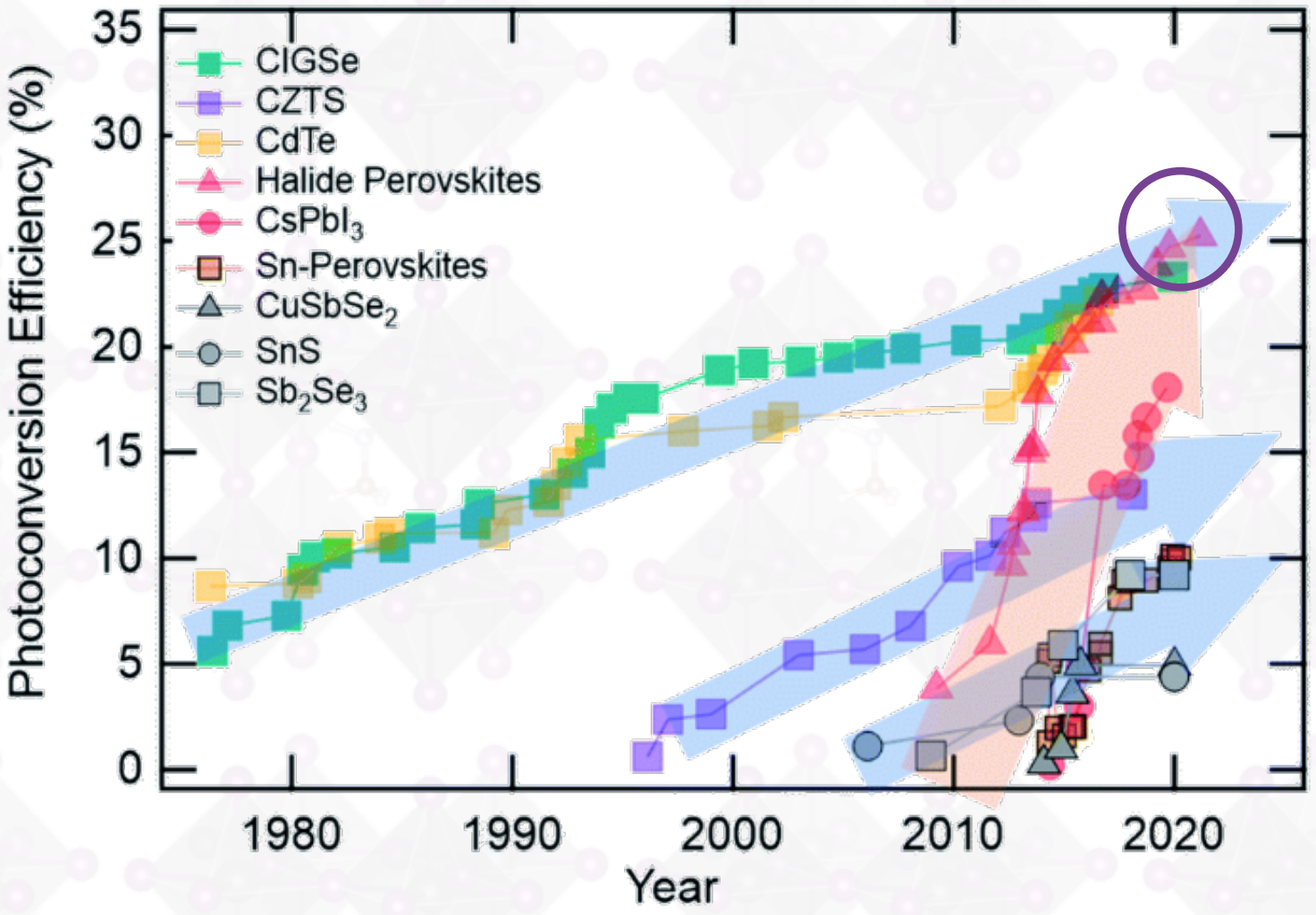
>140 mio calculations

NOMAD is a web-based software for FAIR research data management in materials science

https://nomad-lab.eu



A long way to the roof …



T. Unold, Faraday Discussions 239, 235 (2022).

Classical solar-cell research

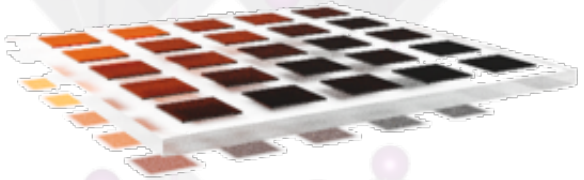
SLOW

Literature

SLOW

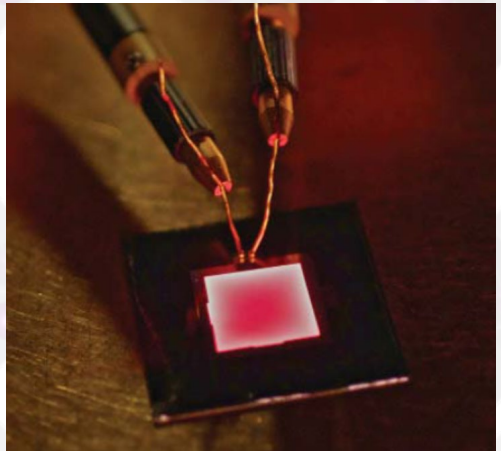
SLOW

Synthesis

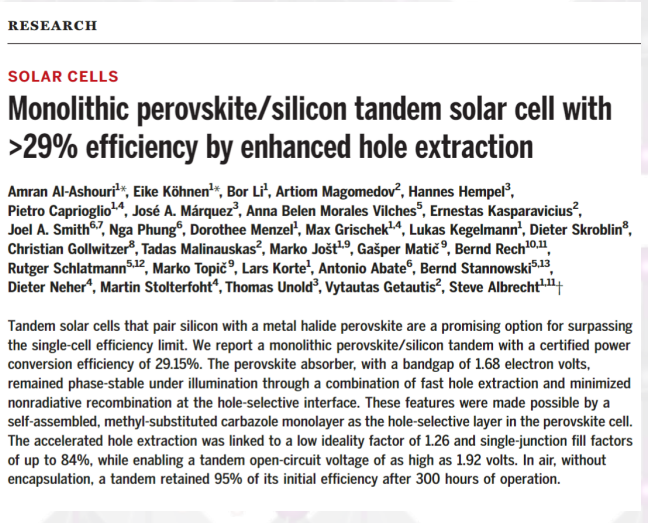


Characterization

Device



Publication



RESEARCH

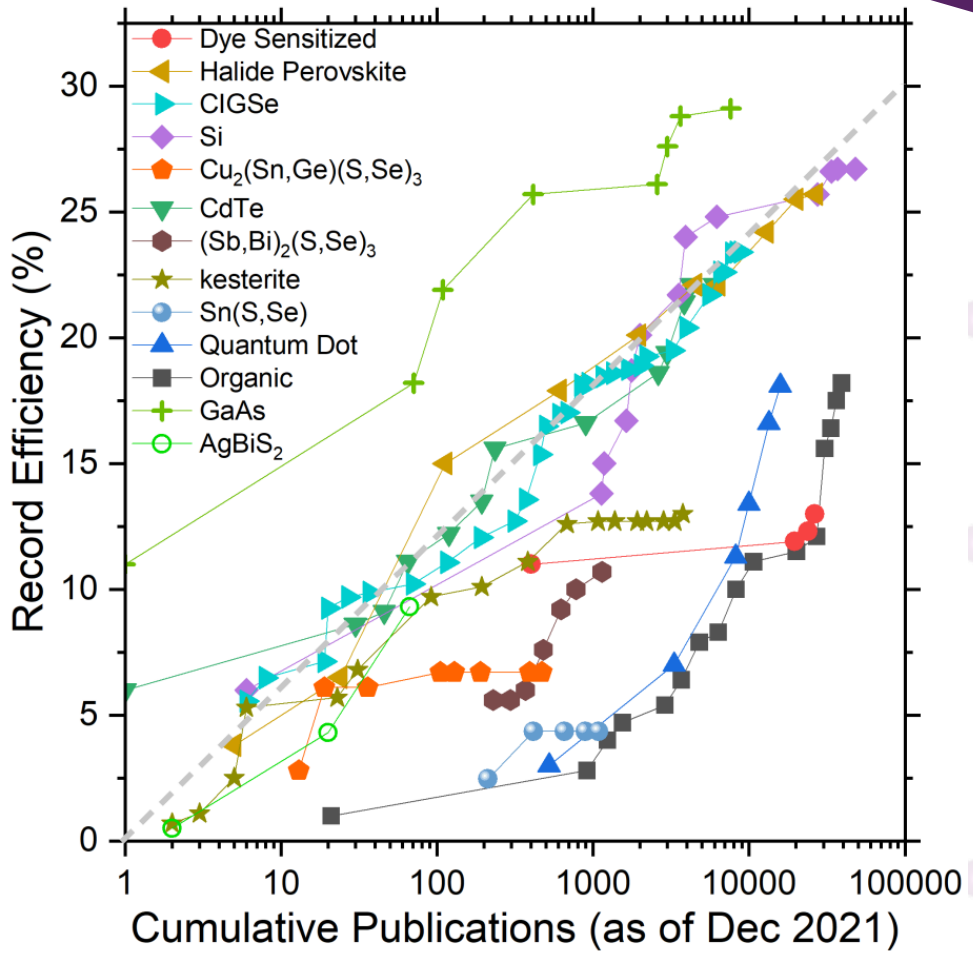
SOLAR CELLS

Monolithic perovskite/silicon tandem solar cell with >29% efficiency by enhanced hole extraction

Amran Al-Ashouri[1]*, Eike Köhnen[1]*, Bor Li[1], Artiom Magomedov[2], Hannes Hempel[3], Pietro Caprioglio[1,4], José A. Márquez[3], Anna Belen Morales Vilches[5], Ernestas Kasparavicius[2], Joel A. Smith[6,7], Nga Phung[6], Dorothee Menzel[1], Max Grischek[1,4], Lukas Kegelmann[1], Dieter Skroblin[8], Christian Gollwitzer[8], Tadas Malinauskas[2], Marko Jošt[1,9], Gašper Matič[9], Bernd Rech[10,11], Rutger Schlatmann[5,12], Marko Topič[9], Lars Korte[1], Antonio Abate[6], Bernd Stannowski[5,13], Dieter Neher[4], Martin Stolterfoht[4], Thomas Unold[3], Vytautas Getautis[2], Steve Albrecht[1,11]†

Tandem solar cells that pair silicon with a metal halide perovskite are a promising option for surpassing the single-cell efficiency limit. We report a monolithic perovskite/silicon tandem with a certified power conversion efficiency of 29.15%. The perovskite absorber, with a bandgap of 1.68 electron volts, remained phase-stable under illumination through a combination of fast hole extraction and minimized nonradiative recombination at the hole-selective interface. These features were made possible by a self-assembled, methyl-substituted carbazole monolayer as the hole-selective layer in the perovskite cell. The accelerated hole extraction was linked to a low ideality factor of 1.26 and single-junction fill factors of up to 84%, while enabling a tandem open-circuit voltage of as high as 1.92 volts. In air, without encapsulation, a tandem retained 95% of its initial efficiency after 300 hours of operation.

