Computational screening of solar energy materials

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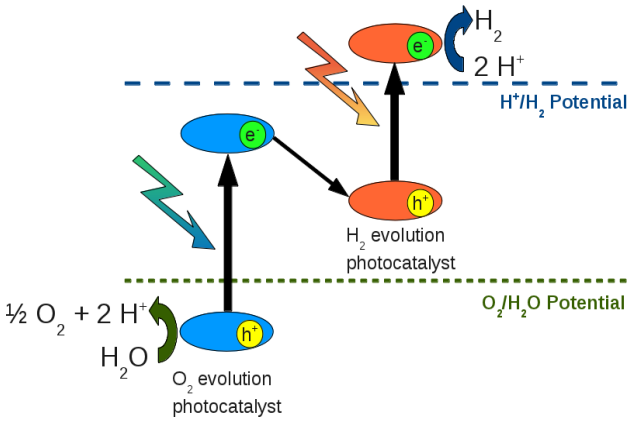
Computational materials screening

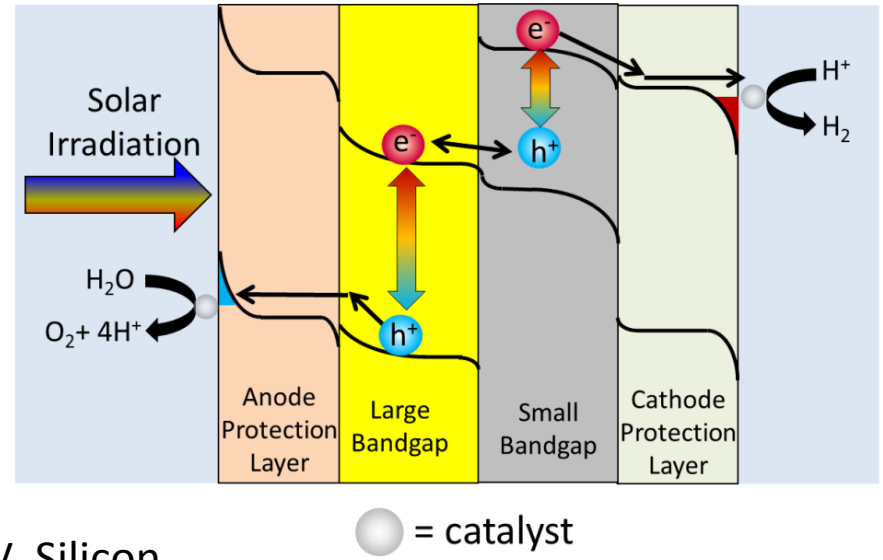
- What is the problem we want to solve? What are the properties of the “dream material”?
- What do we compute – descriptors?
- How do we search in the materials space?

Light induced water splitting
Tandem cells



Produce hydrogen as a fuel
(and save the World)

Light absorbing materials –
small/large band gaps
Protection layers
Catalysts
p-n junctions



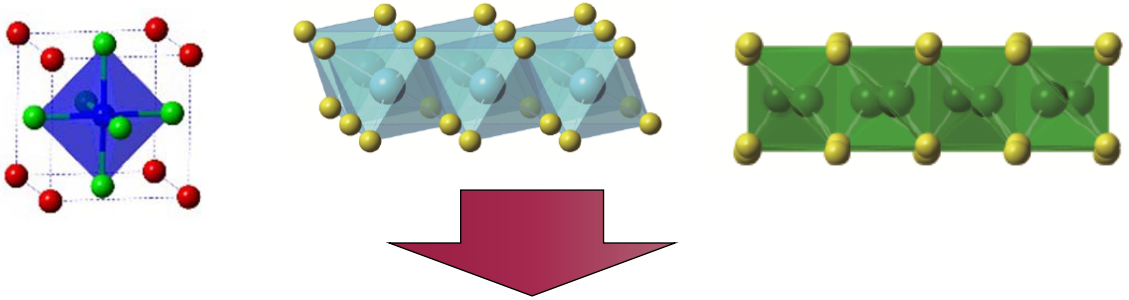
The challenge:
Small bandgap semiconductor: ~1.1 eV Silicon
Large bandgap semiconductor: ~1.8 eV **?????**

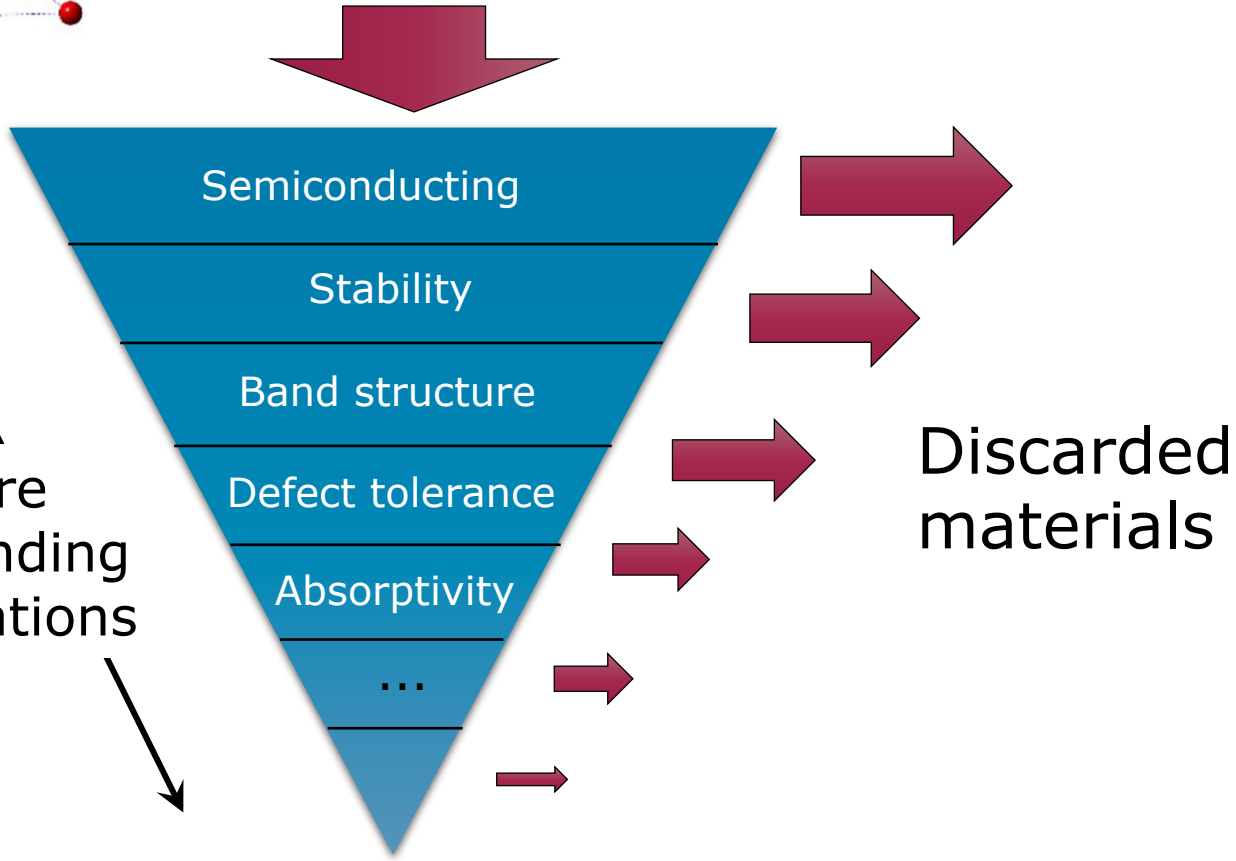
Searching for light absorbing materials

- Mapping out a particular class of materials
  - Perovskites
- Known materials
  - Inorganic crystal structure database (ICSD)
- Searches guided by correlations in materials space
  - Machine learning

Sulfide perovskites
Screening funnel

Initial structures:
(Experimental databases or hypothetical)





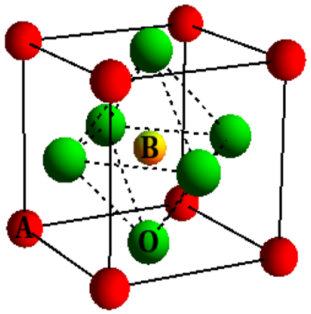
Candidates for experimental investigation

