Data & HPC-driven discovery of novel materials for nanotechnology and sustainable energy

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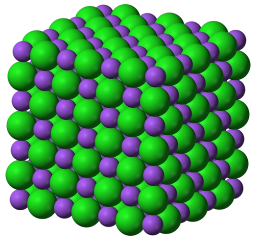
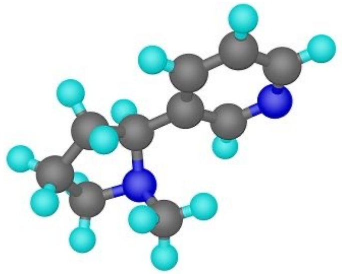


Electronic structure calculations



What?

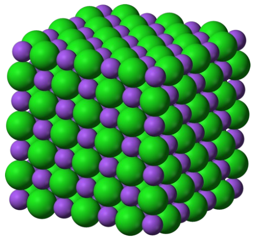
Solve Schrödinger's equation for the motion of electrons in a solid/molecule.

$$\left[\frac{\hbar^2}{2m}\nabla^2 + V(\{\boldsymbol{R}_i\};\boldsymbol{x})\right]\Psi_n\,(\boldsymbol{x}) = E_n\Psi_n(\boldsymbol{x})$$

Why?

The electrons determine everything

- Chemistry (bonds, chemical reactions, ...)
- Thermodynamics (heat capacity, phase diagrams, …)
- Structure and ion dynamics (phonons, …)
- Physical properties (magnetism, conductivity, …)



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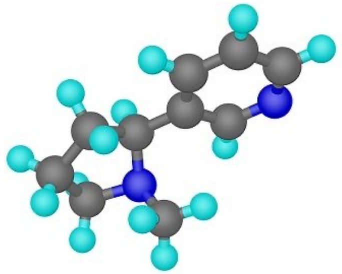

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No parameters.
Only laws of nature and fundamental constants.
Ab initio* / *first-principles

The open source GPAW code

Enkovaara *et al.* J. Phys.:Cond. Mat. **22** (2010) ← **Review article**

https://wiki.fysik.dtu.dk/gpaw/ ← **Free download, GPL**

- ❑ Projector augmented wave (PAW)
- ❑ Three different basis sets (real space, plane waves, LCAO)
- ❑ Efficient parallelization (good scalability up to > 100.000 CPU cores)
- ❑ Time-dependent DFT (linear response + time propagation)
- ❑ Many-body perturbation theory
- ❑ Phonons and electron-phonon coupling
- ❑ Advanced magnetic properties
- ❑ GPU version under development

Jens Jørgen Mortensen

The *Niflheim* supercomputer

DeiC

DeiC resources available (2 year grant period):

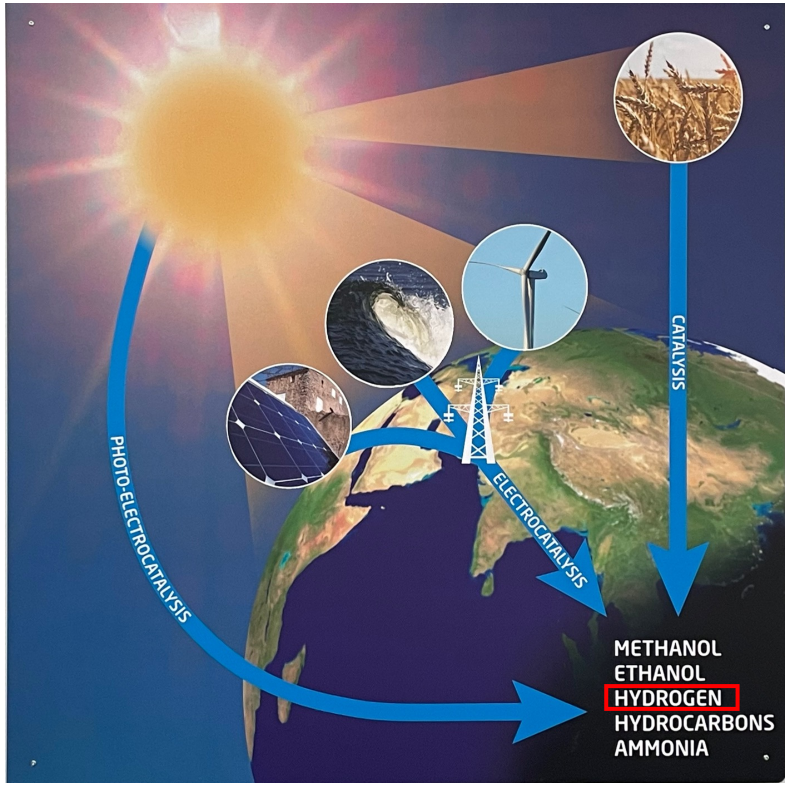
Center	Unit	Resource
DeiC Interactive HPC - CPU	CPU core/h	14,716,800
DeiC Interactive HPC – GPU	GPU core/h	705,100
DeiC Interactive HPC – Storage	TB	1,000
DeiC Throughput HPC	CPU core/h	63,437,500
DeiC Throughput HPC – storage	TB	3,900
DeiC Large Memory HPC	CPU core/h	6,377,200
DeiC Large Mem. HPC – Storage	TB	930

- CPU cores: 24560
- Peak performance: 1.8 PetaFlops
- Co-funded 1:1 by DTU until 2020
- Capacity/2 year: 350 mio core/h
- Investment: DKK 30M over 6 years