Classical solar-cell research



Literature

P. Dale and M. A. Scarpulla,
Sol. Energy Mater. Sol. Cells (2023).

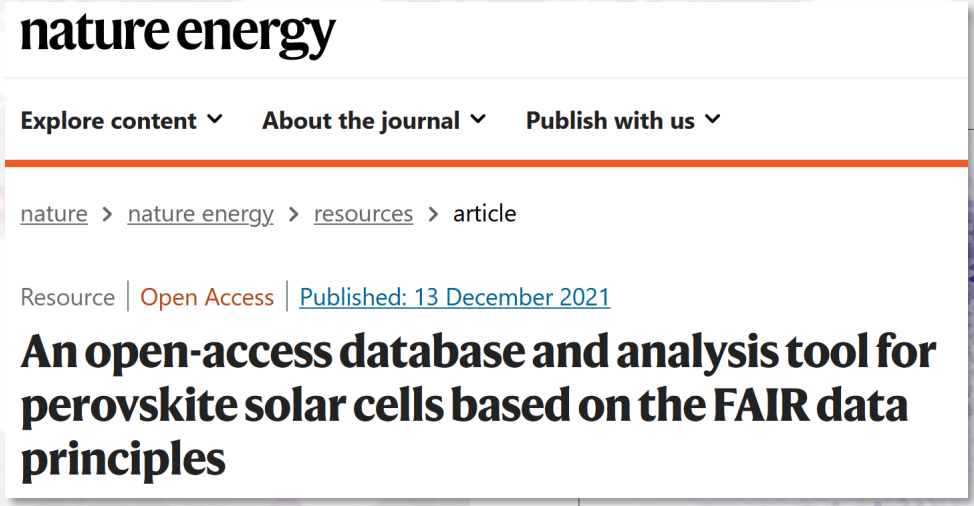
The Perovskite Database Project

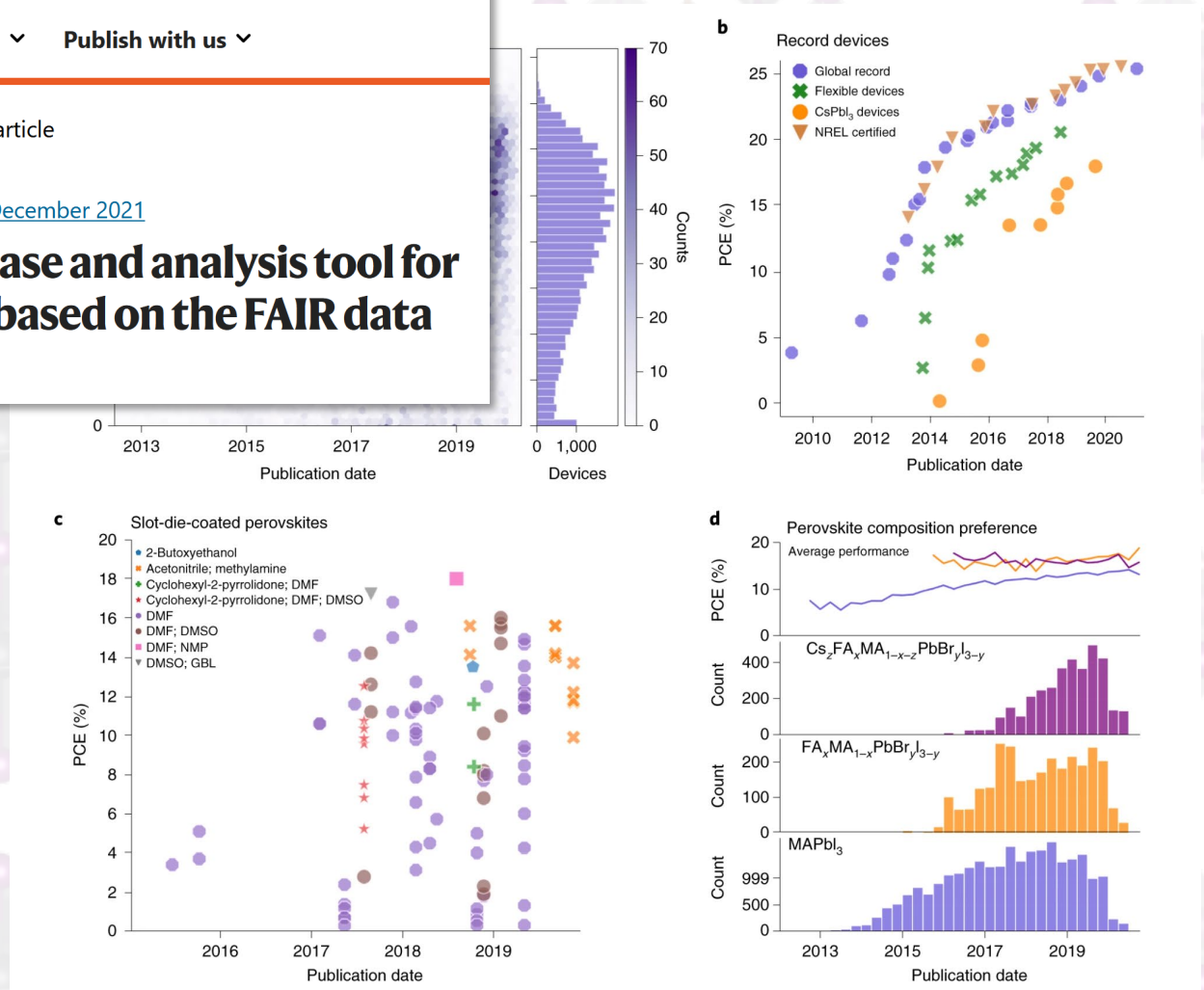
Pioneering project

100 authors
Manual handling of
42000 devices
16000 publications

More than 400 metadata categories
Synthesis information

Difficult to update





J. Jacobsson, et al. Nature Energy 7, 2022.

Heterogenous sources

Efficiency tables

TOPICAL REVIEW • OPEN ACCESS

Emerging inorganic solar cell efficiency tables (version 2)

Andriy Zakutayev[1], Jonathan D Major[2], Xiaojing Hao[3], Aron Walsh[4,5], Jiang Tang[6], Teodor K Todorov[7], Lydia H Wong[8] and Edgardo Saucedo[10,9]

Published 16 April 2021 • © 2021 The Author(s). Published by IOP Publishing Ltd

Journal of Physics: Energy, Volume 3, Number 3

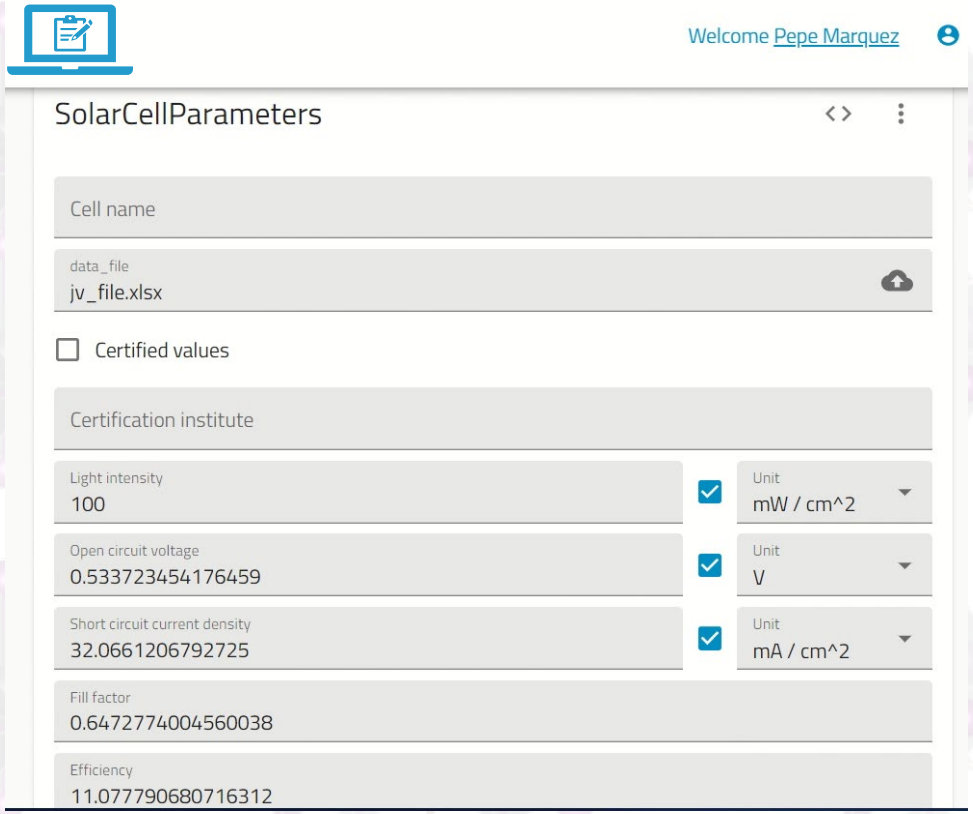
Citation Andriy Zakutayev *et al* 2021 *J. Phys. Energy* **3** 032003

