

CPP 45: Perovskite and photovoltaics I (joint session HL/CPP)

Time: Tuesday 9:30–12:30

Location: POT 251

CPP 45.1 Tue 9:30 POT 251

Properties of $\text{In}_2\text{S}_3\text{:V}$ epitaxially grown on p -Si for Intermediate Band Solar Cell Applications — ●TANJA JAWINSKI¹, MICHAEL LORENZ¹, ROLAND SCHEER², MARIUS GRUNDMANN¹, and HOLGER VON WENCKSTERN¹ — ¹Universität Leipzig, Felix-Bloch-Institut für Festkörperphysik, Germany — ²Martin-Luther-Universität Halle-Wittenberg, Institut für Physik, Germany

The Shockley Queisser limit of single junction solar cells can be overcome by introducing an intermediate band (IB) in wide band gap materials. Thus thermalization losses can be reduced [1]. Furthermore sub-bandgap photons can be absorbed by valence band to IB and IB to conduction band transitions. According to theoretical calculations In_2S_3 hyper-doped with vanadium is a suitable candidate to realize such an IB solar cell [2].

Undoped and V-doped In_2S_3 layers are epitaxially grown by physical co-evaporation of the elements on p -type Si wafers. Using a combinatorial approach, we can cover a wide range of doping concentrations of up to 14 at-% V. Heterostructure pin solar cells are completed using n -ZnO:Al grown by pulsed laser deposition as transparent top electrode. Improved structural properties can be attributed to the epitaxial growth mechanism even for V-doped samples. We compare diode and solar cell parameters of undoped and V-doped sample by analyzing dark and illuminated current-voltage characteristics. Furthermore we use thermal admittance spectroscopy to investigate defect states, that are induced by V-doping. [1] Luque and Martí, *Phys. Rev. Lett.*, 1997, **78**, 5014. [2] Palacios *et al.*, *Phys. Rev. Lett.*, 2008, **101**, 046403.

CPP 45.2 Tue 9:45 POT 251

Time resolved spin dynamics in lead halide hybrid organic perovskite $\text{Fa}_{0.9}\text{Cs}_{0.1}\text{PbI}_{2.8}\text{Br}_{0.2}$ — ●ERIK KIRSTEIN¹, EIKO EVERS¹, VASILLI V. BELYKH^{1,2}, EVGENY A. ZHUKOV¹, DENNIS KUDLACIK¹, INA V. KALITUKHA³, OLGA NAZARENKO⁴, MAXIM V. KOVALENKO^{4,5}, DMTRI R. YAKOVLEV^{1,3}, and MANFRED BAYER^{1,3} — ¹Experimentelle Physik 2, Technische Universität Dortmund, D-44227 Dortmund, Germany — ²P.N. Lebedev Physical Institute, Russian Academy of Sciences, 119991 Moscow, Russia — ³Ioffe Institute, Russian Academy of Sciences, 194021 St. Petersburg, Russia — ⁴Laboratory of Inorganic Chemistry, ETH Zürich, CH-8093 Zürich, Switzerland — ⁵Laboratory for Thin Films and Photovoltaics, Empa-Swiss Federal Laboratories for Materials Science and Technology, CH-8600 Dübendorf, Switzerland

Lead halide hybrid organic perovskites attract increased attention due their promising applications, related to high quantum efficiency and easy synthesis. Spin dynamics in perovskite materials is not studied in detail so far, but shows promising results. The studied $\text{Fa}_{0.9}\text{Cs}_{0.1}\text{PbI}_{2.8}\text{Br}_{0.2}$ bulk sample was grown out of solution of respective ions in polar solvents. Its bandgap of 1.51 eV makes this material well-suited for the resonant excitation with Ti:Sapphire laser. We study the coherent spin dynamics of electrons and holes by means of time-resolved pump-probe Kerr rotation technique at cryogenic temperatures and magnetic fields up to 6 T. We measure longitudinal spin relaxation times T_1 , transverse dephasing times T_2^* , g -factor values and their spread Δg .