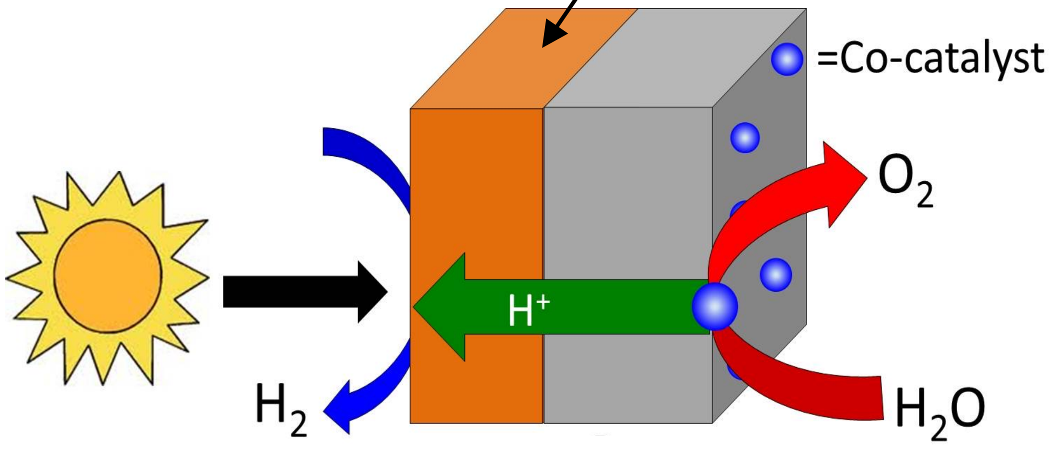
Photo-catalytic water splitting

$$2\,h\nu + H_2O_{(liq)} \rightarrow \tfrac{1}{2}\,O_{2(gas)} + H_{2(gas)}$$

Minimum energy required = 1.23 eV

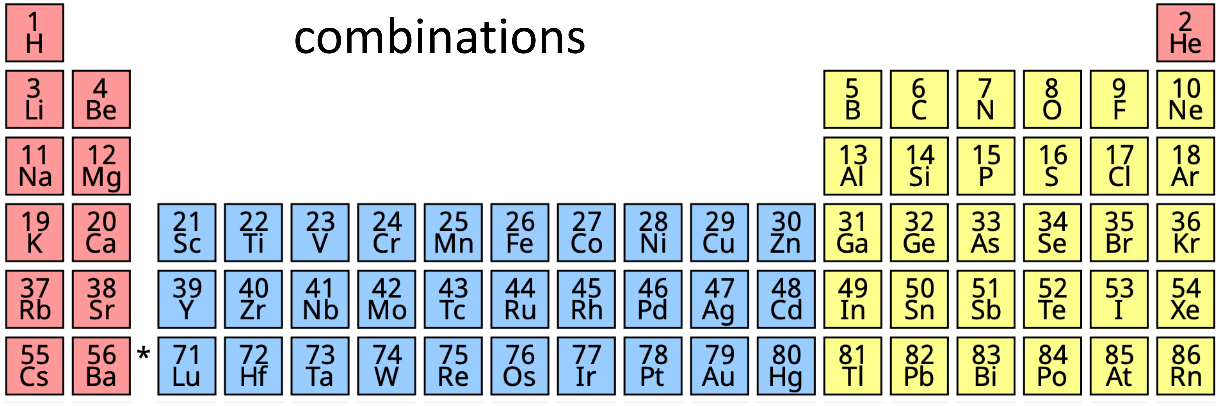


Efficient water splitting requires an absorber material with a band gap ~2 eV



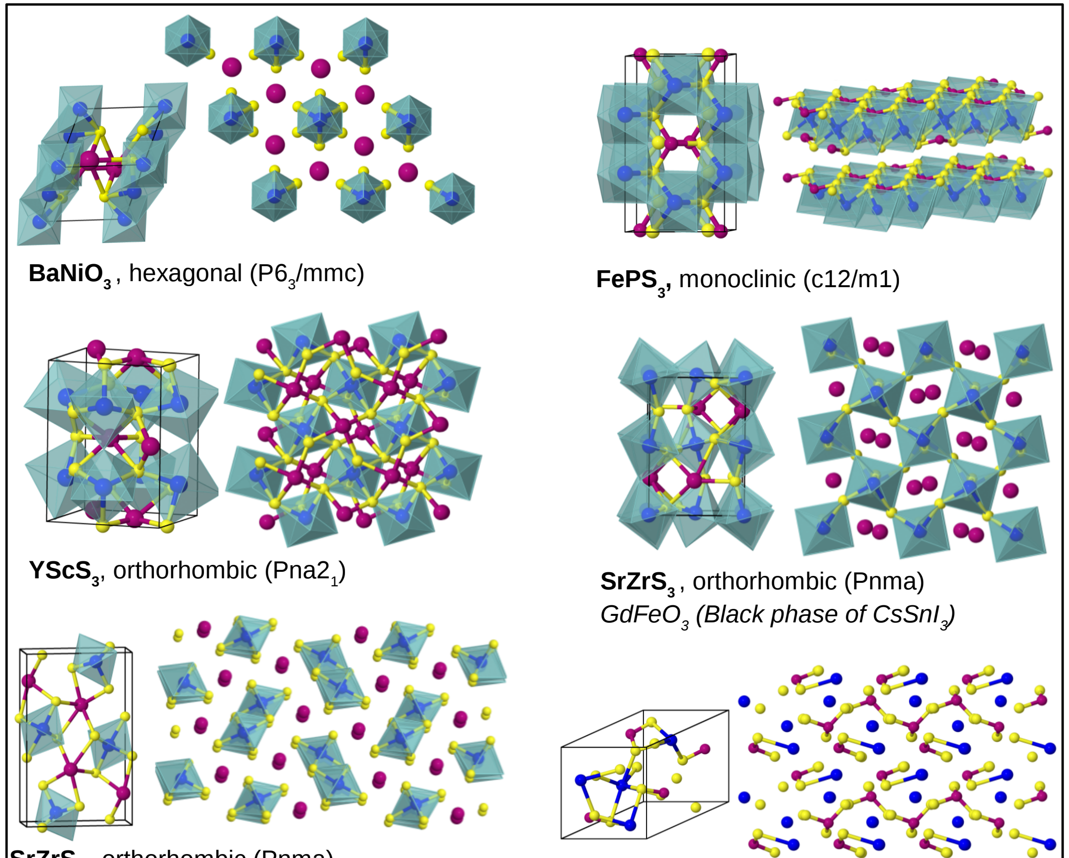
ABS_3 sulphide perovskites

2916 AB cation combinations



17.496 crystals

Six most common ABS_3 crystal structures



$BaNiO_3$, hexagonal ($P6_3/mmc$)