Identify semiconducting ABS_3 compounds

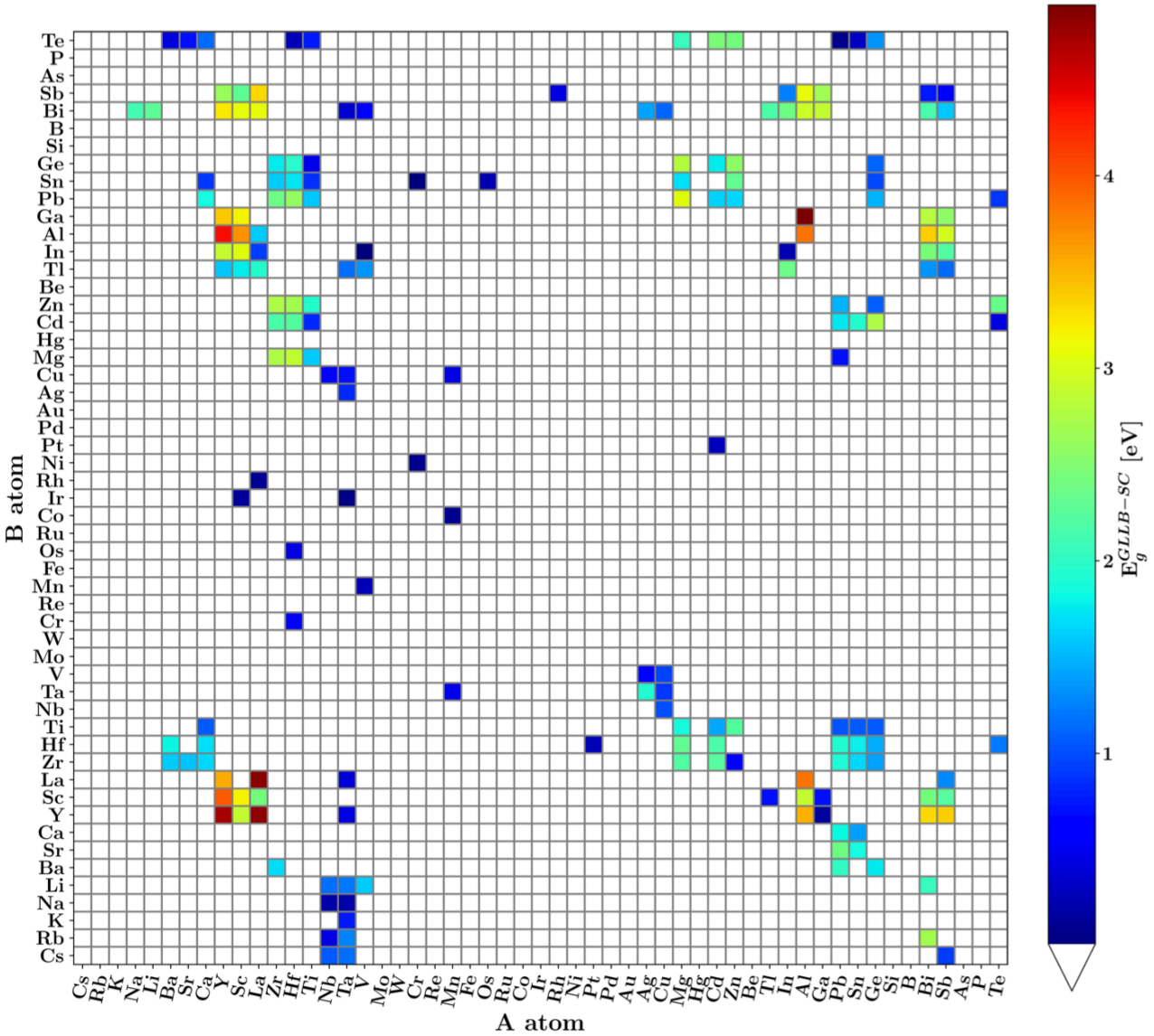
53 metal atoms

53 * 53 = 2809 compounds

Test for bandgap > 0
in distorted 5 atom unit cell

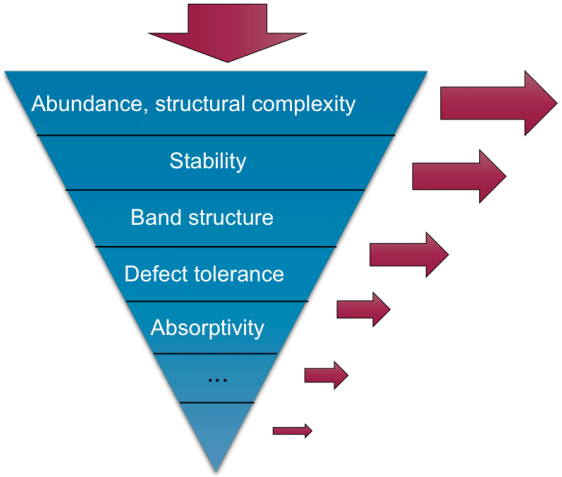


129 compounds identified

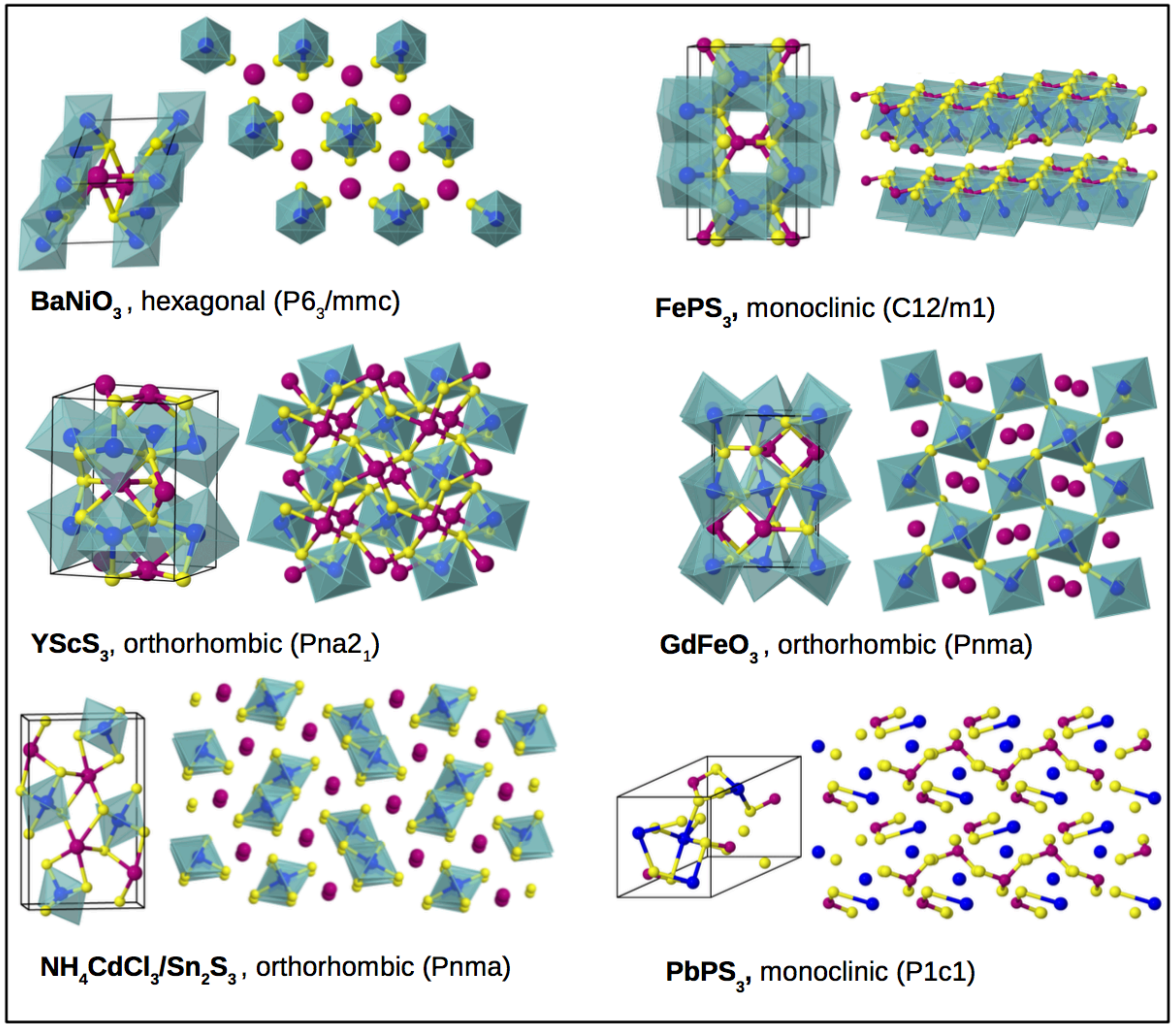


Perovskite sulfides

Screening funnel



Most common ABS_3 structures in ICSD



Y.-Y. Sun et al.,
Nano Lett **15**, 581 (2015)

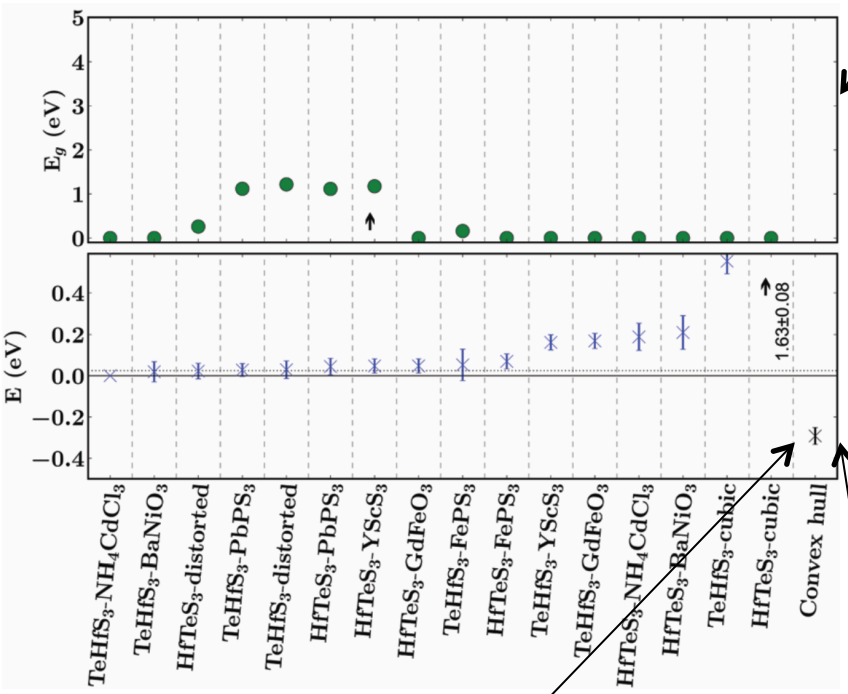
Kuhar, Crovetto, Pandey, Thygesen, Seger, Vesborg, Hansen, Chorkendorff, Jacobsen,
Energy and Environmental Science, **10**, 2579 (2017).

