# Defects in emerging inorganic semiconductors for solar cells

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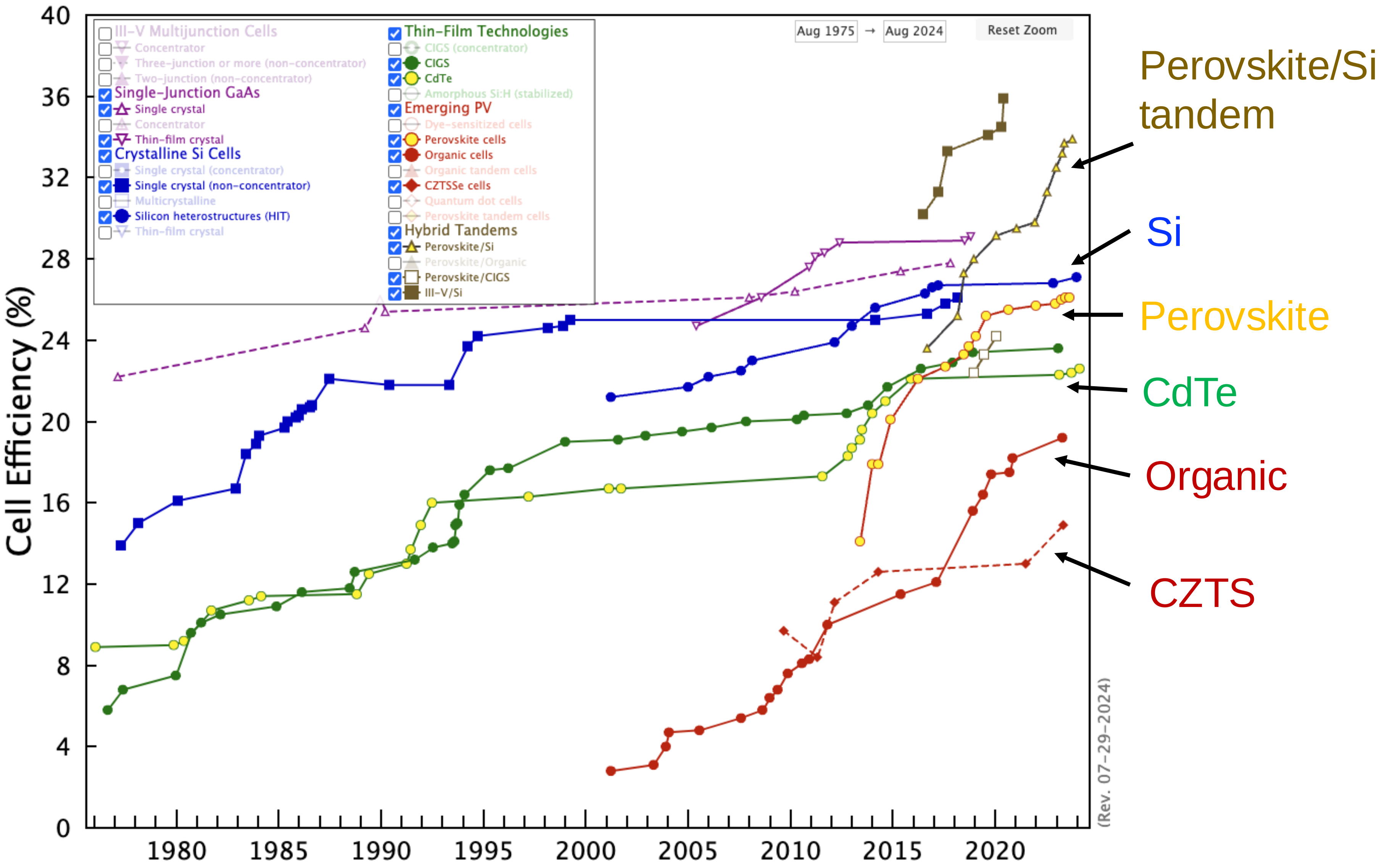
Dartmouth College

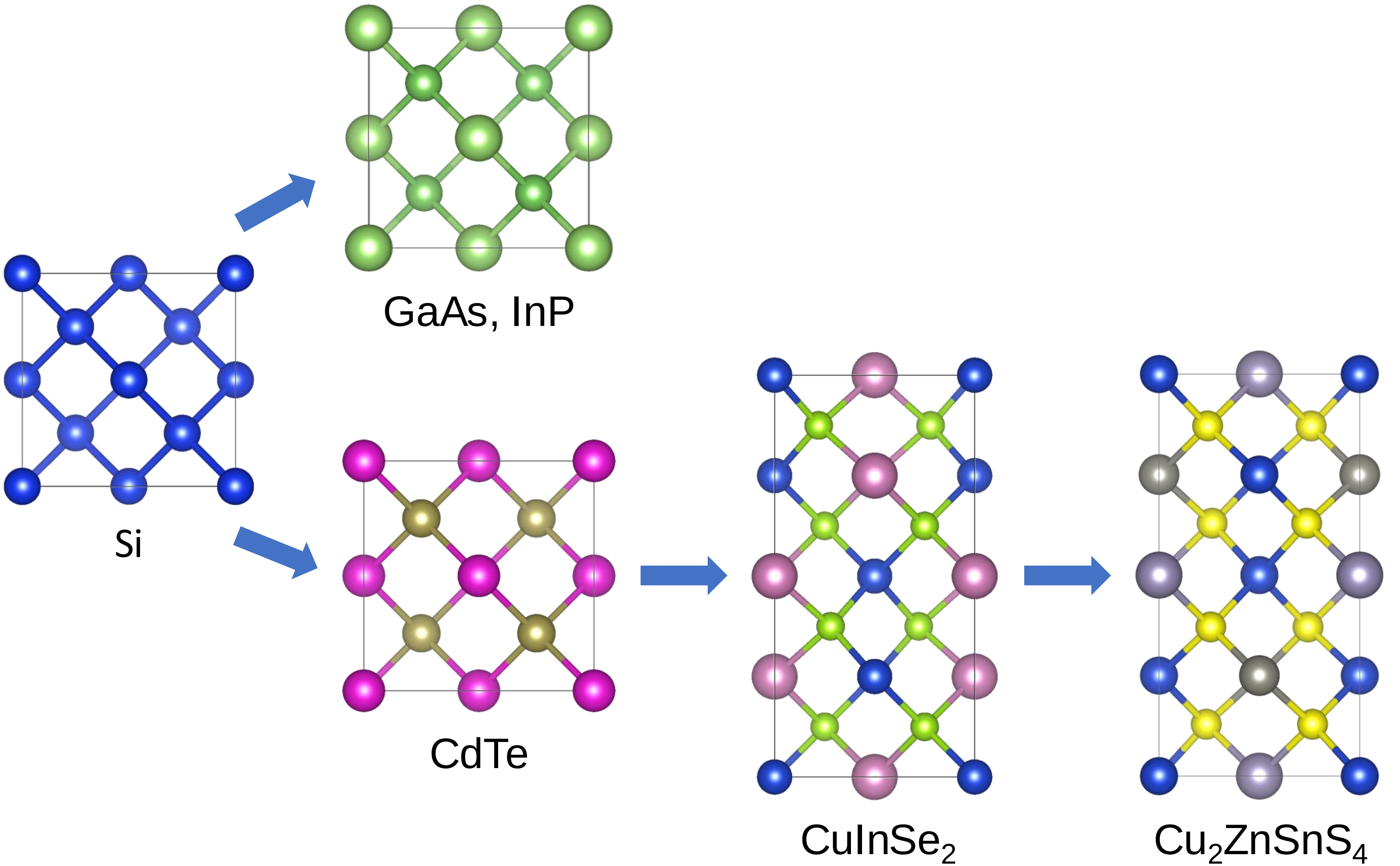
Defects in Semiconductors, 2024 Gordon Research Seminar

