KFM 4: Perovskite and photovoltaics I (joint session HL/KFM)

Time: Monday 9:30–13:00 Location: EW 203

KFM 4.1 Mon 9:30 EW 203

Improving Performance of Two-Step Processed Perovskite Top Cells for Tandem Photovoltaic Applications — •Ronja Pappenberger[1,2], Alexander Diercks[2], Julian Petry[1,2], Paul Fassl[1,2], and Ulrich W. Paetzold[1,2] — [1]Institute of Microstructure Technology, KIT, Germany — [2]Light Technology Institute, KIT, Germany

For high-performance application of perovskite solar cells (PSCs) in monolithic perovskite/silicon tandem configuration, an optimal bandgap and process method of the perovskite top cell is required. While the two-step method leads to regular perovskite film crystallization, engineering wider bandgaps ($E_g > 1.65$ eV) for the solution-based two-step method remains a challenge. This work introduces an effective and facile strategy to increase the bandgap of two-step solution-processed perovskite films by incorporating bromide in both deposition steps, the inorganic (step 1, $PbBr_2$) and the organic (step 2, FABr) precursor deposition. This strategy yields improved charge carrier extraction and quasi-Fermi level splitting with PCEs up to 15.9%. Further improvements are achieved by introducing CsI in the bulk and utilizing LiF as surface passivation, resulting in a stable power output exceeding 18.9% for $E_g = 1.68$ eV. This additional performance boost arises from enhanced perovskite film crystallization, leading to improved charge carrier extraction. Laboratory scale planar monolithic perovskite/silicon tandem solar cells (TSCs) (1 cm^2 active area) achieve PCEs up to 25.7%. In addition, research is being conducted into implementation on textured monolithic perovskite/silicon TSCs.

KFM 4.2 Mon 9:45 EW 203

Optical In-Situ spectroscopy and reactive spin coating for improved control of perovskite thin film fabrication — •Simon Biberger, Maximilian Spies, Konstantin Schötz, Frank-Julian Kahle, Nico Leupold, Ralf Moos, Helen Grüninger, Anna Köhler, and Fabian Panzer — University of Bayreuth, Bayreuth, Germany

Efficient solar cells require a high-quality halide perovskite (HP) film, which is typically achieved through a solution-based solvent engineering spin coating approach. Here, HP crystallization is induced by applying an antisolvent (AS) to the precursor solution film on the spinning substrate after a specific spinning time. The process involves various controllable and uncontrollable parameters that need to be considered. As a result, fabrication recipes (such as spin speed or timing of AS dispensing) are usually developed empirically and vary between labs. Additionally, factors like changes in the atmosphere that are hard to control can introduce substantial variations between and even within batches of devices. In this work we developed a closed-loop feedback system based on our multimodal optical In-Situ spin coater system in combination with a real-time analysis of the optical spectra during spin coating. We monitor the solvent layer thickness as the parameter of interest during the spin coating. When the target level is reached, the HP crystallization is induced by dispensing the AS via a syringe pump. This method compensates for the effects of uncontrolled parameters, like variation in solvent evaporation rate due to atmospheric changes, thus leading to reproducible film quality.

KFM 4.3 Mon 10:00 EW 203

Enhanced Circular Dichroism and Polarized Emission in an Achiral, Low Bandgap Bismuth Iodide Perovskite — •Philip Klement[1], Jakob Möbs[2], Gina Stuhrmann[3], Lukas Gümbel[1], Marius Müller[1], Johanna Heine[2], and Sangam Chatterjee[1] — [1]Institute of Experimental Physics I, JLU Gießen, Germany — [2]Department of Chemistry, PUM, Marburg, Germany — [3]Institute of Nanotechnology (INT), KIT, Eggenstein-Leopoldshafen, Germany

