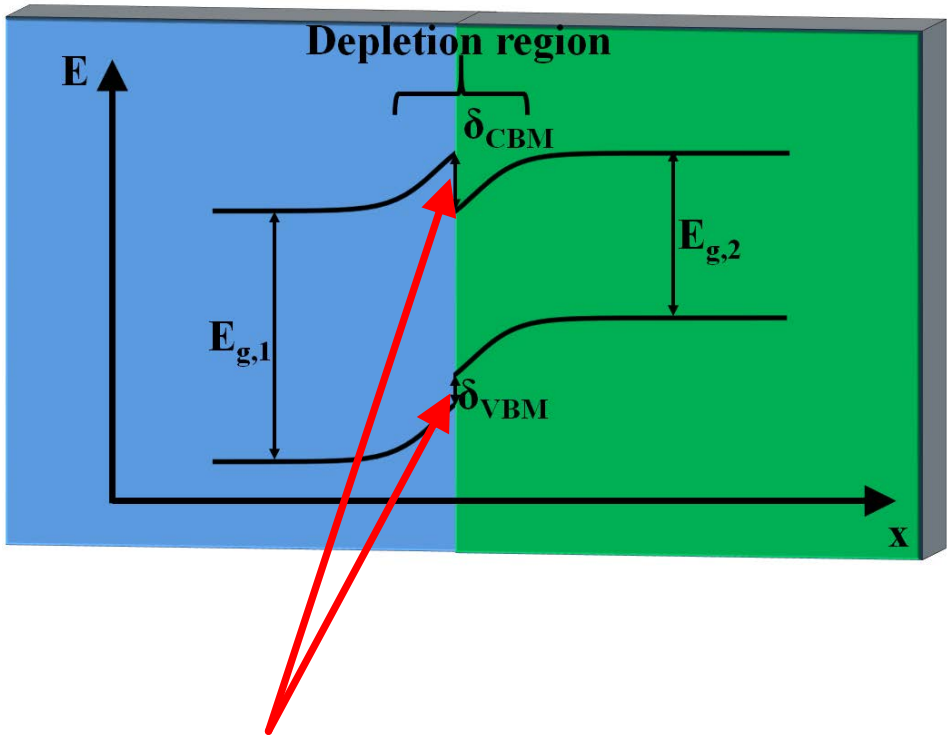
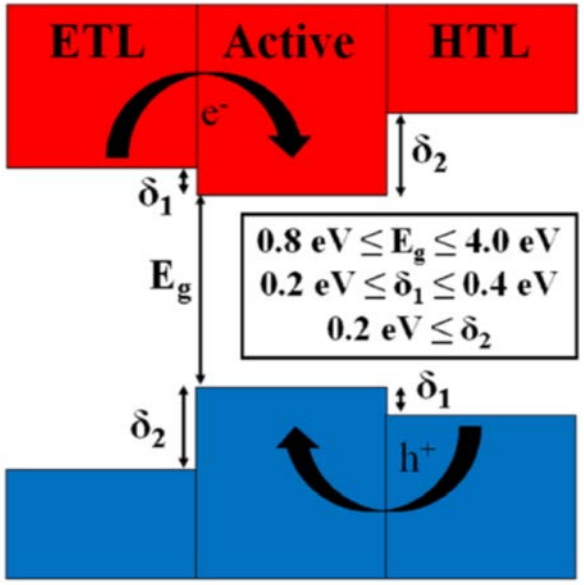
