

CPP 61: Focus: Fundamental Physics of Perovskites II - organized by Lukas Schmidt-Mende and Vladimir Dyakonov

Time: Thursday 9:30–13:00

Location: C 130

Topical Talk

CPP 61.1 Thu 9:30 C 130

Approaching the Shockley-Queisser Limit with Interface Control in Halide Perovskites — ●DAVID GINGER — University of Washington, Seattle, USA

Although remarkable gains in performance have been demonstrated for halide perovskite semiconductors, most current perovskite solar cells are still limited by non-radiative recombination losses. In this talk, I will focus on uncovering and eliminating these loss processes. Experiments suggest that electrical heterogeneities in both the perovskite active layer, as well as the perovskite/electrode interface, affect carrier diffusion and non-radiative recombination processes within perovskite solar cells. I will describe both confocal and conductive atomic force microscopy (cAFM) to explore the role of heterogeneities and grain boundaries on lateral carrier transport, and will demonstrate varying degrees of grain boundary opacity to carrier transport depending on the structure. We will also discuss both ligand exchange and cation exchange experiments in the context of tailoring the surface properties of halide perovskite thin films. We show that with controlled passivation of the perovskite surfaces we are able to obtain carrier lifetimes and PL intensities in solution-processed thin films that rival those in the best single crystals, achieving over 90% PL internal quantum efficiency and quasi-Fermi level splittings that exceed 96% of the Shockley-Queisser limit under illumination. Combining these results with experiments demonstrating contact-induced losses in many common perovskite architectures, we then explore new contact materials and their potential for increased efficiency.

CPP 61.2 Thu 10:00 C 130

Active Materials and interfaces for stable perovskite solar cells — ●ANTONIO ABATE — Helmholtz-Zentrum Berlin für Materialien und Energie

Organic-inorganic perovskites are quickly overrunning research activities in new materials for cost-effective and high-efficiency photovoltaic technologies. Since the first demonstration from Kojima and co-workers in 2009, several perovskite-based solar cells have been reported and certified with rapidly improving power conversion efficiency. Recent reports demonstrate that perovskites can compete with the most efficient inorganic materials, while they still allow processing from solution as a potential advantage to deliver a cost-effective solar technology. Compare to the impressive progress in power conversion efficiency, stability studies are rather weak and often controversial. An intrinsic complication comes from the fact that the stability of perovskite solar cells is strongly affected by any small difference in the device architecture, preparation procedure, materials composition and testing procedure. In the present talk, we will focus on the stability of perovskite solar cells in working condition. We will discuss a measuring protocol to extract reliable and reproducible ageing data. We will present new materials and preparation procedures, which improve the device lifetime without giving up on high power conversion efficiency.

CPP 61.3 Thu 10:15 C 130

Visualizing and suppressing interfacial recombination in high efficiency large area pin-perovskite solar cells — ●MARTIN STOLTERFOHT¹, CHRISTIAN M. WOLFF¹, SHANSHAN ZHANG^{1,2}, JOSÉ A. MÁRQUEZ PRIETO³, CHARLES J. HAGES³, DANIEL ROTHARDT¹, THOMAS UNOLD³, STEVE ALBRECHT⁴, PAUL L. BURN², PAUL MEREDITH⁵, and DIETER NEHER¹ — ¹University of Potsdam, Potsdam-Golm, Germany — ²The University of Queensland, Brisbane, Australia — ³Helmholtz-Zentrum-Berlin, Berlin, Germany — ⁴Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Berlin, Germany — ⁵Swansea University, Swansea, United Kingdom

The current generation of perovskite solar cells (PSCs) is predominantly limited by non-radiative recombination, either through trap-assisted recombination in the absorber layer, or via minority carrier recombination at the interface to the transport layers. Using transient and absolute photoluminescence imaging we are able to visualize all relevant non-radiative recombination pathways in planar pin-type PSCs. We find significant quasi-Fermi level splitting losses (135 meV) in the perovskite bulk, while interface recombination results in an additional energy loss of 80 meV at each individual interface. These losses define the open-circuit voltage of the complete cell. Inserting ultrathin inter-

layers between the perovskite and transport layers allows substantial reduction of these interfacial losses at both the p-and-n-contacts. Using this knowledge and approach, we demonstrate stable 1 cm² PSCs surpassing 20% efficiency (19.83% certified) with record fill factor (> 81%), high V_{OC} (1.17V) and near unity manufacturing yield.