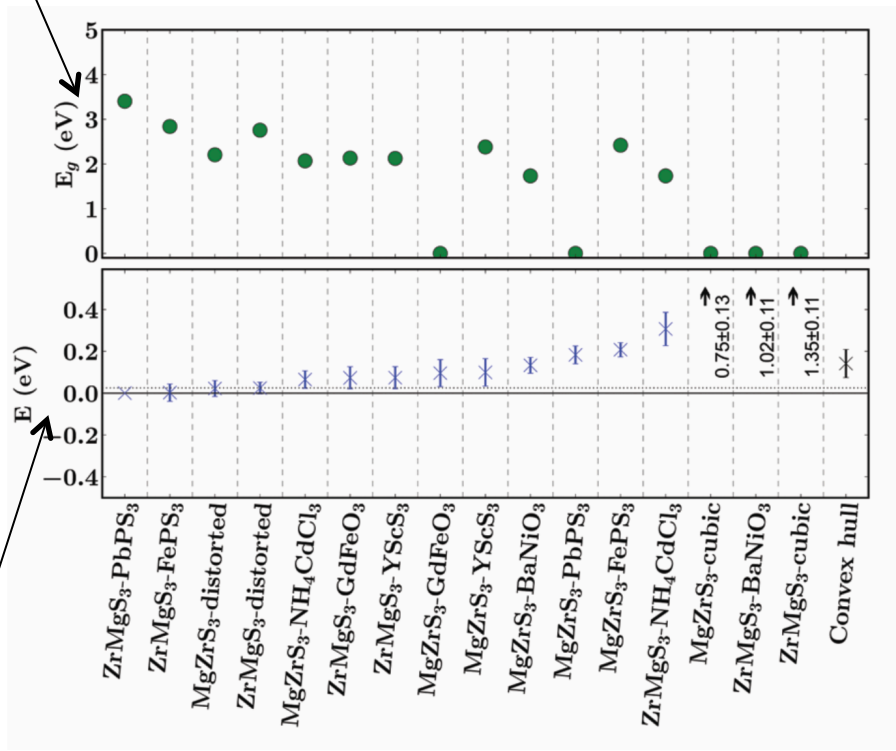
Stability

$TeHfS_3$

Band gap

$ZrMgS_3$





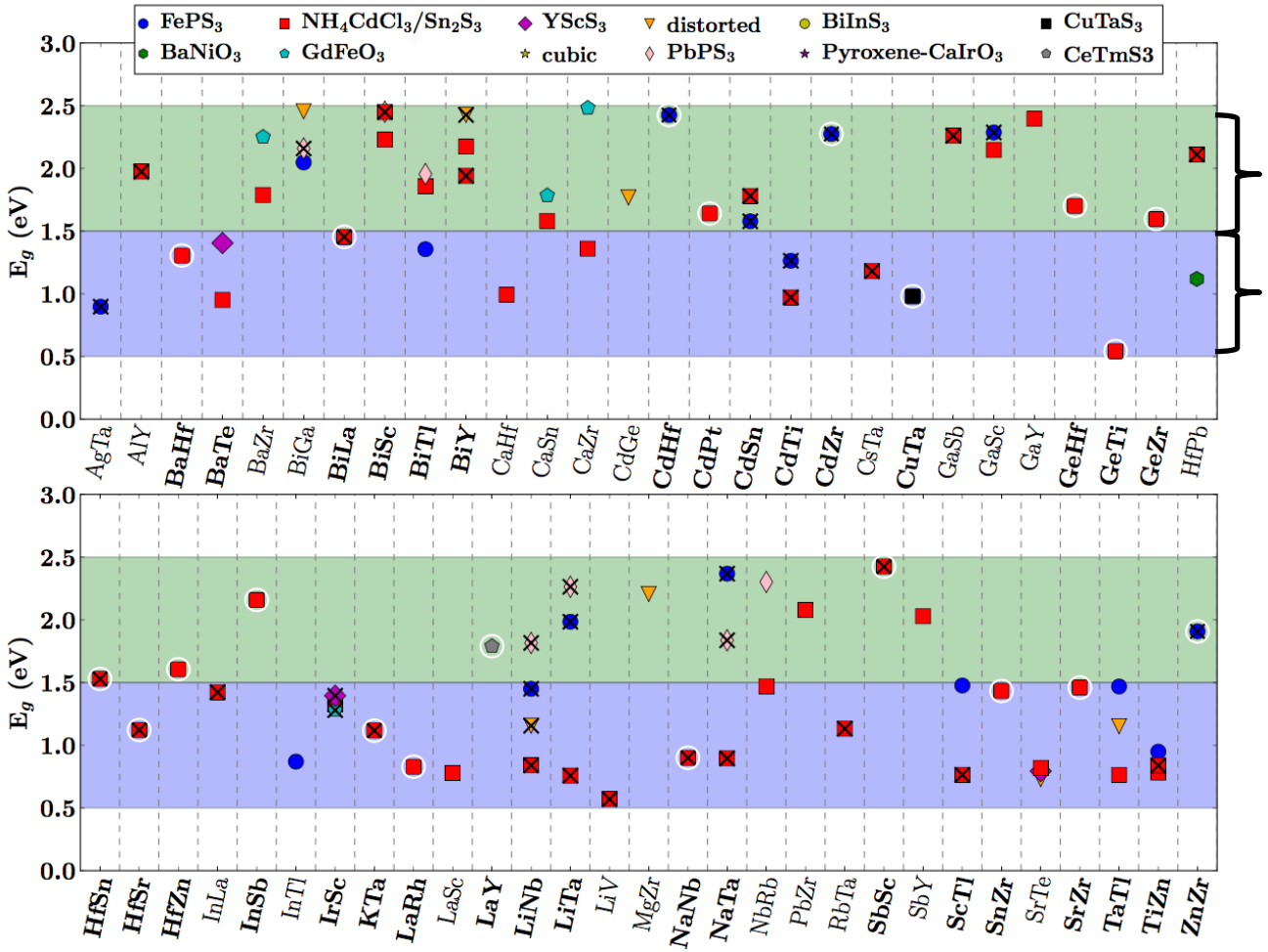
Energy of formation

Unstable relative to decomposition

Use mBEEF uncertainties on energy of formation to improve predictions.

Band gaps (calculated with GLLB)