# PV technologies at various maturity levels

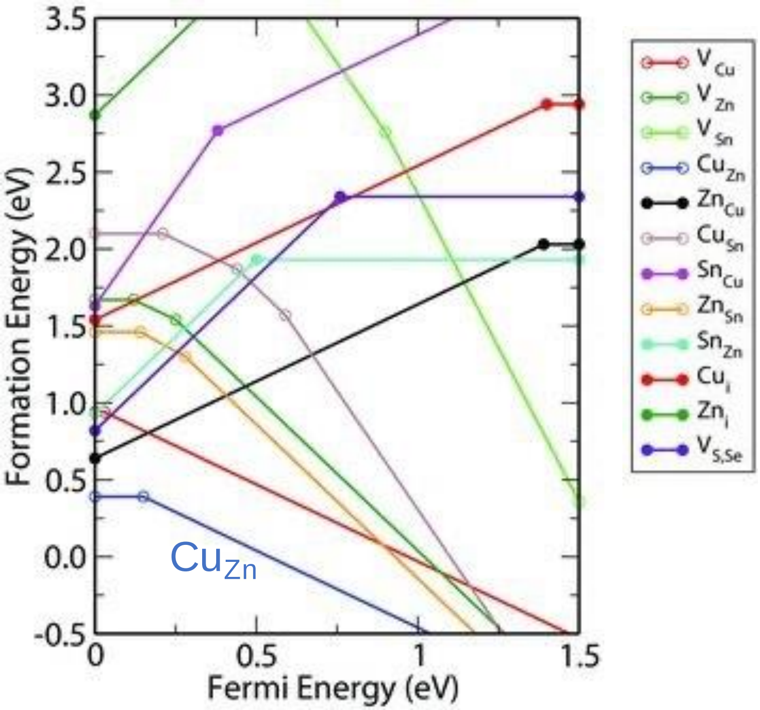


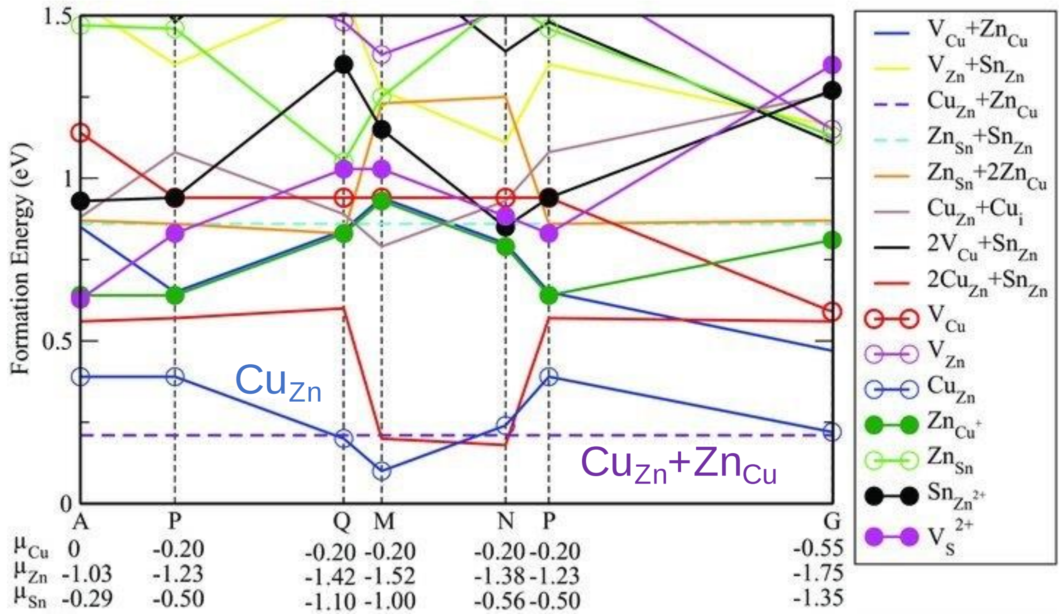

Source: NREL

# Earth-abundant material $Cu_2ZnSnS_4$ (CZTS)



# CZTS has serious defect issues

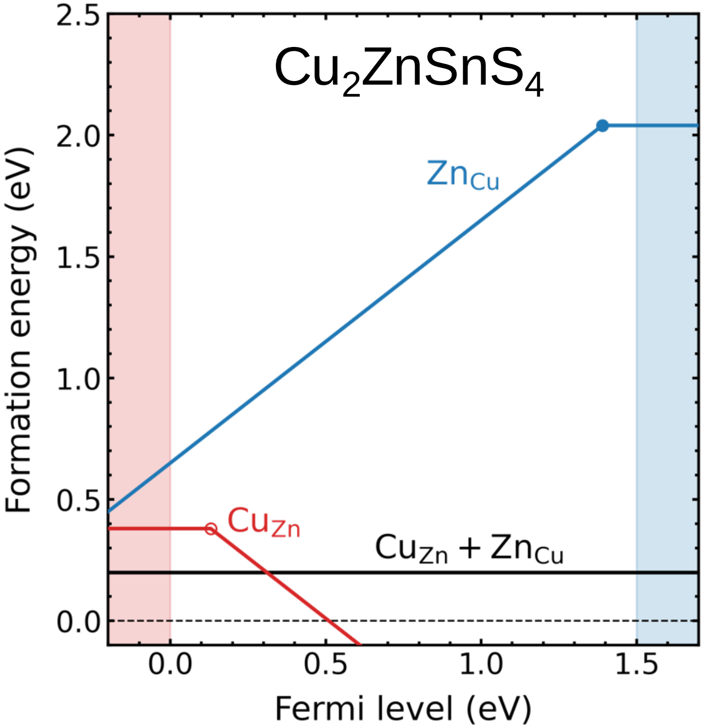


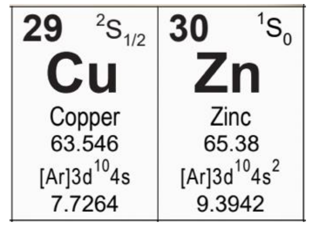

- Many different defects can form
- Cu-Zn antisites ($Cu_{Zn}$ acceptors and $Cu_{Zn}+Zn_{Cu}$ complexes) are easy to form
  - uncontrolled *p*-type doping and disorder

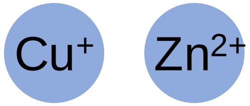
S. Chen, A. Walsh, X. G. Gong, S. H. Wei, *Adv. Mater.* **25**, 1522 (2013)