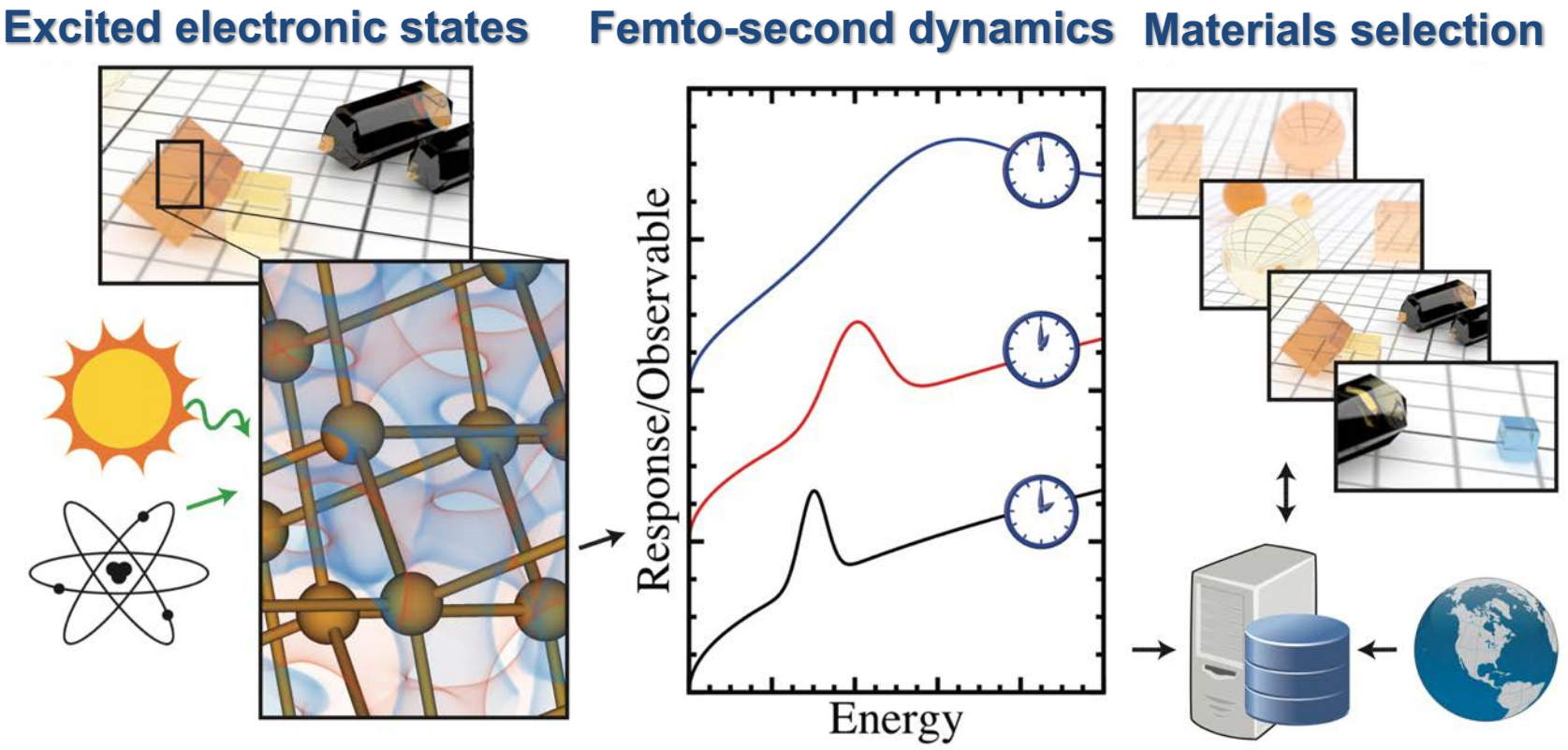
Materials Selection for Semiconductor Heterojunctions





