Finding the Needle in a Haystack: Materials discovery through high-throughput ab initio computing and data mining

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Max conference

January 31, 2018

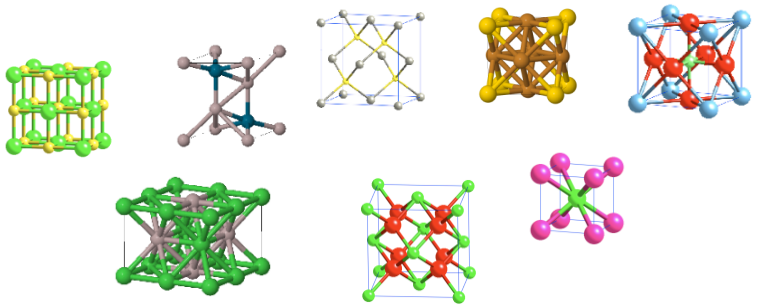
Outline

- **High-throughput computations as an accelerator to materials discovery**

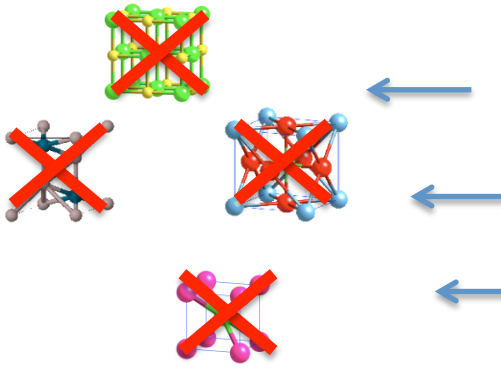
- **High-throughput computing in action:**
  - Transparent conducting oxides and non-oxides
  - electrides

- **The challenge of scaling-up and automation**
  - HT phonons

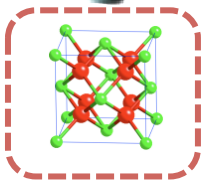
Accelerating materials discovery using high-throughput computations



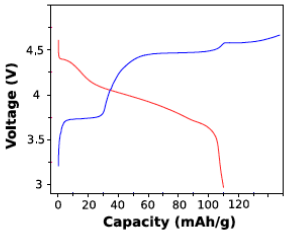
10,000
to 100,000
compounds

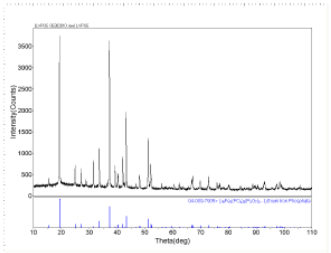


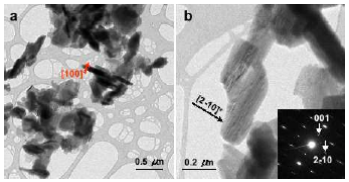
Property 1
Property 2
Property 3
Property 4
Property 5
Property6



10 to 100
compounds to
validate exp.

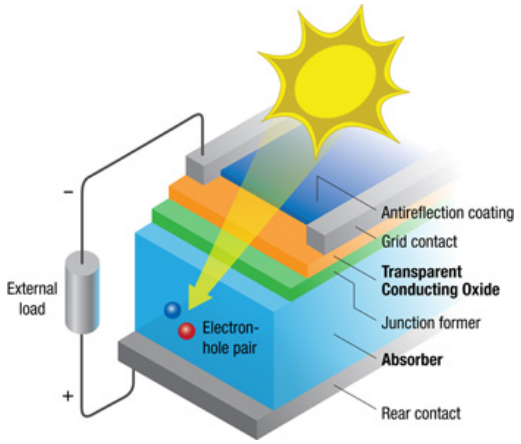






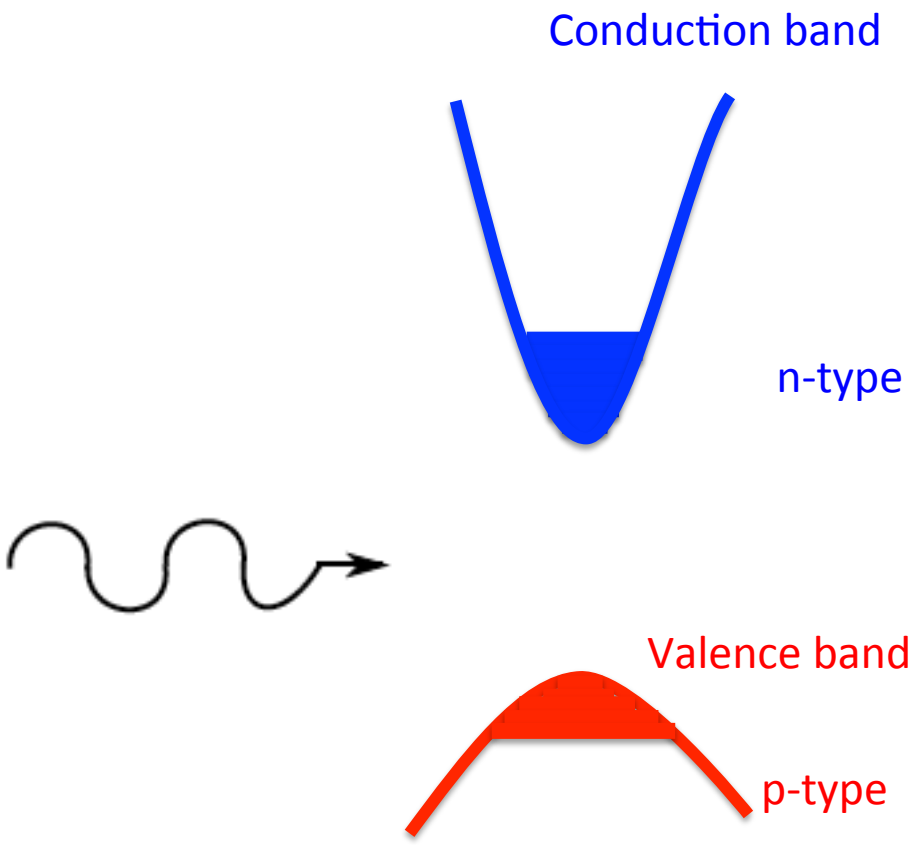
Transparent conducting materials

- Many applications require materials combining transparency with conductivity
  - Touch screens
  - Solar cells
- Those properties are antagonistic and difficult to obtain in the same material



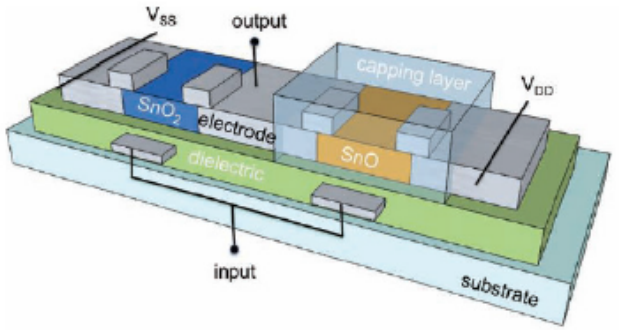
Transparent conducting oxides

- Transparent conducting oxides (TCOs) have
  - Wide band gap (transparency)
  - Doping (carriers)



n- and p-type TCOs

- n-type TCOs are ubiquitous in technology
  - Ex: In_2O_3 doped with Sn (ITO)
- p-type TCOs are lagging behind
  - Best n-type mobility: 100 cm^2/Vs
  - Best p-type mobility: 10 cm^2/Vs
- Fundamental limitations to many technologies (e.g., transparent electronics)



Back to basics: what properties drive TCOs conductivity?

- Conductivity:

Carrier concentration

$$\sigma = ne\mu$$

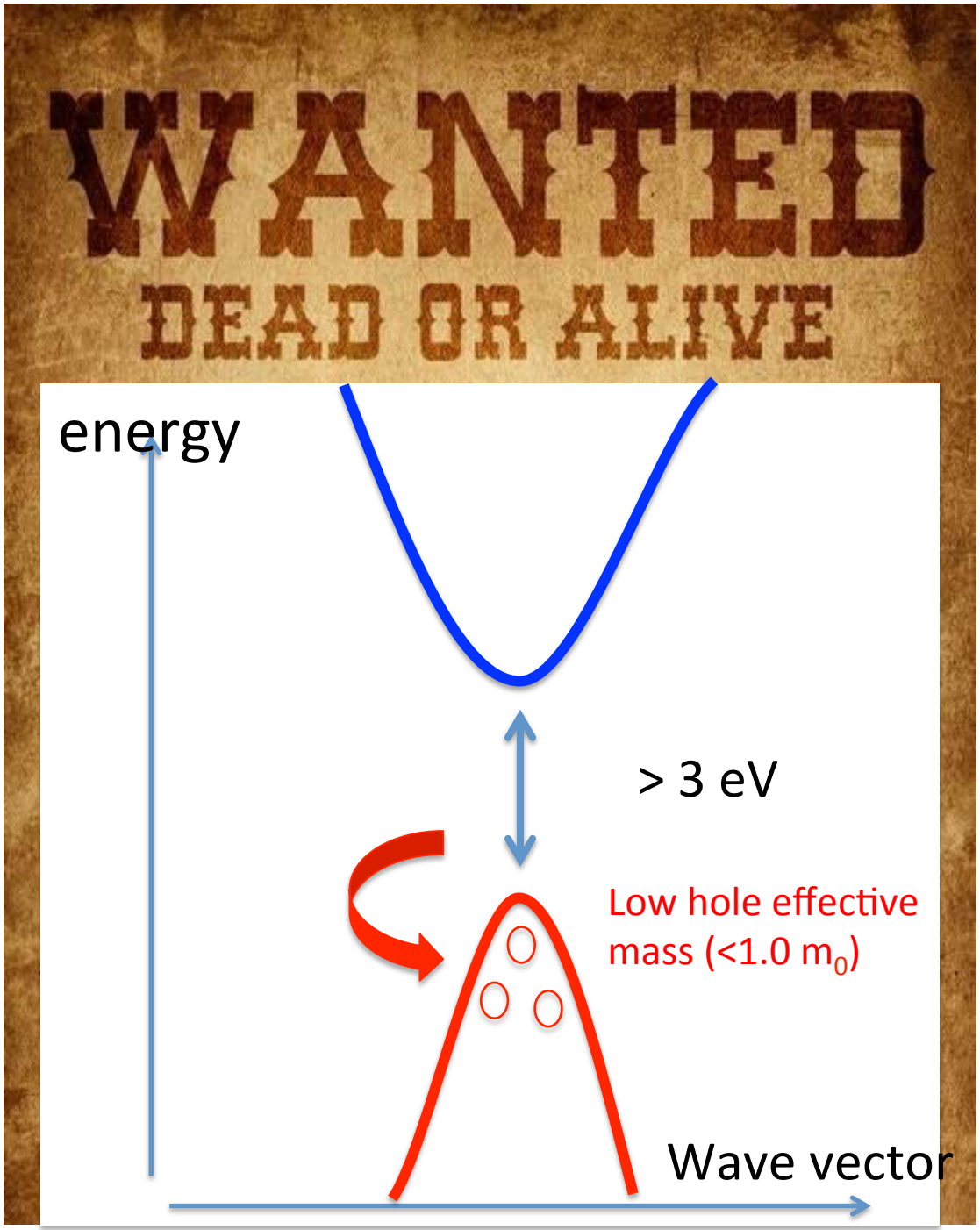
- Mobility:

