

CPP 6: Perovskite and photovoltaics I (joint session HL/CPP)

Time: Monday 9:30–12:30

Location: POT 251

CPP 6.1 Mon 9:30 POT 251

Bandgap engineering of two-step processed perovskite top cells for application in perovskite-based tandem photovoltaics — ●RONJA PAPPENBERGER^{1,2}, ALEXANDER DIERCKS², AHMED FARAG^{1,2}, PAUL FASSL^{1,2}, and ULRICH W. PAETZOLD^{1,2} — ¹Institut für Mikrostrukturtechnologie, KIT, Germany — ²Lichttechnisches Institut, KIT, Germany

Tandem solar cells offer a promising concept of raising the efficiency of silicon solar cells above the theoretical limit of 29%. In this context, silicon is supplemented by a wide-bandgap perovskite top solar cell to make better use of the solar spectrum. Perovskite solar cells come into play given their favorable optoelectronic quality and tunable bandgap. Textured-front perovskite silicon tandem solar cells currently promise the highest energy yield for modules in the field. To avoid shunting, ensure high efficiency and economic production of the perovskite on the μm -sized pyramids, a conformal growth of the perovskite layer as well as a sufficient layer thickness are necessary. A two-step method - containing a separate deposition of the PbI_2 and the organic cations - enables high film quality, flexibility in choice of component/solution and the possibility of upscaling. Here, we investigate different strategies of increasing the bandgap of the perovskite. Thereby the location of the added bromine - cation solution and/or PbI_2 solution - is critical. With our approach, the device performance - PCE of 17.2%, FF of 76% and V_{oc} of 1.156 V ($E_{\text{g}} \approx 1.64$ eV) - and film quality can be maintained. Furthermore, the effect of an increasing bandgap in combination with planar/textured silicon bottom cells is studied.

CPP 6.2 Mon 9:45 POT 251

Application of plasma enhanced atomic layer deposition process of alumina on perovskite film boosts efficiency of solar cells — ●MALGORZATA KOT¹, MAYANK KEDIA², PAUL PLATE³, LUDWIG MARTH³, KARSTEN HENKEL¹, and JAN INGO FLEGE¹ — ¹Applied Physics and Semiconductor Spectroscopy, BTU Cottbus-Senftenberg, Konrad-Zuse-Strasse 1, 03046 Cottbus, Germany — ²Institut fuer Photovoltaik Universitaet Stuttgart, Pfaffenwaldring 47, 70569 Stuttgart, Germany — ³SENTECH Instruments GmbH, Schwarzschildstraße 2, 12489 Berlin, Germany

It is assumed that plasma-enhanced atomic layer deposition (PEALD) cannot be used to prepare thin films on sensitive organic-inorganic perovskites because the plasma destroys the perovskite film and thus deteriorates its photophysical properties. Here, we prove that using an appropriate geometry of the ALD system (SENTECH SI PEALD system) and suitable process parameters it is possible to coat perovskites with alumina by PEALD. Spectromicroscopy followed by electrical characterisation reveal that as long as the PEALD process is not optimized (too long plasma pulses) one gets degradation of the perovskite as well as dissociation of the created iodine pentoxide (during PEALD) under light that causes a valence band maximum (VBM) shift to the Fermi level and thus significantly decreases the solar cell efficiency. However, once the PEALD process parameters are optimized, no VBM shift is observed. Moreover, the solar cell efficiency depends inversely on process temperature and layer thickness.

CPP 6.3 Mon 10:00 POT 251

Tuning Crystallization for Highly Efficient Perovskite Silicon Tandem Solar Cells — ●MOHAMED MAHMOUD^{1,2}, OUSSAMA ER-RAJI^{1,2}, PATRICIA SCHULZE¹, ANNA JULIANE BORCHERT^{1,2}, and ANDREAS W. BETT^{1,2} — ¹Fraunhofer ISE — ²University of Freiburg

