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# Highly Efficient Monolithic Tandem Solar Cells with Metal-Halide Perovskites

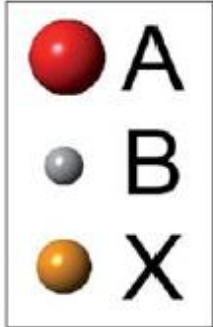
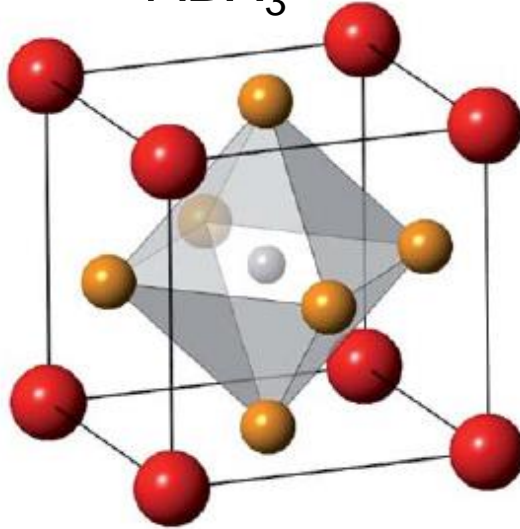
**Prof. Dr. Steve Albrecht + many more**

Investigator Group Perovskite Tandem Solar Cells, Helmholtz-Center Berlin, Germany

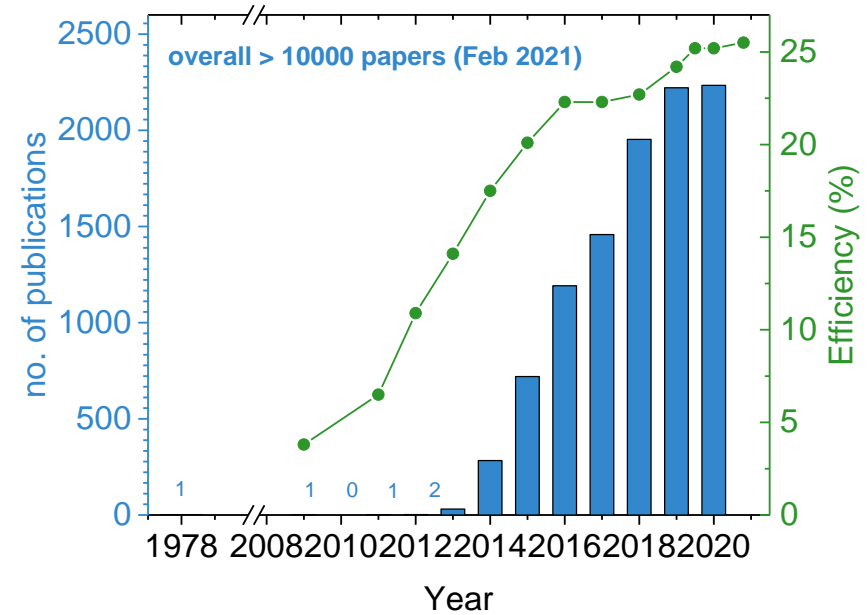
Faculty IV - Electrical Engineering, Technical University Berlin, Germany

**Virtual DPG Meeting “SKM21”**

**Sept. 27, 2021**

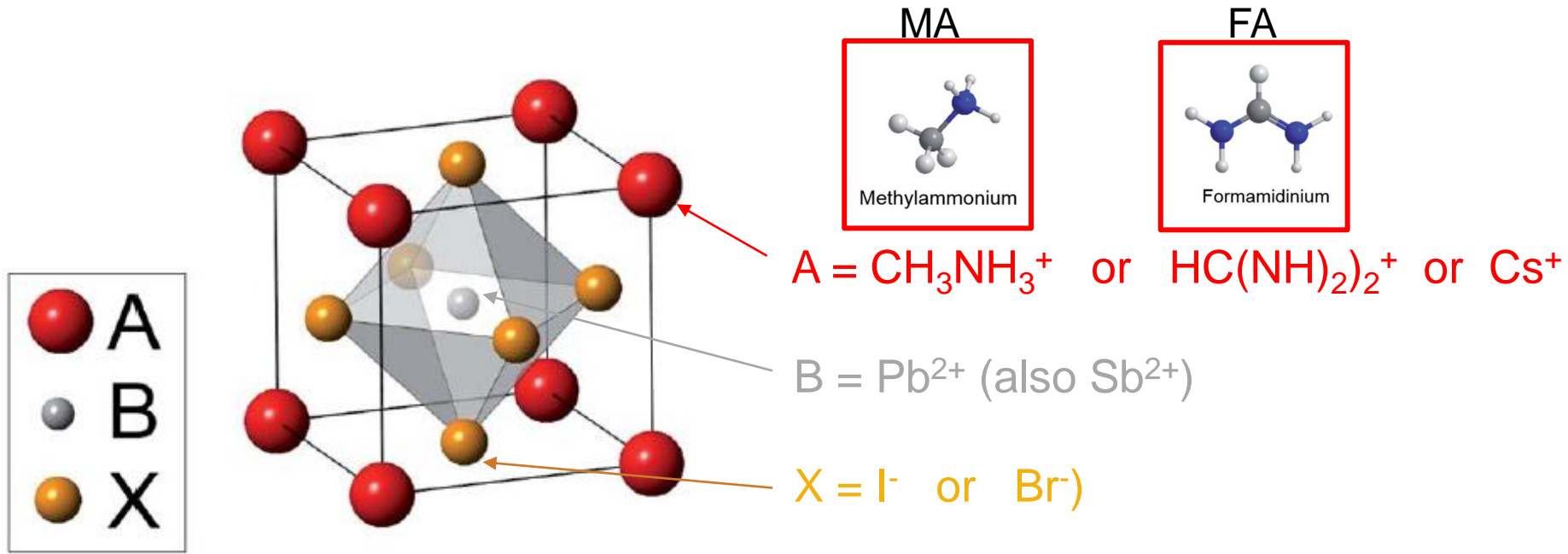


*Snaith et al., Energy Environ. Sci., 2014, 7, 982*



Source: Web of Science  
Search in title „perovskite solar cell“

- First name “perovskite” appeared in 1840 by Gustav Rose for mineral  $\text{CaTiO}_3$
- 2000 perovskites are currently known
- First “organic-inorganic” or “metal-halide” Perovskites by Dieter Weber in 1978
- First application in solar cells late 2008/09 with 3.8%, today over 25% efficiency

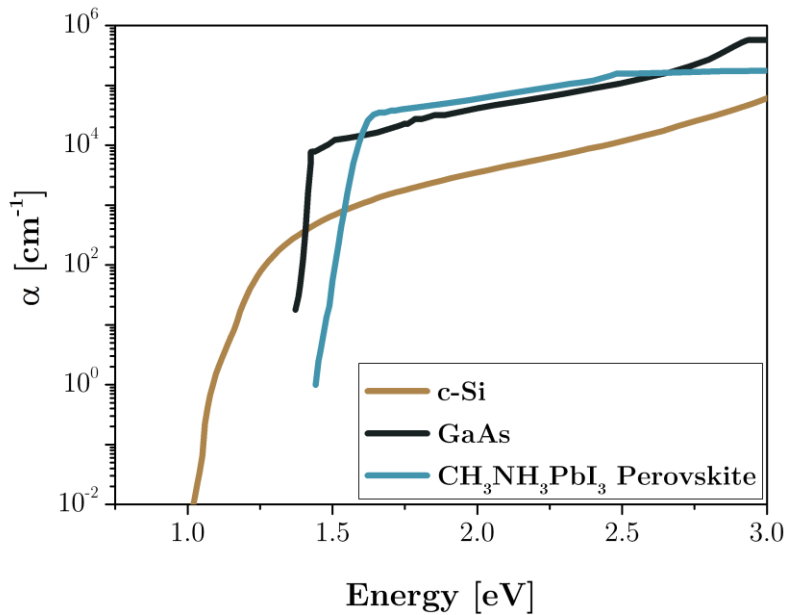


*Snaith et al., Energy Environ. Sci., 2014, 7, 982*

**Methylammonium lead iodide**  
**„Triple cation“**

**$\text{MAPbI}_3$**   
 **$\text{FA}_{0.75}\text{MA}_{0.2}\text{Cs}_{0.05}\text{Pb}[\text{I}_{0.8}\text{Br}_{0.2}]_3$**

*Saliba et al., Energy Environ. Sci., 2016, 9, 1989*



Absorber	$E_g$ (eV)	$\alpha$ ( $\text{cm}^{-1}$ )	Urbach energy (meV)
c-Si	1.1	$10^2$	11
GaAs	1.4	$10^4$	7
CIGS	1.1	$10^3$ - $10^4$	25
CdTe	1.5	$10^3$	10
<b>MAPbI<sub>3</sub></b>	<b>1.55</b>	<b><math>10^3</math>-<math>10^5</math></b>	<b>15</b>

## Organic-inorganic metal halide Perovskites

- Direct, bandgap  $\sim 1.55$  eV for MAPbI<sub>3</sub>
- tunable bandgap via composition
- Very few sub bandgap features
- High absorption  $\rightarrow$  thin layers  $< 1 \mu\text{m}$  required

