COLOSSAL GRAINS FOR A NEW STRUCTURAL PARADIGM IN THIN-FILM PHOTOVOLTAICS

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- *Bowling Green State University*

ACCGE-22
22nd American Conference on Crystal Growth and Epitaxy

OMVPE-20
20th US Workshop on Organometallic Vapor Phase Epitaxy

200 μm

The Premise

- Grain boundaries in polycrystalline PV absorbers are locations of high defect densities and thus high recombination
  - Passivation strategies exist ($CdCl_2$) to mitigate these effects to some extent but has yet to eliminate it entirely

- Single crystals have shown promise for enabling reduced absorber compensation and recombination but are still far too expensive for large scale PV applications (specifically for III-V and II-VI absorbers)

- Large enough grains and/or templates coupled with fast/cheap epitaxy can enable the best of both worlds
  → our new results suggest that these possibilities may be closer than expected

Outline

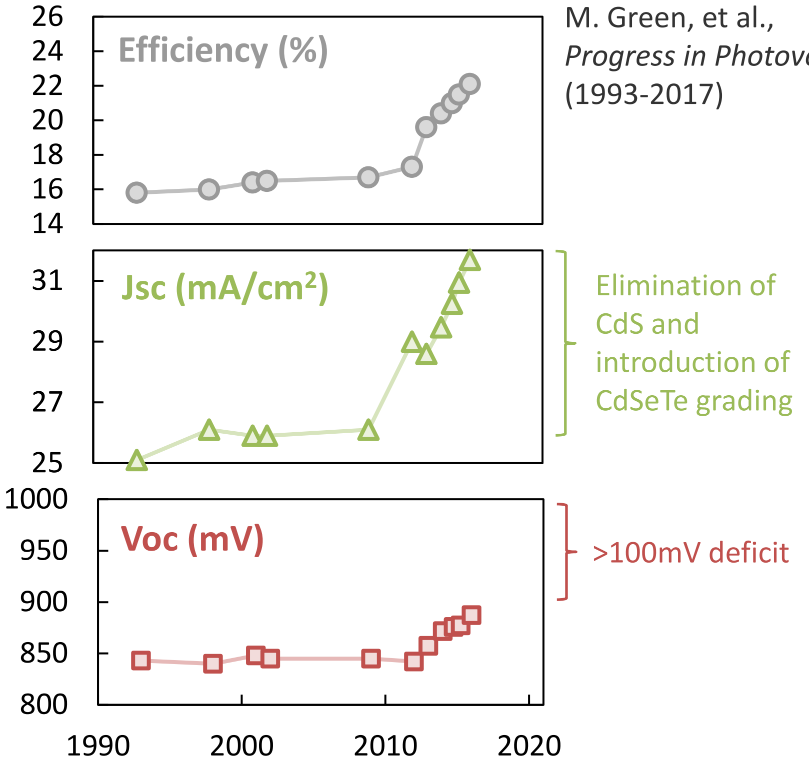
1. Background: CdTe PV and other technologies
2. Fast Epitaxy
3. Grain size, lifetime, and modeling
4. Colossal CdSeTe Grain Growth (CGG)
5. Fast Epitaxy on CGG
6. Doping and Diffusion
7. Conclusions

Outline

Commercial Success – Headroom to Improve

<u>CdTe accounted for 40% of utility-scale PV installations in the US</u> and 8% of PV globally in 2020 & is projected to install 25 GW in the next few years

550 MW CdTe solar farm in the Mojave desert
Photo: First Solar

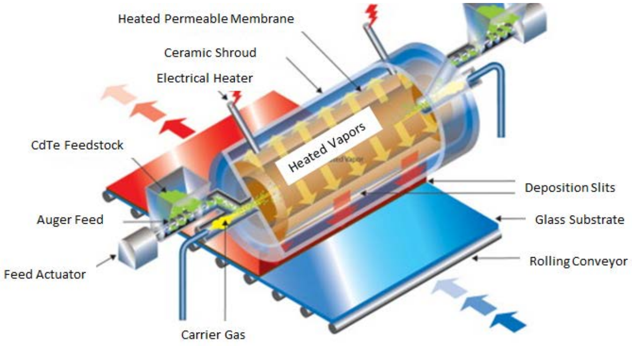


M. Green, et al., *Progress in Photovoltaics* (1993-2017)

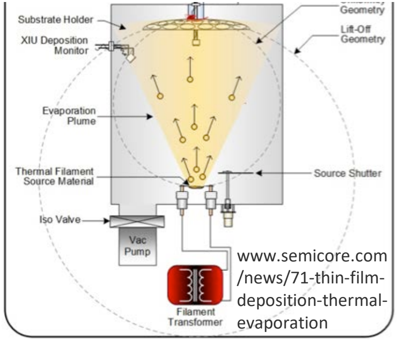
Efficiency >25% should be possible

Typical CdSeTe Absorber Processing

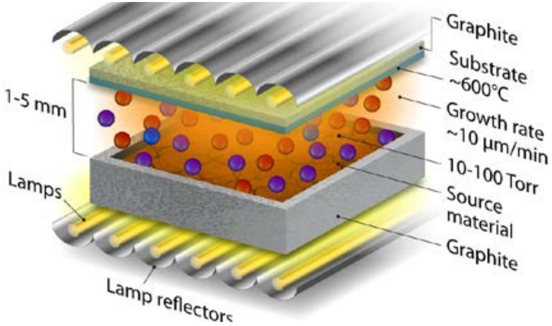
Vapor Transport Deposition (VTD)



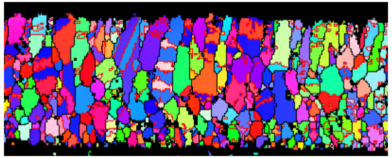
Evaporation

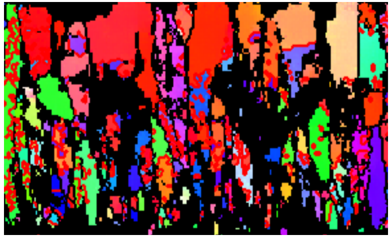


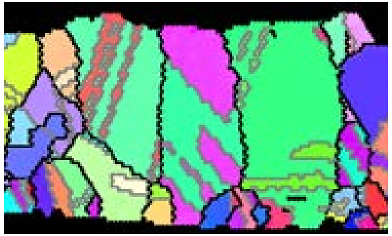
Close Space Sublimation (CSS)