CPP 61.4 Thu 10:30 C 130

Passivating perovskite surfaces by mixtures of PEDOT and dopant-free Spiro-OMeTAD — ●L. KEGELMANN¹, C. M. WOLFF², J. A. MÁRQUEZ PRIETO¹, P. TOCKHORN¹, T. UNOLD¹, D. NEHER², B. RECH¹, and S. ALBRECHT¹ — ¹Helmholtz-Zentrum Berlin, Berlin, Germany — ²University of Potsdam, Institute of Physics and Astronomy, Potsdam, Germany

Perovskite solar cells have achieved efficiencies above 22% within few years. By now, the device parameter with the largest potential for further improvements is the open circuit voltage (V_{oc}), which is mainly limited by recombination losses at the interfaces. Here, a poly(3,4-ethylenedioxythiophene) (PEDOT) layer doped with sulfonated copolymers is utilized as a hole selective contact (HSC) on top of perovskite absorbers. Transient photoluminescence (PL) and steady-state PL quantum yield measurements reveal longer charge carrier lifetimes and larger quasi-fermi level splitting for perovskite films after the PEDOT is deposited. PEDOT thin films therefore seem to suppress charge carrier recombination at the perovskite surface. By blending undoped Spiro-OMeTAD into the PEDOT dispersion, the surface energetics of the resulting films are shifted to larger ionisation energies as measured by photoelectron spectroscopy and the quasi-fermi level splitting is further enhanced. As a result, perovskite solar cells with mixtures of PEDOT and Spiro-OMeTAD as HSC achieve high V_{oc} values above 1.15 V and stabilized efficiencies over 16%. This exceeds the V_{oc} of reference devices with either doped Spiro-OMeTAD or pure PEDOT as HSC.

CPP 61.5 Thu 10:45 C 130

Revealing the impact of Rubidium and Cesium on the electronic trap landscape of mixed cation perovskite solar cells via thermally stimulated current — ●PHILIPP RIEDER¹, YINGHONG HU³, ALEXANDER HUFNAGEL³, MELTEM AYGÜLER³, MICHIEL PETRUS³, PABLO DOCAMPO⁴, KRISTOFER TVINGSTEDT¹, ANDREAS BAUMANN², THOMAS BEIN³, and VLADIMIR DYAKONOV^{1,2} — ¹Experimental Physics VI, University of Würzburg, 97074 Würzburg — ²ZAE Bayern, 97074 Würzburg — ³Department of Chemistry and Center for NanoScience (CeNS), LMU Munich, 81377 Munich — ⁴Physics Department, Newcastle University, Newcastle upon Tyne, NE1 7RU, United Kingdom

In the last years, mixed cation and halide organo-lead perovskite has established itself as the working horse in the perovskite community, due its improved crystallization behavior and therefore reproducible high power conversion efficiencies (PCE) achieved in various solar cell configurations. Recently, this so-called "FAMA" perovskite, whereas methylammonium (MA) and bromide is used to stabilize the crystal structure of formamidinium (FA) lead iodide perovskite, could be further improved by the introduction of small amounts of Rb as well as Cs, boosting the device PCE up to 21.6%, while additionally enhancing the long-term stability. Here, we use thermally stimulated current (TSC) spectroscopy to study the beneficial impact of Cs and/or Rb on the FAMA crystal lattice by directly probing the electronic trap landscape in what has been come to be known as triple and quadruple cation perovskite solar cells.