# How to control Cu-Zn antisites in CZTS?




ionic size

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S. Chen, A. Walsh, X. G. Gong, S. H. Wei, *Adv. Mater*. **25**, 1522 (2013)

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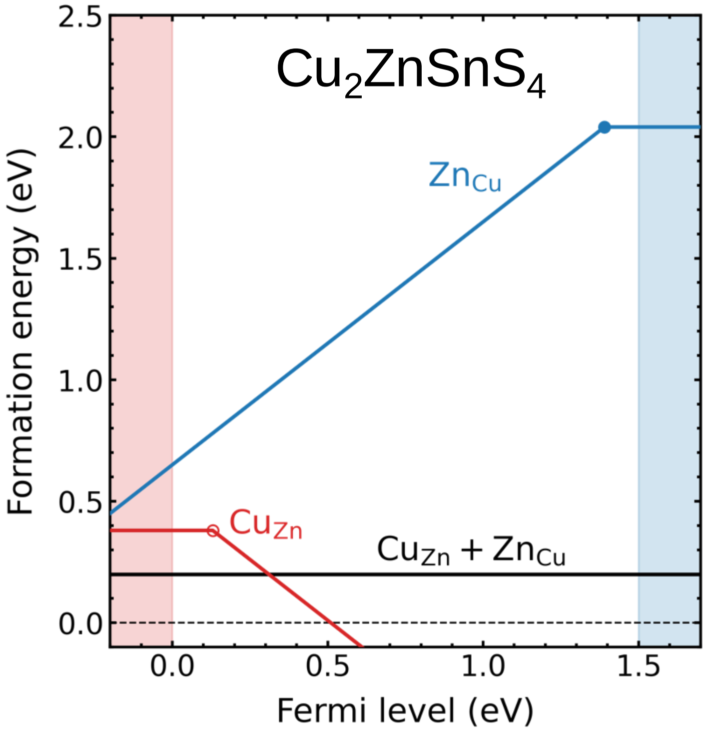
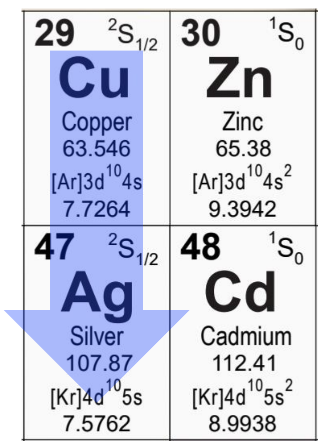
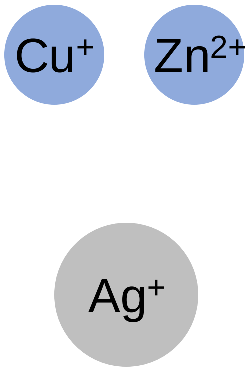



ionic size




$Ag_2ZnSnS_4$ (AZTS)

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*Adv. Mater*. **25**, 1522 (2013)

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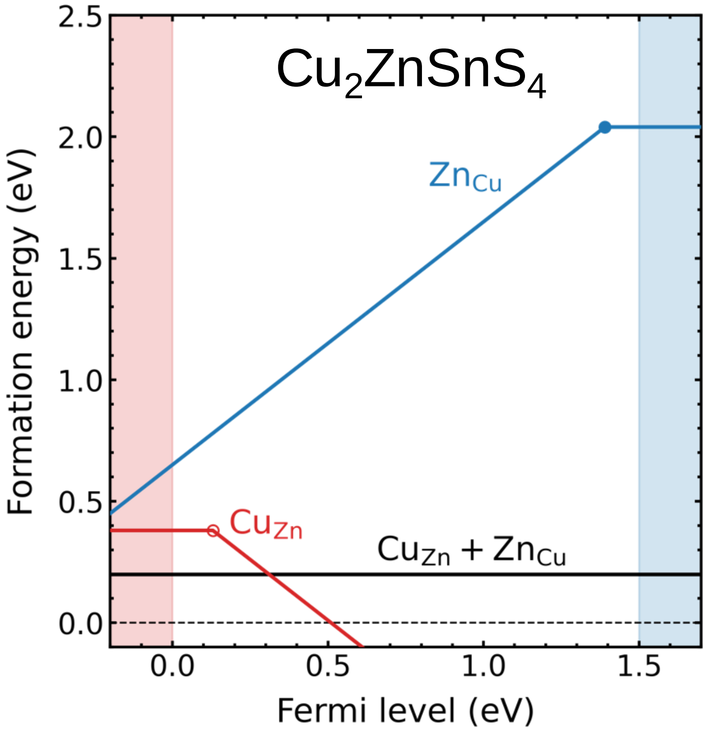


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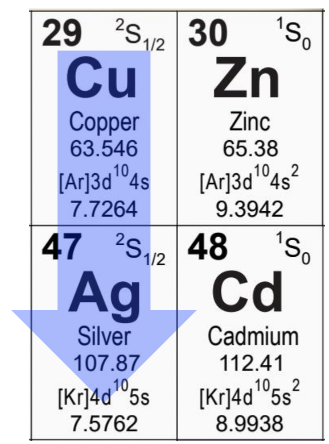
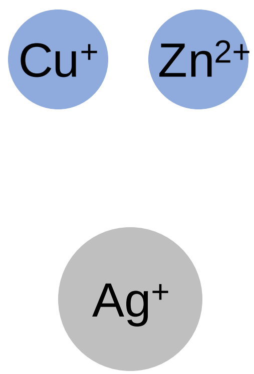
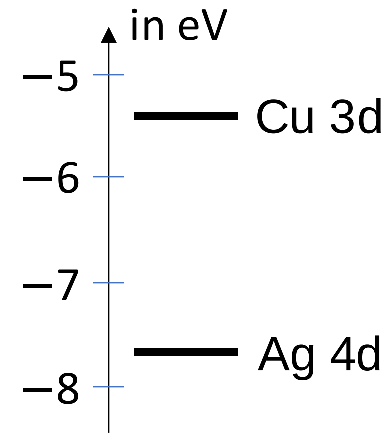



$Ag_2ZnSnS_4$ (AZTS)

