

## HL 29: Hybrid and Perovskite Photovoltaics II (Joint session of CPP, DS and HL, organized by CPP)

Time: Tuesday 10:45–13:00

Location: H37

HL 29.1 Tue 10:45 H37

**Charge Carrier Recombination Dynamics in Perovskite and Polymer Solar Cells probed by Time-Delayed Collection Field (TDCF) Experiments** — ●ANDREAS PAULKE<sup>1</sup>, SAMUEL D. STRANKS<sup>2</sup>, JULIANE KNIEPERT<sup>1</sup>, JONA KUPIERS<sup>1</sup>, CHRISTIAN M. WOLFF<sup>1</sup>, NATALIE SCHÖN<sup>1</sup>, HENRY J. SNAITH<sup>2</sup>, THOMAS J.K. BRENNER<sup>1</sup>, and DIETER NEHER<sup>1</sup> — <sup>1</sup>Institut für Physik und Astronomie, Universität Potsdam, Karl-Liebknecht-Str.24-25, 14476 Potsdam — <sup>2</sup>Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, United Kingdom

Time-Delayed Collection Field (TDCF) experiments are applied to organometal halide perovskite ( $CH_3NH_3PbI_3$ ) based solar cells to investigate charge carrier recombination in a working solar cell at the nanosecond to microsecond timescale. Planar perovskite solar cells ( $ITO/PEDOT : PSS/Perovskite/PCBM/C_{60}/BCP/Al$ ) display a second-order recombination process with a slow-down of the apparent recombination coefficient over several tens of nanoseconds. In contrast, recombination in the  $ITO/TiO_2/mesoporous - TiO_2/Perovskite/Spiro-OMeTAD/Au$  device is governed by a slow first order process, but again with an apparent time-dependence of the recombination coefficient. We also conclude that organometal halide perovskite solar cells differ significantly from prototypical organic bulk heterojunction devices with regard to the mechanism and time-scale of free carrier recombination.

HL 29.2 Tue 11:00 H37

**Recombination of photogenerated charge carriers in planar methylammonium lead halide perovskite solar cells** — ●DAVID KIERNASCH<sup>1</sup>, STEFAN VÄTH<sup>1</sup>, KRISTOFER TVINGSTEDT<sup>1</sup>, ANDREAS BAUMANN<sup>2</sup>, and VLADIMIR DYAKONOV<sup>1,2</sup> — <sup>1</sup>Experimental Physics VI, Julius Maximilian University of Würzburg, 97074 Würzburg — <sup>2</sup>Bavarian Center for Applied Energy Research (ZAE Bayern) Bayern, 97074 Würzburg

In the last years, solar cells based on organo-metal halide perovskites gained a lot of attention. The efficiency of solar cells with a perovskite absorber nowadays reaches more than 20%. With charge carrier mobilities up to tens of  $cm^2V^{-1}s^{-1}$  and diffusion lengths exceeding  $1 \mu m$  transport doesn't seem to be a limiting issue in this new kind of photovoltaic technology. However, recombination of the photogenerated charge carriers is an important factor defining e.g. the open circuit voltage of the solar cell. We prepared different methylammonium lead halide perovskite solar cells in planar configuration. Addressing the recombination dynamics, lifetime and concentration of photogenerated charge carriers are studied by transient photovoltage, charge extraction experiments as well as by the technique of open circuit voltage decay. We measured charge carrier lifetimes in the sub- $\mu s$ -range for high illumination intensities ( $\geq 1$  sun), which is lower than in state of the art bulk heterojunction cells like P3HT:PCBM. Furthermore we discuss our results for different device morphologies and light-absorbing materials.

HL 29.3 Tue 11:15 H37

**Correlating charge carrier mobility, morphology and efficiency in hybrid halide perovskite photovoltaic devices** — ●IRENE GRILL<sup>1,2</sup>, MICHIEL PETRUS<sup>1,2</sup>, NADJA GIESBRECHT<sup>1,2</sup>, THOMAS BEIN<sup>1,2</sup>, PABLO DOCAMPO<sup>1,2</sup>, MATTHIAS HANDLOSER<sup>1,2</sup>, and ACHIM HARTSCHUH<sup>1,2</sup> — <sup>1</sup>Department of Chemistry and CeNS, LMU Munich — <sup>2</sup>Nanosystems Initiative Munich (NIM)