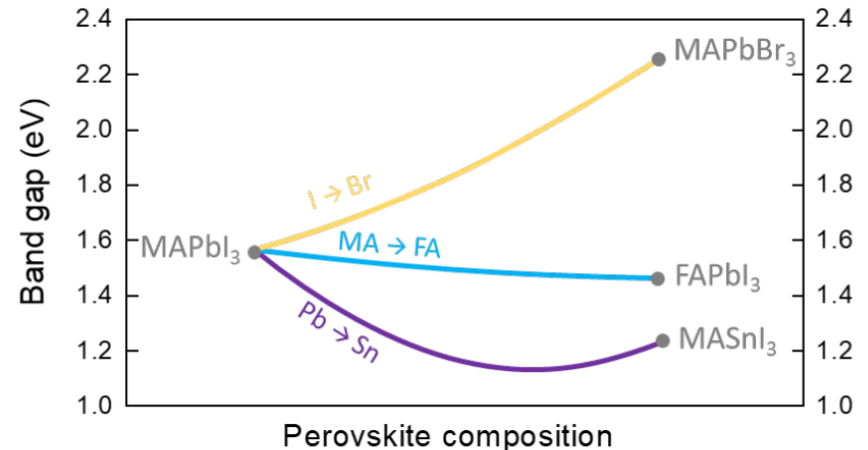
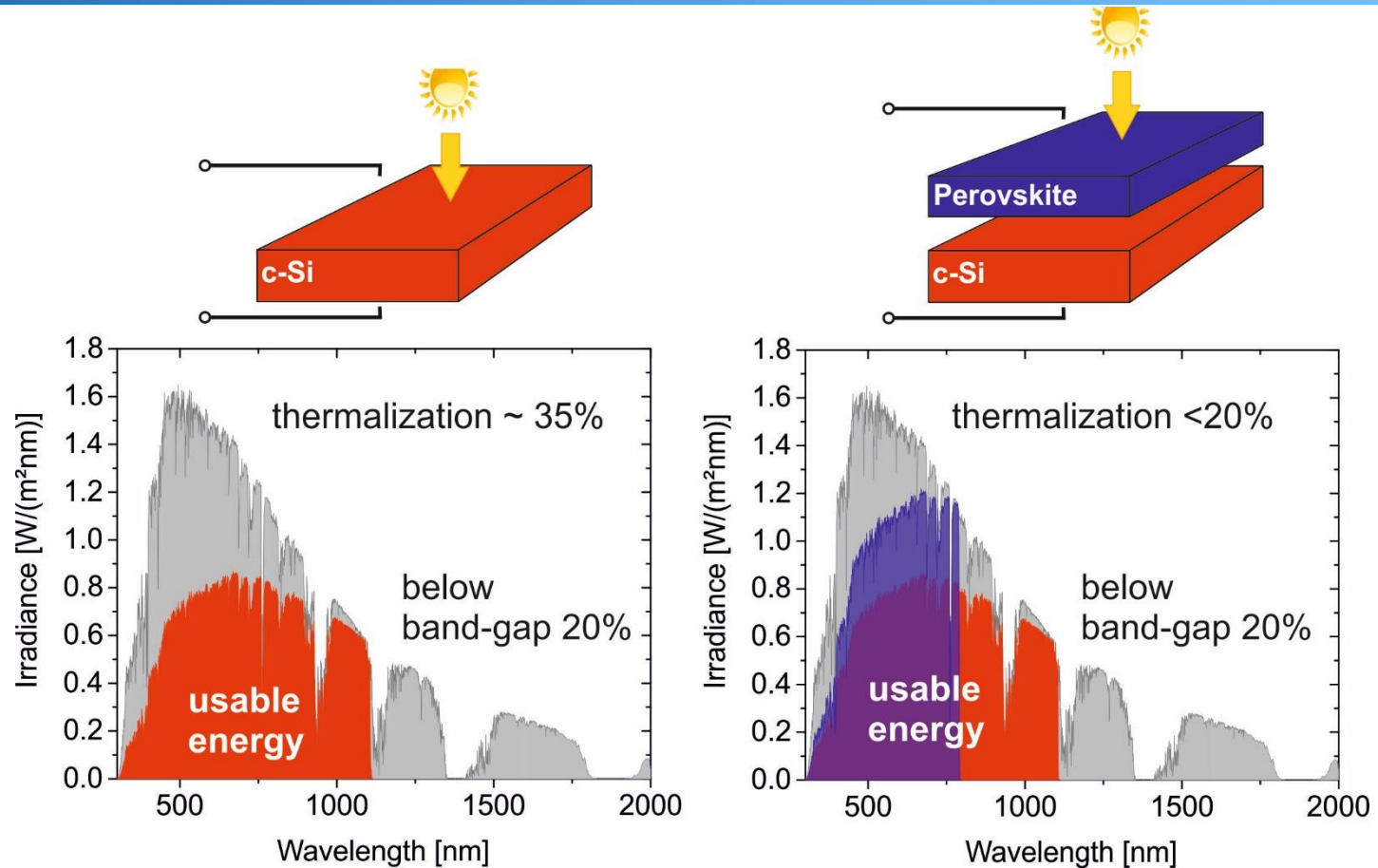
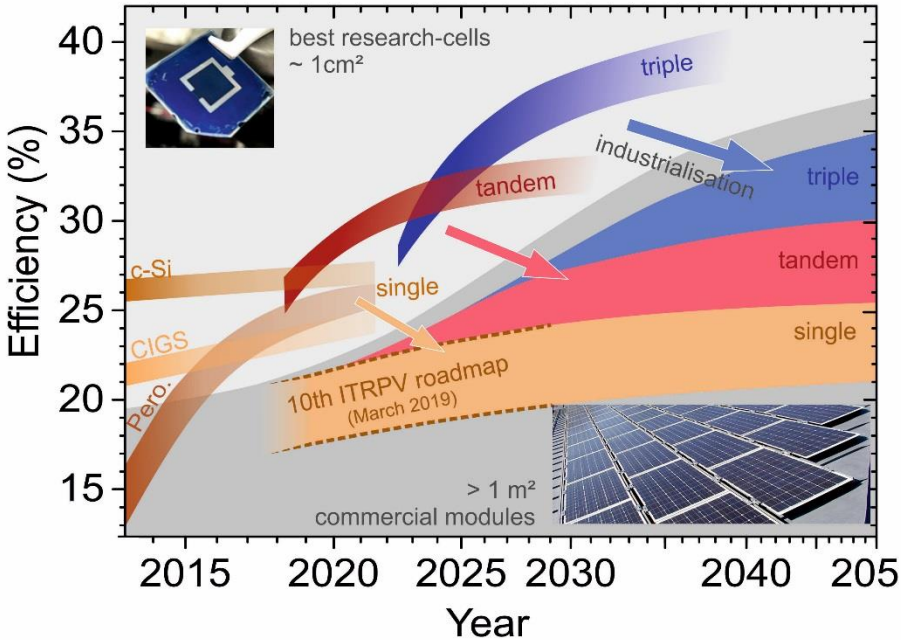


Image courtesy: Selina Olthof (Uni. Cologne)

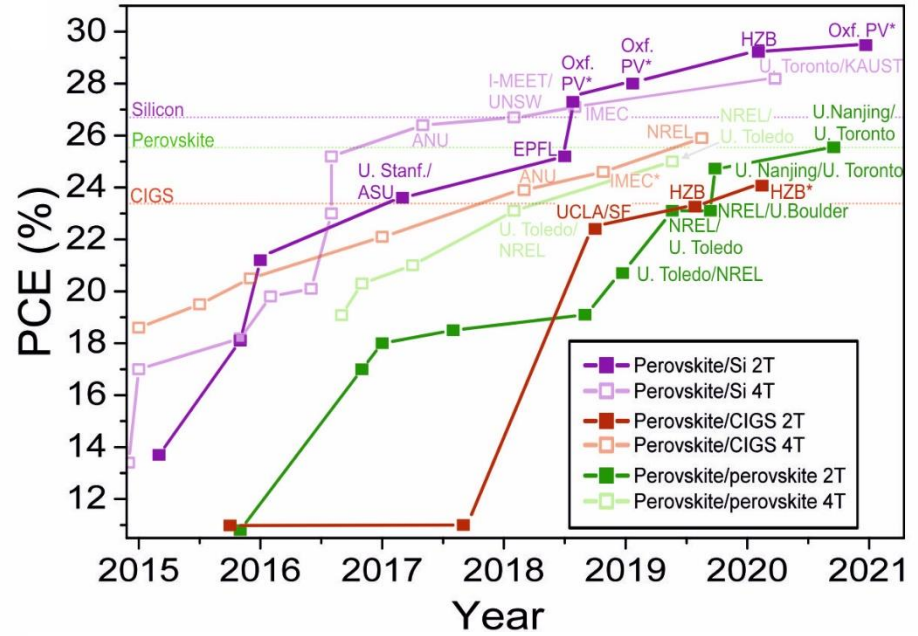


- high loss from thermalization

- high energy photons are absorbed by perovskite
  - converted at a high voltage
  - reduced losses from thermalization
- infrared photons are transmitted into c-Si cover a wide spectral range of absorption



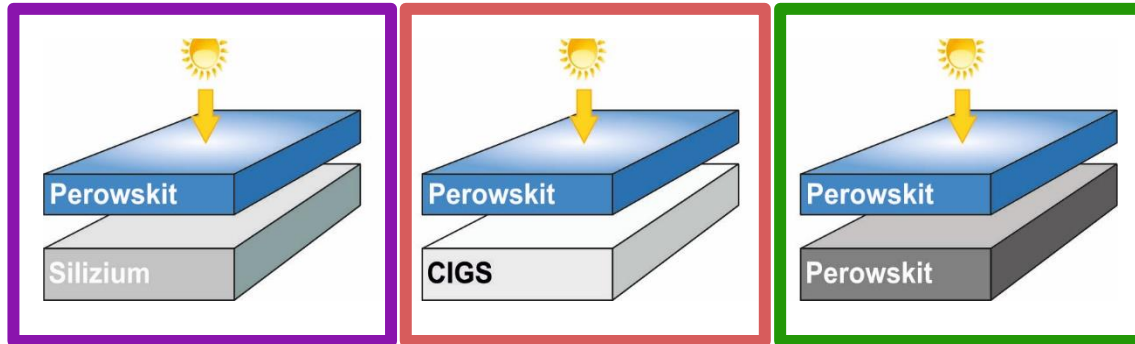
- Single junctions will be limited in efficiency
- Multi-junctions with perovskite top cells can overcome fundamental limitations



