

CPP 19: Hybrid and Perovskite Photovoltaics II

Time: Tuesday 9:30–12:30

Location: H18

CPP 19.1 Tue 9:30 H18

Three-Terminal Perovskite Silicon Tandem Solar Cells with Top and Interdigitated Rear Contacts — ●PHILIPP TOCKHORN, PHILIPP WAGNER, JOHANN CHRISTOPH STANG, LUKAS KEGELMANN, MATHIAS MEWS, STEVE ALBRECHT, and LARS KORTE — Helmholtz Zentrum Berlin, Kekuléstraße 5, 12489 Berlin

So far, perovskite and silicon were paired in two terminal (2T) and four terminal (4T) tandems, yielding power conversion efficiencies (PCE) above 25 %. In a 2T tandem solar cell, the total current density is limited by the subcell yielding the lower current, restricting the overall performance. Independently operated 4T tandem cells need additional transparent conductive layers for lateral conduction and thus parasitic absorption is ultimately higher. Here, we present a novel three terminal (3T) approach for the interconnection of an n-i-p perovskite top cell with an interdigitated back contact (IBC) silicon heterojunction (SHJ) bottom cell. For this, the interconnection of the subcells is realised with SnO₂ and nc-Si:H(n) layers. This allows the independent operation of both subcells in a monolithic device, thus combining the advantages of both 2T and 4T devices into a 3T tandem solar cell. For this, electrons generated in the perovskite top cell are collected at the rear side n-contact of the IBC SHJ. We report the first 3T perovskite/silicon tandem device in this architecture that reaches a combined stabilised PCE of 15.5 %. Despite losses due to hysteresis, our 3T device is still strongly limited by optical losses from high reflection and parasitic absorption. We present the theoretical limits of this tandem approach and highlight further routes to improve efficiency.

CPP 19.2 Tue 9:45 H18

Minimizing Electrical Interconnection Losses in Laser-Patterned Perovskite Solar Cells — ●MARKUS FENSKE¹, CHRISTOF SCHULTZ¹, ANDREAS BARTELT¹, ANTJE NEUBAUER², CORNELIA JUNGHANS², MARKO JOŠT³, BERND RECH⁴, RUTGER SCHLATMANN^{1,5}, STEVE ALBRECHT³, and BERT STEGEMANN¹ — ¹Hochschule für Technik und Wirtschaft Berlin — ²Becker & Hickl GmbH, Berlin — ³Helmholtz-Zentrum Berlin für Materialien und Energie, Young Investigator Group for Perovskite Tandem Solar Cells — ⁴Helmholtz-Zentrum Berlin für Materialien und Energie, Institut für Silizium-Photovoltaik — ⁵Helmholtz-Zentrum Berlin für Materialien und Energie, PVcomB

Successful up-scaling of metal halide perovskite (MHP) solar cells from lab-scale to large module sizes requires monolithic series interconnection. Here we report on recent progress in P2 and P3 laser patterning. By systematically varying the laser pulse energy, pulse duration and laser wavelength, a process window for the patterning could be established. We demonstrate the lateral effect of the laser processing on the chemical composition in the patterned area (by EDX) and on the perovskite semiconductor properties in the vicinity of the scribed laser line (by TR-PL). The results show that shorter laser pulse durations (ps rather than ns) and shorter laser wavelengths (355 nm rather than 532 nm) have a beneficial effect particularly on P2 process control, thereby minimizing electrical interconnection losses.

CPP 19.3 Tue 10:00 H18

Highly Efficient monolithic perovskite silicon tandem solar cells: analysing current-mismatch conditions — ●EIKE KÖHNEN, MARKO JOŠT, PHILIPP TOCKHORN, ANNA B. MORALES VILCHES, AMRAN AL-ASHOURI, BERND STANNOWSKI, and STEVE ALBRECHT — Helmholtz-Zentrum Berlin, Germany

To well exceed the theoretical efficiency limit for silicon single junction solar cells, multiple absorbers with different bandgaps can be combined into tandem solar cells. Recently, metal halide perovskites have attracted significant scientific attention as tandem partner with silicon. Here, we combine a rear emitter silicon cell with a p-i-n perovskite cell into a highly efficient monolithic tandem cell. With this design, a certified tandem efficiency of 25% is realized at highly unmatched sub-cell photocurrents. Further improvements of the top contact and the perovskite thickness, enabled a remarkably efficiency of 26%, the highest yet published value. Additionally, we show how these tandem solar cells behave under various illumination spectra, e.g. when the sun spectrum changes over the day. Using selective illumination, different mismatch conditions are generated, affecting the tandem fill factor (FF). The experimental findings are reproduced by equivalent

circuit simulations based on single cell parametrization. In both experiment and simulation, we find that the FF increases significantly when the sub-cells become more unmatched, an important finding as the FF enhancement slightly compensates the photocurrent reduction due to current mismatch. This will affect the mismatch condition for optimum performance and also affects the energy yield of the tandem solar cell.

