CPP 3: Perovskite and Photovoltaics 1 (joint session HL/CPP/KFM)

Time: Monday 9:30–12:45 Location: H34

CPP 3.1 Mon 9:30 H34

The Electronic Structure of $Cs_2AgBiBr_6$ at Room Temperature — •Julian Gebhardt[1,2] and Christian Elsässer[1,2,3] — [1]Fraunhofer Institute for Mechanics of Materials IWM, 79108 Freiburg — [2]Cluster of Excellence livMatS at FIT - Freiburg Center for Interactive Materials and Bioinspired Technologies, Albert-Ludwigs-University Freiburg, 79104 Freiburg — [3]Freiburg Materials Research Center (FMF), Albert-Ludwigs-University Freiburg, 79104 Freiburg

$Cs_2AgBiBr_6$ is a stable halide double perovskite with a band gap of about 2.2 eV. Therefore, it is intensively studied as possible lead free alternative to hybrid perovskite solar cell absorber materials such as methylammonium-lead iodide. However, power conversion efficiencies of solar cells with this material have not yet exceeded 3%. A detailed understanding of the electronic structure of this material is difficult, due to the variance of reported data and experimental as well as theoretical difficulties that occur in going beyond a qualitative understanding of such an indirect semi-conductor at device operation temperature. Here we combine self-energy corrected electronic-structure theory including spin-orbit coupling and structural dynamics at room temperature to model and understand this compound in a quantitative manner, and we compare our theoretical findings with experimental ones. Based on an achieved good agreement, we propose that the observed low power conversion efficiencies can be attributed to the density of states in the conduction band region. From the relation between dimensionality and electron conductivity, we suggest a general design principle for absorber material search.

CPP 3.2 Mon 9:45 H34

Photon-echo spectroscopy of a $CH_3NH_3PbI_3$ perovskite single crystal — •Stefan Grisard[1], Artur V. Trifonov[1,2], Aleksandr N. Kosarev[1,3], Ilya A. Akimov[1,3], Dmitrii R. Yakovlev[1,3], Julian Höcker[4], Vladimir Dyakonov[4], and Manfred Bayer[1,3] — [1]Experimentelle Physik 2, Technische Universität Dortmund — [2]Spin Optics Laboratory, St. Petersburg State University, Russia — [3]St. Petersburg, Russia — [4]Experimental Physics 6, Julius-Maximilian University of Würzburg

Lead halide perovskites such as $CH_3NH_3PbI_3$ ($MAPbI_3$) show outstanding characteristics important for photovoltaic and optoelectronic applications. However, the peculiarities of light-matter interactions in these materials are far from being fully explored. Here, we applied time-resolved photon echo spectroscopy to a high quality $MAPbI_3$ single crystal highlighting the importance of inhomogeneous broadening of excitonic transitions even at cryogenic temperatures. Furthermore, we developed an experimental photon-echo polarimetry method that unambiguously identifies contributions from exciton and biexciton to the coherent optical response. Most importantly, our method allows to accurately extract the biexciton binding energy of 2.4meV, even though the period of the observed quantum beats exceeds the coherence times of exciton and biexciton.

CPP 3.3 Mon 10:00 H34

Structural properties of (hot-)pressed $MAPbI_3$ films revealed by detailed temperature-dependent optical analyses — •Christina Witt[1], Konstantin Schötz[1], Nico Leupold[2], Simon Biberger[1], Philipp Ramming[1], Ralf Moos[2], and Fabian Panzer[1] — [1]Soft Matter Optoelectronics, University of Bayreuth, Bayreuth 95440, Germany — [2]Department of Functional Materials, University of Bayreuth, Bayreuth 95440, Germany

Halide perovskites attracted much attention in recent years, due to the remarkable increase in corresponding solar cell efficiencies. More recently, hot-pressing has emerged as attractive method for manufacturing and post-treatment of perovskite films [1, 2]. However, a detailed understanding regarding the role of temperature during hot-pressing on resulting film properties is still missing. Thus, we use temperature-dependent PL and absorption measurements of $MAPbI_3$ thin films pressed with different temperatures and in detail analyze their optical properties. This allows us to draw conclusions about structural and optoelectronic properties, revealing that an increased temperature improves film morphology, structural and optoelectronic film properties.