- Main 2T achievements:
- 29.5% record in Dec. 2020
  - Higher than best Si single junction!
- After long time w/o results: Recently promising results 24.2% with Perovskite/CIGS tandems
  - Similar to best CIGS single junction!
- Improvements in Sn-based Perovskites enabled 24.2% Perovskite/Perovskite tandems

certified, > 1cm<sup>2</sup>

Jost et al., *Advanced Energy Materials*, 2020,  
DOI: 10.1002/aenm.201904102



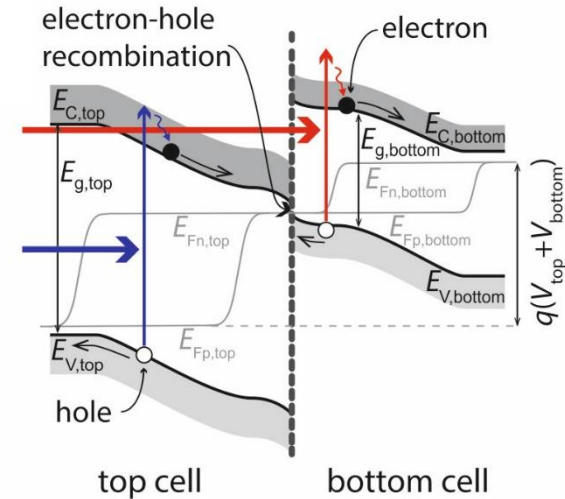
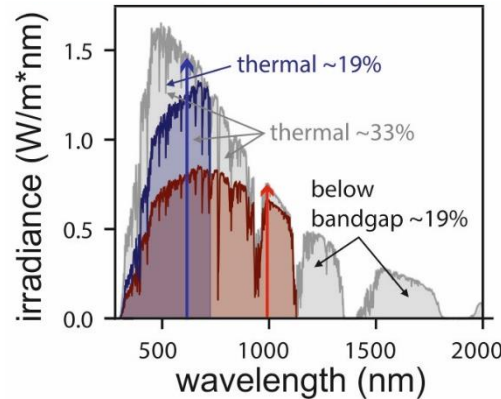
- 1. Results on Perovskite/Silicon Heterojunction (SHJ) Tandems**
- 2. Results on Perovskite/CIGS Tandems**
- 3. Towards all-Perovskite Tandems**
- 4. Upscaling of Perovskite-based Tandems**

# Results on Perovskite/SHJ Tandems



In series connected tandem:

- 2 absorption events ↑ ↑
- 2 electron-hole pairs
- Recombination layer needed



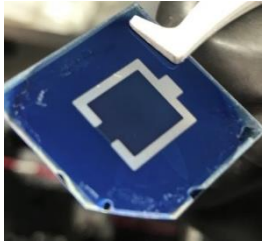
www.istockphoto.com



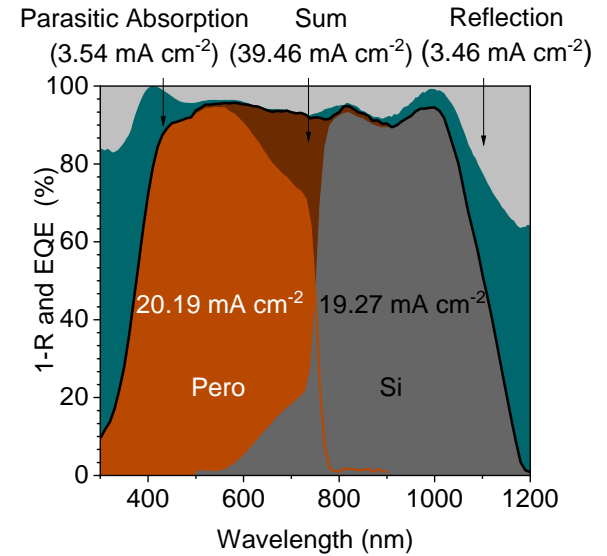
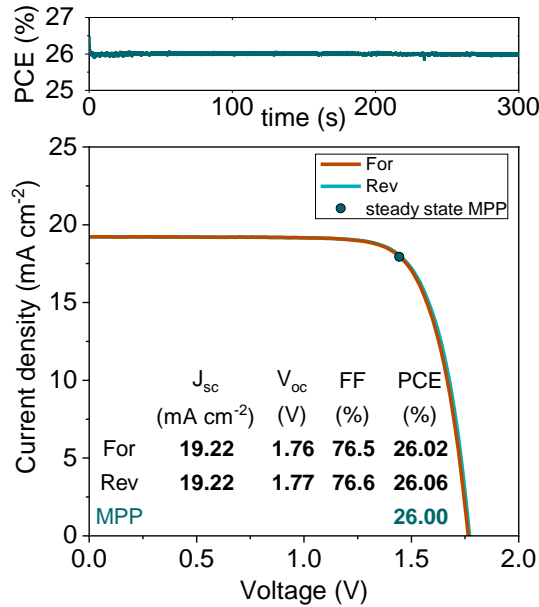
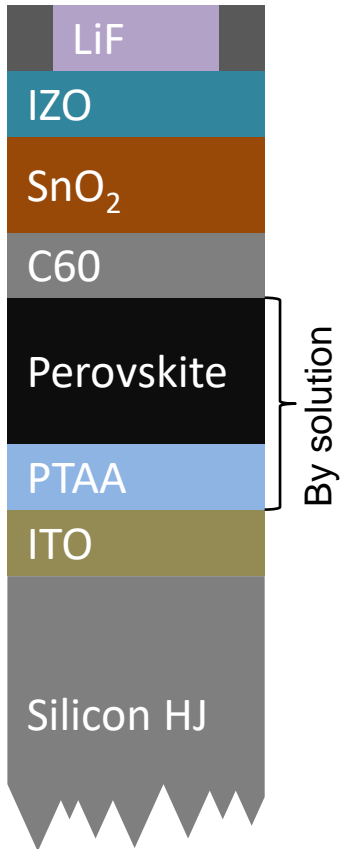
In series connected tandem:

- Voltages add up
- Current limited by limiting sub-cell
- Current matching needed

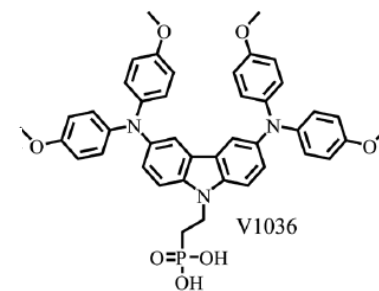
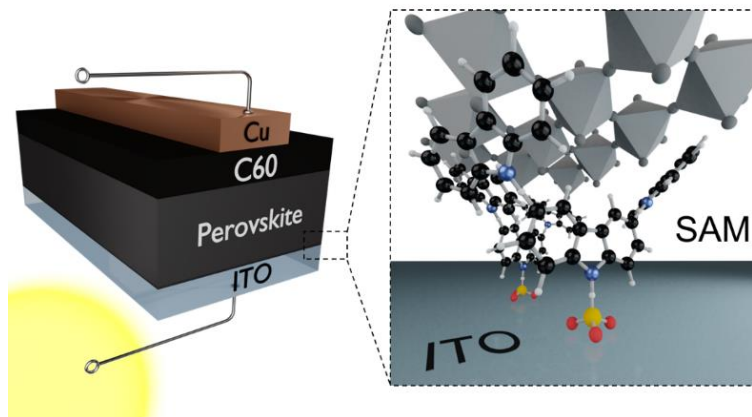
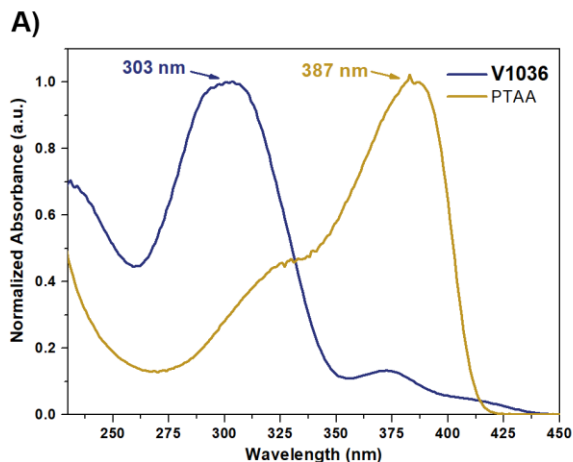
~1 cm<sup>2</sup>



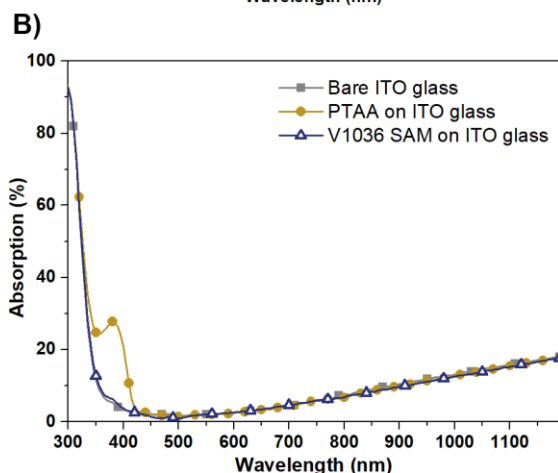
Overall, ~ 15 layers!



- Perovskite band gap ~ 1.63 eV, Voc ~1.1V
- HTM for perovskite Top-cell was polymer PTAA
  - Limited Voc due to interface recombination and non-optimized band gap
  - Limited FF as well!



„SAM 1“



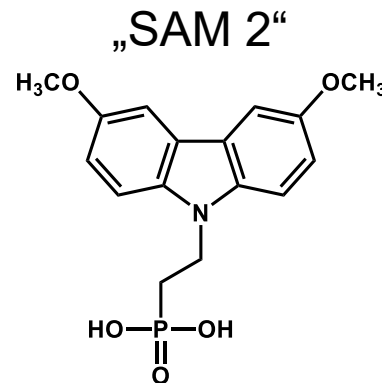
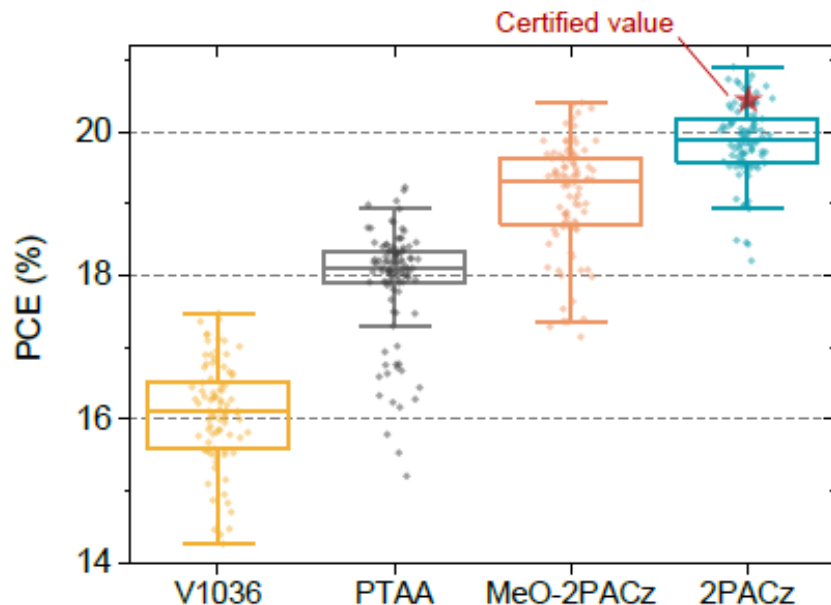
- Covalent bonding to ITO, robust against solution processing
- Reduced absorption loss
- Close to 18% PCE in p-i-n perovskite solar cell

