

CPP 2: Hybrid and Perovskite Photovoltaics I

Time: Monday 9:30–12:45

Location: H18

Invited Talk

CPP 2.1 Mon 9:30 H18

The perovskite/transport layer interfaces dominate non-radiative recombination in efficient perovskite solar cells —•MARTIN STOLTERFOHT¹, PIETRO CAPRIOGLIO¹, CHRISTIAN WOLFF¹, JOSE MARQUEZ², THOMAS KIRCHARTZ³, THOMAS UNOLD², and DIETER NEHER¹ — ¹University of Potsdam — ²Helmholtz-Zentrum-Berlin — ³Forschungszentrum Jülich

Charge transport layers (CTLs) are key components of diffusion controlled perovskite solar cells, however they can induce additional non-radiative recombination pathways. By measuring the quasi-Fermi level splitting (QFLS) of perovskite/CTL heterojunctions, we quantify the interfacial recombination current for a wide range of commonly used CTLs, including various hole-transporting polymers, spiro-OMeTAD, metal oxides and fullerenes. We find that all studied CTLs limit the open-circuit voltage (Voc) by inducing a recombination current that is significantly larger than the loss in the neat perovskite and that the least-selective interface sets the upper limit for the device Voc. The results also show that the Voc equals the internal QFLS in the absorber layer of pin and nip-type cells with selective CTLs and power conversion efficiencies of up to 21.4% (in pin). However, in case of less selective CTLs, the Voc is substantially lower than the QFLS which indicates additional losses at the contacts and/or interfaces. The findings are corroborated by rigorous device simulations which outline several important considerations to maximize the Voc. This work shows that the real challenge to approach the radiative Voc limit lies in the suppression of carrier recombination at the perovskite/CTL interfaces.

CPP 2.2 Mon 10:00 H18

The interplay of interfacial charges, contact layers and hysteresis in perovskite solar cells —ILKA M HERMES¹, YI HOU², CHRISTOPH BRABEC², and •STEFAN A.L. WEBER^{1,3} — ¹MPI for Polymer Research Mainz, Germany — ²Institute of Materials for Electronics and Energy Technology, Friedrich-Alexander University Erlangen-Nürnberg, Germany — ³Institute of Physics, Johannes Gutenberg University Mainz, Germany

The charge selective contact layers in perovskite solar cells affect device properties such as current density-voltage hysteresis. Using fullerene-based electron transport layers (ETLs), hysteresis could be suppressed by a reduction of the mobile ion concentration. However, the effect the ETL on the electronic properties of other constituent device layers remains unclear. Using Kelvin probe force microscopy, we compared potential distributions of methylammonium lead iodide-based solar cells with different ETLs, such as planar titania and a C60-derivative, with vastly different hysteretic behavior. We found the most significant potential differences at the interface to the organic hole transport layer spiroMeOTAD. Our results show that the choice of ETL not only influences the extraction and recombination at the perovskite/ETL interface but also changes the transport properties of the spiroMeOTAD layer. We propose that the higher density of mobile ions in the titania cell caused a reaction between iodide anions with the p-doped spiroMeOTAD to form a neutral interface layer, which increased the interfacial resistance and capacitance at the spiroMeOTAD interface.

CPP 2.3 Mon 10:15 H18

Investigation of Charge Transport Properties of cross-linkable Hole Transport Materials in Perovskite Solar Cell Devices —•SIMON EWERTOWSKI¹, LORENA PERDIGÓN TORO², DIETER NEHER², and KLAUS MEERHOLZ¹ — ¹University of Cologne, Institute of Physical Chemistry, Cologne, Germany — ²University of Potsdam, Institute of Physics, Potsdam, Germany

Organic-inorganic hybrid perovskite materials are known to show impressive properties in charge generation and charge transport. These properties make them useful in optoelectronic thin film devices, such as light emitting diodes or solar cells. Therefore, perovskite solar cells (PSCs) attracted much attention in the last decade due to their rapid evolution in power conversion efficiencies (PCE), today exceeding 20%. [1] In the inverted p-i-n structured PSCs usually PEDOT:PSS, PTAA and poly TPD are used as efficient hole transport materials (HTMs). [2], [3]

In this study, several oxetane-functionalized cross-linkable x-HTMs such as QUPD [4] are implemented in lead based PSCs. Impedance measurements reveal information about the charge transport across

the interfaces between the x-HTMs and the perovskite. These results are taken into account, when analyzing the respective current-voltage characteristics of the built solar cells.

