What Limits the Performance of Wide Bandgap $Cu(In,Ga)S_2$ Solar Cells ?

UNSW SPREE Seminar, May 2022

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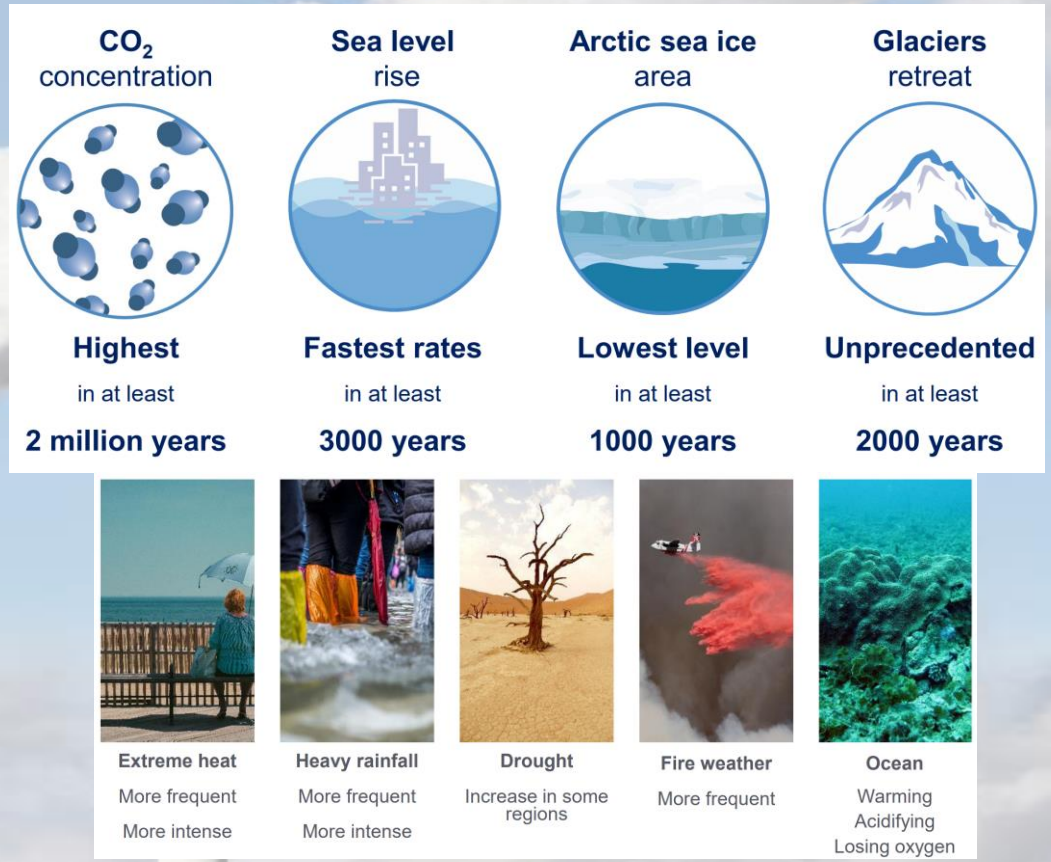
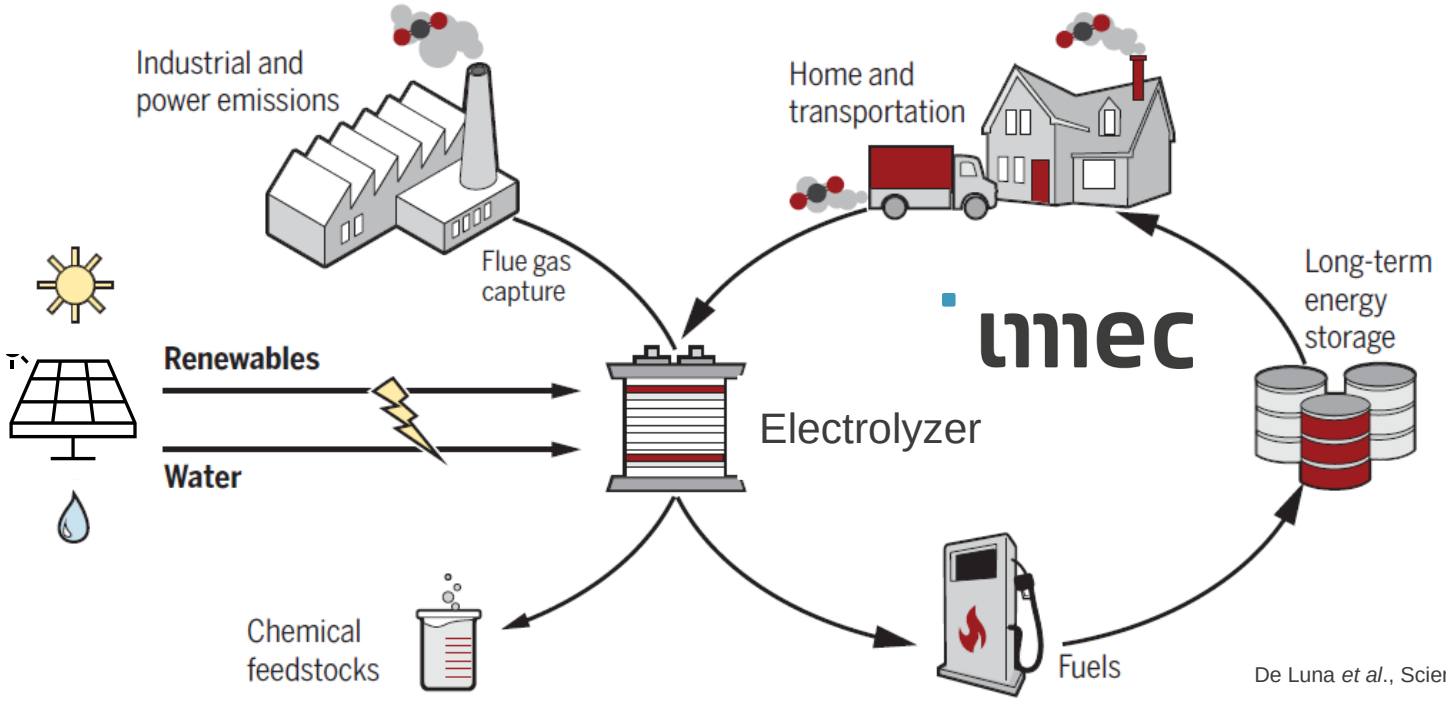
Climate EMERGENCY



Response : Shift from fossil fuels to renewables

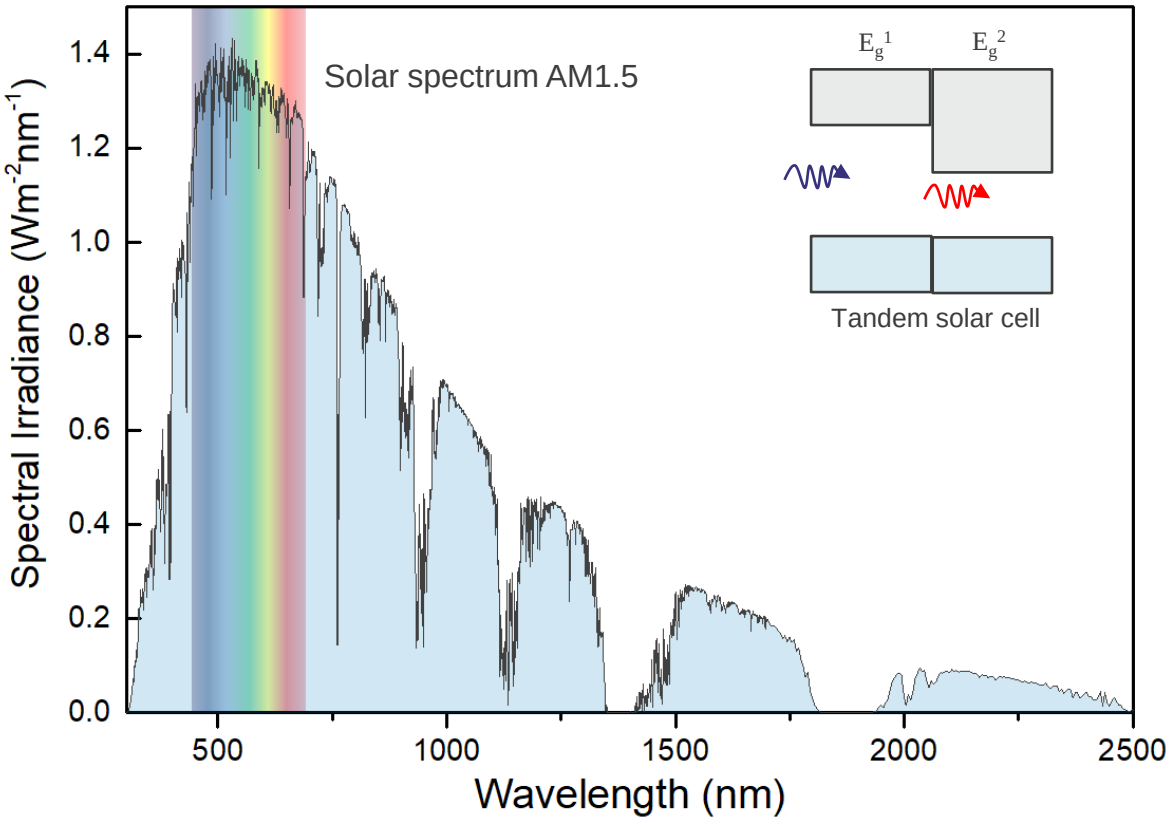
Carbon neutral and circular economy : Solar will be a major player

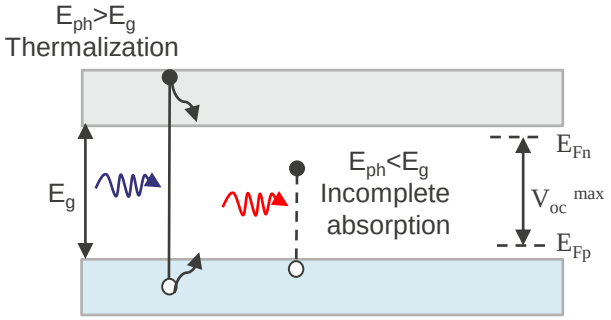


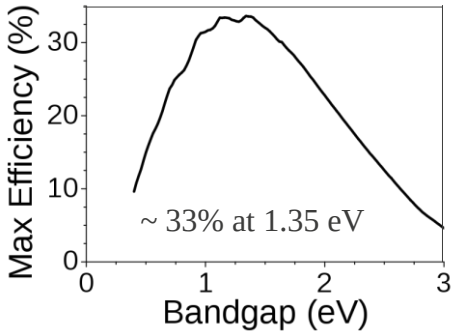
De Luna *et al*., Science 364, 350, 2019