Hybrid perovskites currently represent one of the most promising material systems for incorporation in future solar cell devices since their efficiencies increased enormously in the last few years [1,2]. To date, fundamental physical properties including charge carrier dynamics and transport in these materials are not completely understood and are therefore at the focus of intense research. Here we extract the mobility of charge carriers in working thin film solar cells based on perovskite absorber layers and correlate it to the efficiency of the respective devices. To this end we performed Time-of-flight (ToF) studies on different perovskite thin films serving as photoactive layers. Further, in order to analyze the influence of contacts and other interfaces on charge transport and to identify possible optimization steps in the stacked architecture we carried out additional ToF measurements on each of

the individual layers. Our results are discussed in terms of respective device efficiencies, morphologies and optical properties, allowing for a detailed investigation and identification of the limiting factors for the mobility and the efficiency in perovskite based thin film devices.

[1] M.A. Green and T. Bein, *Nature Mater.* 2015, 14, 559-561.[2] N. Jeon et al., *Nature* 2015, 517, 476-480.

HL 29.4 Tue 11:30 H37

**Analysis of electronic trap states in methylammonium lead halide perovskite solar cells via thermally stimulated current** — ●PHILIPP RIEDER<sup>1</sup>, ANDREAS BAUMANN<sup>1,2</sup>, STEFAN VÄTH<sup>1</sup>, KRISTOFER TVINGSTEDT<sup>1</sup>, and VLADIMIR DYAKONOV<sup>1,2</sup> — <sup>1</sup>Experimental Physics VI, Julius Maximilian University of Würzburg, 97074 Würzburg — <sup>2</sup>Bavarian Center for Applied Energy Research ZAE, 97074 Würzburg

Organolead halide perovskite solar cells have emerged as one of the most promising technologies in thin-film photovoltaics due to their extraordinary increase in performance in only six years. Yet, the working principles of this material class still lack fundamental understanding. To address the possible influence of electronic traps on device performance, we probed solution processed methylammonium lead halide perovskite solar cells via thermally stimulated current (TSC) analysis.[1] Thereby, the sample is heated from 10K to 300K, while monitoring the current flow. This current is attributed to charge carriers being released from previously filled trap states in the semiconductor, allowing drawing conclusions about their distribution and energetic depth. Signals detected at low as well as at high temperatures can be attributed to shallow and deep traps, accordingly. Furthermore, a peak at around  $T=162K$  can be assigned to the reported structural phase transition of the perovskite crystal from orthorhombic to tetragonal crystal lattice structure.

[1] A. Baumann et al., *J. Phys. Chem. Lett.* 6, 2350 (2015)

### 15 min. break

HL 29.5 Tue 12:00 H37

**Tunable ferroelectric polarization and its interplay with spin-orbit coupling in tin iodide perovskite** — ALESSANDRO STROPPA<sup>1</sup>, ●DOMENICO DI SANTE<sup>2</sup>, PAOLO BARONE<sup>1</sup>, MENNO BOKDAM<sup>3</sup>, GEORG KRESSE<sup>3</sup>, CESARE FRANCHINI<sup>3</sup>, MYUNG-HWAN WHANGBO<sup>4</sup>, and SILVIA PICOZZI<sup>1</sup> — <sup>1</sup>CNR-SPIN L'aquila, Italy — <sup>2</sup>CNR-SPIN L'aquila, Italy and Wuerzburg University — <sup>3</sup>Faculty of Physics, Center for Computational Materials Science, University of Vienna, Wien, Austria — <sup>4</sup>Department of Chemistry, North Carolina State University, USA

Ferroelectricity is a potentially crucial issue in halide perovskites, breakthrough materials in photovoltaic research. Using density functional theory simulations and symmetry analysis, we show that the lead-free perovskite iodide (FA)SnI<sub>3</sub>, containing the planar formamidinium cation FA, (NH<sub>2</sub>CHNH<sub>2</sub>)<sup>+</sup>, is ferroelectric. In fact, the perpendicular arrangement of FA planes, leading to a weak polarization, is energetically more stable than parallel arrangements of FA planes, being either antiferroelectric or strong ferroelectric. Moreover, we show that the weak and strong ferroelectric states with the polar axis along different crystallographic directions are energetically competing. Intriguingly, the relatively strong spin-orbit coupling in noncentrosymmetric (FA)SnI<sub>3</sub> gives rise to a co-existence of Rashba and Dresselhaus effects and to a spin texture that can be induced, tuned and switched by an electric field controlling the ferroelectric state.