[1] Brenner, T. M. et al., *Nat. Rev. Mater.* 2016, 1, 15007 [2] Zhao D. et al., *Adv. Energy Mater.* 2014, 1401855 [3] Wolff C. M. et al., *Adv. Mater.* 2017, 29, 1700159 [4] Jhuo H.-J., et al., *J. Mater. Chem. A*, 2015, 3, 9291

CPP 2.4 Mon 10:30 H18

Probing Charge Carrier Dynamics in Perovskite Solar Cells —•CHRISTIAN WOLFF¹, SEAN BOURELLE², SASCHA FELDMANN², FELIX DESCHLER², and DIETER NEHER¹ — ¹Institut für Physik und Astronomie, Universität Potsdam, Karl-Liebknecht-Str. 24-25, Potsdam, Germany — ²Cavendish Laboratory, Department of Physics, University of Cambridge, JJ Thomson Ave., Cambridge, UK

Photogenerated charge carriers in solar cells have different pathways of recombining. The ideal pathway is external recombination while delivering power to a consumer. In parallel charge carriers may recombine radiatively or nonradiatively in the bulk of the absorber, at surface states, within other functional layers, or across internal interfaces. One major challenge in solar cell research is to identify the pathway, order and rate of these processes. In this contribution we probe the fate of charge carriers in fully functioning devices. We employ time-resolved and steady state absorption, luminescence and charge extraction experiments and provide a consistent picture of the dominating recombination processes at relevant carrier densities. We are able to show that the recombination under 1 sun illumination is dominated by a first order process that we attribute to interfacial recombination, while at higher intensities a radiative second order is prevailing. The results suggest that unlike other previous reports the recombination process in full devices follows the simple rate equation $dn/dt = -k_1n + k_2n^2 + k_3n^3$, with $k_1 \sim 10^7 s^{-1}$, $k_2 \sim 10^{-11} cm^{-3} s^{-1}$ and $k_3 \sim 10^{-33} cm^{-6} s^{-1}$ and that there is no need to include mixed-order processes.

CPP 2.5 Mon 10:45 H18

Universal self-assembled monolayers as hole contacts for high-performance p-i-n Perovskite solar cells —•AMRAN AL-ASHOURI¹, ARTIOM MAGOMEDOV², MARCEL ROSS¹, JOSÉ M. PRIETO¹, EIKE KÖHNEN¹, MARKO JOŠT¹, TADAS MALINAUSKAS², THOMAS UNOLD¹, VYTAUTAS GETAUTIS², and STEVE ALBRECHT¹ — ¹HZB, Berlin, Germany — ²KTU, Kaunas, Lithuania

We recently presented a novel concept for hole-selective contacts in perovskite solar cells (PSCs) by utilizing a molecule that forms a self-assembled monolayer (SAM) on the transparent conductive oxide via covalent bonds [1]. Here we present results from a new class of hole-selective SAMs that enable p-i-n PSCs with open-circuit voltages (Voc's) of up to 1.19 V, fill factors of over 80% and thus power conversion efficiencies of up to 21% for three different perovskite compositions. Hyperspectral, absolute and transient photoluminescence spectroscopy reveal that the reduction of non-radiative recombination losses at the hole contact interface, accompanied by high charge carrier lifetimes, is partially the origin of the relatively high Voc. Interestingly, implementing the SAMs also reduces non-radiative recombination at the electron-selective contact (C60). We further analyze the SAMs by IR and X-ray spectroscopy to assess the surface modification and energetic alignments. Our work demonstrates cell performances that rival best published p-i-n PSCs, without any interlayers, doping or perovskite post-treatments and might therefore serve as a suitable strategy for further performance improvements.