"I'd put my money on the sun and solar energy. What a source of power! I hope we don't have to wait till oil and coal run out before we tackle that" – **Thomas Edison**

Utilization of solar spectrum : Efficient light absorption

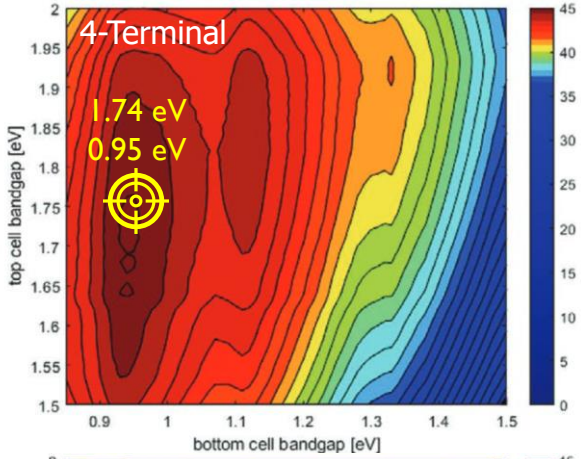


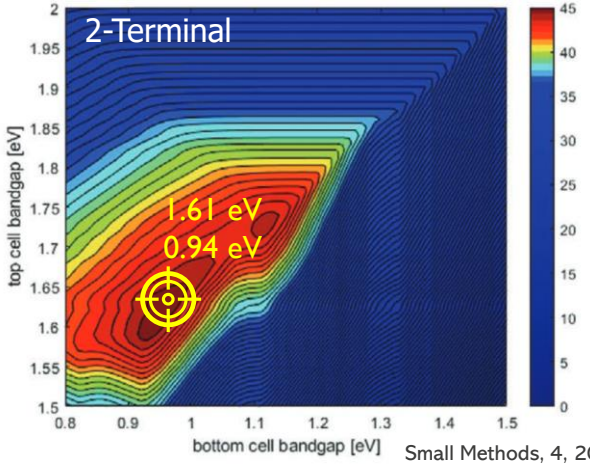


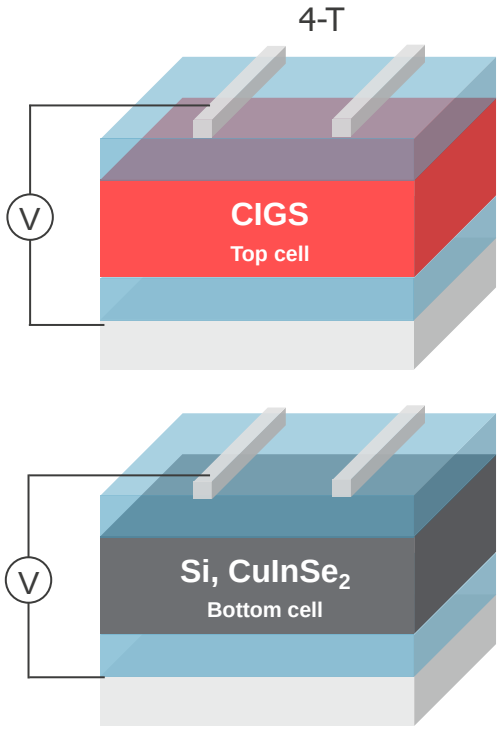


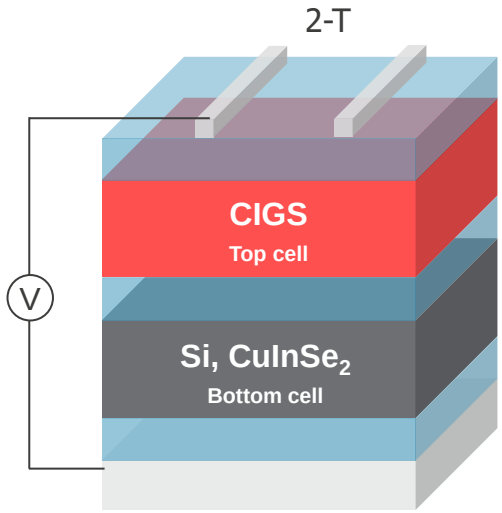
Shockley, Queisser
"Detailed Balanced Limit of Efficiency of –n junction solar cells"
J. Appl. Phys., 32, 510, 1961

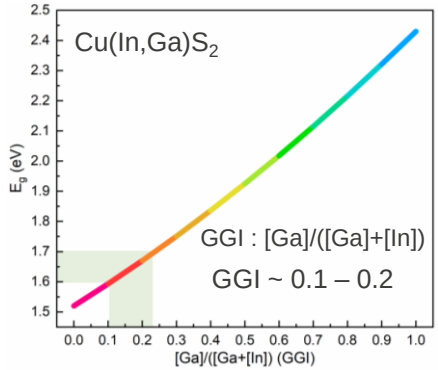
Why Wide band-gap cells ? : Cu(In,Ga)S_2









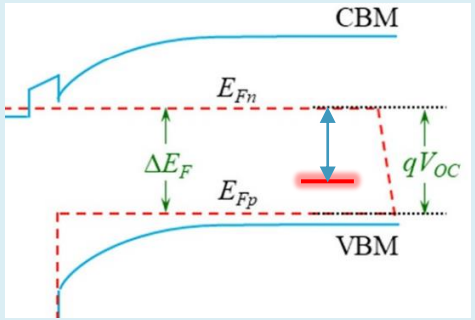


Small Methods, 4, 2000395, 2020

Outline

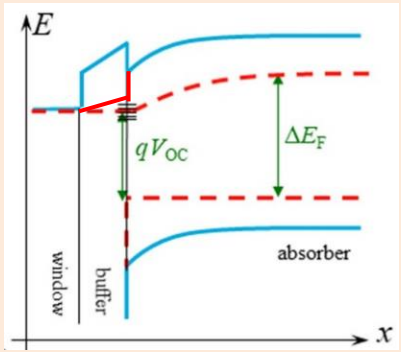
Pure Sulfide $Cu(In,Ga)S_2$ (E_g : 1.5 – 2.4 eV)
Low efficiency compared to CIGSe!

Identification of bulk defects



Anti-site deep defects

Interface passivation



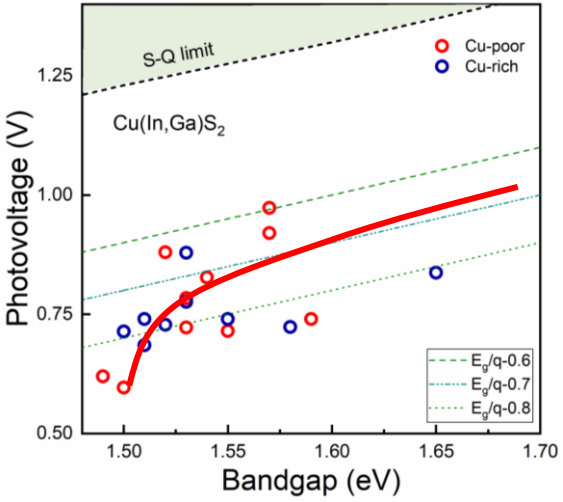
Band alignment

Path forward

- Higher quasi-Fermi level splitting
- Longer carrier lifetimes
- Improved carrier collection
- Growth on TCOs

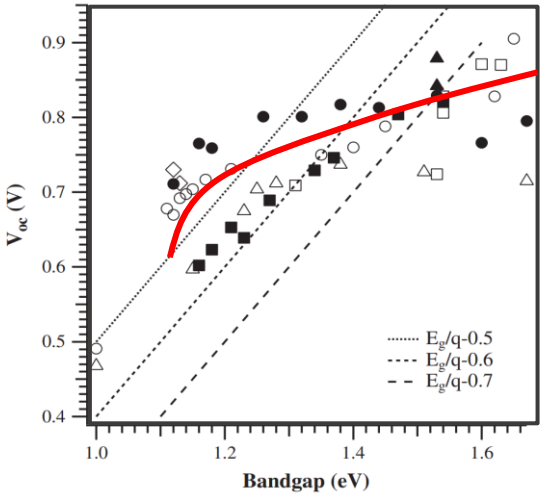
V_{oc} deficit with increasing bandgap : Plateaus

CIGS



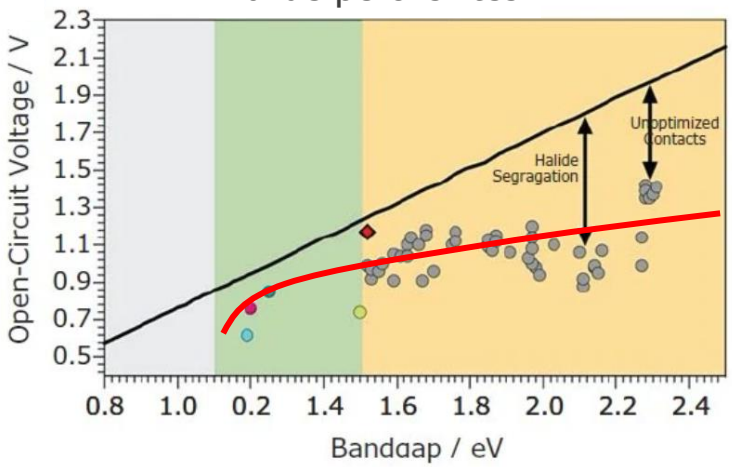
Physical Review Materials 5 (5), 055403, 2021

CIGSe



Prog. Photovolt. Res. Appl. 20, 843, 2012

Halide perovskites



https://www.nrel.gov/pv/cell-efficiency.html

PHYSICAL REVIEW B **93**, 201304(R) (2016)

Role of excited states in Shockley-Read-Hall recombination in wide-band-gap semiconductors