Databases

EMERGING PV

The Perovskite database

Lab notebooks



Explore solar cells in NOMAD

https://nomad-lab.eu

PUBLISH EXPLORE ANALYZE ABOUT

LOGIN / REGIST

Entries

FILTERS

EELS

Workflow

- Molecular Dynamics
- Geometry Optimization

Properties

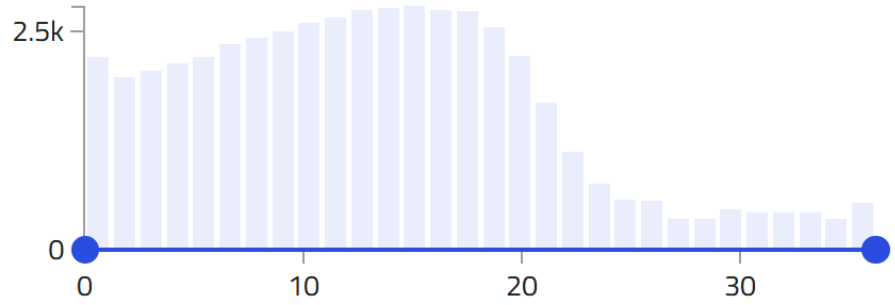
- Electronic
- Vibrational
- Mechanical

Use Cases

12.495.323 RESULTS

SOLAR CELLS

Efficiency zoom 1/4



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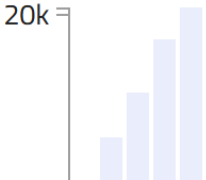
Fill Factor zoom 1/4



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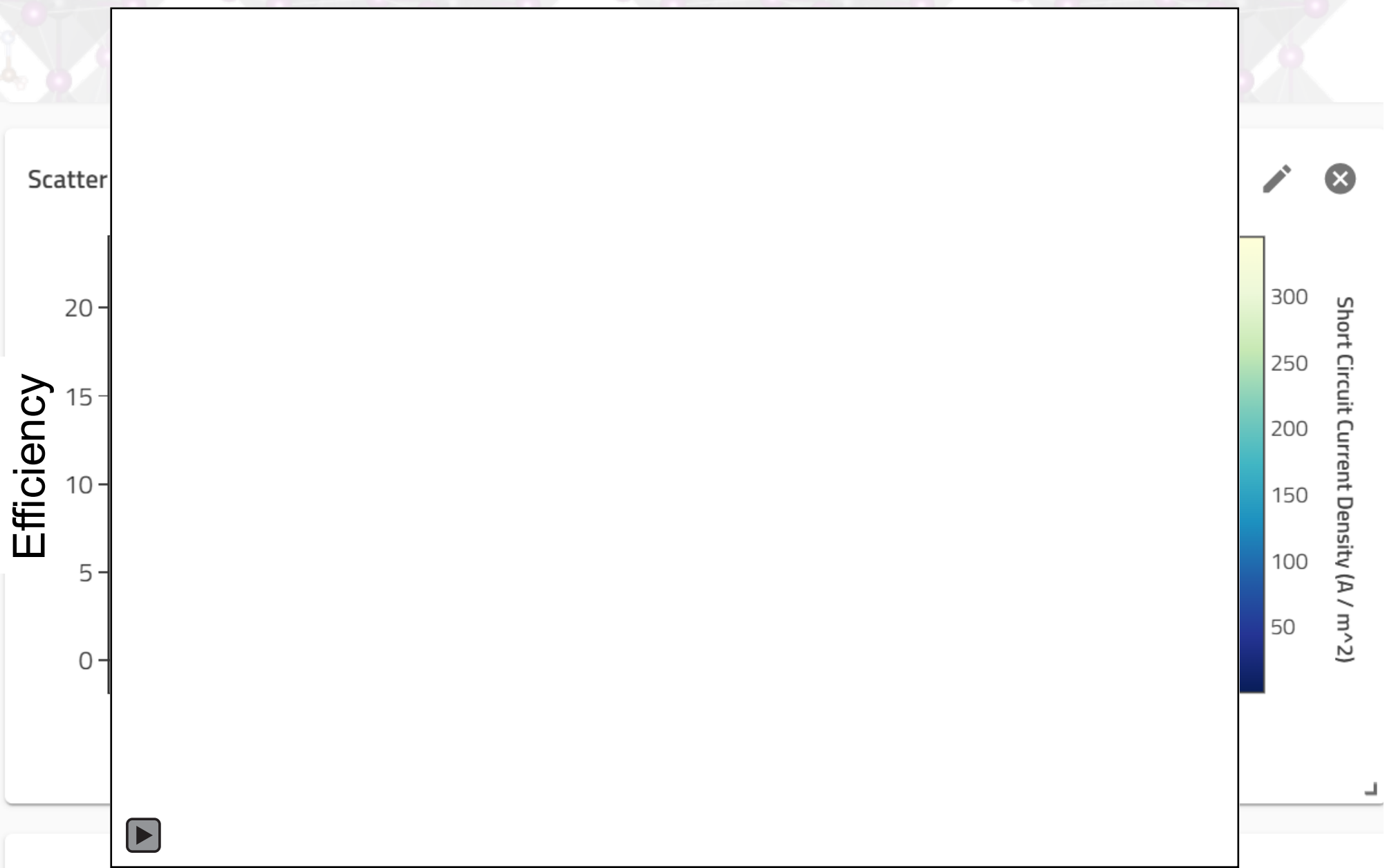
Explore solar cells in NOMAD