$FePS_3$, monoclinic (c12/m1)

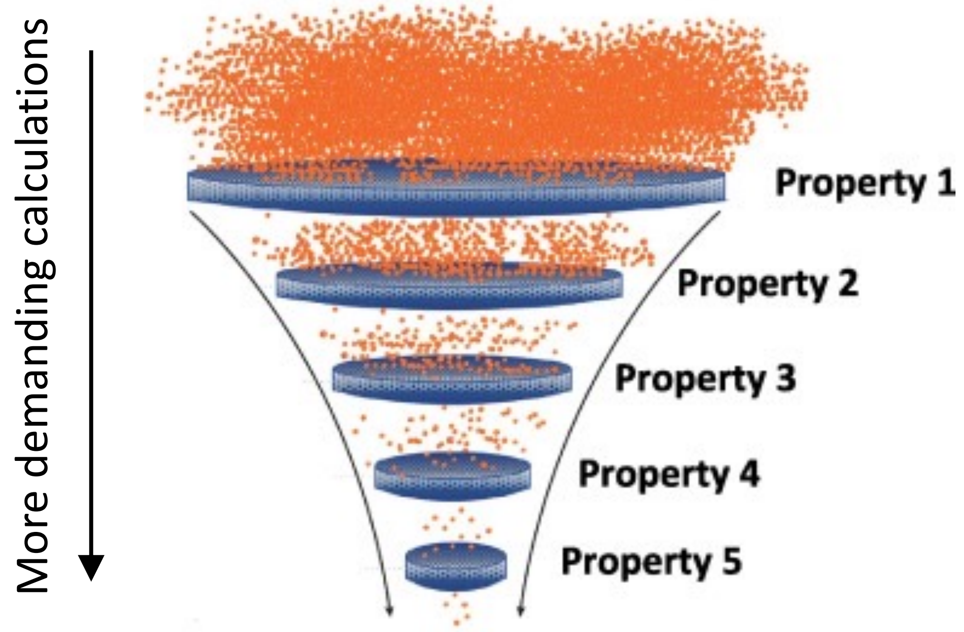
$YScS_3$, orthorhombic ($Pna2_1$)

$SrZrS_3$, orthorhombic (Pnma)
$GdFeO_3$ (Black phase of $CsSnI_3$)

$SrZrS_3$, orthorhombic (Pnma)
NH_4CdCl_3/Sn_2S_3 (Yellow phase of $CsSnI_3$)

$PbPS_3$, monoclinic (P1c1)