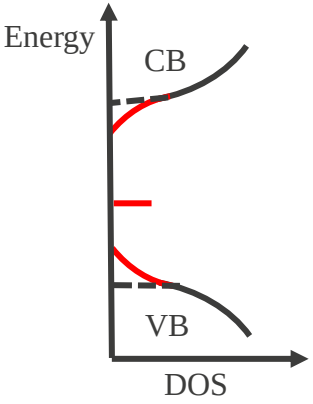
Audrius Alkauskas,[1,*] Cyrus E. Dreyer,[2,†] John L. Lyons,[2,‡] and Chris G. Van de Walle[2]

[1]*Center for Physical Sciences and Technology, Vilnius LT-01108, Lithuania*

[2]*Materials Department, University of California, Santa Barbara, California 93106-5050, USA*

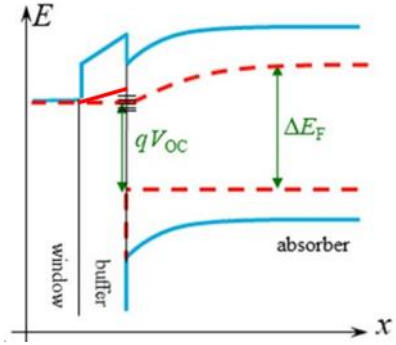
\+ Interface

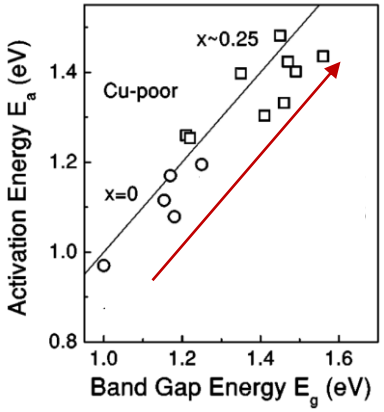
Photovoltage Deficit

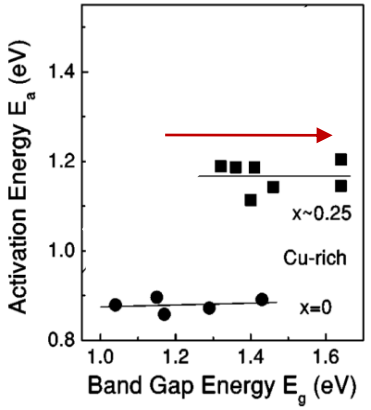


Recombination losses

- **Bulk**
  - Deep defects
  - (Band-) Tail states
  - Potential fluctuations
- **Interface**
  - Fermi level pinning
  - Negative CBO (cliff)
  - Near-interface defects
- **Grain boundaries**



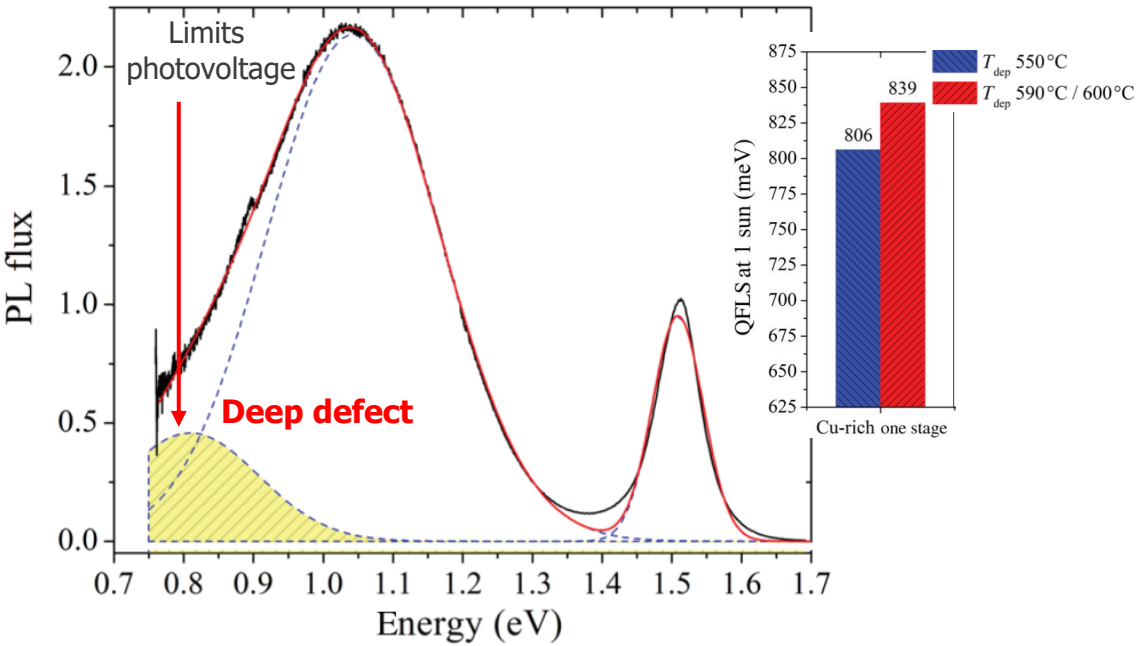




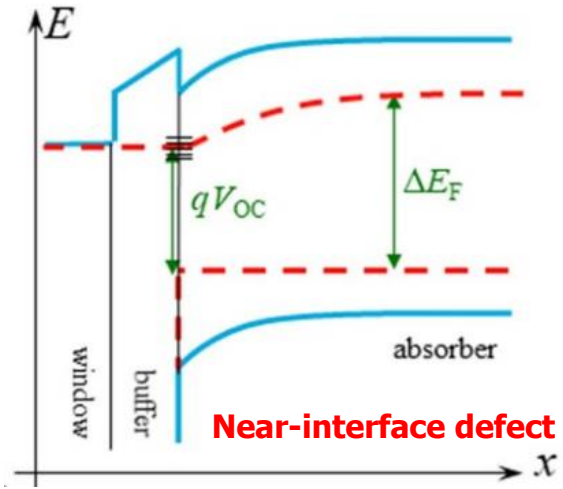
Appl. Phys. Lett. 80, 2598, 2002

Before $Cu(In,Ga)S_2$: $CuInS_2$

Remain limited by deep defects (two) and near interface defects



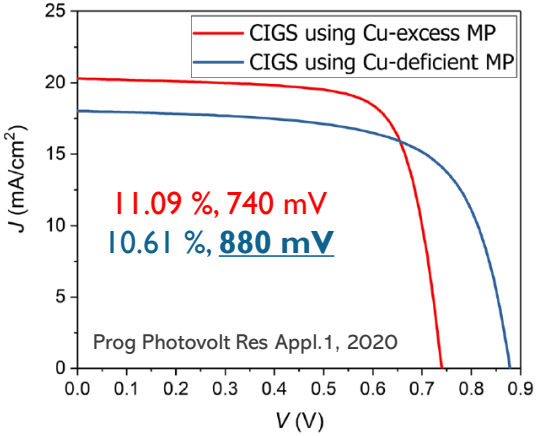
Phys. Rev. Appl., 11, 054052, 2019

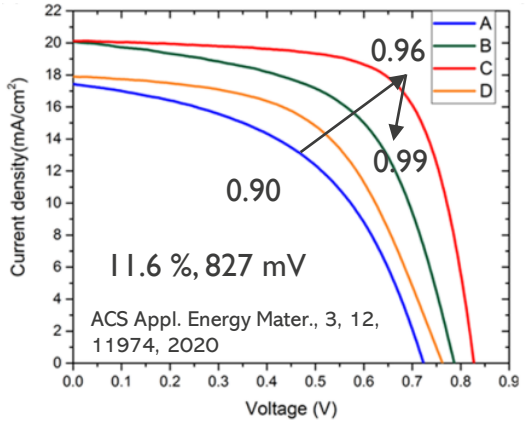


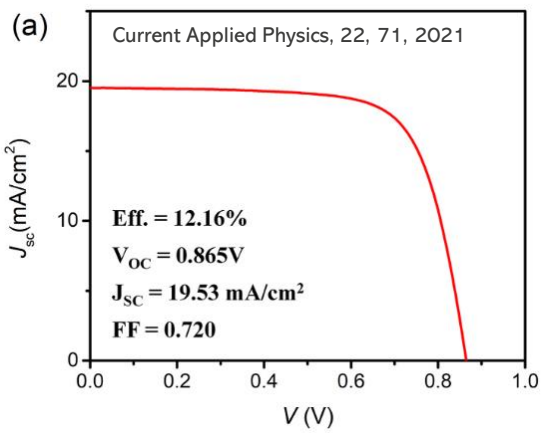
PCE_{max} < 11 %

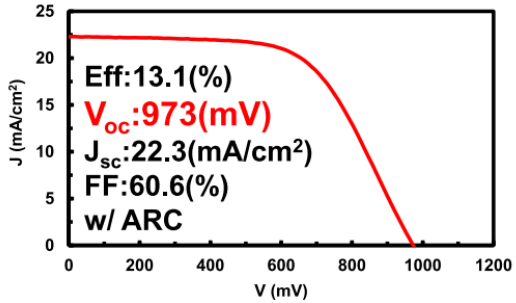
ACS Appl. Mater. Interfaces,14, 7, 9676, 2022