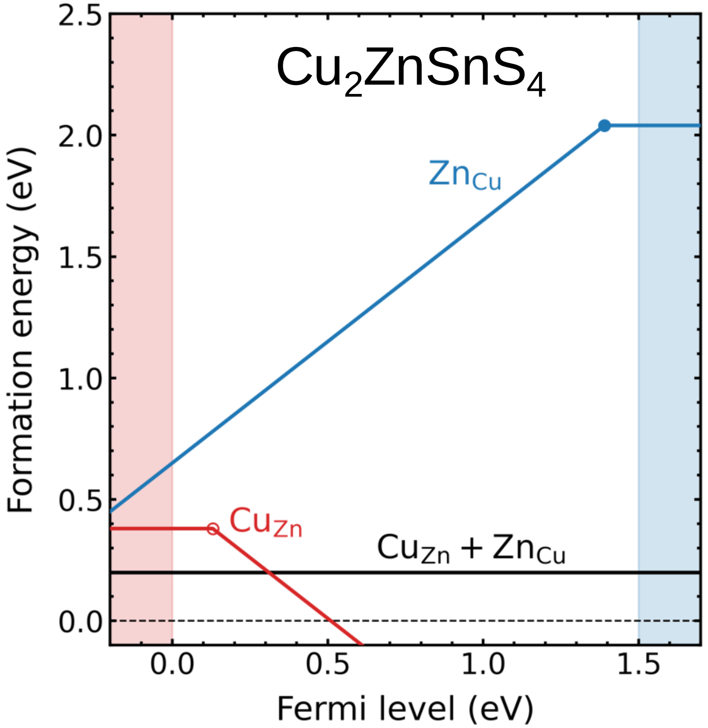
ionic size

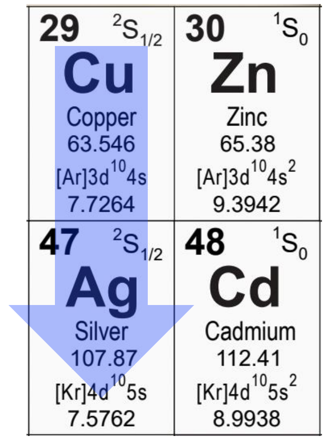



*d* valence orbital



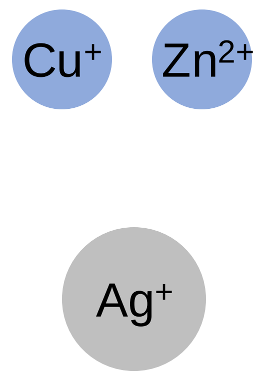
# How to control Cu-Zn antisites in CZTS?



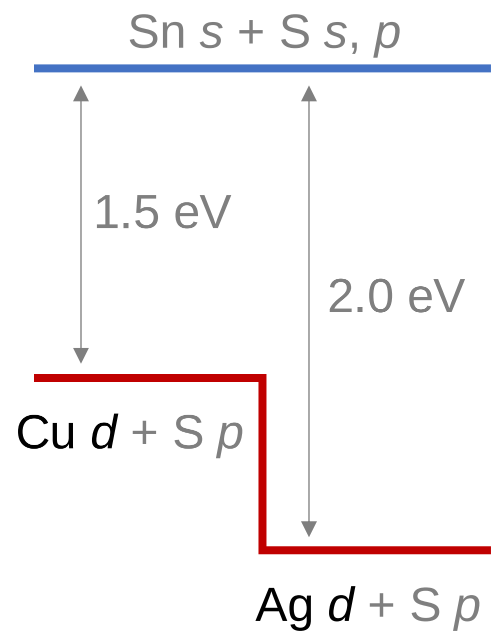




$Ag_2ZnSnS_4$ (AZTS)

ionic size




CZTS AZTS

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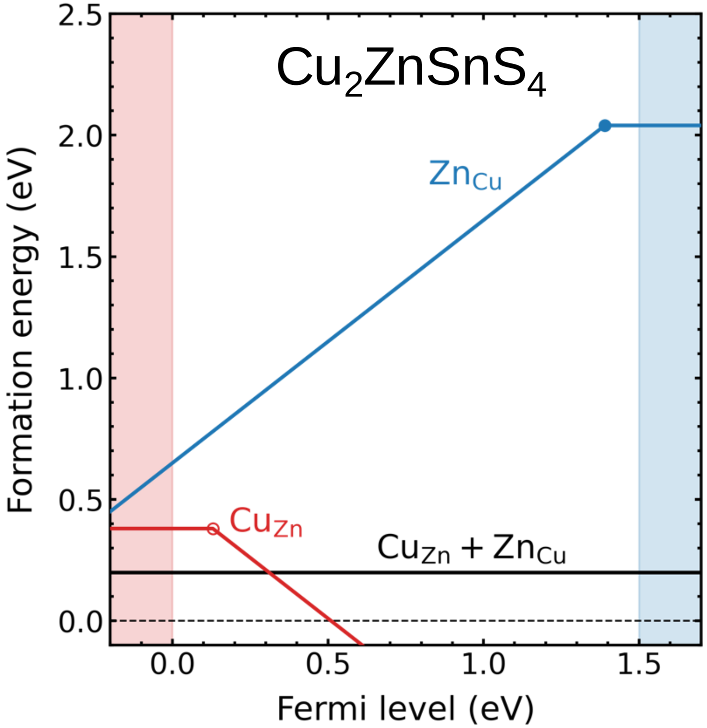
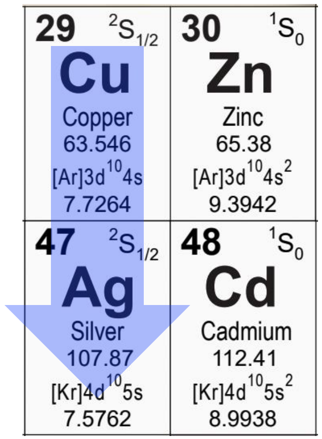
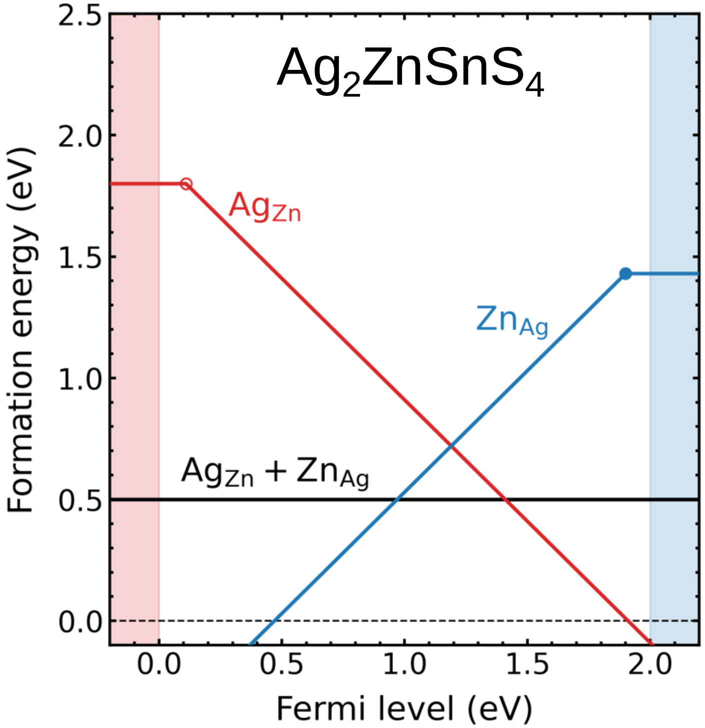
S. Chen, A. Walsh, X. G. Gong, S. H. Wei, *Adv. Mater.* **25**, 1522 (2013)