Computational screening

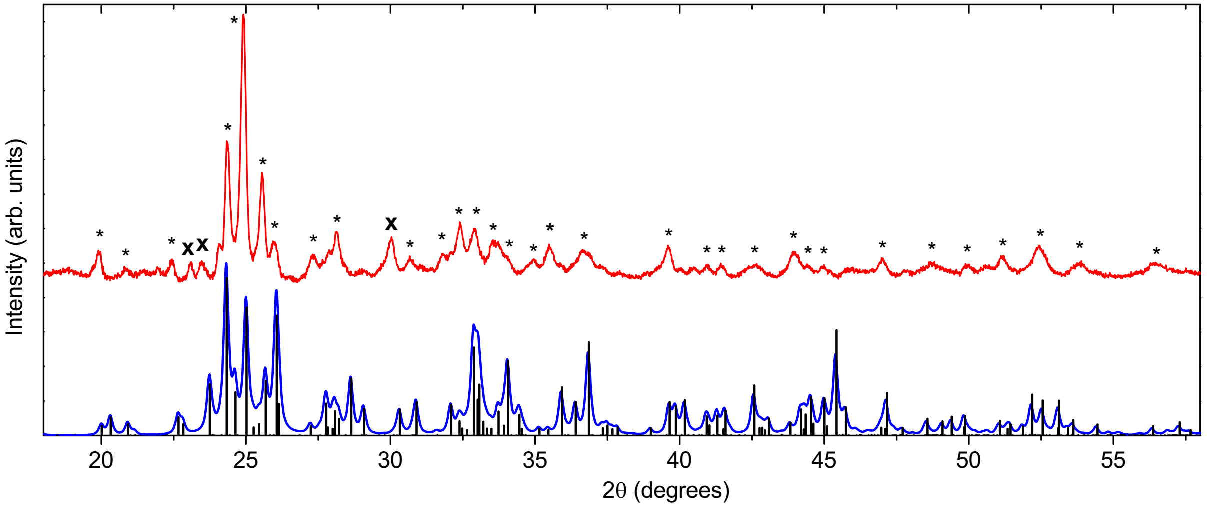


Best candidate materials

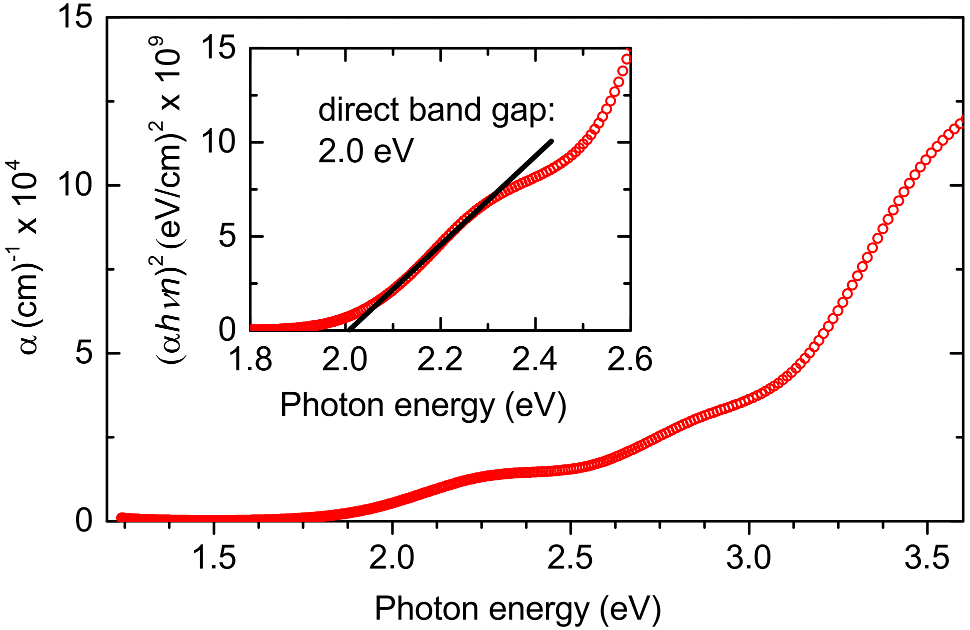
formula	$E_g^{GLLB-SC}$	$E_{g(direct)}^{GLLB-SC}$	E_g^{HSE06}	m^*_h	m^*_e	prototype
$AlLaS_3$	1.67	1.67	1.47	-0.337	0.489	$SrZrS_3$(Y)
$BaHfS_3$	2.32	2.32	2.20	-0.255	0.414	$SrZrS_3$(B)
$BaZrS_3$	2.25	2.25	2.08	-0.749	0.426	$SrZrS_3$(B)
$BiLiS_3$	1.13	1.43	1.08	-0.209	0.455	$FePS_3$
$BiScS_3$	2.45	2.64	2.62	-0.318	0.520	$SrZrS_3$(Y)
$BiTlS_3$	1.36	1.98	1.30	-0.636	0.309	$FePS_3$
$HfGeS_3$	1.70	1.73	1.68	-0.568	0.256	$SrZrS_3$(Y)
$HfPbS_3$	2.11	2.24	1.96	-0.396	0.538	$SrZrS_3$(Y)
$HfSnS_3$	1.53	1.57	1.53	-0.408	0.270	$SrZrS_3$(Y)
$HfZnS_3$	2.03	2.47	1.98	-0.173	0.431	$FePS_3$
$LaSbS_3$	1.23	1.23	0.99	-0.439	0.167	$SrZrS_3$(Y)
$MgZrS_3$	2.21	2.32	2.06	-0.718	0.779	distorted
$PbZrS_3$	1.68	1.91	1.66	-0.434	0.525	$SrZrS_3$(B)
$ScSbS_3$	2.35	2.43	1.99	-0.502	0.258	$SrZrS_3$(Y)
$SnZrS_3$	1.76	1.98	1.56	-0.488	0.802	$PbPS_3$
$SrZrS_3$	2.49	2.49	2.30	-0.768	0.496	$SrZrS_3$(B)
$TaLiS_3$	1.98	2.00	2.06	-0.755	0.985	$FePS_3$
$TlScS_3$	1.60	1.76	1.62	-0.377	0.685	$YScS_3$
$YBiS_3$	2.17	2.24	2.04	-0.428	0.488	$SrZrS_3$(Y)
$YLaS_3$	1.87	1.87	1.57	-0.509	0.438	$SrZrS_3$(Y)
$ZrBaS_3$	1.69	1.96	1.62	-0.453	0.279	distorted
$ZrBaS_3$	1.79	1.79	1.54	-0.402	0.413	$SrZrS_3$(Y)
$ZrZnS_3$	1.91	1.97	1.87	-0.616	0.427	$FePS_3$

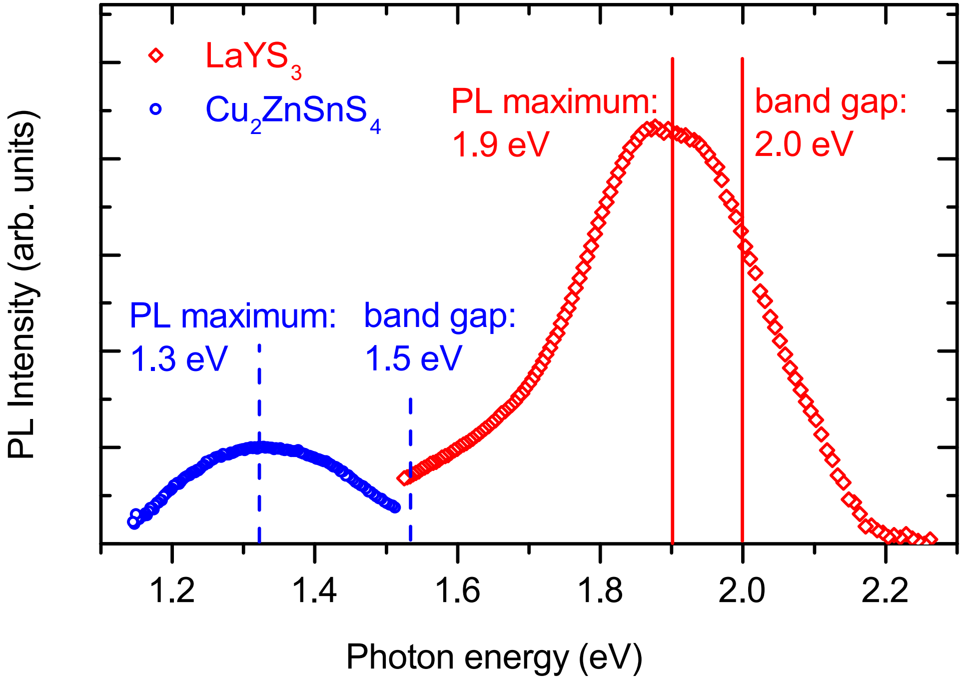
Kuhar, Thygesen, Jacobsen *et al.* Energy Env. Sci. **10**, 2579 (2019)