CPP 19.4 Tue 10:15 H18

Characterisation of inorganic/hybrid lead and tin perovskites for efficient light-emitting diodes — ●INES SCHMIDT, SELINA OLTTHOF, and KLAUS MEERHOLZ — Universität zu Köln

Solution processable semiconducting perovskites have the general structure AMX₃, where A is a cation (Cs, methylammonium (MA) or formamidinium (FA)), M is a metal cation (Sn or Pb) and X is a halide anion (I, Br or Cl). The combination of these results in 18 different primary (i.e. non-mixed) perovskites with bandgaps corresponding to a wide range of the electromagnetic spectrum. This bandgap tunability, very narrow emission characteristic and a high charge carrier mobility make perovskites promising candidates for light-emitting applications.[1] To gain better understanding regarding possible emission wavelengths as well as the absolute energy levels, we optimized and characterized this complete set of 18 material combinations using structural, electrical and optical measurement techniques. With that knowledge, multi-layer perovskite light-emitting diodes with an optimized energy alignment can be fabricated. To this end, we introduced crosslinkable hole-transporting layers into the device stack to improve charge injection. Another important aspect is the enhancement of charge carrier recombination, which is typically done by reducing the grain size in perovskite films. Therefore, we investigated the impact of different small organic molecules and polymer additives in the preparation procedure on perovskite morphology and device performance.

[1] Stoumpos, C. C et al., *Inorg. Chem.*, 52(15), 9019-9038 (2013).

CPP 19.5 Tue 10:30 H18

Pure Bromide-based Perovskites: Film Formation, Morphology and (In)homogeneity — ●CAROLIN REHERMANN^{1,2}, VINCENT SCHRÖDER², SEBASTIÁN CAICEDO-DÁVILA¹, JOSÉ MÁRQUEZ PRIETO¹, ABOMA MERDASA¹, EMIL LIST-KRATOCHVIL^{1,2} and EVA UNGER^{1,2} — ¹Helmholtz-Zentrum Berlin für Materialien und Energie, 12489 Berlin, Germany — ²Humboldt-Universität zu Berlin, 12489 Berlin, Germany

Due to their band gap tunability metal halide perovskites are an interesting material for tandem solar cells and LEDs, high band gaps around 2.3 eV are obtained with pure bromide perovskites.1 For established mixed perovskites wrinkled films are formed depending on preparation conditions. This inhomogeneity in composition and morphology influences the device performance.2, 3 Wrinkles develop likewise during preparation of pure bromides.

In this work, we study the film formation of organic and inorganic bromide-based perovskites with in-situ PL and UV/Vis measurements during different preparation steps and on various contact layers. Morphology and (In)homogeneity in composition of the final films are analyzed by XRD, PLimaging and EDX. Understanding parameters influencing wrinkle formation allows controlling this process and gives the opportunity to systematically provoke/ suppress them. That way resulting optoelectronic properties and detailed influences on device performance of wrinkles bromide perovskites are investigated.

1. Unger et al. *Journal of Materials Chemistry A* 2017, 5 (23), 11401-11409. 2. Braunger et al. *J. Phys. Chem. C* 2018, 122, 17123-17135. 3. Bercegol et al. *J. Phys. Chem. C* 2018, 122, 23345-23351.

CPP 19.6 Tue 10:45 H18

Methylammonium Lead Iodide Perovskite Solar Cells from direct Co-Evaporation — ●MARCEL ROSS, LIDÓN GIL ESCRIG, AMRAN AL-ASHOURI, MARKO JOŠT, and STEVE ALBRECHT — Helmholtz-Zentrum Berlin, Young Investigator Group for Perovskite Tandem Solar Cells