15 min. break

Topical Talk

CPP 61.6 Thu 11:15 C 130

Understanding Hysteresis in Perovskite Solar Cells — ●STEFAN A.L. WEBER^{1,2}, ILKA M. HERMES¹, NIKLAS M. BUDINGER^{1,2}, WOLFGANG TRESS³, ANDERS HAGFELDT³, MICHAEL GRAETZEL³, and RÜDIGER BERGER¹ — ¹MPI for Polymer Research, Ackermannweg 10, 55128 Mainz, Germany — ²Johannes Gutenberg University, Department of Physics, Staudingerweg 10, 55128 Mainz, Germany. — ³Laboratory of Photomolecular Science, Institute of Chemical Sciences and Engineering, École Polytechnique Fédérale de Lausanne,

1015 Lausanne, Switzerland.

We explain the underlying mechanism of hysteresis in a hybrid lead-halide perovskite solar cell by measuring and numerically calculating the dynamics of the charge distribution in an operating device. The origin, magnitude and the dynamics of the charge re-distribution processes that are responsible for hysteresis in the device are still not understood. We developed a method based on Kelvin probe force microscopy that enables mapping charge re-distribution in an operating device upon a voltage- or light pulse with sub-millisecond resolution. A combination of an asymmetric ion migration and the formation of a surface dipole at the anode shielded the electric field in the perovskite layer within 10 ms after applying a forward voltage to the device. After switching off the voltage, the anode surface dipole created a reverse electric field in the cell that lasted for up to 500 ms. This reverse electric field increases charge carrier extraction and thereby directly explains higher photocurrents during reverse bias scans.

CPP 61.7 Thu 11:45 C 130

Recombination in Perovskite Solar Cells — ●WOLFGANG TRESS — Laboratory for Photonics and Interfaces, Institute of Chemical Sciences and Engineering, École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

Solar cells based on metal halide perovskite absorber materials are about to emerge as a high-efficiency photovoltaic technology. At the same time, they are suitable for high-throughput manufacturing characterized by a low energy input and abundant low-cost materials. However, a further optimization of their efficiency, stability and reliability demands for a more detailed optoelectronic characterization and understanding of losses including their evolution with time.

In this work, we analyze perovskite solar cells with different architectures (planar, mesoporous, HTL-free), employing temperature dependent measurements (current-voltage, light intensity, electroluminescence) of the ideality factor to identify dominating recombination processes that limit the open-circuit voltage (V_{oc}). We find that in thoroughly-optimized, high- V_{oc} (> 1.2 V) devices recombination prevails through defects in the perovskite. On the other hand, irreversible degradation at elevated temperature is caused by the introduction of broad tail states originating from an external source (e.g. metal electrode). Light-soaking is another effect decreasing performance, though reversibly. Based on FTPS measurements, this degradation is attributed to the generation of surface defects becoming a new source of non-radiative recombination.

CPP 61.8 Thu 12:00 C 130

Long-range charge extraction in quasi-interdigitated back-contact hybrid perovskite solar cells — ●GREGORY TAINTER^{1,2}, MAXIMILIAN HOERANTNER³, LUIS MIGUEL PAZOS-OUTÓN¹, SUHAS MAHESH³, RICHARD FRIEND¹, HENRY SNAITH³, HANNAH JOYCE², and FELIX DESCHLER¹ — ¹Cavendish Laboratory, Department of Physics, University of Cambridge, JJ Thomson Avenue, Cambridge CB3 0HE, UK — ²Department of Engineering, University of Cambridge, Cambridge CB3 0FA, UK — ³Clarendon Laboratory, University of Oxford, Parks Road, Oxford OX1 3PU, UK.

Understanding charge extraction in photovoltaic devices is critical to engineering practical applications. Here we study individual charge extraction and charge recombination via scanning photocurrent measurements on quasi-interdigitated back-contact (QIBC) hybrid perovskite devices. Lateral separation of carriers is achieved via excitation over charge-selective electrodes and non-ambipolar charge transport is characterised. We model charge carrier diffusion and report electron and hole diffusion constants in operating QIBC devices. A large fraction of holes and electrons are found to propagate through multiple grain boundaries, and carrier recombination is reduced due to charge separation as indicated by our diffusion model.