$Cu(In,Ga)S_2$ Solar Cells

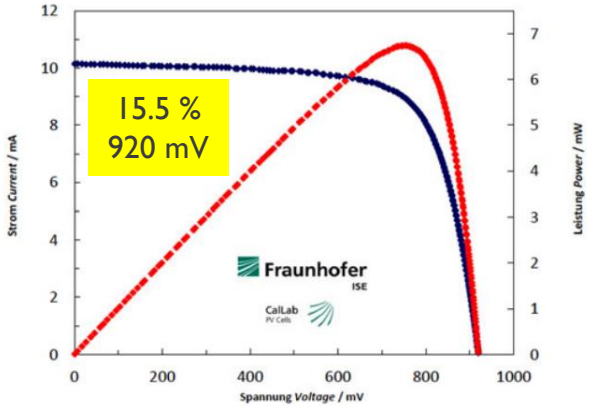




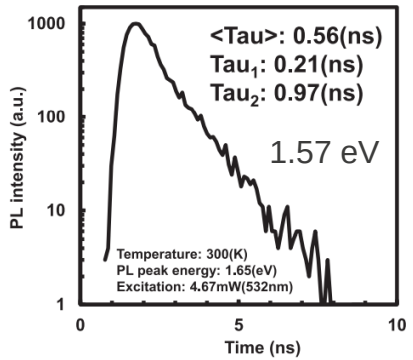




IEEE Journal of Photovoltaics, 6, 6, 1630, 2016



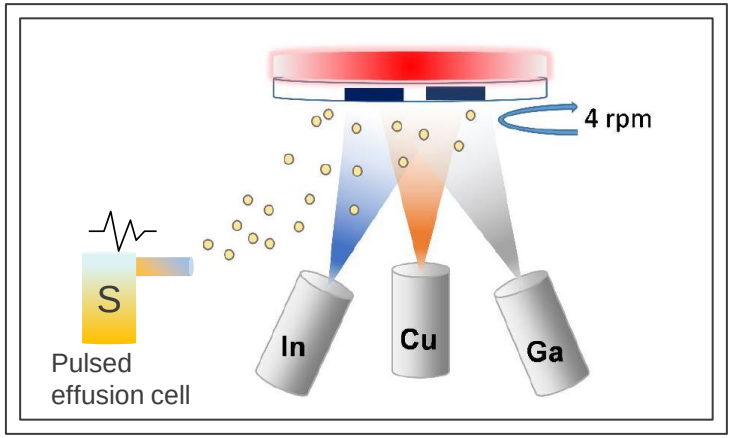
IEEE Journal of Photovoltaics, 6, 3, 760, 2016



Open QUESTIONS

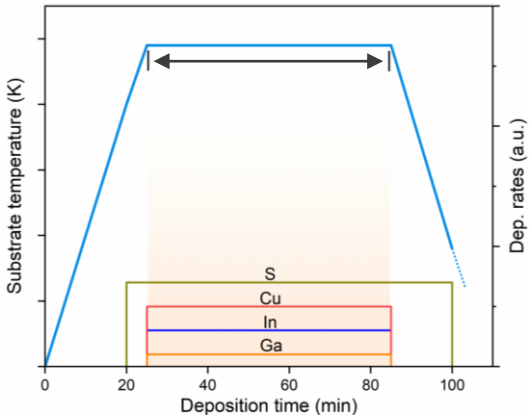
- What limits the V_{oc} in CIGS solar cells → Bulk as well as interface
- What are the deep defects in CIGS ? How can it be passivated ?
- How to fix interface recombination losses in CIGS ?

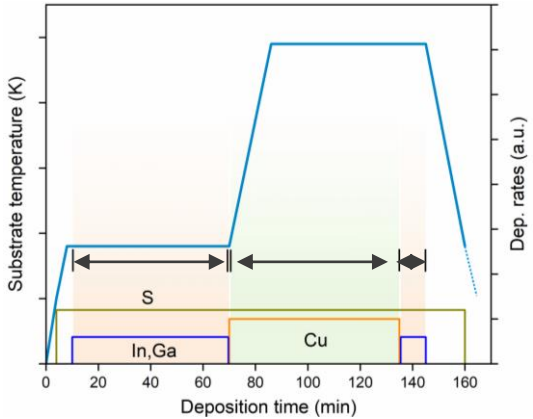
Absorber growth : Co-evaporation



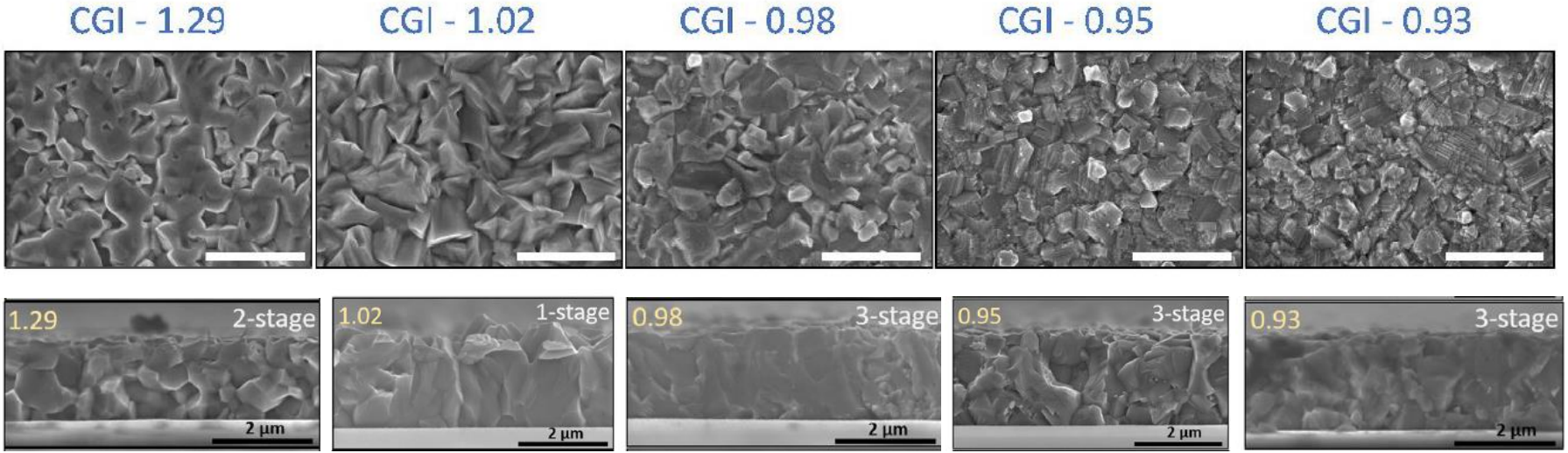
CGI : [Cu]/[Ga+In]
Cu-excess : Stoichiometric CIGS + CuS