Deposition of perovskite solar cell absorbers by co-evaporation offers a variety of advantages over solution based preparation such as homogeneous coating of large substrates and conformal coverage of textured substrates. Up to date the gas-like evaporation behavior of organic precursors such as methylammonium halides makes the rate measurement

and consequently the process control difficult as volatile species might re-evaporate from chamber walls. To overcome these challenge we implement an evaporation setup with a thermal management system to directly co-evaporate volatile precursors for perovskites. Through a combination of actively cooled and heated surfaces, a direct and controlled evaporation process is achieved. This way, high quality Methylammonium Lead Iodide perovskite films are obtained as confirmed by XRD, PL and optical measurements. These perovskite films are implemented into p-i-n solar cells utilizing different hole transport materials (HTMs) such as PTAA, self-assembling monolayer (SAM) molecules and Spiro-TTB. Although the coarse grain morphology measured by SEM is very comparable, the solar cell performance is strongly affected by the used HTM and stabilized efficiency over 20% are realized with the SAMs only. The herein achieved efficiency is comparable to the highest reported value for evaporated perovskite solar cells.

CPP 19.7 Tue 11:00 H18

Structural and optical properties of Cs₂AgBiBr₆, a lead-free perovskite for photovoltaic applications — ●MELINA ARMER¹, JULIAN HÖCKER¹, LIUDMILA KUDRIASHOVA¹, SEBASTIAN HAMMER¹, KRISTOFER TVINGSTEDT¹, ANDREAS BAUMANN², JENS PFLAUM^{1,2}, and VLADIMIR DYAKONOV¹ — ¹Experimental Physics VI, Julius Maximilian University of Würzburg, 97074 Würzburg — ²Bavarian Center for Applied Energy Research (ZAE Bayern), 97074 Würzburg

As conventional perovskite solar cells contain lead and therefore suffer toxicity issues, finding alternative and stable lead-free materials for the application in perovskite photovoltaics has become an essential problem to be solved. In this work, lead-free Cs₂AgBiBr₆ single crystals have been synthesized using a solution based approach, permitting a detailed characterization of the optical and structural properties of this material. The single crystals have been characterized using steady-state and time-resolved photoluminescence (PL) spectroscopy. The morphology and quality of the as prepared crystals has been evaluated using scanning electron microscopy and X-ray diffraction. We observed PL in the visible region characterized by large time constants. Furthermore, Cs₂AgBiBr₆ thin films have been prepared and their optical and structural properties have been investigated, and compared to the corresponding bulk single crystals. Due to the encouraging optical and structural properties Cs₂AgBiBr₆ is a promising material for the application as absorber layer in lead-free perovskite solar cells.

15 min. break

CPP 19.8 Tue 11:30 H18

Improving the thermal stability of co-evaporated lead halide perovskites by replacing MA with Cs: Film growth and decomposition analyzed by *in situ* XRD — ●THOMAS BURWIG, PAUL PISTOR, and ROLAND SCHEER — Martin-Luther-Universität Halle-Wittenberg, Von-Danckelmann-Platz 3, 06120 Halle (Saale), Germany

Perovskites have seen a rising popularity in photovoltaic research due to their outstanding optoelectronic properties, with the most researched perovskite in this field being MAPbI₃. However, practical applications are held back by a lack of stability against atmospheric water, light and temperature. In our work we present fully inorganic caesium-lead-halide-perovskites, in which the organic MA is exchanged for caesium. We deposit thin films via dry, vacuum-based thermal co-evaporation. A specialized *in situ* X-ray diffraction setup allows us to monitor the crystal phase evolution over the course of the experiments. We report on the growth and thermally induced phase changes as well as the thermally induced decomposition of CsPbI₃ and CsPbBr₃ perovskites. CsPbI₃ is grown in a black, orthorhombic perovskite structure at room temperature (α' -CsPbI₃). This phase is stable in vacuum, but unstable under ambient conditions, as it undergoes a phase change to yellow, also orthorhombic δ -CsPbI₃. No such phase change was observed for CsPbBr₃. Our experiments confirm a higher thermal stability for CsPbI₃ and CsPbBr₃ as compared to the MA-lead-halides. Finally, we report on our attempts to synthesize further inorganic perovskites with improved stability.

