Computational Screening for Defect-Tolerant Semiconductors

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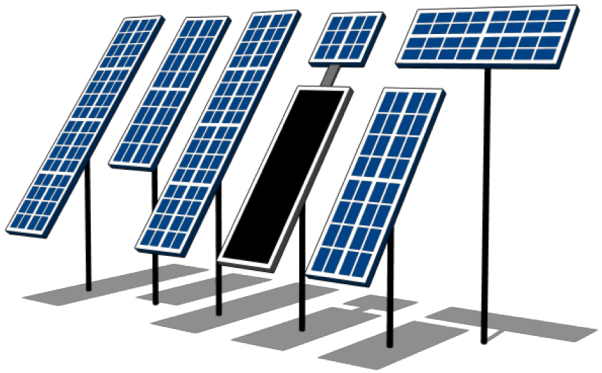
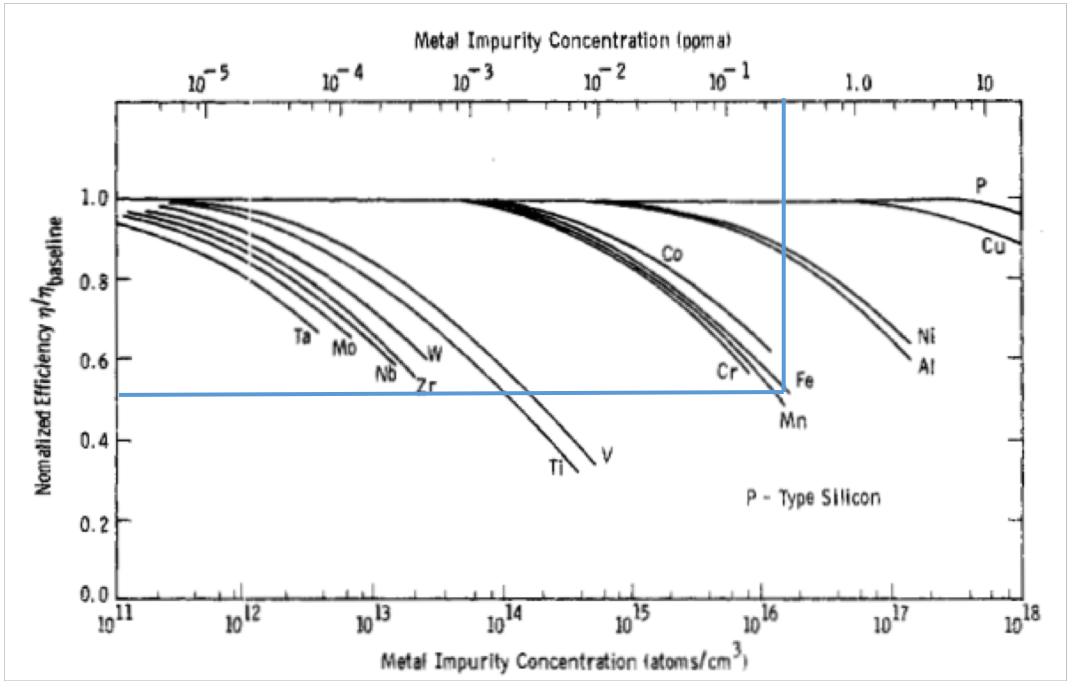
Gordon Research Seminar on Defects in Semiconductors
Colby-Sawyer College, New London, NH
August 18, 2018

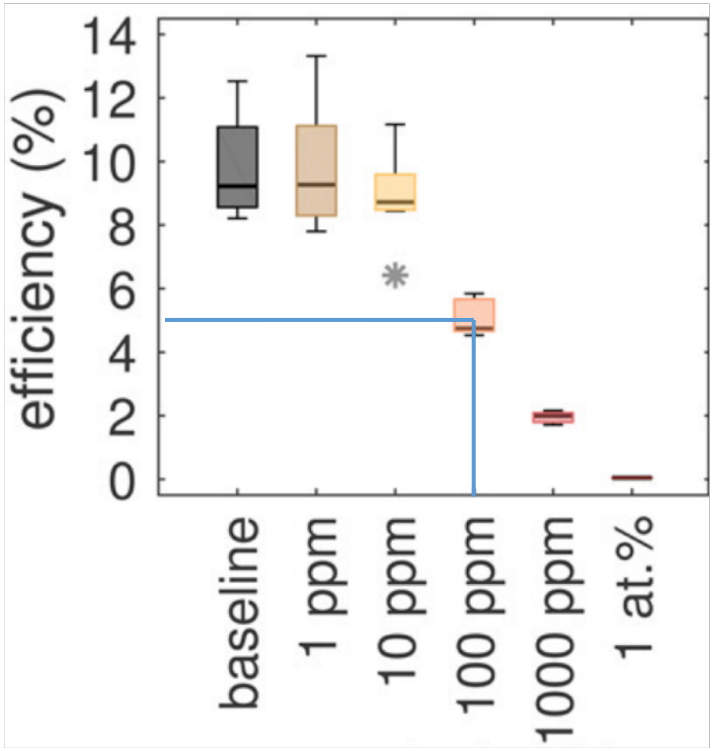


Photovoltaics Research Laboratory

Hybrid perovskites are *defect-tolerant*

defect tolerance
The tendency of a semiconductor to keep its properties (particularly charge carrier transport properties) despite the presence of crystallographic defects.