scattering time

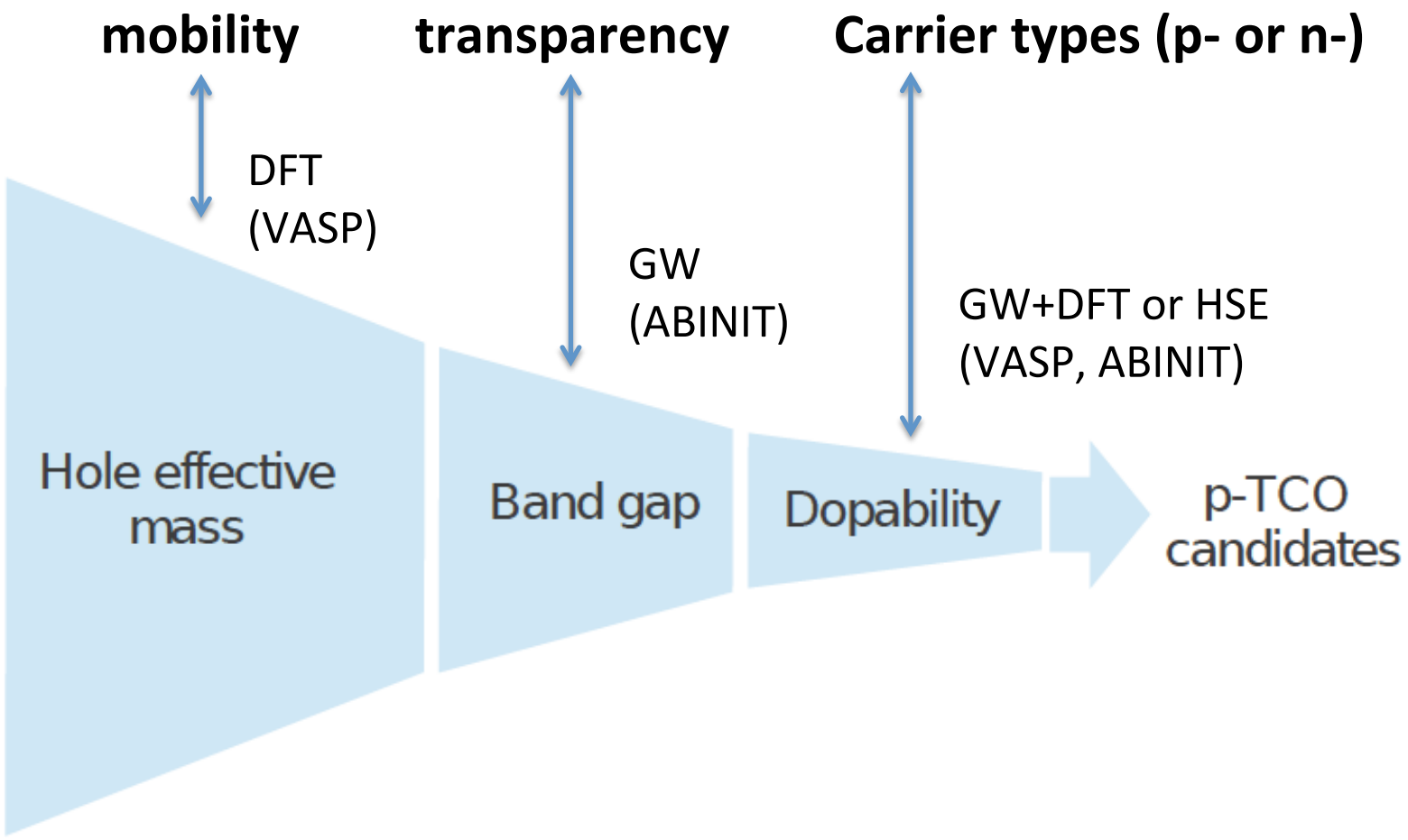
$$\mu = \frac{e\tau}{m^*}$$

Effective mass

$$\frac{1}{m^*} \propto \frac{\partial^2 E}{\partial k^2}$$

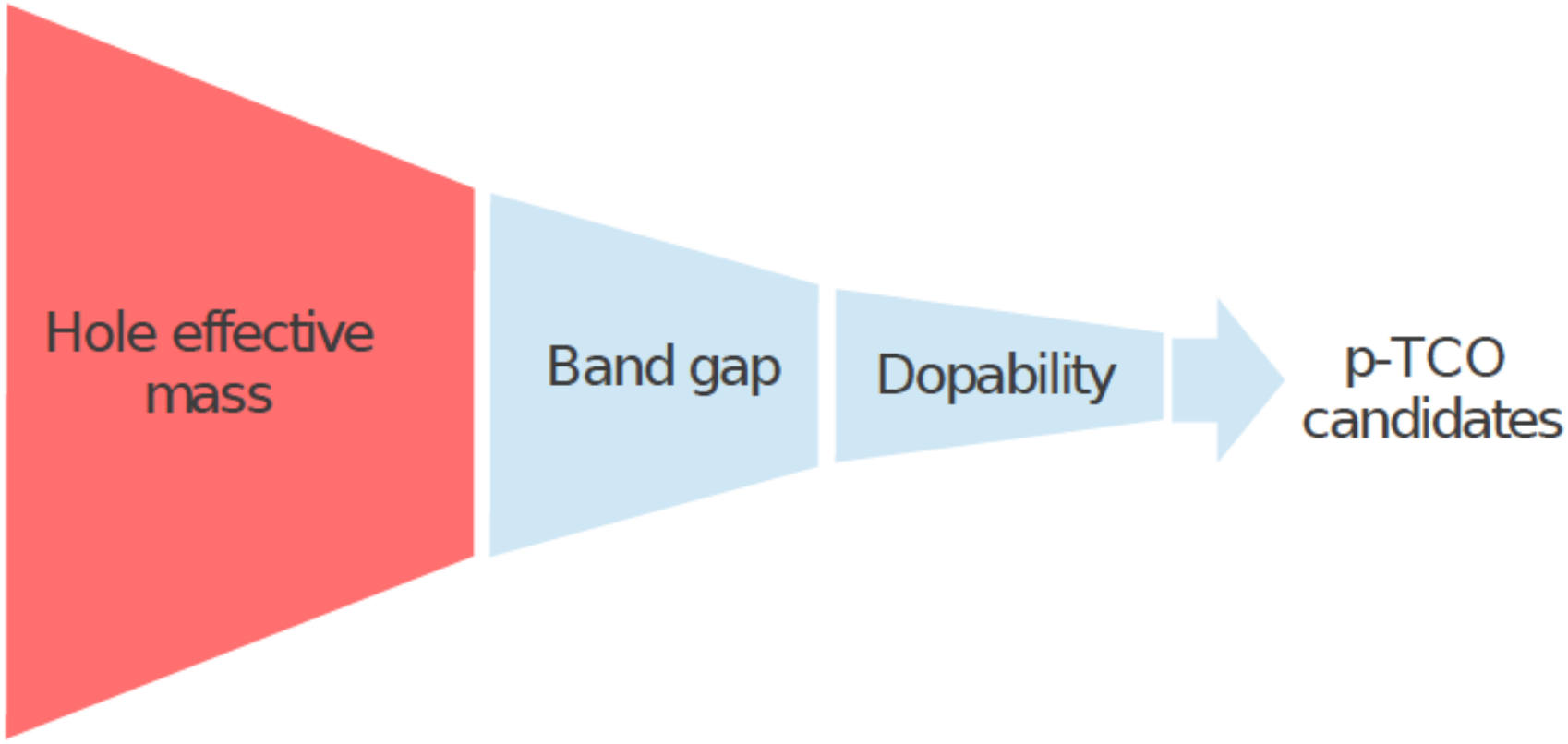

WANTED
DEAD OR ALIVE
energy
> 3 eV
Low hole effective mass (<1.0 m_0)
Wave vector

A tiered high-throughput screening for p-type TCOs discovery



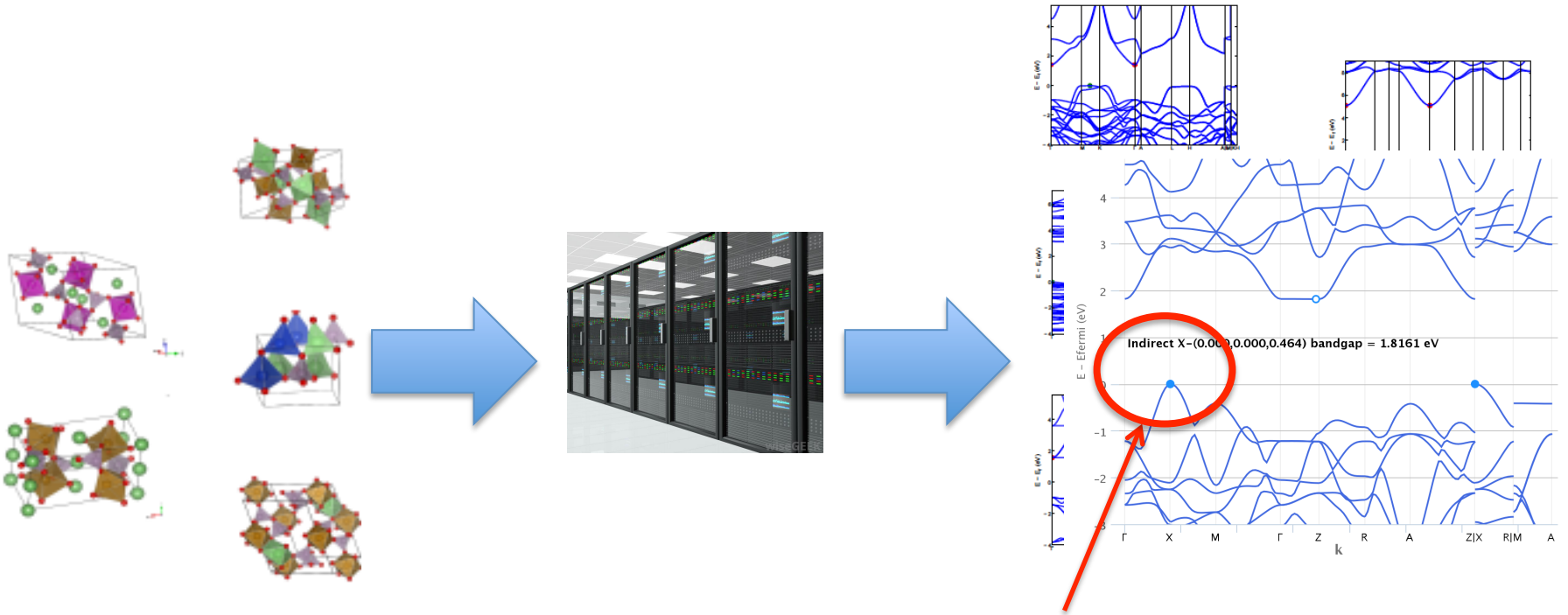
6 000 **known** oxides! (ICSD)

GH, A. Miglio, G. Ceder, G.-M. Rignanese, X. Gonze, *Nature Communications*, 2013


Hole effective mass
Band gap
Dopability
p-TCO candidates

Low hole effective mass screening

- Compute band structures (within DFT) for more than 6,000 oxides
- Known oxides (from the ICSD)
- Extract the effective masses

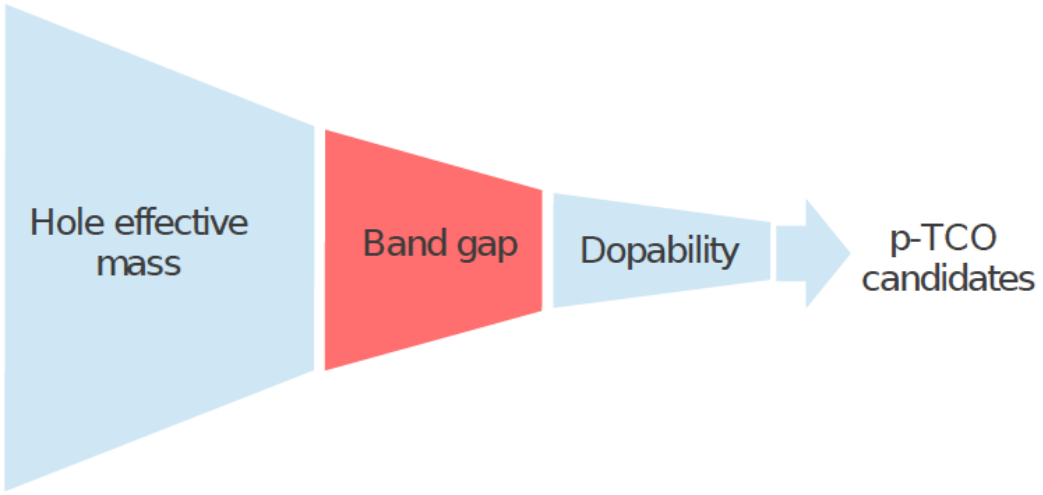


Known materials (from the ICSD)

