Toward Quantitative Metrics to Screen for Defect Tolerance in Novel Semiconducting Materials

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MRS Fall Meeting & Exhibit

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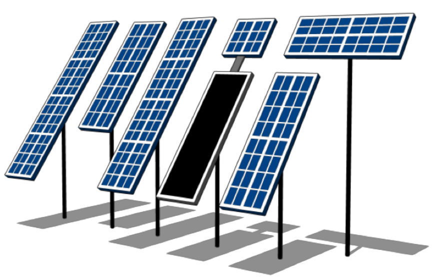
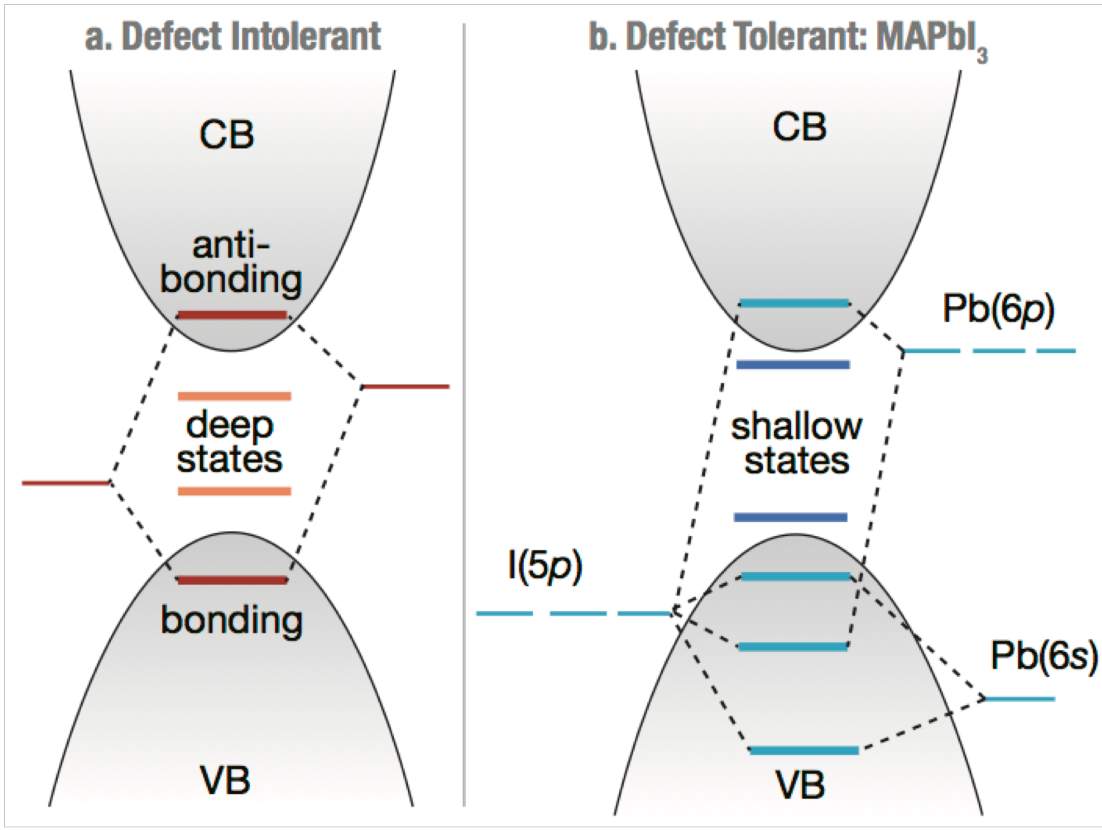
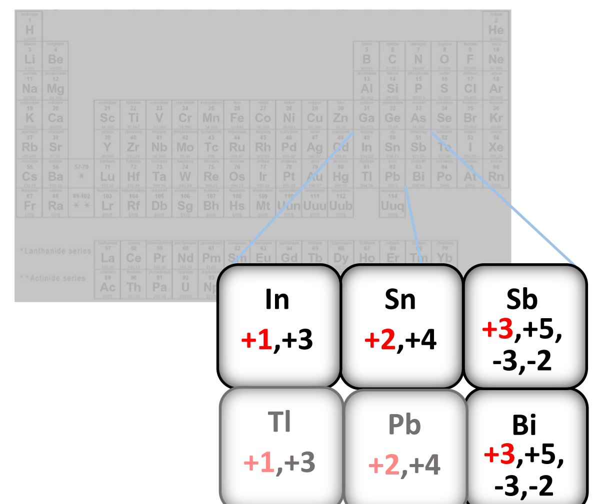


“Perovskite-Inspired” Materials

Inspired by the dramatic success of $MAPbI_3$ perovskites and related compounds, we seek to understand the **general physical principles** underlying that success, so we can use those principles to design **new compounds** that won’t suffer from perovskites’ drawbacks.