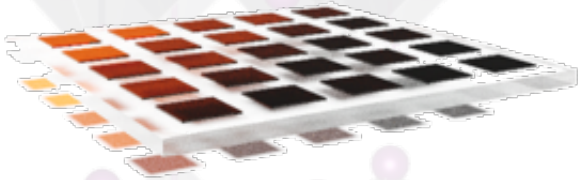


P. Marquez

NOMAD OASIS

SLOW

Synthesis



Characterization

Combinatorial synthesis

High throughput characterization

Autonomous labs

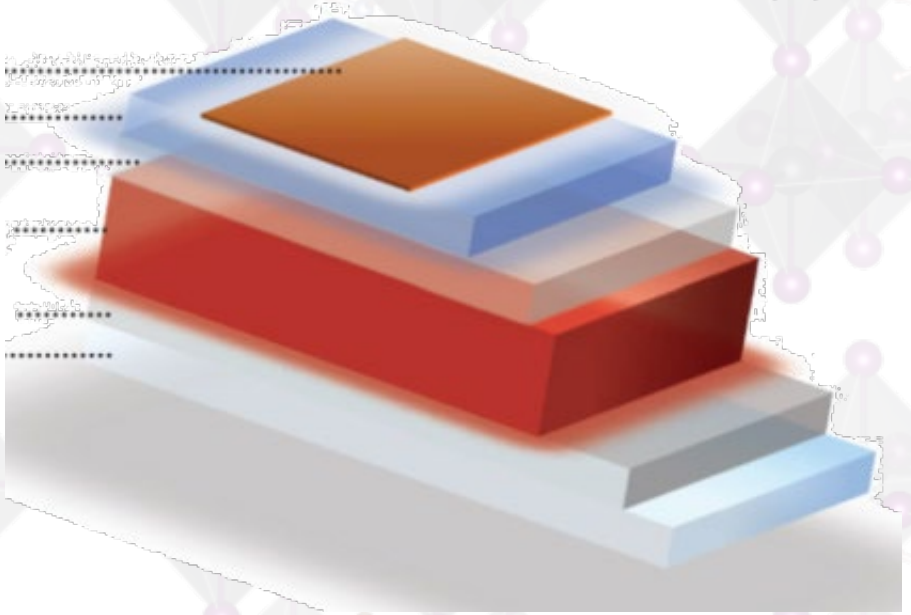
AI for an extra boost

NOMAD
OASIS

Combinatorial explosion

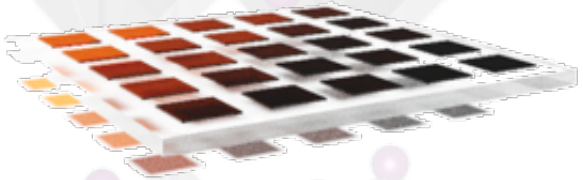
Device stack

contact
ETL
absorber
HTL
contact



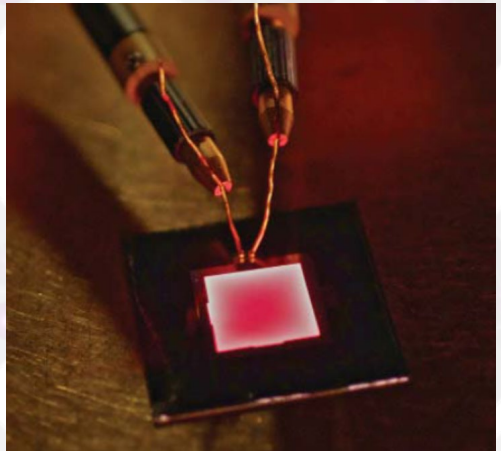
SLOW

Synthesis



Characterization

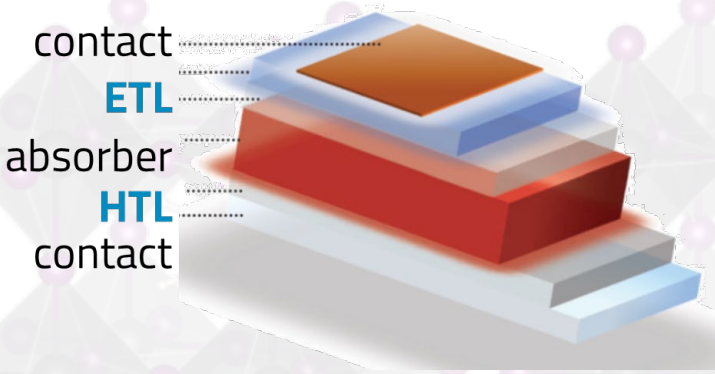
Device

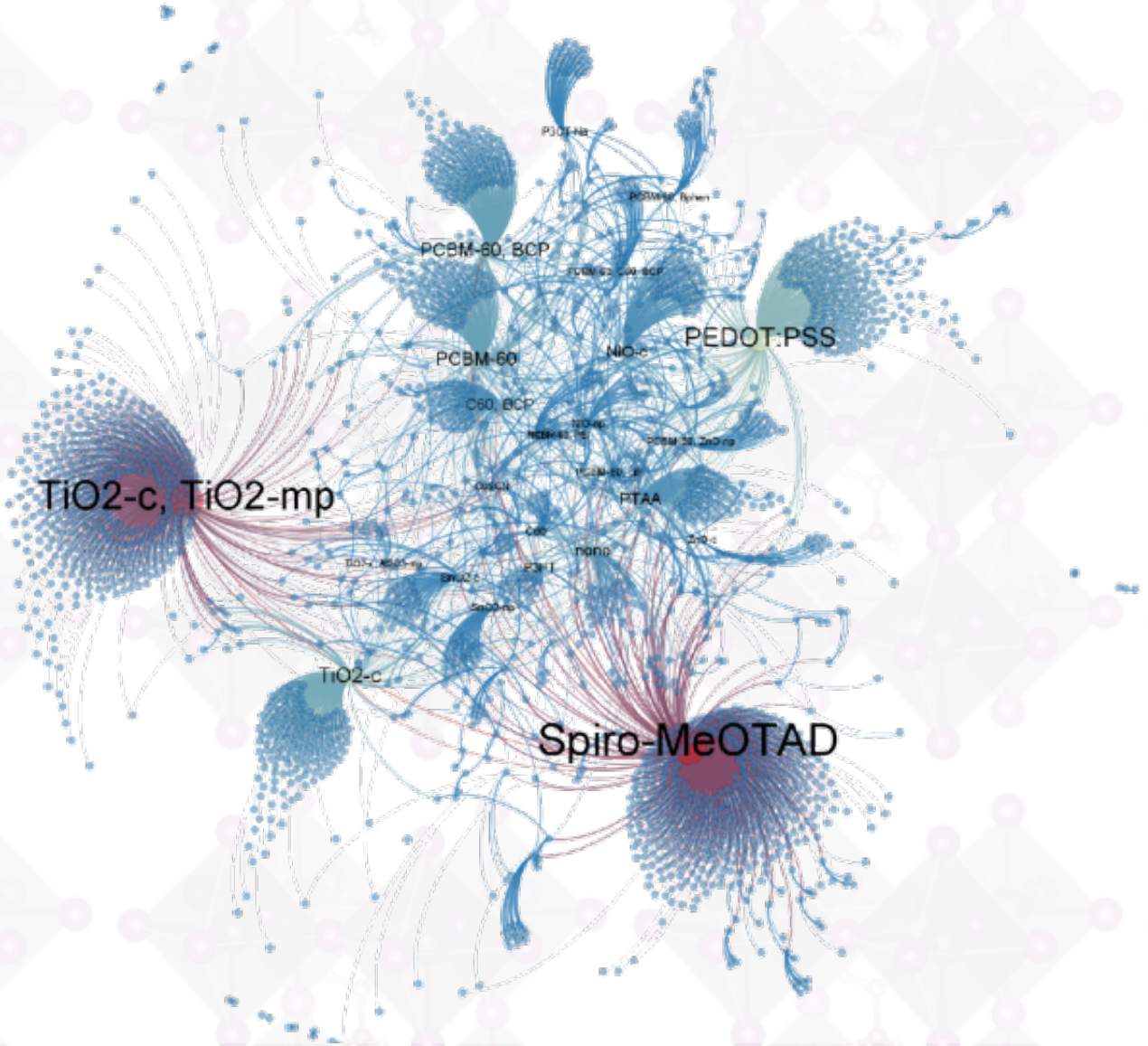


ETL: electron transport layer

HTL: hole transport layer

Charge transport layers





Nodes: 2600

transport layers

Edges: 3126

combinations

Computational high-throughput screening?

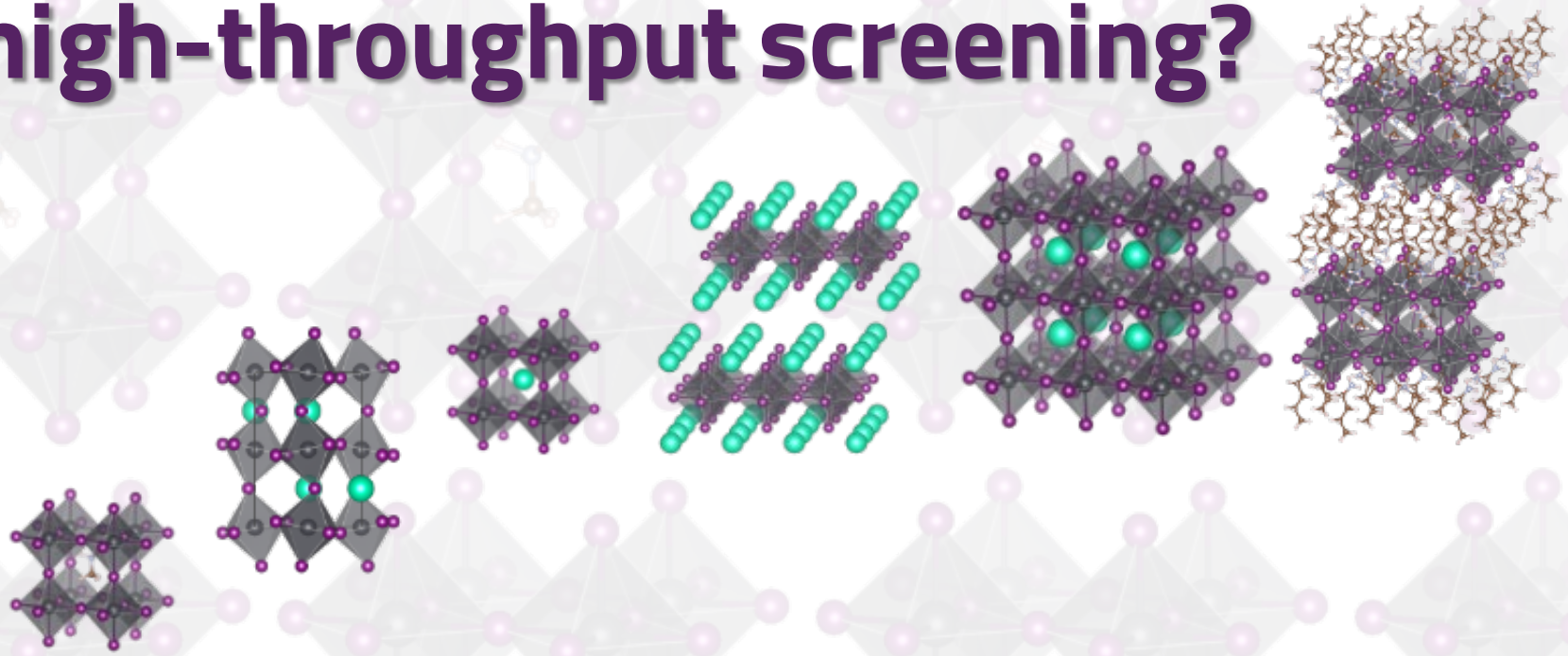
DFT level

Problematic

Many-body level

Not feasible

Problematic



Property 1

Property 2

Property 3

Property 4

Dilemma ...

ADVANCED THEORY AND SIMULATIONS

Research Article | Open Access

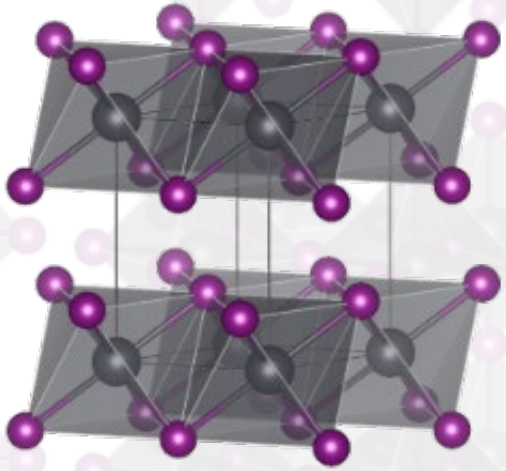
Electronic Structure of (Organic-)Inorganic Metal Halide Perovskites: The Dilemma of Choosing the Right Functional

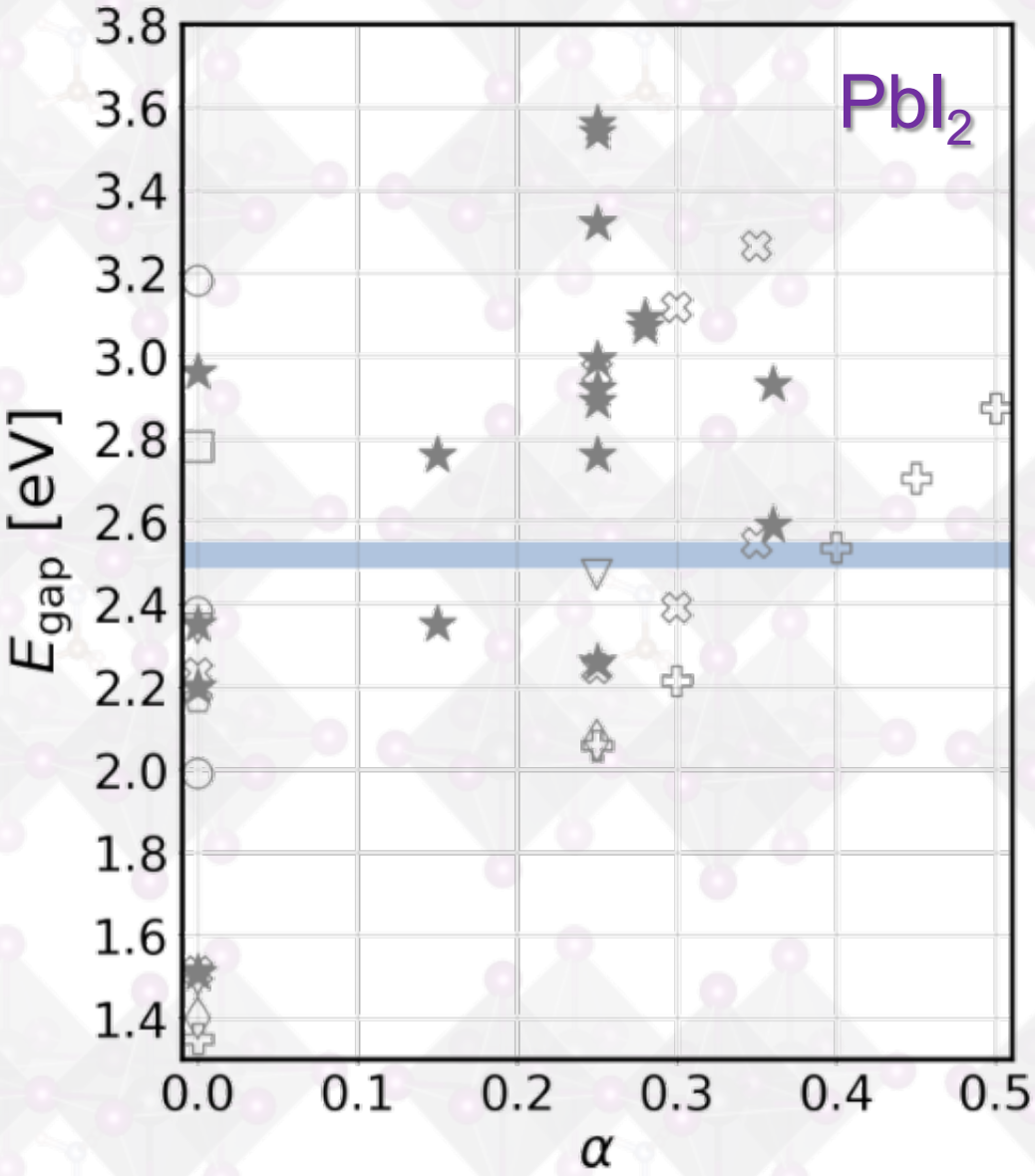
Cecilia Vona, Dmitrii Nabok, Claudia Draxl