GGI : [Ga]/[Ga+In] ~ 0.12 – 0.18



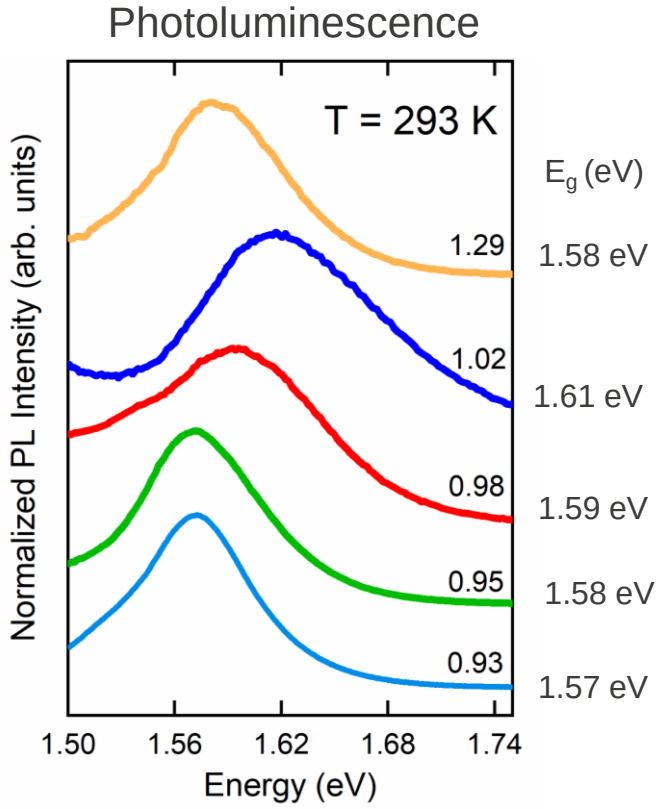


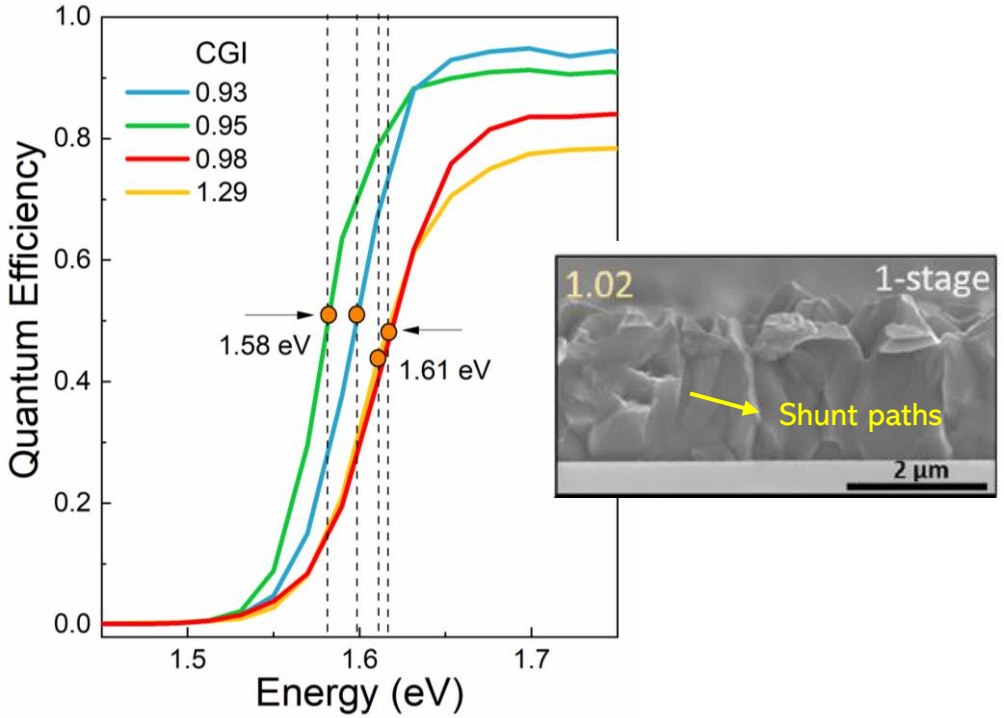
Structure and topography



Shukla, Sood *et al.,* Joule, 5, 1, 2021

Cu-rich and Cu-poor CIGS





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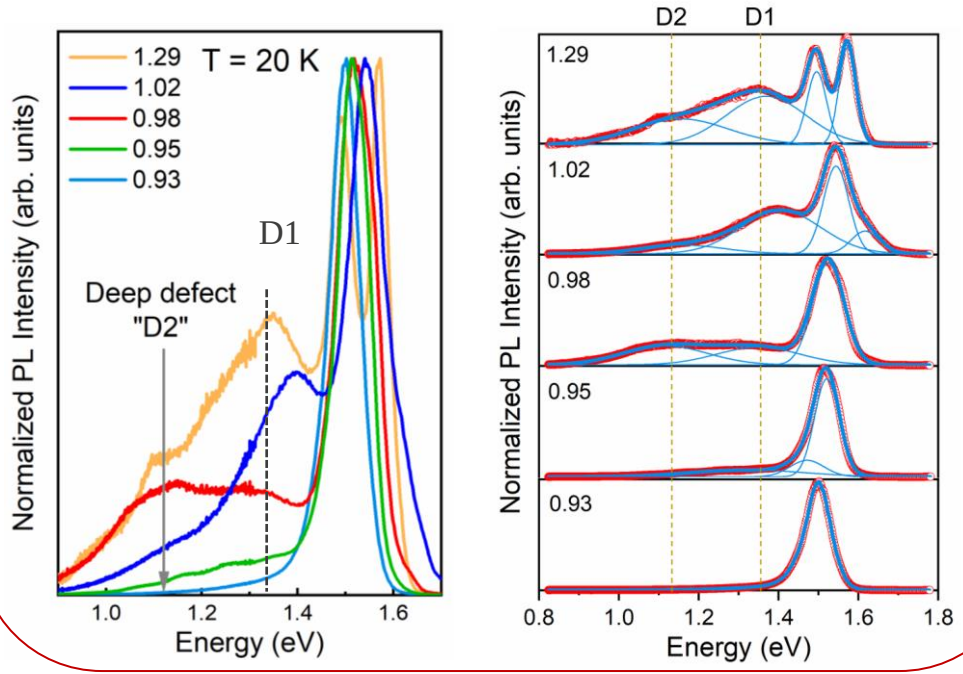
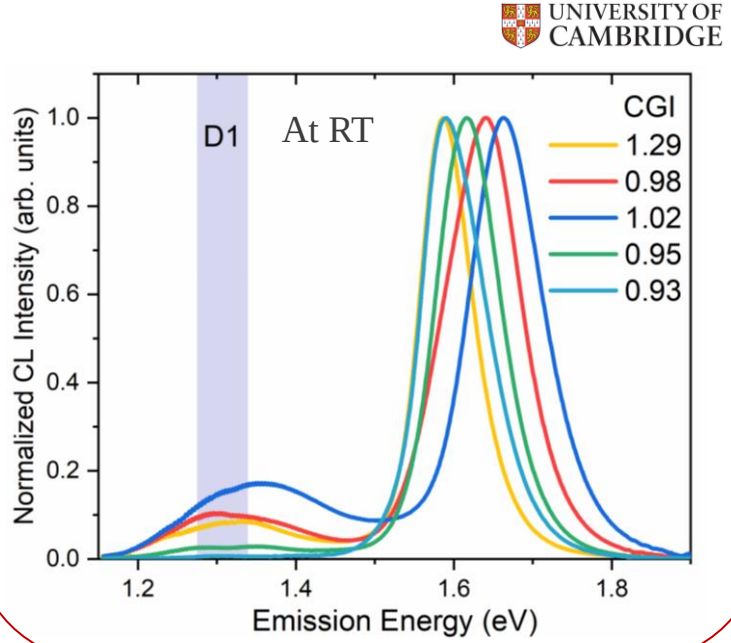
Deep defect : D1 and D2

Photoluminescence



Cathodoluminescence



Two dominant deep defects at ~ 200 meV (D1) and 500 meV (D2) far from the band-edge

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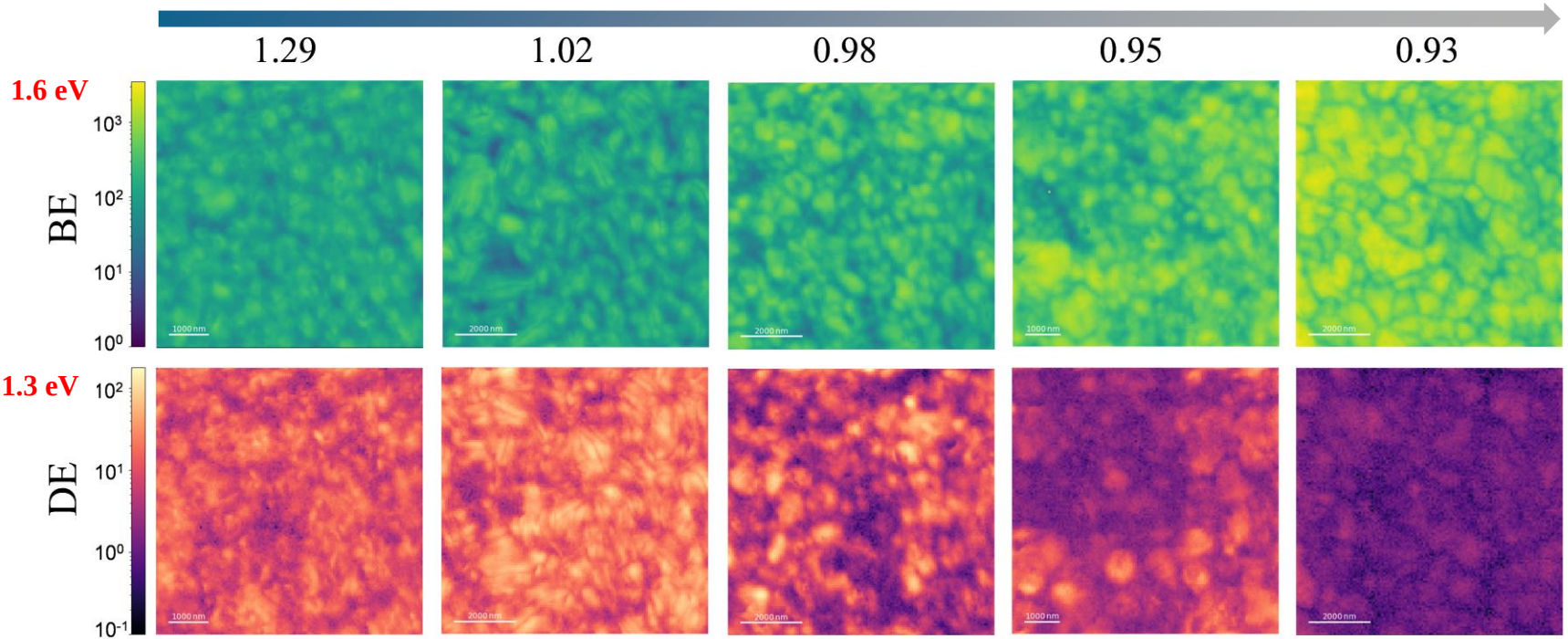
Cathodoluminescence : Band-edge and defect emission



Higher band-edge emission and lower defect band emission for Cu-poor absorbers

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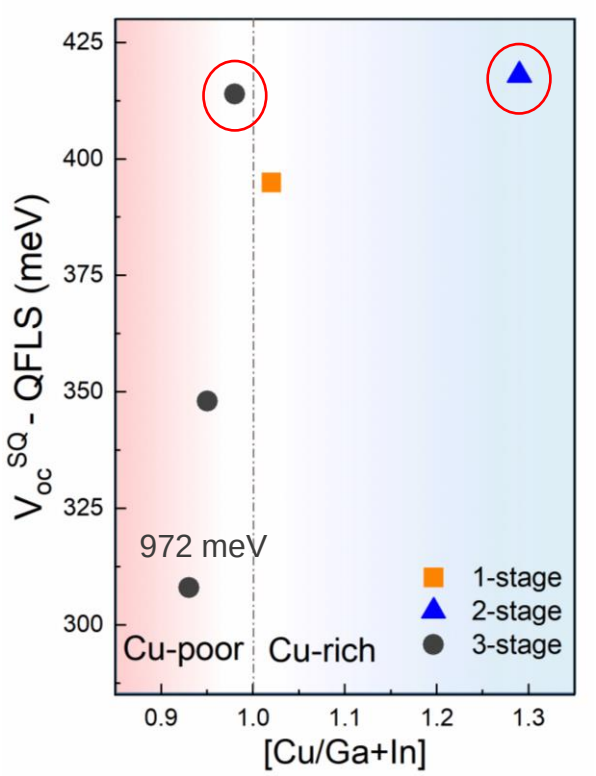
Lower V_{oc} deficit for Cu-poor CIGS



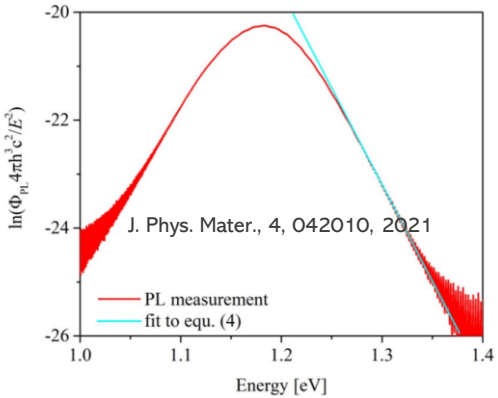
Principle of detailed balance – Emission = absorption

$$\varphi_{PL(E)} = a(E)\varphi_{BB(E)}e^{\Delta EF/kBT}$$

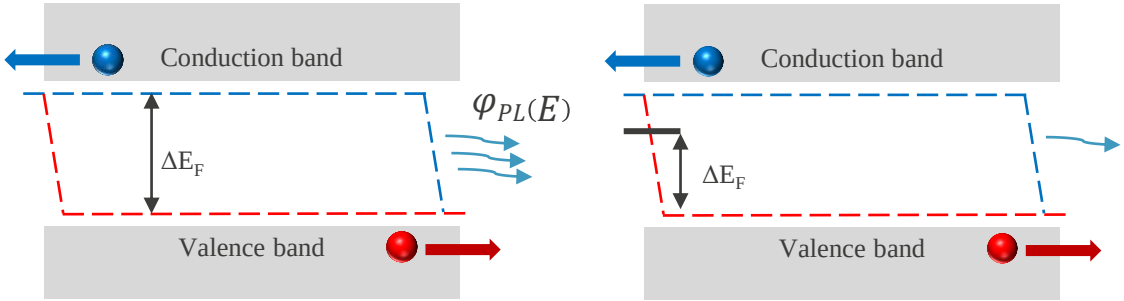
$$\varphi_{BB(E)} = \frac{E^2}{4\pi^2\hbar^3c^2}e^{-E/kBT}$$

