

## HL 35: Perovskite and photovoltaics II (joint session HL/ CPP)

Time: Tuesday 14:00–16:00

Location: POT 251

HL 35.1 Tue 14:00 POT 251

**Temperature-dependent exciton-phonon coupling in CsPbBr<sub>3</sub> crystal using ultrafast two-dimensional electronic spectroscopy** — ●XUAN TRUNG NGUYEN<sup>1</sup>, DANIEL TIMMER<sup>1</sup>, YEVGENY RAKITA<sup>2</sup>, DAVID CAHEN<sup>2</sup>, ALEXANDER STEINHOFF<sup>3</sup>, FRANK JAHNKE<sup>3</sup>, CHRISTOPH LIENAU<sup>1</sup>, and ANTONIETTA DE SIO<sup>1</sup> — <sup>1</sup>Institut für Physik, Carl von Ossietzky Universität, Germany — <sup>2</sup>Weizmann Institute of Science, Israel — <sup>3</sup>Institut für Theoretische Physik, Universität Bremen, Germany

Halide perovskites (HaPs) are promising semiconductor materials for developing efficient solar cells. It is, however, still debated how the unusual electron-lattice couplings in these materials affect their optical and electronic properties. Here, we investigate the temperature dependence of near band gap optical transitions in CsPbBr<sub>3</sub> single crystals by 10fs-time-resolution two-dimensional electronic spectroscopy (2DES). At room temperature, the 2DES maps show rapid charge carrier relaxation within the continuum and the concurrent build-up of exciton screening by free carriers on a <30fs time scale [1]. At 70K, we detect large, persistent oscillations of both amplitude and resonance energy of the exciton peak with periods corresponding to the phonon modes of the lead-halide sublattice. Our results point to strong exciton-phonon couplings and to the importance of many-body interactions for the optical and transport properties in HaPs.

[1] Nguyen, X.T., et al., JPCL, 10, p. 5414-5421, 2019.

HL 35.2 Tue 14:15 POT 251

**Solution-Processed Two-Dimensional Materials for Perovskite Optoelectronics** — ●ANTONIO GAETANO RICCIARDULLI<sup>1,2</sup>, PAUL BLOM<sup>2</sup>, and MICHAEL SALIBA<sup>1</sup> — <sup>1</sup>Technische Universität Darmstadt, Darmstadt, Germany — <sup>2</sup>Max-Planck-Institut für Polymerforschung, Mainz, Germany

The efficiency of perovskite based perovskite solar cells and light-emitting diodes (PSCs and PLEDs, respectively) is limited by hole injection/transport and high leakage current, generated by a high hole injection barrier and poor perovskite morphology, respectively. Black phosphorus (BP), a newly emerged 2D layered semiconductor, is endowed with thickness-dependent bandgap, which spans from 0.3 eV (bulk) to 2.0 eV (monolayer). Hence, by carefully selecting the appropriate thickness, BP can be used as interlayer to enhance charge transport and injection in either PLEDs or PSCs. As a proof of concept, we reported a feasible strategy to reduce both hole injection barrier and defects at the perovskite interface by introducing 2D BP as hole injection layer in a PeLED stack. A continuous film composed of high-quality, ultrathin, and large BP sheets on top of PEDOT:PSS simultaneously improves the hole injection and morphology of the green-emitting CsPbBr<sub>3</sub>. Inclusion of BP enhances over 4 times efficiency of CsPbBr<sub>3</sub> based PeLEDs. Incorporating BP to enhance the hole injection from PEDOT:PSS might also be of interest to increase the open-circuit voltage of PSCs of which the perovskite absorber has deeper valence band. Currently, we are developing novel PSCs based on 2D materials with specific functionalities.

HL 35.3 Tue 14:30 POT 251

**Ultrafast vibrational dynamics in lead halide perovskites** — ●AJAY JHA<sup>1</sup>, HONG-GUANG DUAN<sup>1</sup>, VANDANA TIWARI<sup>1</sup>, GOLIBJON BERDIYOROV<sup>2</sup>, ALEXEY AKIMOV<sup>3</sup>, PABITRA NAYAK<sup>4</sup>, ZHENG LI<sup>1</sup>, HENRY SNAITH<sup>4</sup>, MICHAEL THORWART<sup>5</sup>, MOHAMED MADJET<sup>2</sup>, and R. J. DWAYNE MILLER<sup>1,6</sup> — <sup>1</sup>MPSD, Hamburg — <sup>2</sup>Qatar Environment & Energy Research Institute, Doha — <sup>3</sup>Department of Chemistry, State University of New York — <sup>4</sup>Department of Physics, University of Oxford — <sup>5</sup>I. Institut für Theoretische Physik, University of Hamburg — <sup>6</sup>Departments of Chemistry and Physics, University of Tormoto