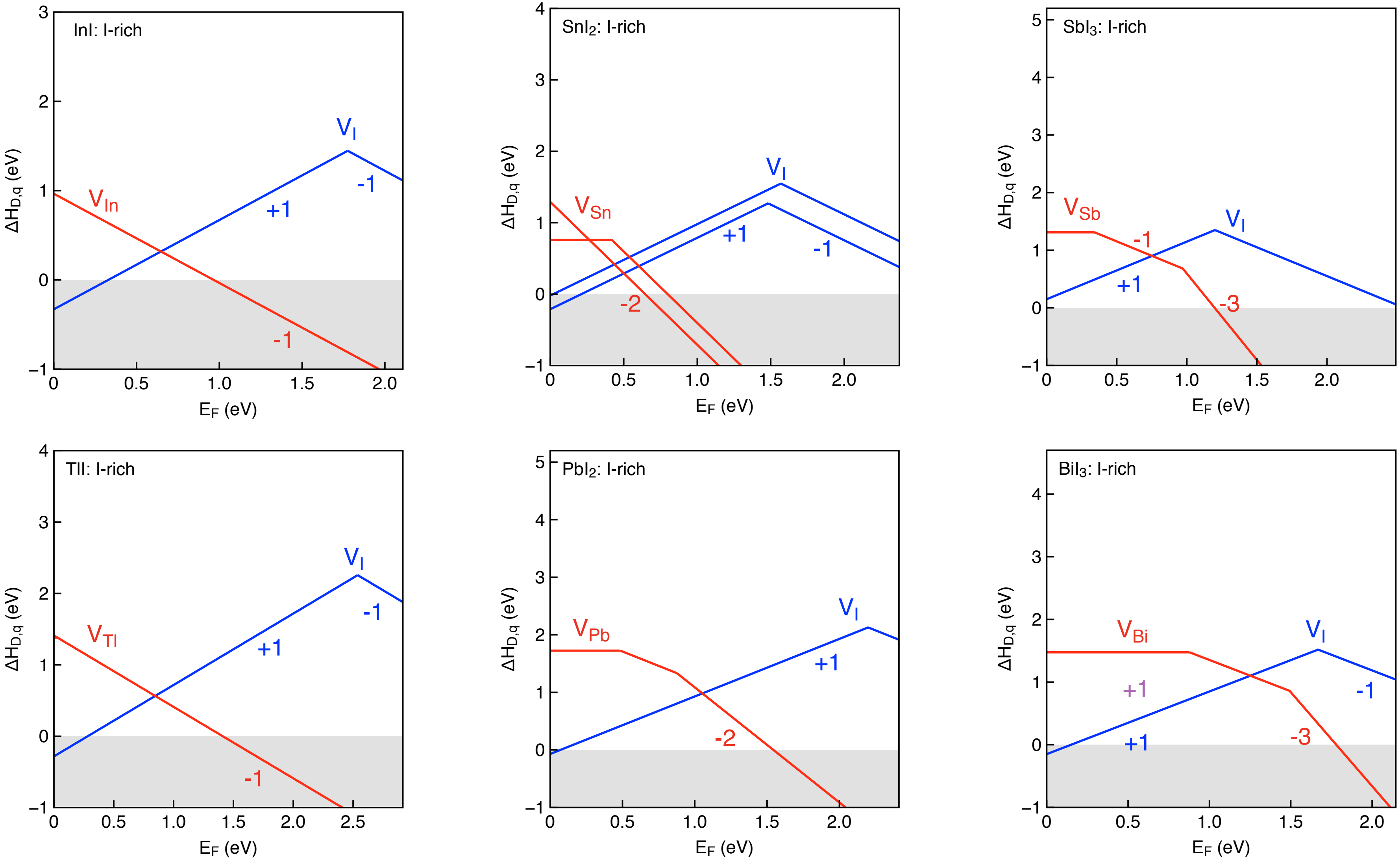


Antibonding VBM enabled by **partially oxidized ns^2 cation**

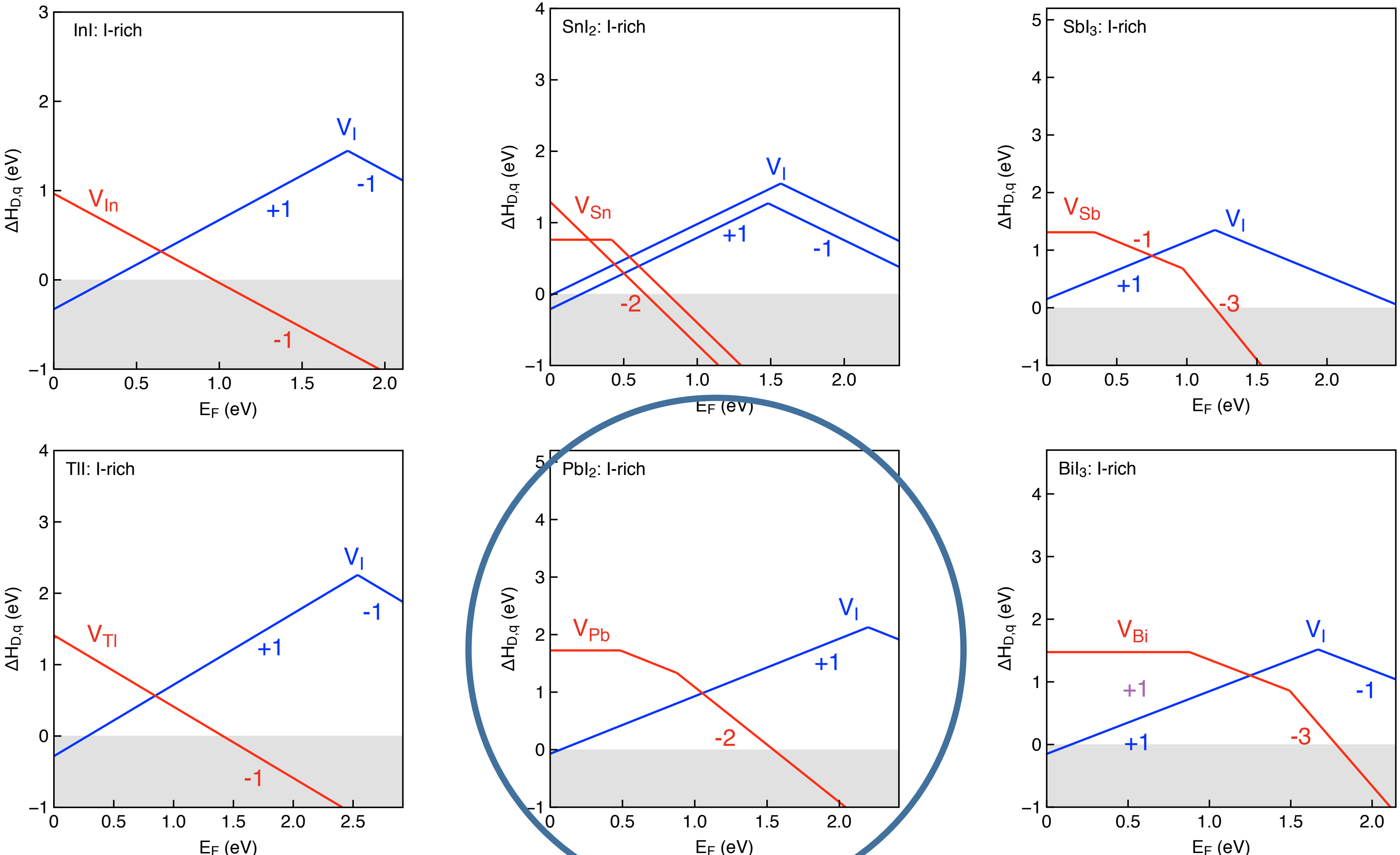


Brandt, R. E., Stevanović, V., Ginley, D. S., & Buonassisi, T. (2015). Identifying defect-tolerant semiconductors with high minority-carrier lifetimes: beyond hybrid lead halide