SAM 1st Gen

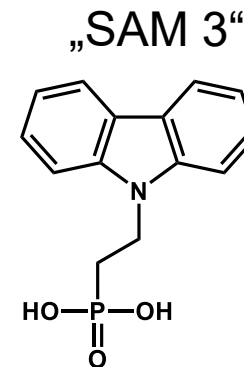
Standard

SAM 2nd Gen

SAM 2nd Gen



**MeO-2PACz**

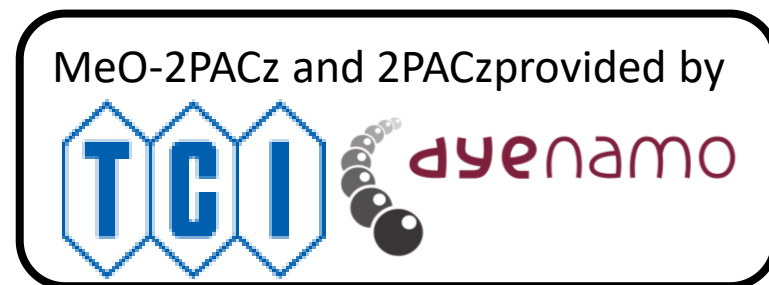


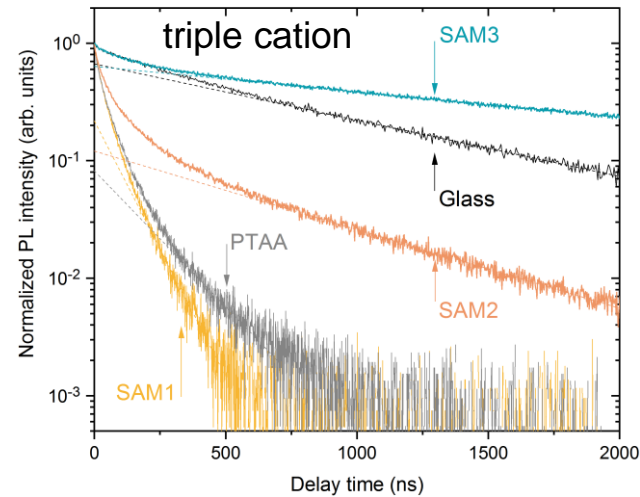
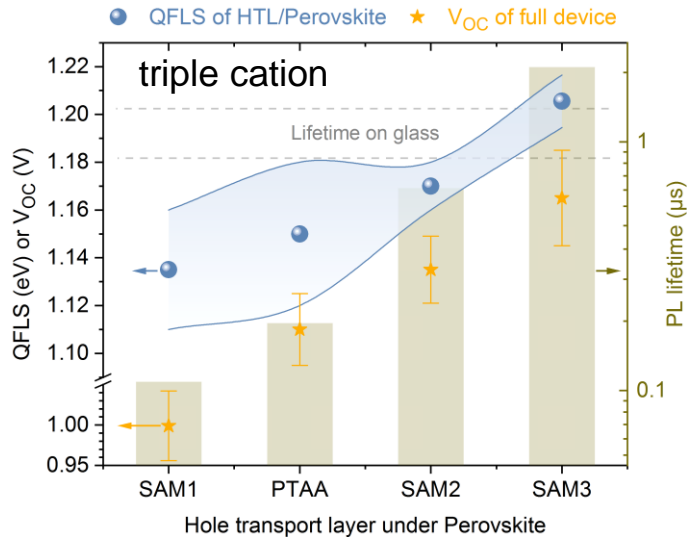
**2PACz**

- Terminated by methoxy groups
- Simplest possible Carbazole SAM

Al-Ashouri et al., *Energy Environ. Sci.*, 2019,  
Advance Article DOI: 10.1039/C9EE02268F

Patents : DE 10 2018 115 379.1,  
PCT/EP2019/060586, DE 10 2019 116 851.1





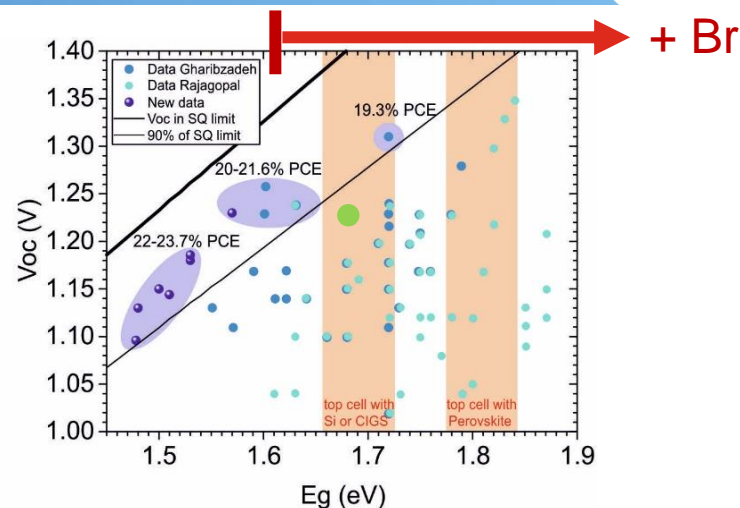
New SAMs 2 and 3 with superior properties:

- Higher PL lifetimes and QFLS and Voc
  - › SAM 3: higher than quartz glass (passivation)
- Perfect energetic alignment

*Al-Ashouri et al., Energy Environ. Sci., 2019, Advance Article DOI: 10.1039/C9EE02268F*

## Problem:

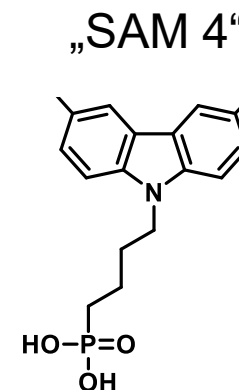
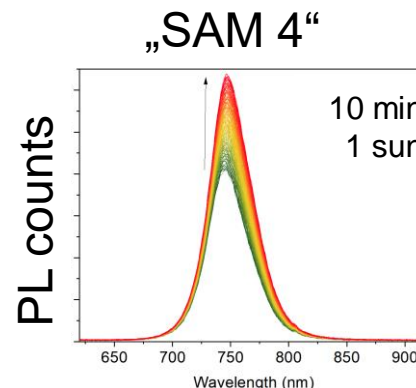
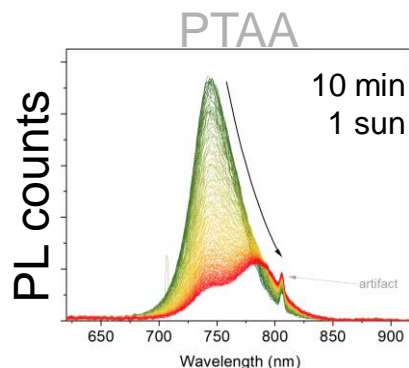
- Highest single cell efficiency with 1.5 eV
- For 1.7 eV compositions
  - › Limited photo-stability
  - › Phase segregation
  - › Non-optimized contacts



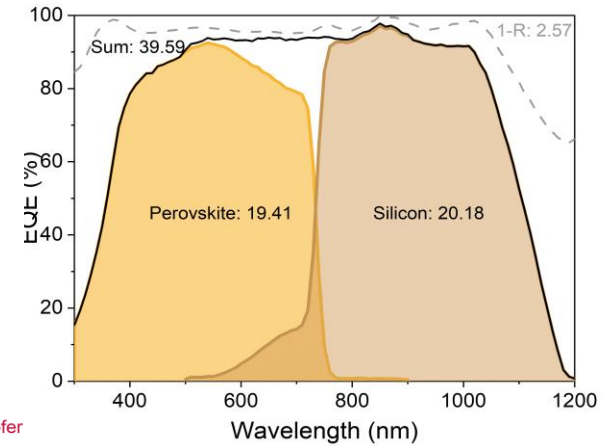
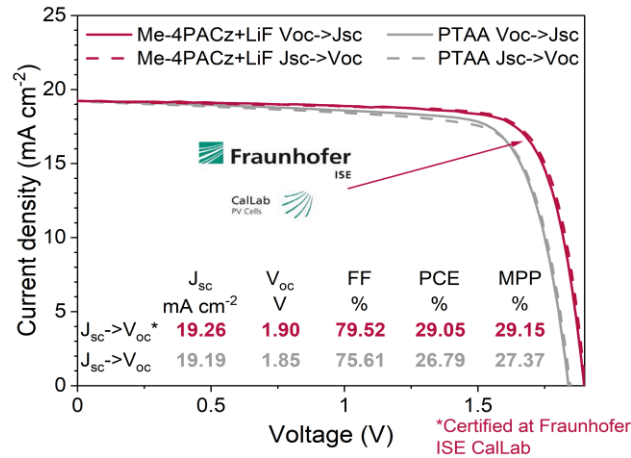
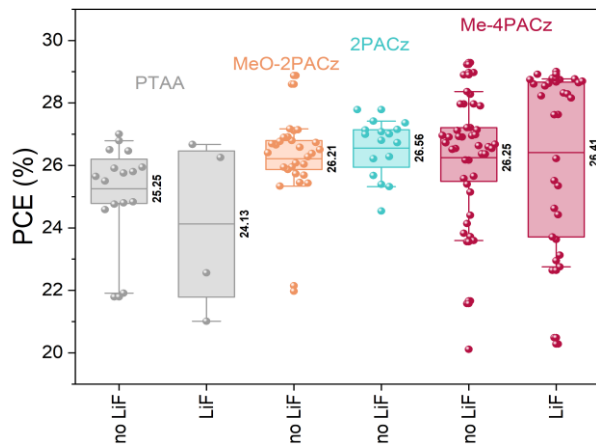
Jost et al., *Adv. En. Mater.*, 2020, DOI: 10.1002/aenm.201904102

## New SAM = Me-4PACz:

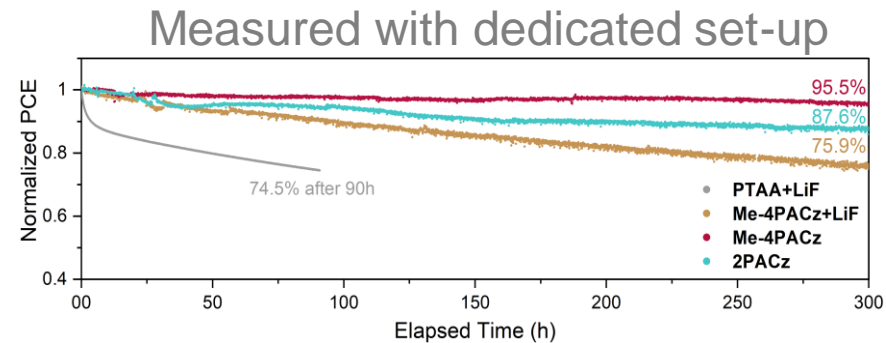
- Efficient passivation
  - › High QFLS → High  $V_{oc}$
- High photo-stability
  - › Reduced phase segregation



