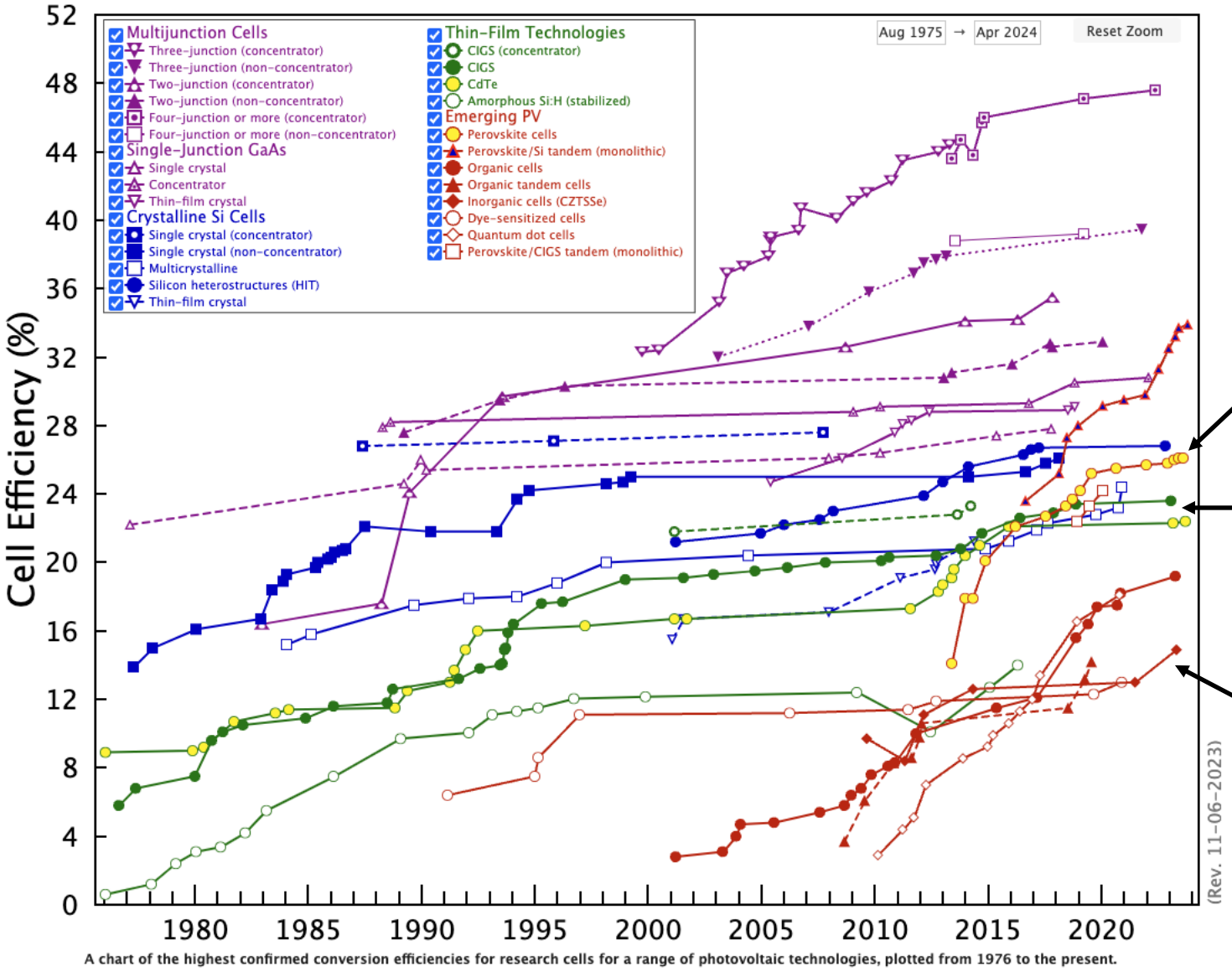
High-throughput computational search for new solar absorbers considering “defect tolerance”

Zhenkun Yuan, Diana Dahliah, Muhammad Rubaiat Hasan, Gideon Kassa, Andrew Pike, Shaham Quadir, Romain Claes, Cierra Chandler, Yihuang Xiong, Victoria Kyveryga, Philip Yox, Gian-Marco Rignanese, Ismaila Dabo, Andriy Zakutayev, David P. Fenning, Obadiah G. Reid, Sage Bauers, Jifeng Liu, Kirill Kovnir, and Geoffroy Hautier

2023 MRS Fall Meeting, Boston

Current solar cell technologies



A chart of the highest confirmed conversion efficiencies for research cells for a range of photovoltaic technologies, plotted from 1976 to the present.

Halide perovskites **stability**

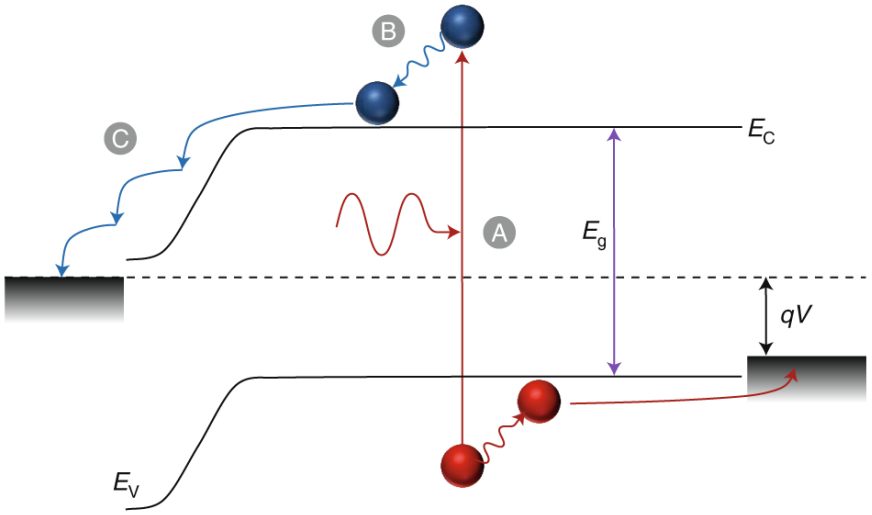
CdTe & CIGS **abundance**

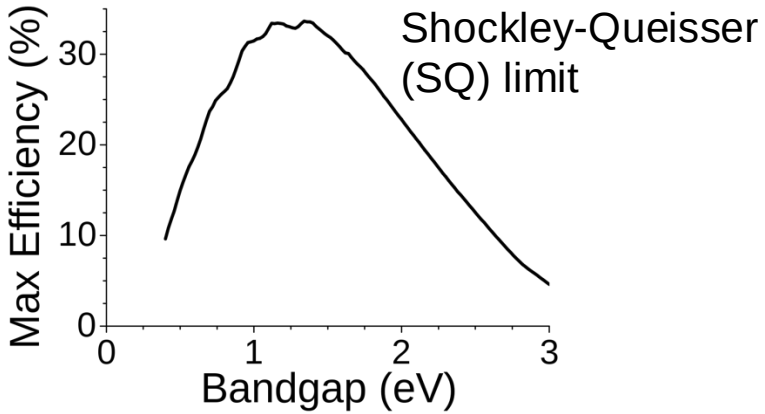
CZTS **efficiency**

Source: NREL

What makes a good solar absorber?