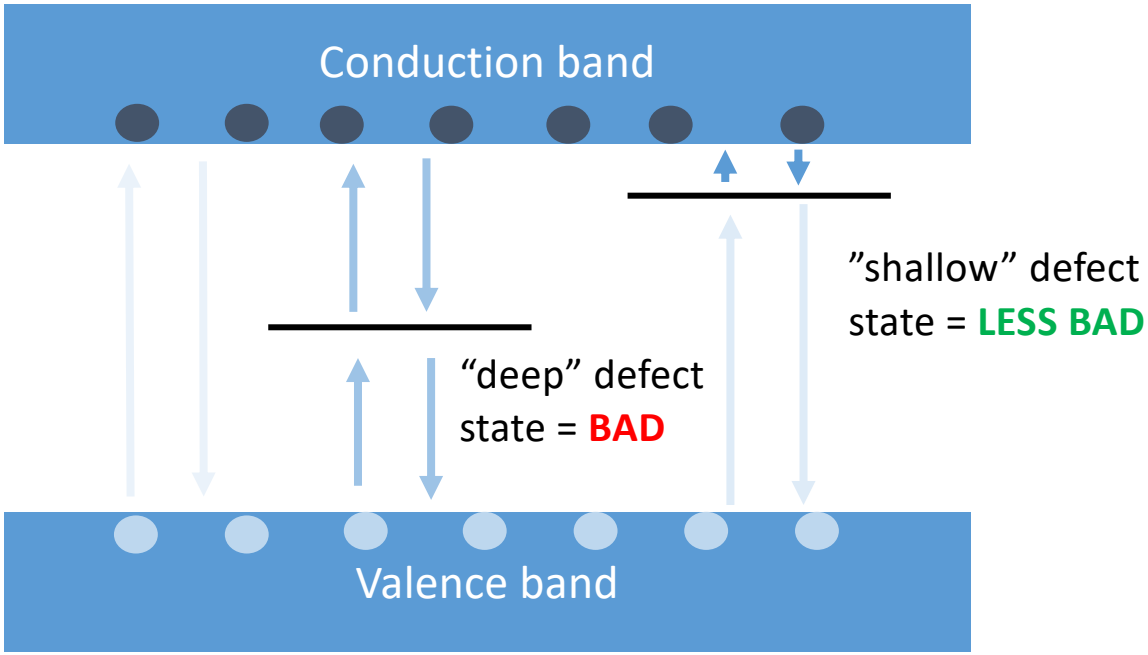




50% relative efficiency loss at over 100 TIMES the concentration of iron!

Zakutayev, A., *et al*. (2014). Defect tolerant semiconductors for solar energy conversion. *Journal of Physical Chemistry Letters*, *5*(7), 1117–1125.
J. R. Davis *et al.* (1980) "Impurities in silicon solar cells," in *IEEE Transactions on Electron Devices*, 27(4), 677-687
Poindexter, J. R., *et al.* (2017). High Tolerance to Iron Contamination in Lead Halide Perovskite Solar Cells. *ACS Nano*, 11(7), 7101-7109

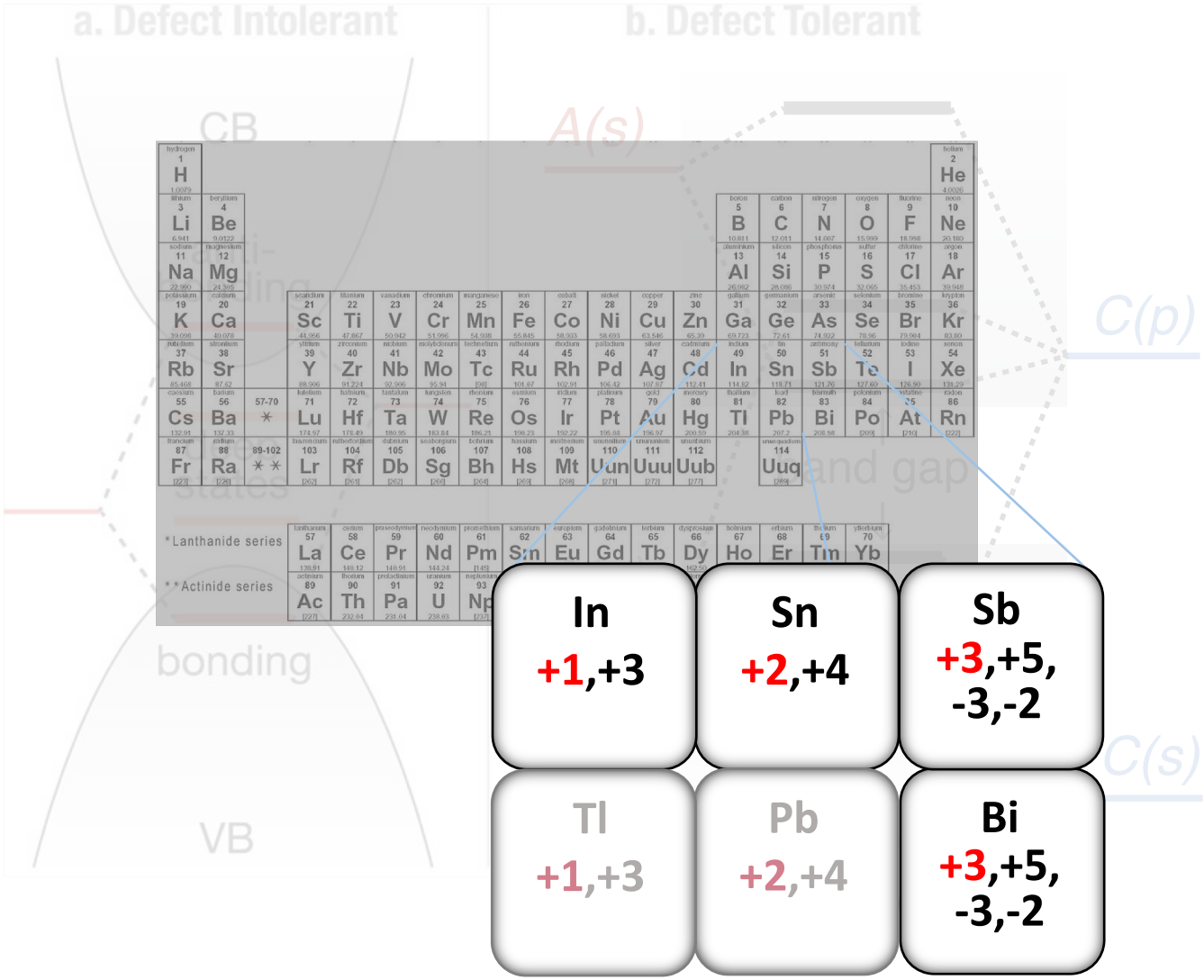
Where does defect tolerance come from?



