Perovskite Based Tandem Solar Cells

Thomas **RIEDL**

Chair of Electronic Devices, University of Wuppertal, Germany Wuppertal Center for Smart Materials & Systems





Motivation – why tandem cells ?

BERGISCHE

UNIVERSITÄT WUPPERTAL





BERGISCHE

UNIVERSITÄT WUPPERTAL

Motivation – why tandem cells ?



hv - E_g is lost for electrical energy conversion



BERGISCHE

UNIVERSITÄT WUPPERTAL

Motivation – why tandem cells ?



Motivation – why tandem cells ?

BERGISCHE

UNIVERSITÄT WUPPERTAL





Tandem Cells









This illustration shows the Mars Helicopter on the surface of Mars.









Infra-Red

6.2

6.0

6.4

0.4

0.0

5.4

5.6

5.8

LATTICE CONSTANT [Å]

DOI: 10.5130/pamr.v3i0.1413

ARTICLES https://doi.org/10.1038/s41560-018-0125-0

Corrected: Author Correction; Author Correction

nature energy

III-V-on-silicon solar cells reaching 33% photoconversion efficiency in two-terminal configuration

Romain Cariou 31.4*, Jan Benick¹, Frank Feldmann^{1,2}, Oliver Höhn¹, Hubert Hauser¹, Paul Beutel¹, Nasser Razek³, Markus Wimplinger³, Benedikt Bläsi¹, David Lackner¹, Martin Hermle¹, Gerald Siefer¹, Stefan W. Glunz^{1,2}, Andreas W. Bett¹ and Frank Dimroth¹



33% efficiency (1.3 x the efficiency of silicon cells)

but: cost approx. 1-10 T€per W (c.f.: Si approx. 1 € per W)



Perovskites – the low-cost alternative?



J. Mater. Chem. A, 2015, 3, 9032–9050

BERGISCHE

UNIVERSITÄT WUPPERTAL

solution processing, low temperature, low cost



BERGISCHE

UNIVERSITÄT WUPPERTAL

Perovskite Band Gap Tunability





Perovskite based tandem cells

CIGS – Perovskite

BERGISCHE

UNIVERSITÄT WUPPERTAL

Pro: CIGS established cell technology, stable

Con: Cd content, abundance of elements, high-T process

Silicon – Perovskite

Pro: high efficiency, Si cell highly developed Con: **high-T process**, not flexible

Perovskite – Perovskite

Pro: low temperature, large area, high throughput Con: narrow-gap cell potentially unstable (Sn-based)

Organic – Perovskite

Pro: low temperature, large area, high throughput, **reduced Pb/Sn** Con: **still low efficiency**?





Efficiency estimates



Bottom Cell Type	Low bandgap(eV)	High bandgap (eV)	Open Circuit Voltage Voc (V)	Short Circuit Current J _{SC} (mA/cm ²)	PCE (%)
Free choice	0.96	1.63	2.032	24.6	44.8
Si	1.12	1.73	2.273	21.6	44.3
CIGS	1.10	1.72	2.246	21.8	44.1
Perovskite	1.22	1.81	2.441	19.3	42.7
OPV	1.33	1.88	2.609	17.4	41.4



Appl. Phys. Rev. 8, 041307 (2021)



Interconnect



Requirements:

- loss-free recombination of electrons from one sub-cell with holes form the other sub-cell → addition of Voltage; high Fill Factor
- optically transparent
- protects the bottom cell against deposition of the subsequent cell



 SnO_{x}/MoO_{x} Junction

coll. with Dr. Selina Olthof



Tandem with only SnO_x / MoO_x junction



 \rightarrow energetic barrier between SnO_x and MoO_x adds a diode-like non-linearity



 \rightarrow with 32 ALD cycles of InO_x (1.5 nm) well behaved J/V characteristcs (no S-shape)





BERGISCHE

UNIVERSITÄT WUPPERTAL

Metals as interconnect





Nature Energy 4, 864-873 (2019)

Joule 4, 1594 (2020)



ALD-InO_x vs. thin metal





Interconnect for all-perovskite tandems

perovskite-**perovskite** tandem



All-perovskite tandem with low gap perovskite \rightarrow PCE of ~ 26%

Adv. Energy Mater. 2023, 13, 2202674





BERGISCHE UNIVERSITÄT WUPPERTAL

Adv. Energy Mater. 2023, 13, 2202674

 $QFLS_{EL} = k_BT \cdot \ln\left(ELQY \cdot \frac{J_{inj}}{J_{0,rad}}\right)$



Sub-cell analysis (electroluminescence): pseudo-JVs from integrated sub-cells match single juctions + add up perfectly to the tandem