First published: 17 December 2021 | https://doi.org/10.1002/adts.202100496

PbI_2 – a precursor

Band gaps from DFT and beyond …

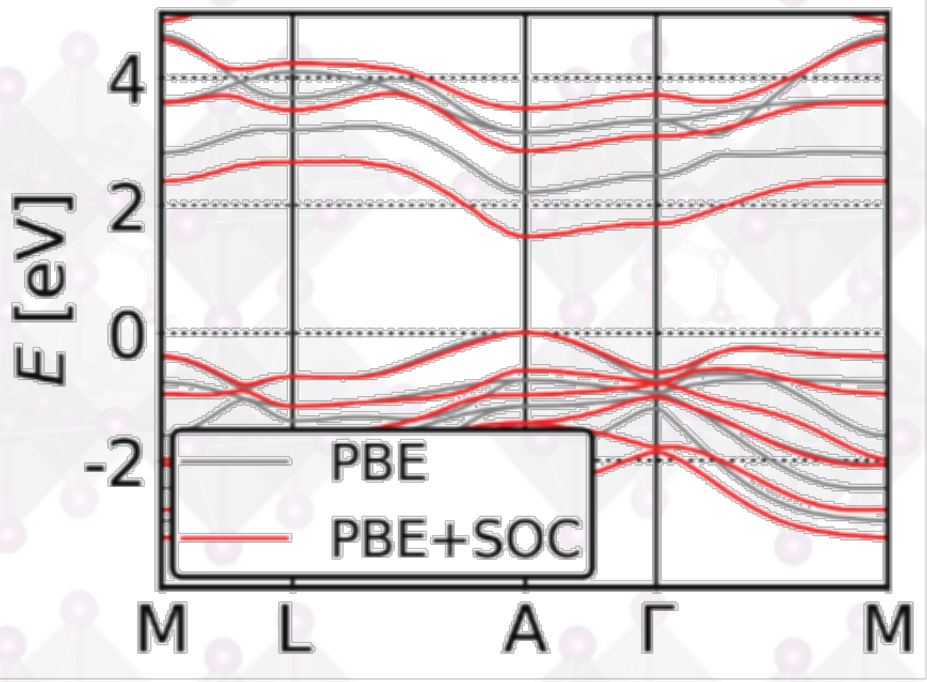


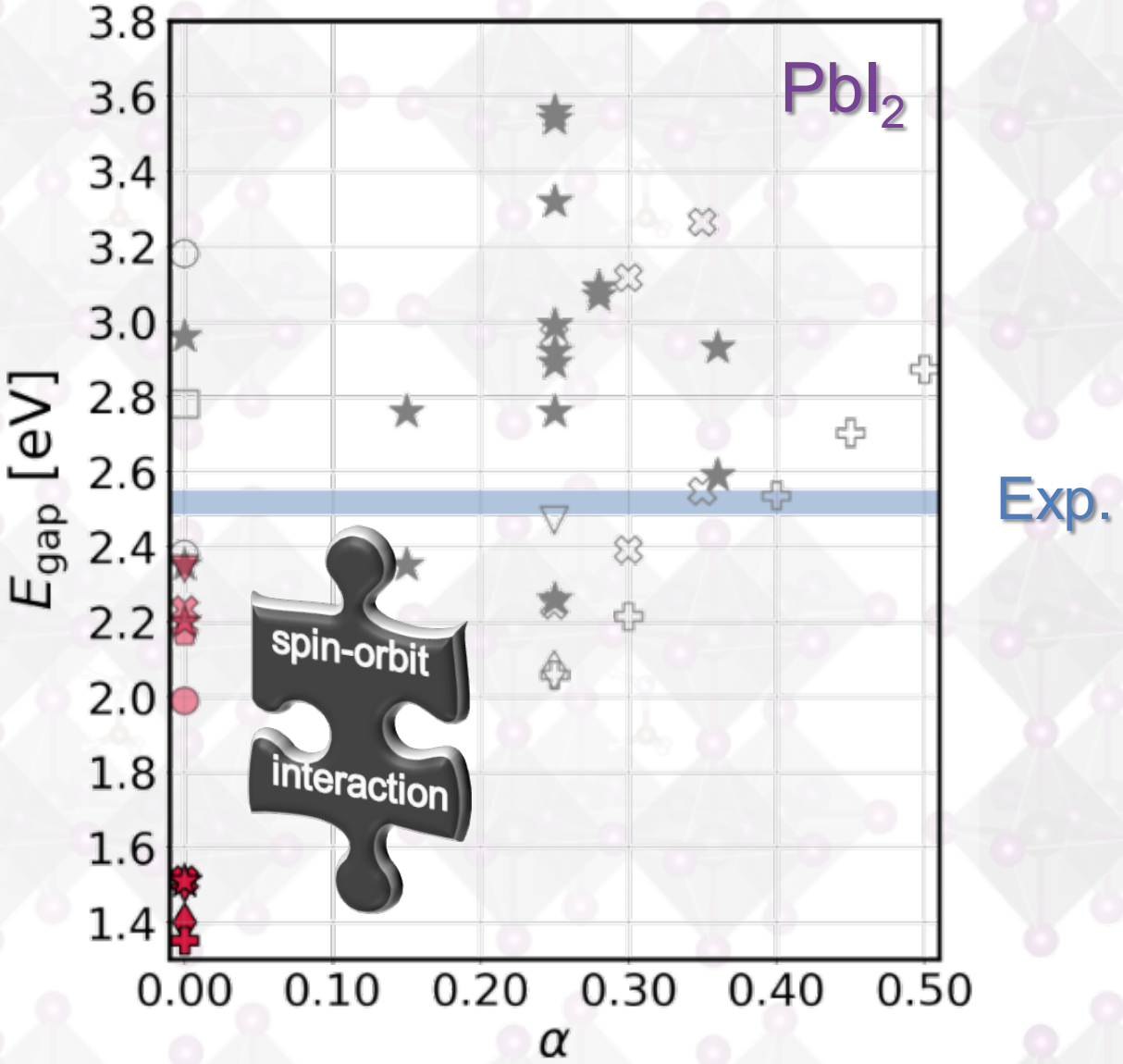


Electronic structure

Semi-local functionals

LDA, GGAs

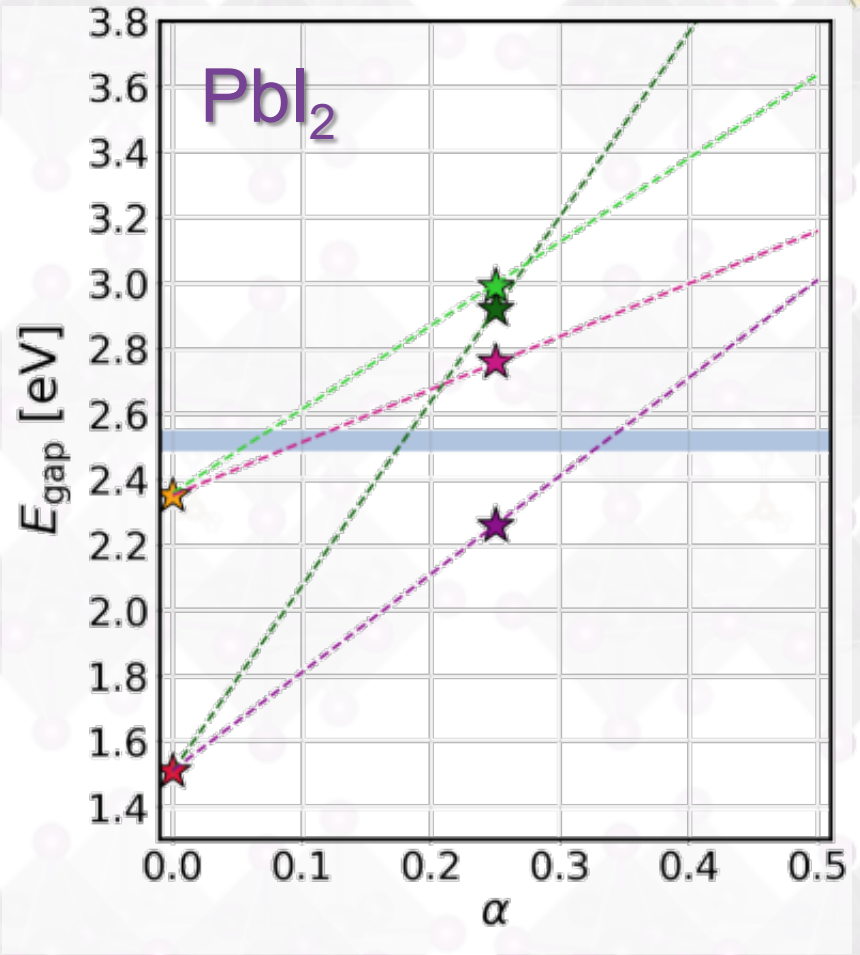


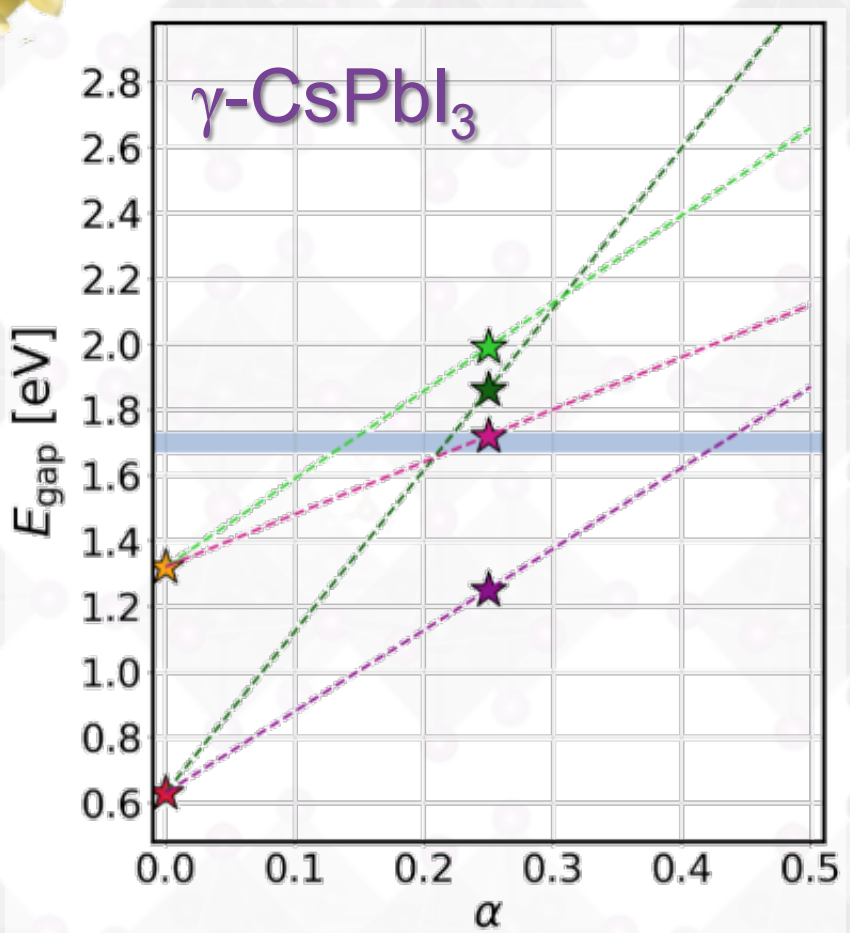


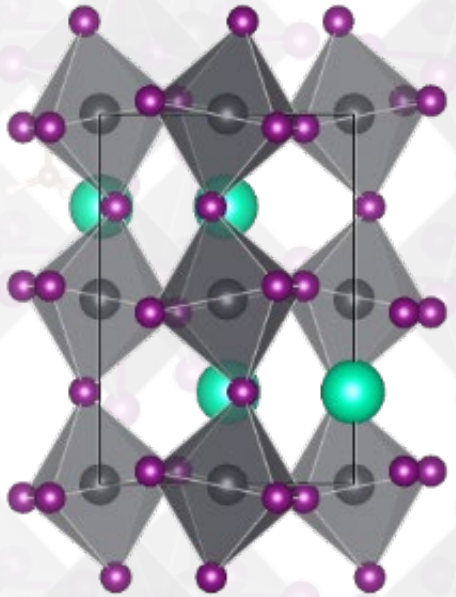