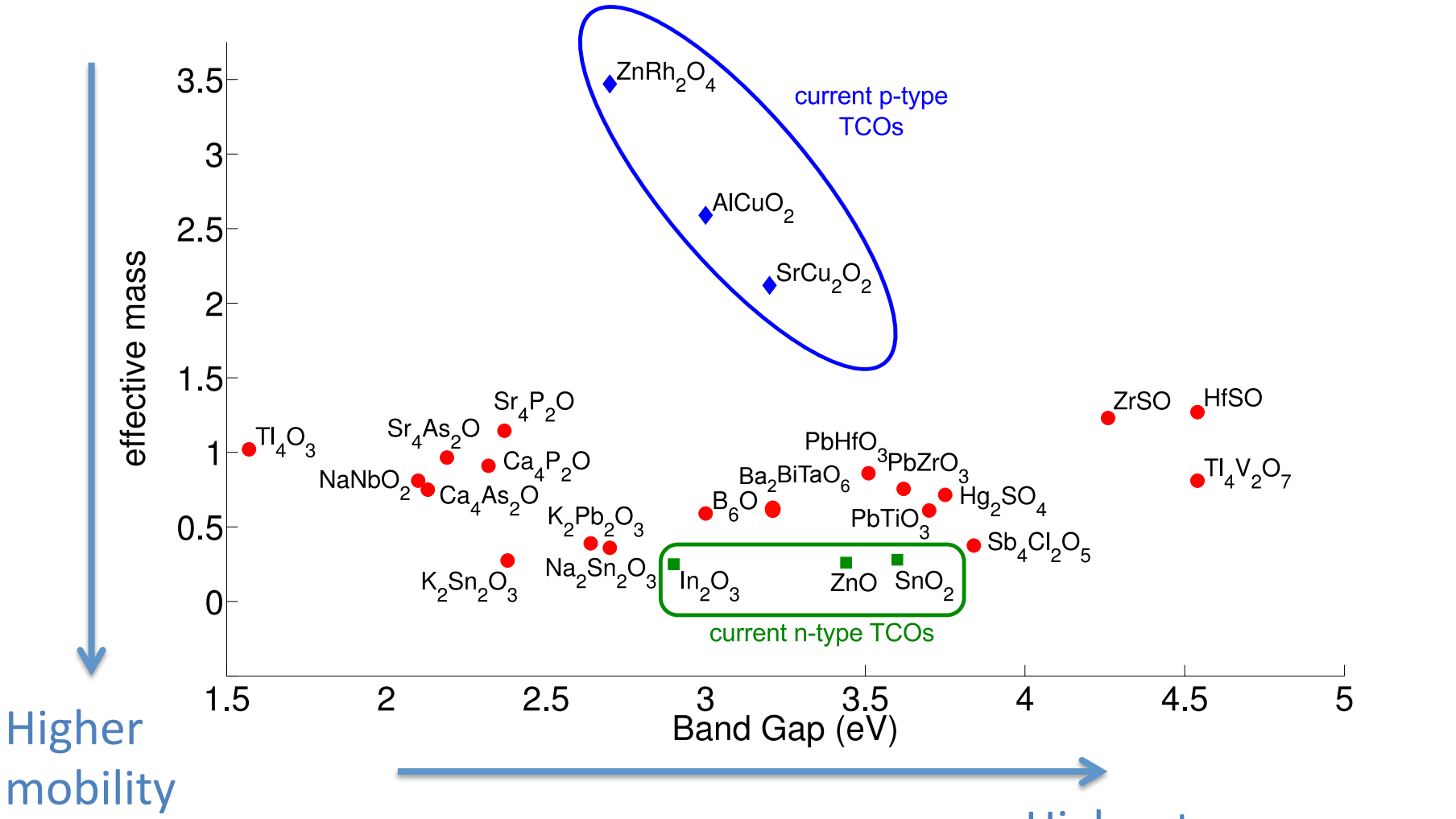
Effective mass, curvature

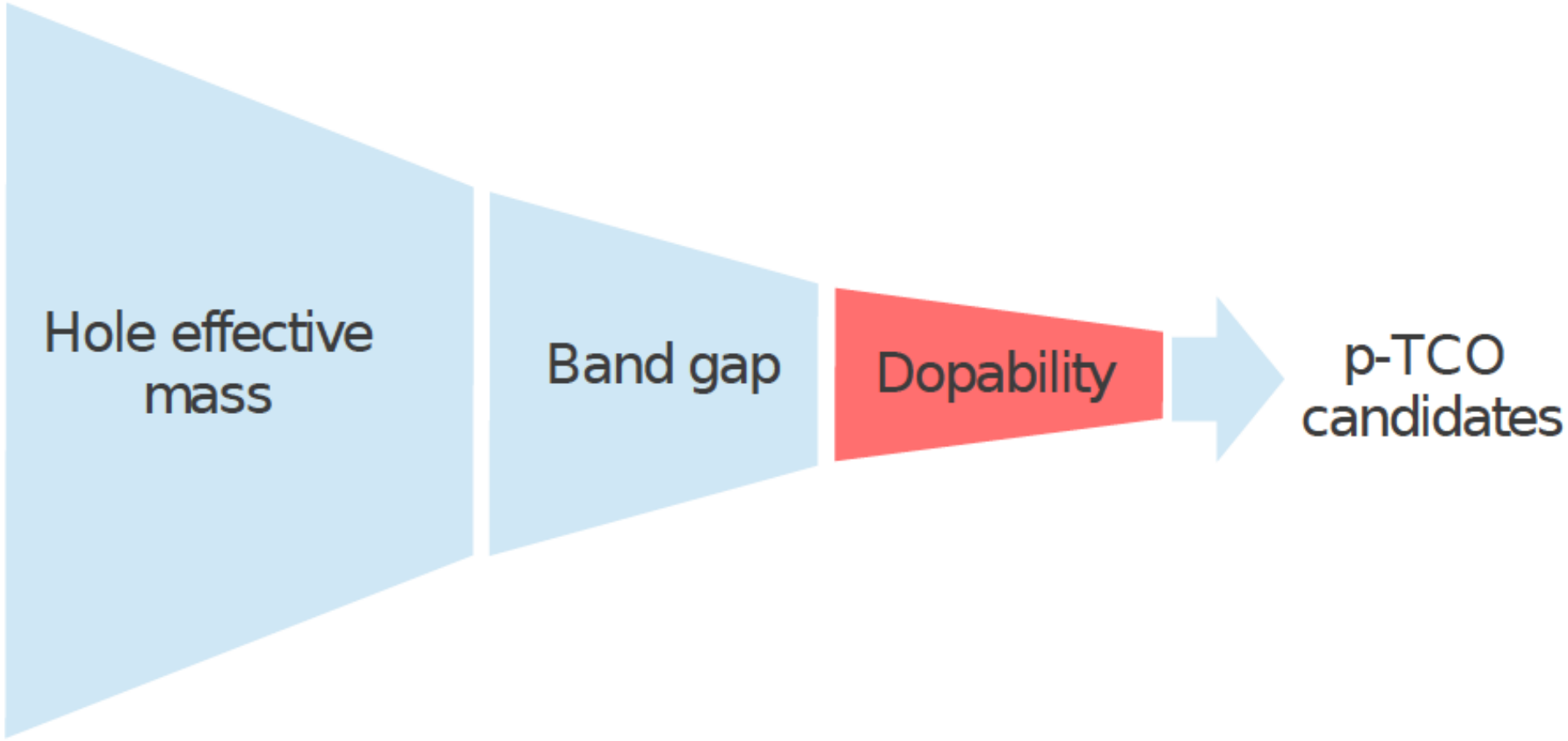
Further screening

- Only keep oxides with hole effective mass < 1.5
  - Around 20 compounds over more than 6,000 oxides!
  - Needle in the haystack problem!
- For those, compute more accurate band gap with one-shot GW (G_0W_0) technique



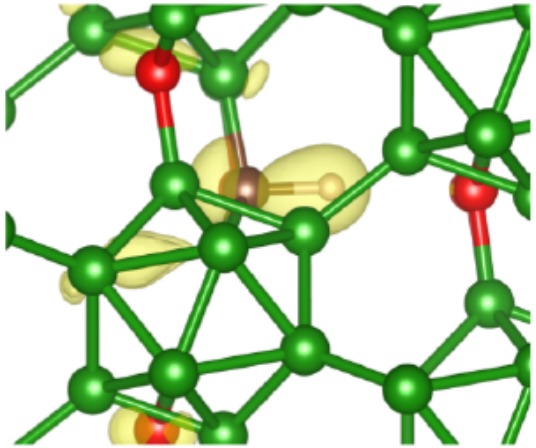
Effective mass vs band gap



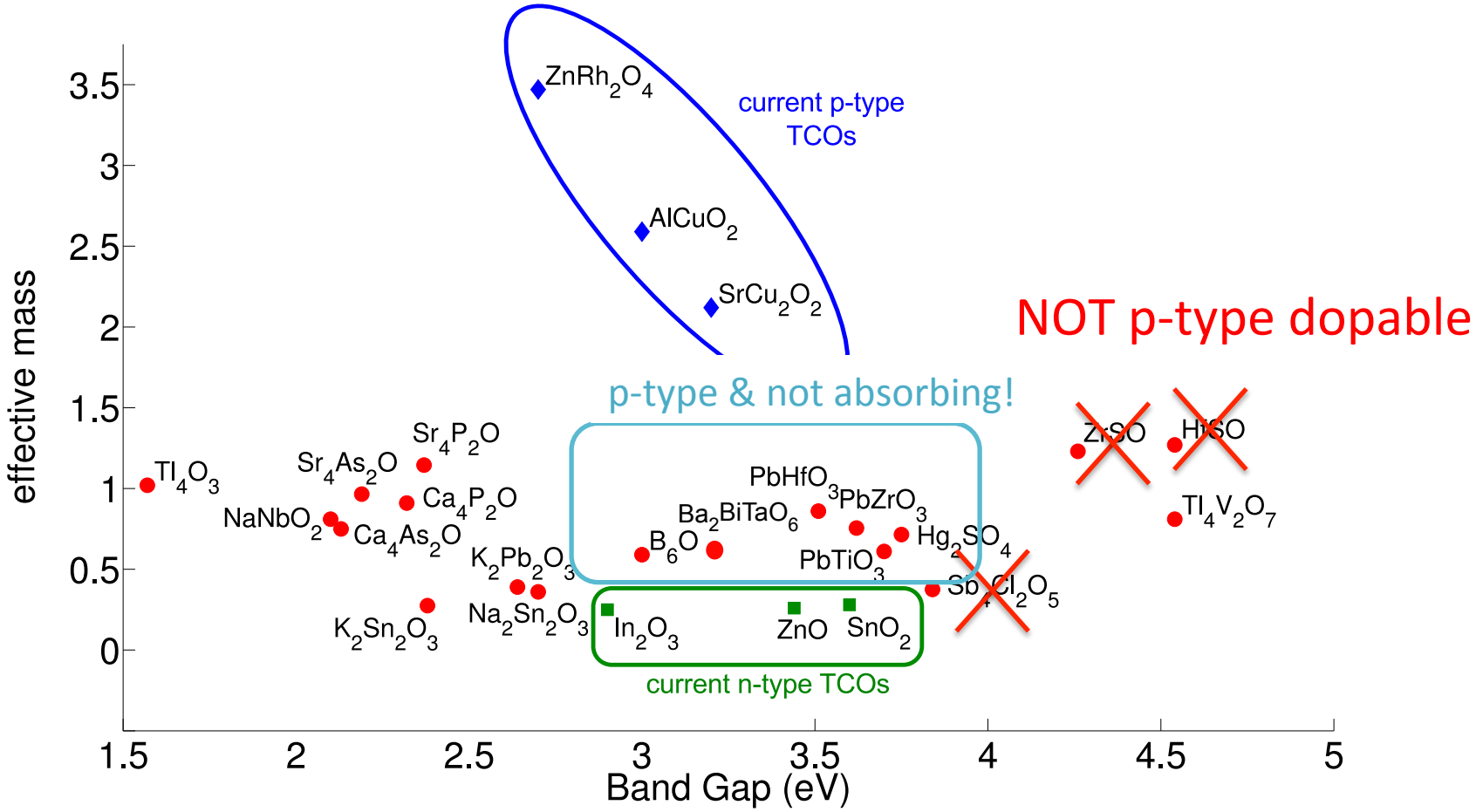

Hole effective mass
Band gap
Dopability
p-TCO candidates

p-type dopability

- Not all oxides can be p-type
- Intrinsic defects often prevent dopability
  - « Hole killers »
  - e.g., the oxygen vacancy
- Defects can be computed through ab initio techniques
  - More expensive
  - GW or hybrid functionals needed