Lead halide perovskites and related materials incorporating chiral organic cations exhibit intriguing properties such as chiroptical activity and chirality-induced spin selectivity enabling the generation and detection of circularly polarized light. However, understanding the structural origin of chiroptical activity presents a challenge due to macroscopic factors and experimental constraints. Here, we present the achiral perovskite derivative $[Cu_2(pyz)_3(MeCN)_2][Bi_3I_{11}]$ (pyz = pyrazine; MeCN = acetonitrile), which displays remarkable circular dichroism (CD) arising from the material*s noncentrosymmetric structure. CuBiI exhibits a low, direct optical band gap of 1.70 eV and both linear and circular optical activity with a substantial anisotropy factor of up to 0.16. Intriguingly, despite the absence of chiral building blocks, CuBiI exhibits a substantial degree of circularly polarized photoluminescence, reaching 4.9%. This value is comparable to the results achieved by incorporating chiral organic molecules into perovskites, typically ranging from 3 to 10% at zero magnetic field. Our findings shed light on the macroscopic origin of CD and provide valuable insights for the design of materials with high chiroptical activity.

KFM 4.4 Mon 10:15 EW 203

Explaining the tetragonal distortion of chalcopyrites and solving the puzzle of the peculiar bond length dependence in (Ag,Cu)(In,Ga)Se_2 — •Hans H. Falk[1], Stefanie Eckner[1], Konrad Ritter[1], Sergiu Levcenko[1], Timo Pfeiffelmann[1], Edmund Welter[2], Jes Larsen[3], William N. Shafarman[4], and Claudia S. Schnohr[1] — [1]Felix Bloch Institute for Solid State Physics, Leipzig University, Germany — [2]Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — [3]Department of Materials Science and Engineering, Uppsala University, Sweden — [4]Department of Materials Science and Engineering, University of Delaware, Newark, USA

(Ag,Cu)(In,Ga)Se_2 is a highly efficient thin film solar cell absorber. EXAFS studies of (Ag,Cu)InSe_2 and (Ag,Cu)GaSe_2 show that the element specific bond length of the common cation (In-Se or Ga-Se) decreases with increasing Ag content even though the lattice expands. This peculiar bond length dependence is not only counter-intuitive but also the opposite of what is commonly observed in other chalcopyrite alloys, like Cu(In,Ga)Se_2 and Ag(In,Ga)Se_2, where the bond length of the common cation (Cu-Se or Ag-Se) increases as the lattice expands. This puzzling difference in the behavior of the common cation bond lengths can be explained by minimizing the distortion energy (Keating potential) of bond angles and bond lengths. With simple calculations, focusing on the local tetrahedral neighborhood, we are able to reproduce both the tetragonal distortion and the bond length behavior of various chalcopyrite alloys, leading to a deepened understanding of these technologically relevant photovoltaic absorber materials.

KFM 4.5 Mon 10:30 EW 203

Nonlocal many-body dispersion for bulk properties: The particular case of cesium-based halide perovskites — Lin Yang[1], Ya Gao[1], Jingrui Li[2], •Guo-Xu Zhang[1], and Zhenbo Wang[1] — [1]Harbin Institute of Technology, Harbin, P. R. China — [2]Xi'an Jiaotong University, Xi'an, P. R. China

Cesium-based all-inorganic halide perovskites have shown excellent photovoltaic properties. However, phase stability and transformations in different polymorphs remain rather poorly understood. Here, we investigate the cohesive, electronic properties and structural instabilities of $CsPbI_3$ and $CsSnI_3$ based on first-principles density-functional theory (DFT) calculations. Care has been taken to choose exchange-correlation functionals and zero-point vibrational effects have been included by performing phonon calculations. In particular, we compare different van der Waals (vdW) corrections to the DFT approaches. Our results demonstrate that nonlocal many-body dispersion interactions play a vital role in predicting the cohesive properties and phonon spectra of halide perovskites studies here. In addition, we find the anharmonicity cannot be neglected in order to correctly predict the phase stability and transitions of polymorphs. The quasiharmonic approximation method that partially include the anharmonicity is able to improve the predictions, in particular for $CsSnI_3$. We stress that both nonlocal many-body dispersion and full anharmonic terms should be considered for accurate studies of such type of materials.