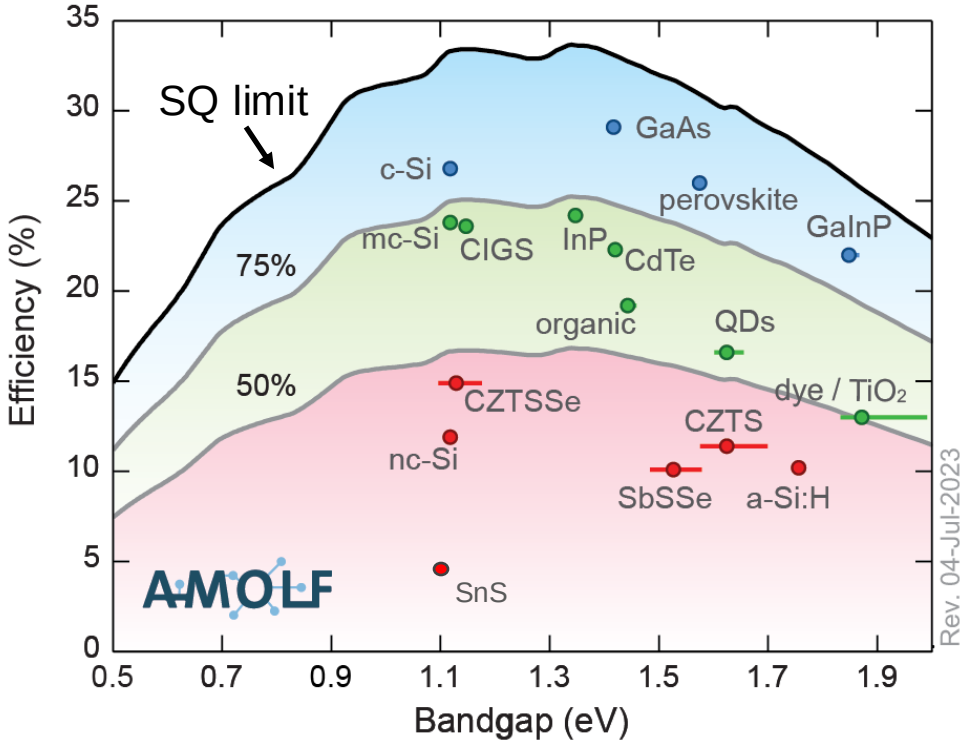
- Light absorption
  - large absorption coefficients
  - suitable band gap
- Carrier transport
  - high mobilities
  - long carrier lifetime
- Good stability in operating conditions

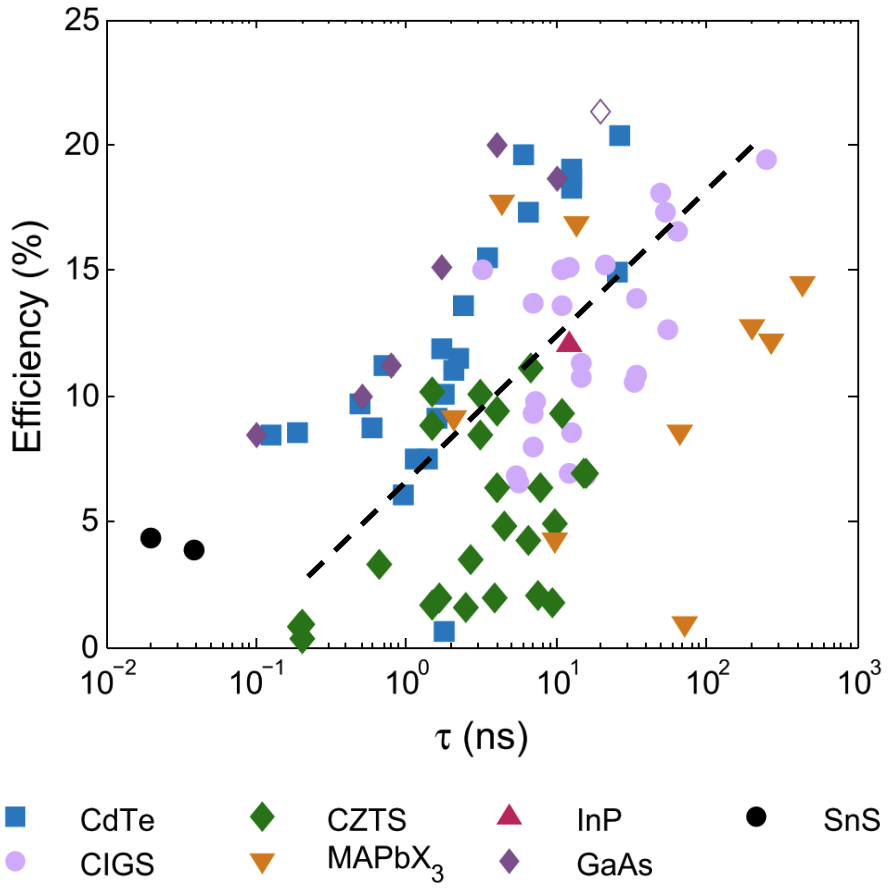




J.-F. Guillemoles et al., *Nat. Photonics* **13**, 501 (2019)