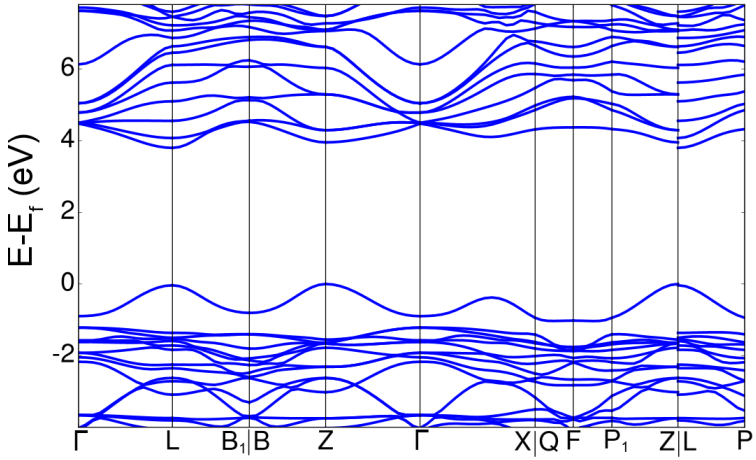


p-type dopability

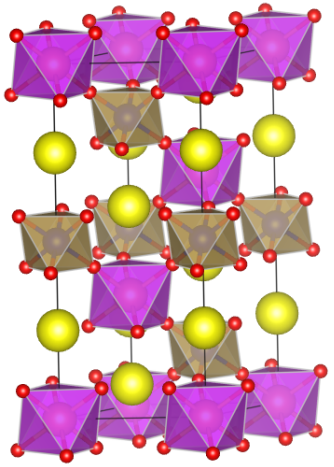


Experimental realization: Ba_2BiTaO_6

- Excellent candidate also:
  - Large band gap (> 3eV)
  - Low hole effective mass
  - p-type dopability

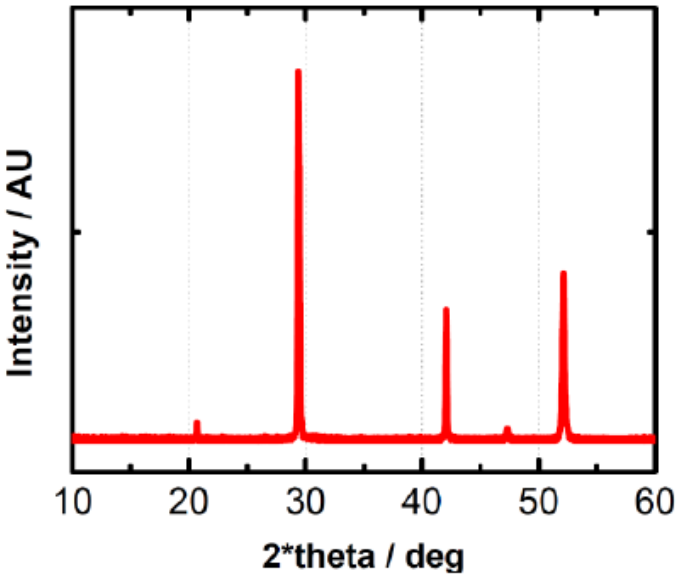


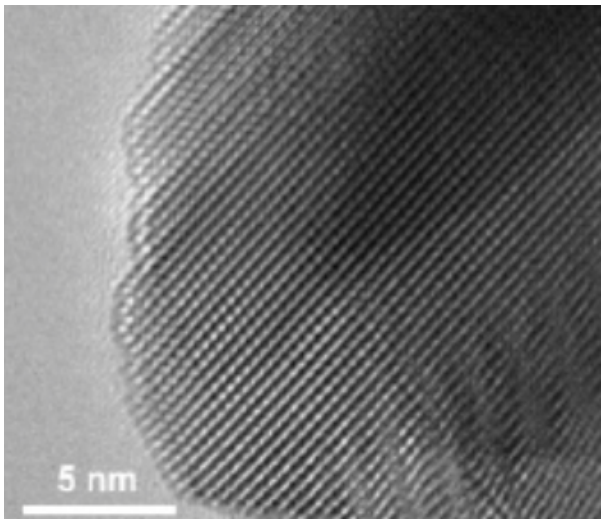
- Perovskite structure
  - Mixed Bi^{3+} and Ta^{5+}



Ba_2BiTaO_6: synthesis

- Solid state synthesis (from BaO, Ta_2O_5 and Bi_2O_3)
- Phase pure pellets and thin films from pulsed laser deposition (PLD) on MgO

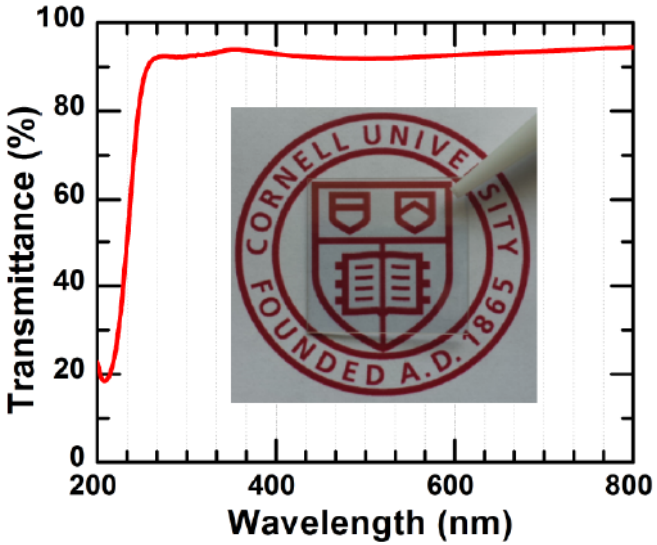




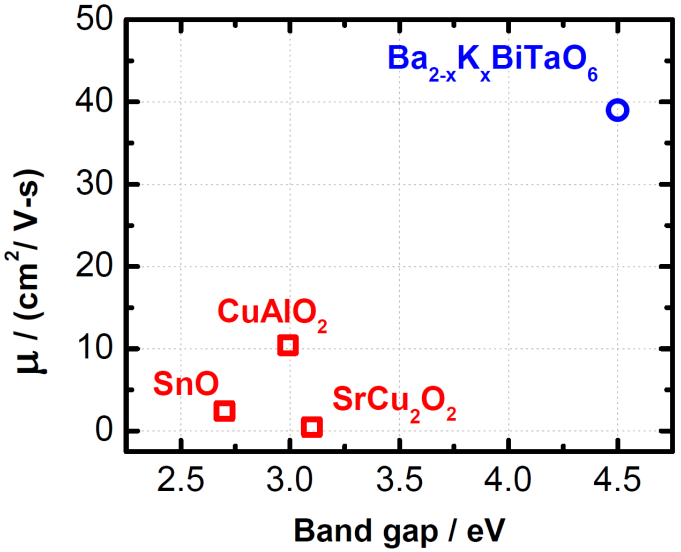
A. Bathia, **GH**, T. Nilgianskul, A. Miglio, J. Sun, H.J. Kim, K.H. Kim, S. Chen, G.-M. Rignanese, X. Gonze and J. Suntivich, *Chemistry of Materials*, 2016

Ba_2BiTaO_6: experimental testing

- Transparent!
- Band gap around 4.5 eV



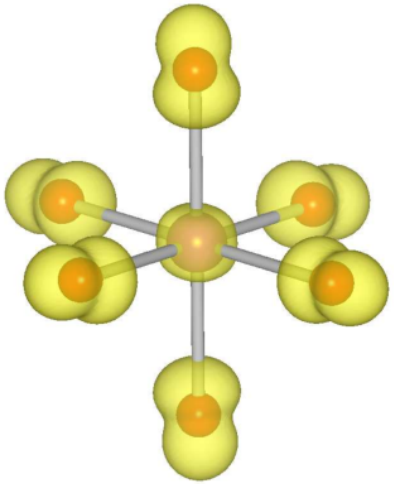
- Made p-type by K doping
- High mobility (**38 cm^2/Vs**)

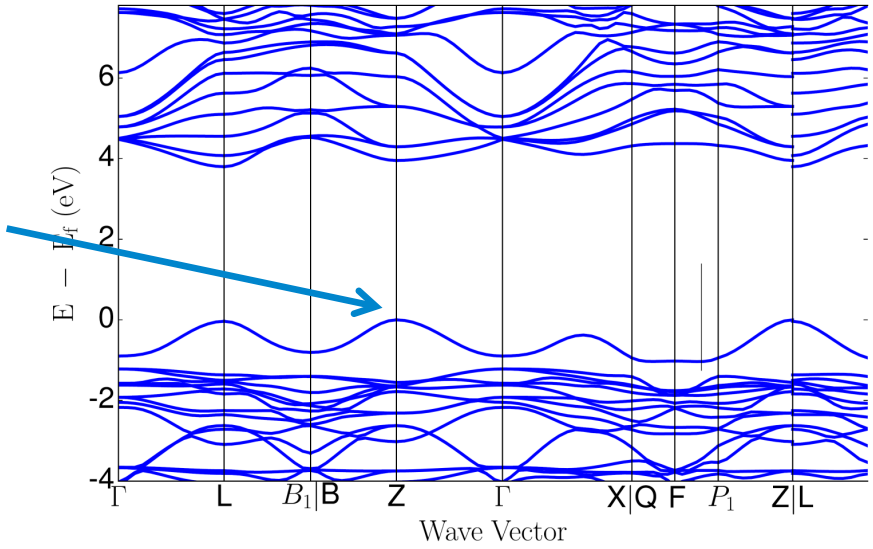


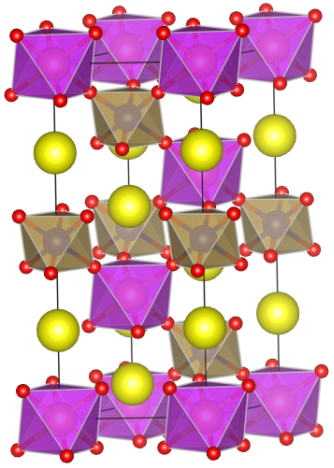
But still very low carrier concentrations (10^{14} cm^2/Vs)