KFM 4.6 Mon 10:45 EW 203

Changes in Polarization Mechanisms Following Dimensional Reduction of the Double Perovskite Octahedral Network in $Cs_2AgBiBr_6$ — •Tim P. Schneider, Jonas Glaser, Jonas Horn, and Derck Schlettwein — Institut für Angewandte Physik, Justus-Liebig-Universität Gießen

Introduction of large organic cations into metal halide perovskites leads to dimensional reduction of the octahedral metal-halide network towards so-called 2D-perovskites with significant changes in the electronic system. Such 2D-perovskites are recently studied to enhance the performance of perovskite solar cells by reducing hysteretic behavior and improving the contact formation of perovskite absorbing layers to transport layers. In this work, 2D $PEA_4AgBiBr_8$ is derived from its 3D counterpart $Cs_2AgBiBr_6$ by replacing Cs^+ by the large organic phenethylammonium (PEA^+) cation. Thin films of both materials, 3D $Cs_2AgBiBr_6$ or 2D $PEA_4AgBiBr_8$, were prepared onto microstructured metal electrode arrays to perform *I-U* measurements and spatially resolved Kelvin probe force microscopy. These experiments serve to characterize the contact formation of both materials and discuss hysteresis caused by ion migration in response to an applied electric field. It was revealed how the dimensional reduction of the octahedral network affects the polarization caused by ionic movements and the formation of contact resistances in the films and at the respective interface to the metal electrodes. Differences among the materials, implications for technical applications in devices and the respective origin of the different observed polarization phenomena will be discussed.

15 min. break

Invited Talk KFM 4.7 Mon 11:15 EW 203

Influence of the Organic Cation Orientation on the Absorption Spectra of 2D Hybrid Organic-Inorganic Perovskites — •Svenja Janke — Department of Chemistry, University of Warwick, Coventry, UK

Hybrid organic-inorganic perovskites (HOIPs) allow combining organic and inorganic materials at the nanoscale and hence open up a wide area of tunability. In two-dimensional HOIPs, both organic and inorganic components can contribute to the electronic frontier levels. For the design of new devices like solar cells, a fundamental understanding of the electronic excitations, their photophysical signatures and the underlying atomic structure is essential. The 2D HOIP inorganic exciton binding energy depends approximately linearly on the inorganic band gap. To estimate the inorganic exciton contribution to the absorption spectrum, I benchmark the amount of Hartree Fock exchange in hybrid density functional theory calculations including spin-orbit coupling.

For the quaterthiophene-based 2D HOIP $(AE4T)PbX_4$, variation of the halide anion leads to structural changes in the organic layer, causing changes in the absorption spectra. The bithiophene-based 2D HOIP $(AE2T)PbI_4$ shows disorder along the stacking direction in X-ray scattering experiments, resulting in several possible atomic structural models for the organic component.

Here, I demonstrate that we can use a Frenkel-Holstein-Hamiltonian-based model to investigate how different orientations of the organic cation alter the organic and inorganic exciton contribution to the absorption spectrum.

KFM 4.8 Mon 11:45 EW 203

Optical Simulations of Light Management in Ultrathin CIGSe Solar Cells with Nanophotonic Back Contacts — •Daniel Jimenez Tejero, Merve Demir, Thomas Schneider, Bodo Fuhrmann, Roland Scheer, Ralf Wehrspohn, and Alexander Sprafke — Martin-Luther-Universität Halle-Wittenberg, Institut für Physik, 06120 Halle (Saale), Germany

Ultrathin $Cu(In,Ga)Se_2$ (CIGSe) solar cells are in development to decrease material usage and broaden the range of applications. However, CIGSe absorber layers with thicknesses below 1 μm suffer from reduced light absorption, especially for wavelengths near the bandgap, leading to decreased power conversion efficiency. To counteract this, we employ nanostructured functional back contacts to effectively increase the optical path length within the CIGSe absorber layer through nanophotonic mechanisms such as scattering and coupling into thin film modes. The focus lies on CIGSe absorber layers with a thickness of 300 nm to 500 nm deposited on SiO_2 nanostructures on a flat aluminium or gold back contact. Our approach employs nano-optical simulations using the finite element method and aims to identify designs with promising photocurrent enhancements feasible for fabrication and to clarify the underlying mechanisms.