Energy discontinuities at interface for band offsets

Why … Materials selection?

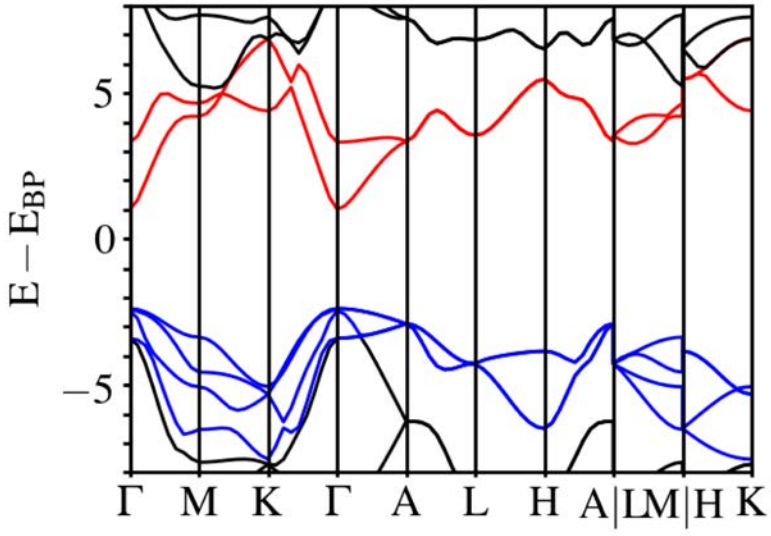


"Pushing The Frontiers Of Modeling Excited Electronic States And Dynamics To Accelerate Materials Engineering And Design", K. Kang, A. Kononov, C.-W. Lee, J.A. Leveillee, E. Shapera, X. Zhang, A. Schleife, Comp. Mat. Sci. **160**, 207–216 (2019); Finalist "Rising Stars in Computational Materials Science"

Heterojunctions: Branch-point energy

- Experimentally and computationally difficult to determine how bands bend and align at interface between materials
- Calculate alignment with branch point energy from online electronic structure data in existing databases

$$E_{BP} = \frac{1}{2N_{\boldsymbol{k}}}\left(\frac{1}{N_c}\sum_{c_i}^{N_c}\varepsilon_{c_i}^{\mathrm{QP}}(\boldsymbol{k}) + \frac{1}{N_v}\sum_{v_i}^{N_v}\varepsilon_{v_i}^{\mathrm{QP}}(\boldsymbol{k})\right)$$



- Band gap corrected with two-tiered scheme
  - Use experimental values if available
  - Linear band-gap correction (Curtarolo) otherwise: $\mathrm{E_g^{corr}} = 1.348\mathrm{E_g^{DFT}} + 0.913\ \mathrm{eV}$
- Use Brus equation for nanocrystals: $\Delta \mathrm{E_g} = \frac{\hbar^2\pi^2}{2\mathrm{R}^2}\left(\frac{1}{\mathrm{m_e^*}} + \frac{1}{\mathrm{m_h^*}}\right)$

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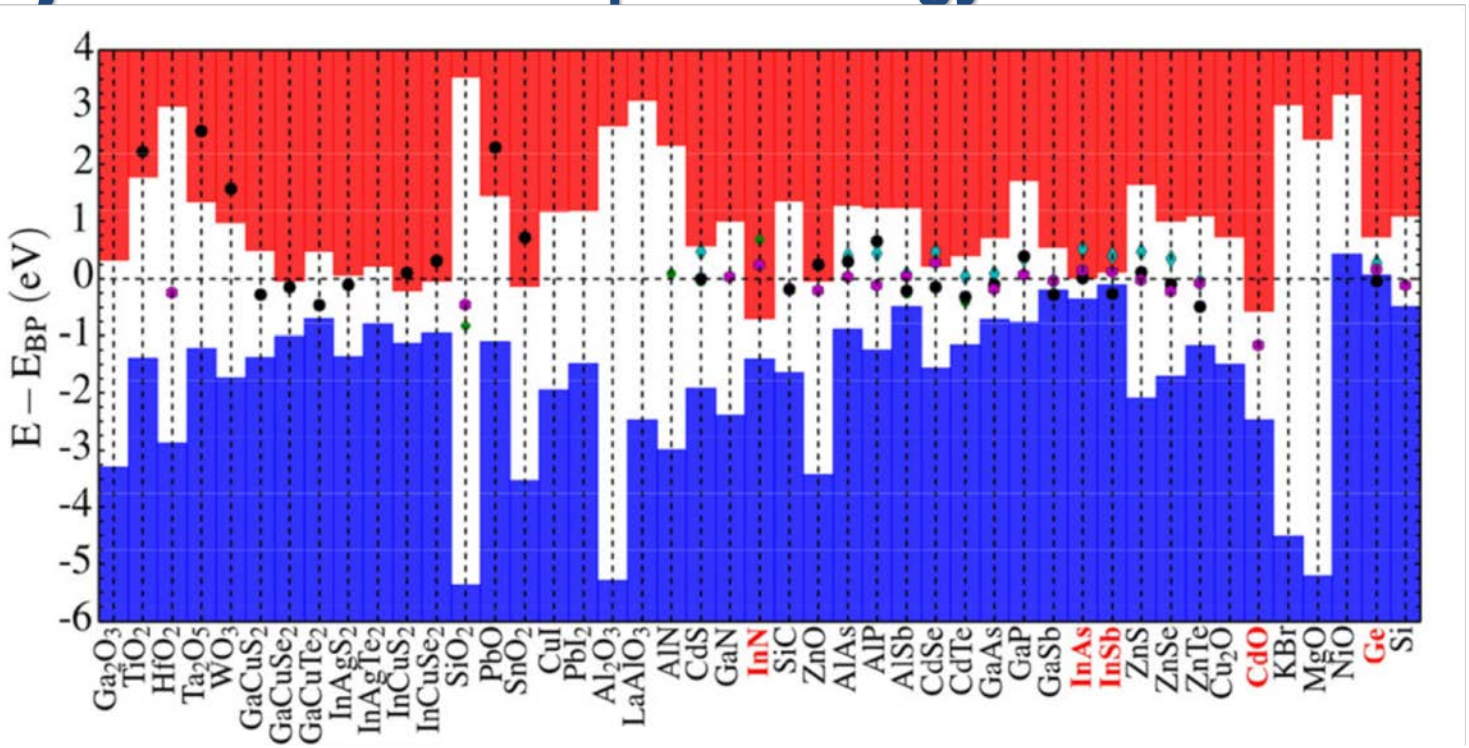
Heterojunctions: Branch-point energy