$$\varphi_{PL(E)} = a(E)\frac{E^2}{4\pi^2\hbar^3c^2}e^{-(E-\Delta EF)/kBT}$$

Generalized Planck's law



V_{oc}^{max} = QFLS = $\Delta E_F/q$



Thinking about even more Cu-poor films ? : OVCs

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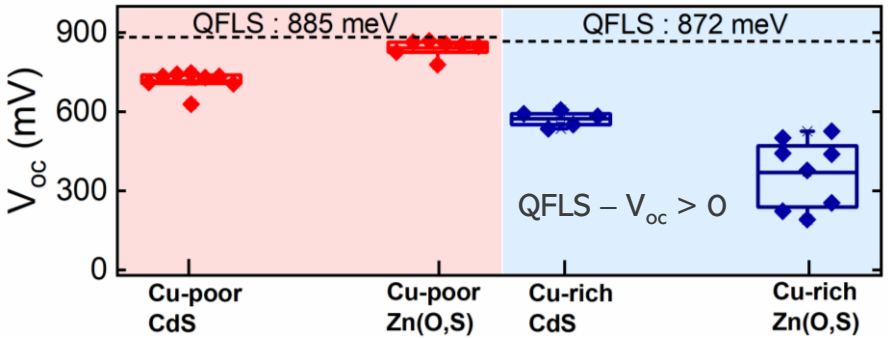
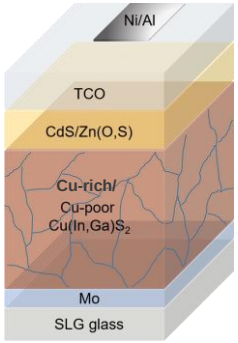
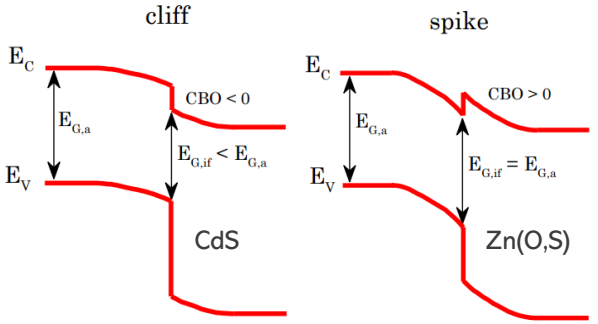
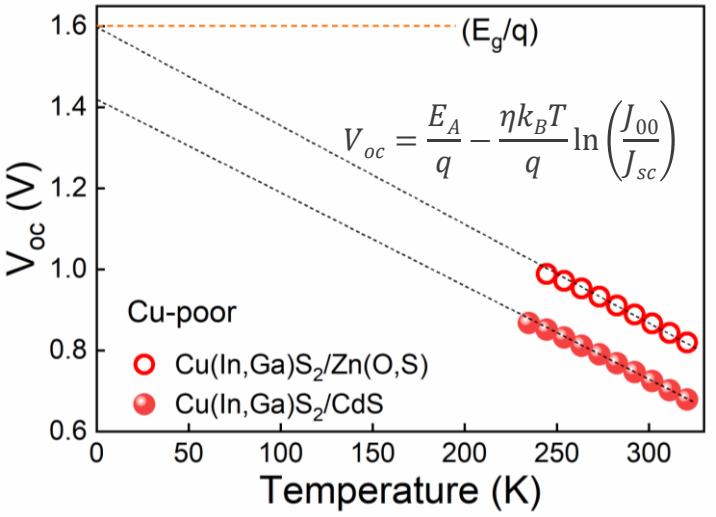
Interface losses

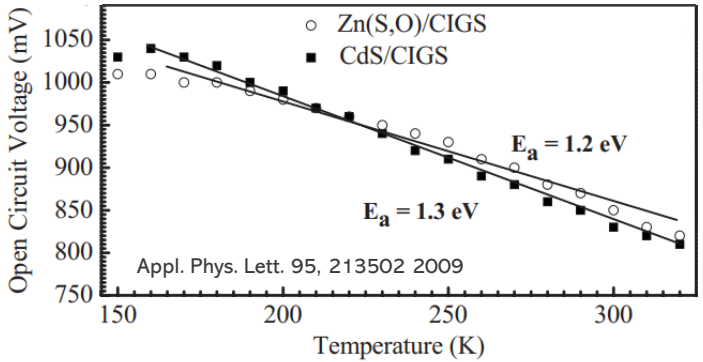






Courtesy - Ph.D. Thesis. A. Lomuscio





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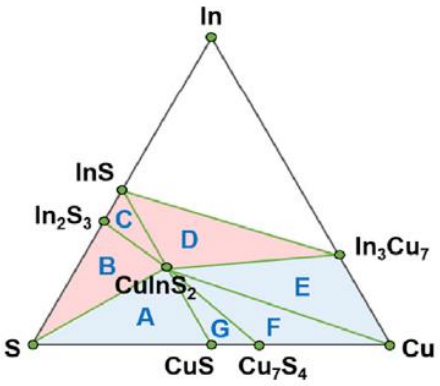
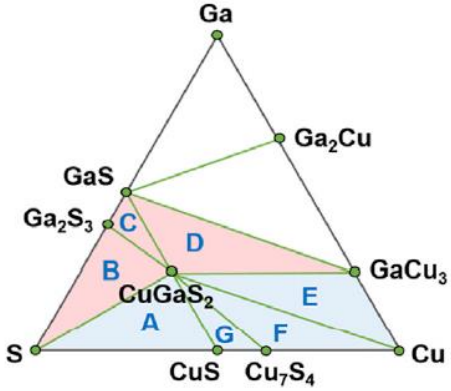
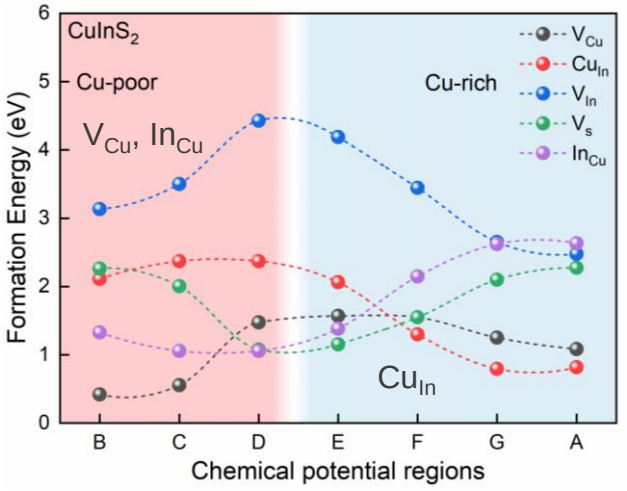
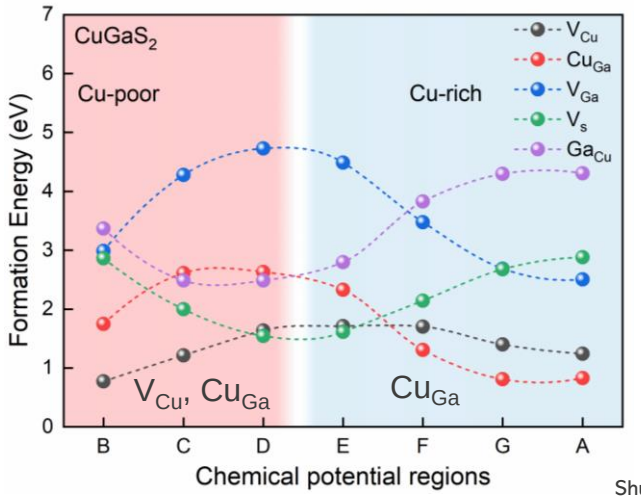
So, what are these defects ?

Defect Formation Energies : Ab-initio calculations









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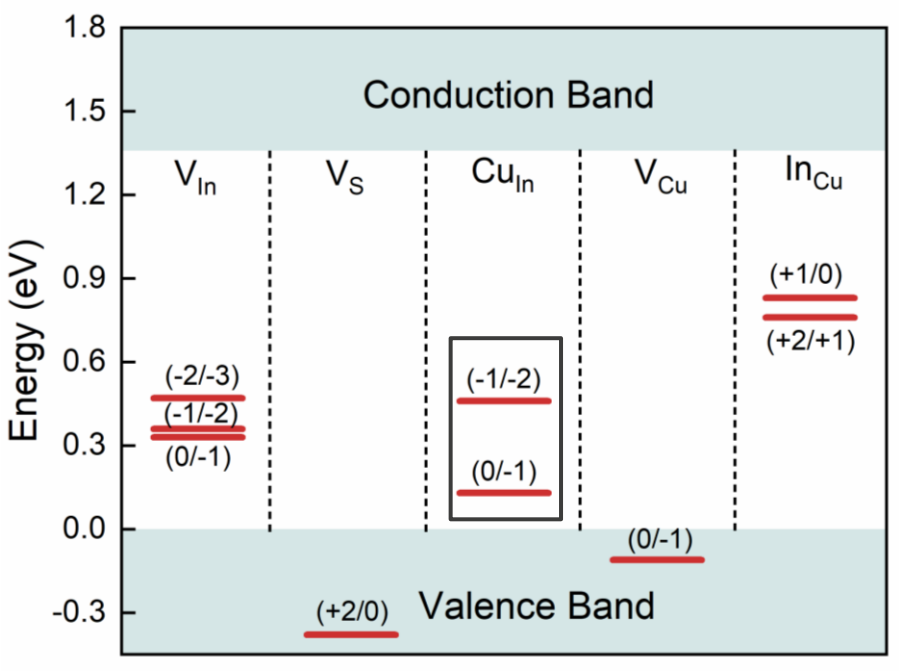
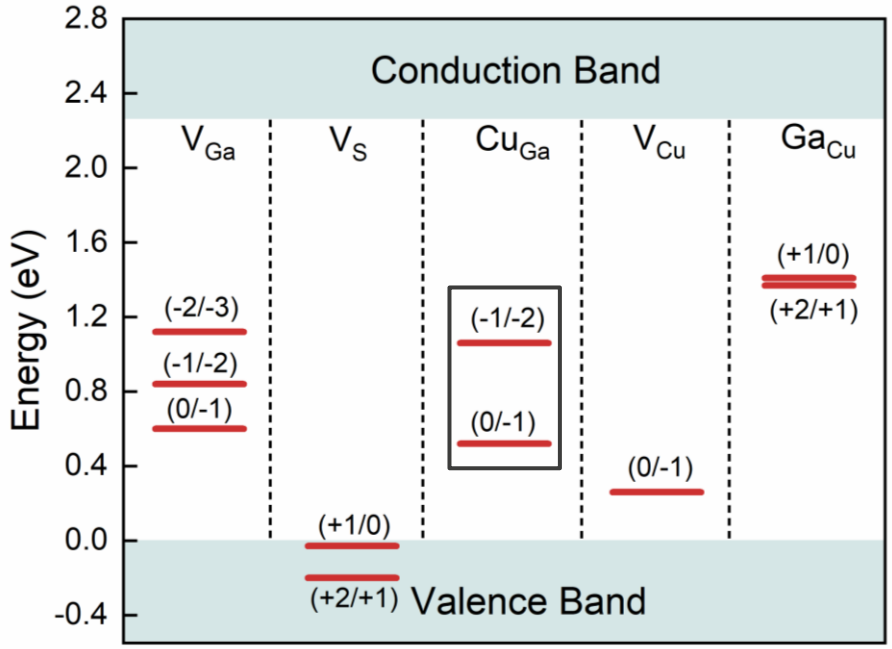
Deep defects : Anti-site defects





