

CPP 61: Perovskite and Hybrid Photovoltaics II (joint session HL/CPP)

Time: Thursday 15:00–17:30

Location: H36

CPP 61.1 Thu 15:00 H36

Valence band structure of CsPbBr₃ inorganic perovskite — ●JANEK RIEGER¹, DANIEL NIESNER¹, ANDRII KANAK², PETRO FOCHUK², IEVGEN LEVCHUK³, CHRISTOPH BRABEC³, and THOMAS FAUSTER¹ — ¹Lehrstuhl für Festkörperphysik, University of Erlangen-Nürnberg, Staudtstr. 7, D-91058 Erlangen, Germany — ²Department of General Chemistry and Material Science, Yuriy Fedkovych Chernivtsi National University, Kotsjubynskyi St. 2, Chernivtsi, 58012, Ukraine — ³I-MEET, Department of Materials Science and Engineering, University of Erlangen-Nürnberg, Martensstr. 7, D-91058 Erlangen, Germany

The impact of the organic ion on the electronic properties of lead halide perovskites is matter of an ongoing debate. With angle-resolved photoelectron spectroscopy we investigated the properties of the valence bands of different perovskite single crystals cleaved in ultra-high vacuum. The talk focusses on recent results from the inorganic perovskite CsPbBr₃. Measurements were carried out in the temperature range from 87 K to 350 K, covering all bulk phase transitions of the material. The width of the valence band and the dispersion of the valence band maximum are discussed. Results are compared to the valence band structure of the organic-inorganic perovskite (HC(NH₂)₂)PbBr₃ as well as to (CH₃NH₃)PbBr₃, which was already investigated in detail by our group [1].

[1] D. Niesner et al., Phys. Rev. Lett. **117**, 126401 (2016).

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Charge Recombination Dynamics in Defect-Engineered Hybrid Organic-Inorganic Halide Perovskites — ●WEN-YU CHENG, CHANG-MING JIANG, and IAN SHARP — Walter Schottky Institut und Physik Department, Technische Universität München, 85748 Garching, Germany

Organic-inorganic halide perovskites, with their ease of fabrication and bandgap-tunability, provide promising prospects in photovoltaic and light-emitting diode applications. While such materials are highly tolerant of defects, the presence of ionic, along with electronic, charge conduction can result in dynamic optoelectronic properties. In this work, we aim at understanding the charge recombination pathways in defect-engineered perovskites. Point defects are intentionally and controllably introduced into the lattice of methylammonium lead iodide (MAPbI₃), and a series of thin films with tunable bandgaps from 1.60 to 1.95 eV are fabricated. The concentrations and energetic positions of defects are quantified by photothermal deflection spectroscopy. The effects of these defects on photoluminescence yield are studied by steady-state and time-resolved fluorescence techniques. In addition to shedding light on the interactions between organics cations and the inorganic lattice, the results provide insights into the factors that underlie defect tolerance in hybrid halide perovskite semiconductors.

CPP 61.3 Thu 15:30 H36

Reversible Bandgap Instabilities in Multiple-Cation Mixed-Halide Perovskite Solar Cells — ●FABIAN RUF¹, PASCAL RIETZ¹, MELTEM F. AYGÜLER², PABLO DOCAMPO³, HEINZ KALT¹, and MICHAEL HETTERICH^{1,4} — ¹Institute of Applied Physics, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany — ²Department of Chemistry and CeNS, University of Munich (LMU), 81377 München, Germany — ³Physics Department, School of Electrical and Electronic Engineering, Newcastle University, Newcastle upon Tyne, NE1 7RU, United Kingdom — ⁴Light Technology Institute, Karlsruhe Institute of Technology (KIT), 76131 Karlsruhe, Germany