In defect-tolerant materials, the abundant (i.e. low-formation-energy) defects tend to be shallow in nature!

Shockley, W., & Read, W. T. (1952). Statistics of the Recombination of Holes and Electrons. *Physical Review*, *87*(46), 835–842.
Hall, R. N. (1952). Electron-Hole Recombination in Germanium. *Physical Review*, *87*(2), 387.

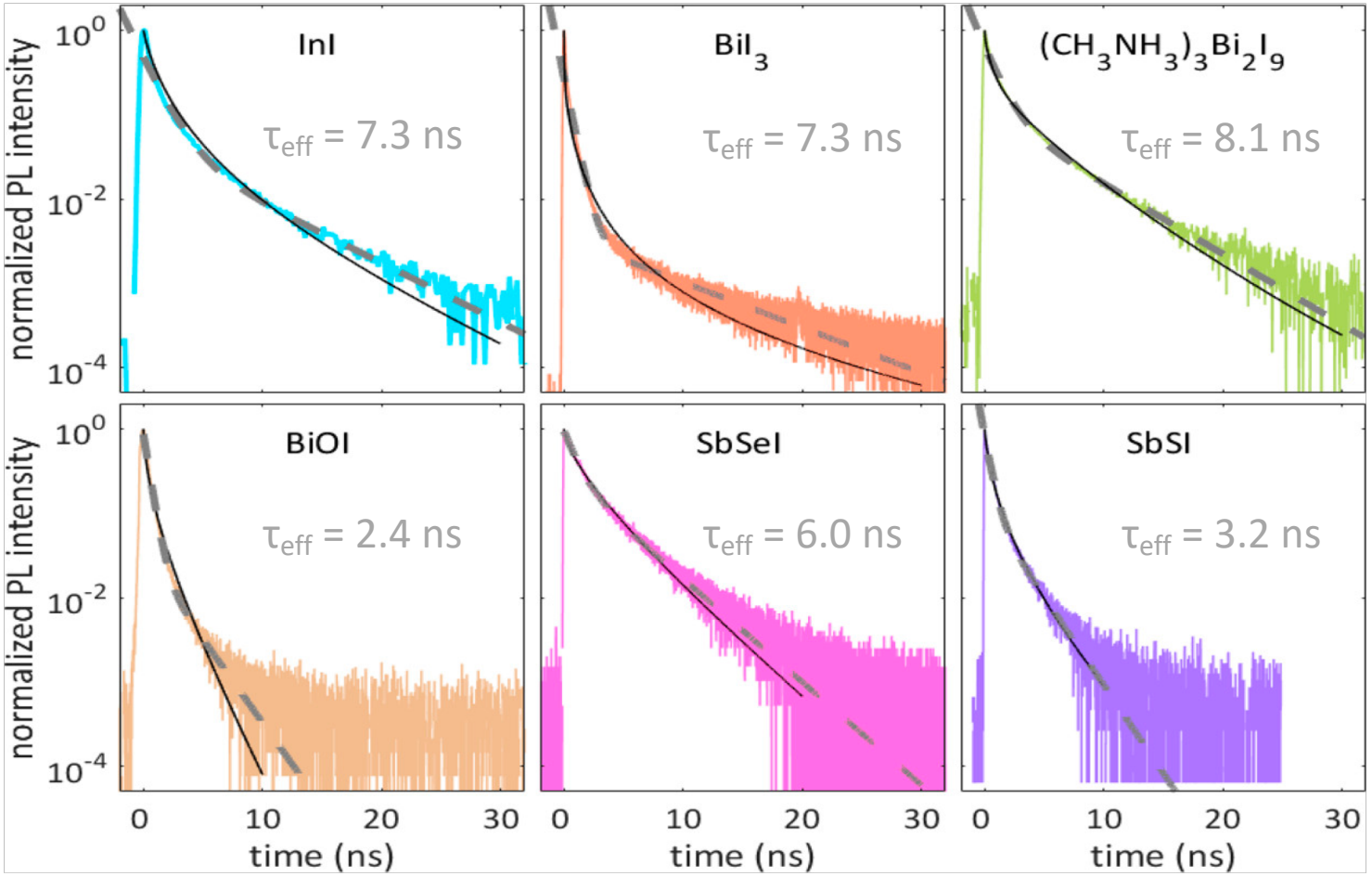
Where does defect tolerance come from?



Brandt, R. E. *et al.* (2017). Searching for " Defect-Tolerant " Photovoltaic Materials: Combined Theoretical and Experimental Screening. *Chemistry of Materials*, 29(11), 4667-4674

Kurchin, R. C. *et al.* (2018). Structural and Chemical Features Giving Rise to Defect Tolerance of Binary Semiconductors *Chemistry of Materials*, just accepted.

…so we tried it!