# Defect *dis*similarity between CZTS and AZTS

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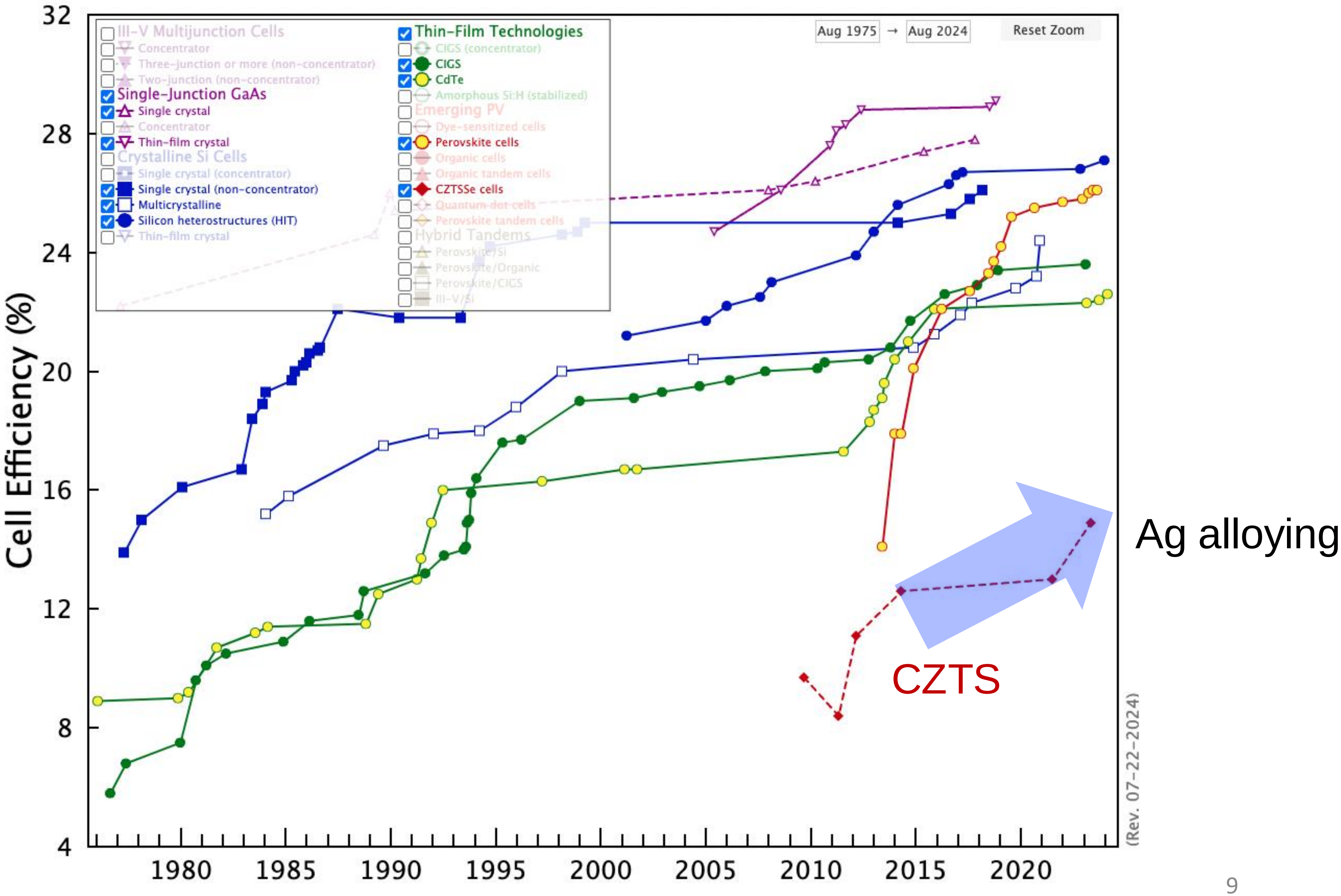
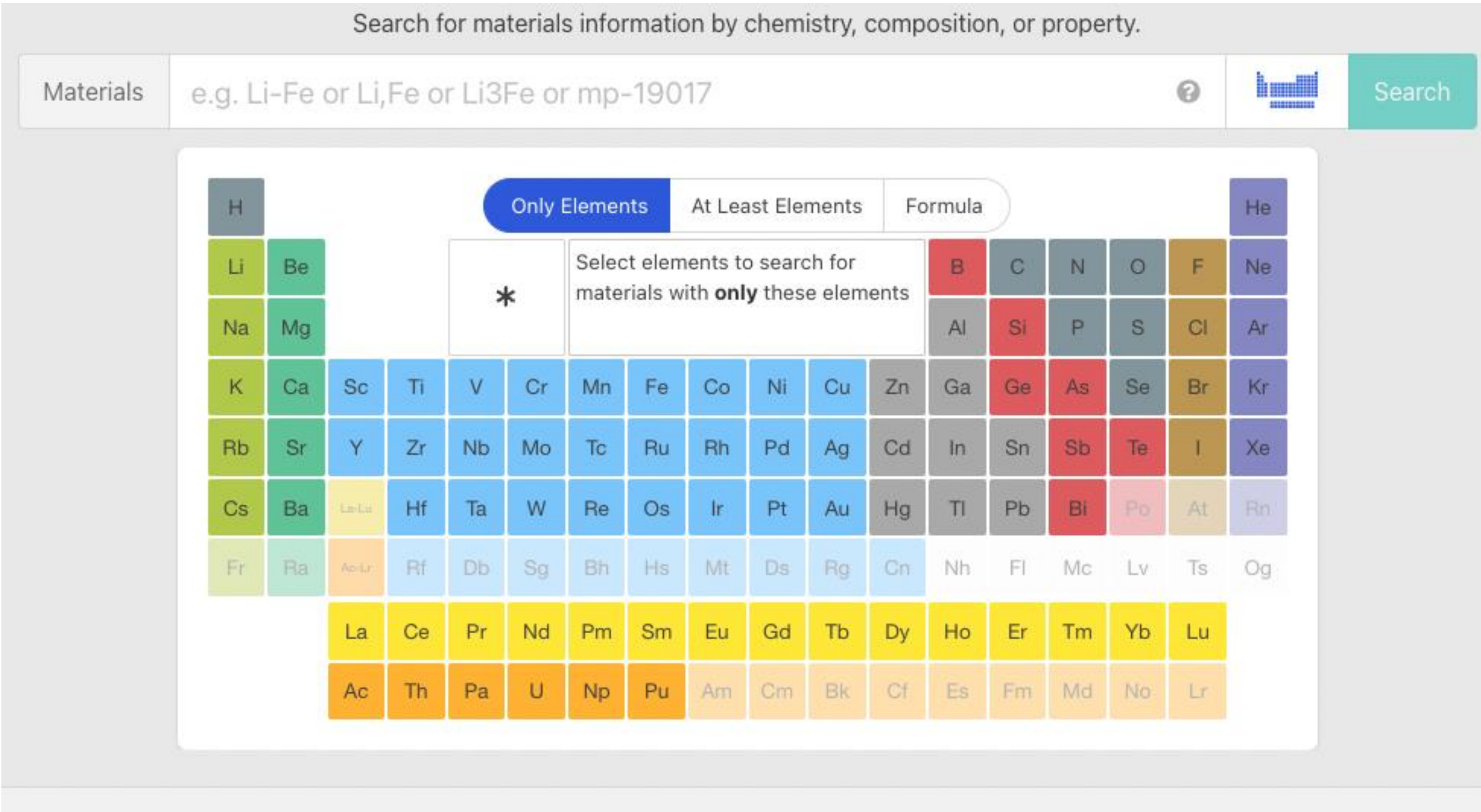
**Z.-K. Yuan**, S. Chen, H. Xiang, X.-G. Gong, A. Walsh, J.-S. Park, I. Repins, S.-H. Wei, *Adv. Funct. Mater.* **25**, 6733 (2015)