perovskites. *MRS Communications*, *5*(2), 265–275.

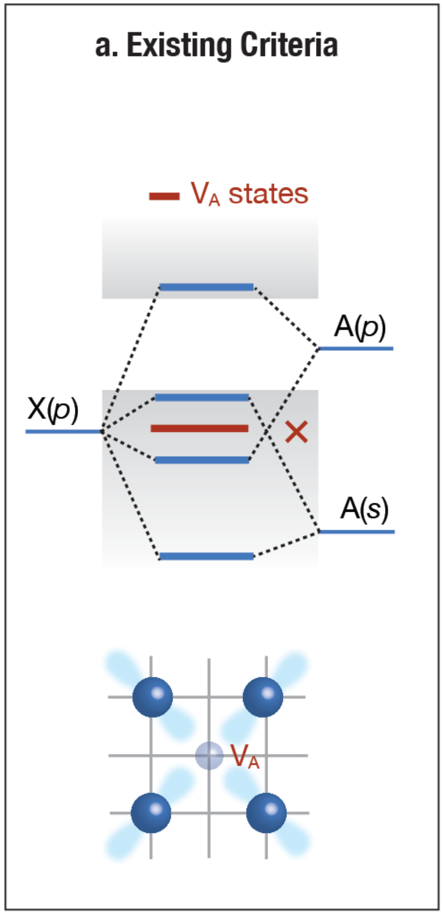
Binary iodides



Observation 1 – PbI_2 doesn't work; MAPI does!



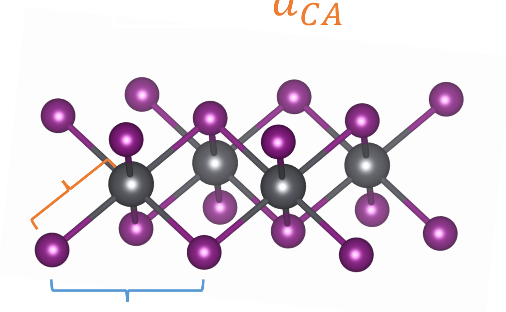
Updating Our Understanding

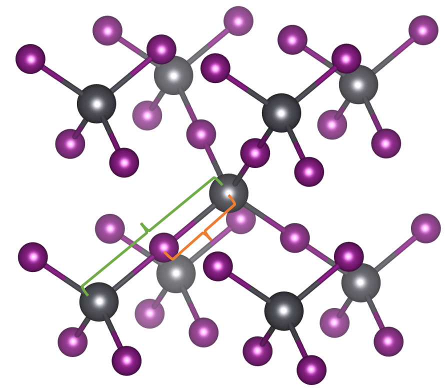


Quantifying the Structural Criterion

Cation vacancy:

$$DR_C = \frac{d_{AA}}{d_{CA}}$$

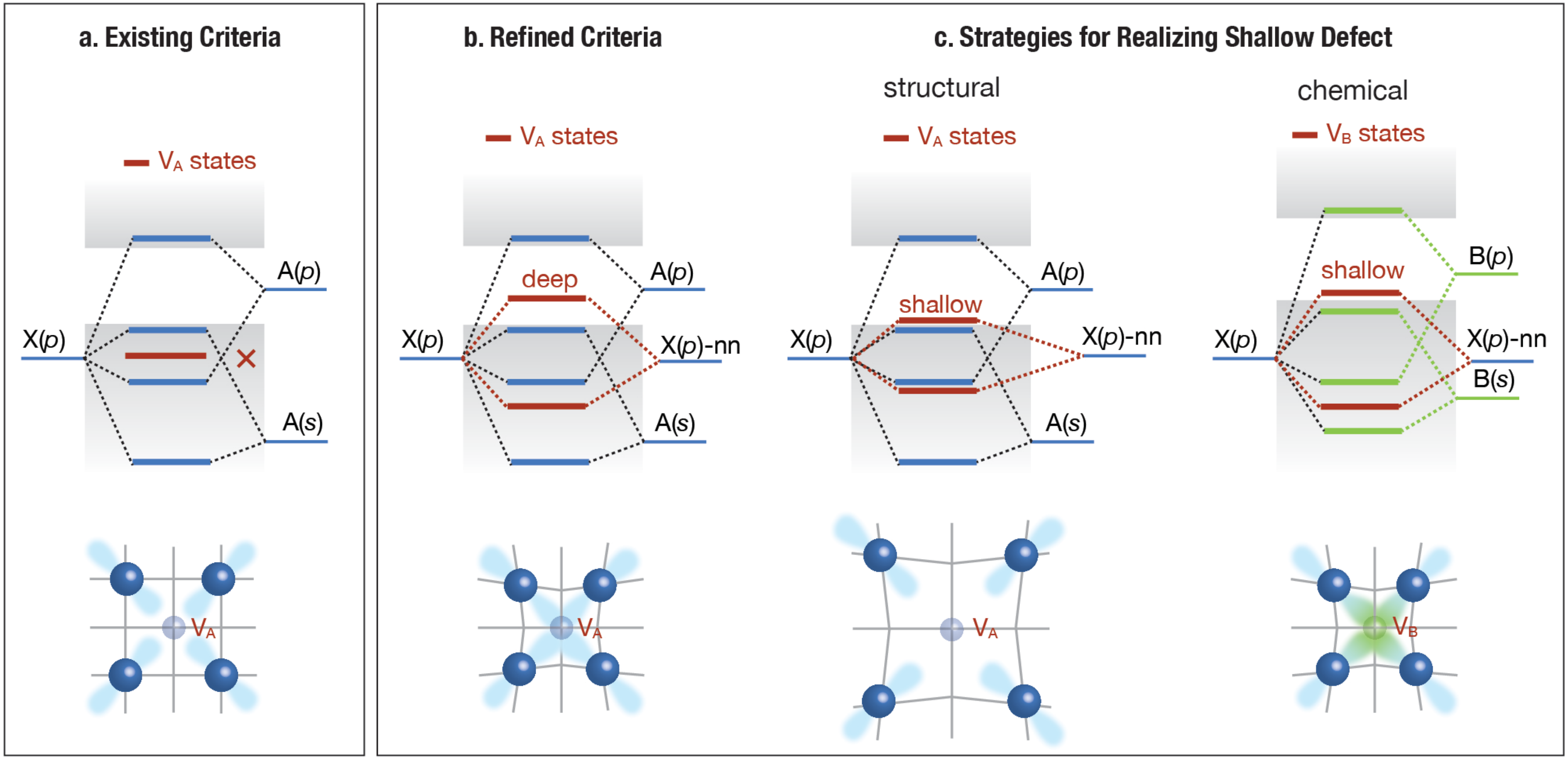




Anion vacancy:

$$DR_A = \frac{d_{CC}}{d_{CA}}$$

Updating Our Understanding

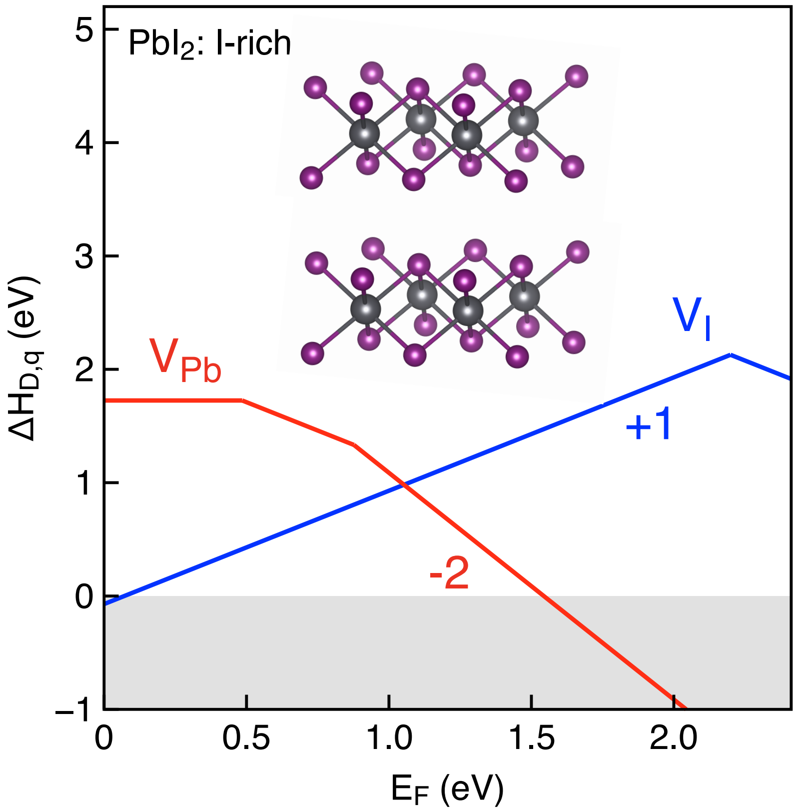


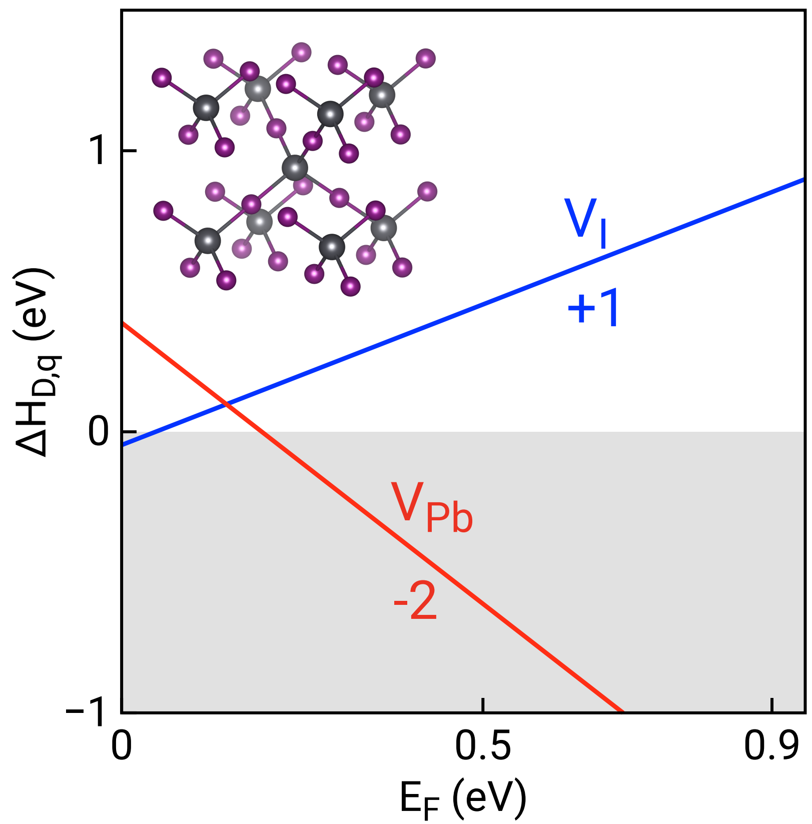
Alternate PbI_2 structures

Ground-state:
$DR_A = 1.4$, $DR_C = 1.4$

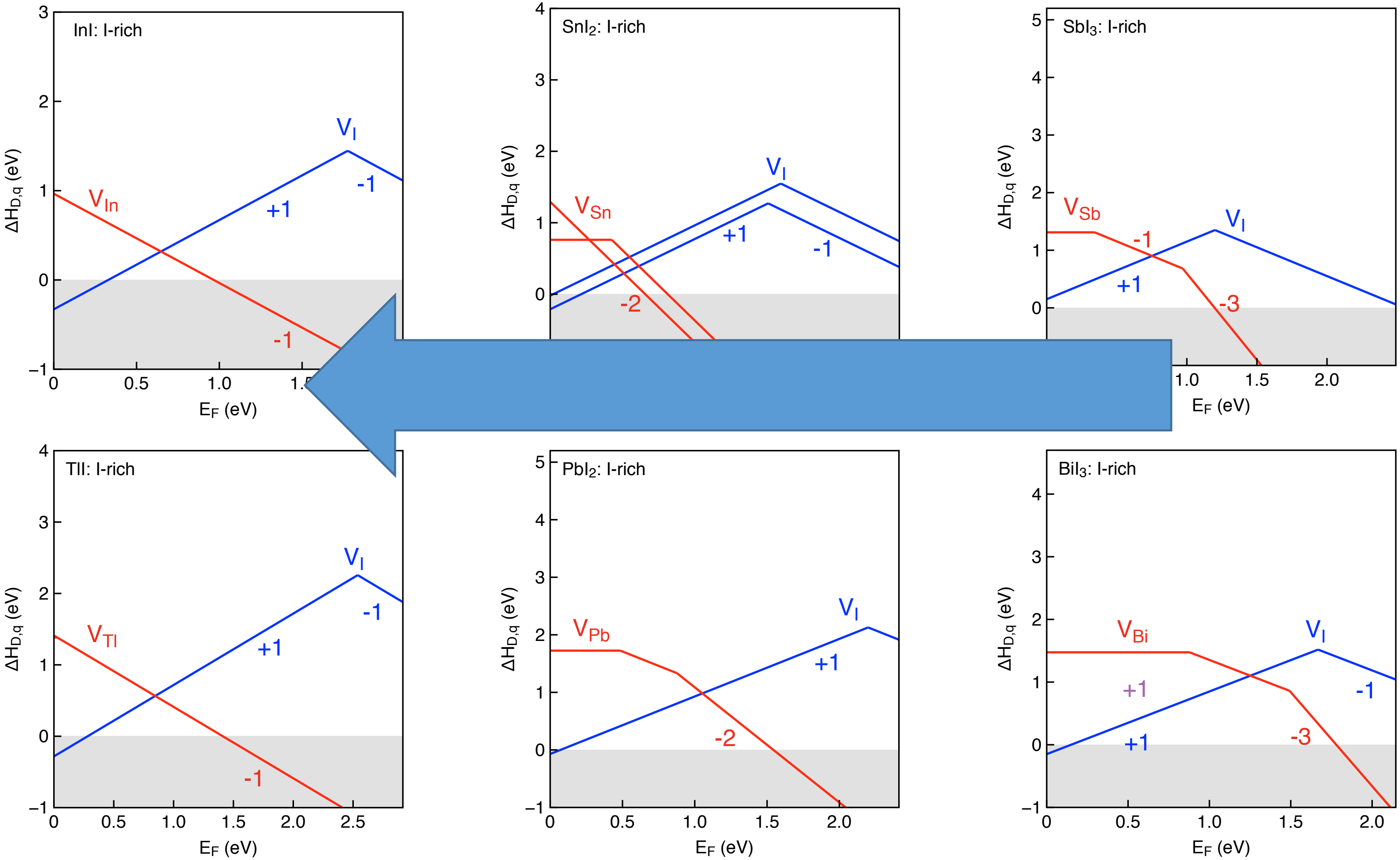
Hypothetical cubic:
$DR_A = 2.0$, $DR_C = 1.6$