[1] Magomedov et al. *Adv. Energy Mater.* 2018, 8, 1801892

CPP 2.6 Mon 11:00 H18

Interfacial Characteristics and Charge Carrier Dynamics in Formamidinium Tin Iodide and Phenylethylammonium/Formamidinium Tin Iodide —•JONAS HORN¹, MIRKO SCHOLZ², KAWON OUM², THOMAS LENZER², and DERCK SCHLETTWEIN¹ — ¹Justus-Liebig-Universität Gießen, Institut für Angewandte Physik — ²Universität Siegen, Physikalische Chemie

Electrical and optical measurements were performed at formamidinium tin iodide ($FASnI_3$) and its phenylethylammonium (PEA) derivative $PEA_{0.08}FA_{0.92}SnI_3$, representing attractive alternatives to lead-

based perovskites. Charge transport across metal-perovskite interfaces and through the perovskite films were studied on microstructured gold electrode arrays. Hysteresis in the current-voltage characteristics and a corresponding current-time behavior indicated limitation by interfacial charge transfer. Smaller persistent changes following prolonged polarization was measured in $PEA_{0.08}FA_{0.92}SnI_3$ compared to $FASnI_3$ which is explained by a 2-dimensional interlayer at the contacts leading to decreased field-induced migration of ions. The rate constants for the Auger- and bimolecular recombination processes were obtained from femtosecond transient absorption experiments. A time constant of 0.5 ps was found for the scattering of charge carriers by optical phonons, even faster than measured in lead-based perovskites. Carrier recombination in the bulk material was not substantially influenced by the substitution of FA by PEA . Changes in the electrical characteristics of $PEA_{0.08}FA_{0.92}SnI_3$ compared to $FASnI_3$ are, therefore, assigned to different contact rather than bulk characteristics.

15 min. break

CPP 2.7 Mon 11:30 H18

Quantitative Analysis of the Transient Photoluminescence of Lead Halide Perovskite with Contact Layers — ●BENEDIKT KROGMEIER¹, LISA KRÜCKEMEIER¹, and THOMAS KIRCHARTZ^{1,2} — ¹IEK5-Photovoltaik, Forschungszentrum Jülich, 52425 Jülich, Germany — ²Faculty of Engineering and CENIDE, University of Duisburg-Essen, Carl-Benz-Str. 199, 47057 Duisburg, Germany

Transient photoluminescence (PL) is a powerful measurement technique to study charge carrier dynamics in perovskite layers. A single layer of lead halide perovskite measured with this technique usually shows a high charge carrier lifetime. However, adding additional contact layers may substantially accelerate the PL decay and therefore decrease the measured lifetime. The higher decay occurs due to the extraction of charge carriers into the contact layer or due to charge carrier recombination at the interface. As the measurements are done on uncontacted samples (i.e. in open circuit), charge carriers may accumulate on the contacts after extraction and thereby repel additional charges at longer times and higher laser fluences. The superposition of extraction, accumulation and recombination makes the use of numerical simulations necessary. We use these simulations to describe experimental results on various structures featuring, zero, one or two contact layers. In addition, we study the effect of low conductivity interface layers (such as PTAA or PCBM) and the effect of high conductivity layers such as indium tin oxide or silver on lateral diffusion and thereby the decay kinetics.

CPP 2.8 Mon 11:45 H18

Charge carrier recombination dynamics and carrier-phonon interactions in bismuth halide semiconductors — ●LISSA EYRE, ROBERT HOYE, TUDOR THOMAS, HANNAH JOYCE, and FELIX DESCHLER — University of Cambridge, JJ Thomson Ave, Cambridge, CB3 0HE, United Kingdom

A promising class of lead-free perovskites for photovoltaic applications include the bismuth halides, such as $MA_3Bi_2I_9$, $BiOI$, and $Cs_2AgBiBr_6$. Although these materials have been predicted to exhibit defect tolerance, as seen in lead-halide perovskites, and already display improved stabilities and long charge carrier lifetimes, the power conversion efficiencies of the corresponding devices have not reached the level of lead-based perovskites. Potential reasons for this are explored, for example, the disconnected nature of the bismuth halide octahedra in the crystal structure, which limits carrier mobility, and the lower levels of absorption due to indirect bandgaps. We probe the behaviour of excited states in many bismuth-halide compounds with various effective dimensionalities using ultrafast transient absorption, Raman, and terahertz spectroscopy. Overall, this work indicates that bismuth-based materials have the potential to be used in efficient optoelectronic devices, but there is a need to account for the effects of strong carrier-phonon coupling and localisation of electronic states on carrier scattering rates. We therefore present charge carrier-lattice interaction strength as an important design criterion for efficient next-generation solar cells.