Al-Ashouri, Köhnen et al, *Science* (2020), DOI: 10.1126/science.abd4016



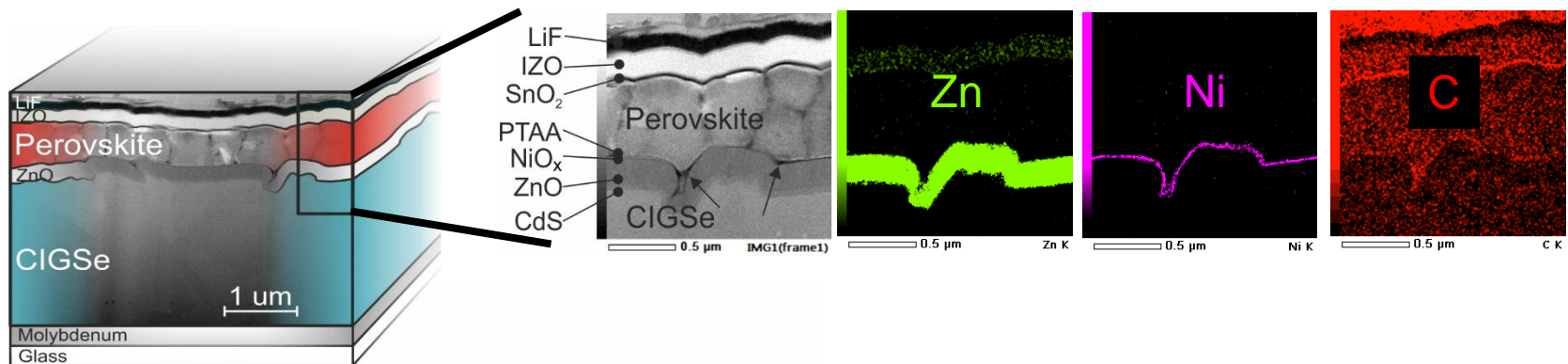
- Fine-tuning and stabilizing of Perovskite band gap to  $\sim 1.68$  eV
- Enhanced hole extraction for higher FFs
- Slightly Perovskite limited in photocurrent
- **Record 29.15% efficiency ( $>1$  cm<sup>2</sup>)**
- **Promising stability data (300 h)**



# Results on Perovskite/CIGS Tandems



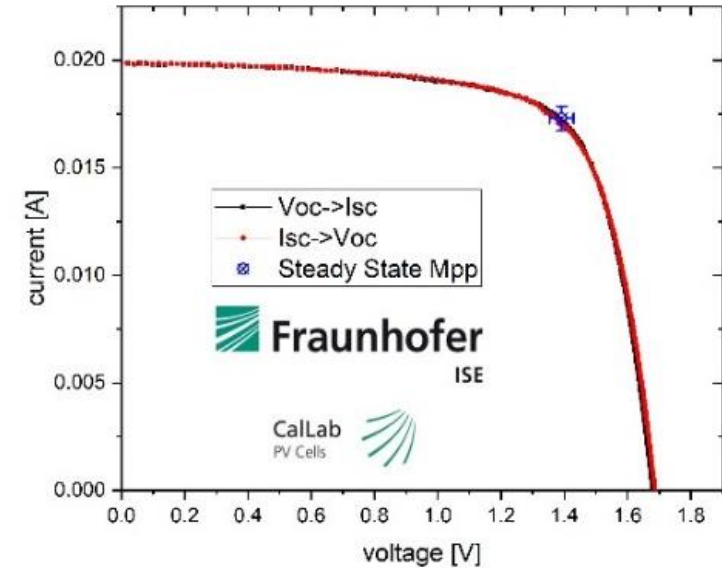
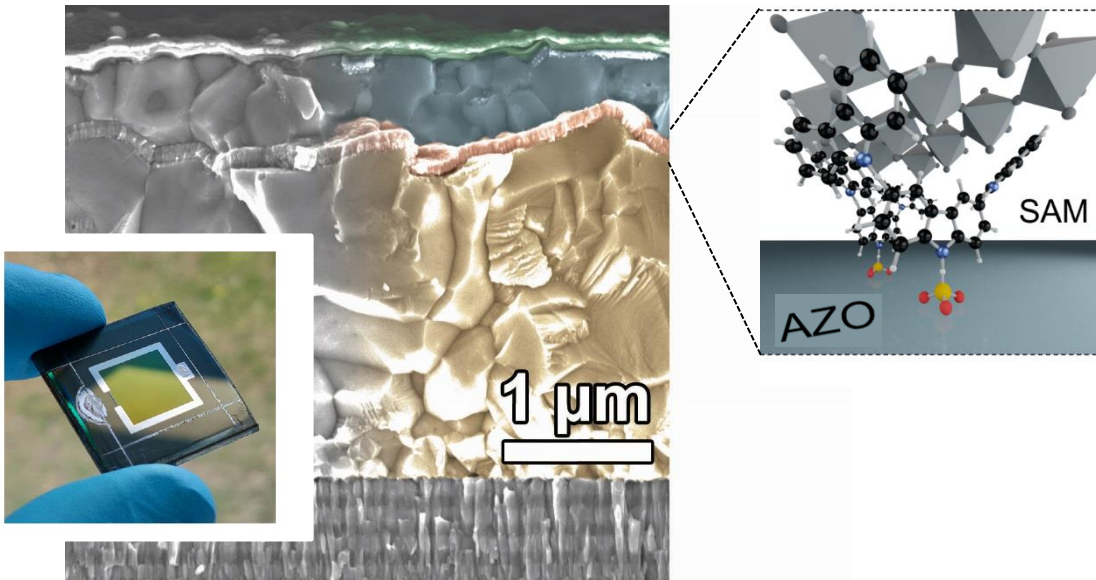
- So far <5 publications on monolithic pero/CIGSe tandems
  - › Rough CIGS surface induces problems with spin-coating of top cell
  - › Recently, polished TCO surface was implemented by UCLA and Solar Frontier [1]
- Here, we use conformal ALD layers of NiOx as hole contact [2]



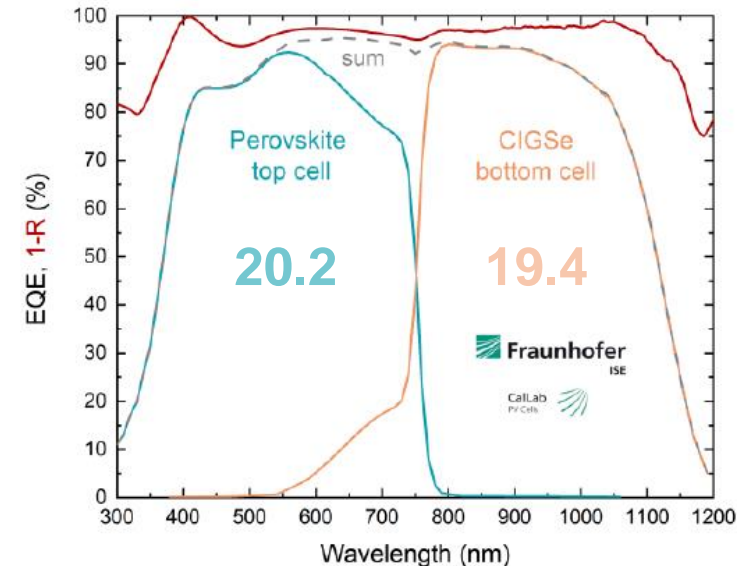
- ALD NiO<sub>x</sub> prevents shorts on rough surface
- Partial coverage with PTAA enables improved Voc and FF
- 21.6% stabilized efficiency (0.8 cm<sup>2</sup> active area)

[1] Han et al., *Science* 2018, 361, 904

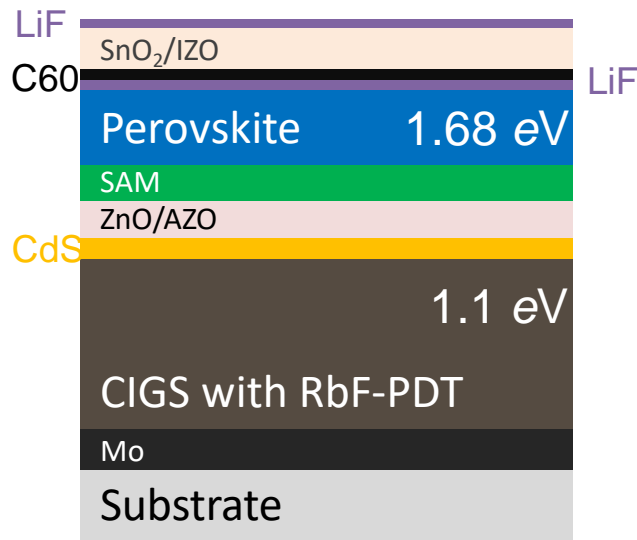
[2] Jost et al., *ACS Energy Lett.* 2019, 4, 2, 583