Perovskite solar cells have the advantages of a strong absorption edge, defect tolerance, and potential cheap production due to easy production methods such as spin coating or slot-die coating as a highly scalable production method. In the industry, double-sided textured (micro-meter sized pyramid) silicon is commonly produced to decrease reflection losses and improve light trapping. Solution-based processing methods of perovskite on top of the textured Si showed low conformality, which resulted in shunts and non-working solar cells. To overcome this issue, the hybrid route was developed, in which inorganic precursors are co-evaporated using the thermal vapor deposition technique and then organic precursors are spin-coated followed by a thermal annealing. By doing that, high conformality of perovskite thin films on top of the textured silicon is achieved. However, the resulting perovskite grain size is rather low, which can lower the bulk quality. In

this work, various additives were used to increase the grain size, and their working mechanisms were studied. In addition, we study the consequences of different grain sizes at the tandem level with respect to device efficiency as well as stability. Moreover, using the thermodynamics fundamentals of crystallization, we hypothesize for the first time a common general explanation for the working mechanism of all the different additives used.

CPP 6.4 Mon 10:15 POT 251

Efficient Modeling Workflow for Accurate Electronic Structures of Hybrid Perovskites — JULIAN GEBHARDT^{1,2}, ●WEI WEI^{1,2}, and CHRISTIAN ELSÄSSER^{1,2,3} — ¹Fraunhofer IWM, 79108 Freiburg — ²Cluster of Excellence livMatS, University of Freiburg — ³Freiburg Materials Research Center, University of Freiburg

Hybrid organic-inorganic halide perovskites are the most promising photovoltaic absorber materials to substitute or complement silicon in high-efficiency solar cells. These hybrid materials are often constrained by their low stability and critical elements like lead. Computational high-throughput screening studies, based on solid-state electronic-structure theory, are useful to identify promising substitute materials with targeted properties. In this work, we present an efficient computational approach based on density-functional theory, which is suitable to predict band gaps for arbitrary compounds reliably and in good quantitative agreement with experimental band gap data for known compounds. This approach is described and demonstrated for the building blocks of one of the most promising hybrid perovskites, namely, $(\text{HC}(\text{NH}_2)_2)_x\text{Cs}_{1-x}\text{Pb}(\text{I}_y\text{Br}_{1-y})_3$, with x and y varied between zero and one.

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CPP 6.5 Mon 10:30 POT 251

Photon Management for Ultrathin Solar Cells: Enabling Waveguide Modes by Structured Back Contact — ●MERVE DEMIR, THOMAS SCHNEIDER, TORSTEN HÖLSCHER, HEIKO KEMPA, and ROLAND SCHEER — Martin-Luther-Universität Halle-Wittenberg, Germany

The recent research based on $\text{Cu}(\text{In,Ga})\text{Se}_2$ (CIGSe) solar cells is focused on thinning down the absorber layer to enable less material consumption and cost effective large scale production. However, having ultra-thin CIGSe solar cells with absorber layer thickness in sub-micron level brings the cost of limited absorption of solar spectra and hence leads to lower energy conversion efficiencies. This problem can be overcome by the cell architecture including functional back contact elements for the enhancement of optical absorption. In this contribution, ultrathin CIGSe solar cells with 500 nm thick absorber layer were combined with nano-textured SiO_2 back contacts together with aluminum back mirror. With this cell design, it is aimed to have increased power conversion efficiency for ultra-thin CIGSe solar cells due to enhanced absorption of long wavelength region photons. The solar cell parameters were extracted and compared with the conventional CIGSe back contact, flat molybdenum, to reveal the effects of functional back contact. The experimental findings on quantum efficiency measurements prove the positive effects of having highly reflective and textured back contact. Furthermore, the growth of CIGSe on textured substrates was examined throughout cross section cuts by scanning electron microscopy and energy dispersive X-ray diffraction.