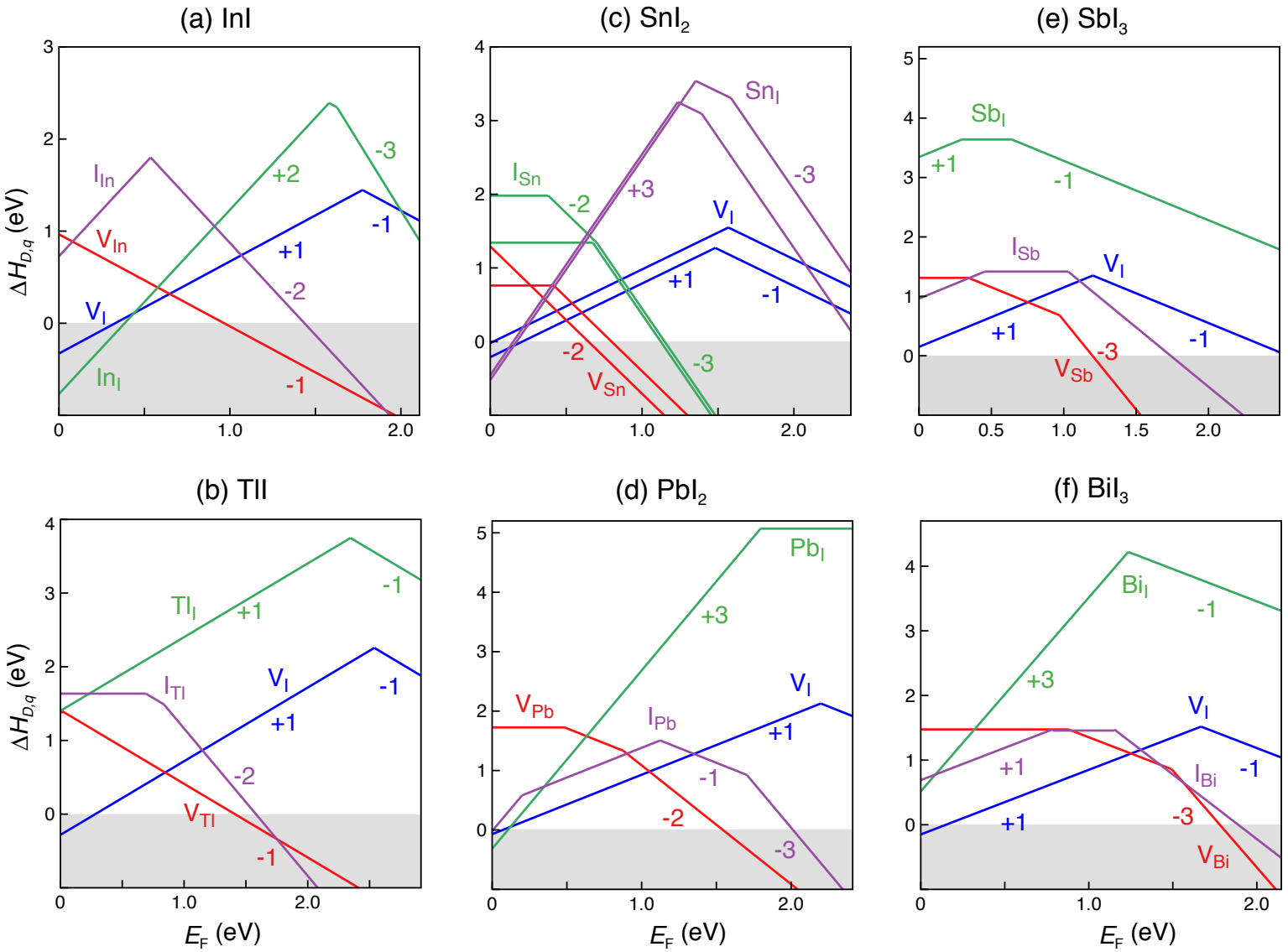


But τ_{eff} in perovskites is $\mathcal{O}$(100) ns…

…something's missing from our story.

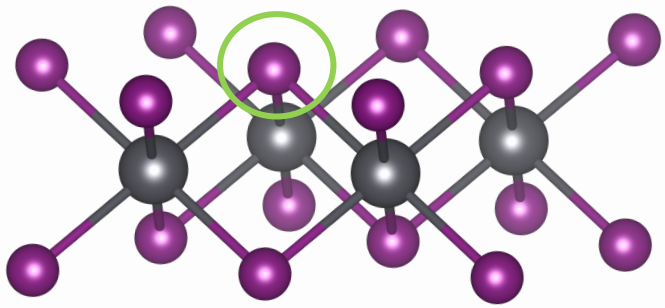
Brandt, R. E. *et al.* (2017). Searching for " Defect-Tolerant " Photovoltaic Materials: Combined Theoretical and Experimental Screening. *Chemistry of Materials*, 29(11), 4667-4674