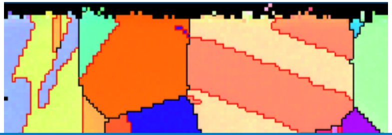
As deposited



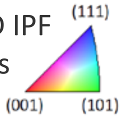




$CdCl_2$



EBSD IPF Maps

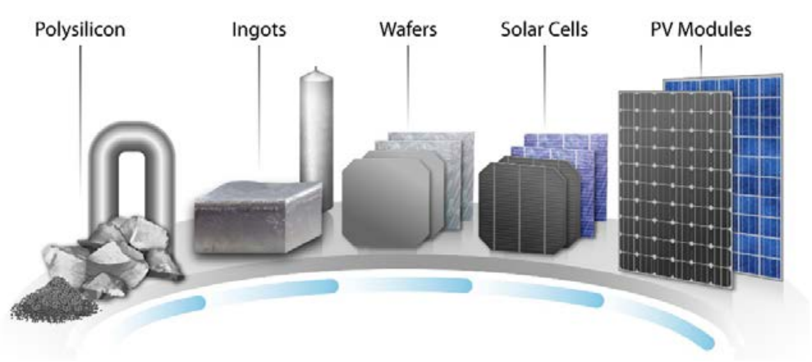


5µm

Small grain poly: 2-5µm grains for 3-6µm films

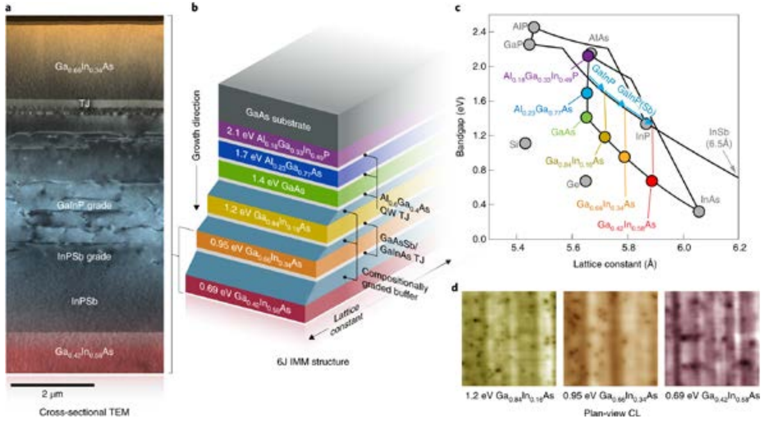
Higher Performance PV → Single Crystals

Silicon:
large grain multi- or single crystal



M. Woodhouse, et al., *NREL* (2018)

III-V:
Single crystal (epi)



J. Geisz, et al., *Nature Energy* (2020)

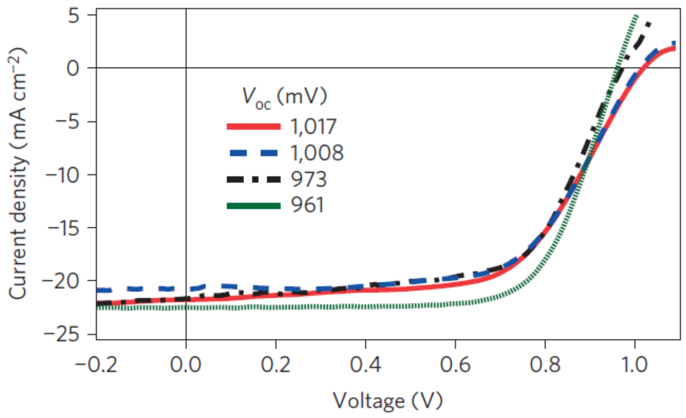
2016: CdTe Single Crystals

Better doping (~10^{16} cm^{-3})

Better lifetime (10-100ns)

Better interface

No $CdCl_2$
(less compensation?)



Burst, et al., *Nature Energy* (2016)

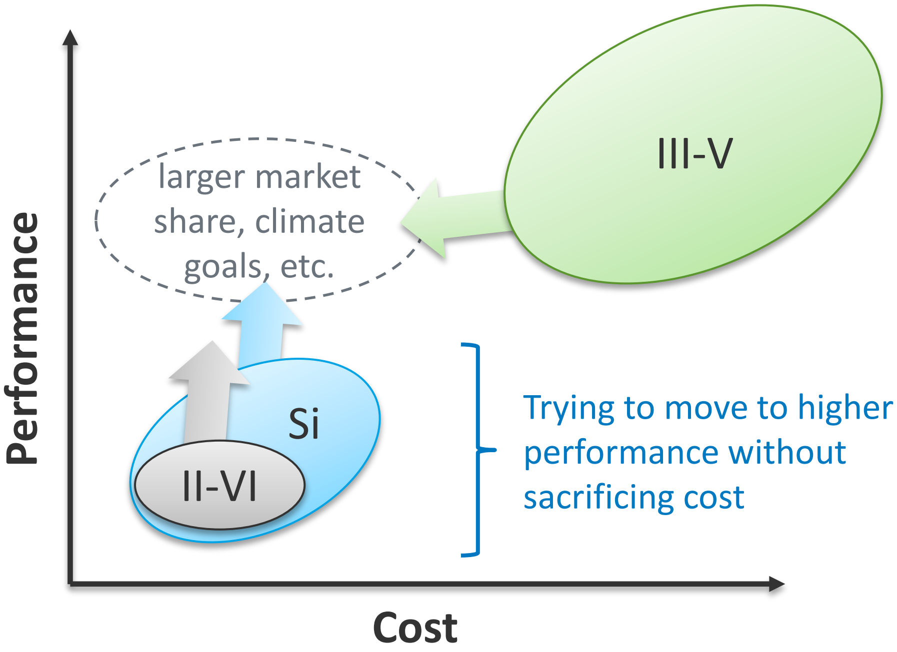
crystals \$250/cm^2

~2.5m^2/module

→ >\$6M/module

How can II-VI PV technology move toward single crystal performance without the associated costs???