CPP 45.3 Tue 10:00 POT 251

Ultrafast Charge Carrier Relaxation in Inorganic Halide Perovskite Single Crystals Probed by Two-Dimensional Electronic Spectroscopy — XUAN TRUNG NGUYEN¹, ●DANIEL TIMMER¹, YEYGENY RAKITA², DAVID CAHEN², ALEXANDER STEINHOFF³, FRANK JAHNKE³, CHRISTOPH LIENAU¹, and ANTONIETTA DE SIO¹ — ¹Institut für Physik, Carl von Ossietzky Universität, Germany — ²Weizmann Institute of Science, Israel — ³Institut für Theoretische Physik, Universität Bremen, Germany

In recent years, halide perovskites have become one of the most intensely studied semiconductors for the development of optoelectronic devices. Regardless, the fundamental understanding of their optical and electronic properties still remains limited. To investigate the ultrafast dynamics of optical excitations we performed pump-probe and two-dimensional electronic spectroscopy (2DES) on CsPbBr_3 single crystals with 10 fs time resolution [1]. While pump-probe spectra show a single bleaching feature near the band edge at 2.4 eV, 2DES maps reveal a more rich structure. In addition to a diagonal exciton bleaching

peak, free carrier signatures in form of an elongated cross peak along the excitation axis appear. The separation of exciton and free carrier contributions allows to derive a rapid cooling rate of ~ 3 meV/fs, indicating a strong electron-phonon coupling. The experimental findings are supported by theoretical simulations based on semiconductor Bloch equations. Our results suggest that strong electron-phonon couplings may substantially contribute to the unusual optoelectronic properties of perovskites. [1] Nguyen, X.T., et al., *JPCL*, 10, p. 5414-5421, 2019.

CPP 45.4 Tue 10:15 POT 251

Two-Dimensional Perovskite Solar Cells with 14.1% Power Conversion Efficiency and 0.68% External Radiative Efficiency — ●WEIFEI FU^{1,2}, ALEX K.-Y JEN¹, and MICHAEL SALIBA² — ¹University of Washington, Seattle, Washington 98195, United States — ²Technical University of Darmstadt, Darmstadt, Germany

Quasi-2D perovskites are attractive due to their improved stability compared to 3D counterparts but they suffer from reduced performance. Here we report an efficient quasi-2D perovskite (PEA)₂(MA)₄Pb₅I₁₆ based optoelectronic device processed with NH₄SCN and NH₄Cl additives, showing a stabilized photovoltaic power conversion efficiency as high as 14.1 % (average value 12.9+_{-0.8} %), among the highest performing quasi-2D perovskite solar cells. These additives increase the perovskite crystallinity and induce a preferred orientation with the (0k0) planes perpendicular to the substrate, resulting in improved transport properties hence increased short-circuit current density. Furthermore, the NH₄Cl treatment enriches the Cl-concentration near the PEDOT:PSS/perovskite interface, which passivates the electron traps leading to an enhanced electroluminescence external quantum efficiency (0.68% at +2.5 V bias). As a result, high open-circuit voltages of 1.21+_{-0.01} V with a record-low non-radiative VOC loss of only ~ 0.16 V could be achieved for the quasi-2D perovskite system.

CPP 45.5 Tue 10:30 POT 251

Structural Influence on the Exciton Fine Structure of Cesium Lead Halide Quantum Dots — ●TORBEN STEENBOCK and GABRIEL BESTER — Institute of Physical Chemistry, University of Hamburg, Grindelallee 117, 20146 Hamburg, Germany.

Cesium lead halide quantum dots (QDs) show a bright emission, which makes them interesting for applications in optoelectronics. The origin of the bright emission is still under debate. One suggestion [1] is based on a theoretical effective mass model assuming a very strong Rashba effect, which would leads to an unusual bright exciton ground state. Another explanation is based on the idea of a phonon bottleneck, which leads to an efficient emission from the higher-energy bright state [2] without dark ground state.