# Ag alloying makes CZTS rise again



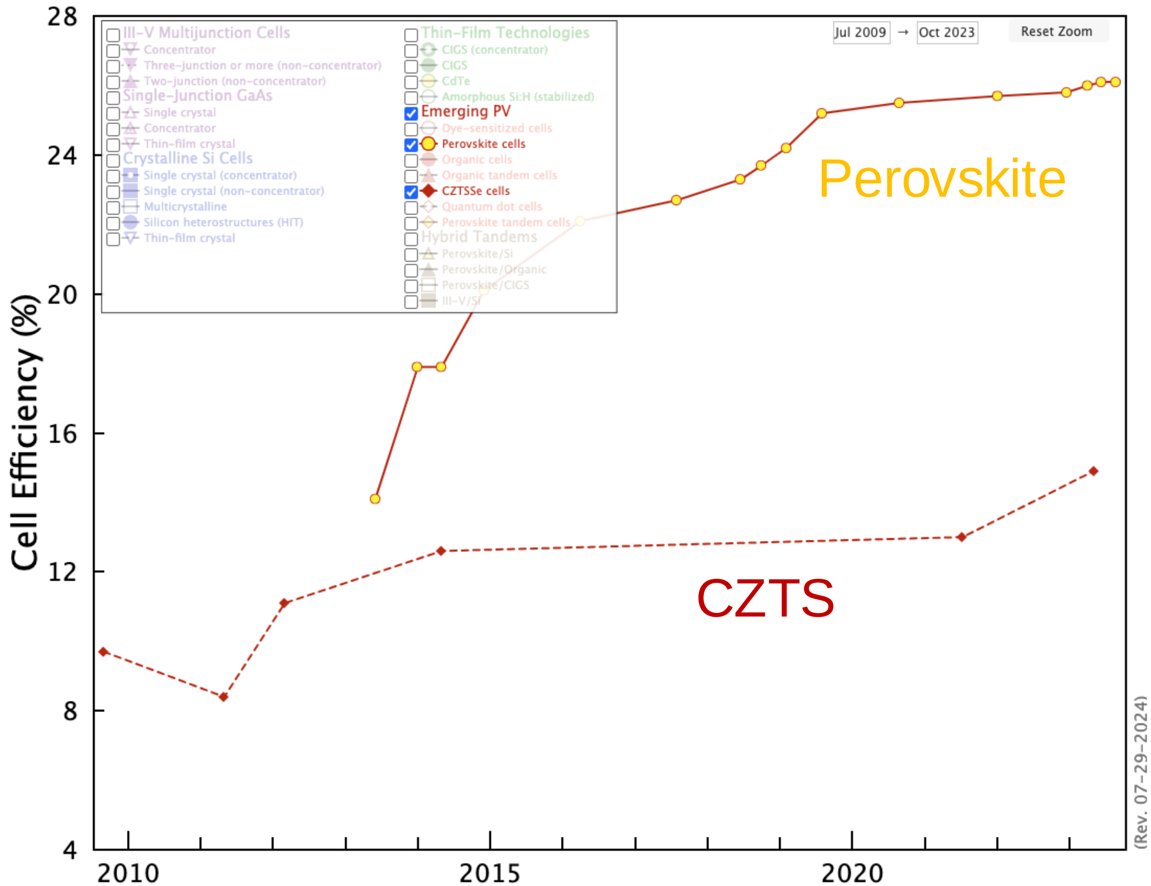

Source: NREL

# Identify new solar absorbers from inorganic materials database



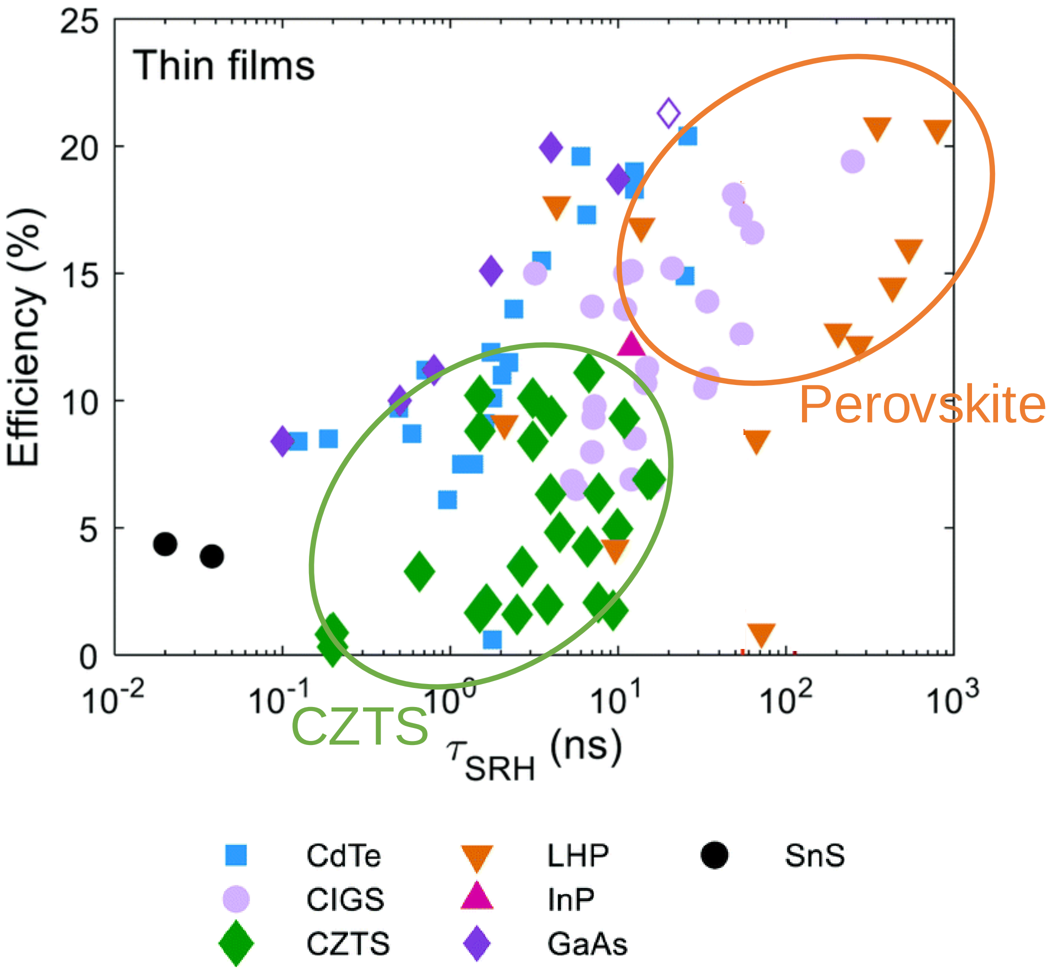

All **153,235 materials**

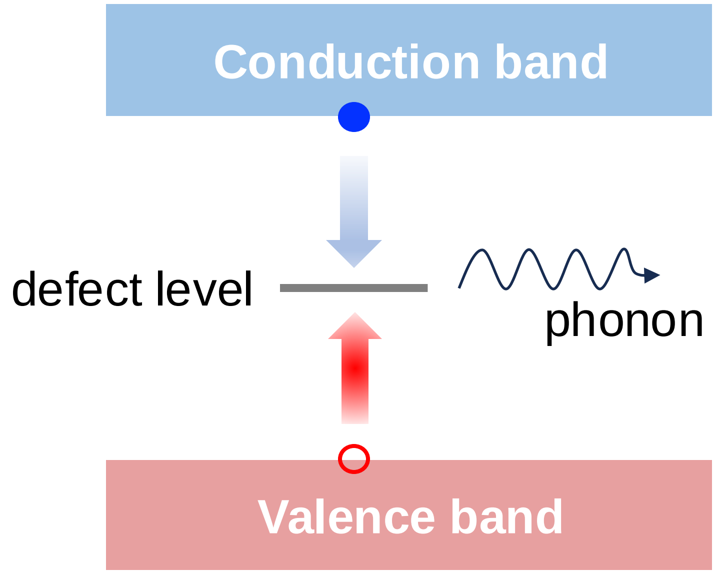
# Search for materials as exceptional as perovskites




Source: NREL

# “Defect tolerance” in perovskites

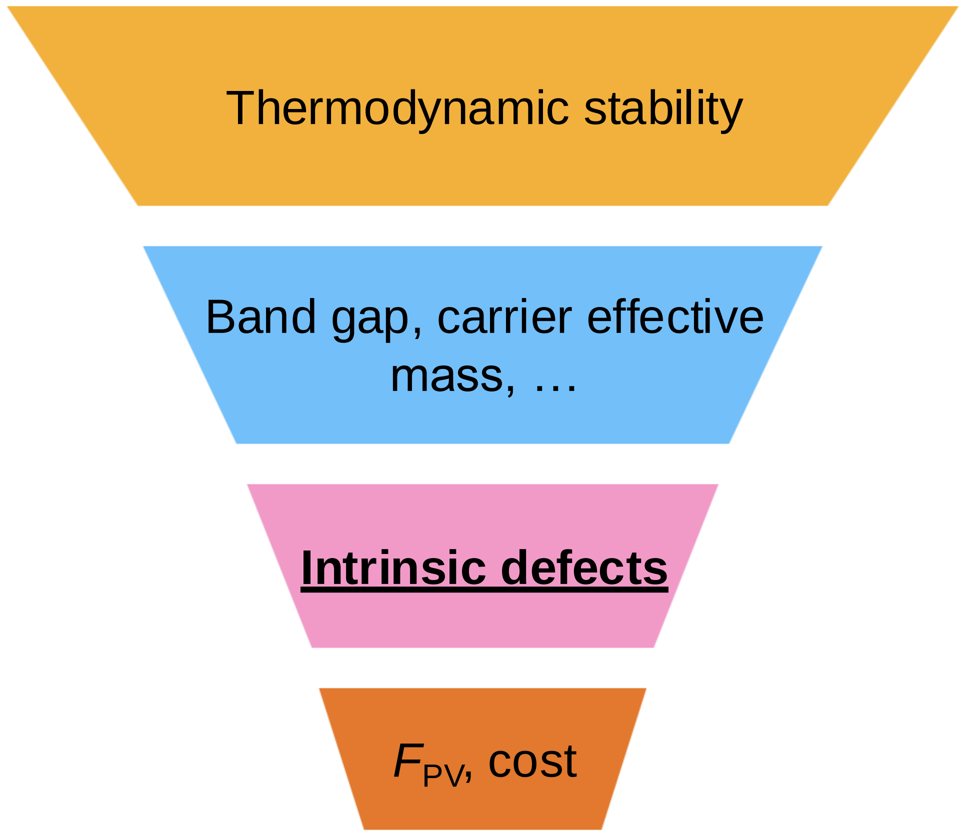



Shockley-Read-Hall (SRH) recombination

K. Ye *et al.*, *Faraday Discuss.*, **239**, 146 (2022)

# Our approach — High-throughput computational screening



# High-throughput defect workflow

**Input** → **DFT calculations** MongoDB document → **Defect data** MongoDB document

“mp-8279”


```
_id: ObjectId('65f5aa34d13c239302d8d97a')
▸ links : Object
▸ parent_links : Object
▸ nodes : Array (113)