[1] Witt, C. et al. Impact of Pressure and Temperature on the Compaction Dynamics and Layer Properties of Powder-Pressed Methylammonium Lead Halide Thick Films. ACS Appl. Electron. Mater. 2020, 2 (8), 2619-2628.

[2] Pourdavoud, N. et al. Room-Temperature Stimulated Emission and Lasing in Recrystallized Cesium Lead Bromide Perovskite Thin Films. Adv. Mater. 2019, 31, 1903717.

CPP 3.4 Mon 10:15 H34

Application of atomic layer deposition and x-ray photoelectron spectroscopy in perovskite solar cells — •Małgorzata Kot[1], Chittaranjan Das[2], Lukas Kegelmann[3], Hans Koebler[3], Mikhailo Vorokhta[4], Carlos Escudero[5], Steve Albrecht[3], Antonio Abate[3], and Jan Ingo Flege[1] — [1]BTU Cottbus-Senfteberg, Cottbus, Germany — [2]KIT, Eggenstein-Leopoldshafen, Germany — [3]HZB, Berlin, Germany — [4]Charles University, Prague, Czech Republic — [5]ALBA Synchrotron, Cerdanyola del Vallès, Spain

In this work we have utilized near-ambient pressure and ultra-high vacuum X-ray photoelectron spectroscopy as well as atomic layer deposition to investigate perovskite solar cells (PSCs). We have demonstrated that ultrathin room temperature atomic layer-deposited aluminium oxide on the perovskite surface very effectively suppresses iodine migration[1] and improves the long term stability and efficiency of PSCs [2,3]. Furthermore, exposure to light proves more detrimental to the perovskite film than exposure to water vapor.[2] Absorbed photons create Frenkel defects in the perovskite crystal and their number strongly depends on the used illumination. The higher the photon flux, the higher the concentration of Frenkel defects, and thus the stronger the degradation of power conversion efficiency and the stronger the hysteresis in the J-V characteristics. [1] C. Das, M. Kot et al., Cell Reports Physical Science 2020, 1, 100112. [2] M. Kot et al., ChemSusChem 2020, 13, 5722. [3] M. Kot et al., ChemSusChem 2018, 11, 3640.

CPP 3.5 Mon 10:30 H34

Chemical Engineering of Ferroelastic Twin Domains in MAPbI3 Thin Films — •Yenal Yalcinkaya[1], Ilka Hermes[1], Tobias Seewald[2], Katrin Amann-Winkel[1], Lothar Veith[1], Lukas Schmidt-Mende[2], and Stefan A.L. Weber[1] — [1]Max Planck Institute for Polymer Research, Ackermannweg 10, 55128 Mainz, Germany — [2]Department of Physics, University of Konstanz, Universitätsstr. 10, 78464, Germany

In this study, we introduce a new chemical method for controlling the strain in methylammonium lead iodide (MAPbI3) perovskite crystals by varying the ratio of Pb(Ac)2 and PbCl2 in the precursor solution. We used a combination of piezoresponse force microscopy (PFM) and X-ray diffraction (XRD) to observe the effect on crystal strain. We observed larger ferroelastic twin domains upon increasing the PbCl2 content, indicating increased crystal strain via PFM images. We confirmed the increased crystal strain via the XRD patterns with strong crystal twinning features. We suggest that this behaviour is caused by different evaporation rates of methylammonium acetate and methylammonium chloride which led to a strain gradient during the crystallization as revealed by time-of-flight secondary ion mass spectroscopy (ToF-SIMS) and grazing incidence x-ray diffraction (GIXRD) measurements. We observed films with larger twin domain structures show an increased carrier via time-resolved photoluminescence (TRPL). The results demonstrate the potential of chemical strain engineering as an easy method for controlling strain-related e*ects in lead halide perovskites.