30 min. break

CPP 6.6 Mon 11:15 POT 251

Employing three-dimension structure analysis: digital twin for studying grain boundary effects in thin film solar cells — ●CHANG-YUN SONG¹, MATTHIAS MAIBERG¹, HEIKO KEMPA¹, ALI GHOLINIA², WOLFRAM WITTE³, DIMITRIOS HARISKOS³, DANIEL ABOU-RAS⁴, and ROLAND SCHEER¹ — ¹Martin-Luther-University Halle-Wittenberg, Halle, D — ²University of Manchester, Manchester, UK — ³Zentrum für Sonnenenergie- und Wasserstoff-Forschung, Stuttgart, D — ⁴Helmholtz-Zentrum Berlin, Berlin, D

Grain boundaries (GBs) in polycrystalline $\text{Cu}(\text{In,Ga})\text{Se}_2$ (CIGSe), are believed to be one of the performance limiting factors of current record efficiency CIGSe solar cells. Numerous simulation studies have been conducted to unveil their effects. Most simulations, however, have

been done within two-dimensions (2D), thereby presumably using the simplified GBs shapes. In this study, we thus present a realistic three-dimensional (3D) GB model for a high-efficiency CIGSe layer. To this end, a combination of electron backscatter diffraction and focused ion beam was applied to obtain 3D data of the CIGSe layer, which then allowed the reconstruction of the 3D grain structure into a computer model. By using the computer model as input for 3D optoelectronic simulations, we study the electronic effects of GBs on the high-efficiency solar cell under investigation bulk parameter values for the simulations were obtained through a combination of simulation and experiments, such that the solar cell is consistently described. As an outcome, the 3D simulations confirm that the effect of GB was indeed underestimated in earlier conventional 2D simulations.

CPP 6.7 Mon 11:30 POT 251

Interface Engineering to reduce non-radiative recombination losses at the perovskite/C60 interface in monolithic perovskite silicon tandem solar cells — ●JOHANNA MODES, PATRICIA S. C. SCHULZE, KAITLYN MC MULLIN, MARYAMSADAT HEYDARIAN, CHRISTOPH MESSMER, JULIANE BORCHERT, and ANDREAS BETT — Fraunhofer ISE

Metal halide perovskites have emerged in recent years as a promising absorber material for solar cells with the potential to combine high power conversion efficiency with low production costs. However, significant non-radiative charge carrier recombination occurs at the perovskite interface to the contacts, thus preventing the full potential of the solar cell from being exploited. Photoluminescence quantum yield measurements clearly show that the Quasi-Fermi level splitting is reduced by evaporation of the electron contact C60 onto perovskites, leading to limited open-circuit voltage in devices. In recent literature, as well as in our investigations, different passivation layers are deposited between perovskite and the electron contact to reduce non-radiative recombination and to improve the open-circuit voltage. This is on the one hand pursued by increasing the selectivity at the contacts through field effects and band alignment and secondly by reducing defects at the interface through chemical passivation.

CPP 6.8 Mon 11:45 POT 251

Spontaneous Polarization in NaNbO_3 — ●KISUNG KANG¹, SAUD BIN ANOOZ², JUTTA SCHWARZKOPF², MATTHIAS SCHEFFLER¹, and CHRISTIAN CARBOGNO¹ — ¹The NOMAD Laboratory at the FHI of the Max-Planck-Gesellschaft and IRIS-Adlershof of the Humboldt-Universität zu Berlin — ²Leibniz-Institut für Kristallzüchtung (IKZ)

The perovskite NaNbO_3 is regarded as a promising lead-free piezoelectric material, also because its polarization properties can be further tailored via strain engineering. [1] In thin films, lattice strain can be incorporated by the heteroepitaxial growth on lattice mismatch. By tuning the epitaxial strain in the films, different polymorphs with distinctively different polarization strength and orientation can be realized [2, 3]. We investigate this question by using density-functional theory at the semi-local level of theory, which we carefully validate with hybrid-functional calculations. By this means, we compute the spontaneous polarization for ten phases of NaNbO_3 as a function of stress and strain. In line with experiments, we confirm that the monoclinic Pm phase features a non-vanishing in-plane polarization, the orientation of which is independent of the strain. Conversely, the polarization direction of the orthorhombic $Pmc2_1$ phase depends on the applied tensile strain. We analyze the underlying electronic and atomistic mechanism and discuss how the relevant properties are influenced

by phase transformations.

[1] N. Bein, *et al.*, *Phys. Rev. Mater.* **6**, 084404 (2022).

[2] J. Schwarzkopf, *et al.*, *J. Appl. Cryst.* **45**, 1015 (2012).

[3] S. B. Anooz, *et al.*, *Appl. Phys. Lett.* **120**, 202901 (2022).