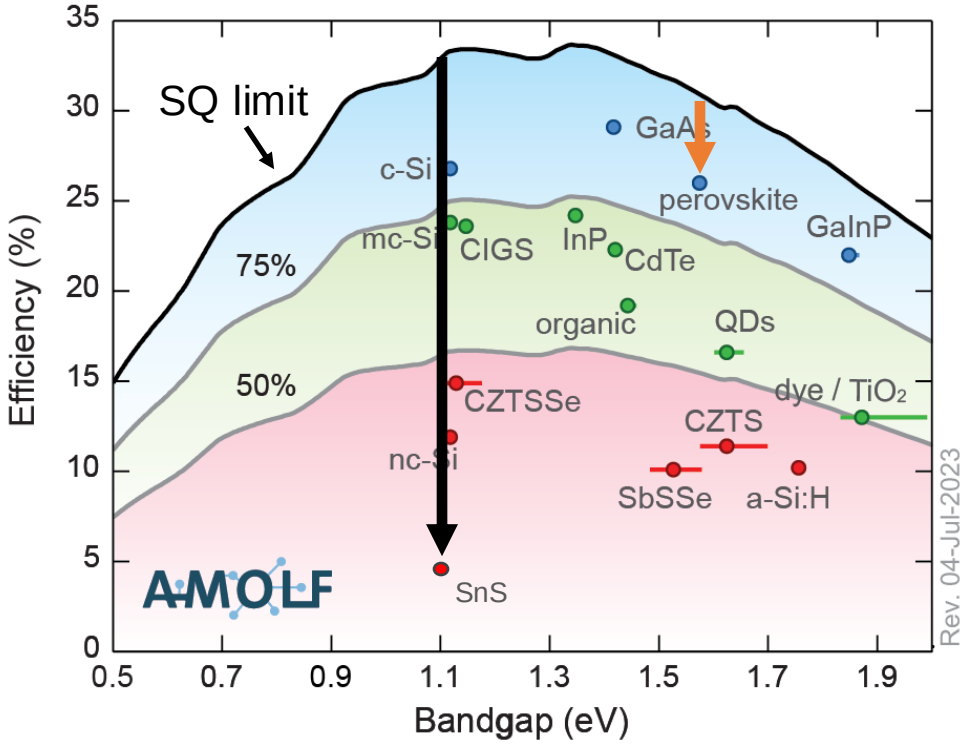
The importance of long carrier lifetime

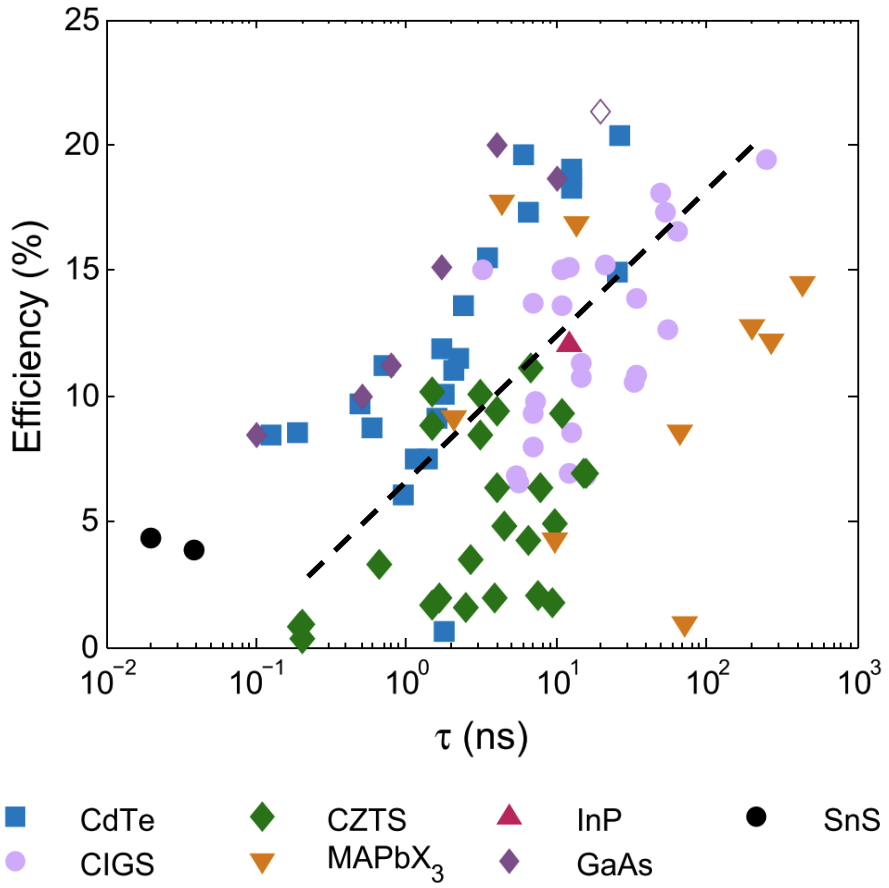




R. Jaramillo *et al.*, *J. Appl. Phys.* **119,** 035101 (2016)

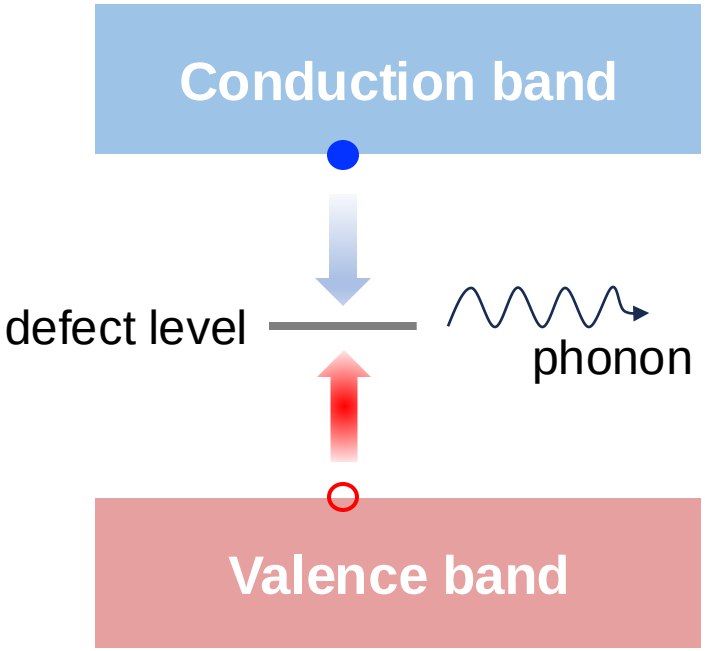
The importance of long carrier lifetime





R. Jaramillo *et al.*, *J. Appl. Phys.* **119,** 035101 (2016)

Defect-assisted nonradiative recombination



Nonradiative lifetime

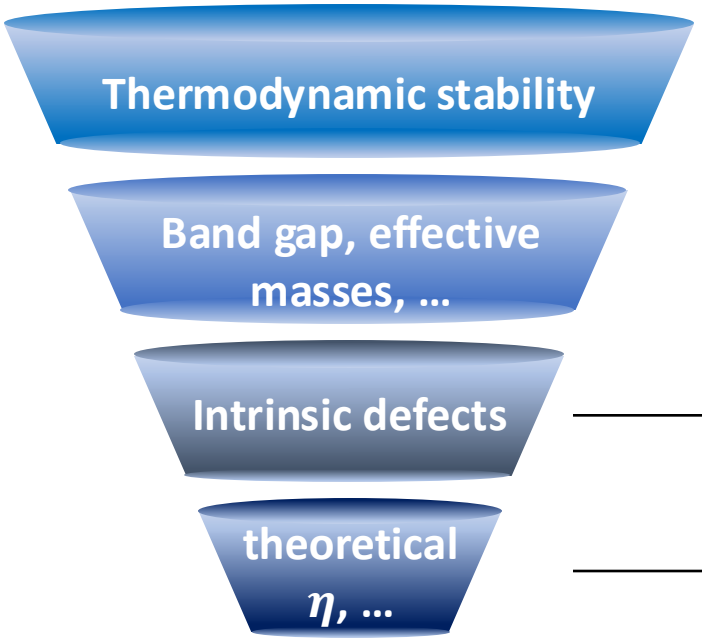
$$\tau = \frac{1}{N_{\mathrm{d}} * C}$$

deep-defect density

carrier capture coefficient

S. Kim *et al.*, *Energy Environ. Sci.*, **14**, 5057 (2021)

Going to high-throughput (HT) search



- vacancies, cation antisites
- τ, assuming constant C
- PBE; HSE (for those with $\tau > 1$ ns)