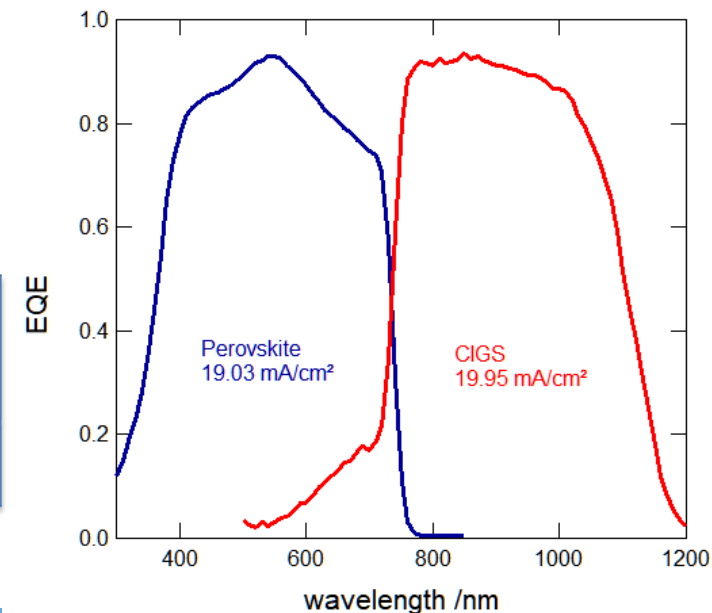
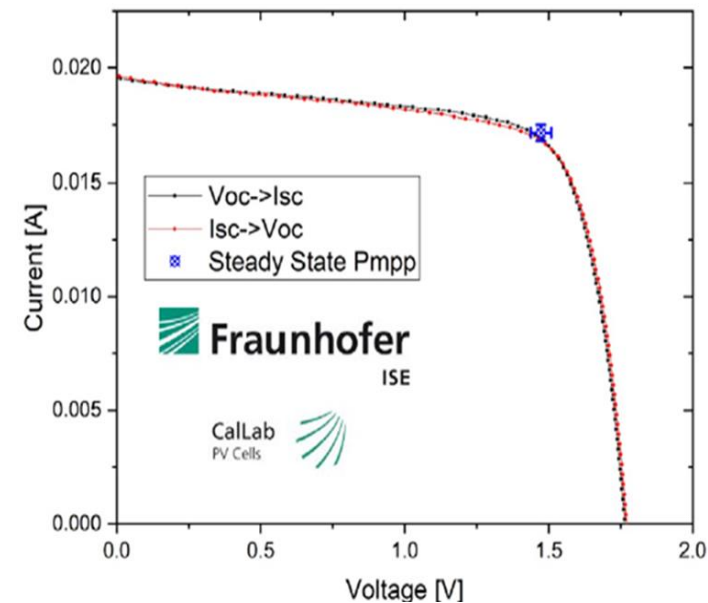
- 2<sup>nd</sup> Gen SAM integrated directly on top of rough CIGSe
- Optimized optics and SAM-improved Voc



Area (cm <sup>2</sup> )	MPP (%)	Jsc (mA/cm <sup>2</sup> )	Voc (V)	FF (%)
1.035	23.26	19.17	1.68	72

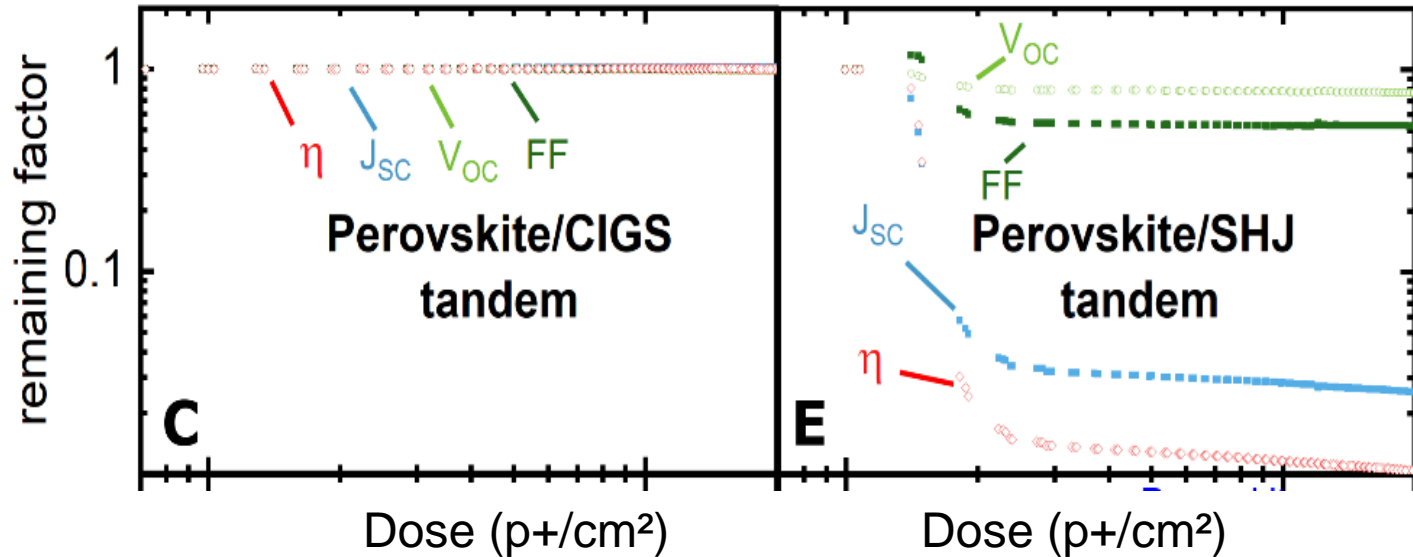


- 3<sup>rd</sup> Gen SAM integrated directly on top of rough CIGSe
- Optimized perovskite band gap - higher Voc



Area (cm <sup>2</sup> )	MPP (%)	Jsc (mA/cm <sup>2</sup> )	Voc (V)	FF (%)
1.045	24.16	18.8	1.77	72

*Jost et al, manuscript in prep.*

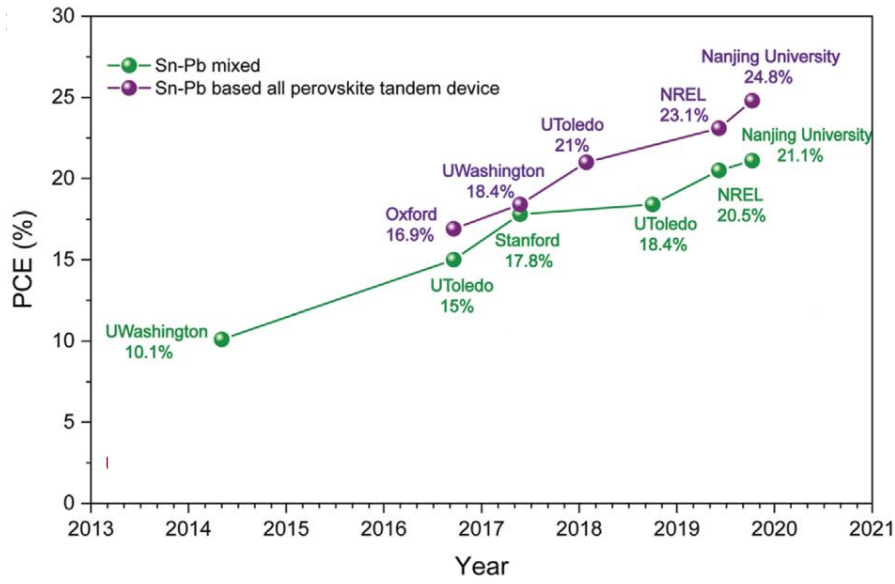


- Pero/CIGS is stable under proton dose of >50 years at the ISS orbit!

Lang, et al., Joule, (2020),5, 1054

Lang et al., Advanced Materials (2016), 28, 8726

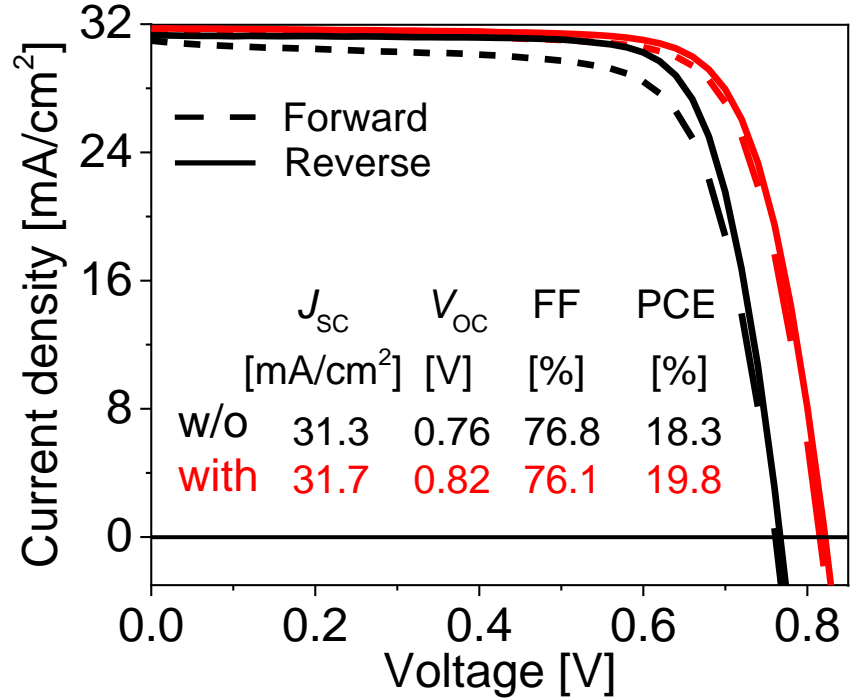
# Towards all-Perovskite Tandems



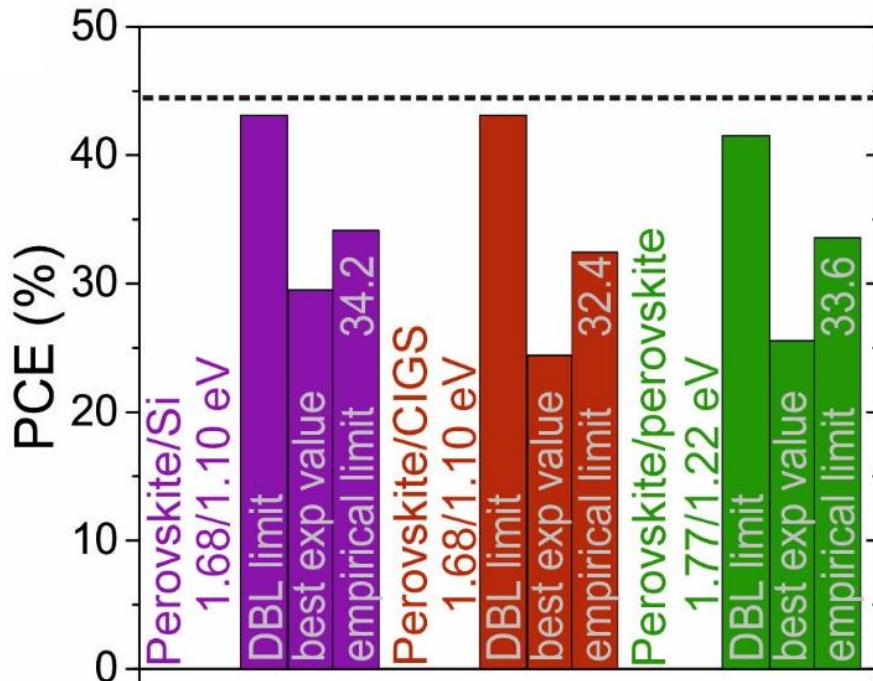
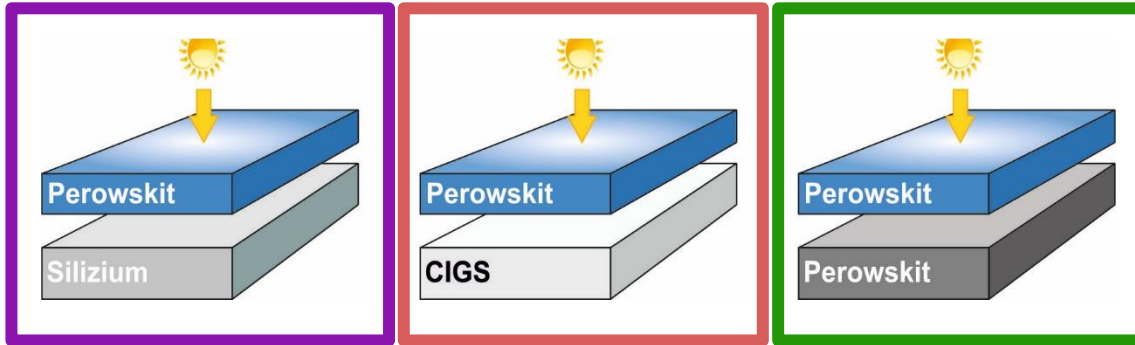
S, Gu, et al. *Adv. Mater.* **2020**, 1907392.