$$\left[T + V_{ext}(\mathbf{r}) + V_H(\mathbf{r}) + V_{xc}(\mathbf{r})\right]\psi_i^{KS}(\mathbf{r}) = \epsilon_i^{KS}\psi_i^{KS}(\mathbf{r})$$

Best performance?

exciting







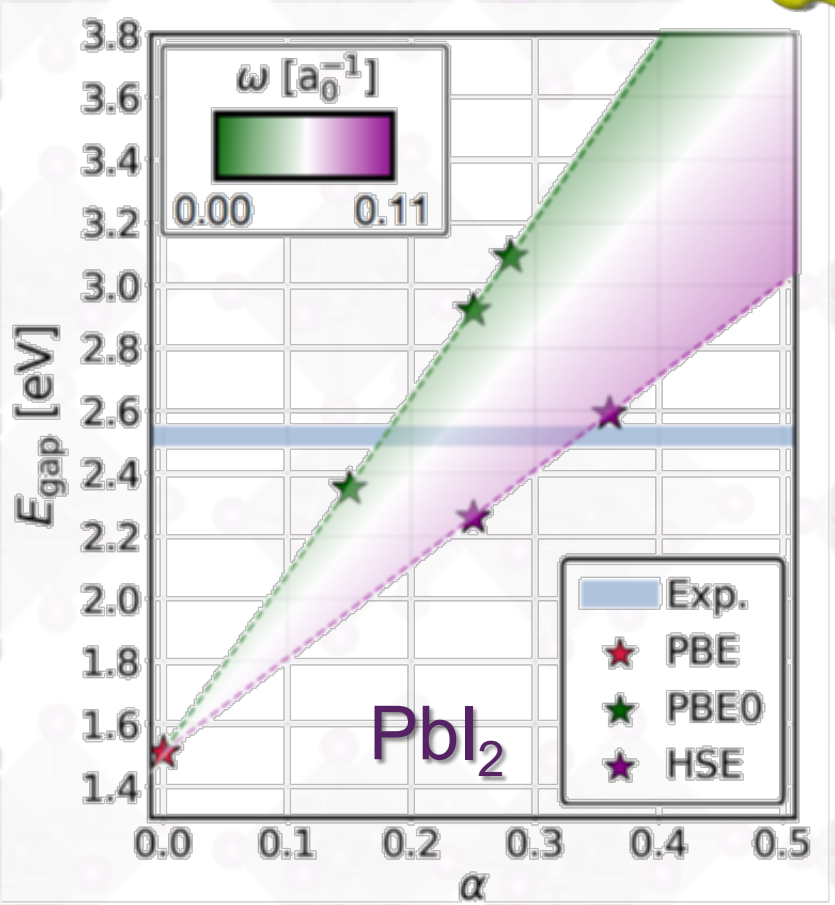
$$E_{xc}^{PBE0} = E_{xc}^{PBE} + \alpha(E_x^{HF} - E_x^{PBE})$$

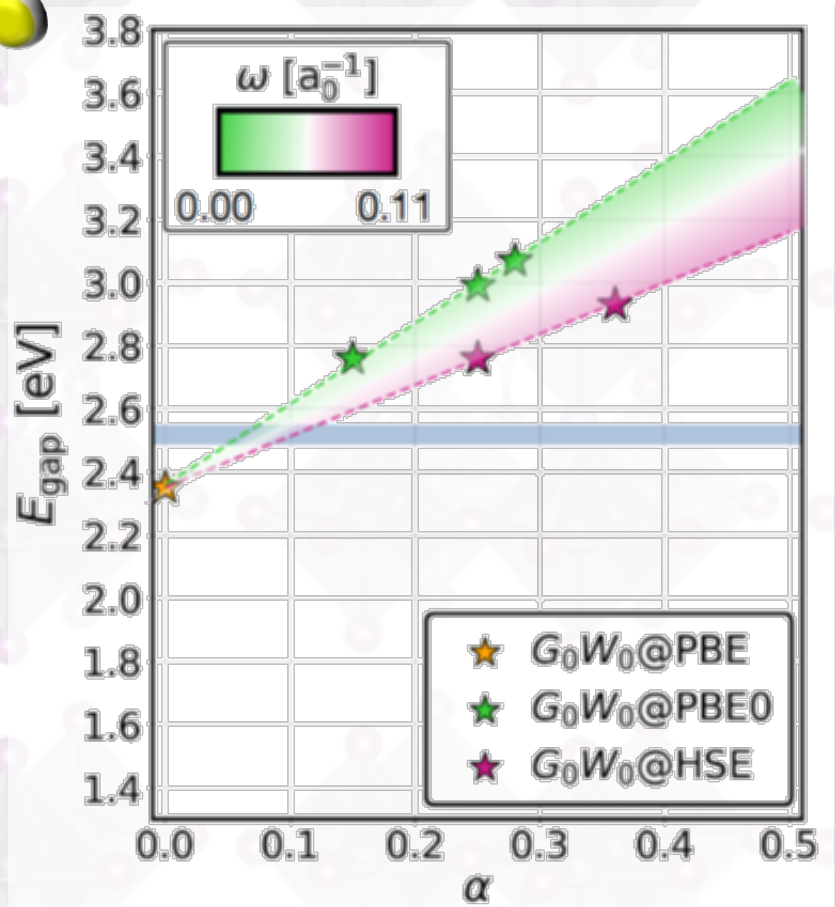
$$E_{xc}^{HSE06} = E_{xc}^{PBE} + \alpha[E_x^{HF,SR}(\omega) - E_x^{PBE,SR}(\omega)]$$

More to tune …

$$E_{xc}^{HSE06} = E_{xc}^{PBE} + \alpha[E_x^{HF,SR}(\omega) - E_x^{PBE,SR}(\omega)]$$