We calculate the exciton fine structure splitting (FSS) based on ab-initio density functional theory combined with a screened configuration interaction approach for small CsPbX_3 (X=Cl,Br,I) QDs. We find that the bright-dark splitting for Cl, Br, and I CsPbX_3 perovskites amount to 17, 12 and 5 meV, respectively, with only small changes due to the symmetry (cubic, tetragonal, orthorhombic) so that we always obtain a dark ground state. The FSS (splitting between the bright states) is sensitive to the symmetry with splittings of up to 5 meV for the lowest symmetry orthorhombic structures.

[1] P.C. Sercel *et al.*, *Nano Lett.* **2019**, *19*, 4068–4077.[2] P. Tamarat *et al.*, *Nat. Mater.* **2019**, *18*, 717–724.

30 min. break

CPP 45.6 Tue 11:15 POT 251

Understanding the Role of Antisolvent Quenching in Film Formation, Device Performance, and Reproducibility of Triple Cation Perovskite Solar Cells — ●ALEXANDER TAYLOR^{1,2}, QING SUN^{1,2}, KATELYN GOETZ^{1,2}, MAXIMILIAN LITTERST^{1,2}, FABIAN PAULUS^{1,2}, and YANA VAYNZOF^{1,2} — ¹Integrated Center for Applied Physics and Photonics, TU Dresden — ²cfaed, TU Dresden

Organic-inorganic perovskite materials are promising candidates for high-efficiency solar cells, quickly approaching the performance of current state-of-the-art materials. However, irreproducibility between devices made by different research labs, even world leading labs, continue to plague the field. The causes of this issue seem to be related

to small, hard to control details, such as stoichiometry variations of fractions of a percent. Herein, we seek to further the understanding of the irreproducibility in cutting edge "triple cation" perovskite solar cells by examining the minute technical differences exhibited by different researchers. We reveal that subtle differences during the crucial antisolvent step dramatically affect the resulting film microstructure, and therefore the final PV performance. By simulating this device-to-device variation, we can reliably produce devices with widely disparate power conversion efficiencies * as low as 15% and as high as 21%. Crucially, these devices would currently be reported in the literature with the exact same recipe. These results challenge some of the prevailing beliefs currently held by the research community, regarding not only what the highest performing antisolvents are, but also the general role of the antisolvent in fabricating high performance perovskite solar cells.

CPP 45.7 Tue 11:30 POT 251

Efficient and Stable Hybrid Triple-Cation Perovskite/PbS Quantum Dot Solar Cells — •MIGUEL ALBALADEJO-SIGUAN¹, DAVID BECKER-KOCH¹, ALEX TAYLOR¹, QING SUN², VINCENT LAMI², POLA GOLDBERG-OPPENHEIMER³, FABIAN PAULUS¹, and YANA VAYNZOF¹ — ¹Integrated Centre for Applied Physics and Photonic Materials and cfaed, Technical University Dresden — ²Kirchhoff Institute for Physics, Heidelberg University — ³School of Biochemical Engineering, University of Birmingham, United Kingdom

Solution-processed quantum dots (QDs) are promising for fabricating flexible, low cost and large-scale solar cells. Researchers have shown that QD devices employing a single monovalent cation perovskite shell exhibit an increased PCE when compared to standard ligand passivation. Herein we demonstrate that the use of a triple cation Cs_{0.05}(MA_{0.17}FA_{0.83})_{0.95}Pb(I_{0.9}Br_{0.1})₃ perovskite composition for surface passivation of the QDs results in highly efficient solar cells, which maintain 96 % of their initial performance after 1200h shelf storage. We trace the perovskite shell formation around the core by a range of spectroscopic techniques as well as high-resolution TEM. We find that the triple cation shell results in a favorable energetic alignment to the core of the dot, resulting in reduced recombination due to charge confinement without limiting transport in the active layer. Consequently, photovoltaic devices reached a maximum AM1.5G power conversion efficiency of 11.3 % surpassing previous reports of PbS solar cells employing perovskite passivation.