Band gaps



White circle: One structure significantly more stable

Bold: All low-energy structures with interesting gaps

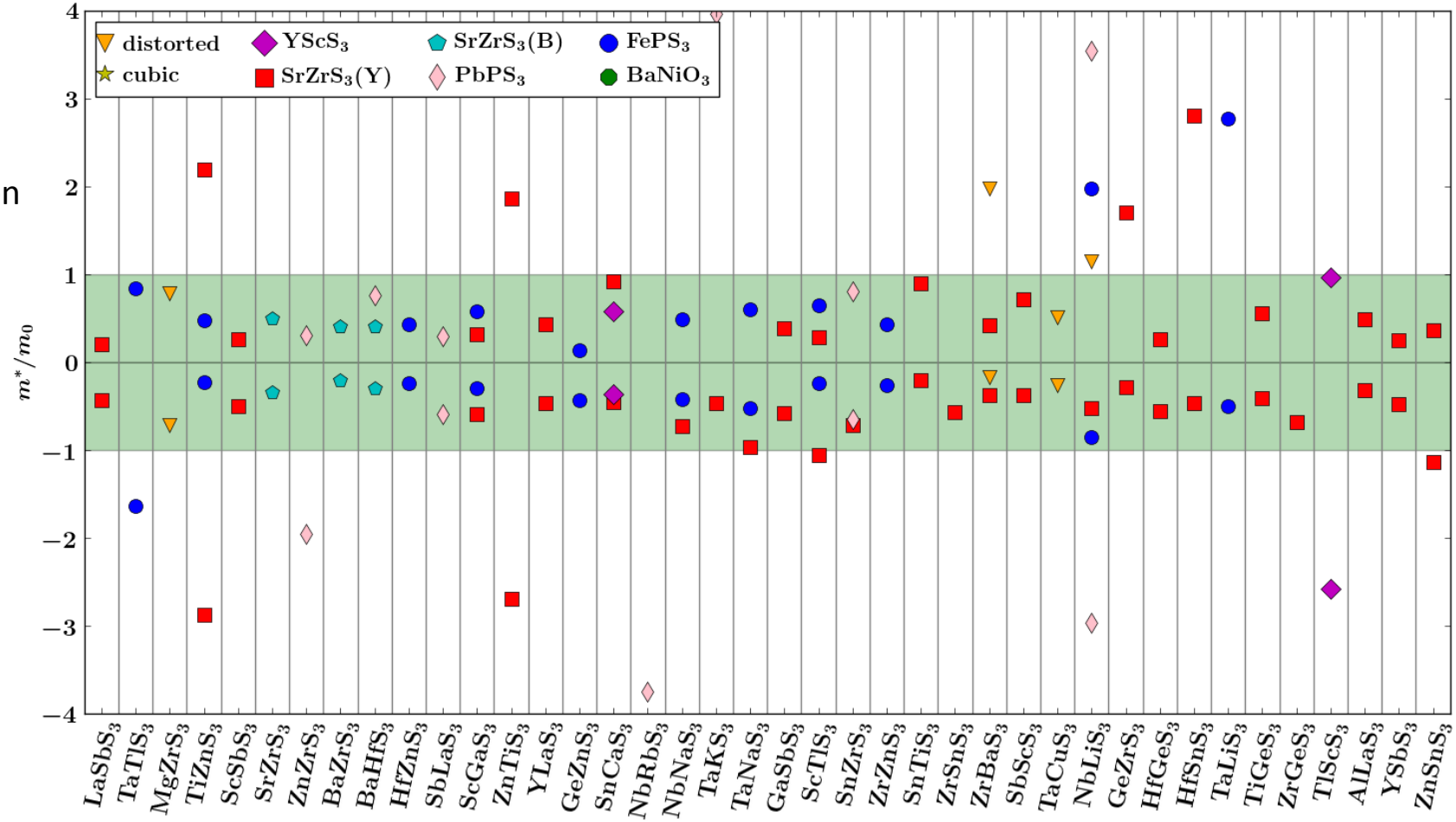
Crosses: AB->BA

Mobility – effective masses

Mobility $\mu \propto 1/m$

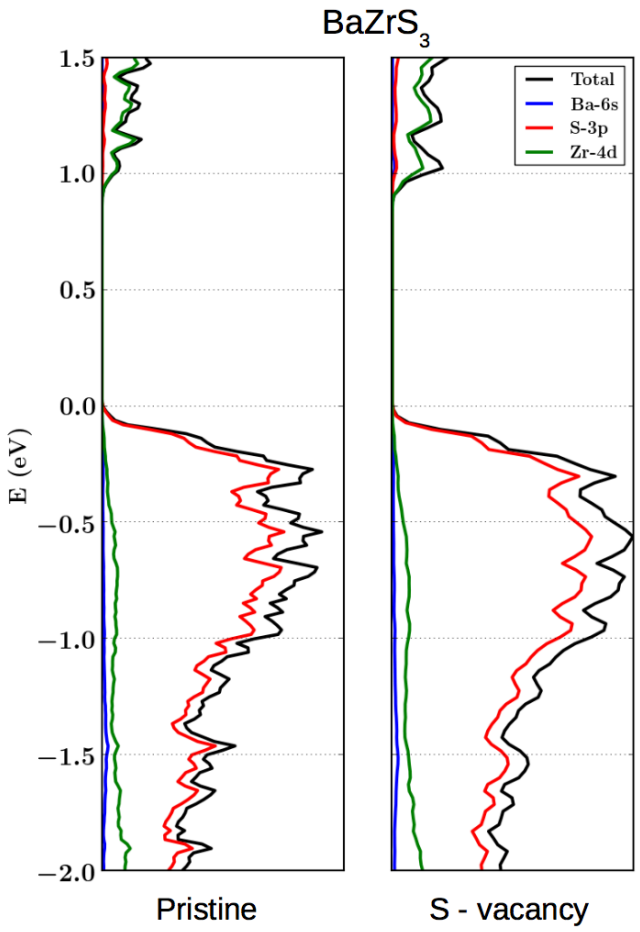
electron

hole

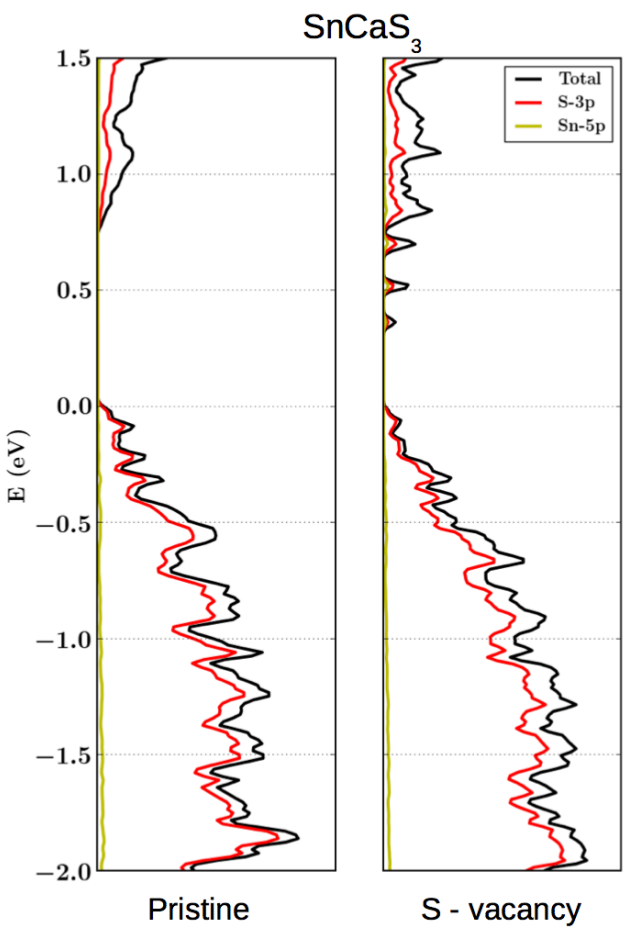


Defect tolerance/sensitivity of ABS_3 compounds
Electronic density-of-states

Defect tolerant



No mid-gap states introduced by vacancy



Mid-gap states introduced by vacancy

Defect sensitive

Interesting sulfides for photoabsorption (15 candidate materials)

	formula	$E_g^{GLLB-SC}$	$E_{g(direct)}^{GLLB-SC}$	E_g^{HSE06}	m^*_h	m^*_e	prototype
Ba-Hf	$BaHfS_3$	1.31	1.31	1.60	-0.347	0.943	NH_4CdCl_3/Sn_2S_3
Ba-Zr	$BaZrS_3$	2.25	2.25	2.08	-0.749	0.426	$GdFeO_3$
Bi-Tl	$BiTlS_3$	1.36	1.98	1.30	-0.636	0.309	$FePS_3$
Ca-Hf	$CaHfS_3$	0.99	0.99	1.36	-0.336	0.759	NH_4CdCl_3/Sn_2S_3
Ca-Sn	$CaSnS_3$	1.58	1.93	1.45	-0.606	0.943	NH_4CdCl_3/Sn_2S_3
Ca-Zr	$CaZrS_3$	1.36	1.36	1.32	-0.765	0.884	NH_4CdCl_3/Sn_2S_3
Hf-Sr	$SrHfS_3$	1.12	1.12	1.45	-0.327	0.811	NH_4CdCl_3/Sn_2S_3
Hf-Pb	$HfPbS_3$	1.12	1.63	0.94	-0.275	0.235	$BaNiO_3$
La-Y	$LaYS_3$	1.79	1.79	1.47	-0.670	0.490	$CeTmS_3$
Li-Ta	$TaLiS_3$	1.98	2.00	2.06	-0.755	0.985	$FePS_3$
Mg-Zr	$MgZrS_3$	2.21	2.32	2.06	-0.718	0.779	distorted
Sb-Y	$SbYS_3$	2.03	2.09	1.67	-0.372	0.484	NH_4CdCl_3/Sn_2S_3
Sr-Zr	$SrZrS_3$	1.46	1.46	1.37	-0.644	3.115	NH_4CdCl_3/Sn_2S_3
Ta-Tl	$TaTlS_3$	1.15	1.15	1.35	-0.297	0.241	distorted
Zn-Zr	$ZrZnS_3$	1.91	1.97	1.87	-0.616	0.420	$FePS_3$

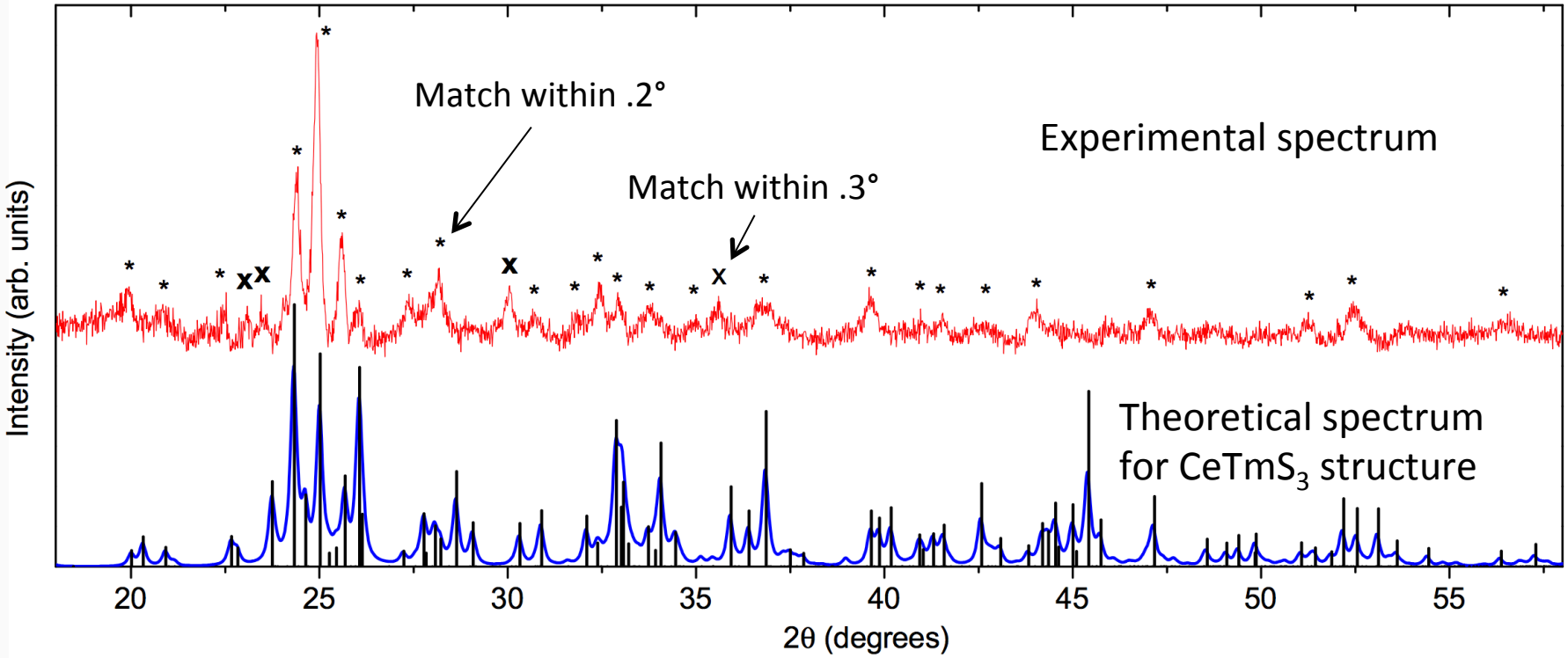
Bold: all low-energy phases have relevant band gaps

$BaZrS_3$: W. Meng et al., *Chem Mater*, **28**, 821 (2016)

Kuhar, Crovetto, Pandey, Thygesen, Seger, Vesborg, Hansen, Chorkendorff, Jacobsen, Energy and Environmental Science, **10**, 2579 (2017).

$LaYS_3$
Synthesis and characterization

Thin films produced by deposition of La and Y followed by sulfurization.

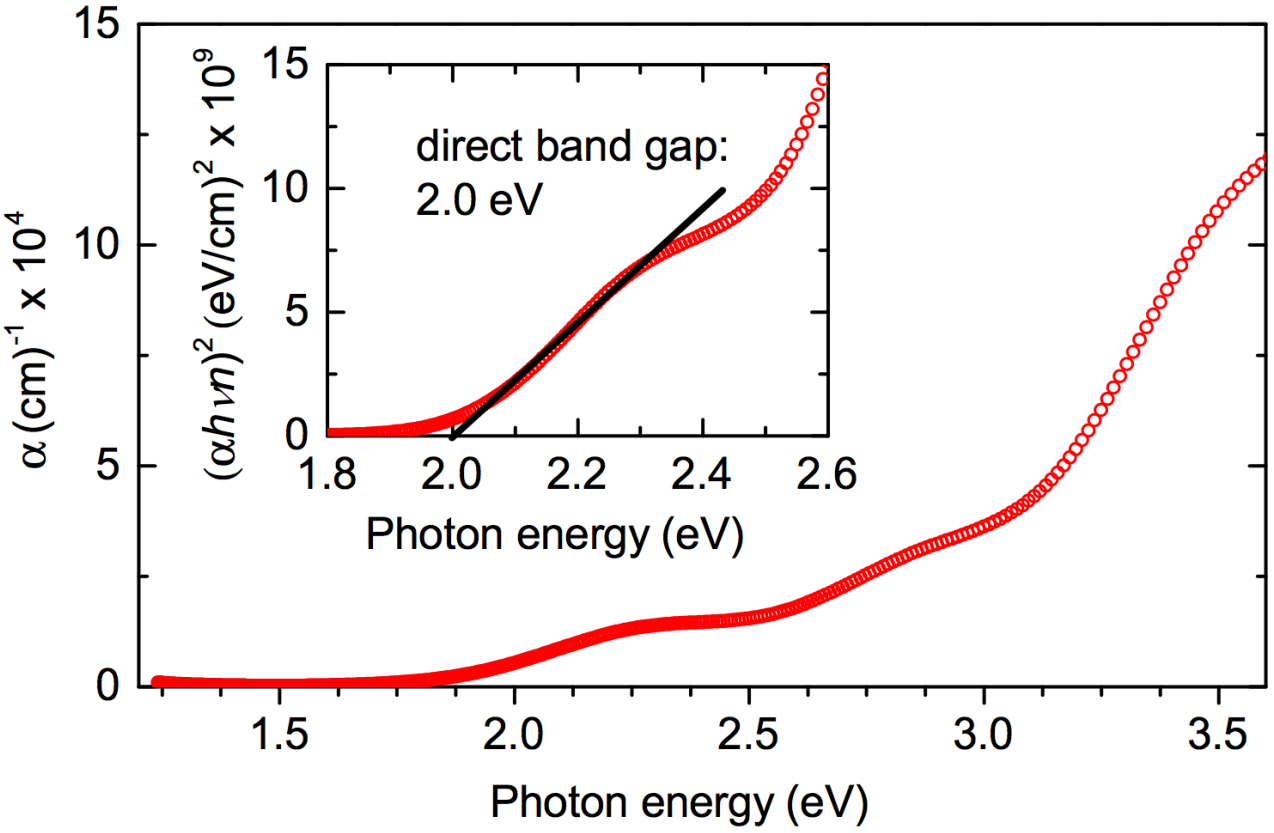


Theoretical spectrum calculated for random orientation.
Blue curve with .2° smearing.