CPP 3.6 Mon 10:45 H34

Inspecting the local structure of cubic phase halide perovskites from first-principles — •Xiangzhou Zhu, Sebastián Caicedo-Dávila, Christian Gehrmann, and David A. Egger — Department of Physics, Technical University of Munich, Garching, Germany

Halide perovskites (HaPs) have been identified as one of the most promising optoelectronic materials in recent years. Different from the conventional inorganic semiconductors, HaPs exhibit profound deviations from their average atomic structure at finite temperature, which have important consequences for their optoelectronic properties. However, a detailed understanding of these local structural fluctuations, the underlying physical mechanisms as well as their consequences is far from complete. Here, we perform molecular dynamics (MD) calculations based on density functional theory (DFT) to investigate the local structure and anharmonic dynamics of $CsPbBr_3$ in the cubic phase at T=425 K and 525K. We find that motions of neighboring Cs-Br atoms interlock within a nominal cubic unit cell. This manifests in the most likely Cs-Br distance being significantly shorter than what is inferred from an ideal cubic structure. Furthermore, we use the statistical information on the dynamic atomic distributions to quantify the effective potential associated with certain atomic motions at two temperatures. We find that Br motions occur in a dynamically disordered potential energy landscape and relate the Cs motion as well the Cs-Br coupling to $PbBr_6$ octahedral rotations.

30 min. break

CPP 3.7 Mon 11:30 H34

Distinct Resonances in Absorption Spectra of Lead Halide-based Quantum Dots — •Anja Barfüsser, Quinten A. Akkerman, Sebastian Rieger, Amrita Dey, Ahmet Tosun, Tushar Debnath, and Jochen Feldmann — Chair for Photonics and Optoelectronics, Nano-Institute Munich and Department of Physics, Ludwig-Maximilians-Universität (LMU), Königinstr. 10, 80539 Munich, Germany

In recent years, perovskite nanocrystals have attracted much attention for their unique optical properties. Here, we discuss sphere-like lead halide-based quantum dots with diameters in the range of 4.5-12 nm featuring a multitude of distinct resonances in their absorption spectra. We have investigated the nature of these resonances by comparing experimental data with model calculations based on weak and strong confinement. In transient absorption experiments, bleaching and induced absorption signals are observed, which we discuss in terms of confined excitons and biexcitonic contributions.

CPP 3.8 Mon 11:45 H34

Revealing the doping density in perovskite solar cells and its impact on device performance — •Francisco Peña-Camargo and Martin Stolterfoht — Physik weicher Materie, Institut für Physik und Astronomie, Universität Potsdam, Karl-Liebknecht-Str. 24-25, 14776 Potsdam, Germany

Inorganic semiconductors can be electronically doped with high precision. Conversely, there is still conjecture regarding the assessment of the electronic doping density in metal-halide perovskites, not to mention of a control thereof. This study presents a multifaceted approach to determine the electronic doping density for a range of different lead-halide perovskite systems. Optical and electrical characterisation techniques comprising intensity-dependent and transient photoluminescence, AC Hall effect, transfer-length-methods, and charge extraction measurements were instrumental in quantifying an upper limit for the doping density. The obtained values are subsequently compared to the electrode charge per cell volume at short-circuit conditions (CU_{bi}/eV), which amounts to roughly 10^{16} cm^{-3}. This figure of merit represents the critical limit below which doping-induced charges do not influence the device performance. The experimental results demonstrate consistently that the doping density is below this critical threshold ($< 10^{12}$ cm^{-3} which means $\ll CU_{\mathrm{bi}}/eV$) for all common lead-based metal-halide perovskites. Nevertheless, although the density of doping-induced charges is too low to redistribute the built-in voltage in the perovskite active layer, mobile ions are present in sufficient quantities to create space-charge-regions in the active layer.