Different Approaches – Similar Goal



Trying to move to lower cost without sacrificing performance

Trying to move to higher performance without sacrificing cost

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2 **Fast Epitaxy**

3 Grain size, lifetime, and modeling

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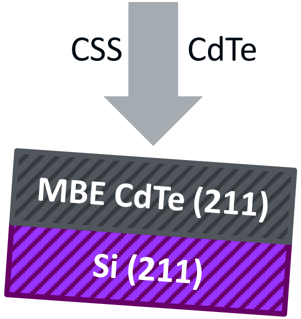
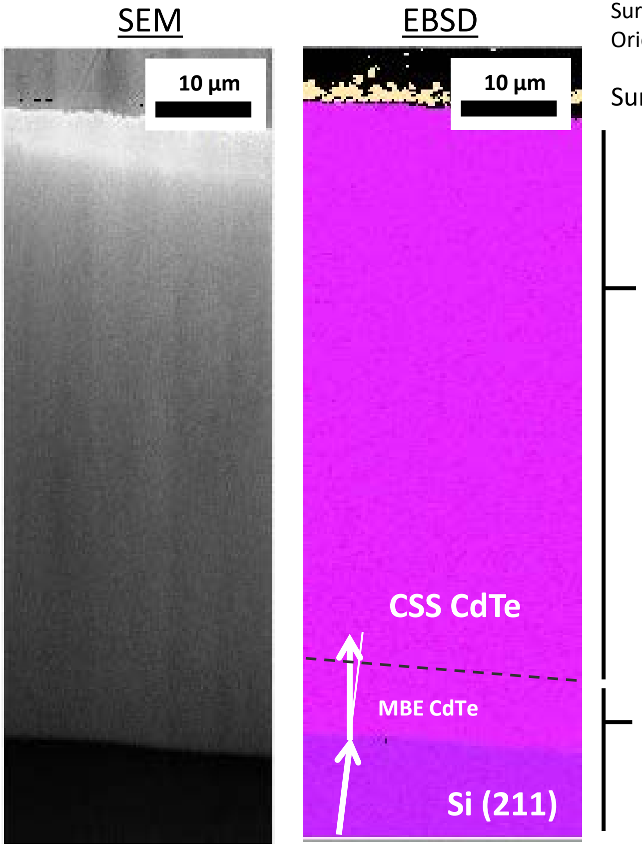
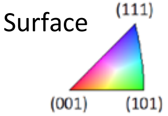
Fast Epitaxy in low-cost CSS

Use a single crystal template in CSS





Surface Normal Orientation



57µm CSS CdTe

Seed Layer

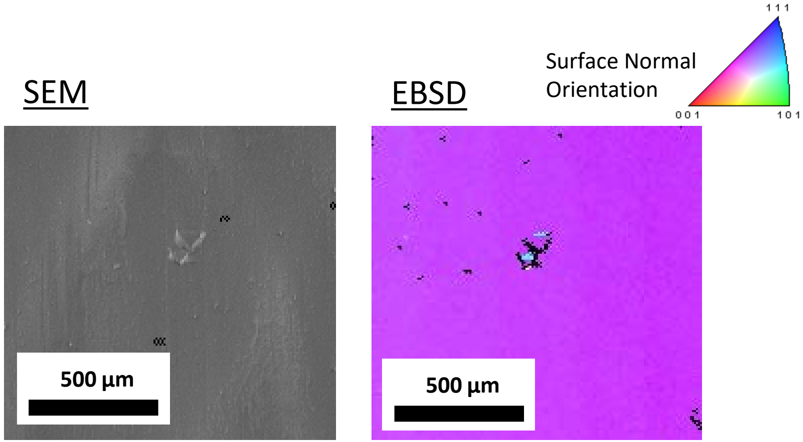
Clean Si/MBE CdTe interface

Seamless MBE/CSS interface

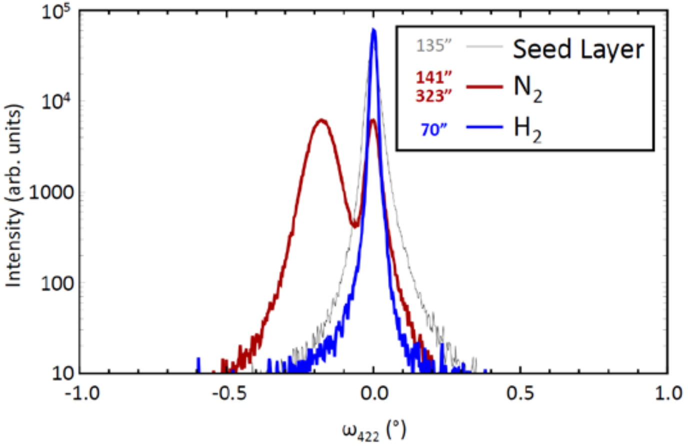
57 µm CSS layer – deposited in 8 minutes

E. Colegrove, et al., *Scientific Reports* (2020)

Large Area Epitaxy in CSS



E. Colegrove, et al., *Scientific Reports* (2020)



High-quality epitaxy possible

Epitaxy maintained over very large areas with high growth rates

CSS has been scaled to 2 ft x 4 ft

… still have the problem of templates.

What size grains to we need?

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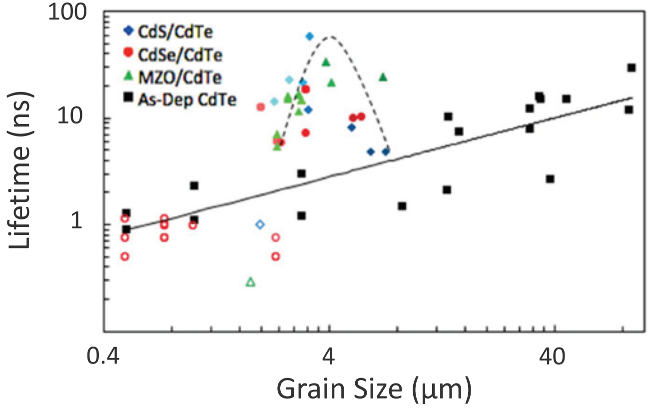
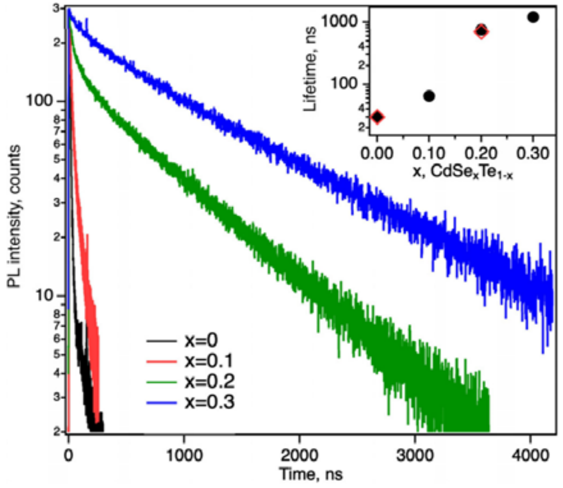
$CdCl_2$, Grain Size, Se, and Lifetime