Kuhar, Crovetto, Pandey, Thygesen, Seger, Vesborg, Hansen, Chorkendorff, Jacobsen,
Energy and Environmental Science, **10**, 2579 (2017).

$LaYS_3$
Band gap

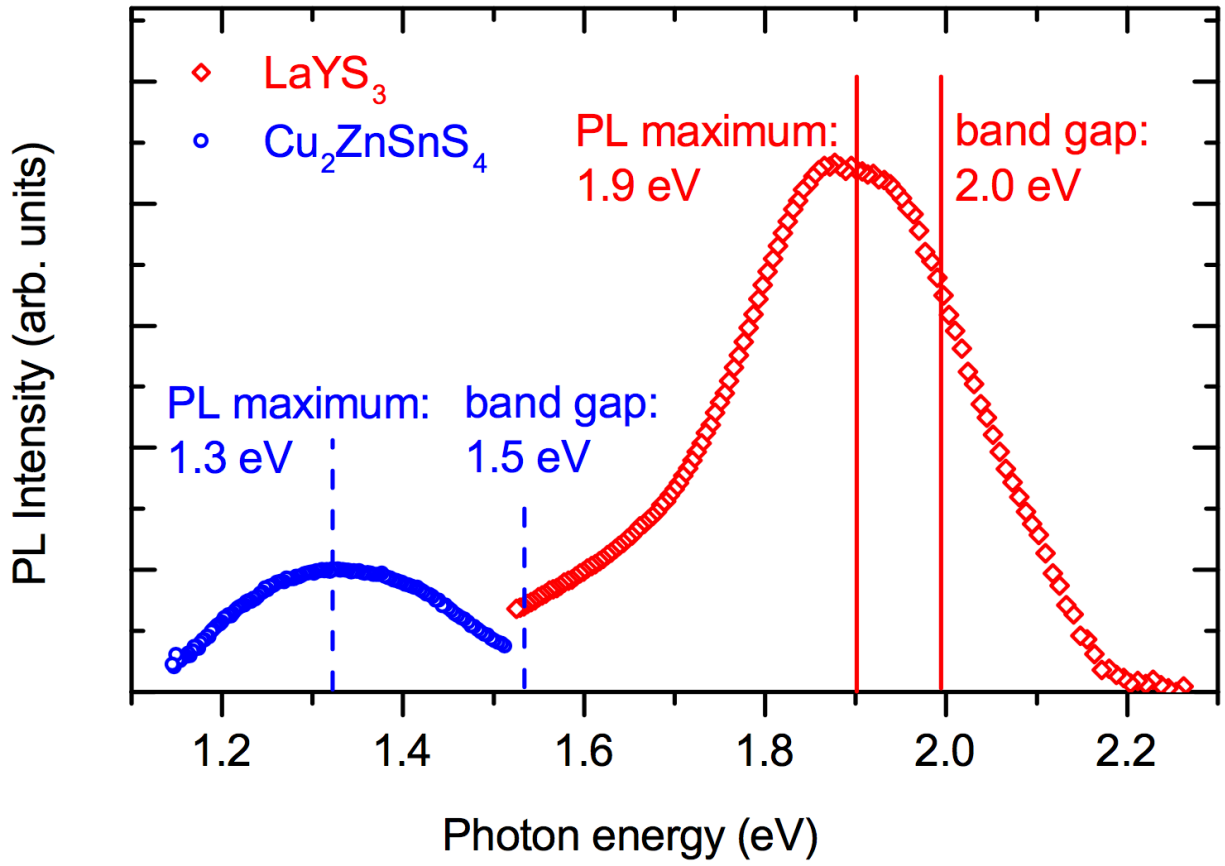
Spectroscopic ellipsometry – light absorption coefficient



Direct band gap determined from absorption coefficient and refractive index

Kuhar, Crovetto, Pandey, Thygesen, Seger, Vesborg, Hansen, Chorkendorff, Jacobsen,
Energy and Environmental Science, **10**, 2579 (2017).

$LaYS_3$ Photoluminescence



Strong photoluminescence signal

Defects not giving rise to non-radiative processes.

Performance in water-splitting device being investigated. (Andrea Crovetto).

Limitations

- Limited compositions (ABS_3)
- Limited number of structures (6)
- Absorption: band gap with GLLB (±0.4 eV)
- Mobility: effective mass
- Defects: only neutral vacancies

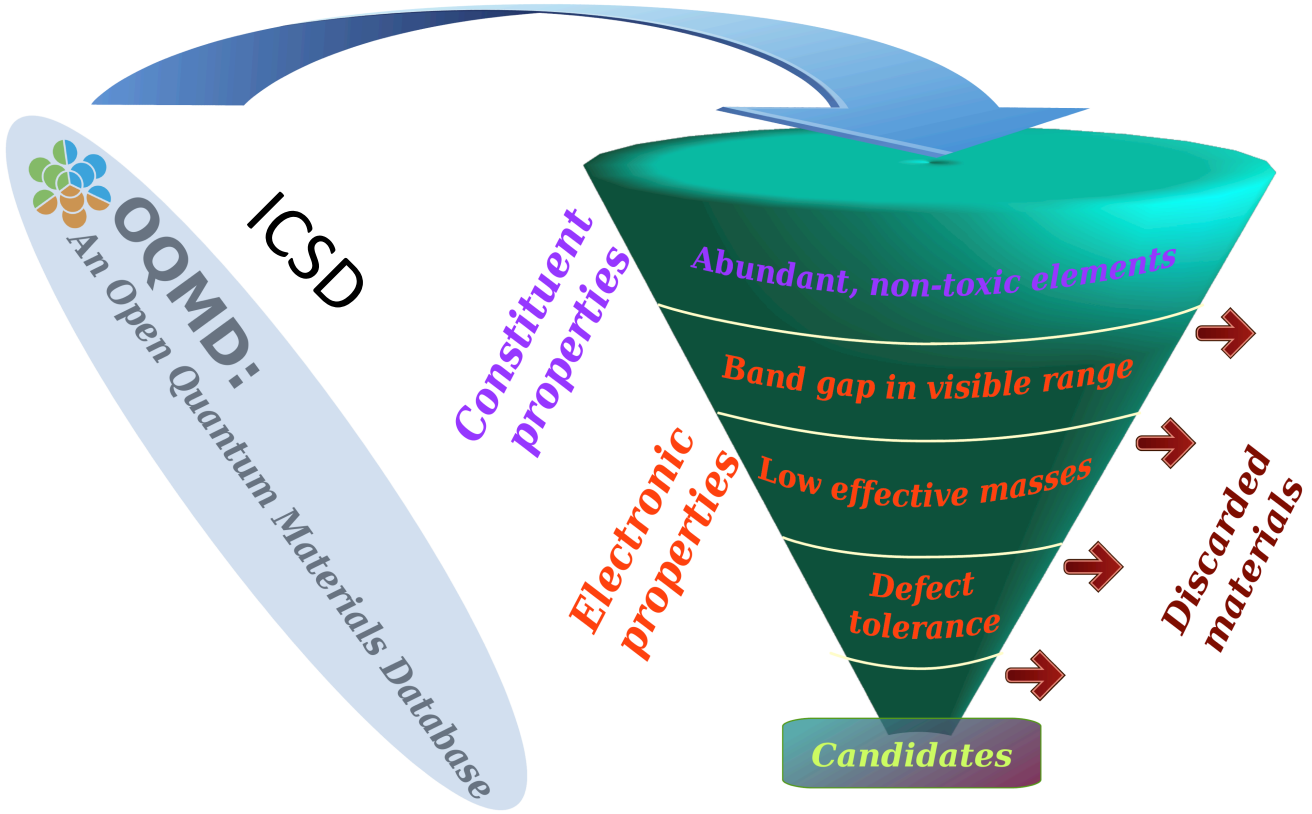
- But still useful:
  - 2809 -> 15 materials

Screening of known materials for photovoltaics or water splitting

Advantages:

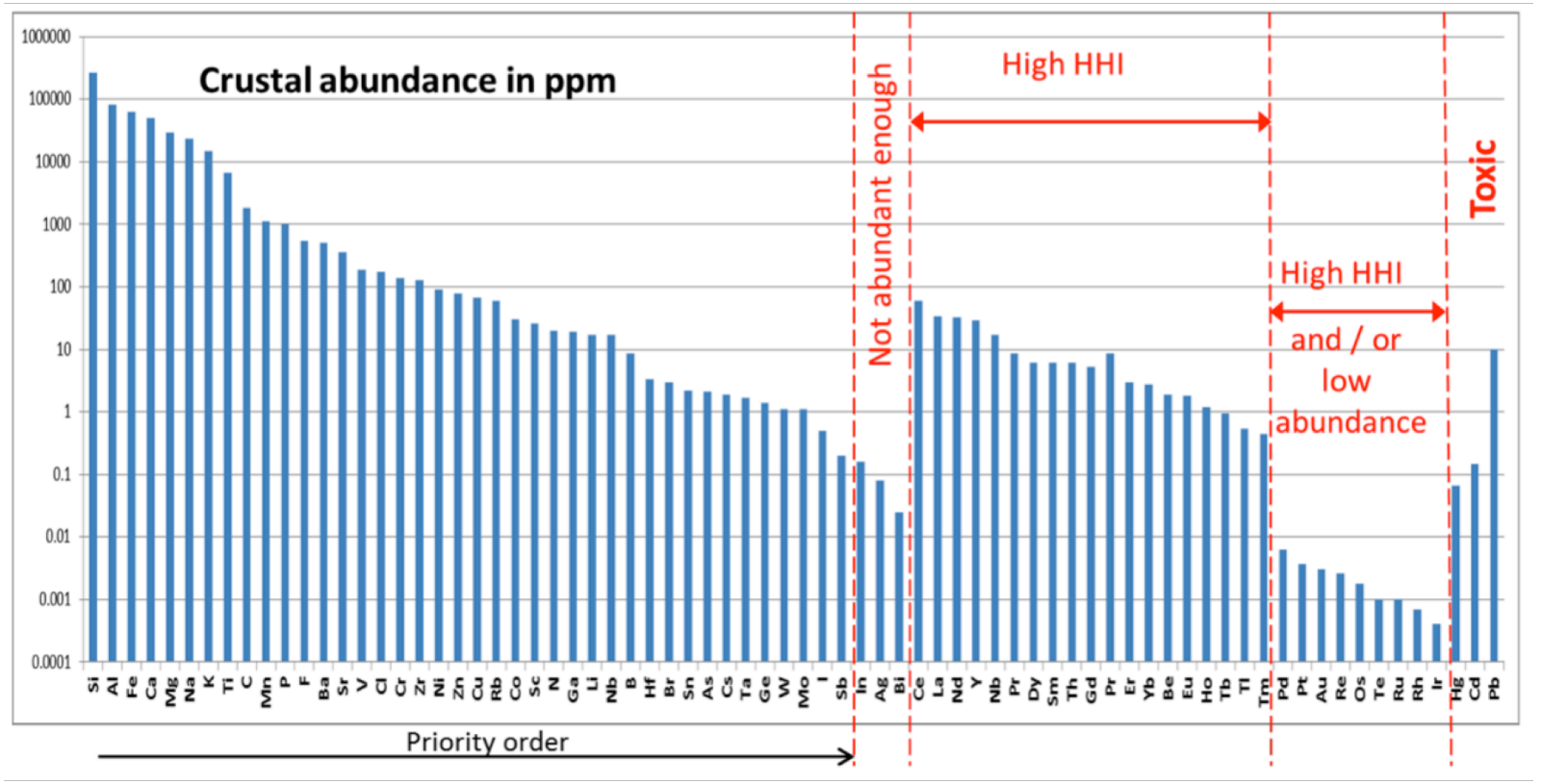
Materials known to be stable or metastable

Known synthesis procedures



Kuhar, Pandey, Thygesen, Jacobsen, ACS Energy Letters, doi:10.1021.acsenergylett.7b01312 (2018).

Abundance and Herfindahl–Hirschman index



Only abundant elements without a monopoly market.

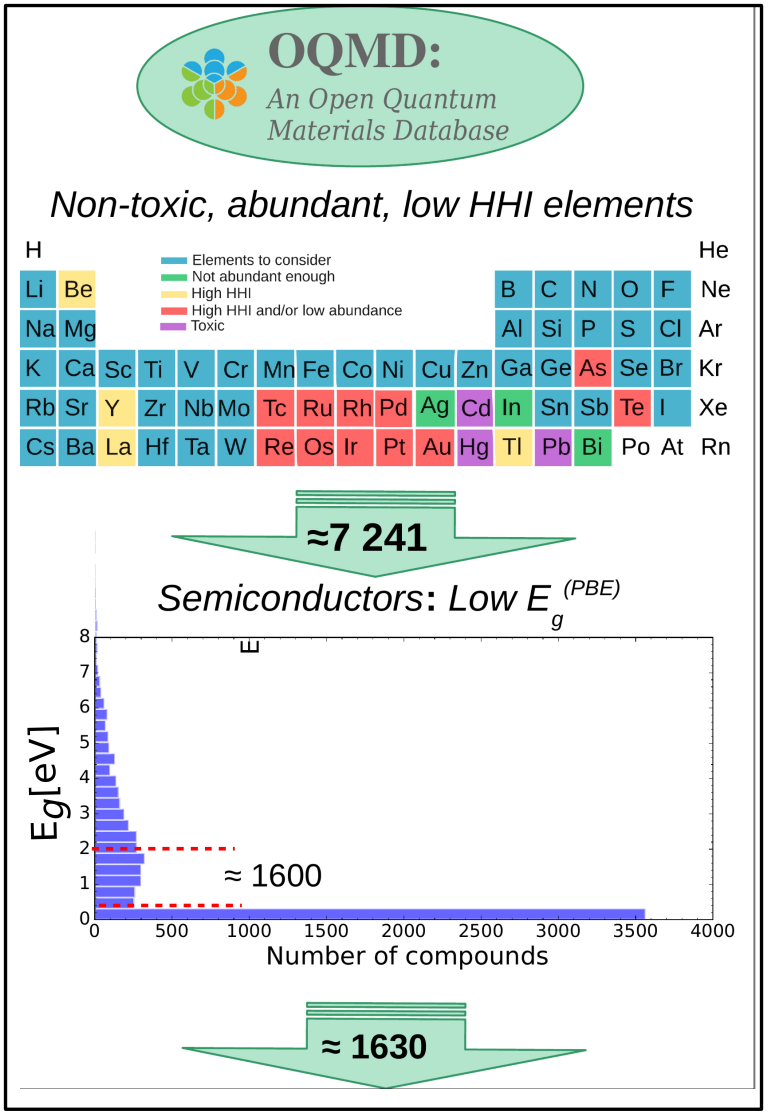
Screening funnel

Toxicity, abundance, HHI

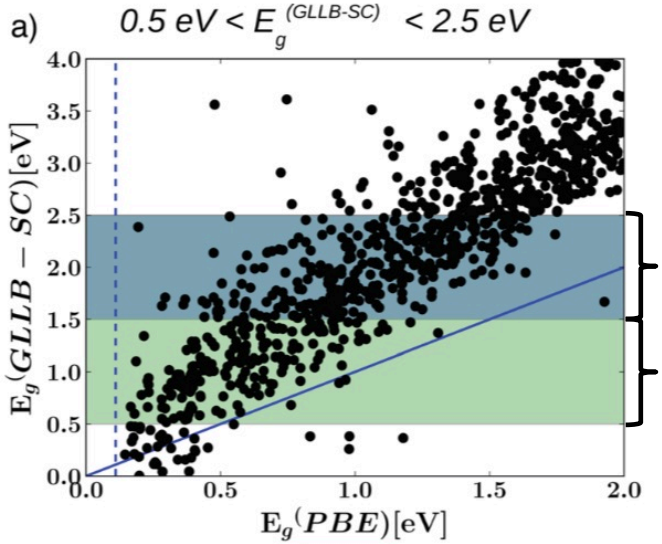
Binary or ternary

Materials in ICSD and OQMD

PBE band gap available in OQMD
$0 < E_g < 2$ eV



Screening funnel (cont.)



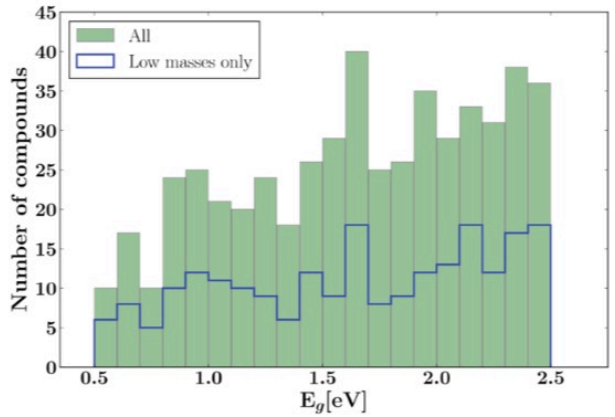
Band gap calculated with GLLB

Water splitting

Photovoltaics

518

b) *Low carrier effective masses*



222

222

Defect sensitivity

c)

Tolerant

Sensitive

74

Candidates

Kuhar, Pandey, Thygesen, Jacobsen,
ACS Energy Letters, doi:10.1021.acsenergylett.7b01312 (2018).

Screening results (74 materials)

formula	$E_g^{GLLB-SC}$ (eV)	$E_{g(direct)}^{GLLB-SC}$ (eV)	m_h^* (m_e)	m_e^* (m_e)
Al_2MgSe_4*	2.47	2.47	0.38	0.21
$(B_{12}S)$*	0.58	0.75	0.40	0.29
Ba_3P_4	1.07	1.07	0.95	0.97
Ba_3SbN	2.05	2.05	0.18	0.25
Ba_5Sb_4	0.94	1.27	0.66	0.36
Ba_4SnP_4	1.78	1.79	0.32	0.47
BaCaSn	0.88	0.88	0.34	0.73
BaLiP	1.98	1.98	0.16	0.16
$BaZrN_2$	2.45	2.45	0.38	0.28
$BaZrS_3$	2.34	2.34	0.35	0.43
Ca_3NP	2.46	2.46	0.21	0.29
CaLiSb	1.36	1.36	0.13	0.40
Cs_2SnI_6*	0.77	0.77	0.84	0.26
Cs_3Sb	2.45	2.75	0.76	0.23
Cs_6AlSb_3	2.11	2.21	0.91	0.28
Cs_6GaSb_3	1.84	1.94	0.99	0.29
$CsCuSe_4$	1.94	2.01	0.48	0.26
$CsGe\ Cl_3$	2.31	2.31	0.27	0.29
$CsNaGe_2$	2.48	2.51	0.35	0.51
$CsSnBr_3$	0.99	0.99	0.09	0.08
•	•	•	•	•
•	•	•	•	•

Antiperovskite →

Known perovskites →

Some interesting candidates for water splitting:
Hf_3N_4, NbI_5, **SrS_3**, Zr_3N_4
Ba_3SbN, $BaZrN_2$,
Cs_6GaSb_3, $CsGeCl_3$,
Rb_2SnBr_6, Sr_3GaN_3, and
Sr_3SbN