CPP 6.9 Mon 12:00 POT 251

Peculiar bond length dependence and its impact on the band gap bowing in $(\text{Ag,Cu})(\text{In,Ga})\text{Se}_2$ thin film alloys — ●HANS H. FALK¹, STEFANIE ECKNER¹, KONRAD RITTER¹, SERGIU LEVCENKO¹, TIMO PFEIFFELMANN¹, EDMUND WELTER², JES LARSEN³, WILLIAM N. SHAFARMAN⁴, and CLAUDIA S. SCHNOHR¹ — ¹Felix Bloch Institute for Solid State Physics, Leipzig University, Germany — ²Deutsches Elektronen-Synchrotron DESY, Germany — ³Department of Materials Science and Engineering, Uppsala University, Sweden — ⁴Department of Materials Science and Engineering, University of Delaware, USA

Incorporation of Ag into $\text{Cu}(\text{In,Ga})\text{Se}_2$ thin film solar cells improves several of their properties. However, with increasing Ag content, the band gap of $(\text{Ag,Cu})\text{GaSe}_2$ increases even though the lattice expands and the Ga-Se bond length is predicted to decrease. This is counter-intuitive, since in other chalcopyrite alloys all bond lengths increase and the band gap decreases as the lattice expands. Therefore, we studied the element-specific average bond lengths of $(\text{Ag,Cu})\text{GaSe}_2$, $(\text{Ag,Cu})\text{InSe}_2$ and $\text{Ag}(\text{In,Ga})\text{Se}_2$ using X-ray absorption spectroscopy for thin films grown on Mo-coated soda lime glass by a single stage co-evaporation process. As predicted, the Ga-Se bond length decreases with increasing Ag content in $(\text{Ag,Cu})\text{GaSe}_2$. While the In-Se bond length of $(\text{Ag,Cu})\text{InSe}_2$ shows the same behavior, $\text{Ag}(\text{In,Ga})\text{Se}_2$ exhibits a dependence similar to that of $\text{Cu}(\text{In,Ga})\text{Se}_2$, demonstrating that the peculiar behavior is related to mixing the group-I lattice site. Using the bond lengths we model the anion positions and estimate their effect on the band gap bowing.

CPP 6.10 Mon 12:15 POT 251

Fast diffusion of spin polarized excitons in bulk lead halide perovskites — ●SERGIU ANGHEL¹, DMITRI R. YAKOVLEV¹, DMITRY N. DIRIN², MAKSYM V. KOVALENKO², MANFRED BAYER¹, and MARKUS BETZ¹ — ¹Experimentelle Physik 2, Technische Universität Dortmund, Otto-Hahn-Straße 4a, D-44227 Dortmund, Germany — ²Laboratory of Inorganic Chemistry, Department of Chemistry and Applied Biosciences, ETH Zürich, Zürich CH-8093, Switzerland

We investigate the spin diffusion of the free "hot" excitons in $\text{FA0.9Cs0.1PbI2.8Br0.2}$ bulk lead halide perovskite crystal at cryogenic temperatures by employing ultrafast time- and spatial-resolved magneto-optical Kerr microscopy. We measure the spin diffusion coefficient (D_s) of the free excitons of $D_s \sim 50 \text{ cm}^2/\text{s}$, which is a very surprising outcome, especially in the light of the recent results obtained on bulk or two-dimensional perovskites [1,2], where D_s is at least two orders of magnitude lower. D_s shows a roughly linear dependence on pump energy whereas the dependence on pump power is much more intricate - after a certain pump power threshold we observe an anomalous, nonlinear spatial dependence of D_s . We discuss our findings in view of efficient exciton-phonon coupling and Auger processes.

[1] A. Baldwin, G. Delport, K. Leng, R. Chahbazian, K. Galkowski, K. P. Loh, and S. D. Stranks, *J. Phys. Chem. Lett.* **12**, 4003 (2021).

[2] S. D. Stranks, G. E. Eperon, G. Grancini, C. Menelaou, M. J. P. Alcocer, T. Leijtens, L. M. Herz, A. Petrozza, and H. J. Snaith, *Science* **342**, 341 (2013).