CPP 45.8 Tue 11:45 POT 251

Microscopic Insights into the Ionic Defect Migration in Metal Halide Perovskites — •NGA PHUNG¹, AMRAN AL-ASHOURI¹, SIMONE MELONI², ALESSANDRO MATTONI³, STEVE ALBRECHT¹, EVA L. UNGER¹, ABOMA MERDASA¹, and NGA PHUNG¹ — ¹Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, Kekuléstr. 5, Berlin D-12489, Germany — ²Dipartimento di Scienze Chimiche e Farmaceutiche (DipSCF), Università degli Studi di Ferrara (Unife), Via Luigi Borsari 46, I-44121, Ferrara, Italy — ³Consiglio Nazionale delle Ricerche, Istituto Officina dei Materiali, CNR-IOM, Cagliari, Cittadella Universitaria, Monserrato 09042-I (CA), Italy

Halide perovskites are emerging as revolutionary materials for optoelectronics. Their ionic nature and the presence of mobile ionic defects have a dramatic influence on the operation of thin-film optoelectronics. This study combines photoluminescence (PL) experiments and molecular dynamics simulations to demonstrate that the grain boundaries within a perovskite film inhibit the movement of ions. Based on experimental observation and theoretical calculation, we find that the

diffusion of ions is inhibited by grain boundaries, at the same time, the simultaneous removal of methylammonium and iodine causes a blue-shift of the PL spectrum. This blue-shift is reproduced by simulations, revealing that it is caused by a structural distortion of the perovskite's Pb-I octahedral network induced by an increased concentration of ionic defects. These findings provide critical information for modeling and explaining the dynamic behavior of perovskite-based optoelectronics.

CPP 45.9 Tue 12:00 POT 251

Highly stable perovskite nanocrystals in polymer micelles — •JURI G. CRIMMANN, CAROLA LAMPE, and ALEXANDER S. URBAN — Nanospectroscopy Group, Nano-Institute Munich, Department of Physics, Ludwig-Maximilians-Universität München, Königinstraße 10, 80539 Munich, Germany

Halide perovskite nanocrystals (NCs) have been synthesized through many approaches. Comprising a variety of geometrical shapes, these NCs exhibit excellent optoelectronic qualities, such as near-unity quantum yields and tunable photoluminescence emission. However, perovskite nanocrystals suffer from stability problems as well as extremely mobile halide ions, both resulting in shifts of the photoluminescence emission and instant degradation in water. By using a polymer-templated synthesis we massively improve the stability and suppress halide ion migration by encapsulating NCs individually instead of entire devices. During our synthesis perovskite NCs are formed inside diblock copolymer micelles. The micelles act not only as nanoreactors, but also encapsulate and passivate the NCs, protecting them from the environment. The stability against water degradation increases significantly. Accordingly, even after 75 days of complete submersion in water, characteristic photoluminescence is observable. Furthermore, we have shown that halide ion migration through the polymer shell is nonexistent. We explore the extension of this synthesis to other perovskite compositions, comparing efficiencies and stability. And finally, we explore strategies for incorporating these highly stable NCs into working optoelectronic devices, such as light-emitting diodes (LEDs).

CPP 45.10 Tue 12:15 POT 251

Optical Printing of Single Perovskite Nanoparticles — •ANDREAS NIKLAS RANK, CAROLA LAMPE, MORITZ GRAMLICH, and ALEXANDER URBAN — Nanospectroscopy Group, Nano-Institute Munich, Department of Physics, Ludwig-Maximilians-Universität München, Königinstraße 10, 80539 Munich, Germany

The investigation of single perovskite nanocrystals in different temperature regimes can reveal the fundamental optical properties of the individual nanocrystals and enable further improvement for optical applications. Spin-coating diluted dispersions is the typical approach to obtain films with single, separated nanocrystals. Another versatile method is optical printing, an already established approach for gold, silver and silicon nanoparticles. In this method, the optical forces of a focused laser beam are utilized to localize nanoparticles at specific positions on a substrate. A specific surface coating prevents unwanted binding of other nanoparticles. The remaining solution can be washed away, leaving only the printed nanoparticles bound to the substrate via Van der Waals forces. Here, we investigate the applicability of this method to perovskite nanocrystals. We look into different nanocrystal morphologies and compositions as well as important laser parameters and substrate materials. Adapting this method to perovskite nanocrystals would not only unlock facilitate the investigation of single perovskite nanocrystals but could also lead to complex, nanostructured substrates.