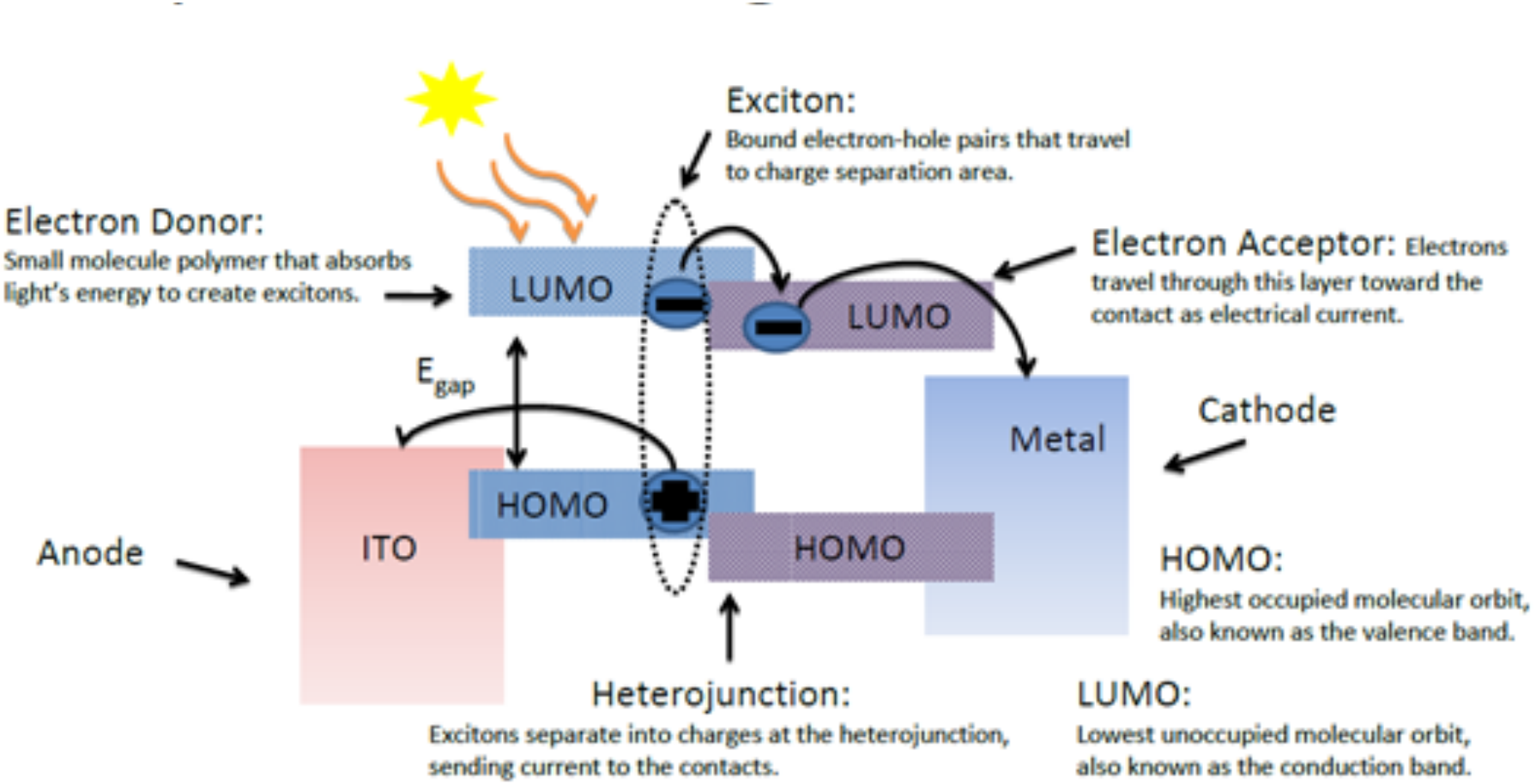
Kuhar, Pandey, Thygesen, Jacobsen, ACS Energy Letters, doi:10.1021.acsenergylett.7b01312 (2018).

Machine learning new materials

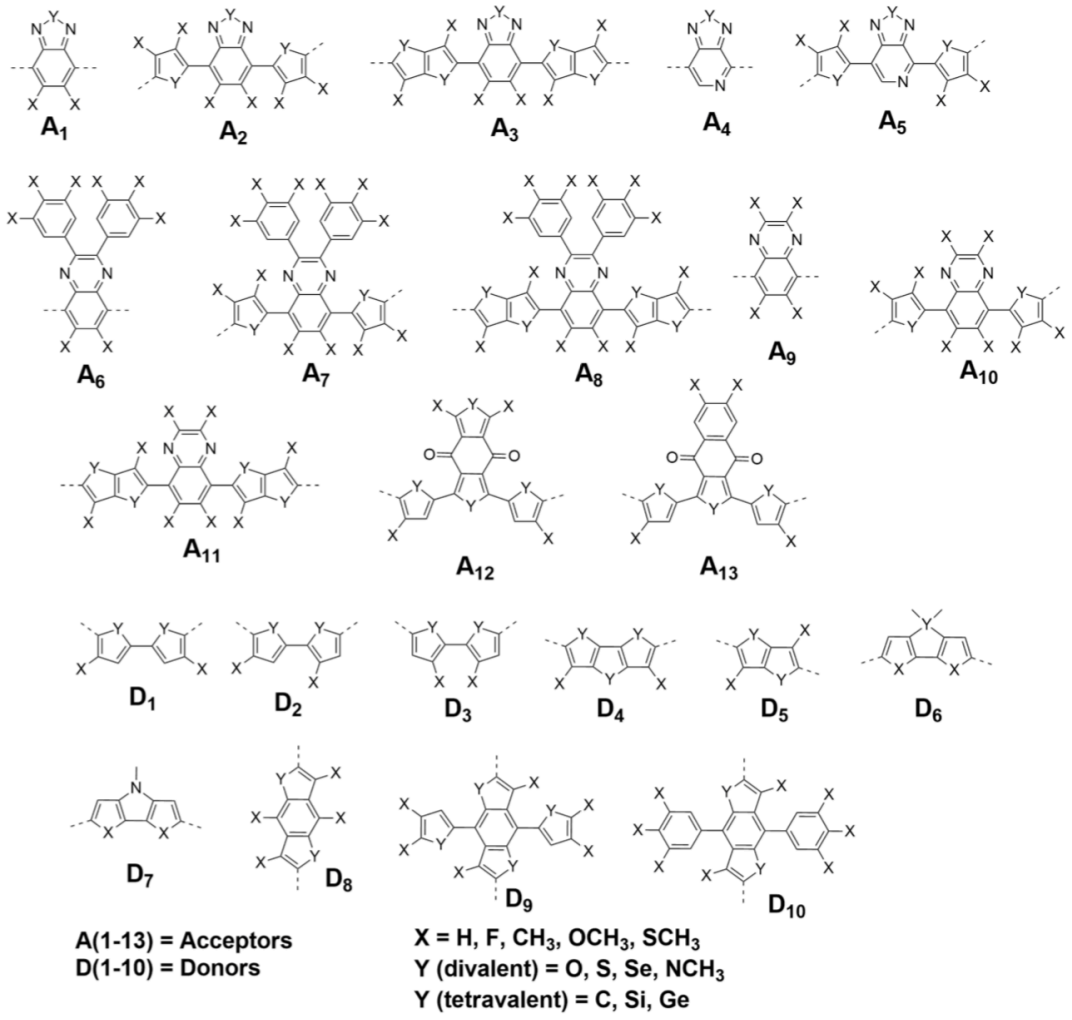
- Many different techniques
  - Representation of materials (fingerprints, ⋯)
  - Kernel regression, neural networks, ⋯
- Two challenges:
  - Can we predict material properties without knowing where the atoms are?
  - Can we avoid evaluation of properties of many (irrelevant) materials? Can we directly identify relevant materials?

Organic solar cell
(PCBM-based blended polymer solar cell)

PCBM = Phenyl-C'61-Butyric-Acid-Methyl-Ester



Donor-acceptor molecules (polymer units)



What is the position of the LUMO and the optical gap for these molecules?

In principle 10^{14} molecules!

Training set with ~4000 molecules (Gaussian, B3LYP)

Jørgensen, Mesta, Shil, García Lastra, Jacobsen, Thygesen, and Schmidt, (2018).

Data representation

String representation of molecules.

Grammatical production rules.

No specification of atomic coordinates.

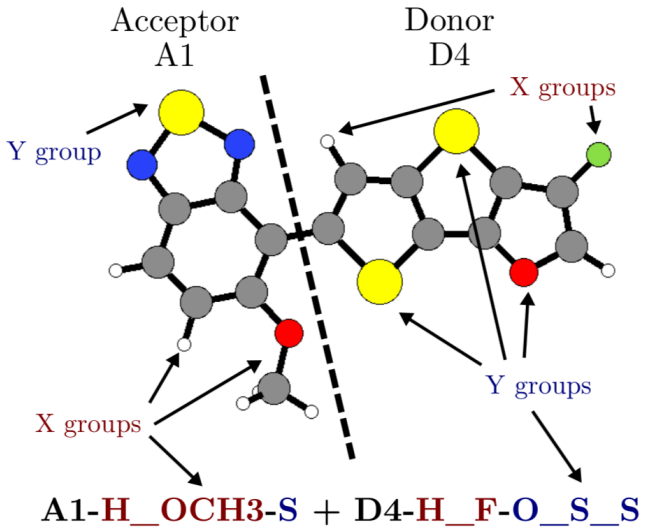


FIG. 2: String representation of one of the molecules of the solar cell dataset: "Acceptor backbone"-"X groups"-"Y groups"+"Donor backbone"-"X groups"-"Y groups". Whenever no side groups are present "*" character is used instead.