The first Bi-based high mobility p-type oxide

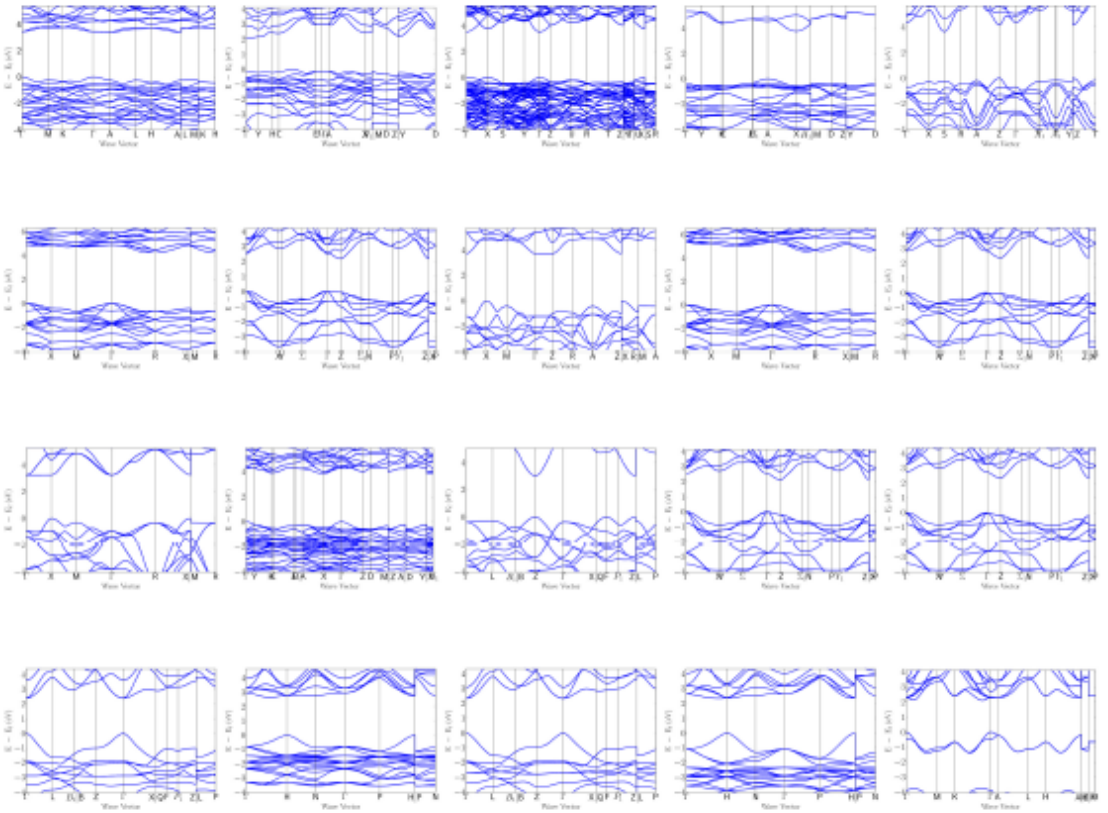
- Promising hole mobility confirmed experimentally
- Future work on improving dopability
- Bi-s/O-p mixing at the VBM





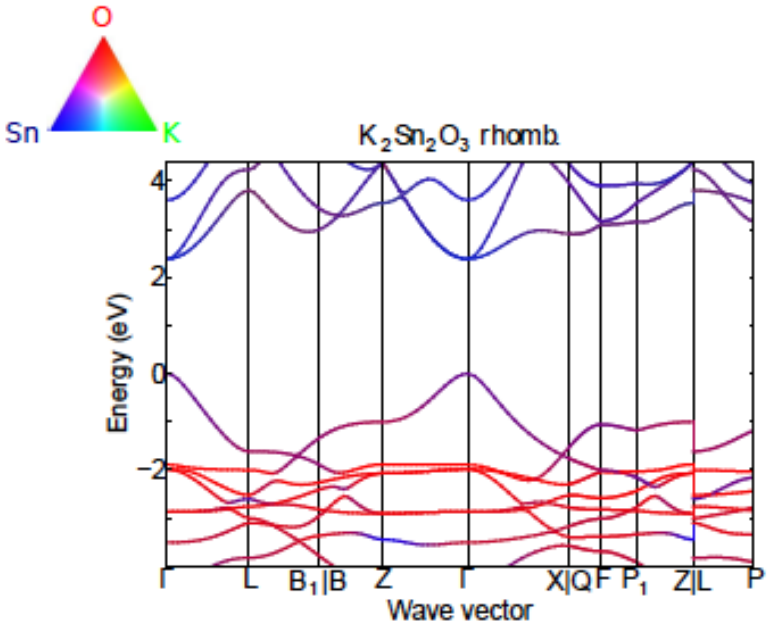


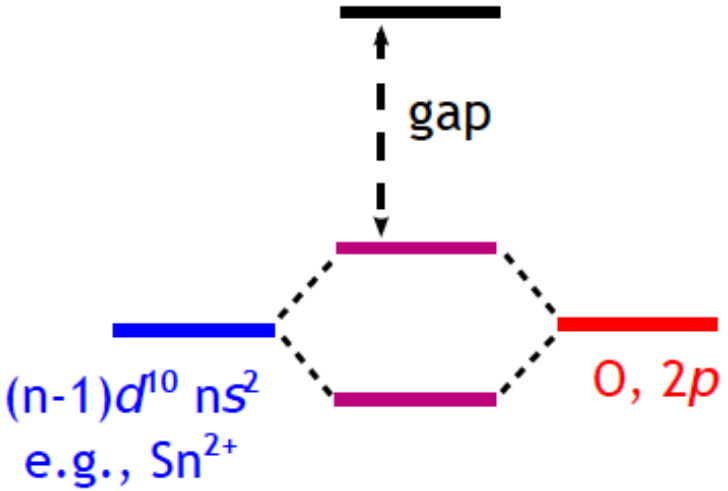
Design principles for low hole effective masses oxides?



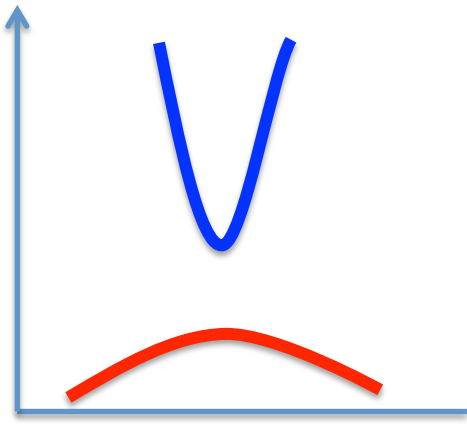
design principle: (n-1)$d^{10}ns^2$ ions

- Reduced main group elements: Sn^{2+}, Pb^{2+}, Bi^{3+},…
- Why? Mixing with O-p in the valence band
  - AND s orbital give a large overlap with the O-p





Beyond oxides?

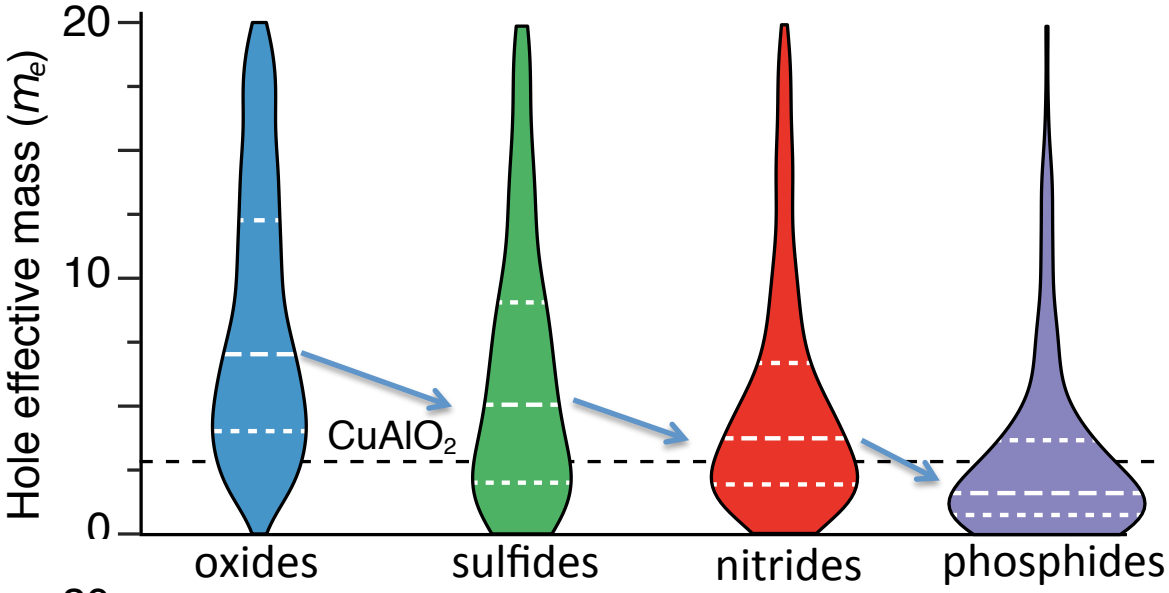


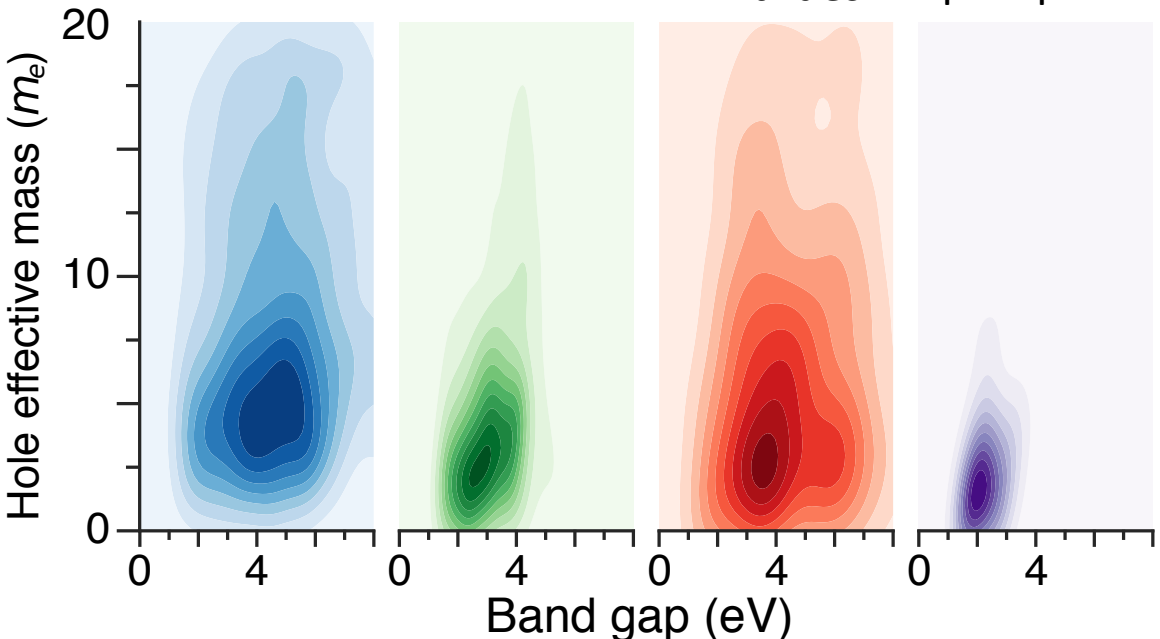
- Oxides have intrinsic issues
  - Flat oxygen p valence band
  - Only few materials (20 over 6000 !) can lead to low hole effective masses

- Do we really need to stick to oxides?
  - Similar study on non-oxides (30 000 compounds)

J. Varley, A. Miglio, V.A. Ha, G.-M. Rignanese and **GH**, *Chemistry of Materials*, 2017

Hole effective mass in alternative chemistries





Changing the anion chemistry lowers the hole effective mass!

What about the gap?