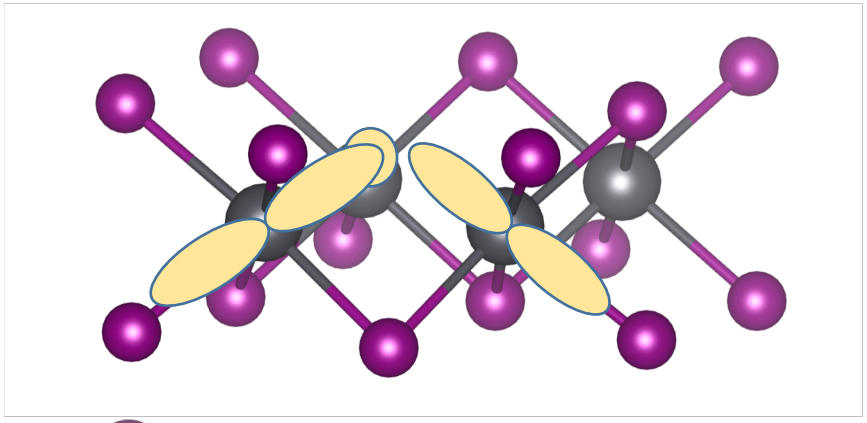
Defect Calculations

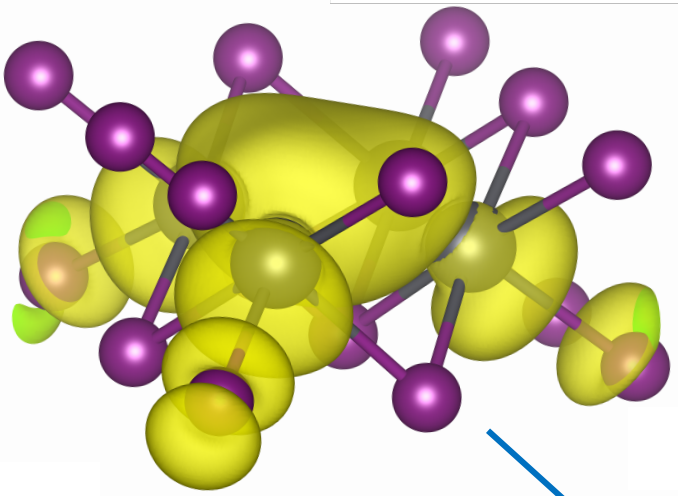


Kurchin, R. C. *et al.* (2018). Structural and Chemical Features Giving Rise to Defect Tolerance of Binary Semiconductors *Chemistry of Materials*, DOI:10.1021/acs.chemmater.8b01505.

Back to the drawing board!



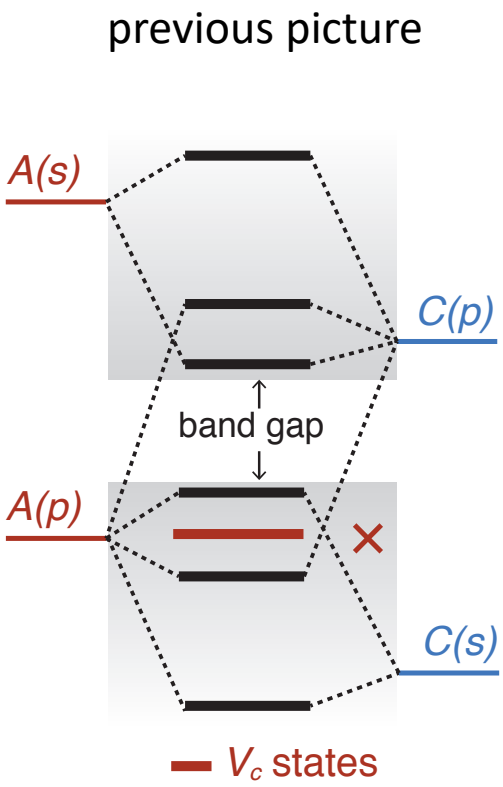


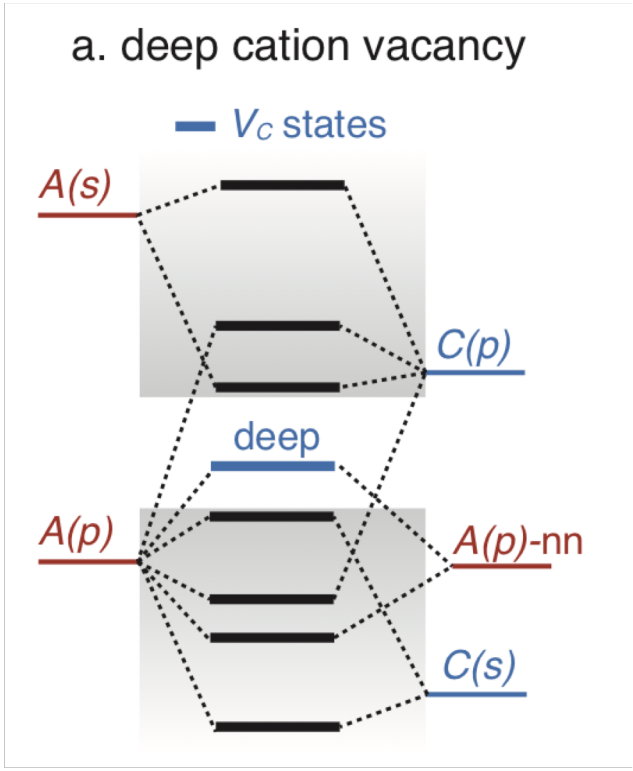


change the **structure**

change the **energies** (chemistry)