CPP 19.9 Tue 11:45 H18

Kinetics of Ion-Exchange Reactions in Hybrid Organic-Inorganic Perovskite Thin Films Studied by *In Situ* Real-Time X-ray Scattering — ●ALESSANDRO GRECO¹, ALEXAN-

DER HINDERHOFER¹, M. IBRAHIM DAR², NEHA ARORA², JAN HAGENLOCHER¹, ANDREY CHUMAKOV³, MICHAEL GRÄTZEL², and FRANK SCHREIBER¹ — ¹Institute for Applied Physics, University of Tübingen, Auf der Morgenstelle 10, 72076 Tübingen, Germany — ²Laboratory of Photonics and Interfaces, Institute of Chemical Sciences and Engineering, École Polytechnique Fédérale de Lausanne, Switzerland — ³European Synchrotron Radiation Facility, 71, Avenue des Martyrs, CS 40220, 38043 Grenoble Cedex 9, France

The exchange of ions in hybrid organic-inorganic perovskites with the general formula APbX₃ (A = MA, FA; X = I, Cl, Br) is studied in five different systems using *in situ* real-time X-ray diffraction. In systems where the organic cation is exchanged, we find a continuous shift of the lattice parameter. The relative shift compared to the pure materials is used to quantify the exchange. Whether or not a conversion is possible, as well as the amount of exchanged cations, depends on the halide used. In the case of the interconversion of MAPbI₃ and MAPbCl₃, we observe a decay of the diffraction peaks of the original perovskite and the emergence of new peaks corresponding to the structure with the alternative halide. Moreover, we determined the relevant time scales of the growth and decay of the perovskite structures. [1]

[1] A. Greco *et al.*, J. Phys. Chem. Lett., **2018**, 6750 (DOI: 10.1021/acs.jpcclett.8b02916)

CPP 19.10 Tue 12:00 H18

Manipulating the excited state of CsPbBr₃ nanoplatelets for superior optical properties — ●THOMAS MORGENSTERN¹, MATTHEW JUROW^{2,3}, CARISSA EISLER², JUN KANG³, MANUEL ENGELMAYER¹, PAUL ALIVISATOS^{2,3}, YI LIU³, and WOLFGANG BRÜTTING¹ — ¹University of Augsburg, Institute of Physics, 86135 Augsburg, Germany — ²College of Chemistry, University of California, Berkeley, California 94720, United States — ³Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States

Lead halide perovskite (LHP) nanocrystals (NCs) and their light emitting features have recently moved into the focus of optoelectronic applications. However many properties of the nanoparticles remain to be investigated for a complete understanding of their optoelectronic attributes. Of crucial importance for lighting applications is the alignment of the emissive transition dipole moment with respect to the substrate normal, which limits the maximum achievable device performance. [1] To investigate this property we measured the photoluminescence radiation pattern of CsPbBr₃ nanoplatelets. As a consequence of the nanocrystal shape, the energy of the emission peak shifts to shorter wavelengths, resulting in a deep blue emission maximum at 460 nm for the investigated nanoplatelets. Further the orientation of the transition dipole moments is confined in space and therefore aligns within the plane of the substrate. Hence the NCs have the potential to outperform existing solutions for blue emitting devices within display and lighting applications. [1] M. JUROW, ET. AL.: *Nano Lett.* **17**(7) (2017), 4534-4540

CPP 19.11 Tue 12:15 H18

Modeling MPa pressure-induced changes in photoluminescence spectra of organic-inorganic halide perovskites — ANDREA CASTELLI¹, ●GIULIA BIFFI^{1,2}, LUCA CESERACCIU¹, DAVIDE SPIRITO¹, MIRKO PRATO¹, DAVIDE ALTAMURA³, CINZIA GIANNINI³, SERGEY ARTYUKHIN¹, ROMAN KRAHNE¹, LIBERATO MANNA¹, and MILENA ARCINIEGAS¹ — ¹Istituto Italiano di Tecnologia, Via Morego 30, 16163 Genova, Italy — ²Dipartimento di Chimica e Chimica Industriale, Università degli Studi di Genova, Via Dodecaneso, 31, 16146, Genova, Italy — ³Istituto di Cristallografia, Consiglio Nazionale delle Ricerche, via Amendola 122/O, 70126 Bari, Italy

High compressive strains have been used as a way to tune physical properties of three-dimensional perovskite crystals. Pressure in the GPa range generates structural distortions in perovskite lattice, affecting the electronic structure and modifying optical and transport properties. Surprisingly, stacks of hybrid organic-inorganic perovskite flakes demonstrate tunability of optical spectra at much lower pressures, in the range of tens of MPa. Pristine flakes are near-white emitting, and *in-situ* photoluminescence experiments during loading and unloading in a mechanical pressure cell reveal drastic change in their optical emission spectrum, particularly an enhancement of the blue emission. Using first principles simulations, we are able to reproduce the main features of the optical properties and to analyze their variations under compressive strain.