Synthesis of $LaYS_3$

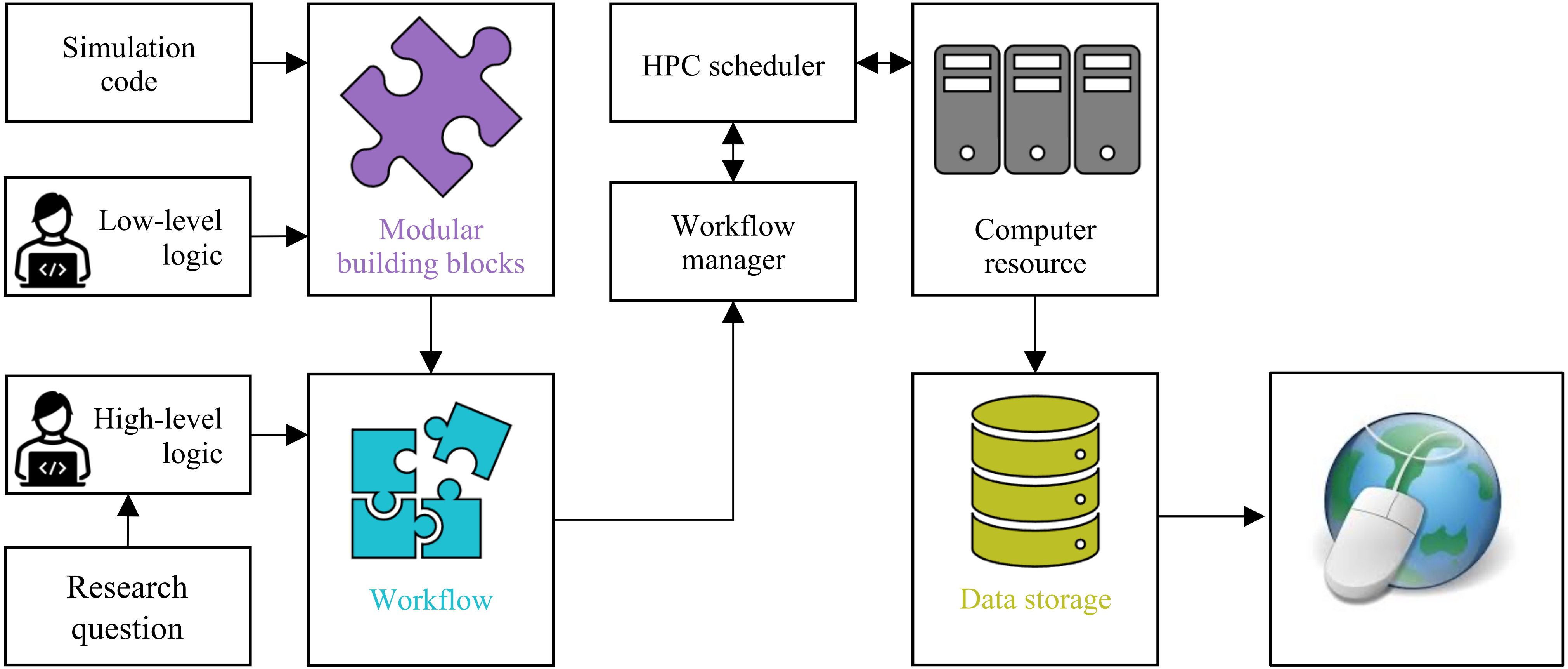


- XRD confirms the structure
- Optical absorption confirms the band gap of 2.0 eV
- PL confirms direct gap and absence of defects





TaskBlaster: Python framework for workflows

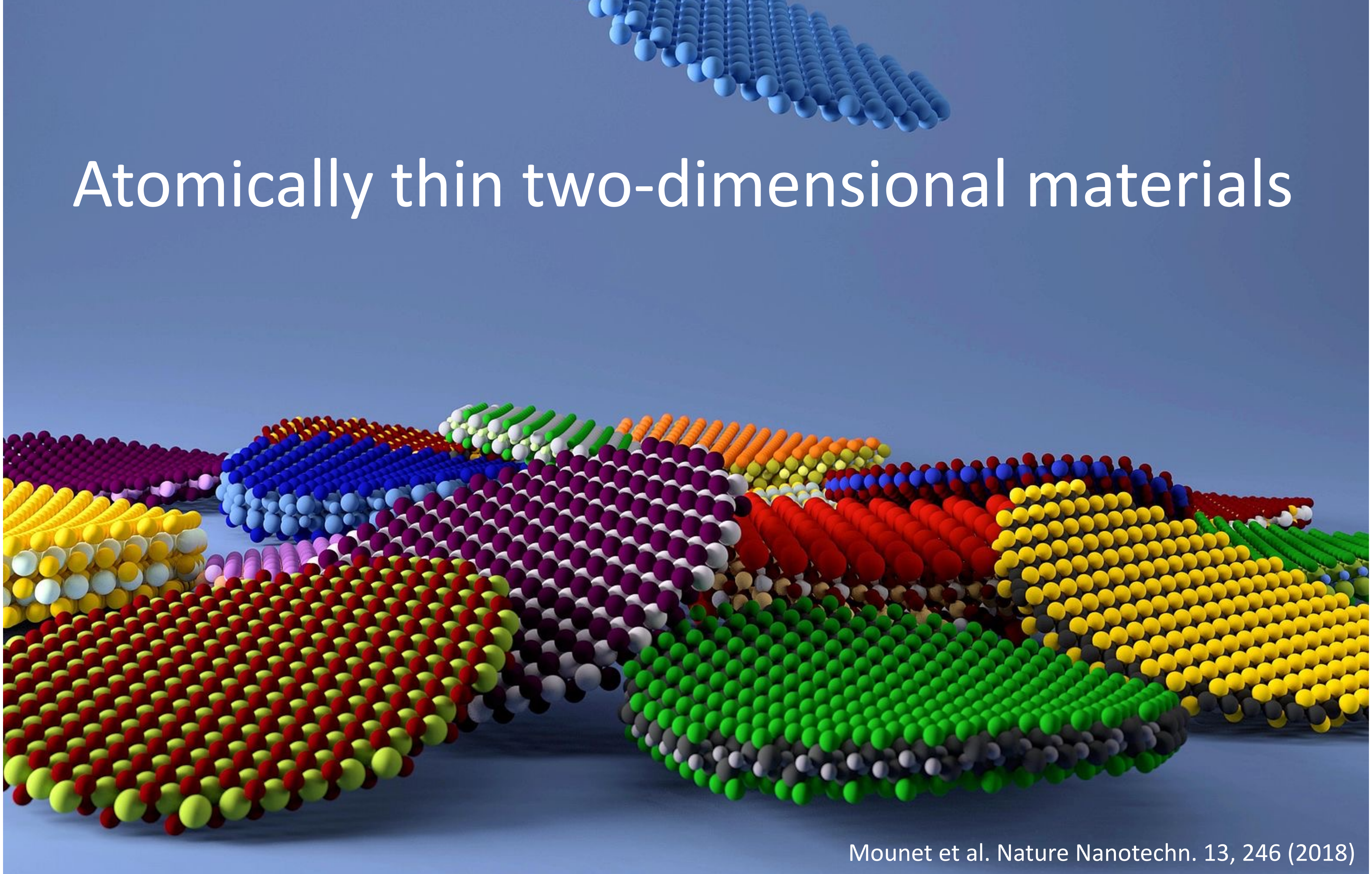


Scaling of TaskBlaster

- Demonstrated scaling of TaskBlaster to the entire LUMI supercomputer (g-partition).
- Running ca. 14000 GPAW-GPU calculations concurrently.
- Not trivial! Experience used to develop a LUMI-compatible version of ASR