Chemistry





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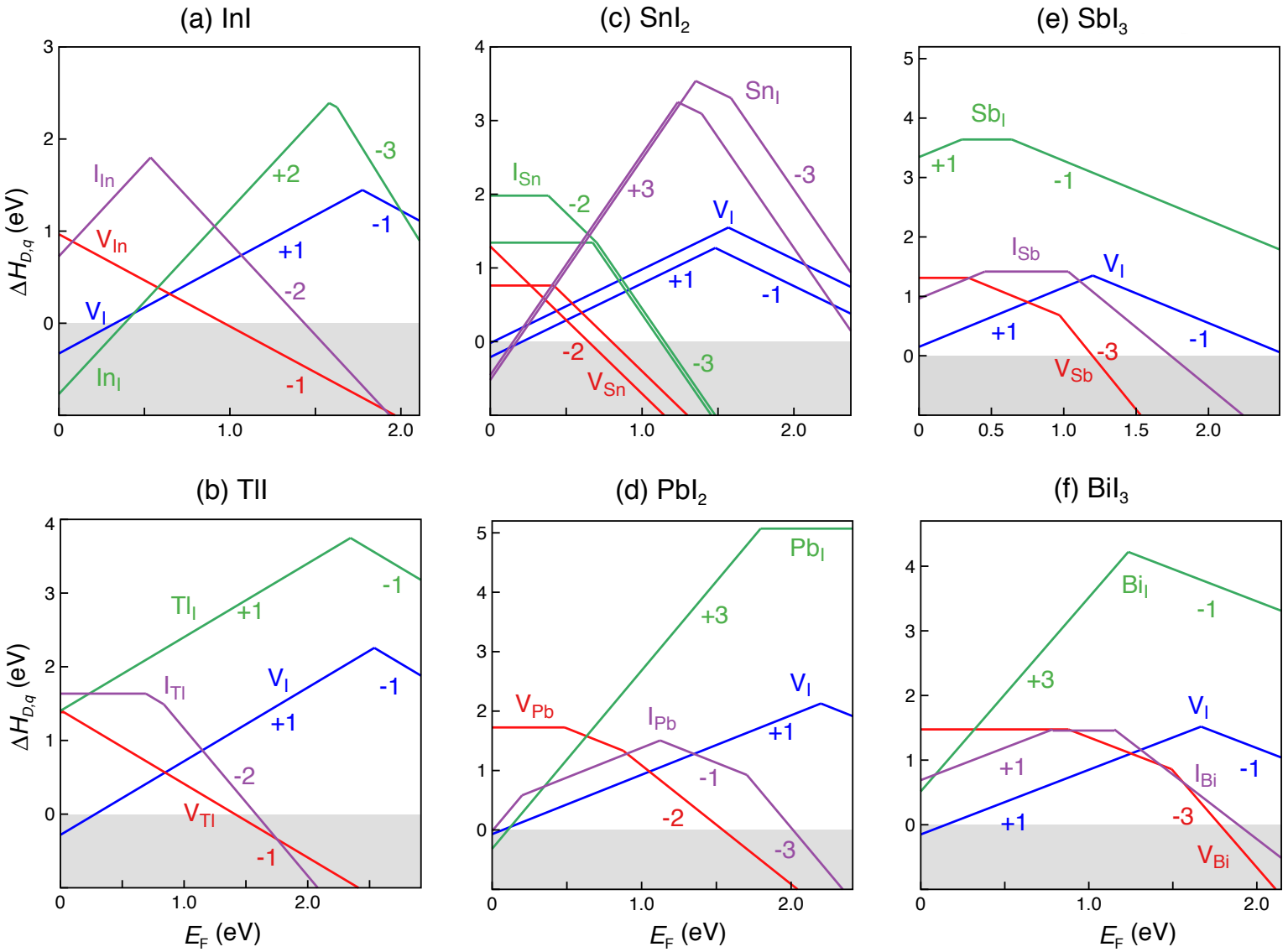
Defect Calculations



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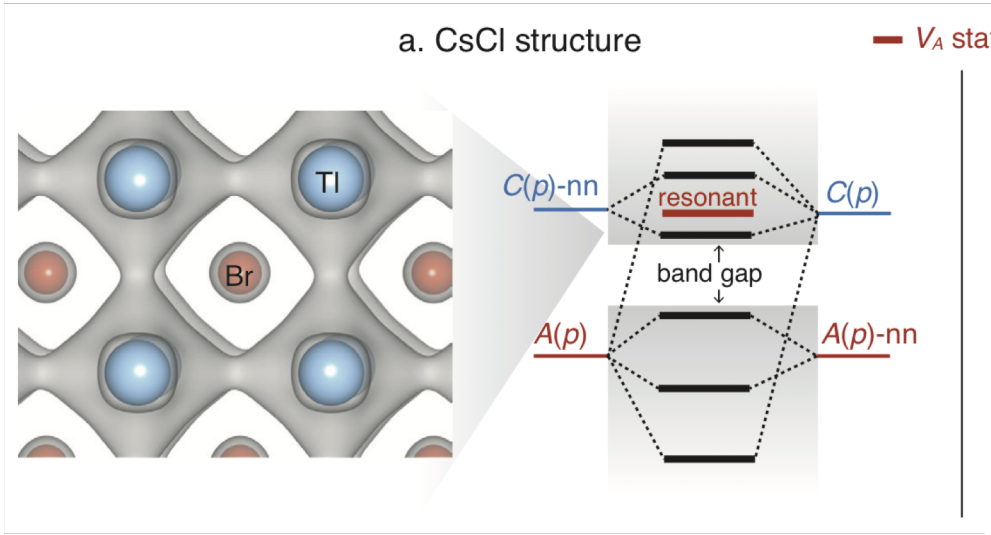
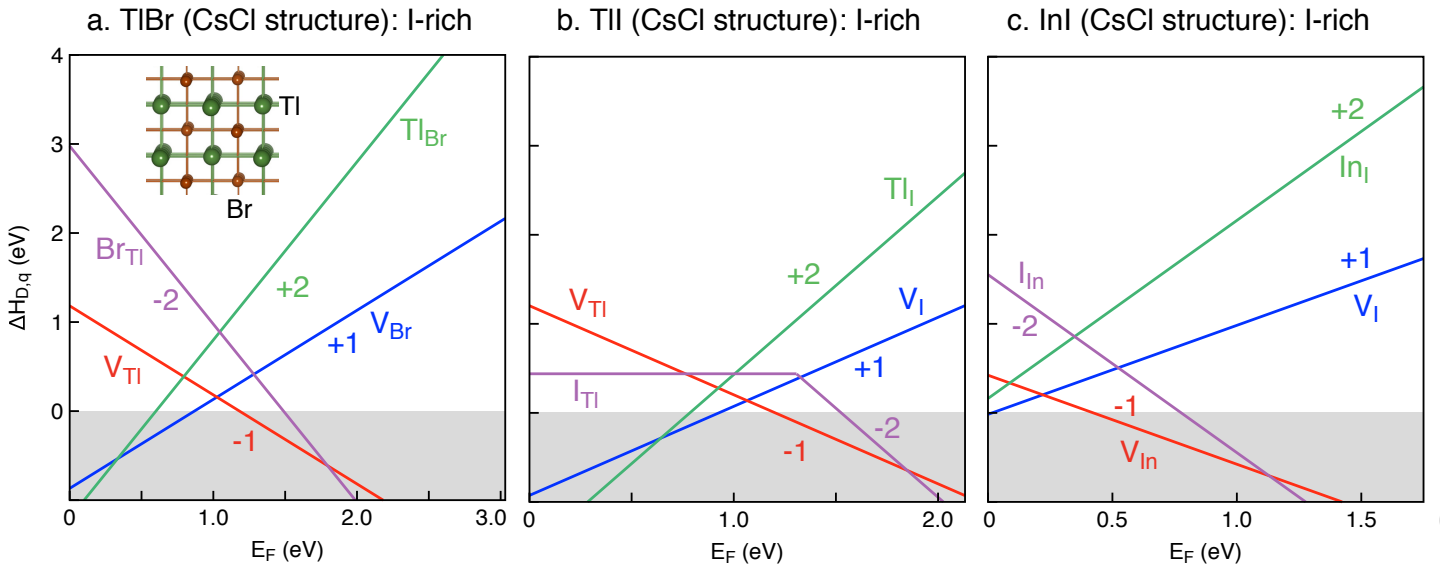
Structure