A. Stroppa, D. Di Sante et al., *Nature Commun.* 5, 5900 (2014)

HL 29.6 Tue 12:15 H37

**Mixed Pb:Sn methyl-ammonium halide perovskites: Thermodynamic stability and optoelectronic properties** — ●LARS WINTERFELD, KSENIA KORSHUNOVA, WICHARD J.D. BEENKEN, and ERICH RUNGE — Institut für Physik, Technische Universität Ilmenau, 98693 Ilmenau, Germany

Using density functional theory, we investigate systematically mixed  $MA(Pb : Sn)X_3$  perovskites, where  $MA$  is  $CH_3NH_3^+$ , and  $X$  is  $Cl$ ,  $Br$  or  $I$ . Our results cover optoelectronic properties, structural and thermodynamic stability. Ab initio calculations of the orthorhombic,

tetragonal and cubic perovskite phases show that the substitution of lead by tin has a much weaker influence on both structure and cohesive energies than the substitution of the halogen. The thermodynamic stability of the  $MA(Pb : Sn)X_3$  mixtures at finite, non-zero temperatures is studied within the Regular Solution Model. We predict that it will be possible to create iodide mixtures at any temperature. Mixing is unlikely for the low-temperature phase of bromide and chloride compounds, where instead local clusters are more likely to form. We further predict that in the high-temperature cubic phase,  $Pb$  and  $Sn$  compounds will mix for both  $MA(Pb : Sn)Br_3$  and  $MA(Pb : Sn)Cl_3$  due to the entropy contribution to the Helmholtz free energy. We calculated optoelectronic properties using both DFT and post-DFT methods (including self-consistent GW) with and without spin orbit coupling. Interestingly, the optoelectronic properties are not just a linear combination of the non-mixed parent structures and are not limited by the non-mixed values, which allows band gap engineering.

HL 29.7 Tue 12:30 H37

**A model Hamiltonian for perovskite solar cells** — ●MARTIN SCHLIPF, MARINA R. FILIP, MIGUEL A. PÉREZ-OSORIO, and FELICIANO GIUSTINO — Department of Materials, University of Oxford, Parks Road, Oxford OX1 3PH, United Kingdom

Hybrid organic-inorganic halide perovskites emerge as one of most promising materials for new solar cells based on their high energy-conversion efficiency. The most commonly investigated materials include a large spin-orbit coupling, which may give rise to Rashba and Dresselhaus effects and makes them suitable for spintronic applications. In this contribution, we develop a simple model Hamiltonian that allows to study the impact of the spin-orbit coupling on the band structure. In particular, we focus on the physics near the bottom of the conduction band. Using group-theoretical considerations, we can reduce the number of parameters that the model exhibits. We ap-

ply this model to the prototype material  $MAPbI_3$  ( $MA = CH_3NH_3$ ) and show how different parameters of the model can be obtained from first-principles density functional theory (DFT) calculations. We discuss how the model can assist in designing improved perovskite solar cells.

HL 29.8 Tue 12:45 H37

**Stability and electronic properties of novel perovskites for photovoltaics from high-throughput ab initio calculations** — ●SABINE KÖRBEL<sup>1,2</sup>, MIGUEL A L MARQUES<sup>2,3</sup>, and SILVANA BOTTI<sup>1,2</sup> — <sup>1</sup>Institut für Festkörpertheorie und -optik, Friedrich-Schiller-Universität Jena, Max-Wien-Platz 1, 07743 Jena, Germany — <sup>2</sup>Institut Lumière Matière, UMR5306 Université Lyon 1-CNRS, F-69622 Villeurbanne Cedex, France — <sup>3</sup>Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, D-06099 Halle, Germany

Using a high-throughput approach based on density functional theory, we perform an extensive search for stable  $ABX_3$  perovskites, where  $X$  is a non-metallic anion and  $A$  and  $B$  are cations spanning the largest portion of the periodic table. Our search is motivated by the necessity to improve the thermodynamic stability of hybrid organo-metal halide perovskite absorbers. We calculate the ternary phase diagram for each composition and we discuss the thermodynamic stability of the perovskite phases. We find a large number of  $ABX_3$  perovskites which are still absent from databases, and which are stable with respect to decomposition into known ternary, binary or elementary phases. For these structures, we then calculate electronic band gaps, hole effective masses, and the spontaneous ferroelectric polarization as relevant material properties for an application as photovoltaic absorbers. We find several novel perovskites which exhibit promising properties for photovoltaic applications. Based on our findings, we discuss possible strategies to improve the thermodynamic stability of perovskite absorbers.