Multiple-cation mixed-halide perovskites have demonstrated their great potential as promising candidates for next-generation thin-film photovoltaics due to further improved power-conversion efficiency and stability. In this contribution, we investigate the stability of the compositional and resulting electronic structure of Cs_{0.05}(FA_{0.83}MA_{0.17})_{0.95}Pb(I_{0.83}Br_{0.17})₃ solar cell absorbers using electroreflectance (ER) spectroscopy as sensitive and non-invasive measurement technique. Despite the enhanced compositional stability, these compounds show a decrease of the bandgap up to 70 meV under AM1.5 illumination and applied voltage. We attribute this to segregation of the intermixed perovskite into iodide-rich and bromide-rich domains which is also confirmed by in-situ X-ray diffraction. A detailed study under operation-relevant conditions (illumination and

bias) is conducted in various environments (different oxygen contents and relative humidity) to evaluate the consequences for photovoltaic applications.

CPP 61.4 Thu 15:45 H36

Exciton Spin Dynamics and Their Dependence on Monovalent Cation Dipole Moment in Layered 2D Metal-Halide Perovskites — ●SEAN BOURELLE, RAVICHANDRAN SHIVANNA, and FELIX DESCHLER — University of Cambridge

Strong spin-orbit coupling in metal-halide perovskites connects optical transitions to spin states via the conservation of total angular momentum. These self-assembled quantum well structures of high-performance, defect tolerant semiconductors are broadly tunable through chemical composition. Here, we use ultrafast circularly-polarised broadband transient absorption spectroscopy to study the effect of perovskite composition on the polarisation and dynamics of exciton spin-states in two-dimensional (2D) Ruddlesden-Popper perovskites. We spectrally resolve the dynamic circular dichroism from a photoinduced polarisation in the secondary total angular momentum quantum number. This dichroism is used to determine the impact of the monovalent cation's dipole moment on the exciton spin-state depolarisation time.

CPP 61.5 Thu 16:00 H36

Vacuum-deposited Bismuth and 2D perovskites as absorber material for solar cells — ●MARTIN KROLL — TU Dresden, IAPP, Dresden Deutschland

Organic-inorganic halide perovskites are a fast developing absorber material class for thin-film solar cells. The state of the art material combinations with record efficiencies all contain methyl-ammonium (MA) and lead as cations, which both display unfavorable properties. Cells with methyl-ammonium as organic cation all show rapid degradation due to its instability under exposure to light and moisture. Lead is known for its toxicity, which is bound to cause problems during cell marketing and legislation. In order to circumvent these issues, current research discusses less toxic material combinations like MA/Cesium-Bismuth-Iodide as well as self-layering 2D perovskites for better stability, which are formed by introducing large organic cations like butyl-ammonium (BA). So far, these approaches have only been shown using solution processing. We show the prospects of multi-source vacuum evaporation, which excels at material purity and film control. For MA/Cs₃Bi₂I₉ as well as BA/MAPbI₃, we present XRD and absorption data for thin-films as well as functional solar cells with efficiencies of > 1% and > 10%, respectively.

15 min. break

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The missing long range order in point-dipole based hybrid perovskite models — ●MENNO BOKDAM and JONATHAN LAHNSTEINER — University of Vienna, Faculty of Physics and Center for Computational Materials Sciences, Vienna, Austria

The crystal structure of hybrid perovskites forms an intricate electrostatic puzzle. Using density functional theory (DFT) calculations we study the ordering of A={MA, Cs} cations in the APbI₃ perovskite framework. We show that previously proposed model Hamiltonians do not capture the long range structural order observed in large scale *ab-initio* molecular dynamics calculations. We attempt to improve on these models by applying an Ewald summation to sum the dipole-dipole interaction and by introducing a distance dependent screening function, however with only limited success. We envision that well-trained force fields would be able, and are likely necessary, to fully describe the finite temperature behaviour of hybrid perovskites.