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Conclusions

- Defect tolerance is a critical characteristic making a PV material amenable to low-cost, scalable manufacturing techniques
- The prior screening criteria were necessary but not sufficient to ensure defect tolerance
- We have described additional structural and chemical criteria contributing to defect-tolerant behavior with respect to intrinsic defects:
  - Valence energy levels of cation and anion should be well aligned
  - Crystal structure should inhibit unfavorable interactions
- Future work will generalize these updated criteria to ternary and multinary systems and apply them in materials search/design efforts

Thank you!

- Questions?

BLUE WATERS

CNGMD
Incorporating Metastability

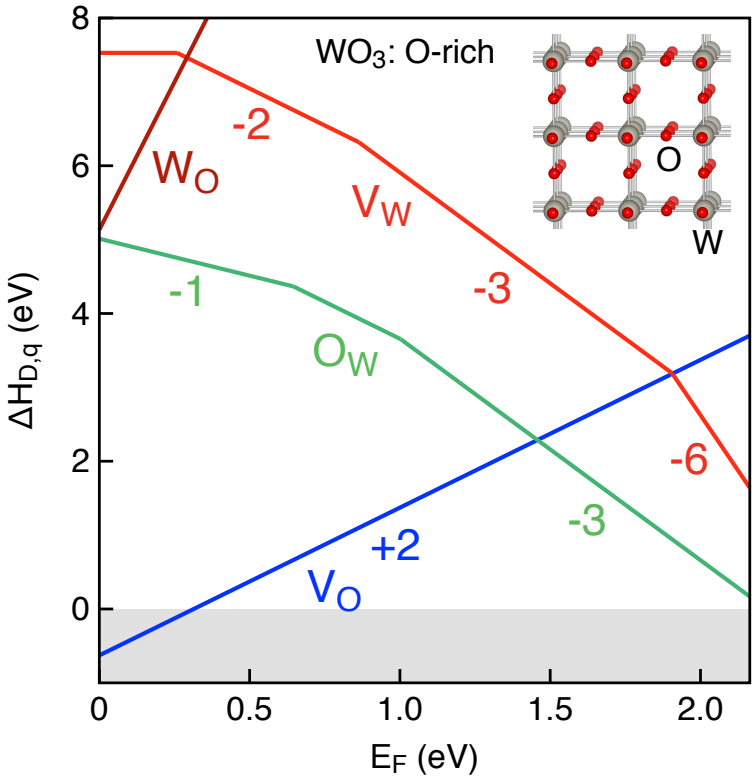
SENER
SECRETARÍA DE ENERGÍA

ESTADOS UNIDOS MEXICANOS

MASSACHUSETTS
CLEAN ENERGY
CENTER

Backup slides!