M. Amarasinghe, et al.,
Advanced Energy Materials (2018)



M. Amarasinghe, et al.,
Applied Physics Letters (2021)



Lifetime and grain size typically improved through $CdCl_2$

6-10µm grains is the best we can do with typical $CdCl_2$

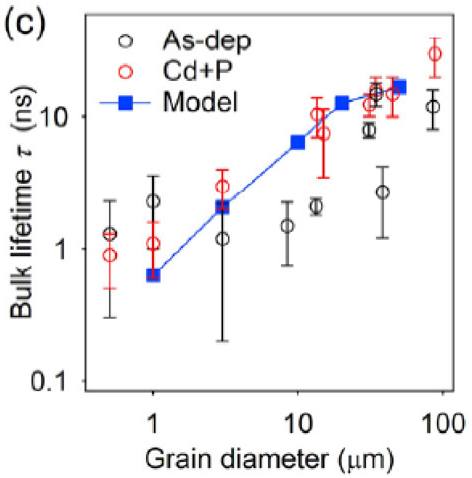
Se alloying also significantly improves lifetime
… still requires $CdCl_2$

Cl-free Grain Size vs Lifetime

Long carrier lifetimes in large-grain polycrystalline CdTe without $CdCl_2$ **(2016)**

Cite as: Appl. Phys. Lett. **108**, 263903 (2016); https://doi.org/10.1063/1.4954904
Submitted: 06 May 2016 . Accepted: 15 June 2016 . Published Online: 27 June 2016

S. A. Jensen, J. M. Burst, J. N. Duenow, H. L. Guthrey, J. Moseley, H. R. Moutinho, S. W. Johnston, A. Kanevce, M. M. Al-Jassim, and W. K. Metzger

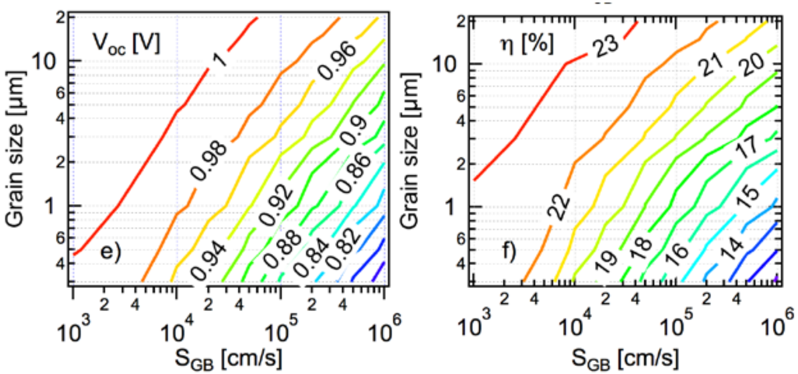


Increasing thickness

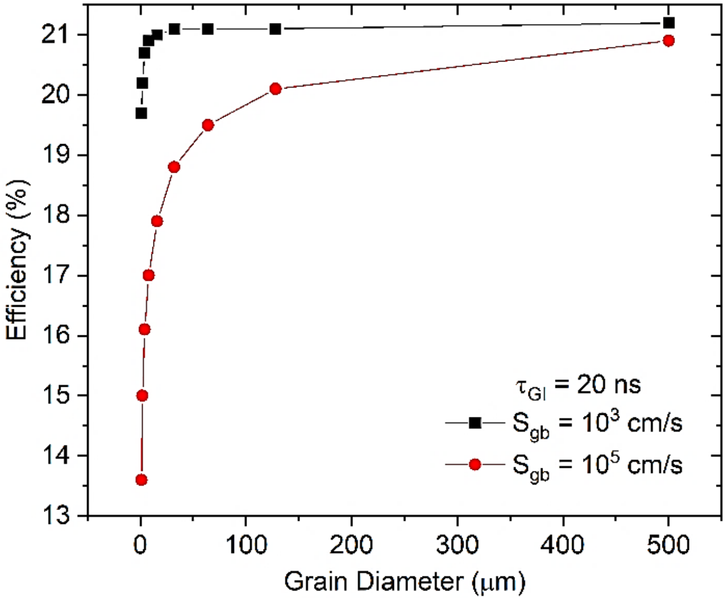
Long lifetime possible with large grains without $CdCl_2$

No $CdCl_2$ modeling

A. Kanevce, et al.,
Journal of Applied Physics (2017)



2021 - Marco Nardone - BGSU

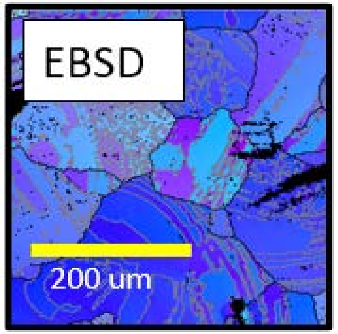


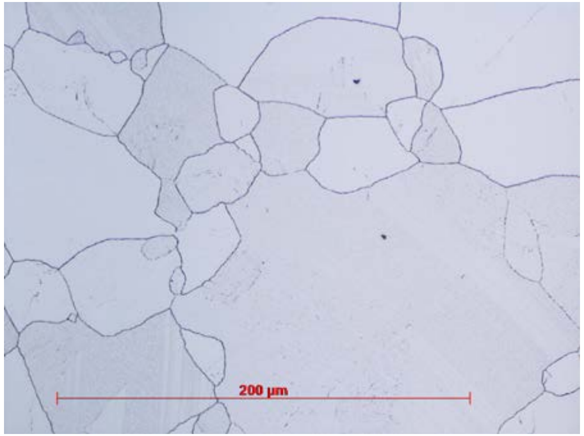
If we want to eliminate $CdCl_2$ grains on the order of 500 µm are necessary