 $\rightarrow InO_x$ interconnect almost lossless



Perovskite based tandem cells

CIGS – Perovskite

BERGISCHE

UNIVERSITÄT WUPPERTAL

Pro: CIGS established cell technology, stable

Con: Cd content, abundance of elements, high-T process

Silicon – Perovskite

Pro: high efficiency, Si cell highly developed Con: **high-T process**, not flexible

Perovskite – Perovskite

Pro: low temperature, large area, high throughput Con: narrow-gap cell potentially unstable (Sn-based)

Organic – Perovskite

Pro: low temperature, large area, high throughput, **reduced Pb/Sn** Con: **still low efficiency**?





BERGISCHE

UNIVERSITÄT WUPPERTAL





BERGISCHE

UNIVERSITÄT WUPPERTAL

Mixed-Halide Perovskites



ACS Energy Lett. 3, 204 (2018)

Nat. Comms. 12, 6955 (2021)



V_{oc} loss in wide-gap perovskite cells



Rajagopal et al., Nano Lett. 2018, 18, 3985–3993



 $FA_{0.8}Cs_{0.2}Pb(I_{0.5}Br_{0.5})_3 E_g = 1.85 eV$

coll. with Dr. Martin Stolterfoht





1.40

GIWAXS by L. Merten, U Tuebingen

Wide-gap perovskite cell

BERGISCHE

UNIVERSITÄT

WUPPERTAL

Universitäx

coll. with

Dr. Martin Stolterfoht



Losses at interfaces more critical than in the bulk!

3D/2D interface – electronic structure

Universität

zu Köln







Wide-gap perovskite cell



 \rightarrow high $V_{\rm oc}$ and high *FF*

 \rightarrow record for wide-gap PSCs with Br/l > 2/3 (E_{q} = 1.8 - 1.9 eV)





Adv. Energy Mater. 2023, 13, 2202674







 $\mathsf{FA}_{0.78}\mathsf{Cs}_{0.22}\mathsf{Pb}(\mathsf{Br}_{0.3}\mathsf{I}_{0.7})_3 + 10\%\mathsf{MAPbCI}_3$



N N N N

piperazinium iodide

J. Am. Chem. Soc. 2020, 142, 47, 20134–20142

Piperazinium iodide reduces recombination loss in 1.80 eV triple-halide perovskites

Nature Energy (under review)





Record high V_{oc} of 1.36 V for a 1.80 eV wide gap perovskite solar cell ($\Delta V_{oc} = 0.44$ V) Nature Energy (under review)





 V_{oc} of 1.35 V of a 1.725 eV wide gap perovskite solar cell (ΔV_{oc} = 0.375 V)

Adv. Energy Mater. 2021, 11, 2003386



Perovskite based tandem cells

CIGS – Perovskite

UNIVERSITÄT WUPPERTAL

Pro: CIGS established cell technology, stable

Con: Cd content, abundance of elements, high-T process

Silicon – Perovskite

Pro: high efficiency, Si cell highly developed Con: **high-T process**, not flexible

Perovskite – Perovskite

Pro: low temperature, large area, high throughput Con: narrow-gap cell potentially unstable (Sn-based)

Organic – Perovskite

Pro: low temperature, large area, high throughput, reduced Pb/Sn Con: still low efficiency?







http://pubs.acs.org/journal/aelccp

Perovskite/CIGS Tandem Solar Cells: From Certified 24.2% toward 30% and Beyond

Marko Jošt,* Eike Köhnen, Amran Al-Ashouri, Tobias Bertram, Špela Tomšič, Artiom Magomedov, Ernestas Kasparavicius, Tim Kodalle, Benjamin Lipovšek, Vytautas Getautis, Rutger Schlatmann, Christian A. Kaufmann, Steve Albrecht,* and Marko Topič

Read Online



+ The main improvement: increased open-circuit voltage of PSC (1.68 eV) (SAM Me-4PACz HTL, PEAI additive, and LiF interlayer)

- poor fill factor (FF = 71.2%; shunts) and short-circuit current density only 18.8 mA/cm²

	nonencapsulated				encaps	
	$E_{\rm g} = 1.65 {\rm eV}$	$E_{\rm g} = 1.68 {\rm eV}$	$E_{\rm g} = 1.70 {\rm eV}$	$E_{\rm g} = 1.72 {\rm eV}$	$E_{\rm g} = 1.69 {\rm eV}$	
Pero $J_{\rm SC SIM}$ (mA cm ⁻²)	19.92	19.93	19.92	19.33	18.8	
CIGS J_{SC} SIM (mA cm ⁻²)	19.92	19.92	19.93	19.92	18.8	
$J_{\rm SC SIM}$ (mA cm ⁻²)	19.9	19.9	19.9	19.3	18.8	
FF (%)	80	80	80	80	80	
$V_{\rm oc}$ (V)	1.96	1.99	2.01	2.03	2.00	
PCE (%)	31.2	31.6	32.0	31.3	30.0	