Another test: good structure but bad chemistry



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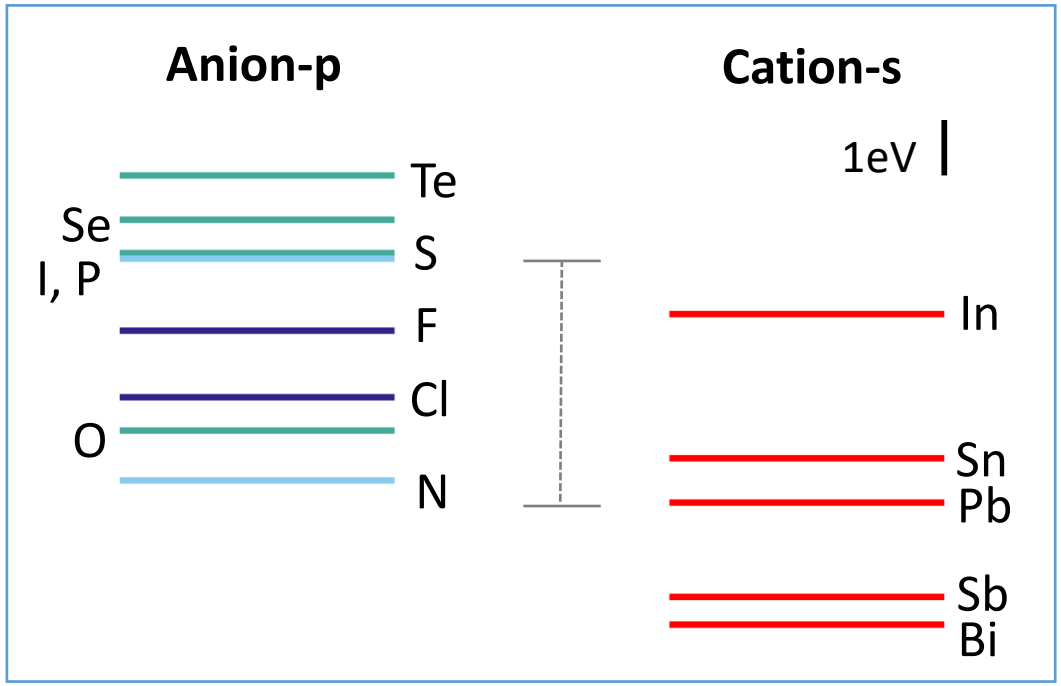
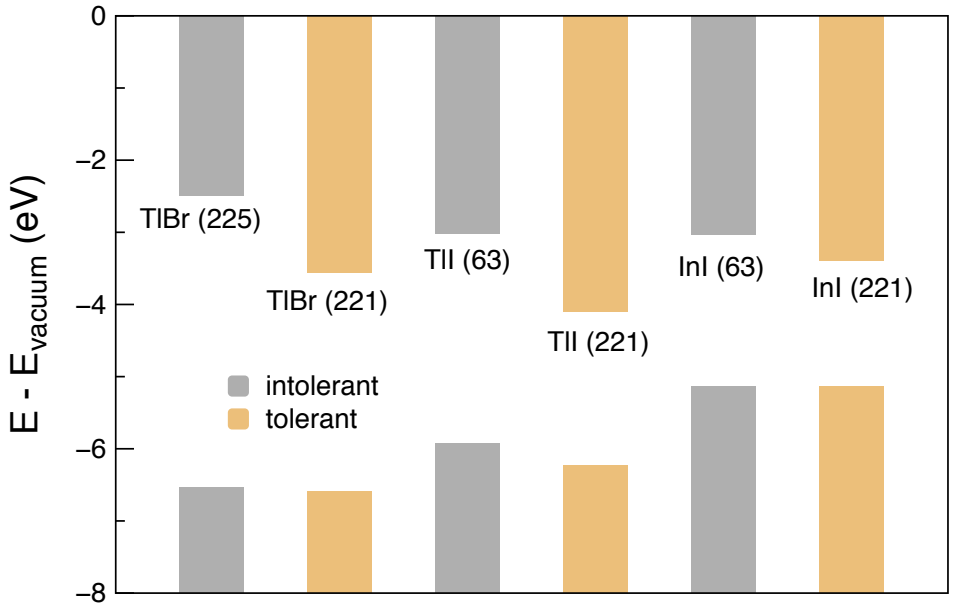
Approximate atomic energy levels



Trends in Band Alignment



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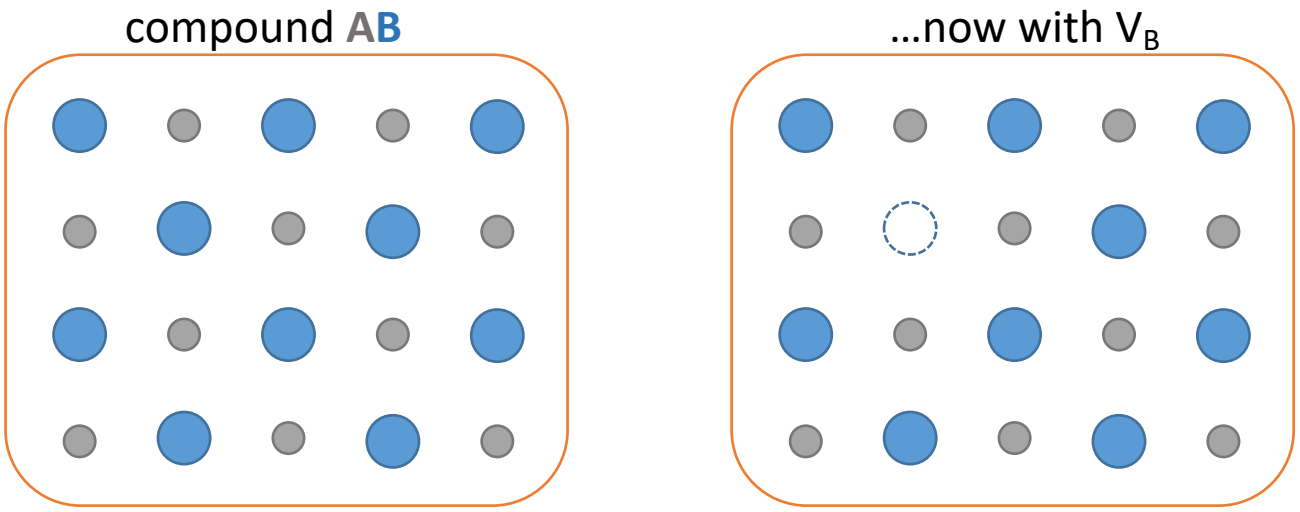
DFT Defect Calculations



$$\Delta H_{\mathrm{D,q}}(E_F) \approx [E_{\mathrm{D,q}} - E_{\mathrm{host}}] + qE_F + \sum_{\alpha} n_\alpha \mu_\alpha + E_{\mathrm{corr}}$$

Electrochemical potential *(electrons)*

Chemical potential *(for atoms added/removed)* (here 1 x μ_B)

Compensate for supercell size effects

Lany, S. and Zunger, A. (2008) 'Assessment of correction methods for the band-gap problem and for finite-size effects in supercell defect calculations: Case studies for ZnO and GaAs', *Physical Review B - Condensed Matter and Materials Physics*, 78(235104), pp. 1–25.