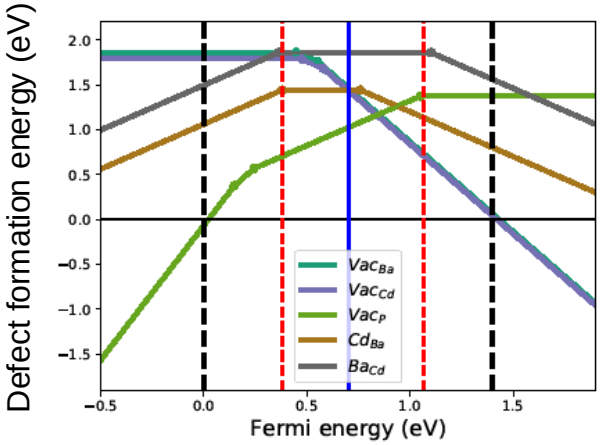
- solar cell efficiency (η) from device modeling using τ
- stability (empirical rules, synthesis report, …)
- materials cost

HT Infrastructure:
PyCDT, Atomate, Fireworks, Materials Project, …

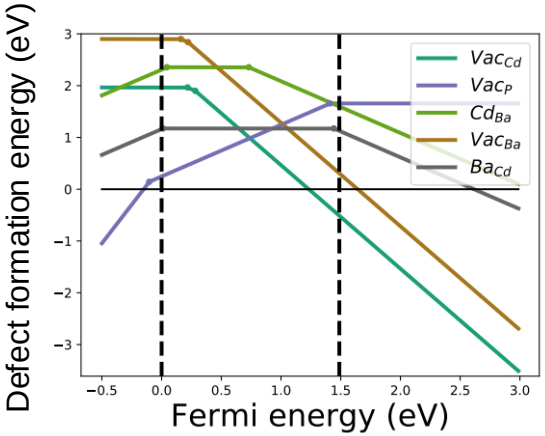
D. Dahliah *et al.*, *Energy Environ. Sci.*, **14**, 5057 (2021)