- New additive for Sn-Pb perovskites
- improved lifetime, PL yield and Voc
- Up to 20% with a band-gap of 1.25 eV

- Progress of all-perovskite monolithic tandem solar cells
- Depends on low band gap Sn-Pb bottom cell



Fengjiu Yang, in preparation



## What we learn:

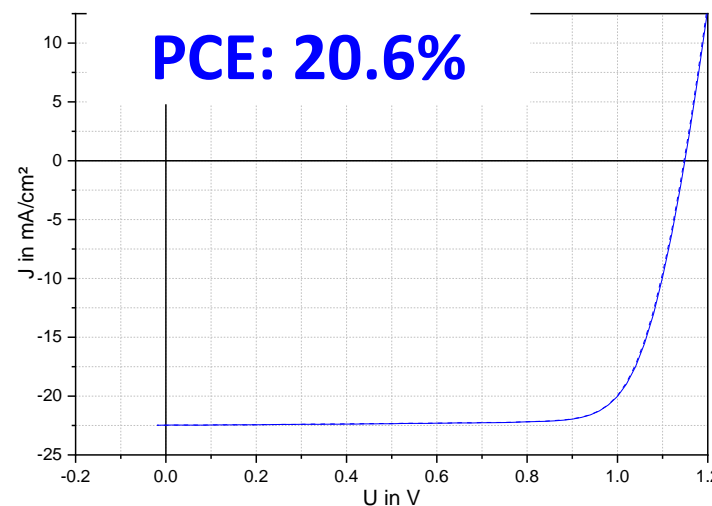
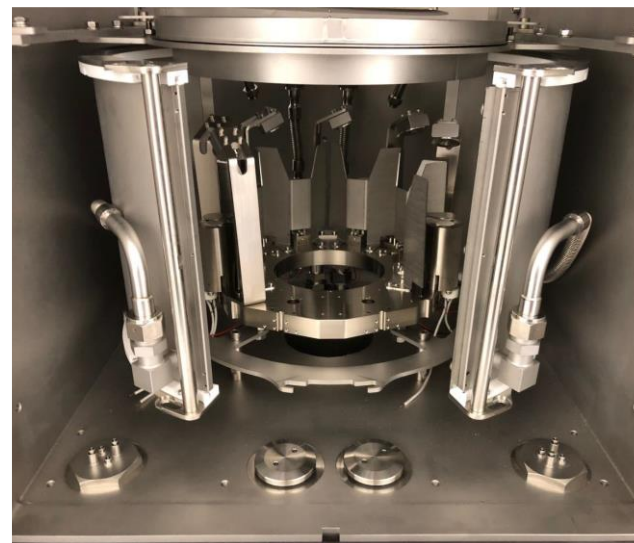
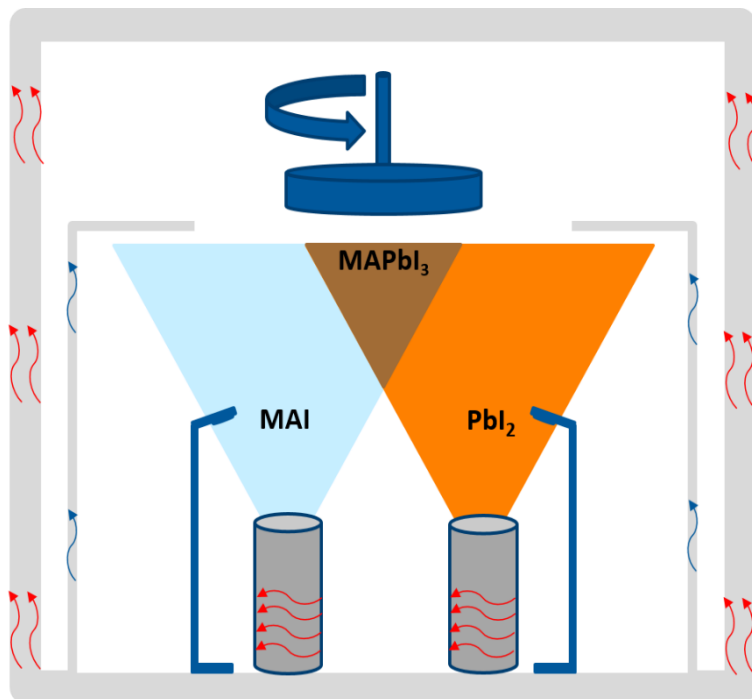
- Perovskite-based tandem solar cells with **Silicon**, **CIGS** or **perovskite** bottom cells have 32-24% efficiency potential

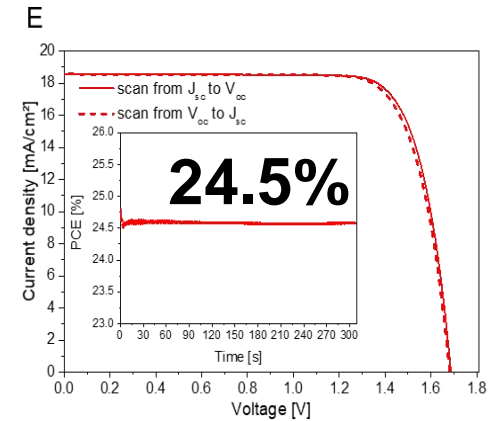
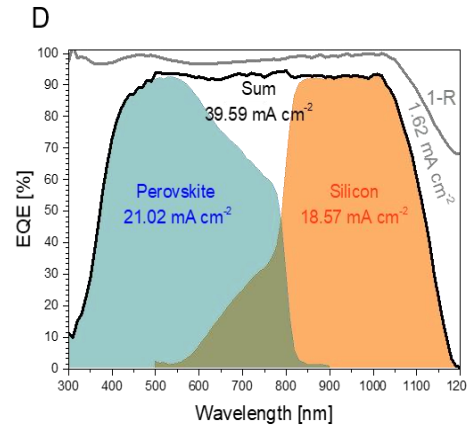
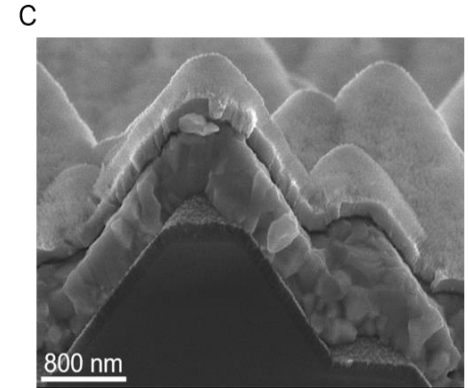
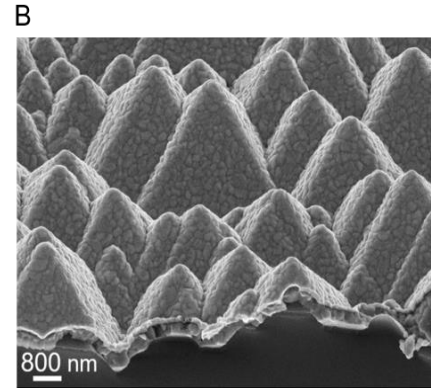
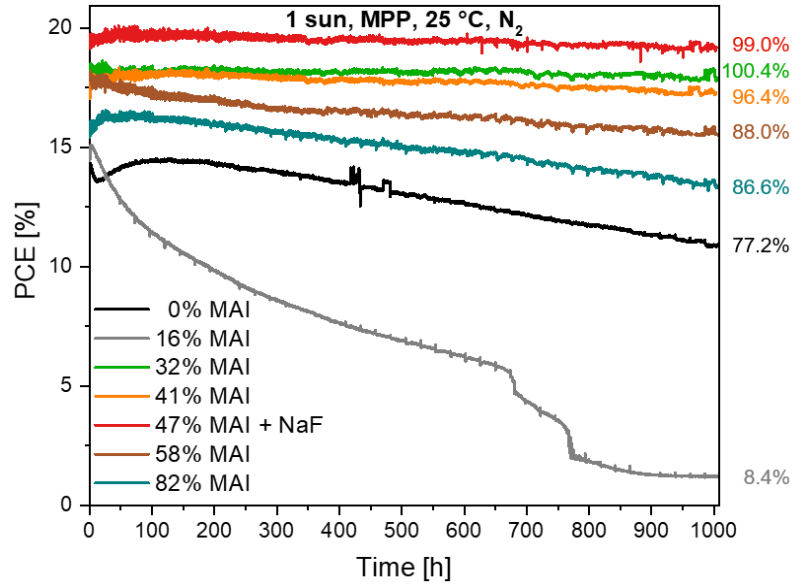
# Upscaling of Perovskite Solar Cells



## New concept:

- thermal management system:
  - actively cooled and heated surfaces
- trapping of volatile MAI on cooled surfaces:
  - no re-evaporation from chamber walls
  - direct co-evaporation