both:
$OA = 4.4$ eV

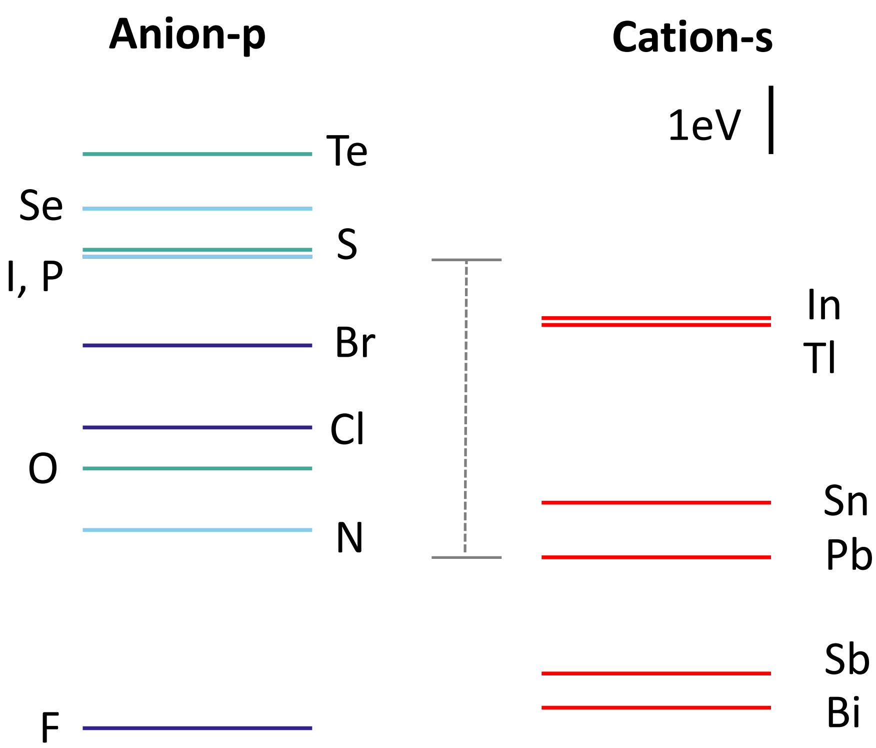




Observation 2 – defects get shallower left→right



Quantifying the Chemical Criterion

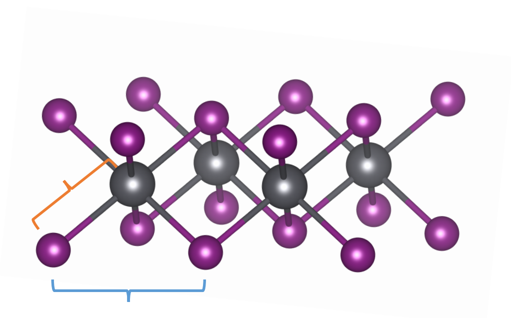


Orbital alignment:

$$OA = |E_{cat} - E_{an}|$$

Quantifying the New Criteria

Structural: distance ratio

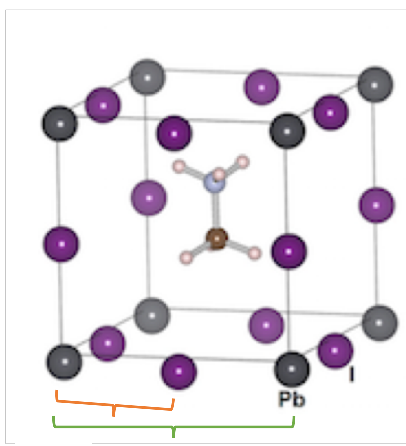


Cation vacancy:

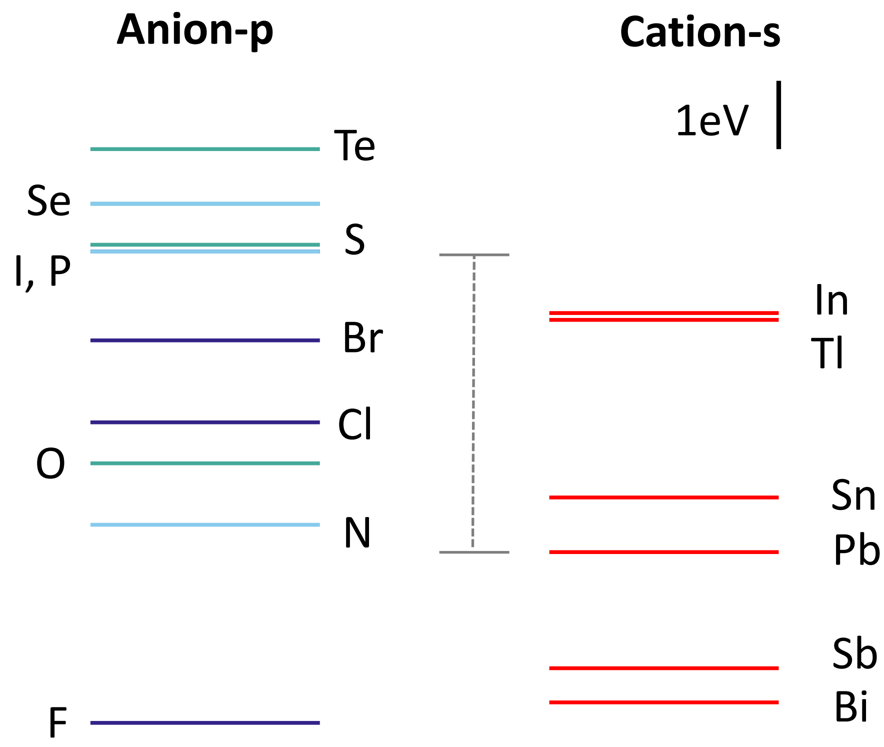
$$DR_C = \frac{d_{AA}}{d_{CA}}$$

Anion vacancy:

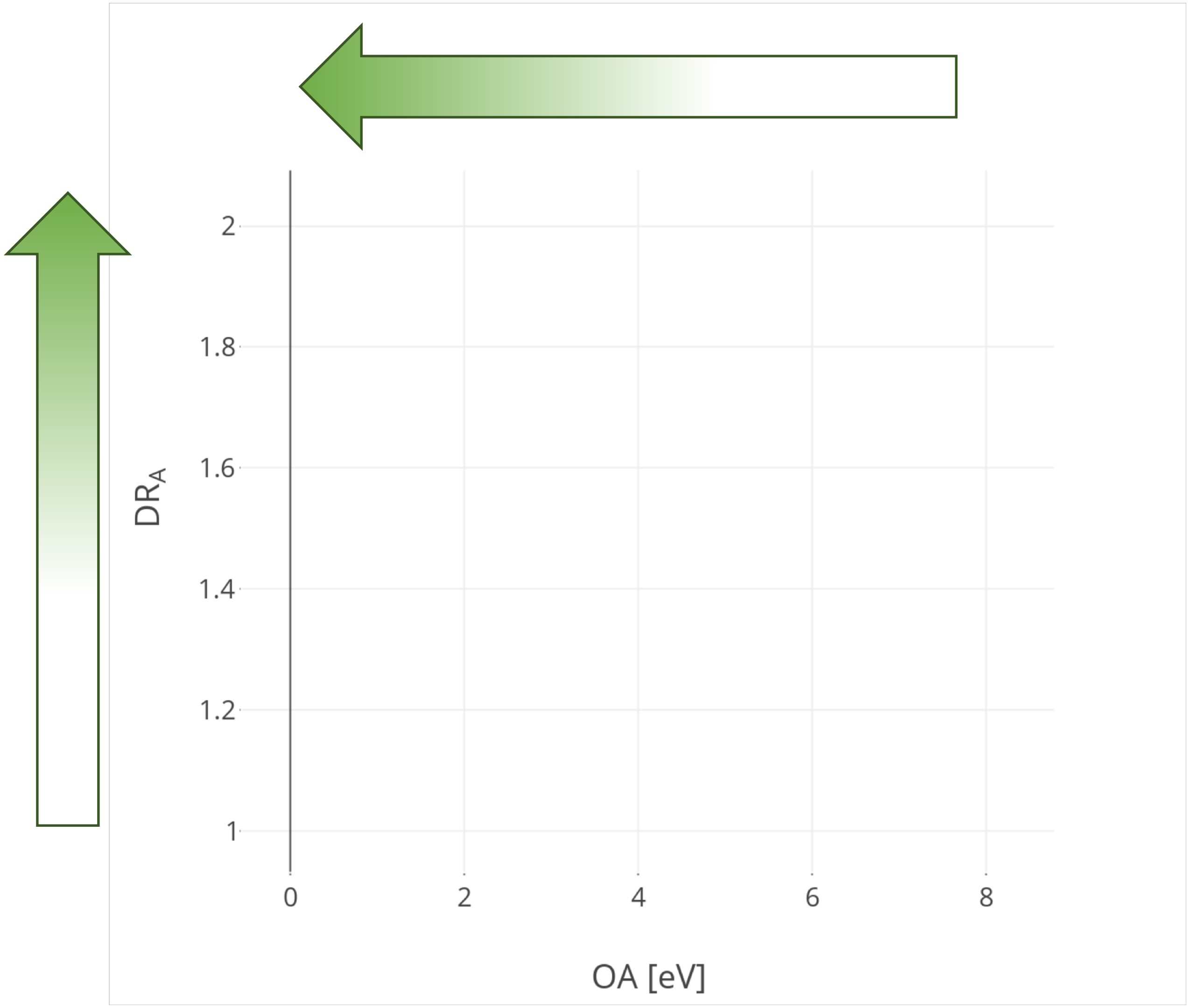
$$DR_A = \frac{d_{CC}}{d_{CA}}$$



Chemical: orbital alignment

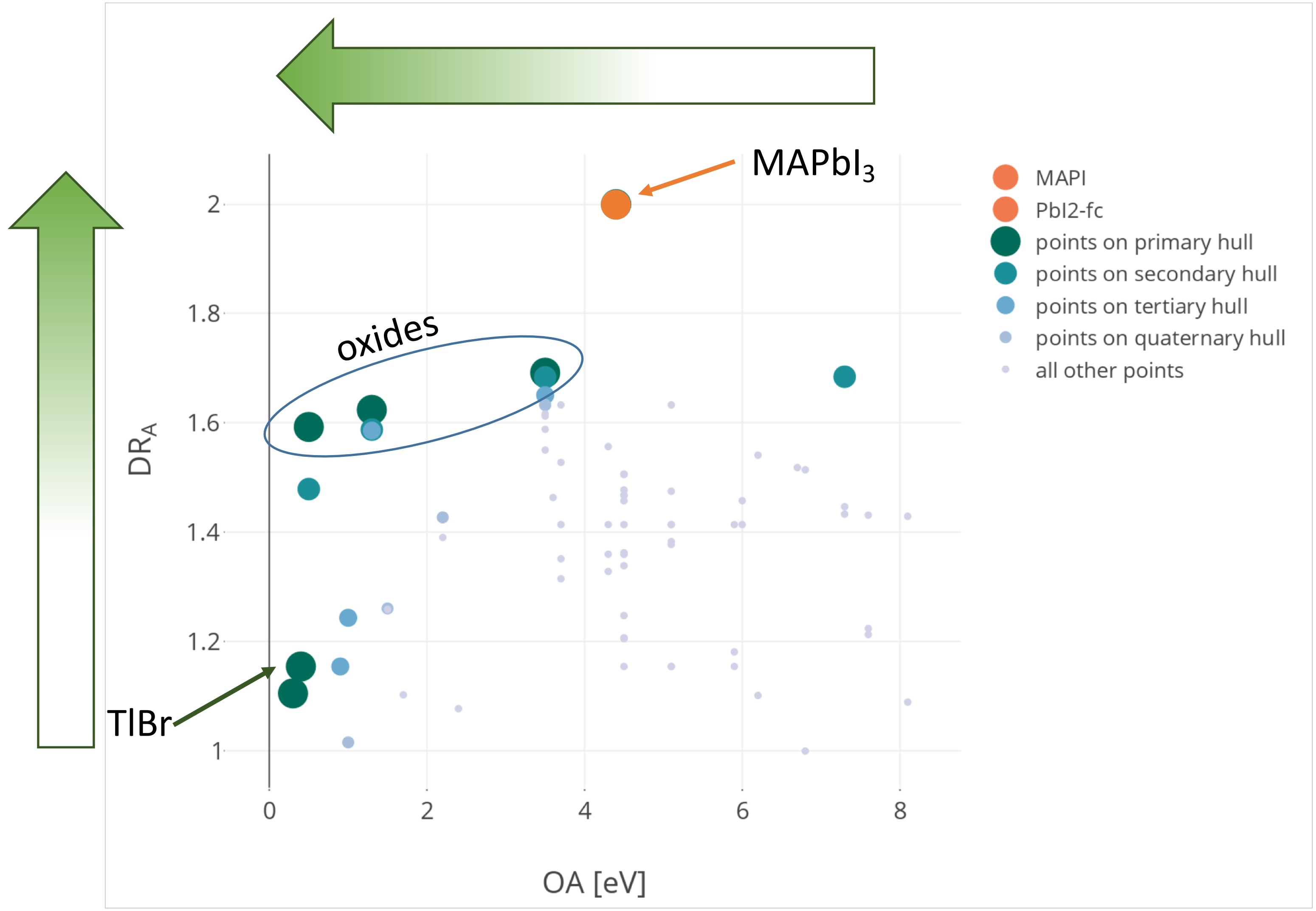


Searching all ICSD binaries with ns^2 cations



Shi, H., & Du, M.-H. (2014). Shallow halogen vacancies in halide optoelectronic materials. *Physical Review B*, *90*(17), 174103.

Searching all ICSD binaries with ns^2 cations



Conclusions/Next Steps

- Interactions between atoms at the site of a vacancy (or other defect) must be considered when developing generalized predictions of defect energetics
- If we seek a nontoxic, defect-tolerant material with an appropriate bandgap for photovoltaic applications, we likely need to look beyond binaries
- Work is ongoing to apply these new screening criteria to multinary systems

Thank you for your attention!

Binary iodides w/antisites