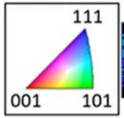
Outline

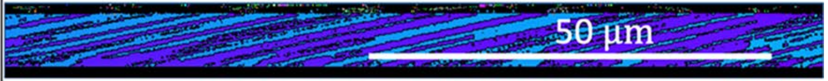
Colossal Grain Growth (CGG)

D. S. Albin, et al.,
Journal of Physics: Energy (2021)





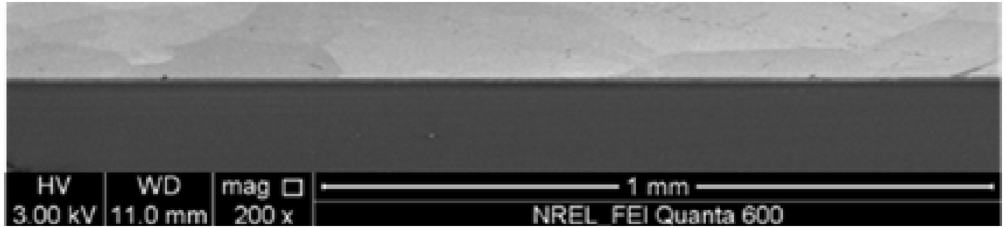




Evaporated $CdSe_{0.1}Te_{0.9}$ films annealed at 500-600C exhibit explosive recrystallization → CGG

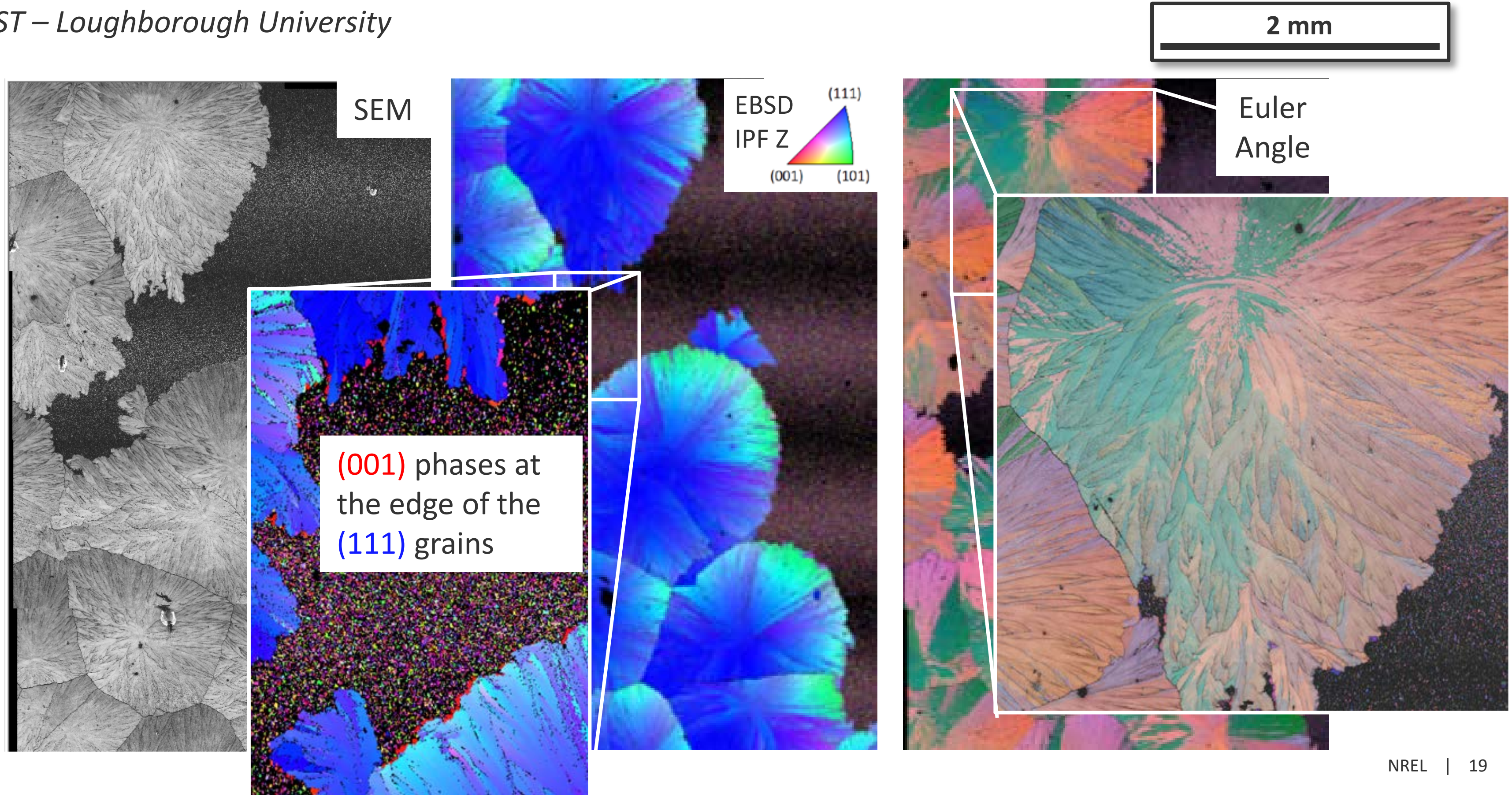
Film thickness and composition maintained

Grain sizes from 100µm to >500µm possible with film <5µm thick



Extreme CGG Provides Propagation Insights

CREST – Loughborough University



Limitations and Challenges related to CGG

1. CGG most prominent in $CdSe_xTe_{1-x}$ film with x=0.1
   - Some CGG observed with x<0.1, but limited
   - No CGG observed for x>0.1

   Best lifetime and device performance for $0.2 < x < 0.3$

2. Initial film thickness needs to be >2 µm

   Best devices use ≤ 1µm CdSeTe layers

   (Items 1–2:) Best devices still require CdTe

3. Surface roughness (R_a) of substrate needs to be ≤ 1nm

   Commercial glass/TCOs have R_a~10-20 nm

Intra-grain defects currently still require Cl passivation

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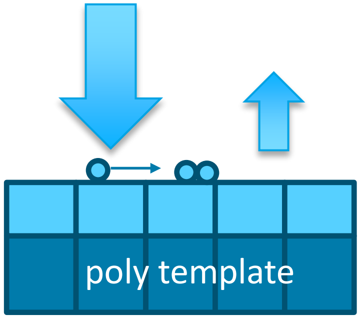
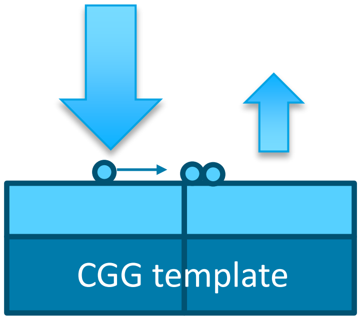
Epitaxy maintained at high rates by CSS

Source flux must exceed re-evaporation (sticking coefficients, adatom mobility, etc.)