- Approx. 10k GPUs
- 375 petaflops (or 1.5 mio. laptops)
- #3 in the world
- Operated by CSC, Finland

Atomically thin two-dimensional materials



Mounet et al. Nature Nanotechn. 13, 246 (2018)

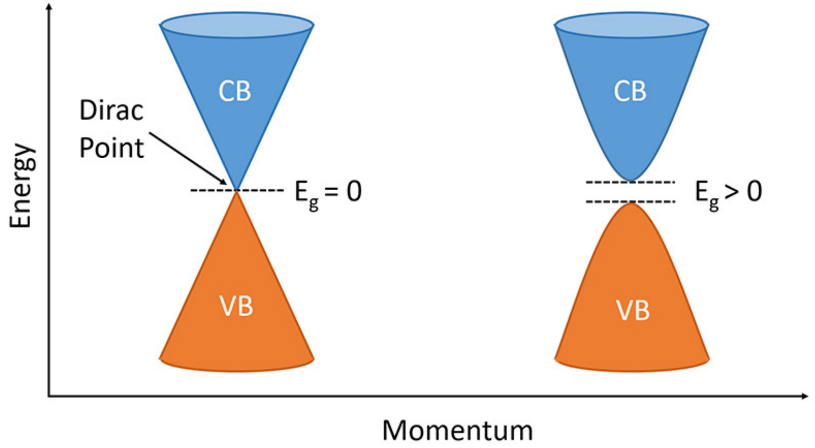
Graphene – the wonder material

Particle with mass:

Massless particle:

$$E = v_F p$$

$$E = \frac{p^2}{2m}$$



Electrons in graphene move as if their mass was zero!

- Almost transparent (absorbs ca 2% light)
- Best electrical conductor
- Strongest material (100 x steel)
- Extremely flexible
- **Exotic physics**