- Visual comparison of band alignment to different experimental (purple, black) and theoretical techniques (green, cyan), 45 semiconductors
- Comparing to Exp. for 21 materials: Mean absolute error = 0.19 eV
- Vacuum-level alignment for 17 materials: MAE = 0.28 eV (us: 0.12 eV)
- Hydrogen-level alignment: MAE = 0.32 eV (us: 0.31 eV)

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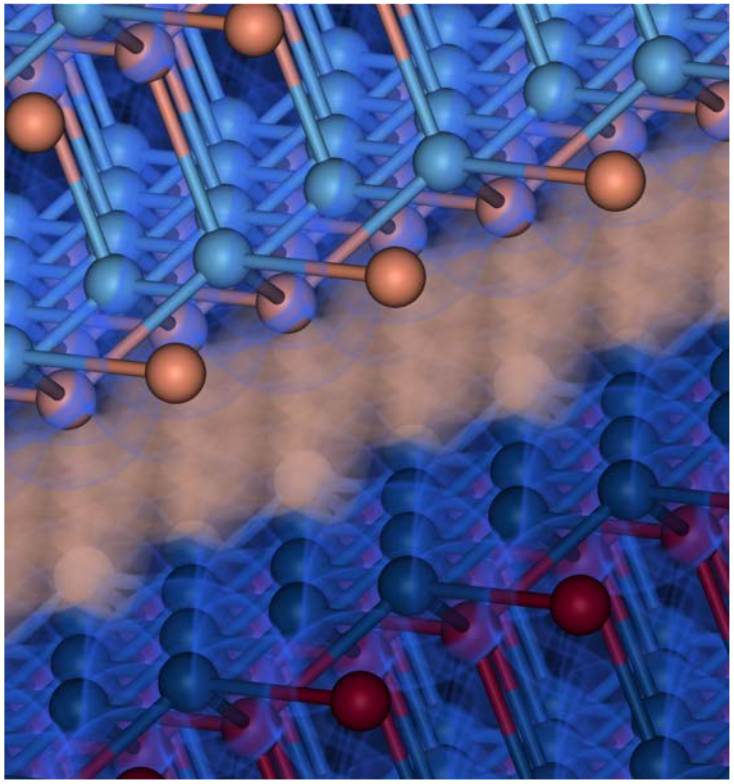
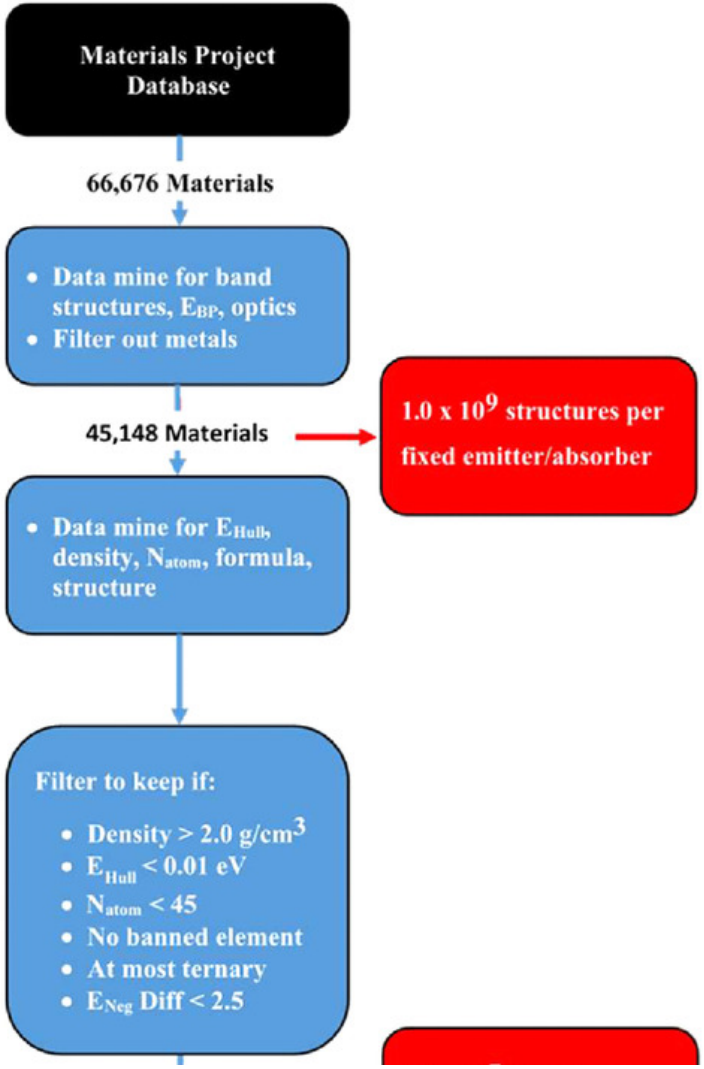
Semiconductor heterojunctions: Results