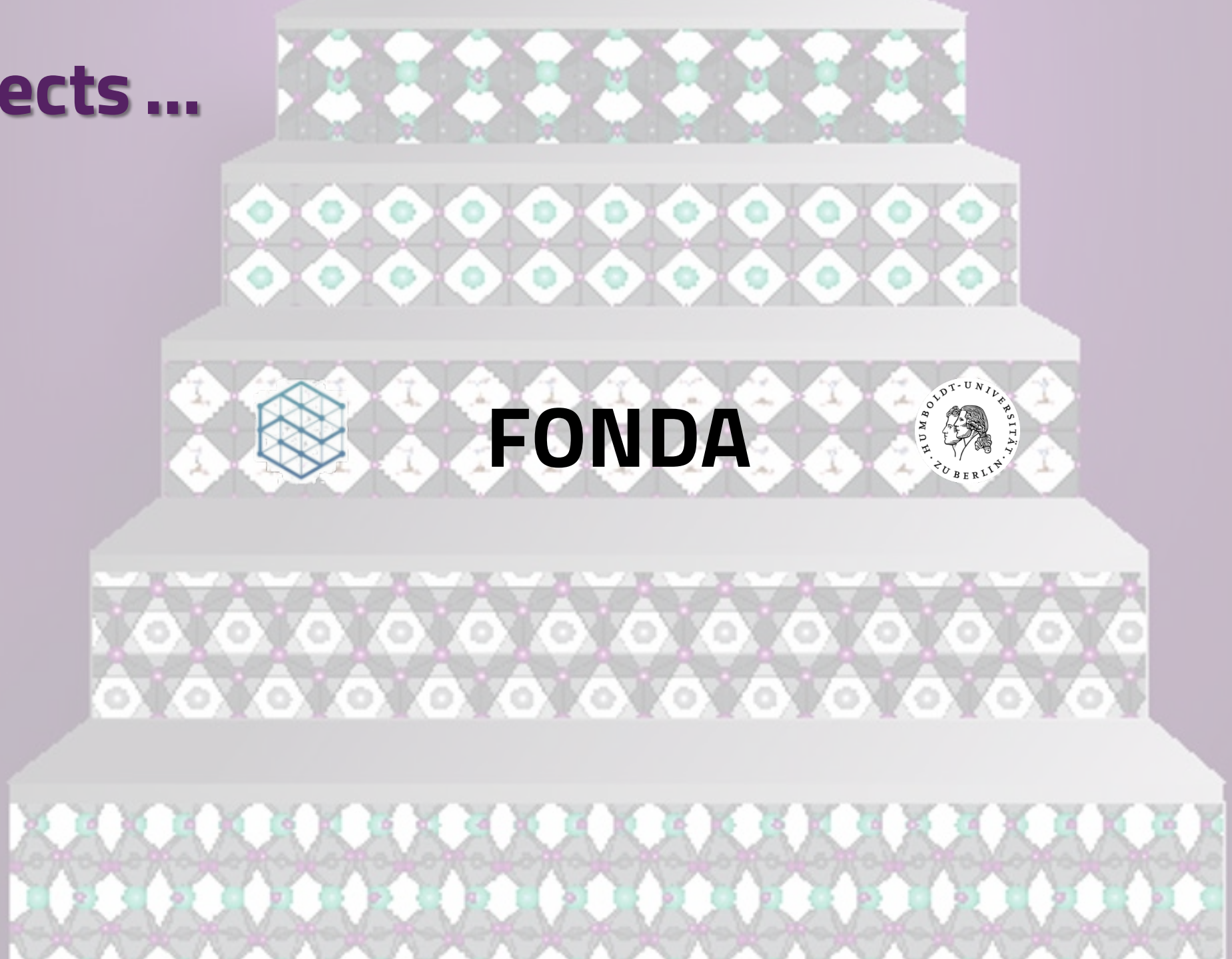
HSE: mixing α, screening ω





Exp.

3 projects ...