▸ metadata : Object
  state : "COMPLETED"
  name : "mp-8279_Ba(CdP)2_defects_GGA"
  created_on : 2024-03-16T14:18:27.981+00:00
  updated_on : 2024-03-17T11:03:47.252+00:00
▸ fw_states : Object
```



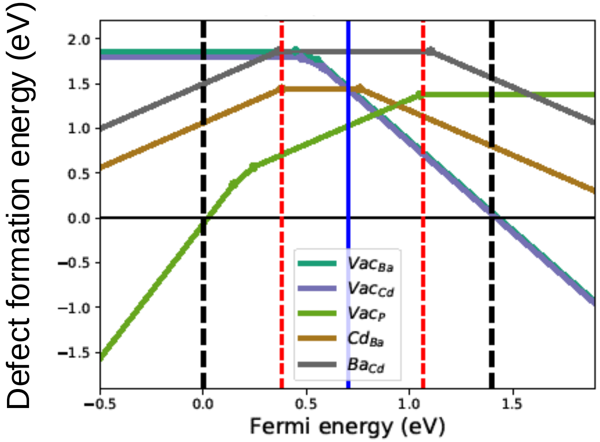
```
_id: ObjectId('66636dfd28308bc46a4e932f')
uuid : "93ecc23d-7747-4ff2-98ad-5ace1e7a0c74"
index : 1
name : "mp-8279_Ba(CdP)2_defects_data"
▾ parsed_data : Object
  ▸ conventional_unitcell : Object
  ▸ bulk_supercell : Object
  ▸ dropbox_links : Object
  ▸ band_edges : Object
  ▸ dielectric_tensor : Object
  ▸ density_of_states : Object
  ▸ delta_Qs : Object
  ▸ chemical_potentials : Object
  ▸ formation_energies : Object
  ▸ defect_thermo : Object
  ▸ transition_levels_PBE : Object
  ▸ carrier_lifetime : Object
▸ units_log : Object
created_on : "2024-06-07 16:30:53"
```