Hybrid organic-inorganic perovskites have gathered much attention owing to their unprecedented success in photovoltaics. To unravel the secrets to this success, we have studied the ultrafast dynamics of lead halide perovskites using heterodyne-detected transient grating and 2D spectroscopy in thin films for tetragonal (room temperature) and orthorhombic phases (T = 20 K). Our measurements capture the different ultrafast exciton dynamics in two phases of perovskite. In addition, we distinctly observe the ground and excited state vibrational modes corresponding to organic and inorganic sub-lattices in

two phases. The interplay of strongly coupled dominant vibrational modes to ultrafast carrier generation process will be discussed. To rationalize our observations, we have employed density functional theory (DFT) predicting changes in the vibrational observations highlighting the importance of interaction between organic cation and inorganic sub-lattice in stabilization of charges upon photoexcitation.

HL 35.4 Tue 14:45 POT 251

**Lead-free co-evaporated perovskites for photovoltaic applications.** — ●TIM SCHRAMM, MARTIN KROLL, FREDERIK NEHM, ZONGBAO ZHANG, KARL LEO, and YANA VAYNZOF — Dresden Integrated Center for Applied Physics and Photonic Materials (IAPP), Technische Universität Dresden

The performance of lead-based perovskites has dramatically increased over the past decade reaching power conversion efficiencies of over 25%. Despite these remarkable advances, lead-based perovskite suffer from serious challenges, most notably lead related toxicity and poor stability, which remain significant obstacles for their integration into industrial applications.

One promising route to circumvent these issues is by turning to novel absorber material systems such as the lead-free double perovskites. Herein, we present our recent results on the formation of Cs<sub>2</sub>AgBiBr<sub>6</sub> perovskites by thermal evaporation. In particular, we employ a triple-source co-evaporation of the precursor materials, followed by thermal annealing and characterize their microstructure, crystallinity and optical properties. We identify processing conditions that lead to high quality crystalline films with large domains and investigate their performance in photovoltaic devices. Our results pave the route for further development of double perovskites for optoelectronic applications.

HL 35.5 Tue 15:00 POT 251

**Ion Migration Induced Modification of the Interfacial Energetics in Perovskite Solar Cells and its Effect on Device Performance** — ●JOSHUA KRESS<sup>1,3,4</sup>, QINGZHI AN<sup>1,3,4</sup>, NIR TESSLER<sup>2</sup>, and YANA VAYNZOF<sup>1,3,4</sup> — <sup>1</sup>TU Dresden — <sup>2</sup>Technion, Haifa — <sup>3</sup>Integrated Center for Applied Physics and Photonics — <sup>4</sup>Center for Advancing Electronics Dresden

The migration of ions in perovskite materials has been linked to many negative phenomena such as hysteresis and device degradation. Herein, we demonstrate that, under certain conditions, the migration of ions may also lead to an improvement in device performance. Specifically, by performing ultra-violet photoemission spectroscopy depth profiling experiments, we show that upon the application of an electric field during PV characterization of MAPbI<sub>3</sub> inverted architecture solar cells, the migration of iodine ions to the interface between the perovskite active layer and electron extraction layer results in the modification of the electronic structure at that interface. The increase in the density of iodine ions leads to a band bending, resembling the electronic structure of iodine-rich overstoichiometric MAPbI<sub>3</sub> films. This change in the interfacial electronic structure results in a significant enhancement of the built-in potential and open-circuit voltage of the photovoltaic cells in subsequent measurements, commonly observed by researchers in the field. Finally, our results are corroborated by numerical device simulations which include the presence and migration of ions.

HL 35.6 Tue 15:15 POT 251

**Reduction of s-shape formation induced by transparent top-contact sputter deposition in Perovskite solar cells** — ●MARLENE HÄRTEL<sup>1,2</sup>, RUSLAN MUYDINOV<sup>1</sup>, STEVE ALBRECHT<sup>1,2</sup>, and BERND SZYSZKA<sup>1,2</sup> — <sup>1</sup>Technische Universität, Berlin, Deutschland — <sup>2</sup>Helmholtz-Zentrum, Berlin, Deutschland