🔤 😳 🚺



RESEARCH

SOLAR CELLS

Monolithic perovskite/silicon tandem solar cell with >29% efficiency by enhanced hole extraction

Amran Al-Ashouri^{1*}, Eike Köhnen^{1*}, Bor Li¹, Artiom Magomedov², Hannes Hempel³, Pietro Caprioglio^{1,4}, José A. Márquez³, Anna Belen Morales Vilches⁵, Ernestas Kasparavicius², Joel A. Smith^{6,7}, Nga Phung⁶, Dorothee Menzel¹, Max Grischek^{1,4}, Lukas Kegelmann¹, Dieter Skroblin⁸, Christian Gollwitzer⁸, Tadas Malinauskas², Marko Jošt^{1,9}, Gašper Matič⁹, Bernd Rech^{10,11}, Rutger Schlatmann^{5,12}, Marko Topič⁹, Lars Korte¹, Antonio Abate⁶, Bernd Stannowski^{5,13}, Dieter Neher⁴, Martin Stolterfoht⁴, Thomas Unold³, Vytautas Getautis², Steve Albrecht^{1,11}

Perovskite used (E_g 1.68 eV): $Cs_{0.05}(FA_{0.77}MA_{0.23})_{0.95}Pb(I_{0.77}Br_{0.23})_3$

Science 370, 1300-1309 (2020)



Efficiency > 31% predicted from sub-cell analysis

Science 370, 1300–1309 (2020)



Perovskite/Silicon

33.2%! KAUST Team Sets World Record for Tandem Solar Cell Efficiency

APRIL 14, 2023 BY ALEINA IN TECHNOLOGY

f 🎔 🖇 🔊 in

PVTIME – Researchers at the King Abdullah University of Science and Technology (KAUST) Photovoltaics Laboratory (KPV-Lab) of the KAUST Solar Center, a leading institute of research excellence in the Kingdom of Saudi Arabia, claimed on 13 April that it has produced a perovskite-silicon tandem solar cell with a power conversion efficiency of 33.2%, which was confirmed by the European Solar Test Installation (ESTI) as the top of the National Renewable Energy Laboratory's (NREL) Best Research-cell Efficiency List.

Meyer Burger to commercialize 29.6%-efficient perovskite tandem solar cells

Meyer Burger is working with several research institutes in Switzerland and Germany to integrate perovskite tandem technology into its manufacturing processes.

DECEMBER 14, 2022 SANDRA ENKHARDT





Multijunction Cells (2-terminal, monolithic)

- LM = lattice matched
- MM = metamorphic
- IMM = inverted, metamorphic
- Three-junction (concentrator)
- Three-junction (non-concentrator)
- Two-junction (concentrator)
- Two-junction (non-concentrator)
- Four-junction or more (concentrator)
- Four-junction or more (non-concentrator)

Single-Junction GaAs

- ▲ Single crystal
- ▲ Concentrator
- Thin-film crystal

Crystalline Si Cells

- Single crystal (concentrator)
- Single crystal (non-concentrator)
- Multicrystalline
- Silicon heterostructures (HIT)
- ▼ Thin-film crystal

Thin-Film Technologies

- CIGS (concentrator)
- CIGS
- CdTe
- Amorphous Si:H (stabilized)

Emerging PV

- O Dye-sensitized cells
- Perovskite cells
- A Perovskite/Si tandem (monolithic)
- Organic cells
- Organic tandem cells
- Inorganic cells (CZTSSe)
- Quantum dot cells (various types)
- Perovskite/CIGS tandem (monolithic)







Adv. Mater. 2023, 35, 2207883



Carbon footprint





Perovskite/Perovskite



All-perovskite tandem with low gap perovskite \rightarrow PCE of \sim 26%

Adv. Energy Mater. 2023, 13, 2202674



Perovskite/Perovskite



Nature Energy (under review)



Perovskite/Perovskite



Sub-cell analysis projects efficiency > 30%

Nature Energy (under review)

Narrow Bandgap Perovskite

Sargent, Tan et al. Nature 603, 73–78 (2022)

BERGISCHE

UNIVERSITÄT WUPPERTAL



Narrow gap cell needs to be very thick $\sim \mu m$ Problem: limited carrier diffusion length



