Highly Efficient Monolithic Tandem Solar Cells with Metal-Halide Perovskites

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Virtual DPG Meeting “SKM21”

Sept. 27, 2021
Perovskite: Crystal Structure and History

- First name “perovskite” appeared in 1840 by Gustav Rose for mineral 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*
Metal Halide Perovskites

\[ A = \text{CH}_3\text{NH}_3^+ \quad \text{or} \quad \text{HC(NH)}_2\text{H}_2^+ \quad \text{or} \quad \text{Cs}^+ \]

\[ B = \text{Pb}^{2+} \quad \text{(also} \quad \text{Sb}^{2+} \text{)} \]

\[ X = \text{I}^- \quad \text{or} \quad \text{Br}^- \]

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

Methylammonium lead iodide

„Triple cation“

\[ \text{MAPbI}_3 \]

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

Organic-inorganic metal halide Perovskites

- Direct, bandgap $\sim 1.55$ eV for MAPbI$_3$
- tunable bandgap via composition
- Very few sub bandgap features
- High absorption $\rightarrow$ thin layers $<1$ µm required
Motivation for Tandems

- 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
Motivation for Pero Tandems

• 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

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
How does a 2T-Tandem Solar Cell work?

In series connected tandem:

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

In series connected tandem:

- Voltages add up
- Current limited by limiting sub-cell
- Current matching needed
Further optimized monolithic tandem

- 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!

Köhnen et al., Sustainable Energy & Fuels, 2019,3,1995
1st Generation hole transporting SAM V1036

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

Magomedov, Al-Ashouri et al., Advanced Energy Materials, 2018, 8, 1801892
2nd Generation SAM Meo-2PACz, 2PACz

- Terminated by methoxy groups
- Simplest possible Carbazole SAM


MeO-2PACz and 2PACz provided by TCI

MeO-2PACz and 2PACz provided by dyenamo
2nd Generation SAM Meo-2PACz, 2PACz

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

3rd Generation of SAM: Stable 1.68 eV Perovskite

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

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
3rd Generation of SAM: Record Tandem

- Fine-tuning and stabilizing of Perovskite band gap to ~1.68 eV
- Enhanced hole extraction for higher FFs
- Slightly Perovskite limited in photocurrent
- Record 29.15% efficiency (>1 cm²)
- Promising stability data (300 h)

Al-Ashouri, Köhnen et al, Science (2020), DOI: 10.1126/science.abd4016
Results on Perovskite/CIGS Tandems
Recent 2T Perovskite/CIGSe Tandem

- 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 NiOx prevents shorts on rough surface
- Partial coverage with PTAA enables improved Voc and FF
- 21.6% stabilized efficiency (0.8 cm² active area)

[1] Han et al., Science 2018, 361, 904
2nd Gen SAM integrated directly on top of rough CIGSe

Optimized optics and SAM-improved Voc

<table>
<thead>
<tr>
<th>Area (cm²)</th>
<th>MPP (%)</th>
<th>Jsc (mA/cm²)</th>
<th>Voc (V)</th>
<th>FF (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.035</td>
<td>23.26</td>
<td>19.17</td>
<td>1.68</td>
<td>72</td>
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</table>

World Record 2T Perovskite/CIGSe Tandem 2020

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

<table>
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<tr>
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<th>Voc (V)</th>
<th>FF (%)</th>
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<tr>
<td>1.045</td>
<td>24.16</td>
<td>18.8</td>
<td>1.77</td>
<td>72</td>
</tr>
</tbody>
</table>

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
Sn-Pb Low Band Gap Perovskite Solar Cells

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

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

![Graph showing PCE (%) vs Year]


<table>
<thead>
<tr>
<th></th>
<th>J_{SC} [mA/cm^2]</th>
<th>V_{OC} [V]</th>
<th>FF [%]</th>
<th>PCE [%]</th>
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<tbody>
<tr>
<td>Ref. w/o</td>
<td>31.3</td>
<td>0.76</td>
<td>76.8</td>
<td>18.3</td>
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<tr>
<td>RbI with</td>
<td>31.7</td>
<td>0.82</td>
<td>76.1</td>
<td>19.8</td>
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</tbody>
</table>

Fengjiu Yang, in preparation
Finally >30%

What we learn:

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

Yang and Albrecht et al., APL Materials submitted
Upscaling of Perovskite Solar Cells
Direct Co-Evaporation of Perovskite

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

Roß et al., ACS Appl. Mater. Interfaces 2020, 12, 35, 39261–39272
Fully textured co-evaporated pero/Si tandems

- High stability MAFA$\text{PbI}_3$ perovskite composition
- High robustness against top contact processing
- First demonstration of fully textured perovskite/silicon tandems from co-evaporation

$\text{Roß et al., 2021, submitted}$
**Large area Perovskite/Silicon Tandem**

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

- 60 cm² active area
- Perovskite absorber fully evaporated
- Front contact screen-printed at low temp.
- 21% PCE including shadowing losses

![Current density vs. Voltage graph](image)

**Köhnen and Roß, EUPVSEC-2020**
New Perovskite Cluster Tool “KOALA”

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

• 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 cm² perovskite/silicon tandem with 21%
  • New cluster tool for 6” perovskite-based tandems
Acknowledgements

Funding provided by:

- Federal Ministry of Education and Research
- Federal Ministry for Economic Affairs and Energy
- HELMHOLTZ ASSOCIATION
- DFG Deutsche Forschungsgemeinschaft

Mesa-ZUMA, PEROWIN
PersiST, P³T, Presto, PeroQ

HySPRINT, PeroSeed, TAPAS

SPP 2196, HIPSTER
Acknowledgements

Eike Köhnen
Amran Al-Ashouri
Marcel Ross
Lars Korte
Philipp Tockhorn
Philipp Wagner
Anna Belen Morales
Bernd Stannowski
Rutger Schlatmann
Tobias Bertram
Christian Kaufmann
Iver Lauermann
Thomas Unold
José Márquez Prieto
Hannes Hempel
Klaus Jäger
Christiane Becker
Bernd Rech

Tadas Malinauskas
Vytautas Getautis
Artiom Magomedov

Thomas Riedl
Kai Brinkmann

Bart Macco
Dibyashree Koushik
Mariadriana Creatore

Dieter Neher
Christian Wolff
Martin Stolterfoht
Pietro Caprioglio

Marko Topic
Janez Krc
Marko Jost

Lidon Gil-Escrig
Henk Bolink
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