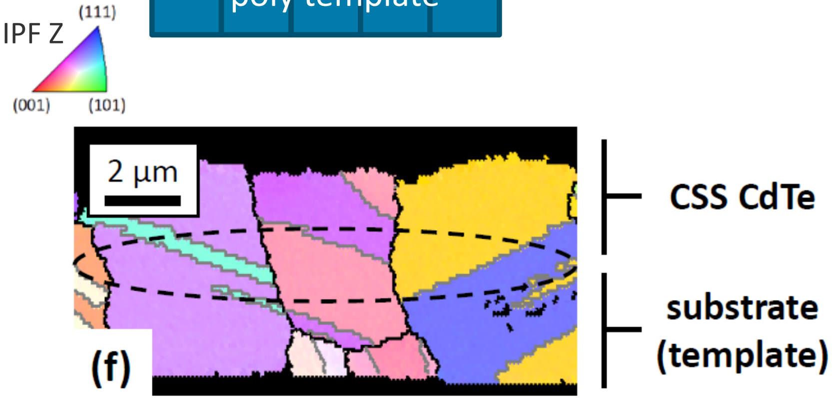
Controlled by:

- Temperatures
- Background pressures
- Chemistry
- Template crystallinity (lattice parameters/orientation)

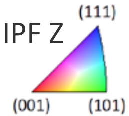


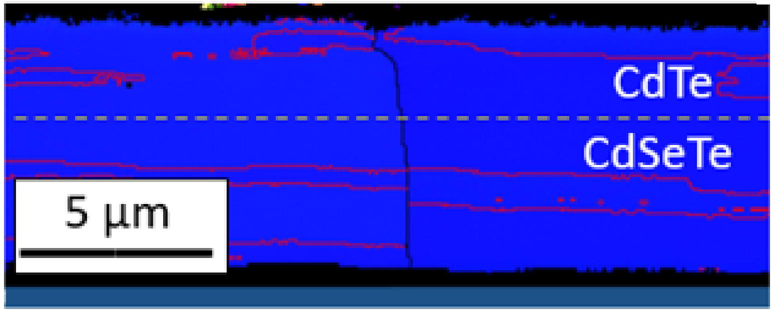


Lattice mismatch <1% for CdTe on $CdSe_{0.1}Te_{0.9}$



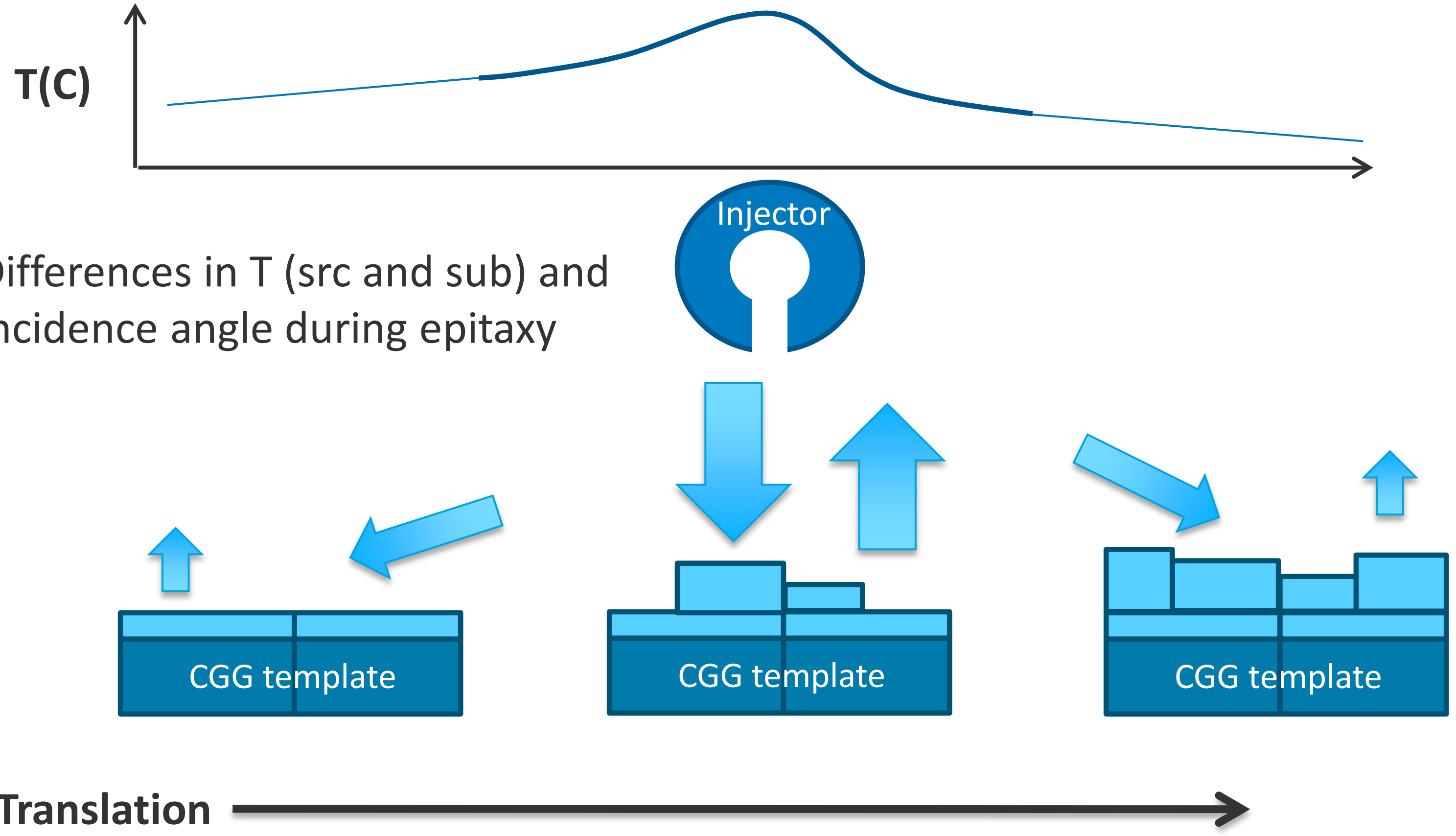
E. Colegrove, et al., *Scientific Reports* (2020)