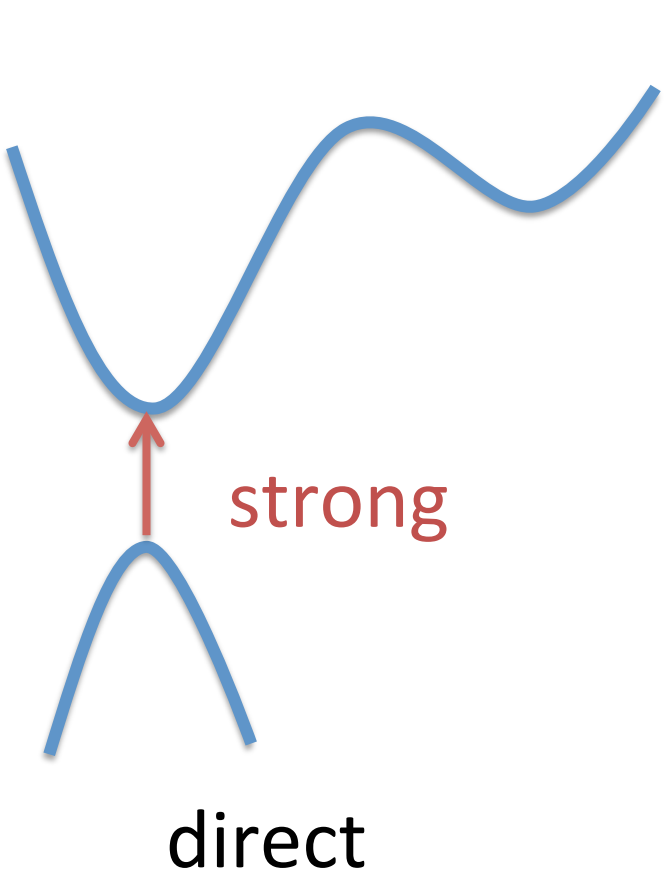
Hole effective mass ↘

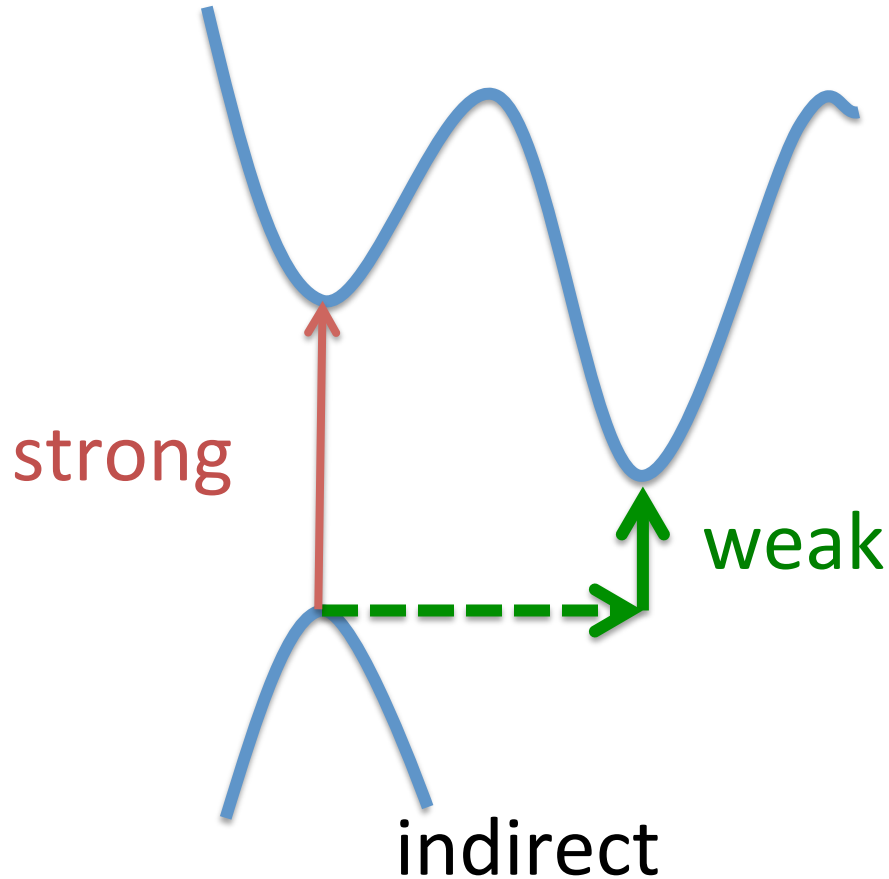
Band gap ↘

Oxides are especially good in transparency (> 3eV)

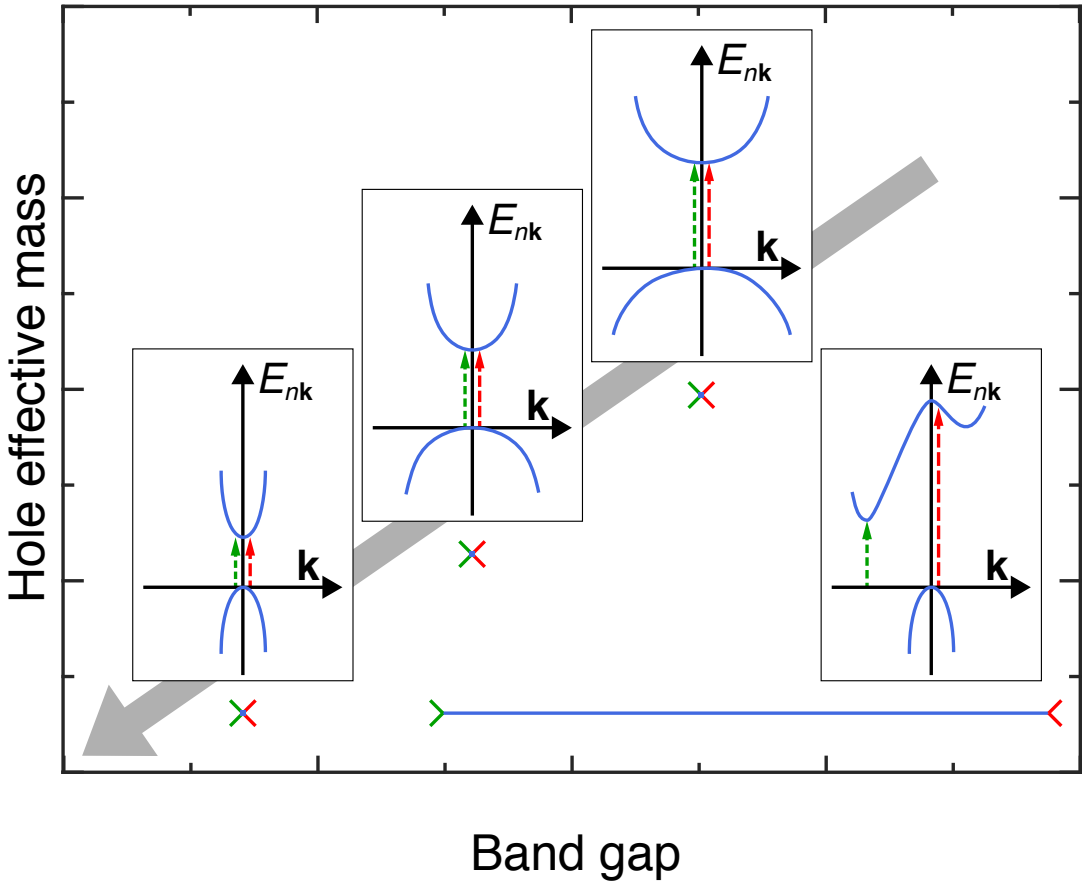
A strategy to break the effective mass-band gap correlation

- Optical absorption and transparency is mainly driven by direct absorption, indirect ones tend to be very weak



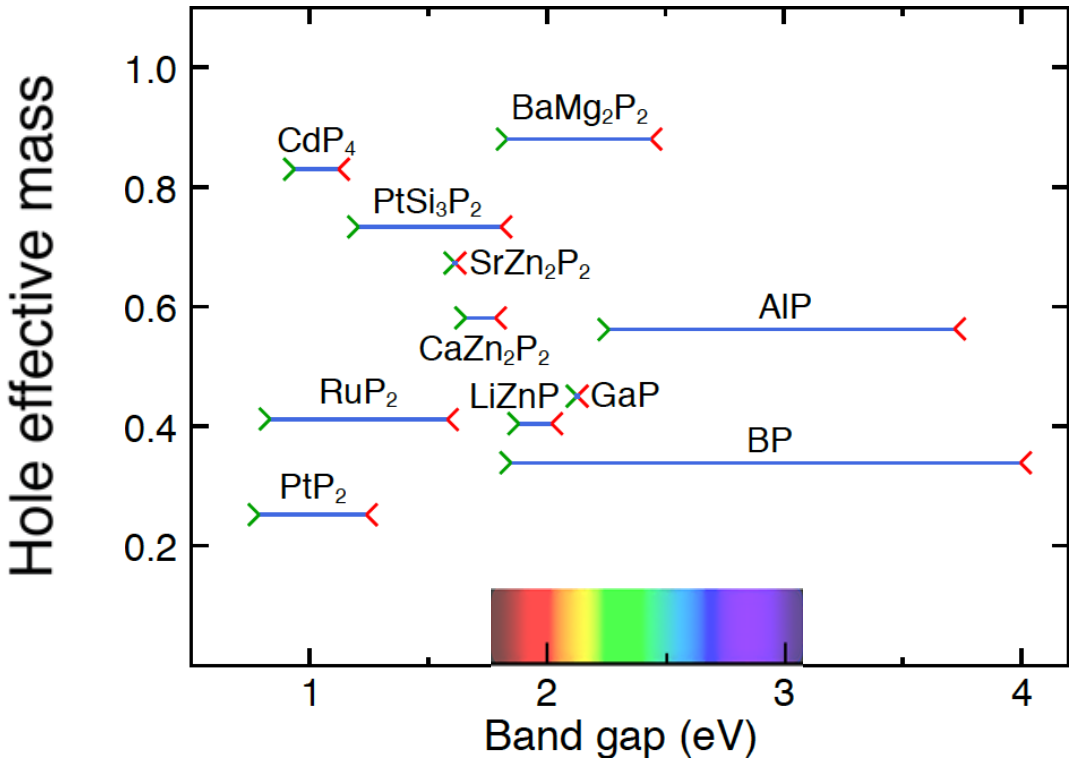


A strategy to break the effective mass-band gap correlation



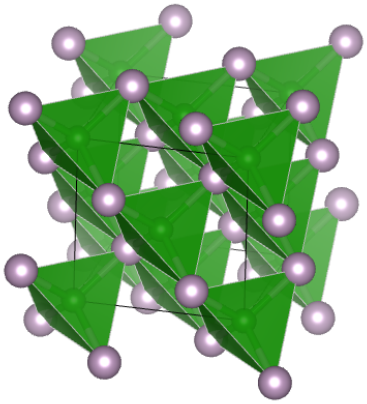
Search for large indirect band gap phosphides

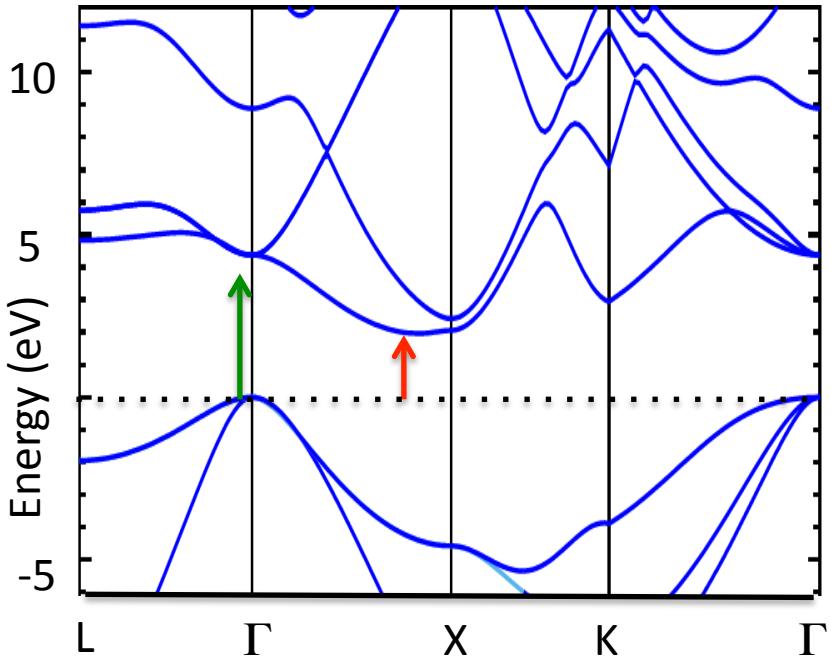
- Phosphides have very low hole effective mass
- Focus on the most promising effective mass materials
- Compute band gaps with more accurate GW technique.