Narrow Bandgap Perovskite

Sargent, Tan et al. Nature 603, 73–78 (2022)





Narrow Bandgap Perovskite



Sargent, Tan, et at. Nat. Energy 4, 864–873 (2019)





J. Phys. Chem. C 2018, 122, 25, 13926–13936



Nat. Comms, **12**, 2853 (2021)





Article nature Perovskite-organic tandem solar cells with indium oxide interconnect

https://doi.org/10.1038/s41586-022-04455-0
Received: 9 December 2020
Accepted: 24 January 2022
And the second

K. O. Brinkmann^{1,2,8}, T. Becker^{1,2,8}, F. Zimmermann^{1,2}, C. Kreusel^{1,2}, T. Gahlmann^{1,2},
M. Theisen^{1,2}, T. Haeger^{1,2}, S. Olthof³, C. Tückmantel^{1,2}, M. Günster^{1,2}, T. Maschwitz^{1,2},
F. Göbelsmann^{1,2}, C. Koch³, D. Hertel³, P. Caprioglio⁴, F. Peña-Camargo⁴, L. Perdigón-Toro⁴,
A. Al-Ashouri⁵, L. Merten⁶, A. Hinderhofer⁶, L. Gomell⁷, S. Zhang⁷, F. Schreiber⁶, S. Albrecht⁵,
K. Meerholz³, D. Neher⁴, M. Stolterfoht⁴ & T. Riedl^{1,2}

Check for updates









BERGISCHE

UNIVERSITÄT WUPPERTAL

Our OSCs based on PM6:Y6 (+PC₆₀BM)



PM6

PM6:Y6 (1:1.2) in CHCl₃ +0.5% Cl-naphthalene



adding some fullerene PC₆₀BM (20%) boosts efficiency to **17.5%** mainly due to increased J_{sc}





continuous MPP tracking in N2



BERGISCHE

UNIVERSITÄT WUPPERTAL

- degradation linked to VIS illumination
- no degradation under NIR illumination
- \rightarrow beneficial for tandems
 - perovskite cell acts as low-pass filter











 \rightarrow photo-degradation of PL-QY of donor polymer PM6

 \rightarrow Y6 acceptor mostly unaffected

BERGISCHE

UNIVERSITÄT WUPPERTAL

L. Perdigón Toro and M. Stolterfoht, U Potsdam





Perovskite/organic tandem semi-empirical simulation



With organic $E_g = 1.33 \text{ eV}$, perovskite cell with min. $E_g \approx 1.85 \text{ eV}$ required



Perovskite/organic tandem



Received: 9 May 2021; Accepted: 2 December 2021; nature Published online: 20 January 2022 energy

ARTICLES 0.1038/s41560-021-00966

Monolithic perovskite/organic tandem solar cells with 23.6% efficiency enabled by reduced voltage losses and optimized interconnecting layer

Wei Chen 01.2.3.4.8, Yudong Zhu 2.5.8, Jingwei Xiu², Guocong Chen², Haoming Liang 3.4, Shunchang Liu 3.4, Hansong Xue 4, Erik Birgersson 4.6, Jian Wei Ho4, Xinshun Qin1, Jingyang Lin17, Ruijie Ma⁵, Tao Liu⁵, Yanling He^{1,7}, Alan Man-Ching Ng⁷, Xugang Guo⁰⁰², Zhubing He⁰²²⁰, He Yan⁰⁵, Aleksandra B. Djurišić 01 and Yi Hou 034

RESEARCH ARTICLE

() Check for updates

Article

Constructing Monolithic Perovskite/Organic Tandem Solar Cell with Efficience of 22.0% via Reduced Open-Circuit Voltage Loss and Broadened Absorption Spectra

Shucheng Qin, Chenxing Lu, Zhenrong Jia, Yiyang Wang, Siguang Li, Wenbin Lai, Pengju Shi, Rui Wang, Can Zhu, Jiaqi Du, Jinyuan Zhang, Lei Meng,* and Yongfang Li*

Adv. Funct. Mater. 2021, 2109321 RESEARCH ARTICLE

www.afm-journal.de

Surface Reconstruction for Stable Monolithic All-Inorganic Perovskite/Organic Tandem Solar Cells with over 21% 21% Efficiency

Weijie Chen, Dong Li, Xu Chen, Haiyang Chen, Shuo Liu, Haidi Yang, Xingi Li, Yunxiu Shen, Xuemei Ou, Yang (Michael) Yang, Lin Jiang, Yaowen Li,* and Yongfang Li

Joule

Joule 4, 1594 (2020)