Screening materials on their intrinsic defects

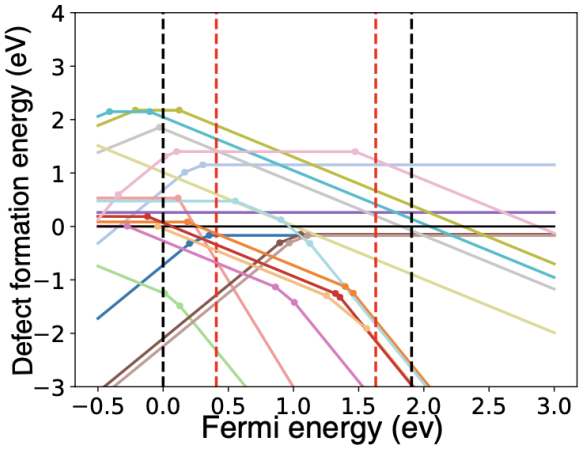
DFT-PBE



HSE verify

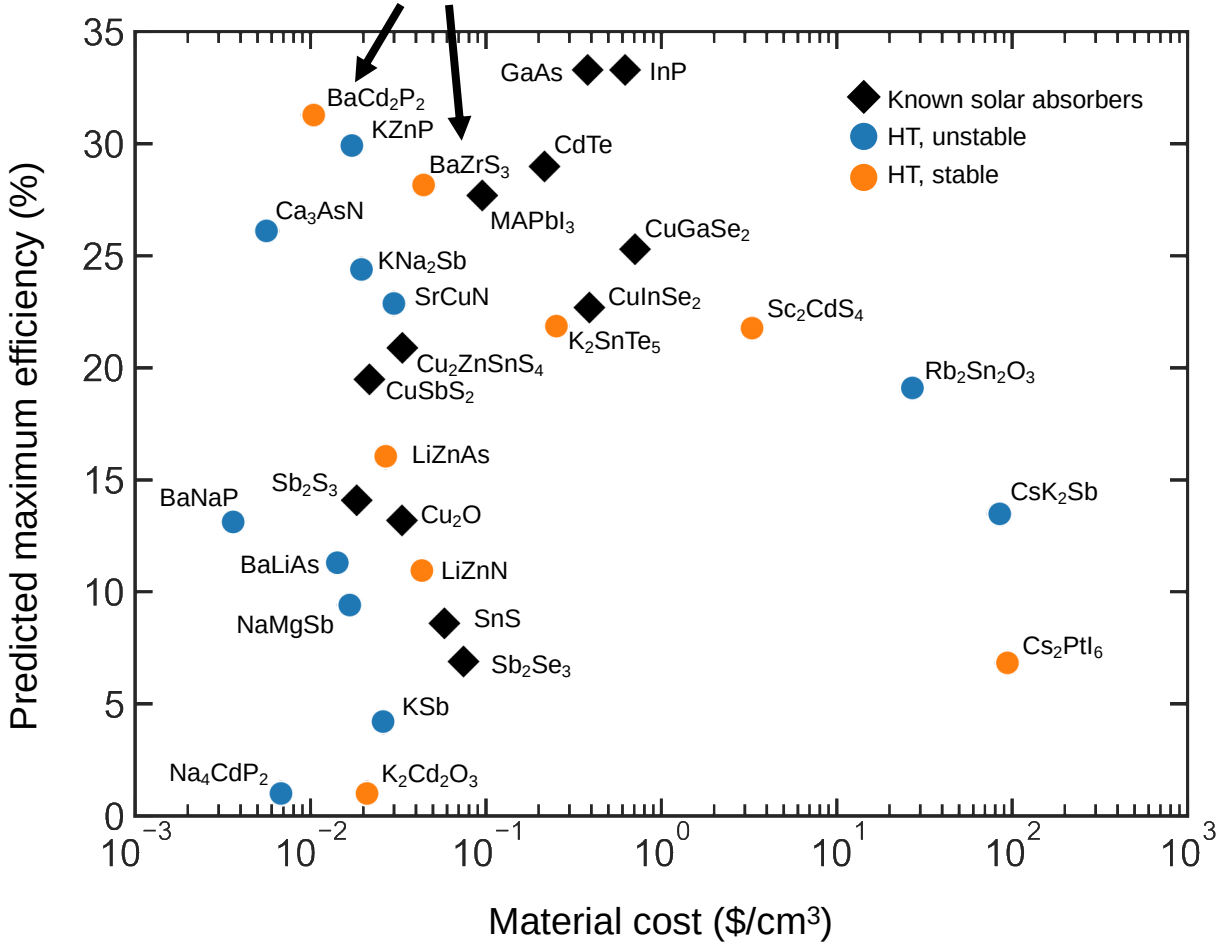


DFT-PBE



Best candidates among ~40,000 inorganic materials

High-efficiency, stable, and low-cost candidates



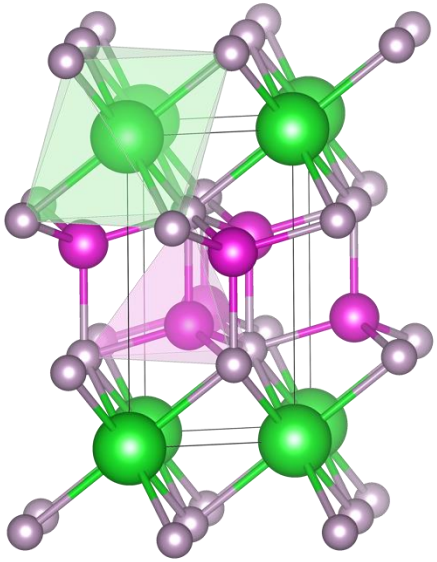
$BaCd_2P_2$

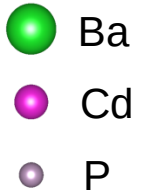
P. Klüfers, 1980

$BaZrS_3$

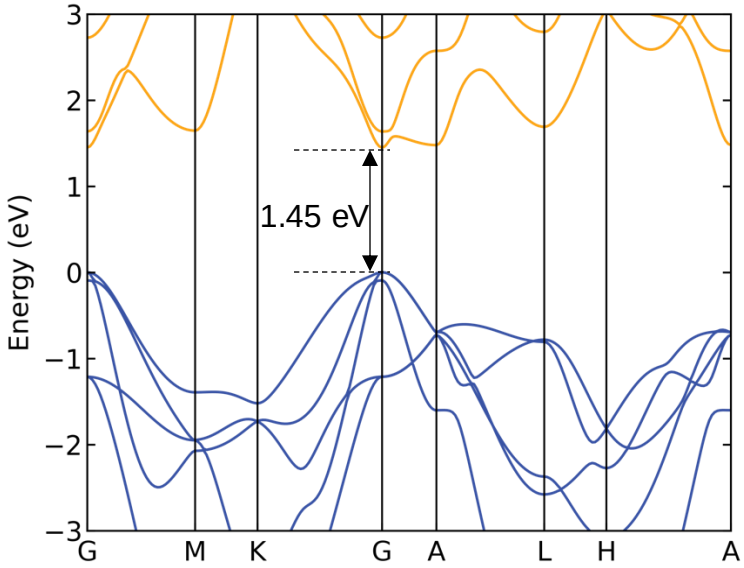
Y. Sun *et al.*, *Nano Lett.* **15**, 581 (2015)