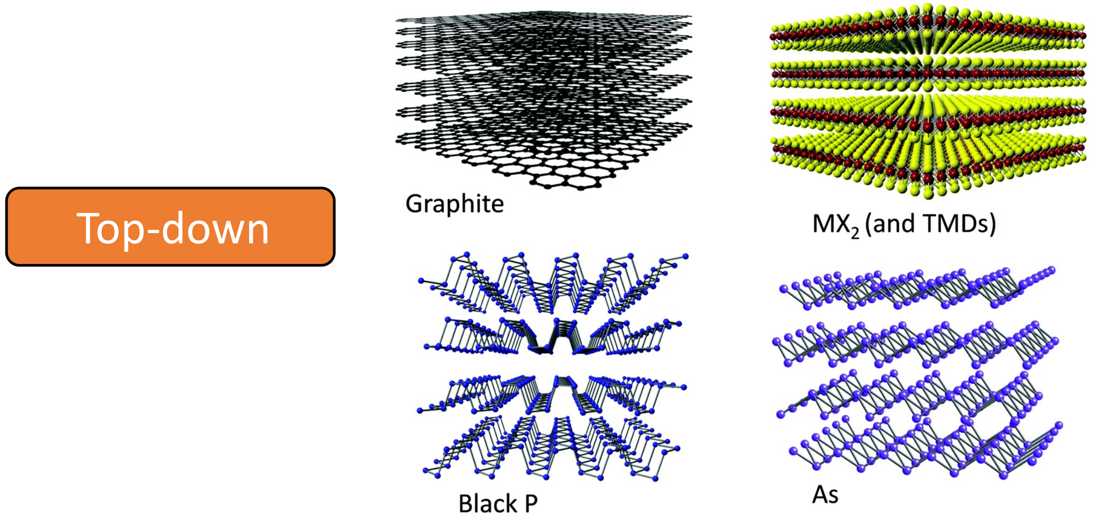
Nobel Prize in physics 2010

K. Novoselov

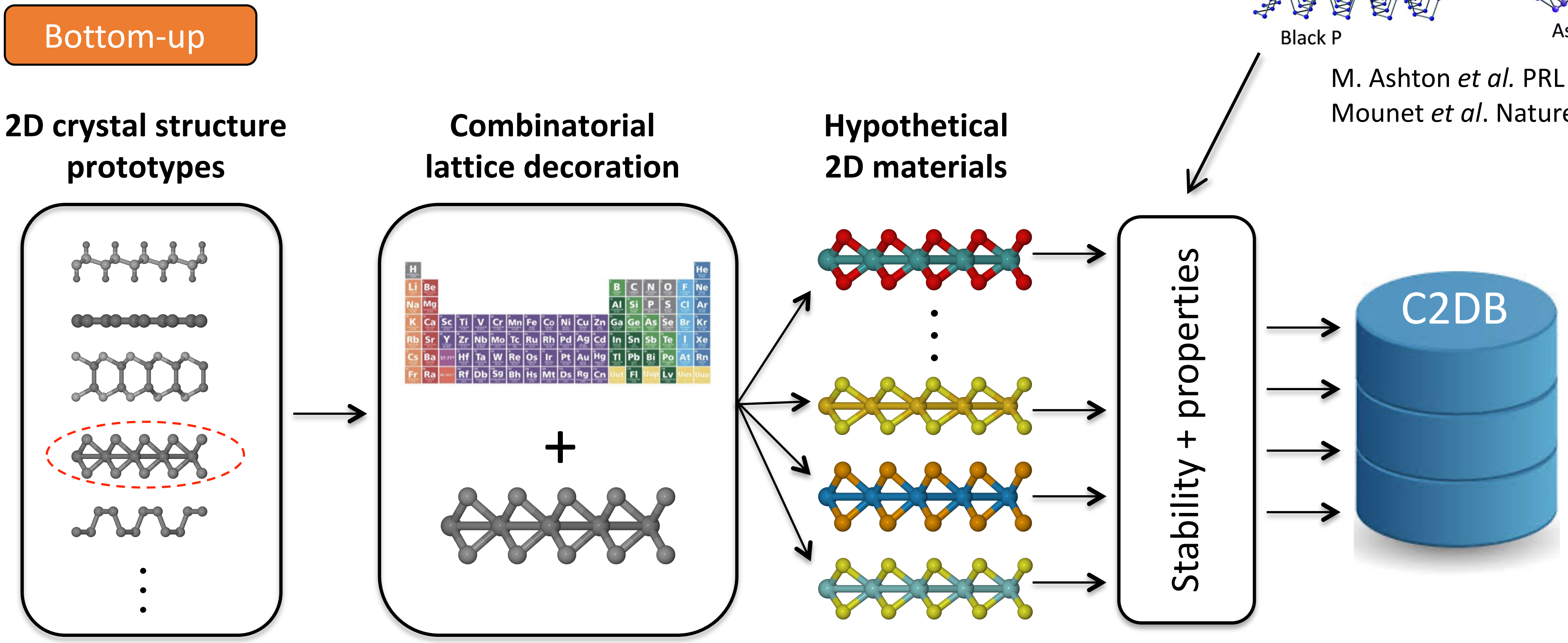
A. Geim

C2DB The Computational 2D Materials Database

Known layered bulk compounds (~600)



M. Ashton *et al.* PRL **118**, 106101 (2017)
Mounet *et al*. Nature Nano. **13**, 246 (2018)

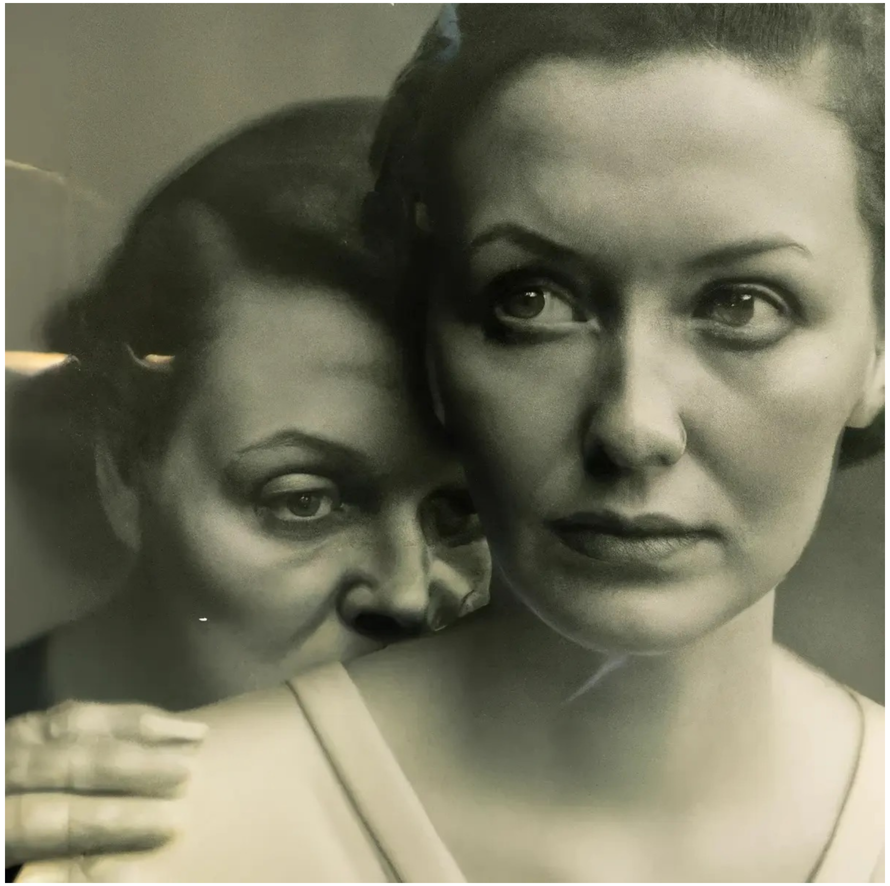


Deep generative models

Artistic style transfer

Natural language processing

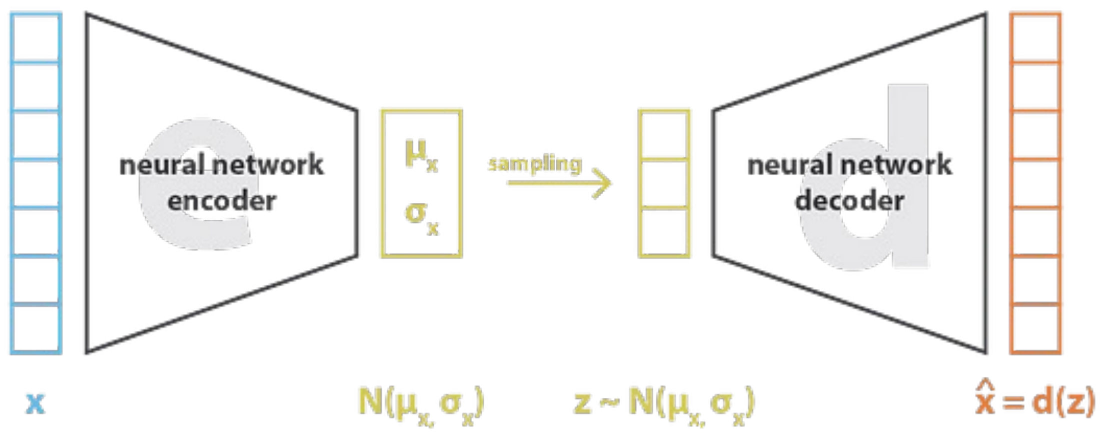
Image generation



Boris Eldagsen refuses to receive Sony World Photography Award for this AI-generated image