- Acceptor like **Cu_{In} and Cu_{Ga}** have the lowest formation energy in **Cu-rich domain** → killer defects
- Donor like **In_{Cu} and Ga_{Cu}** defects have low formation energy in **Cu-poor domain** → compensation ?
- More Cu-poor CIGS : Think about defect complexes - V_{Cu} + (In_{Cu} + Ga_{Cu})

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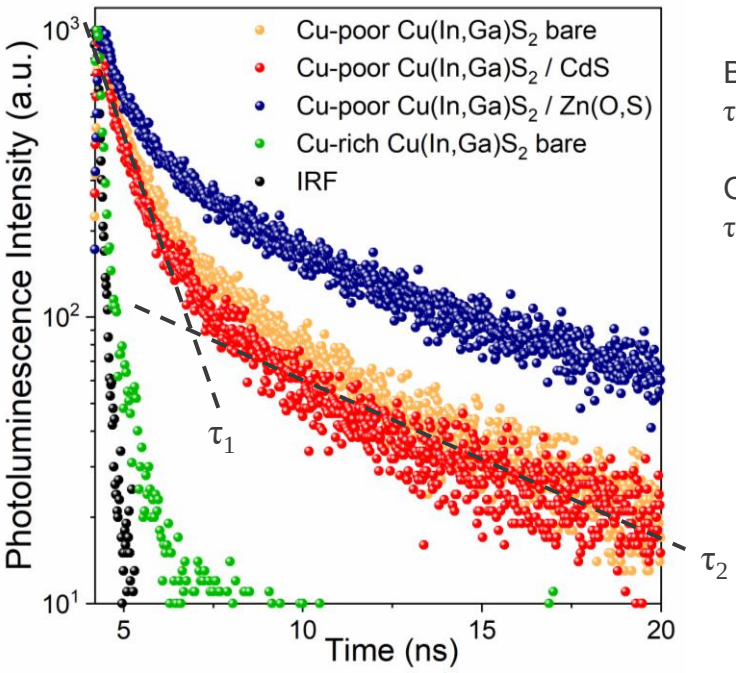
Charge carrier lifetime

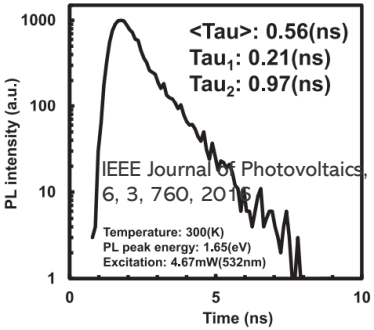


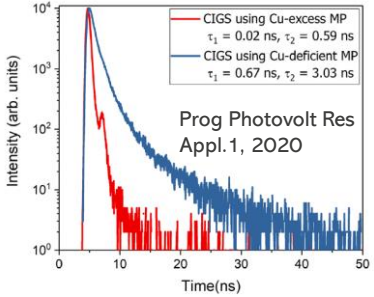
λ_{ex} : 640 nm
PL @ 790 nm (1.56 eV)

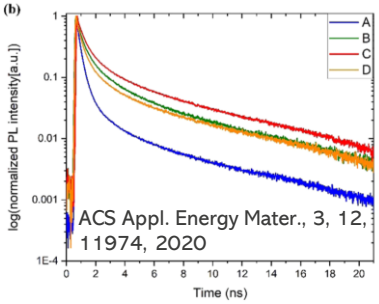
Bare CIGS :
τ_1 : 0.5 ns and τ_2 : 3.2 ns

CIGS/Zn(O,S) :
τ_1 : 0.6 ns and τ_2 : 4.5 ns

High non-radiative recombination
- Deep defects must be passivated







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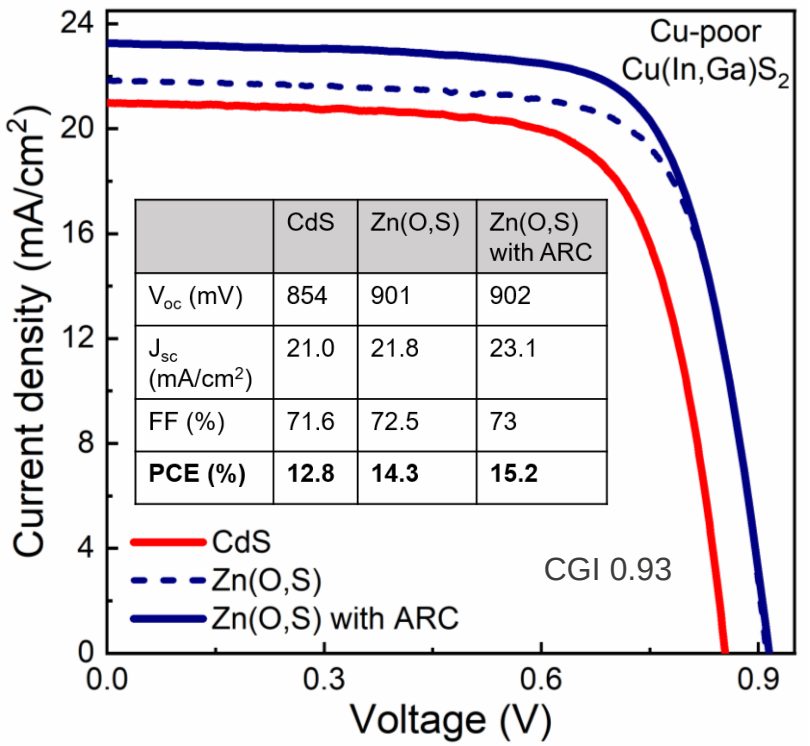
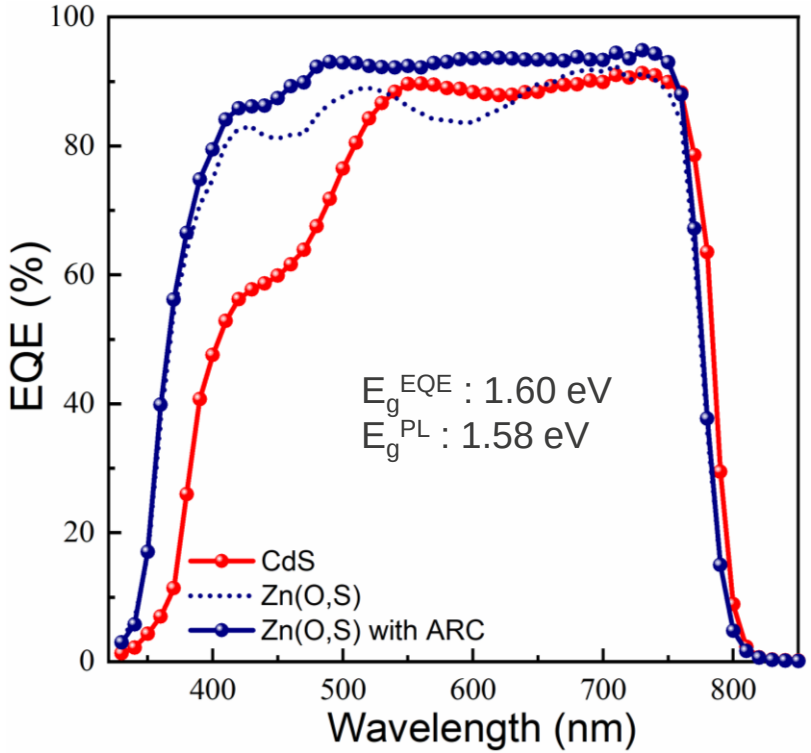
Best solar cell performance



	CdS	Zn(O,S)	Zn(O,S) with ARC
V_{oc} (mV)	854	901	902
J_{sc} (mA/cm^2)	21.0	21.8	23.1
FF (%)	71.6	72.5	73
PCE (%)	**12.8**	**14.3**	**15.2**