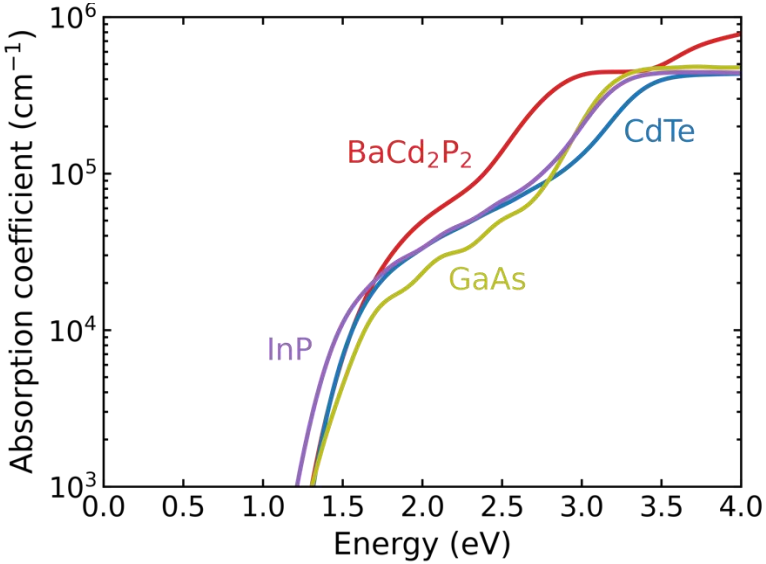
$BaCd_2P_2$ as a new solar absorber



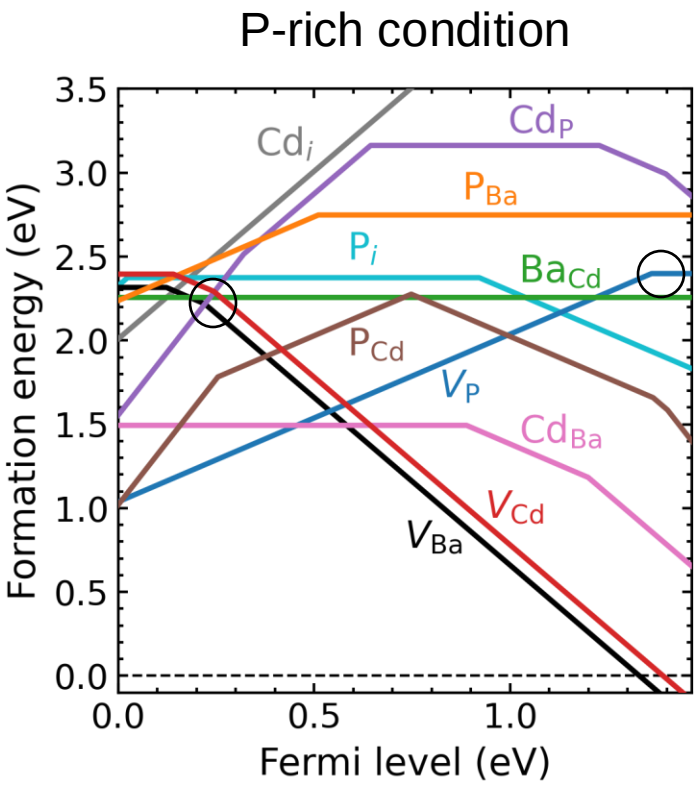


Zintl structure





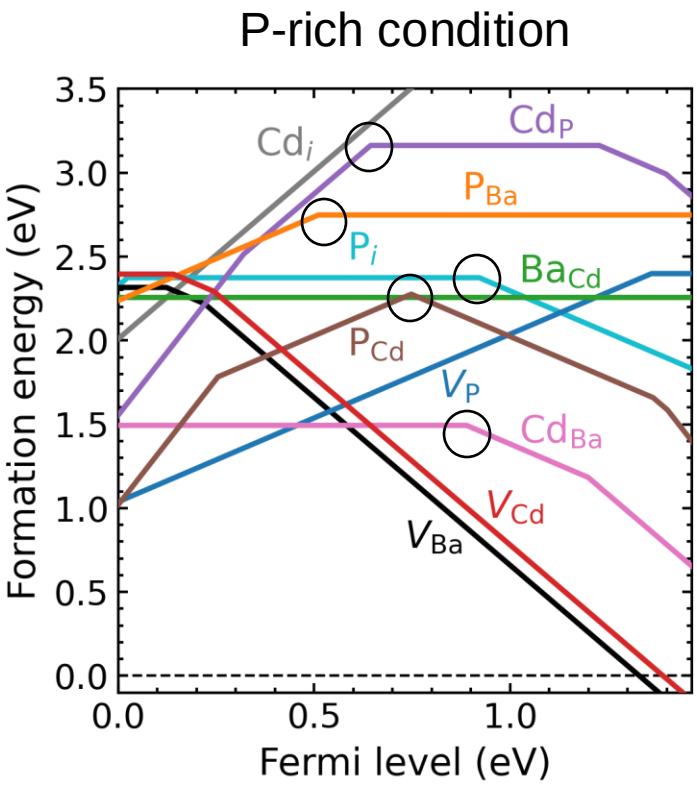
Intrinsic defects in $BaCd_2P_2$



- Vacancies (V_{Ba}, V_{Cd}, and V_P) are major defect types, but they are all shallow
  - V_P is shallow in InP, but very deep in Zn_3P_2

Z. Yuan *et al.*, *J. Mater. Chem. A*, **11**, 20592 (2023)

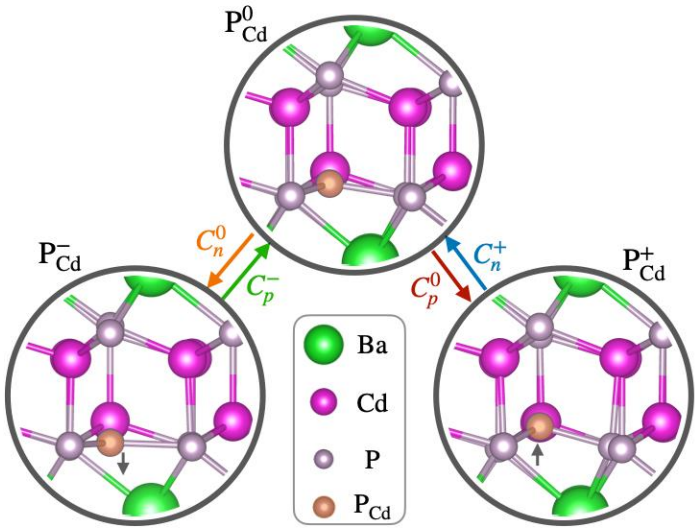
Intrinsic defects in $BaCd_2P_2$



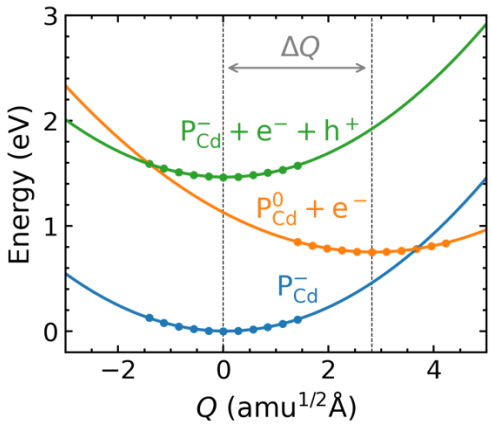
- Vacancies (V_{Ba}, V_{Cd}, and V_{P}) are major defect types, but they are all shallow
  - V_{P} is shallow in InP, but very deep in Zn_3P_2
- Deep-level defects ($\mathrm{Cd_{Ba}}$, $\mathrm{P_{Cd}}$, $\mathrm{P_{Ba}}$, P_i, and $\mathrm{Cd_P}$) are generally high in formation energy

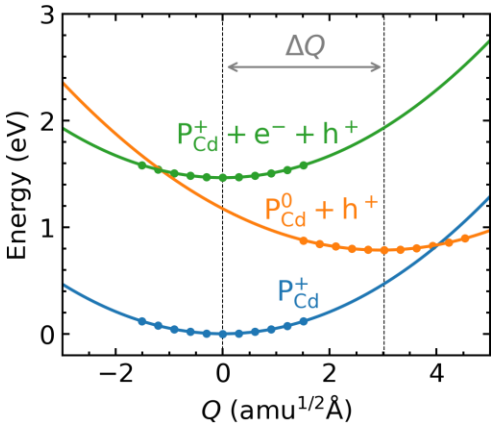
Z. Yuan *et al.*, *J. Mater. Chem. A*, **11**, 20592 (2023)

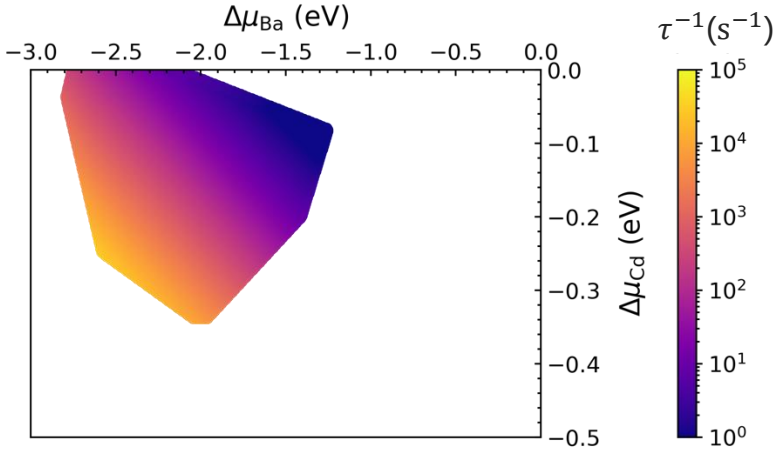
Nonradiative recombination in $BaCd_2P_2$