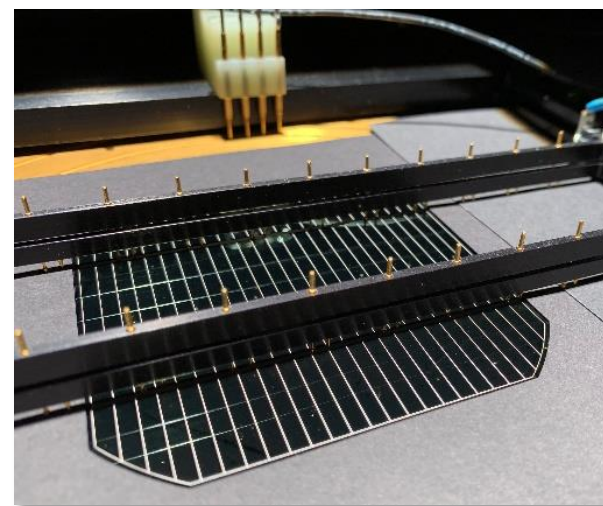
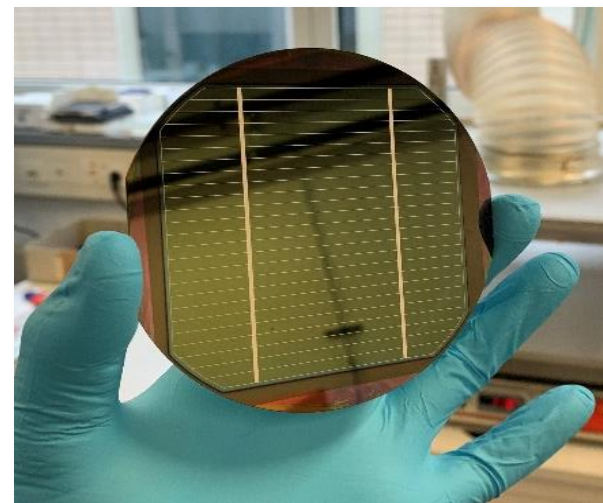
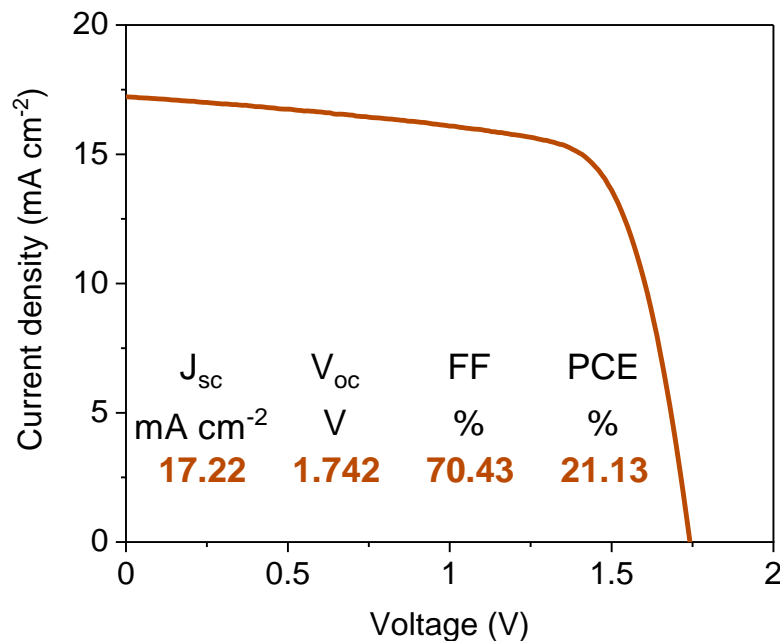




- High stability MAFAPbI<sub>3</sub> perovskite composition
- High robustness against top contact processing
- First demonstration of fully textured perovskite/silicon tandems from co-evaporation

## Proof-of-concept large area Perovskite/Si tandem:

- 60 cm<sup>2</sup> active area
- Perovskite absorber fully evaporated
- Front contact screen-printed at low temp.
- 21% PCE including shadowing losses



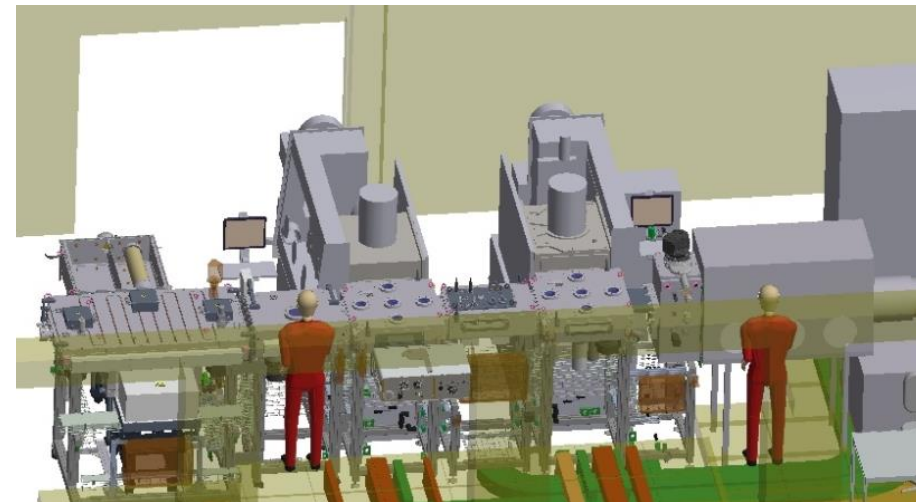
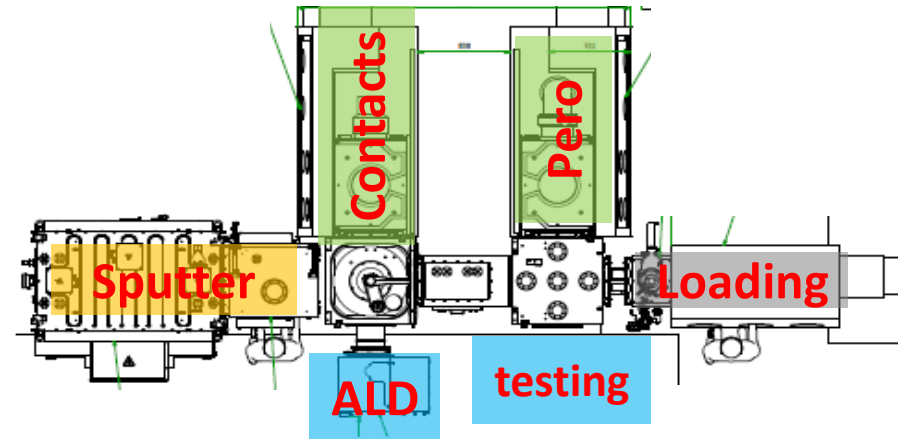
- Designed for 6" perovskite-based tandem solar cells
- Focus on vacuum deposition
  - Co-evaporation
  - Sputter deposition
  - ALD
- Baseline integration + industry collaboration
- Planned operation: 2021

VON ARDENNE

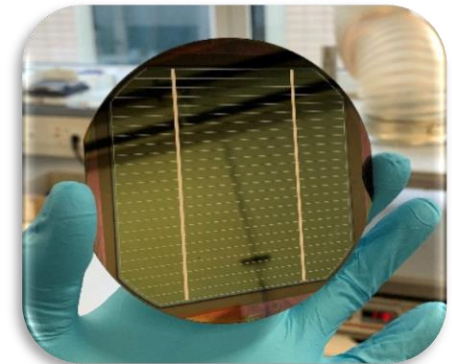
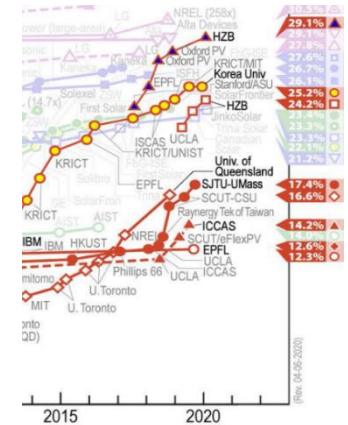
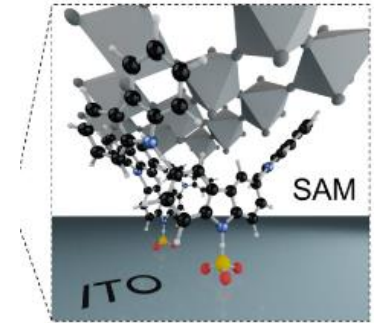


Federal Ministry  
for Economic Affairs  
and Energy

**CREAPHYS**  
MBRAUN GROUP



- Perovskite-based tandem solar cells: promising for next generation modules
- New versatile hole transporting SAMs
  - High passivation, selectivity and fast extraction
  - Stabilizing the 1.68 eV band gap
  - Record certified 24.16% 2T Pero/CIGS tandem
  - Record certified 29.15% 2T Pero/Si tandem
- Upscaling of perovskite via printing and thermal evaporation
  - Promising tandem results with scalable methods
  - First proof-of-concept on textured Si
  - 60 cm<sup>2</sup> perovskite/silicon tandem with 21%
  - New cluster tool for 6" perovskite-based tandems





Funding provided by:



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PersiST, P<sup>3</sup>T, Presto, PeroQ



HySPRINT, PeroSeed, TAPAS



SPP 2196, HIPSTER



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Henk Bolink

Lidon Gil-Escrig

Eike Köhnen  
Amran Al-Ashouri  
Marcel Ross  
Lars Korte  
Philipp Tockhorn  
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Christian Kaufmann  
Iver Lauermann  
Thomas Unold  
José Márquez Prieto  
Hannes Hempel  
Klaus Jäger  
Christiane Becker  
Bernd Rech



Tadas Malinauskas  
Vytautas Getautis

Artiom Magomedov

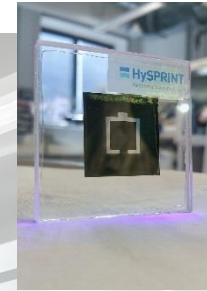
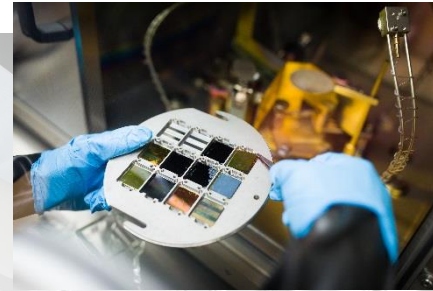


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**Thank you for your attention!**

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