Article

Efficient and Reproducible Monolithic Perovskite/Organic Tandem Solar Cells with Low-Loss Interconnecting Layers



CelPress

Perovskite-organic tandem solar cells with indium oxide interconnect 24 %

nature

https://doi.org/10.1038/s41586-022-04455-0	K. O. Brinkmann ^{1,2,8} , T. Becker ^{1,2,8} , F. Zimmermann ^{1,2} , C. Kreusel ^{1,2} , T. Gahlmann ^{1,2} ,
Received: 9 December 2020	 M. Theisen^{1,2}, T. Haeger^{1,2}, S. Olthof³, C. Tückmantel^{1,2}, M. Günster^{1,2}, T. Maschwitz^{1,2}, F. Göbelsmann^{1,2}, C. Koch³, D. Hertel³, P. Caprioglio⁴, F. Peña-Camargo⁴, L. Perdigón-Toro⁴, A. Al-Ashouri⁵, L. Merten⁶, A. Hinderhofer⁶, L. Gomell⁷, S. Zhang⁷, F. Schreiber⁶, S. Albrecht
Accepted: 24 January 2022	
	K. Meerholz ³ , D. Neher ⁴ , M. Stolterfoht ⁴ & T. Riedl ^{1,2}
Check for updates	







Stability of the tandem limited by the wide-gap perovskite cell





Need organic cell with lower gap (1.15 eV) combined with 1.7-1.75 eV perovskite

BERGISCHE UNIVERSITÄT WUPPERTAL

Jiang et al., Science 378, 1295–1300 (2022)



$$E_g = 1.75 \, eV$$

great MPP stability

MOLECULAR PHYSICS, 1970, VOL. 18, No. 2, 145–164

The energy gap law for radiationless transitions in large molecules

by ROBERT ENGLMAN Theoretical Solid State Physics Group, Soreq Nuclear Research Centre, Yavne, Israel

and JOSHUA JORTNER Department of Chemistry, Tel-Aviv University, Tel-Aviv, Israel

$$W = \frac{C^2 \sqrt{2\pi}}{\hbar \sqrt{\hbar\omega_M \Delta E}} \exp\left(-\gamma \Delta E/\hbar\omega_M\right).$$

Rate for non-rad recombination W increases exponentially as the energy gap ΔE decreases



Figure 2. Plot of in k_{nr} vs. E_{em} for the MLCT excited states of the series of complexes fac-[Re(bpy)(CO)₃L](PF₆) at 23 °C in methylene chloride solution.

The Journal of Physical Chemistry, 87, 6 (1983)



Nat. Rev. Mater. (in preparation)



 $E_{g} = 1.2 \text{ eV}$



Table 1 | Photovoltaic parameters of the optimal single-junction OSCs based on PTB7-Th:BTPSV-4F and PTB7-Th:BTPSeV-4F, under the illumination of AM 1.5G, 100 mW cm⁻²

Device	V _{oc} [V]	J _{sc} [mA·cm ⁻²]	J _{cal} from EQE [mA·cm ⁻²]	FF [%]	PCE [%]
PTB7-Th:BTPSV-4F	0.66 (0.66±0.01)	28.4 (28.1±0.3)	27.76	69.5 (69.2±0.6)	13.0 (12.8 ± 0.2) ^a
PTB7-Th:BTPSeV-4F	0.66 (0.66 ± 0.01)	30.1 (29.8 ± 0.2)	29.30	71.4 (71.0 ± 0.4)	14.2 (13.9±0.2)

"The average values and deviations are calculated from 10 independent devices.

Nature Communications 14, 1236 (2023)

Univ. Cologne

Dr. C. Koch Dr. S. Olthof Dr. D. Hertel Prof. K. Meerholz

Univ. Potsdam

P. Caprioglio Dr. M. Stolterfoht Prof. D. Neher

Univ. Tübingen

L. Merten Dr. A. Hinderhofer Prof. F. Schreiber

HZB Berlin

Dr. F. Yang Dr. A. Al-Ashouri Prof. S. Albrecht

MPI Eisenforschung

L. Gomell Dr. S. Zhang

DFG Deutsche Forschungsgemeinschaft

RI1551/12-1 (MUJUPO) RI1551/15-1 (HIPSTER)





Horizon 2020 European Union funding for Research & Innovation **Univ. Wuppertal**



Dr. K. Brinkmann F. Zimmermann C. Kreusel T. Becker Dr. T. Gahlmann M. Theisen Dr. T. Haeger M. Günster F. Göbelsmann T. Maschwitz

Grant no. 951774 (FOXES)