Small carrier capture barriers







$$\tau^{-1} = N_d * C$$

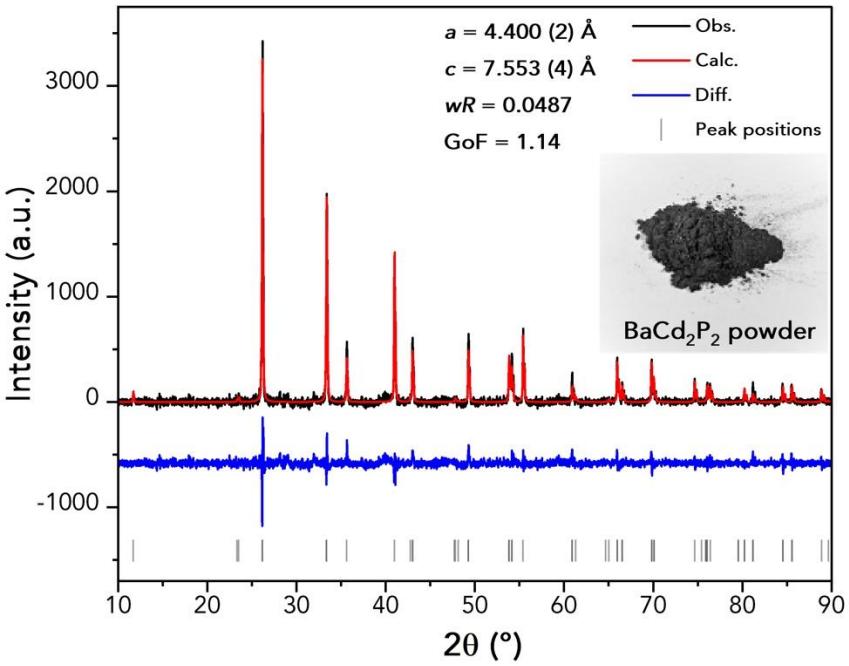
$CH_3NH_3PbI_3$

$\tau^{-1} \sim 10^7\ s^{-1}$

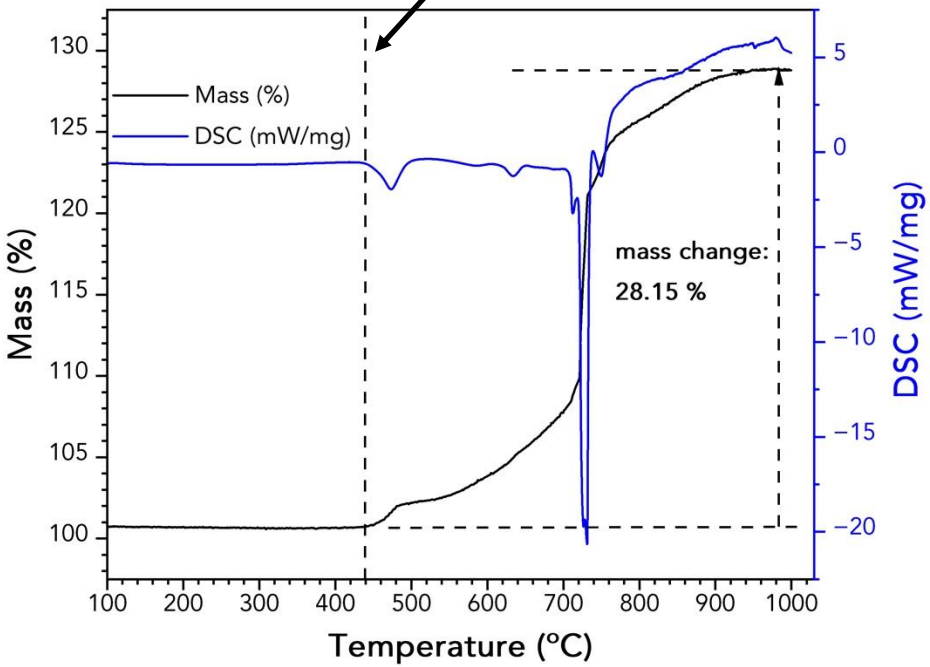
X. Zhang *et al.*,
PRB **101**, 140101(R) (2020)