CPP 2.9 Mon 12:00 H18

Origin of Efficient Photoluminescence in Alloyed Perovskites: Photodoping from Localized Charge Accumulation Regions — ●SASCHA FELDMANN¹, STUART MACPHERSON¹, JASMINE RIVETT¹, MOJTABA ABDI-JALEBI¹, GUANGJUN NAN², GREGORY TAINTER¹, RICHARD FRIEND¹, HENNING SIRRINGHAUS¹, MICHAEL SALIBA³, DAVID BELJONNE⁴, SAMUEL STRANKS¹, and FELIX DESCHLER¹ — ¹Cavendish Laboratory, University of Cambridge, Cambridge, UK — ²Department of Physics, Zhejiang Normal University, Jinhua 321004, China — ³Adolphe Merkle Institute, University of Fribourg, CH-1700 Fribourg, Switzerland — ⁴Chimie des Matériaux Nouveaux, Université de Mons, B-7000 Mons, Belgium

Metal-halide perovskites have emerged as exceptional semiconductors for optoelectronic applications. Here, we reveal the origin of the improved optoelectronic performance arising from compositional engineering by tracking charge carrier dynamics in alloyed perovskites with time-resolved spectroscopy. In contrast to single-halide perovskites, we find that luminescence yields show weak dependence on carrier density and remain high even for photo-excited carrier densities far below solar illumination conditions. We show that the carrier recombination regime changes from second to first order within the first tens of nanoseconds after excitation. Supported by evidence of microscale variations in optical bandgap, electrically-gated transport measurements and first-principles calculations, we propose that spatially-varying energetic disorder in the electronic states leads to local accumulation of one charge carrier type, creating photodoped p- and n-type regions.

CPP 2.10 Mon 12:15 H18

Charge Carrier Transport in Organo-Metal Halide Perovskites Probed by Transient Microwave Conductivity — ●ANDREAS BAUMANN¹, MANUEL ULLRICH², NATHANIA HENNING², PATRICK DÖRFLINGER², VLADIMIR DYAKONOV², and ANDREAS SPERLICH² — ¹Bavarian Center for Applied Energy Research (ZAE Bayern), 97074 Würzburg — ²Experimental Physics VI, Julius Maximilian University of Würzburg, 97074 Würzburg

The charge carrier mobility is one of the important intrinsic parameters for the performance of solar cell devices as it together with the charge carrier lifetime defines the diffusion length of the charge carriers and hence sets a limit to the device active layer thickness. For lead halide perovskite (HaP) solar cells, long charge carrier diffusion lengths up to micrometer scale have been reported which is primarily due to the long charge carrier lifetime. However, the charge carrier mobility is found to be smaller compared to the theoretically predicted values. In order to investigate the fundamental charge carrier transport properties in HaP materials we used the contact-less technique of transient microwave conductivity (TRMC). We analysed different HaPs including methylammonium lead iodide ($MAPbI_3$), lead bromide ($MAPbBr_3$) and mixed cation and halide perovskite by TRMC at room temperature and find the highest charge carrier mobility for $MAPbI_3$. Finally, TRMC mobilities in $MAPbI_3$ tested in a wide temperature range show a clear negative temperature coefficient in the tetragonal phase as well as a mobility drop at the phase transition.

CPP 2.11 Mon 12:30 H18

Anisotropic charge carrier diffusion in methylammonium lead iodide — ILKA M. HERMES¹, MARKUS MEZGER^{1,2}, DAVID S. GINGER³, RÜDIGER BERGER¹, KALOIAN KOYNOV¹, and ●STEFAN A.L. WEBER^{1,2} — ¹MPI for Polymer Research, Mainz, Germany — ²Institute of Physics, JGU Mainz, Germany — ³University of Washington, Seattle, USA

In this combined piezoresponse force microscopy (PFM) and local time-resolved photoluminescence (PL) study, we correlate the charge carrier dynamics on large ($>10\mu m$) isolated methylammonium lead iodide grains to the arrangement of ferroelastic twin domains[1]. Using PL-microscopy, we measured the carrier diffusion parallel and perpendicular to the twin domain stripes. The diffusion parallel to the domains was significantly faster than perpendicular to the domains, resulting in a distinct anisotropy in the carrier dynamics. Our results demonstrate that controlling the strain during crystal growth could offer a way to enhance carrier extraction in perovskite solar cells. [1] J. Phys. Chem. C, 2016, 120, 5724.