Depositing transparent top electrodes in Perovskite tandem solar cells without adequate buffer layers leads to damaging of the sensitive organic charge transport layers (CTL), due to harsh conditions during the sputter process. However, commonly used buffer layers utilize thermal atomic layer deposition, which is time consuming and could induce reactions with moisture. Therefore, direct sputter deposition is preferred, but sputter damage is mirrored in an s-shaped current-voltage characteristic, accompanied by a fill factor loss. In this contribution, the origin of the s-shape is analyzed by employing photoluminescence, and intensity and temperature dependent current-voltage measurement, and identified as an energetic barrier, which is formed at

the CTL. The findings are supported by electrical simulations based on SCAPS. Moreover, two different sputter deposition techniques, namely radio-frequency magnetron and hollow cathode gas flow sputtering, are implemented and compared with regard to their application in damage-free sputter-deposition. By varying sputter process conditions, such as the power, or sample to target position, the s-shape in the solar cell devices can be reduced, thereby improving their electrical performance. Sputter damage-free TCO deposition allows for future enhancements in tandem device fabrication and their commercialization.

HL 35.7 Tue 15:30 POT 251

**Structural and optical properties of defect-engineered organic-inorganic halide perovskites** — •CHANG-MING JIANG<sup>1</sup>, WEN-YU CHENG<sup>1</sup>, MICHAEL EHRENREICH<sup>2</sup>, GREGOR KIESLICH<sup>2</sup>, and IAN SHARP<sup>1</sup> — <sup>1</sup>Walter Schottky Institut, Technische Universität München — <sup>2</sup>Fakultät für Chemie, Technische Universität München

Incorporating large organic cations into the ABX<sub>3</sub> hybrid perovskite is known to yield the so-called 'hollowed perovskite' structure. To accommodate the oversized cation onto the A-site, certain concentrations of B<sup>2+</sup> cation and X<sup>-</sup> anion vacancies form. The presence of these point defects and associated dangling bonds are expected to affect the electronic properties, charge carrier dynamics, and phase stability. In the case of FASnI<sub>3</sub>, incorporation of ethylenediammonium (en<sup>2+</sup>) cations enlarges the optical bandgap and significantly improves the photovoltaic efficiency and stability. In this work, we aim at understanding the composition-dependent optical properties and relative phase stability in the defect-engineered (en)MAPbI<sub>3</sub> system. Thin films with tunable bandgaps from 1.60-1.85 eV are fabricated, and the non-radiative recombination pathways associated with intentionally added point defects are studied by time-resolved PL techniques. Additionally, by measuring temperature dependence of PL, we find that both the orthorhombic-to-tetragonal and tetragonal-to-cubic

phase transition temperatures are affected by the extent of large organic cation incorporation. This work sheds light on the interactions between organic cations and the inorganic lattice and provides insights into the defect tolerance in hybrid halide perovskite semiconductors.

HL 35.8 Tue 15:45 POT 251

**Band Gap Engineering of Double Perovskite Halides Cs<sub>2</sub>AgBiCl<sub>6</sub> through Different Sites Alloying** — •DAN HAN, MASAKO OGURA, ANDREAS HELD, and HUBERT EBERT — Department of chemistry, Ludwig-Maximilians-Universität München, Munich, Germany

Recently, double perovskite halides have been proposed as potential Pb-free photovoltaic materials. However, many experimentally or theoretically reported double perovskite halides have indirect and large band gaps, hindering efficient sunlight absorbing. Thus, tuning the band gap of double perovskite halides is required for their future photovoltaic application. Forming alloys is a practical way for band gap engineering. Here, we presented a systematic study of alloying on different sites for a representative double perovskite halide Cs<sub>2</sub>B'B''X<sub>6</sub> (B'=Ag, B''=Bi, X=Cl) using the fully relativistic Korringa-Kohn-Rostoker (KKR) Green function in combination with the coherent potential approximation (CPA) method. Cs<sub>2</sub>Na<sub>x</sub>Ag<sub>1-x</sub>BiCl<sub>6</sub>, Cs<sub>2</sub>AgSb<sub>x</sub>Bi<sub>1-x</sub>Cl<sub>6</sub> and Cs<sub>2</sub>AgBi(Br<sub>x</sub>Cl<sub>1-x</sub>)<sub>6</sub> (x=0.25, 0.5, 0.75) all show a band gap bowing behavior, i.e., a nonlinear band gap dependence on the chemical composition. Additionally, we evaluated the mixing thermodynamical stability of Cs<sub>2</sub>AgSb<sub>x</sub>Bi<sub>1-x</sub>Cl<sub>6</sub> and show Cs<sub>2</sub>AgSb<sub>x</sub>Bi<sub>1-x</sub>Cl<sub>6</sub> alloy is thermodynamically stable at room temperature. Bloch spectral functions of alloys with different compositions exhibit broadening due to CPA technique. Alloys at B' and B'' sites could tune the finite lifetime of carrier more effectively than alloys at X site.