Synthesis and stability of $BaCd_2P_2$

Direct reaction of elements in sealed ampoule at 1000 ℃ and annealed at 800 ℃

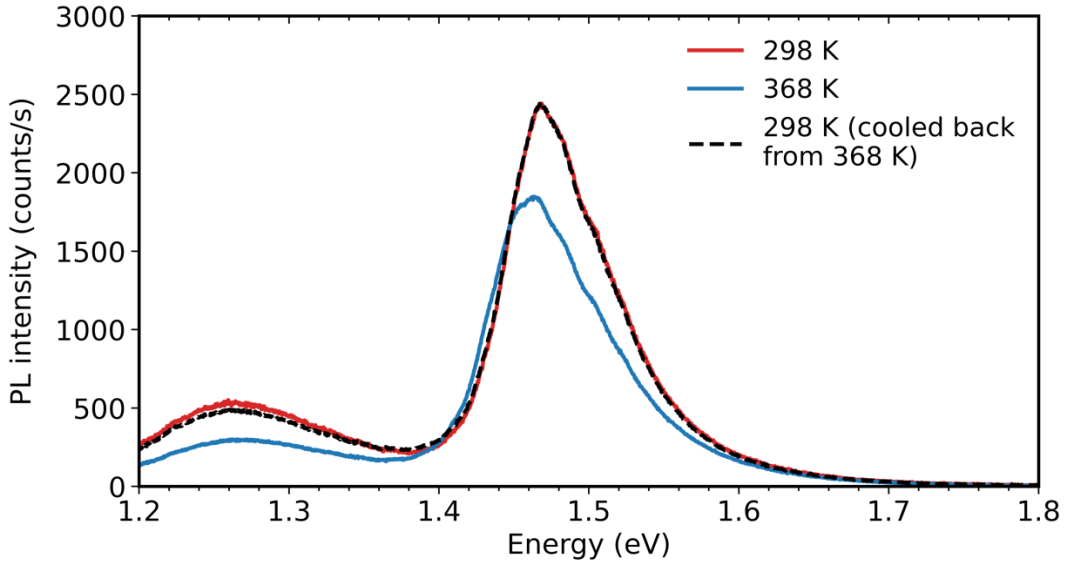


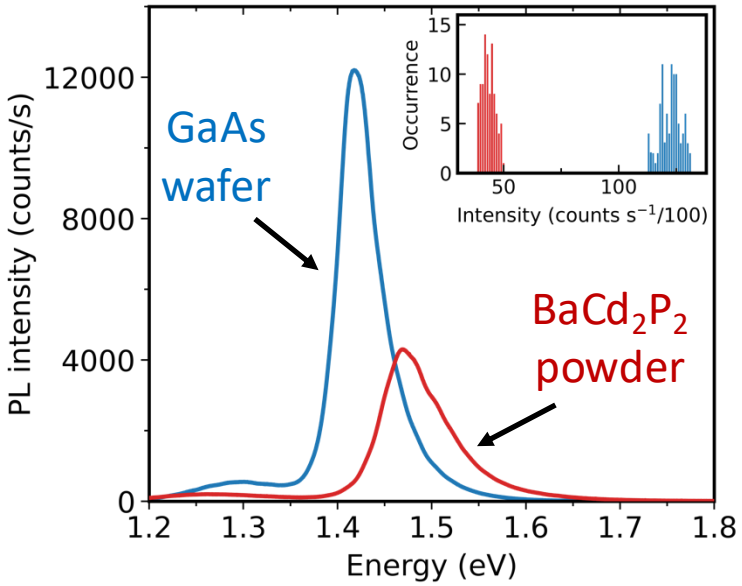
Oxidation starts for $T > 425℃$



Thermal analysis in ambient air

Photoluminescence (PL) of $BaCd_2P_2$ powder

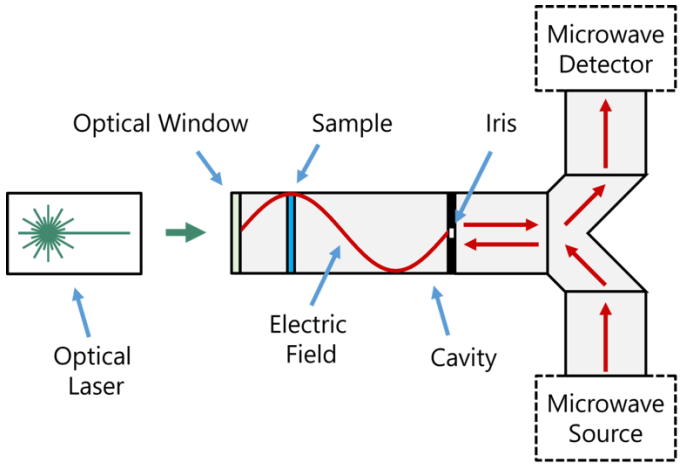


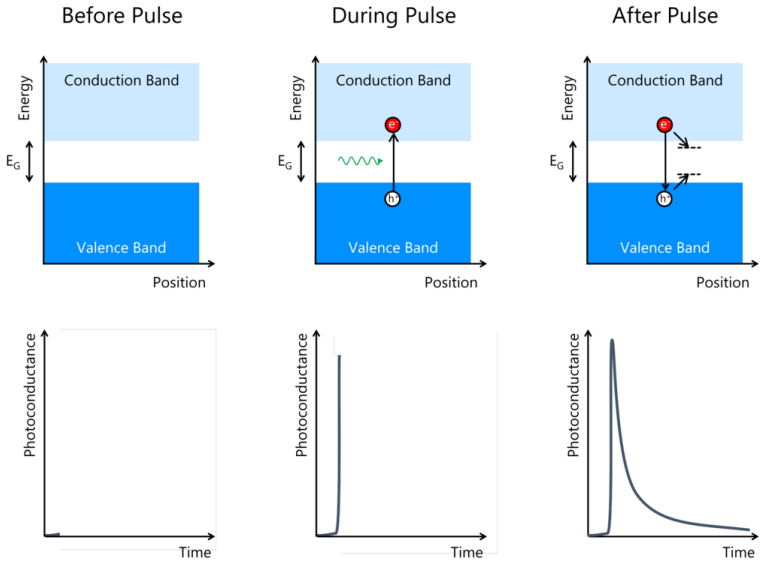


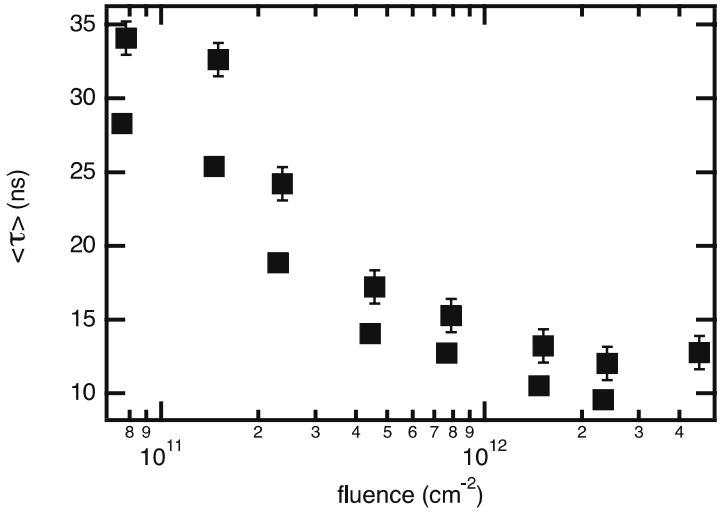
- Confirm the computed band gap
- Bright, stable PL

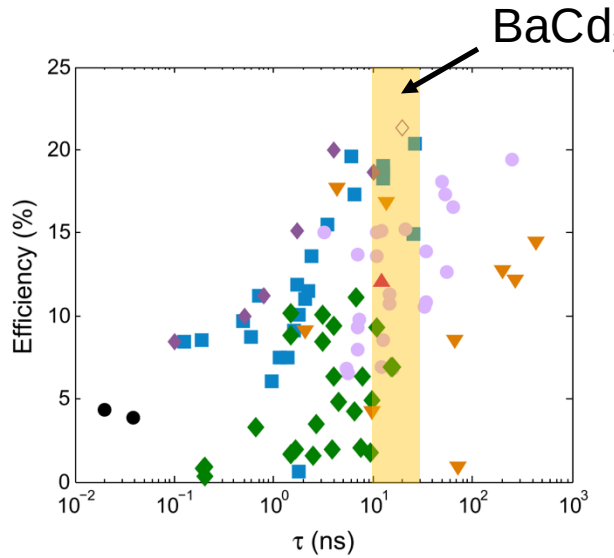
Carrier lifetime in $BaCd_2P_2$ powder pellet

Time resolved microwave conductivity









R. Jaramillo *et al.*, 2016

CdTe, CIGS, CZTS, $MAPbX_3$, InP, GaAs, SnS

Conclusions

- Expand the scope of high-throughput computational search for solar absorbers to include intrinsic defects
- Discover $BaCd_2P_2$ as a long carrier lifetime and highly stable solar absorber, confirmed by follow-up experiment

Acknowledgments

U.S. DEPARTMENT OF ENERGY

DE-SC0023509

NERSC

BES-ERCAP0020966

Hautier's group at Dartmouth

Exploring the Family of AM_2Pn_2 Materials and Their Alloys for Use as Solar Absorbers

Dec 1, 2023
9:00am - 9:15am

Hynes, Level 2, Room 207

Chemical synthesis at Iowa State U & Ames Laboratory

M. Hasan, K. Kovnir

PL characterization at Dartmouth

G. Kassa, J. Liu

Thin film growth at NREL and U. Colorado Boulder

S. Quadir, S. Bauers, A. Zakutayev

Photo-activity characterization at UCSD

G. Esparza, D. Fenning

Surface & interface studies at Penn State

C. Chandler, I. Dabo