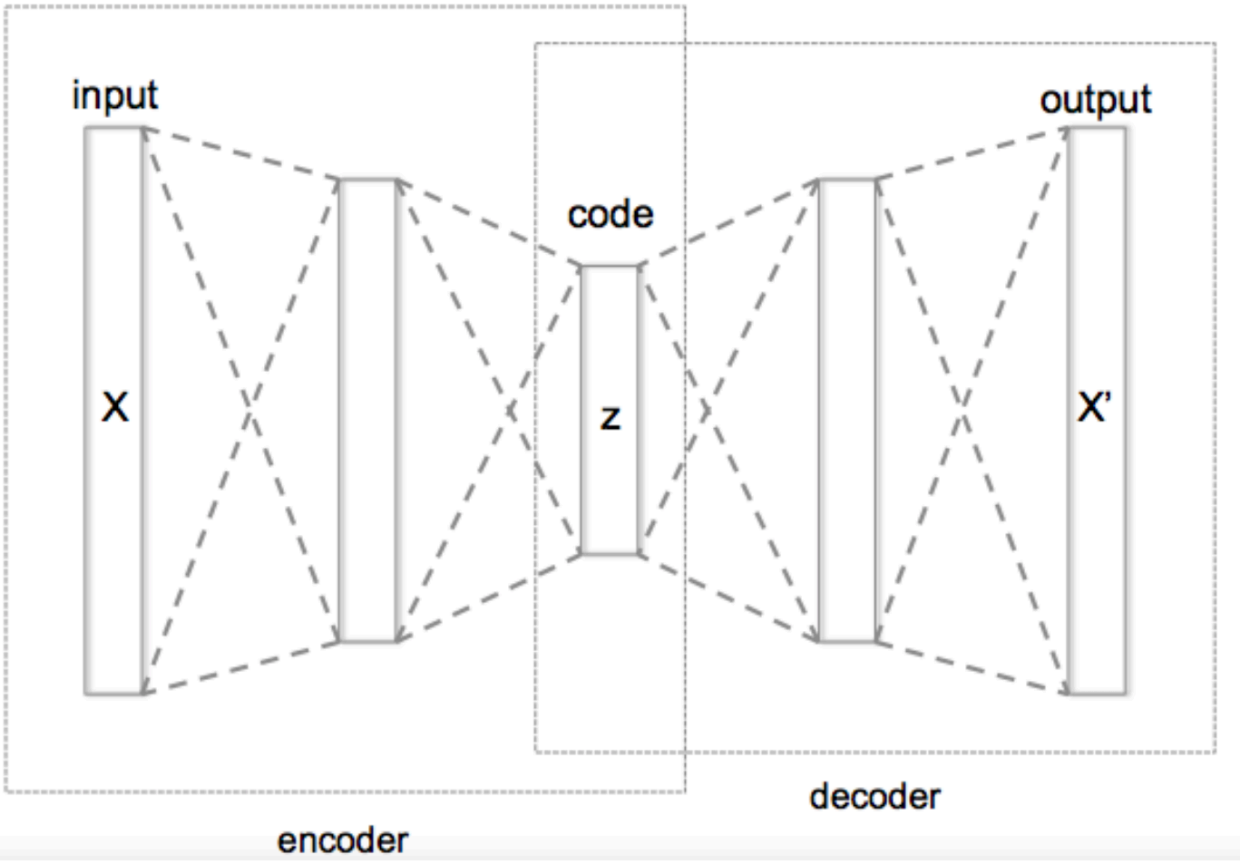
Earlier work uses SMILES to represent molecules:

Gómez-Bombarelli et al. (2016), arXiv:1610.02415 [cs.LG].
Kusner et al. (2017), arXiv:1703.01925 [stat.ML].

Grammar Variational AutoEncoder

32-dimensional
vector space
(“latent” space)

Strings
(grammar)



Jørgensen, Mesta, Shil, García Lastra, Jacobsen, Thygesen, and Schmidt, (2018).

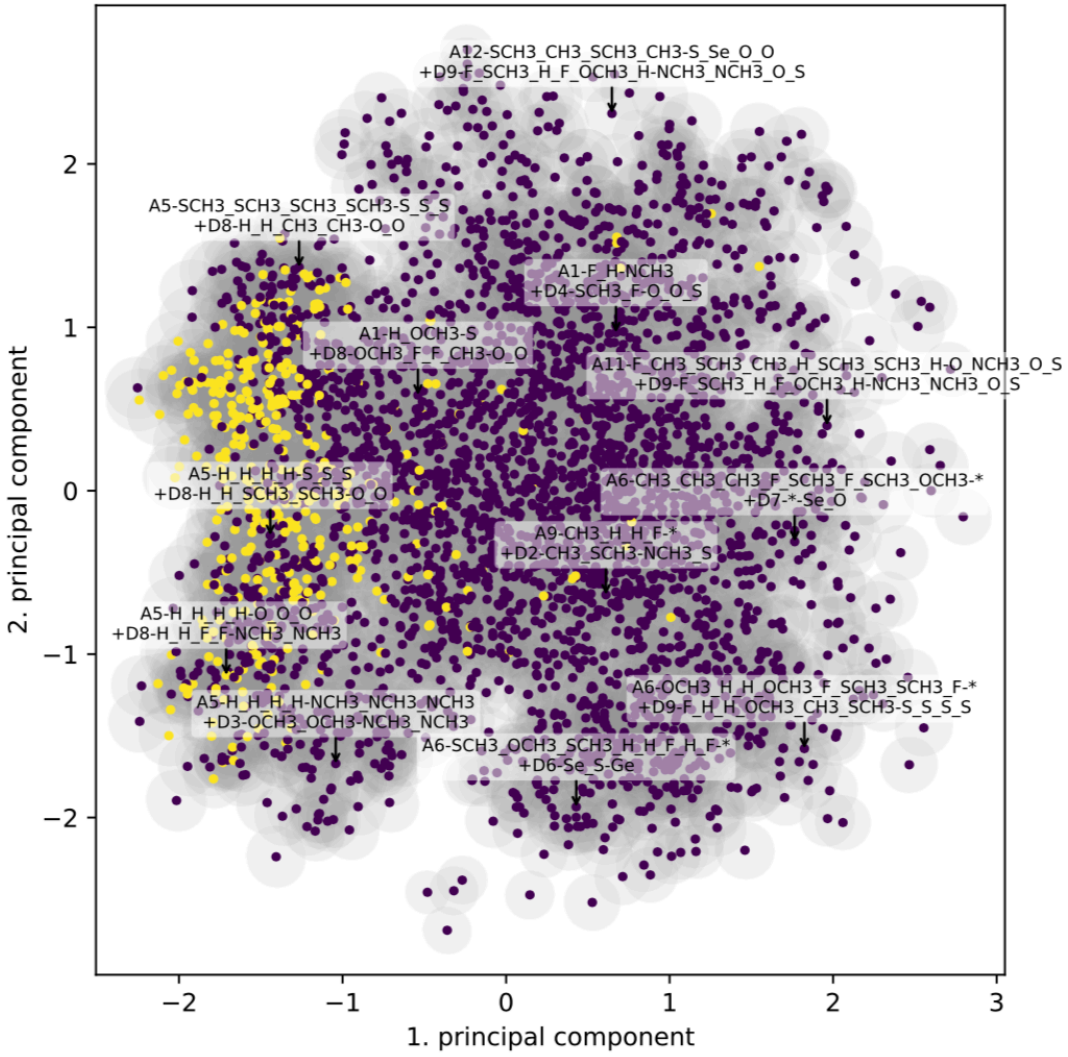
Latent space

First two principal components.

Bright points are within target range for LUMO energy and optical band gap.

New materials can be predicted by optimization or interpolation in the latent vector space.

Decoder:
Latent space -> strings
-> molecules

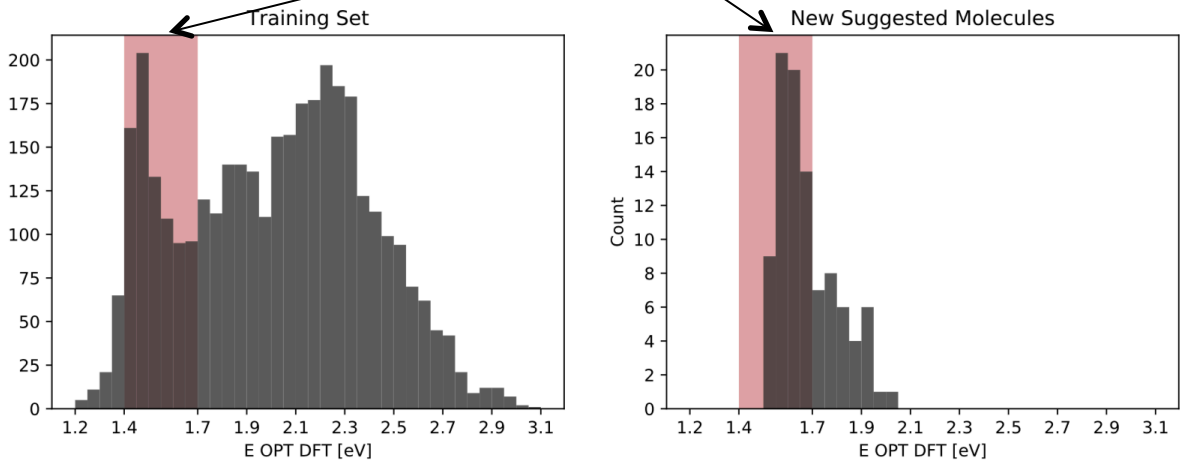


Prediction of new molecules

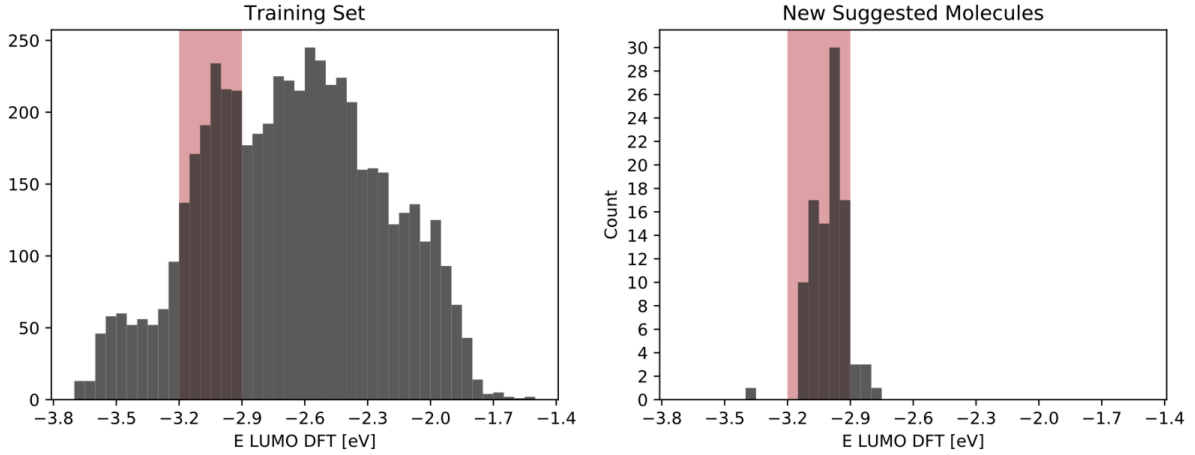
Target region

100 new molecules predicted

Optical band gap



LUMO energy



Computational screening and exa-scale: Some naïve considerations

- Opportunities
  - Screening more different materials
  - More accurate calculations
    - Light absorption, PBE -> GLLB –> GW -> BSE
  - New properties
    - Carrier lifetimes
- Challenges
  - Better and more descriptors
    - 1% of 1000 materials -> 10 candidates
    - 1% of 10^6 materials -> 10000 candidates

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Mikkel N. Schmidt

SURFCAT/DTU:
Andrea Crovetto
Brian Seger
Søren Dahl
Peter Vesborg
Ole Hansen
Ib Chorkendorff

CASE
Catalysis for Sustainable Energy

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Center on Nanostructuring for Efficient Energy Conversion

The VILLUM Center for the Science of Sustainable Fuels and Chemicals