Semiconductor Heterojunctions: Materials selection



- Data-mine Materials Project database for electronic, crystal properties
- Filtering based on experimental criteria

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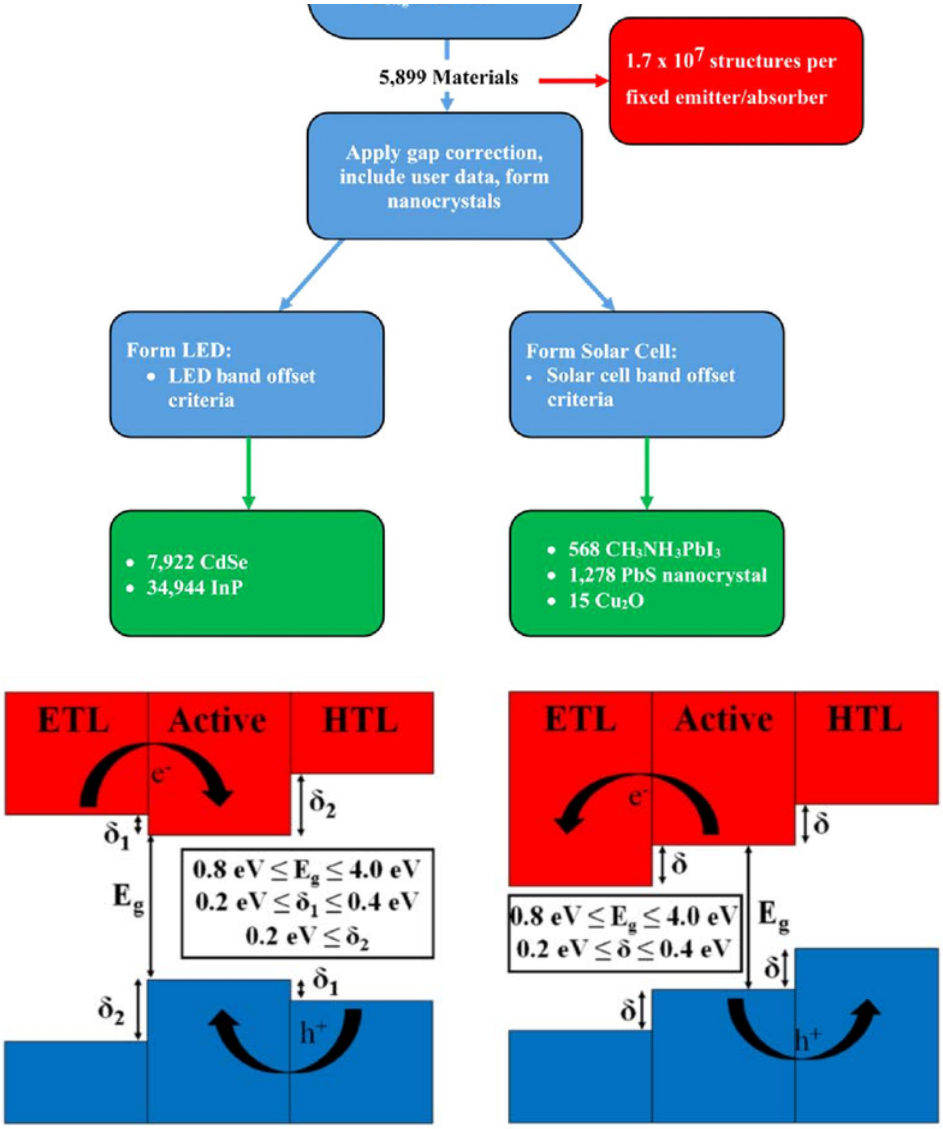
Semiconductor Heterojunctions: Materials selection