**HT software infrastructure**:
PyCDT, Atomate, Fireworks,
Pymatgen, py-sc-fermi,…

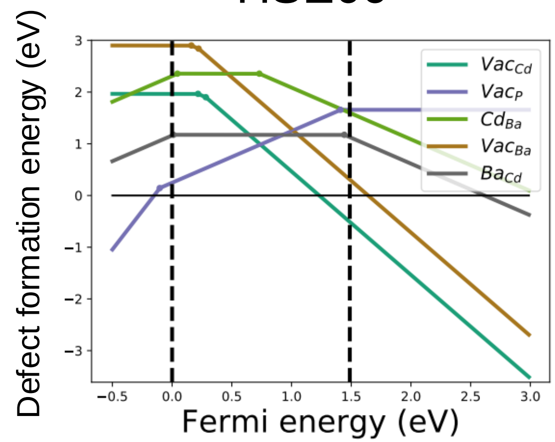
# Screen materials on intrinsic defects

✓

DFT

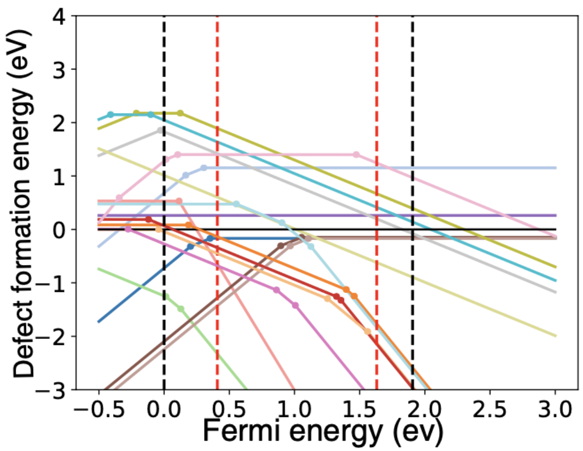



HSE06

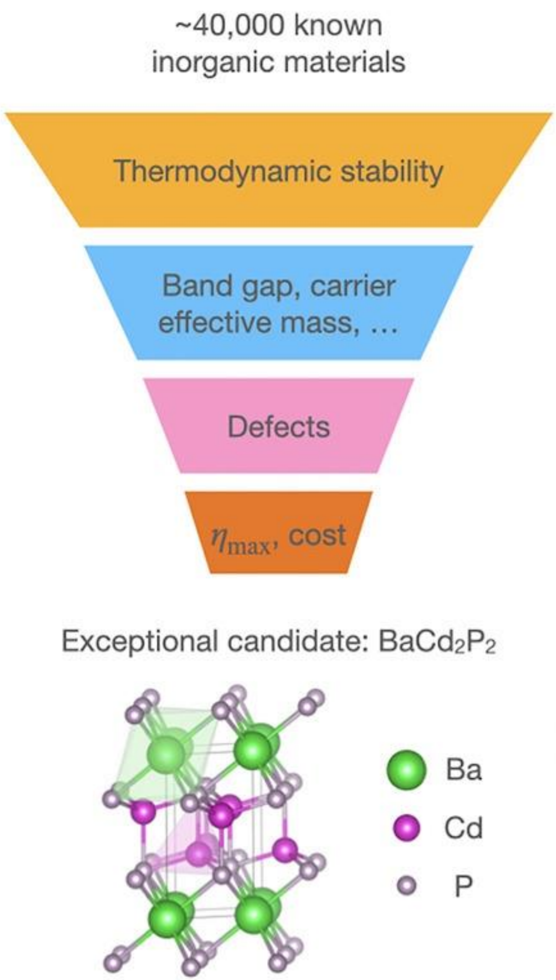


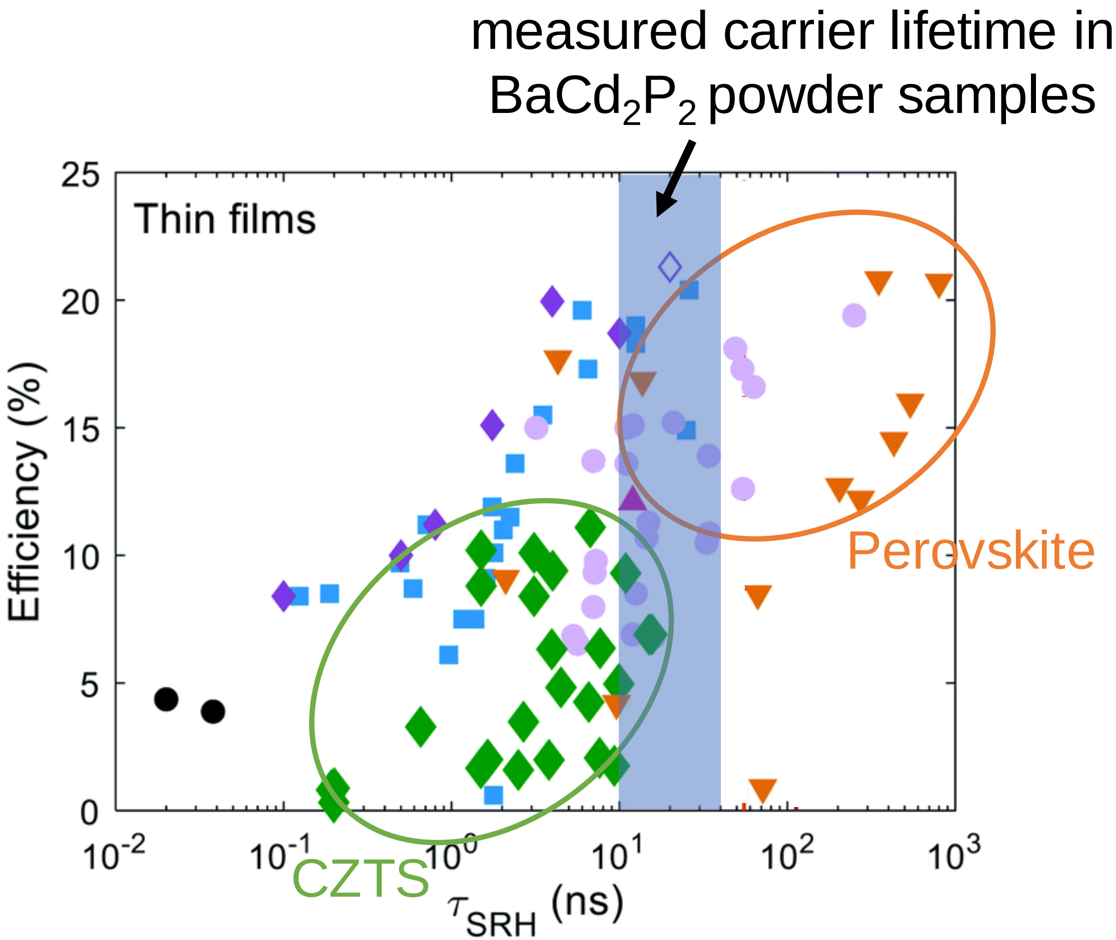

✗

DFT



# $BaCd_2P_2$ identified as a long carrier lifetime solar absorber




**Z. Yuan**, D. Dahliah, M. R. Hasan, G. Kassa, A. Pike, S. Quadir, R. Claes, C. Chandler, Y. Xiong, V. Kyveryga, P. Yox, G.-M. Rignanese, I. Dabo, A. Zakutayev, D. P. Fenning, O. G. Reid, S. Bauers, J. Liu, K. Kovnir, and G. Hautier, *Joule* **8**, 1412 (2024)

# Conclusions

- Chemical intuitive guided defect control to optimize existing solar absorbers
- Identify new exceptional solar absorbers, through high-throughput defect computations


# Acknowledgments

U.S. DEPARTMENT OF ENERGY

NERSC