Boron phosphide is a promising p-type TCM

- Low hole effective mass (0.3 m_0)
- Large direct gap (4eV)
- P-type dopable





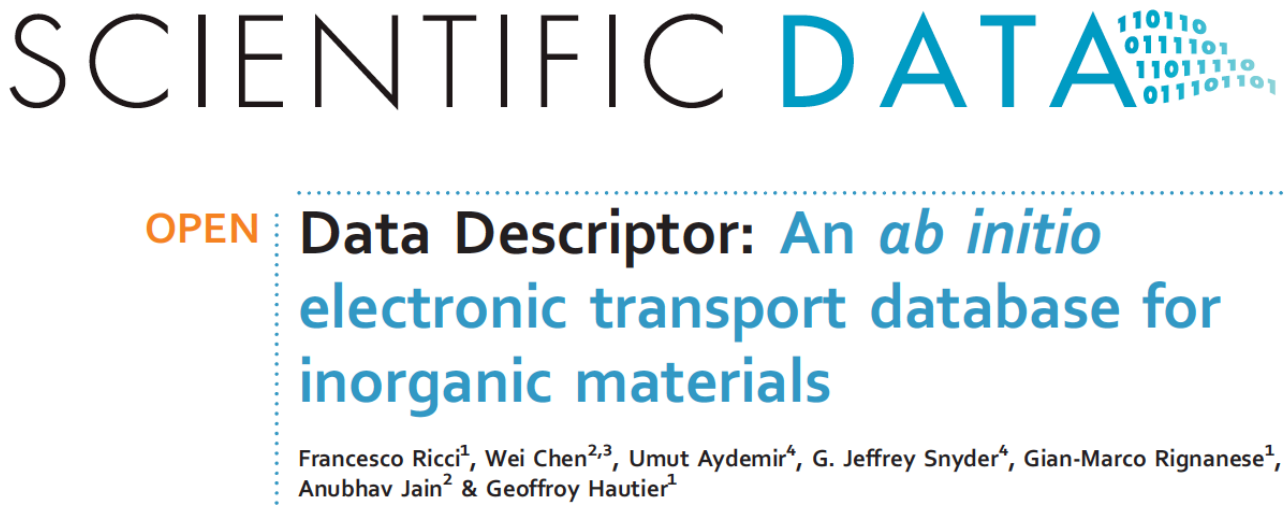
Boron phosphide previous experimental data

- BP was studied in the 60s, 70s but not much since then
- Experimental results
  - (undirect) Band gap: 2 eV [1]
  - P-type dopable: 10^{16} cm^{-3} to 10^{18} cm^{-3} [2]
  - Hole mobility measured: 8 to 350 cm^2/Vs [1,2,4]
  - Absorption coefficient: 1000 cm^{-1} (2% absorption for 200 nm film) [2,3]
  - Hole conductivity: 2800 Ω^{-1} cm^{-1} [4]
- In agreement with theory
- Our work motivates a reinvestigation of this material!

[1] Iwami, Tohda, Kawabe, Electr. Eng. Japan, 1975
[2] Stone, Hill, PRL, 1960
[3] Anantharayanan, Mohanty, Gielisse, J. Cryst. Growth, 1973
[4] Shohno, Takigawa, Nakada, J. Cryst. Growth, 1974

Sharing our large high-throughput database

- We computed electronic transport properties for more than 30,000 compounds
- Useful to many fields: TCOs, thermoelectric, photovoltaics,…
- Data publicly available



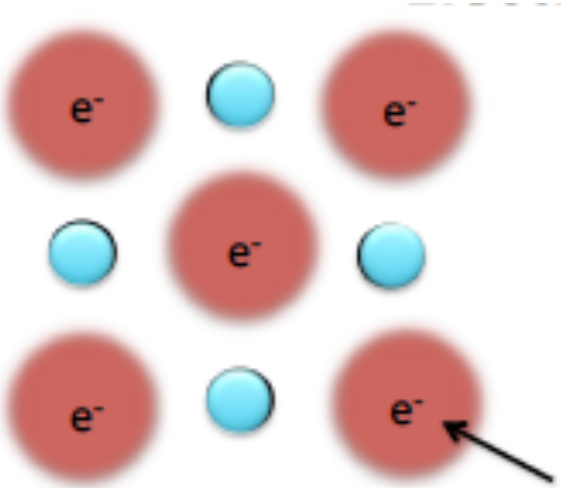
Soon on the Materials Project

http:://www.materialsproject.org

MATERIALS PROJECT

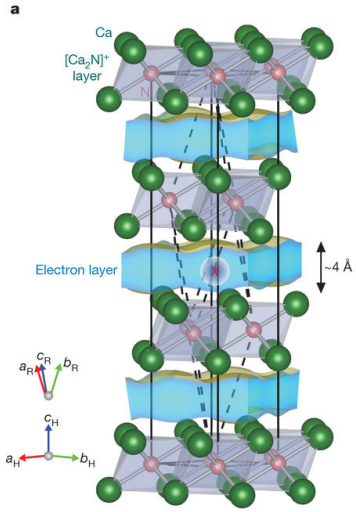
Even more unusual properties: electrides

- Electrides are materials where electrons localize off the nuclei
- Anion = electron



- Growing field in chemistry and Physics
  - Catalyst support
  - Transparent conducting materials
  - …

- Only a handful electrides are known:
  - Ca_2N, Y_2C, Y_5Si_3, and LaH_2….

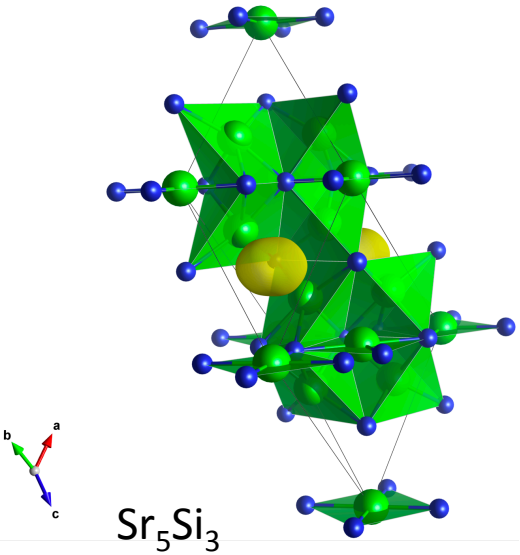


K. Lee et al., *Nature*, 2013

High-throughput search for electrides

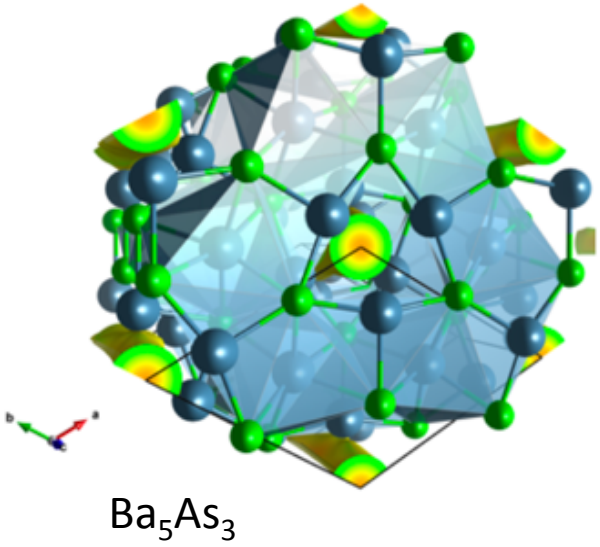
- Browse a database of 30,000 DFT computations on known ICSD compounds
  - Metals only
  - Purely based on DFT-PBE partial charge density, Baader analysis
- Discover more than 60 new electrides!

0D electride



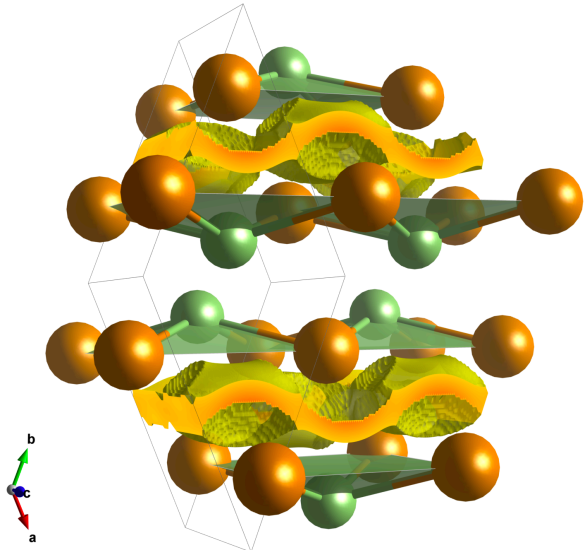
Sr_5Si_3

1D electride



Ba_5As_3

2D electride



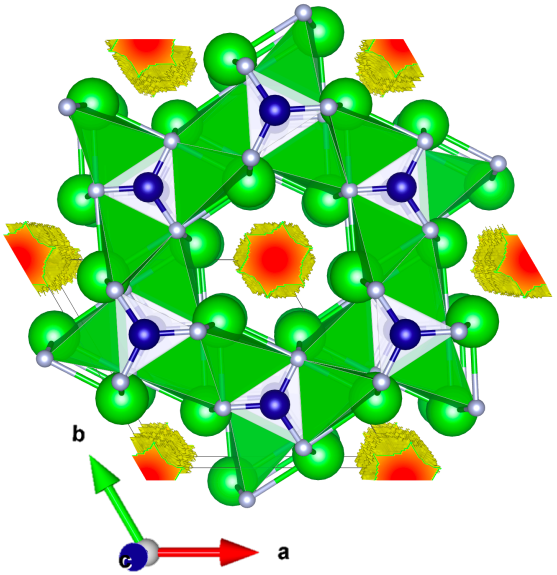
The first transition metal containing electride

- Chemical « rule »:

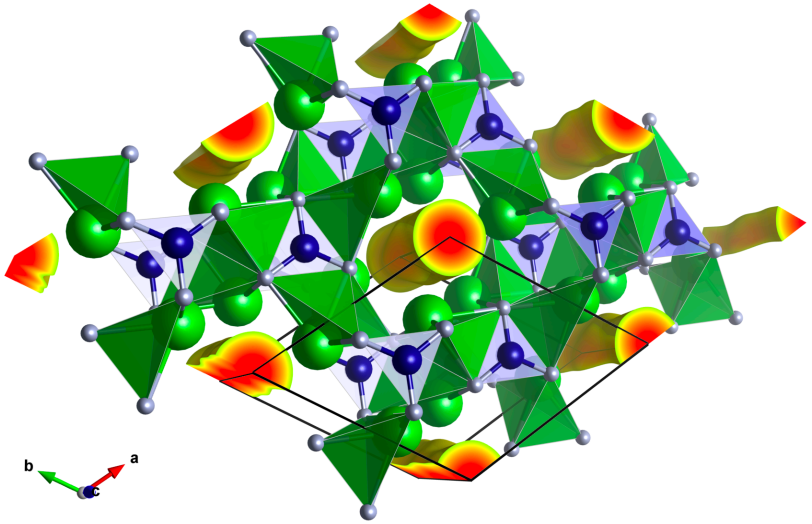
(partially filled d-shell) transition metal cannot lead to electride behavior

- Exception to the rule found through HT computing!