CPP 61.7 Thu 16:45 H36

Recent progress in the vapour deposition of organic-inorganic hybrid metal-halide perovskite solar cells — ●JULIANE BORCHERT¹, IEVGEN LEVCHUK², LAVINA C. SNOEK¹, MATHIAS ULLER ROTHMANN¹, HENRY J. SNAITH¹, LAURA M. HERZ¹, CHRISTOPH J. BRABEC², and MICHAEL B. JOHNSTON¹ — ¹Clarendon Laboratory, Department of Physics, University of Oxford — ²Materials for Electronics and Energy Technology (i-MEET), FAU Erlangen-

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Hybrid metal-halide perovskite semiconductors are promising absorber materials for single junction and as well as for tandem solar cells. They have attracted a lot of research attention in recent years, due to the rapid rise of solar cell efficiencies for these materials. Record efficiencies have now reached to above 23%. Co-evaporation of perovskite thin-films for solar cells offers many advantages such as precise thickness control, pinhole free planar films and compatibility with a large range of different substrates. Efficient co-evaporated solar cells have been demonstrated with different perovskite materials, for example methylammonium lead iodide (MAPbI₃) and formamidinium lead iodide (FAPbI₃). Furthermore co-evaporation is a promising technique for the upscaling of perovskite solar cells to commercial scales. Some challenges remain, including problems with the process control and relatively small crystallites in the deposited films. Additionally, it is more challenging to achieve mixed compositions with co-evaporation than with solution processing. Here we present recent progress made towards solving these challenges.

CPP 61.8 Thu 17:00 H36

Growth Monitoring and Evolution of Optoelectronic Properties during the Formation of CH₃NH₃PbI₃-xCl_x — ●KLARA SUCHAN¹, JUSTUS JUST¹, EVA L. UNGER^{1,2}, JOSÉ MARQUEZ PRIETO², and THOMAS UNOLD² — ¹Lund University, Paradisgatan 2, 22350 Lund, Sweden. — ²Helmholtz-Zentrum Berlin, Hahn-Meitner-Platz 1, 14109 Berlin, Germany

We present a detailed investigation of the formation mechanism and the evolution of optoelectronic properties during annealing of chlorine derived methylammonium lead iodide (MAPIC). MAPIC layers are prepared by a one-step solution based process. While solution based processes do not need cost intensive vacuum technology and no high-temperature annealing steps, little is known about the exact mechanisms during synthesis. This prohibits identification and exact control of all relevant variables, leading to widely reported poor reproducibility. Using in-situ optical reflection we are able to monitor the evolution

of the MAPIC phase as a function of time and processing temperature. The extracted formation kinetics are fitted using an altered Johnson-Mehl-Avrami-Kolmogorov model and show that the presence of Chlorine induces a delayed formation of MAPIC. This is verified by X-ray diffraction and X-ray fluorescence measurements. From absolute photoluminescence measurements we determine the quasi-Fermi level splitting during film formation. Correlating structural and electronic properties, we will present a detailed view on the reaction route during synthesis and specifically the influence of annealing conditions on the process and its intermediate states

CPP 61.9 Thu 17:15 H36

The versatility of polyelemental perovskite compositions — ●MICHAEL SALIBA — University of Fribourg, Switzerland

Perovskite solar cells (PSCs) have created much excitement in the past years and attract spotlight attention from research groups all over the world with many thousands of publications every year. This talk will provide an overview on the reasons for this unique success story highlighting the historic development as well as the specific material properties that make perovskites so attractive for the research community.

The current challenges are exemplified using a high-performance model systems for PSCs (multication Rb, Cs, methylammonium (MA), formamidinium (FA) perovskites).(1,2) The triple cation (Cs, MA, FA) achieves power conversion efficiencies (PCEs) close to 21% due to suppressed phase impurities. This results in more robust materials enabling breakthrough reproducibility. Through multication engineering, the seemingly too small Rb can be integrated (unsuited as a single-cation perovskite).(2) This results in a stabilized efficiency of 21.6%with one of the smallest differences between band gap and voltage ever measured for any PV material. Polymer-coated cells maintained 95% of their initial performance at elevated temperature for 500 hours under working conditions, a crucial step towards industrialisation of PSCs. The last part elaborates on a roadmap on how to extend the multication to multicomponent engineering providing a series of new compounds that are highly relevant candidates for the coming years.