CPP 61.9 Thu 12:15 C 130

Surface State Density Determines the Energy Level Alignment at Hybrid Perovskite/Electron Acceptor Interfaces — ●FENGSHUO ZU^{1,2}, PATRICK AMSALEM¹, MARYLINE RALAIARISOA^{1,2}, THORSTEN SCHULTZ¹, RAPHAEL SCHLESINGER^{1,2}, and NORBERT KOCH^{1,2} — ¹Institut für Physik, HU Berlin — ²HZB, 12489 Berlin

Substantial variations of the surface electronic structure and thus con-

flicting energetics at interfaces between hybrid perovskites and charge transport layers can be found in literature. In an attempt to unravel the origin of these variations, we demonstrate that donor-like surface states stemming from reduced lead (Pb0) directly impact the energy level alignment at perovskite and molecular electron acceptor interfaces, using photoelectron spectroscopy. When forming the interfaces, it is found that electron transfer from surface states to acceptor molecules occurs, leading to a strong decrease in the density of ionized surface states. As a consequence, for perovskites with low surface state density, the initial band bending at the pristine perovskite surface can be flattened upon interface formation. In contrast, for perovskites with a high surface state density, the Fermi-level is strongly pinned at the conduction band edge and only minor changes in surface band bending are observed upon acceptor deposition. Consequently, depending on the initial surface state density, very different interface energy level alignment situations (variations over 0.5 eV) are demonstrated and rationalized. Our findings help explaining the rather dissimilar reported energy levels at interfaces and refining our understanding of the operating principles in devices comprising this material.

CPP 61.10 Thu 12:30 C 130

Self-assembled Monolayers Enhance the Stability and Efficiency of p-i-n Perovskite Solar Cells — ●CHRISTIAN WOLFF¹, MARTIN STOLTERFOHT¹, YOHAI AMIR¹, ANTONIO ABATE², and DIETER NEHER¹ — ¹Institut für Physik und Astronomie, Universität Potsdam, Potsdam, 14476, DE — ²Institut für Silizium-Photovoltaik, Helmholtz-Zentrum Berlin für Materialien und Energie, 12489 Berlin, DE

Perovskite solar cells with all-organic transport layers have shown efficiencies rivalling their counterparts that employ high temperature treated inorganic transport layers [1,2,3]. One of the main issues associated with perovskite solar cells is their instability, in particular towards long periods of illumination or moisture and closely related the lack of control of the nature and electronic structure of the interface between the active perovskite and the adjacent transport layers. In this contribution we show that in p-i-n cells interjection of a self-assembling monolayer between the perovskite and the adjacent electron transport layer allows us to produce solar cells with efficiencies above 20% on small areas (6 mm²) as well as 1 cm² with almost no loss by scaling. Importantly, these cells exhibit enhanced resilience towards heat and light illumination as exemplary shown by keeping the solar cells at 85°C under full load (MPP, simulated 1 sun illumination) for 250h without appreciable efficiency loss.

[1] Wolff et al., Adv.Mater., 2017, 29, 1700159 [2] Stolterfoht et al., Energy and Environ. Sci., 2017, 10, 1530-1539 [3] Zheng et al., Nat. Energy, 2017, 2, 17102

CPP 61.11 Thu 12:45 C 130

Structure and electronic properties characterization of CH₃NH₃PbI_{3-x}Cl_x mixed halide perovskite films upon massive solvents exposure — ●MARYLINE RALAIARISOA¹, YERILA RODRIGUEZ², INGO SALZMANN¹, LIDICE VAILLANT², and NORBERT KOCH¹ — ¹Humboldt-Universität zu Berlin, Institut für Physik & IRIS Adlershof — ²Physics Faculty, University of Havana

We combined photoelectron spectroscopy (PES), grazing incidence X-ray diffraction, UV-Vis absorption spectroscopy, and atomic force microscopy to characterize the effect of massive solvent exposure on CH₃NH₃PbI_{3-x}Cl_x mixed halide perovskite films. We selected solvents that can potentially be used to solution-deposit organic charge transport layers on top of the perovskite: water, dimethylformamide (DMF), chloroform (CF), and chlorobenzene (CB). Water and DMF instantly dissolved the perovskite films. Particularly, water degraded the perovskite film into a yellow PbI₂ film, along with the generation of reduced Pb⁰ precipitates. Furthermore, PES results indicate the presence of methyl ammonium or related compounds even after water dissolution. Although the crystal structure and morphology of perovskite films were preserved upon CF and CB exposure, the valence band and therefore the electronic properties of the films were strongly altered, as a result of surface state modifications. In fact, the film surfaces become less n-type upon exposure, possibly due to a reduction of surface Pb⁰-species. Thus, the use of CB and CF for solution deposition of organic charge transport layers is possible, but can already substantially modify the perovskite properties at the interface.