FONDA

Science of workflows

FONDA

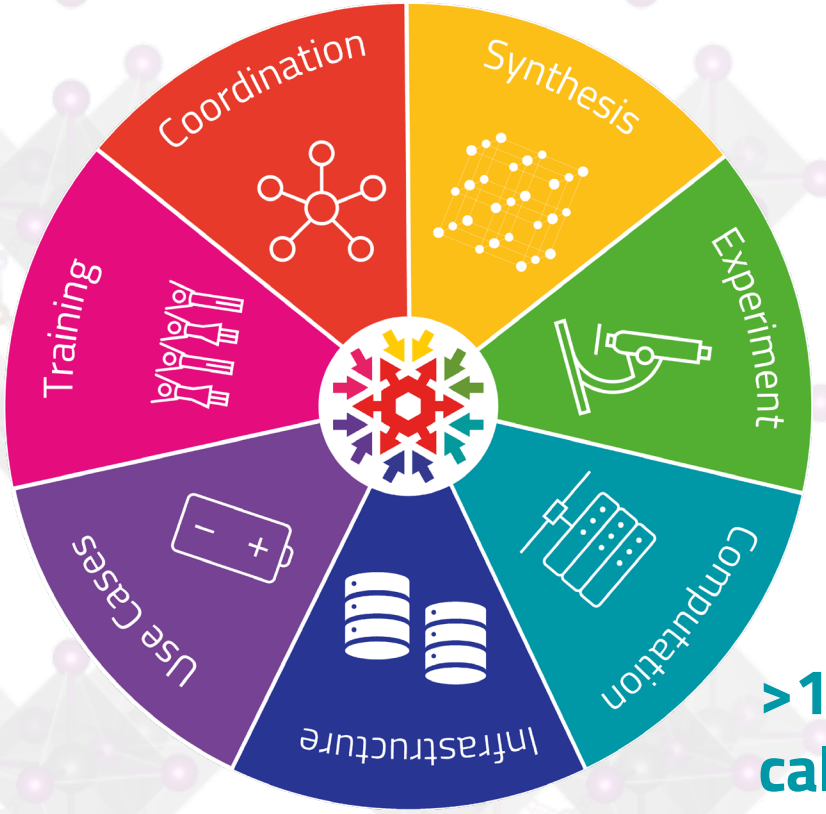
FONDA

Foundation of Workflows for Large-Scale Scientific Data Analysis

Specific project

Deriving Trust Levels for
Multi-Choice Data Analysis Workflows

- Verification & validation
- Data quality
- Configuration-space sampling



>140 mio calculations

Similarity measures

Example: spectral fingerprints*

Tailored weighting of spectral features

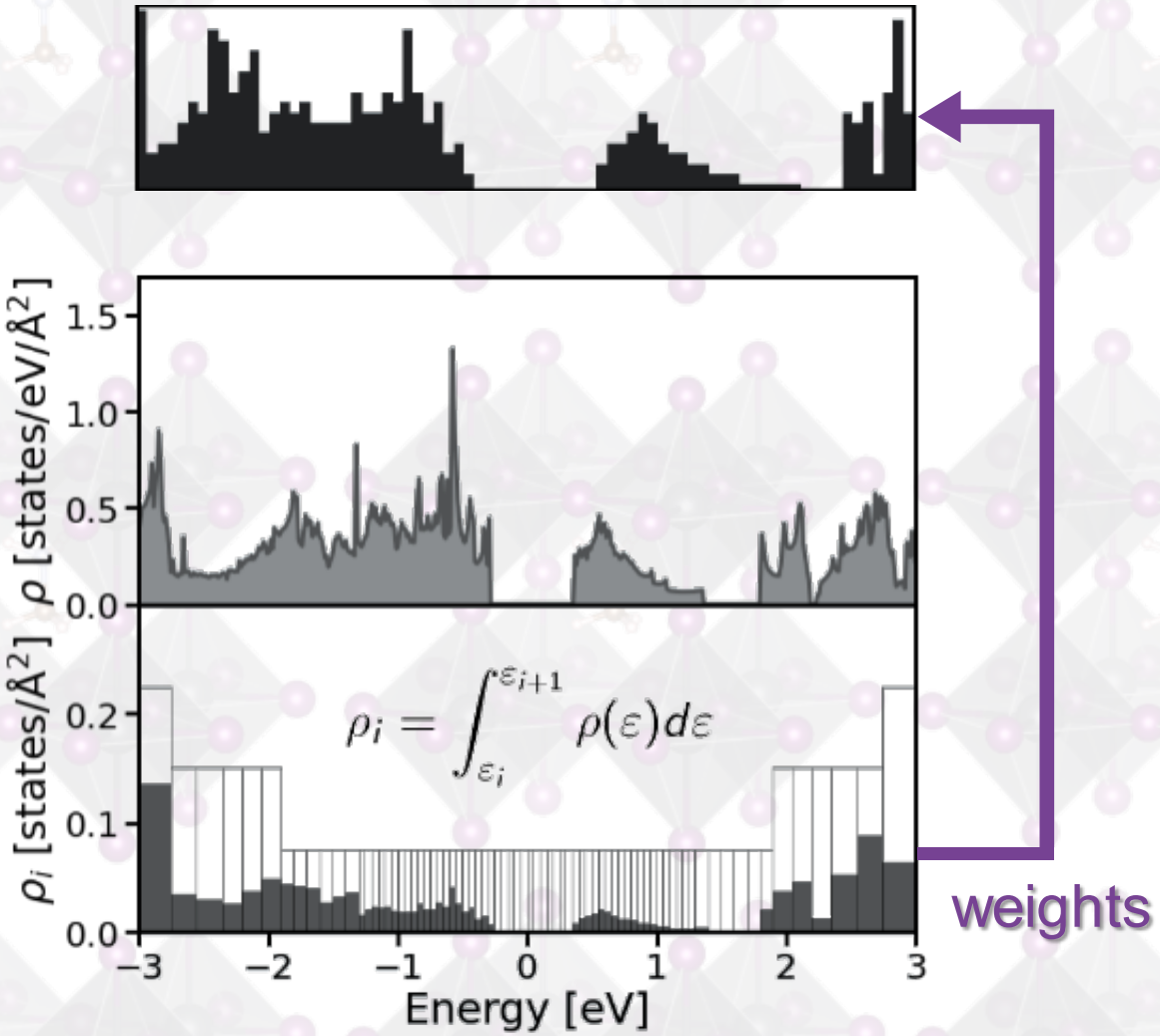
Choose energy range, binning

Focus on particular region

Tanimoto's similarity coefficient

$$Tc(i,j) = \frac{f_i \cdot f_j}{|f_i|^2 + |f_j|^2 - f_i \cdot f_i}$$

0 ≤ Tc ≤ 1



M. Kuban, S. Rigamonti, M. Scheidgen, and C. Draxl, Sci. Data 9, 646 (2022).

Spectral fingerprints

Data quality and error estimates

Example: bulk PbI_2

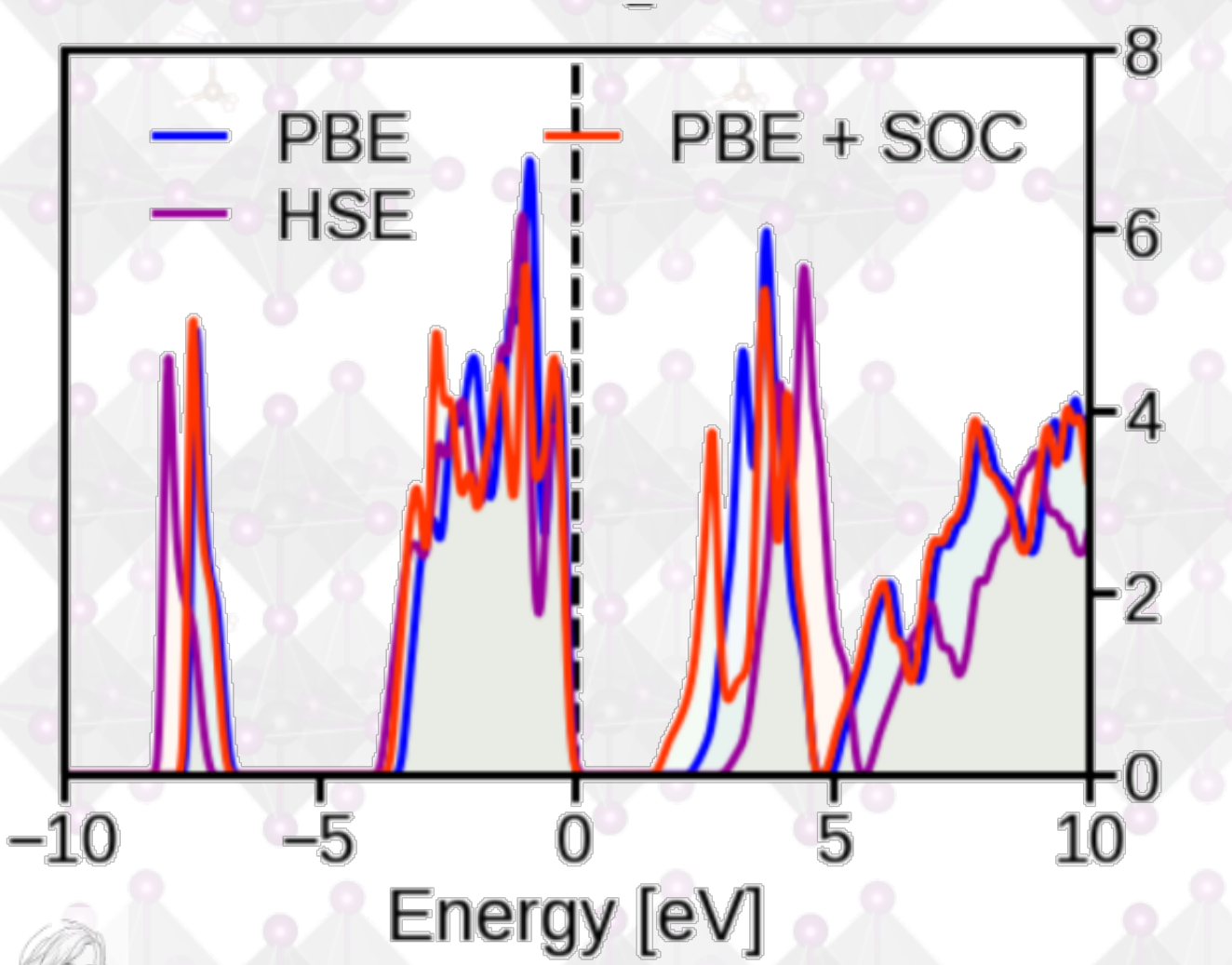
PBE – HSE

Tc=0.60

PBE – PBE+SOC

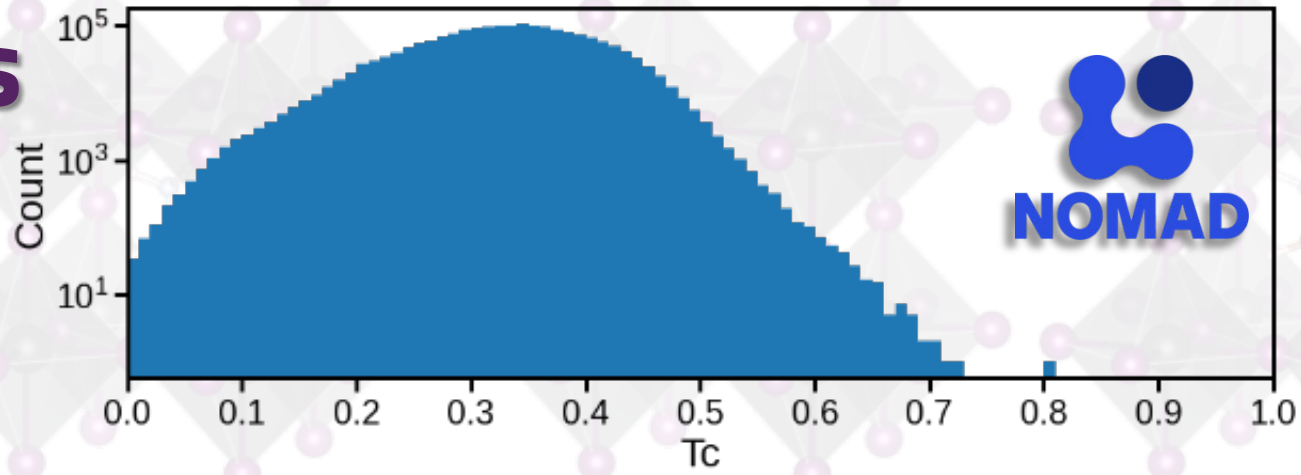
Tc=0.71

Cancellation of effects



M. Kuban, S. Gabaj, W. Aggoune, C. Vona, S. Rigamonti, and C. Draxl, MRS Bulletin Impact 47, 1 (2022).

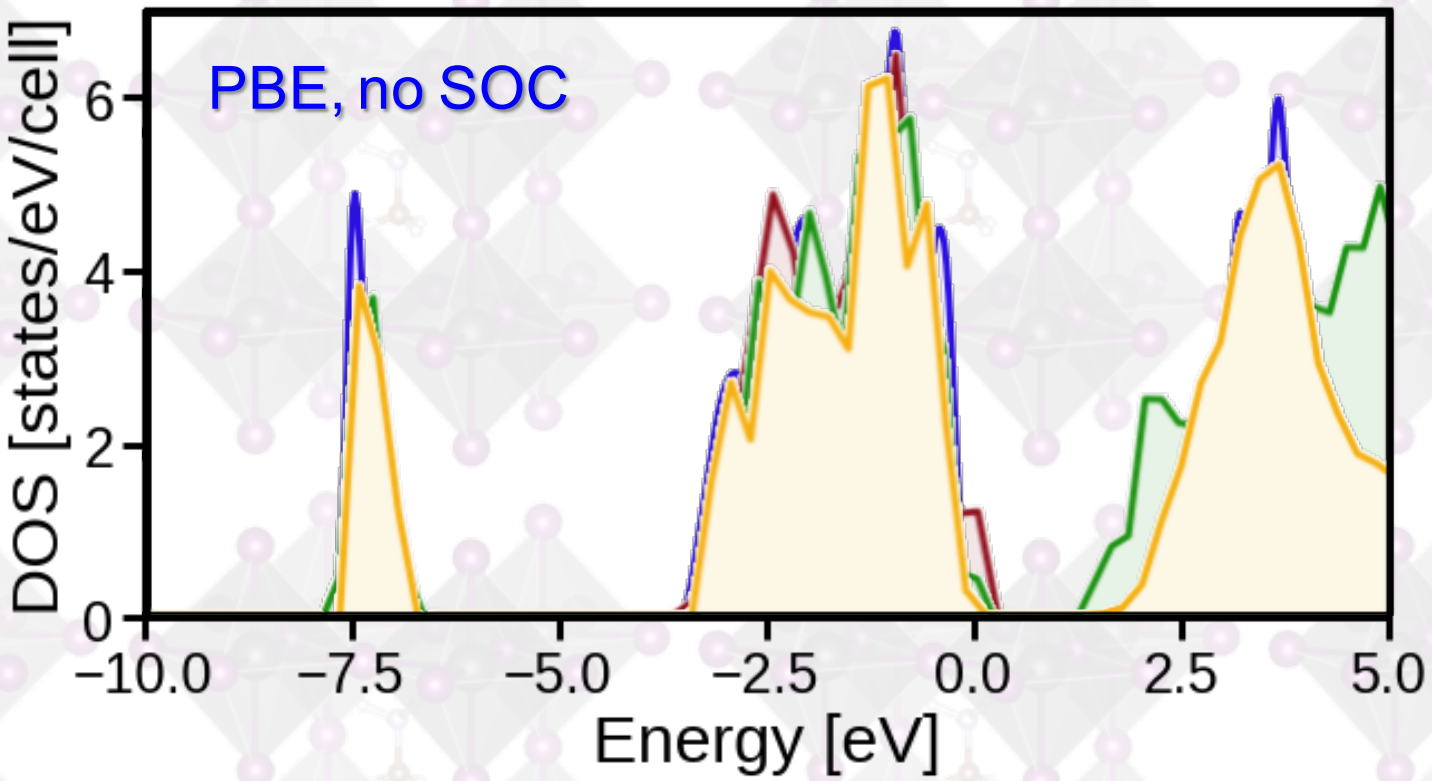
Finding similar materials



Example

Materials similar to PbI_2

Based on 1,899,783 mio calculations



PbI_2	reference
$PbSe_2Sr$	Tc=0.80
$PbSe_2Sr$	Tc=0.72
Ca_2S_2	Tc=0.72

M. Kuban

Wrap-up ...

FONDA

FAIRmat

NOMAD CoE

Customize your workflows ...

Thanks !!