Deep generative models

Variational autoencoder

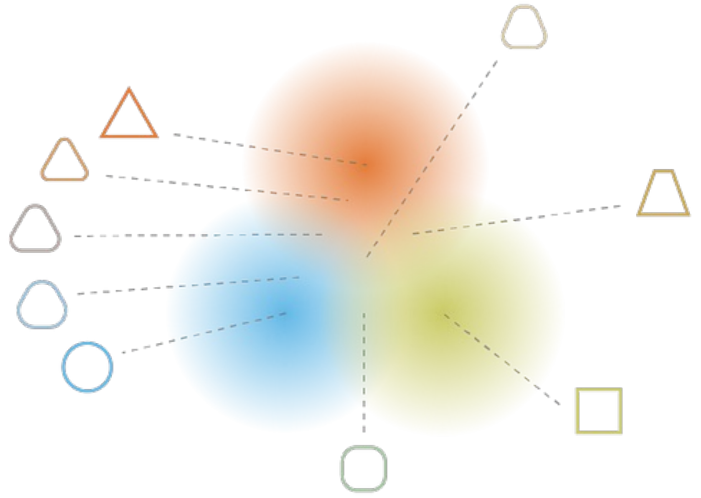


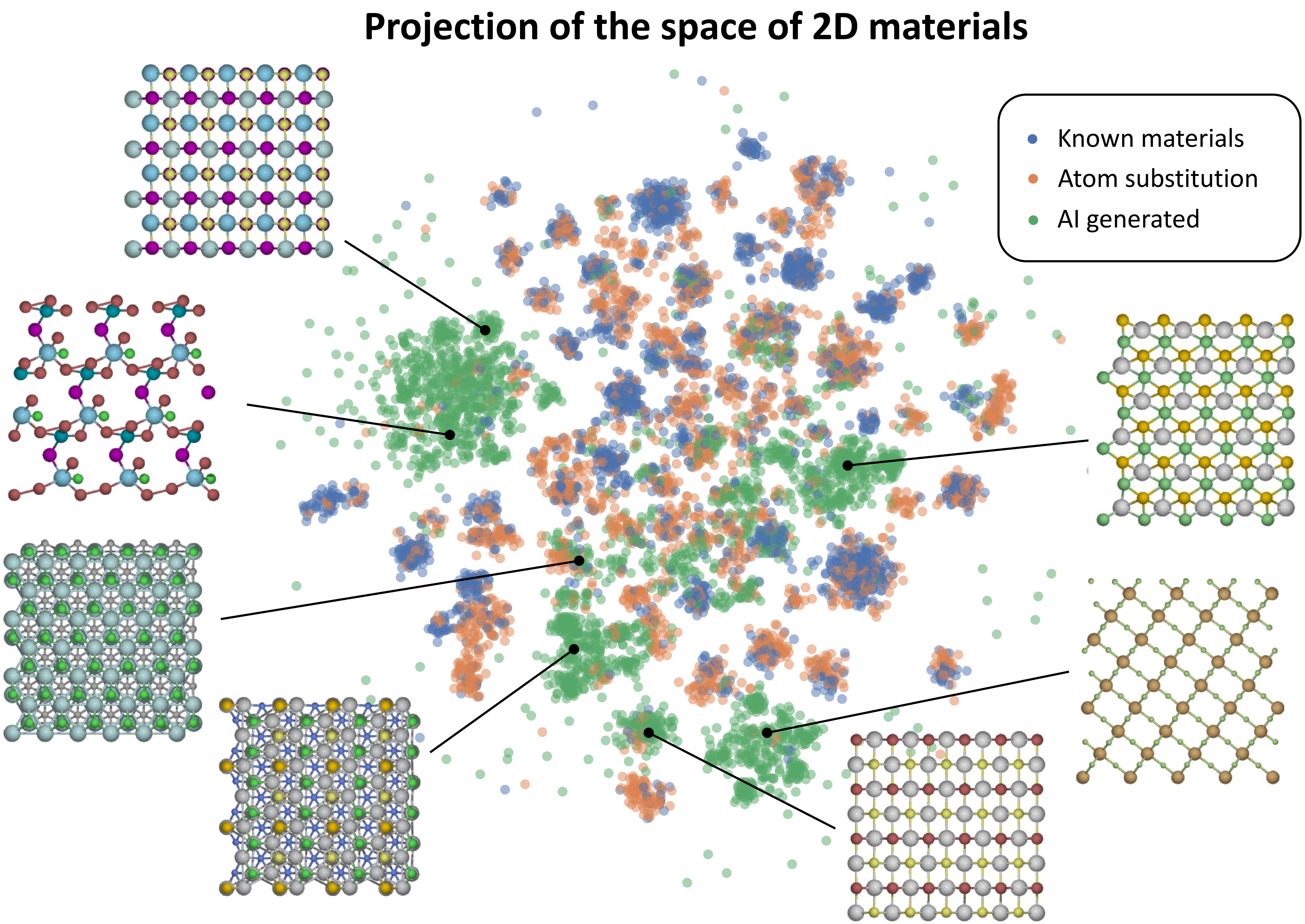
$$\text{loss} = \| x - \hat{x} \|^2 + \mathrm{KL}[\, N(\mu_x, \sigma_x), N(0, I)\,] = \| x - d(z) \|^2 + \mathrm{KL}[\, N(\mu_x, \sigma_x), N(0, I)\,]$$

Reconstruction term

KL-regularisation (relative entropy)

Input, x, is encoded as a normal distributions, $N(\mu_x, \sigma_x)$, over latent space




Projection of the space of 2D materials
Known materials
Atom substitution
AI generated

CAMD section members

Kristian Thygesen
Prof. Head of section

Jakob Schiøtz
Professor

Karsten Jacobsen
Professor

Thomas Olsen
Assoc. Professor

Jens Jørgen Mortensen
Software developer

Ole Holm Nielsen
Senior HPC officer

erc European Research Council

VILLUM FONDEN

HORIZON EUROPE

novo nordisk fonden