Ba_3**Cr**N_3 and Sr_3**Cr**N_3



Apparently $Ba_3Cr^{3+}N_3$ but actually $Ba_3Cr^{4+}N_3$ e^-



L. Burton, F. Ricci, G.-M. Rignanese, **GH**, *submitted*, http://arxiv.org/abs/1801.02948

The scaling up issue in high-throughput computing

- « Baby sitting » a few computations is fine…

the scaling up issue in high-throughput computing

- « Baby sitting » thousands of them becomes a challenge

Growing number of properties accessible high-thoughput

- Every new properties require the development of an automatic work-flows and tools.
- More and more « complex » computations

  - **HT GW**: M. Van Setten, M. Giantomassi, X. Gonze, G.-M. Rignanese, GH, *Phys. Rev. B*, 2017

  - **HT defects**: in progress, D. Brodberg, B. Medasani, N. Zimmermann, A Canning, M. Haranczyk, M. Asta, GH, accepted in *Computer Physics Communications*

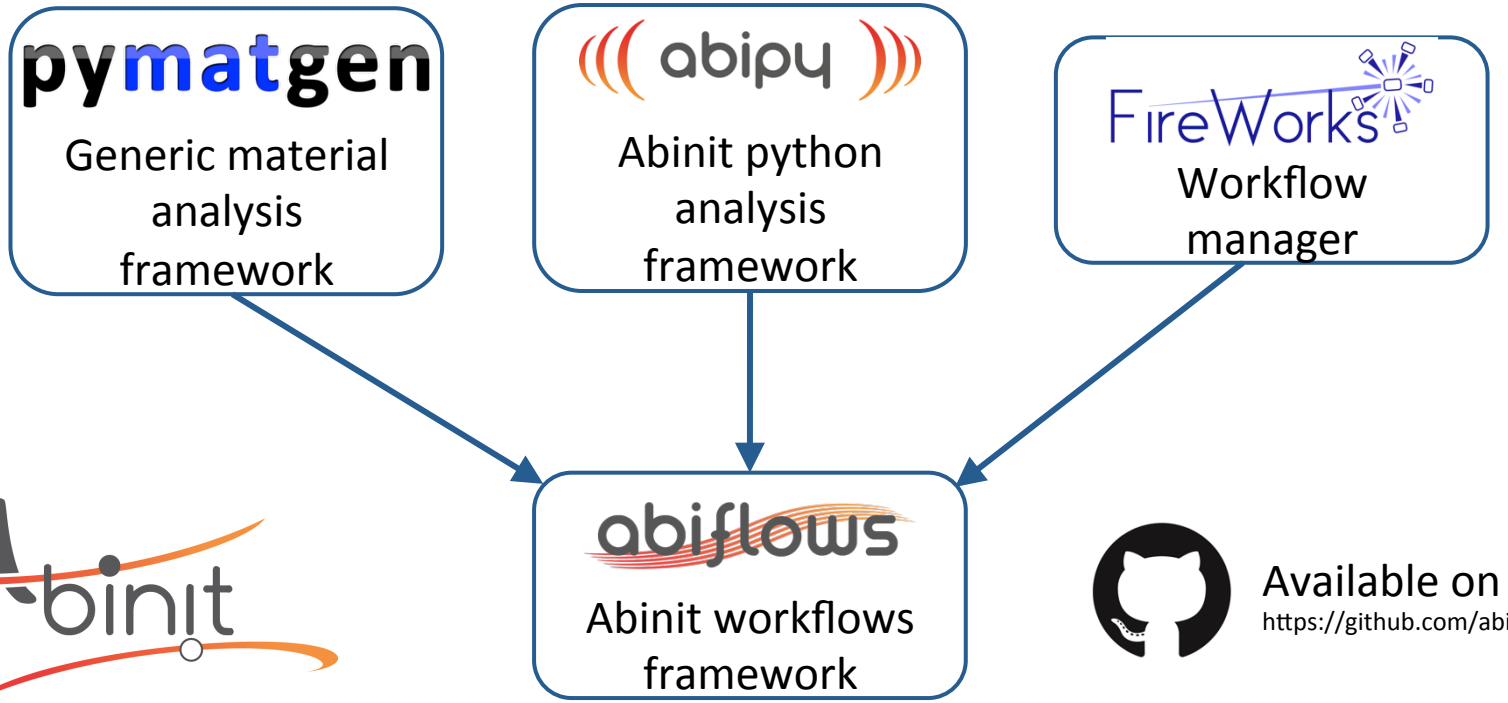
- HT phonons

High-throughput phonons within DFPT: automation, work-flows and software

- First step, automation algorithm (convergence in k-, q-points etc…)

  G. Petretto, X. Gonze, **GH**, G.-M. Rignanese, *Computational Materials Science*, 2018

- Second, coding it

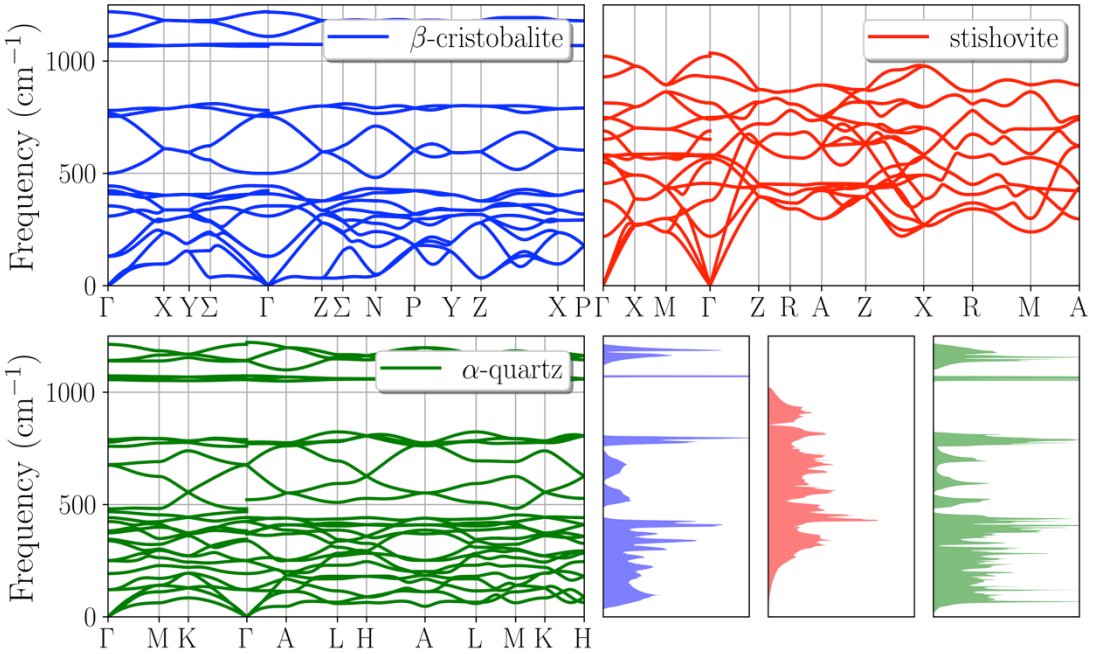


Abinit

Available on Github:
https://github.com/abinit/abiflows

A database of phonon band structures

- More than **1 500** phonon band structures computed automatically (DFPT, GGA-PBEsol)

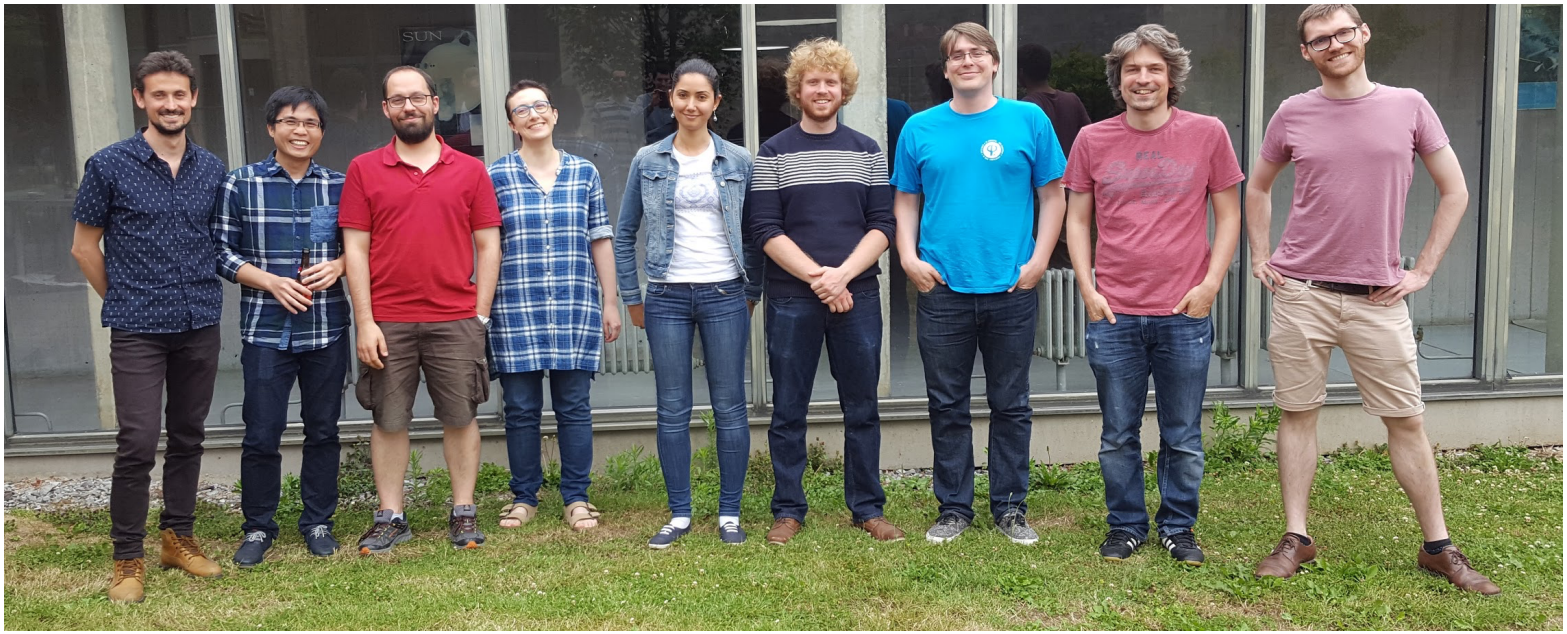


- Data soon available
  - Materials Project
  - G. Petretto, S. Dwaraknath, H. P. C. Miranda, D. Winston, M. Giantomassi, M. J. van Setten, X. Gonze, K. A. Persson, **GH**, G.-M. Rignanese, *submitted in Scientific Data*

Conclusions

- Materials Discovery can be tremendously accelerated by high-throughput computing
- Examples of TCOs and electrides
  - Thinking outside the box (new chemistries, new concepts…)
  - Questions known « rules »
  - Experimentally confirmed predictions
- The automation challenge
  - HT phonons
- More info:
  - http://perso.uclouvain.be/geoffroy.hautier
  - geoffroy.hautier@uclouvain.be

Acknowledgments



- **My group at UCL:** especially V.-A. Ha, F. Ricci, A. Miglio, H. Miranda, G. Petretto, L. Burton
- **Collaborators**
  - A. Jain, G. Ceder, K. Persson (Berkeley and LBNL), J. Varley (LLNL)
  - J. Suntivich, A. Bathia (Cornell)
  - M. Van Setten, G.-M. Rignanese, X. Gonze (UCL)
- **Funding**
  - FNRS
  - EU: Marie-Curie Integration grant (CIG): HT4TCO
  - US department of energy (DOE)