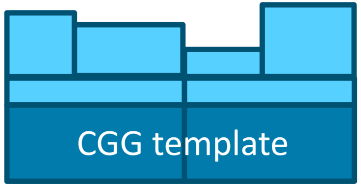
D.S. Albin, et al., *Journal of Physics: Energy* (2021)

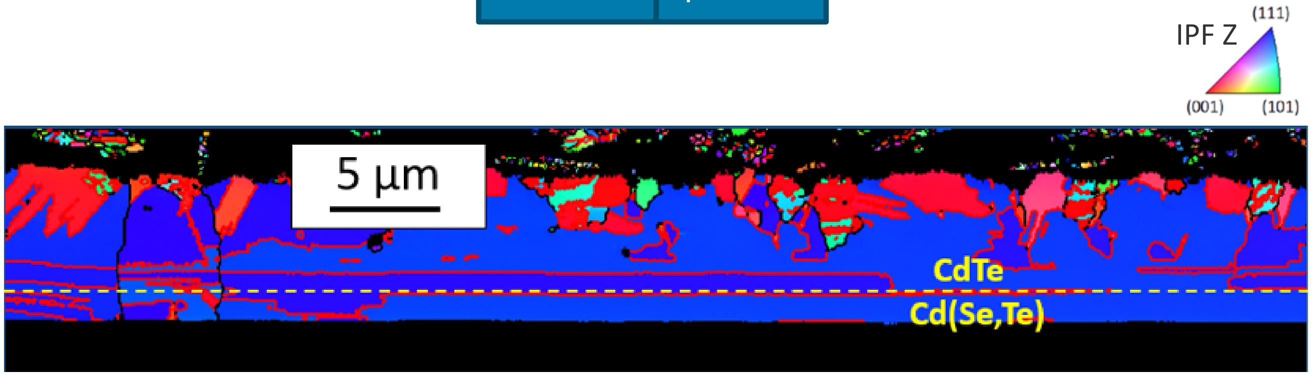
VTD is much more dynamic



Differences in T (src and sub) and incidence angle during epitaxy

VTD is much more dynamic

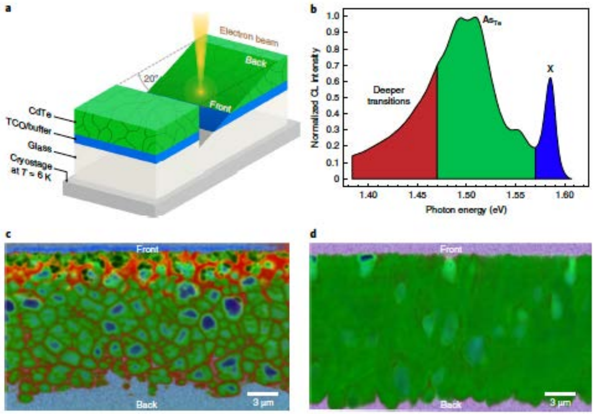


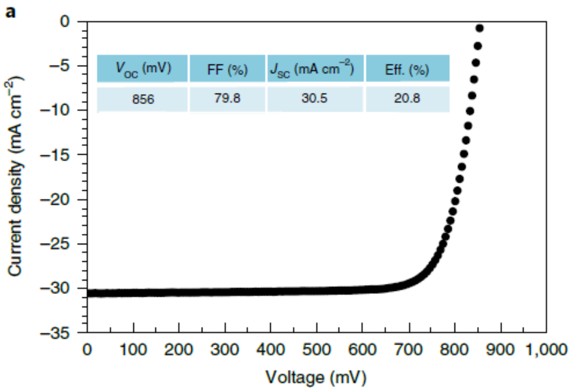


Early deposition results in come epitaxy, but this breaks down in the dynamic process