- Impose band offset requirements, determined by application
- Layered structure of semiconductors, electron transport layer, active layer, and hole transport layer
- Rank by figure of merit: Drude model conductivity
- Published as jupyter notebook

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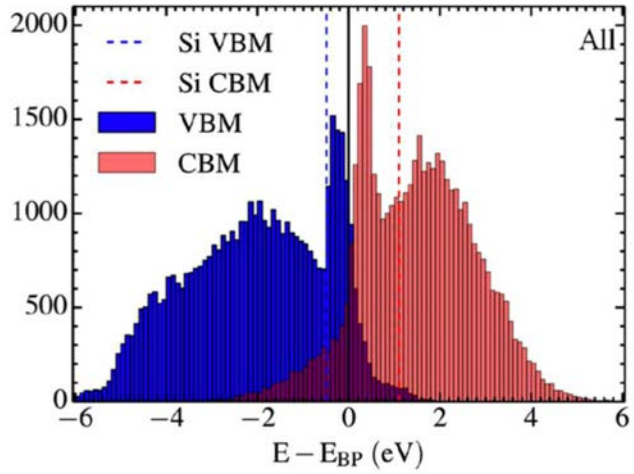
Semiconductor Heterojunctions



- More than 500 materials available for any branch-point energy between 0 and 3.5 eV below the CBM as well as 0 and 4.3 eV above the VBM
- Many candidates! Explore?

Example	Binary	Ternary
LED (CdSe)	264	7922
LED (InP)	1764	34 944
Solar Cell ($CH_3NH_3PbI_3$)	144	568
Solar Cell (PbS nanoparticle $R = 5$ nm)	212	1278
Solar Cell (Cu_2O HTL)	4	15

Example	ETL	Active	HTL
LED	Ca_3N_2, Mg_3N_2, $ZnSeO_4$	CdSe	CdS,[83] WO_3,[84] MoO_3,[85] SiC[86]
LED	ZnSe,[87] CuI, $NaBiS_2$[88]	InP	GaSe, V_2O_5,[89] $SrCuO_2$[90]
Solar cell	WO_3, In_2S_3[91]	$CH_3NH_3PbI_3$	Mn_3O_4, Cr_2O_3[92]
Solar cell	PdS, Fe_3Si[93]	PbS, $R = 5$ nm	MnP, Cr_3S_4
Solar cell	$Ca_2Cu_2O_5$	CdS[94]	Cu_2O

- Found materials that are actually used in practice!
- Found new candidates! Test?
- Problems: Sensitively depends on band-gap correction, branch-point energy, and alignment criteria
- Currently: Fixable by user input

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