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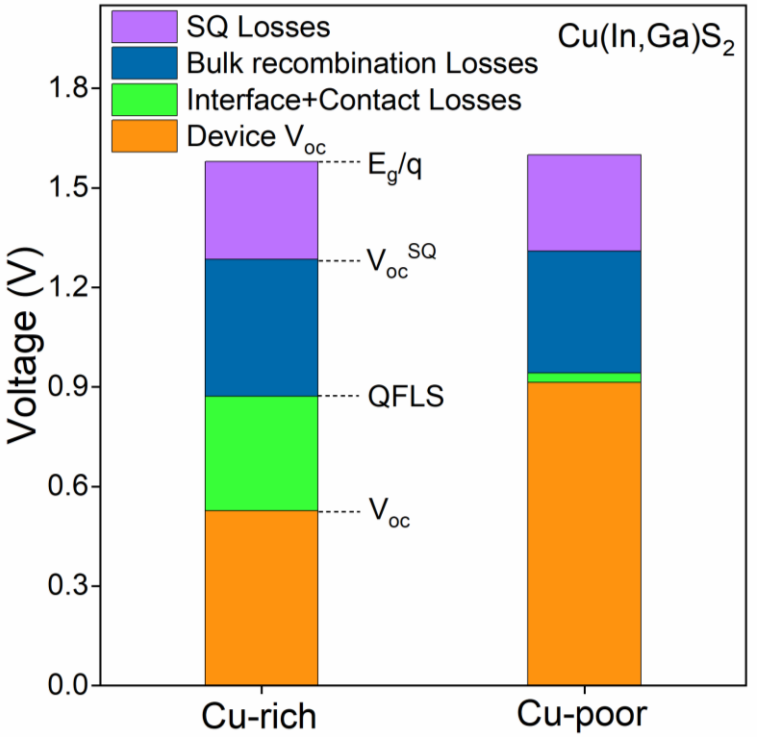
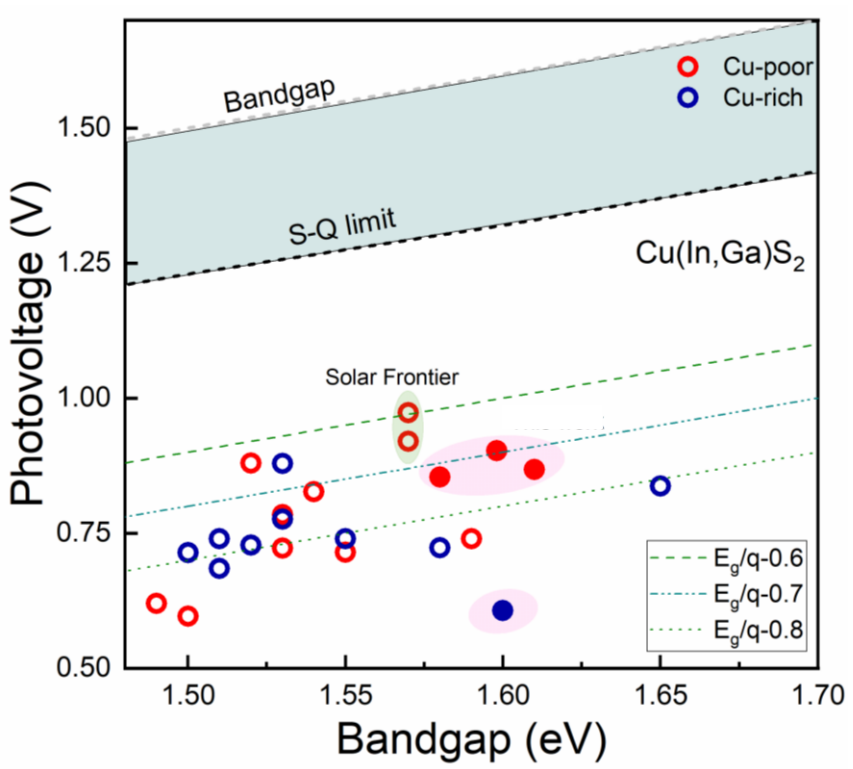
Reduced V_{oc} deficit





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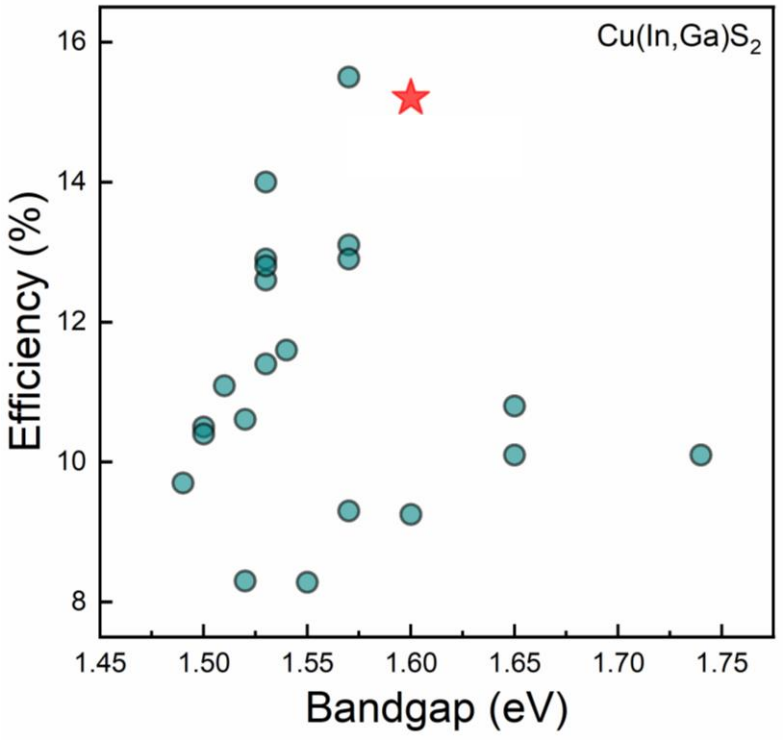
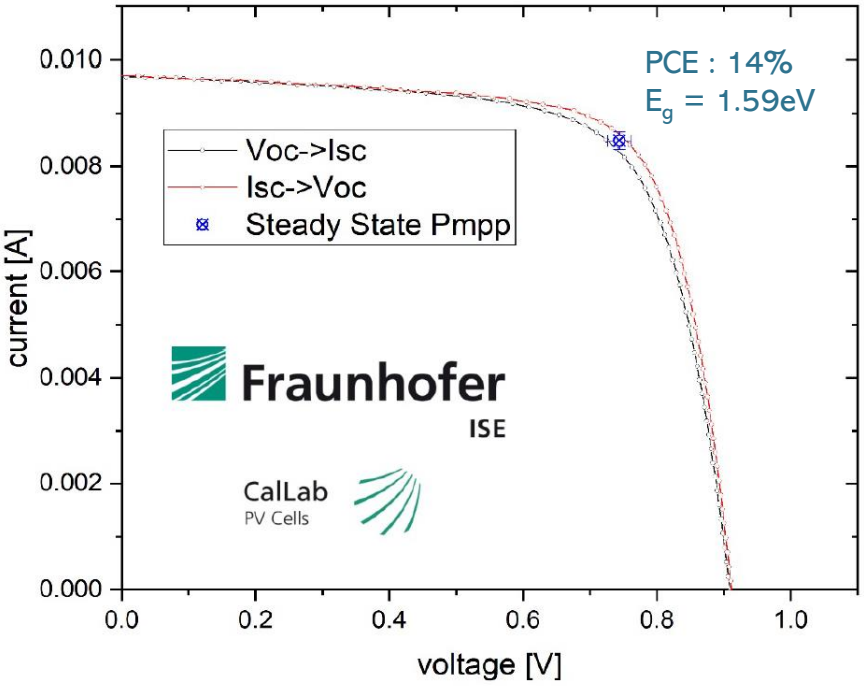
Current status





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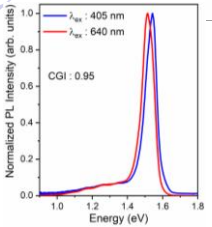
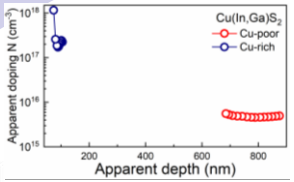
What's next ? A possible roadmap

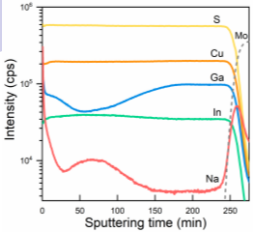
Deep defect Passivation
- Chemical nature
- Sulfur content
- Temperature



Doping density
- Alkali treatment
- Ag doping

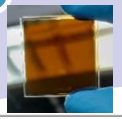


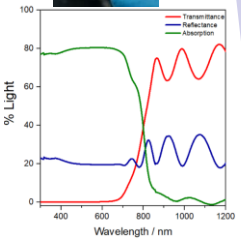
Carrier collection
- Ga grading



GB passivation
- Na, K, Cs

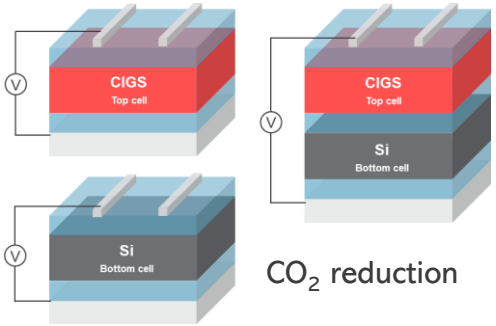
On TCO





Projects involving higher E_g materials

MSCA

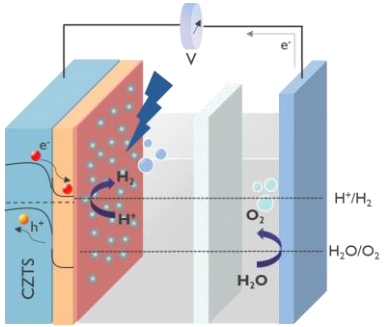


CO_2 reduction

Solar to Fuel Conversion

M-ERA.NET

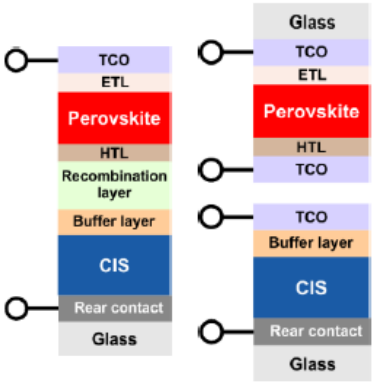
Kesper



Kesterite for water splitting

PERCISTAND

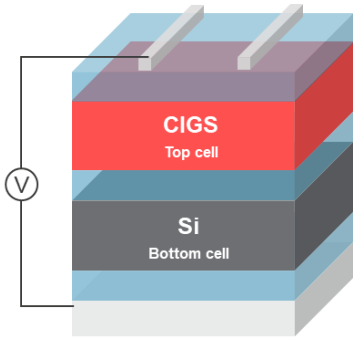
Horizon Europe



Development of all thin-film PERovskite on CIS TANDem photovoltaics

SITA

Horizon Europe



Stable Inorganic TAndem solar cell with superior device efficiency and increased durability

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Gian-Marco Rignanese

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Thank you for your attention

Reduce your carbon footprint

Prof. Bart Vermang

Fonds National de la Recherche Luxembourg