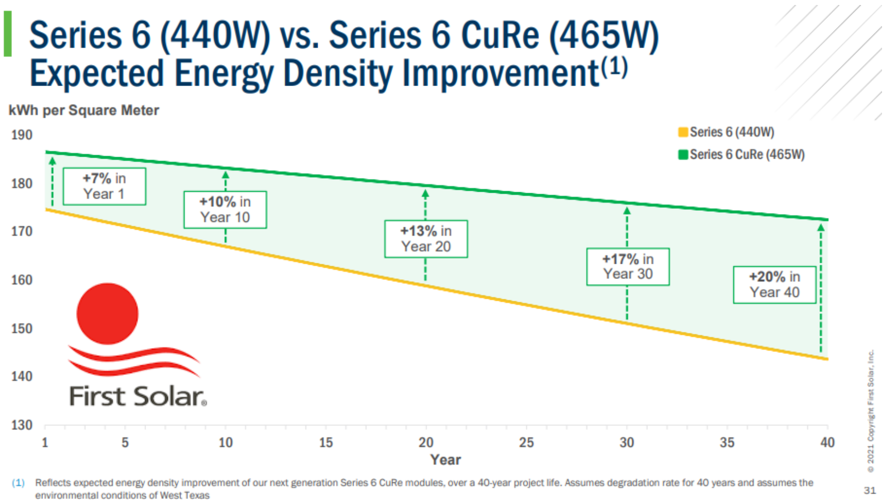
Outline

Current status of As doping





W. K. Metzger, et al., *Nature Energy* (2019)



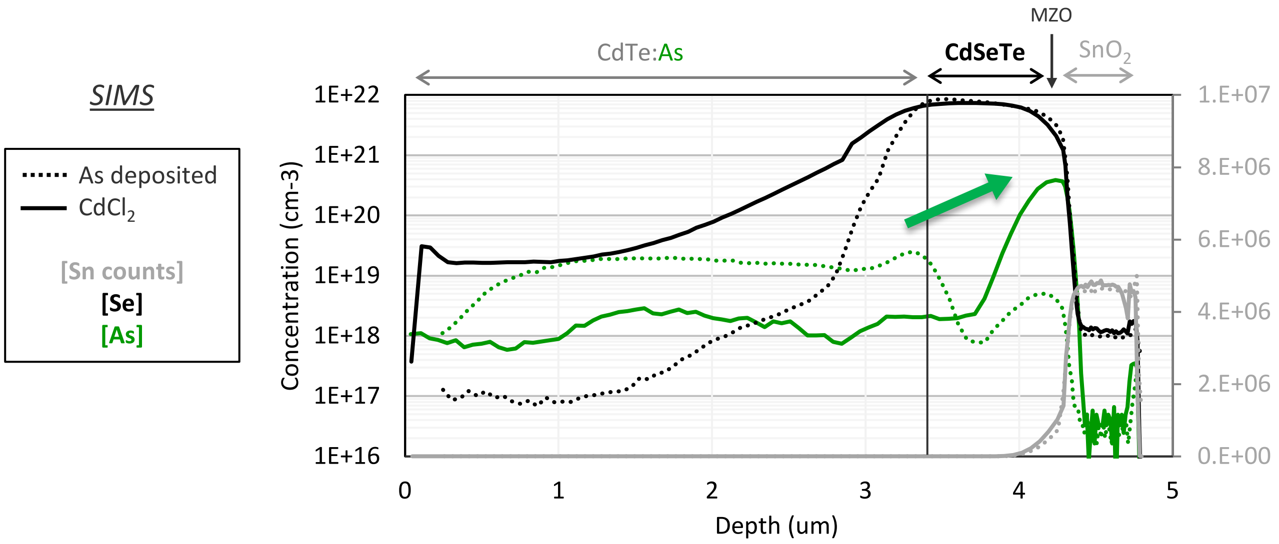
https://s2.q4cdn.com/646275317/files/First-Solar-Investor-Overview-(May-2021)-vF.pdf

As doping has enabled thin film CdSeTe devices with supplier stability
→ **production lines being converted to CuRe (“Cu Replacement”)**

~21% devices with 10^{16} cm^{-3} absorber hole density,
but Voc deficit still present
→ **junction interface issues are the most likely cause**

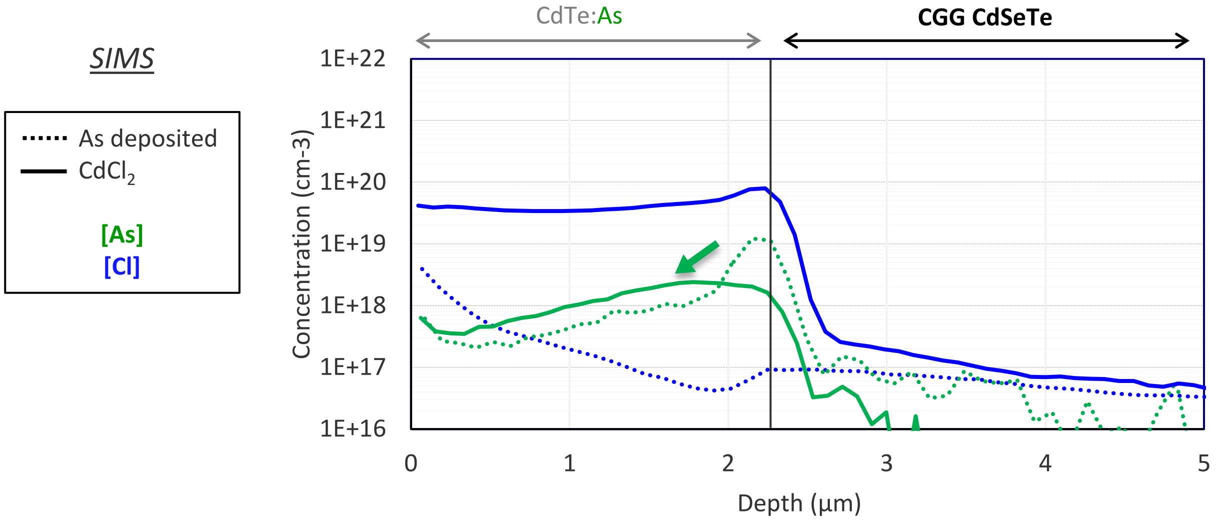
As incorporation and diffusion

- As accumulates at deposition surface followed by lower, uniform, incorporation
- After $CdCl_2$ treatment, As redistributes, diffusing into Se rich region and accumulating at the oxide interface



CGG and diffusion

- As accumulates at deposition surface followed by lower, uniform, incorporation
- After $CdCl_2$ treatment, As diffuses more uniformly through CdTe, but **does not penetrate CGG CdSeTe**



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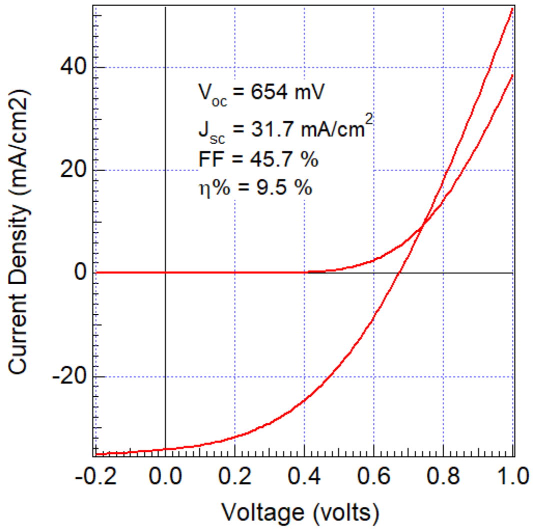
Conclusions

- CGG coupled with fast epitaxy may enable II-VI PV technology to transition from a micro-crystalline structural paradigm into a multi-crystalline regime without the need for $CdCl_2$

- Current limitations of CGG and low intra-grain material quality need to be addressed to realize a direct impact on PV performance



M. Amarasinghe, et al., *Spring MRS 2021*

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Thank You!

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