CPP 3.9 Mon 12:00 H34

Ground-state structures, electronic structure, transport properties and optical properties of anion-ordered anti-Ruddlesden-Popper phase oxide perovskites — •Dan Han, Shizhe Wang, Thomas Bein, and Hubert Ebert — Department Chemie, Ludwig-Maximilians-Universität München, Germany

Anti-Ruddlesden-Popper (ARP) phase oxide perovskites Ca4OA2 (A = P, As, Sb, Bi) have recently attracted great interest in the field of ferroelectrics and thermoelectrics, while their optoelectronic application is dominantly limited by their indirect band gaps. In this work, we consider A-site anion ordering in Ca4OA2 (A = P, As, Sb, Bi), and find that it induces an indirect-to-direct band gap transition. Using first-principles calculations, we study the ground-state structures, electronic structure, transport properties and optical properties of anion-ordered ARP phase oxide perovskites Ca4OAA'. Based on an analysis of the lattice dynamics, the ground-state structures of Ca4OAsSb, Ca4OAsBi, Ca4OPSb and Ca4OPBi are identified. In contrast to the Ruddlesden-Popper (RP) phase oxide and halide counterparts, Ca4OAA' show larger band dispersion along the out-of-plane direction, smaller band gaps and highly enhanced out-of-plane mobilities, which is ascribed to the short interlayer distances and enhanced covalency of the pnictides. Although the out-of-plane mobilities of these n = 1 ARP phase perovskites highly increase, comparatively strong polar optical phonon (POP) scattering limits the further enhancement of their mobilities. This work shows that these anion-ordered Ca4OAA' exhibit the potential for optoelectronic applications.

CPP 3.10 Mon 12:15 H34

Including light management concepts in performance prediction modelling of perovskite-silicon tandem solar cells by implementing transfer matrix method — Aminreza Mohandes[1,2], Peymaneh Rafieipour[1,2], Mohammad Moaddeli[1], and •Mansour Kanani[1] — [1]Department of Materials Science and Engineering, School of Engineering, Shiraz University, Shiraz, Iran — [2]Department of Physics, Shiraz University, Shiraz, Iran

The 2-T monolithic perovskite-silicon tandem design holds a record efficiency of 29.80%, recently. To perform more accurate, complete and experimentally reliable modelling of tandem solar cell, we adopt the transfer matrix method (TMM) which incorporates the interfacial reflections, light scattering and parasitic absorption losses in the calculation of the light transmitted from the top perovskite solar cell. The results reveal that the light scattering and interfacial reflection losses cannot be ignored and the previously used Beer-Lambert exponential relation is insufficient for studying tandem configuration. Including TMM method in the performance optimization of the tandem solar cells lets to consider light management concepts more extensively. Therefore, identifying and reducing optical losses in each layer/interface and designing appropriate anti-reflection coatings in a multilayer tandem simulation can be achieved. In this study, standalone and tandem devices have been analyzed and the effect of absorber layer thickness variation, J-V curves, external quantum efficiency (EQE), filtered spectra, current matching, and tandem performance parameters on the cell efficiency is considered.

CPP 3.11 Mon 12:30 H34

Highly Efficient Perovskite-on-Silicon Tandem Solar Cells on Planar and Textured Silicon — •Christian M. Wolff[1], Xin Yu Chin[2], Kerem Artuk[1], Deniz Türkay[1], Daniel Jacobs[1], Quentin Jeangros[2], and Christophe Ballif[1,2] — [1]École polytechnique fédérale de Lausanne, STI IEM PVLAB, Rue de la Maladière 71b, 2000 Neuchâtel — [2]Centre Suisse d'Electronique et de Microtechnique, Rue Jaquet-Droz 1, 2002 Neuchâtel

Multi-junction devices offer the possibility to harness the sun‘s light beyond the limitations of single-junction solar cells. Among the different combinations perovskite-on-silicon (Pk/Si) tandems hold the great promise of high efficiencies >30%, while maintaining low cost. I will report on our latest progress in the development of Pk/Si tandems comparing our efforts on single-side and double-side textured Pk/Si tandems, reaching a V_{OC} up to 1.95V, summed short-circuit currents above 41mA/cm^2, and certified efficiencies >29%, on an active area of 1cm^2. We achieved these results by dedicated electrical and optical optimizations of all layers within the stack. Specifically, we reduced recombination and transport losses in the Pk absorbers through process and additive engineering for both solution-processed one-step and hybrid two-step deposited Pks, and improved the transparency of the front stack electrodes and contacts through simulation-guided optimizations of the front grid and layer thicknesses. Furthermore, we investigated the stability of single-junction Pk and tandem devices under reverse-bias and standardized accelerated aging conditions.