KFM 4.9 Mon 12:00 EW 203

Multi-Phase Optical Exciton Dynamics in Mixed (2D/3D) Inorganic-Organic Hybrid Semiconductors — •Mohammad Adnan[1,2], Pawan Kumar Kanaujia[2], Kshetra Mohan Dehury[2], and Gaddam Vijaya Prakash[2] — [1]Institute of Physics, University of Münster, Wilhelm-Klemm-Straße 10 48149 Münster, Germany — [2]Nanophotonics Lab, Department of Physics, Indian Institute of Technology Delhi, Hauz Khas, New Delhi 110016 India

Inorganic-organic (IO) hybrid semiconductors are promising optoelectronic materials due to their unique crystal packing, the wide variety of crystal phases, the wide bandgap tunability and the associated optical exciton characteristics. Here, we investigate the digitized intercalation process of various organic moieties, resulting in a mixed IO hybrid system of type (R-NH3)2(R'-NH3)n-1PbnI3n+1. By employing continuous photoluminescence monitoring, we observe a progressive and dynamic structural evolution and elucidate the underlying mechanisms occurring during the intercalation process. The interplay of (i) cyclic (ii) long alkyl chain and (iii) small alkyl amine based organic moieties during the intercalation leads to the formation of either 2D ((R-NH3)2PbI4) or 3D (R'-NH3PbI3) IO hybrid networks and causes significant structural phase variations within the 2D and 3D crystal packings.

KFM 4.10 Mon 12:15 EW 203

Exploring Mixed-Metal Chalcohalides $M(II)_2M(III)Ch_2X_3$ Compounds for Photovoltaic Applications — •Pascal Henkel[1], Jingrui Li[2], and Patrick Rinke[1] — [1]Department of Applied Physics, Aalto University, P.O.Box 11100, FI-00076 AALTO, Finland — [2]School of Electronic Science and Engineering, Xi'an Jiaotong University, Xi'an 710049, China

New photovoltaic materials are needed to increase power conversion efficiencies (PCEs), reduce costs, and improve device longevity to facilitate the renewable energy transformation. In this context, perovskite-inspired quaternary mixed-metal chalcohalides $M(II)_2M(III)Ch_2X_3$ have emerged as an interesting materials class, that has the potential to overcome the stability and toxicity problems of the currently favoured halid perovskites [1], and still deliver high PCEs [2].

In this study, we apply density functional theory to identify new $M(II)_2M(III)Ch_2X_3$ compounds. We considered a total of 54 materials each in three different space groups ($Cmcm$, $Cmc2_1$ and $P2_1/c$) for which we computed the energetic stability and the band gaps with the HSE06 hybrid functional. We identified a total of 22 $M(II)_2M(III)Ch_2X_3$ materials, which fulfill our stability requirements and have a direct band gap in the range 0.7 eV to 2 eV. Out of the 22, 8 lead-free and 9 lead-containing materials are new [3]. Overall for all 54 compounds, $P2_1/c$ is the thermodynamically preferred phase, whereas direct band gaps occur predominantly for $Cmcm$ and $Cmc2_1$.

[1] *Z. Anorg. Allg. Chem.* **468**, 91-98 (1980). [2] *Mater. Horiz.* **8**, 2709 (2021), [3] *Chem. Mater.* **35**, 7761-7769 (2023).

KFM 4.11 Mon 12:30 EW 203

Effective model for charge transport in hybrid organic-inorganic materials dominated by molecule-lattice interactions — •Florian Kluibenschedl, Georgios Koutentakis, Ragheed Alhyder, and Mikhail Lemeshko — Institute of Science and Technology Austria (ISTA), Am Campus 1, 3400 Klosterneuburg, Austria

The growing importance of crystalline hybrid materials with embedded molecular entities, such as hybrid organic-inorganic perovskites (HOIPs) or molecular organic frameworks, requires extensions of the conventional picture of charge transport based on electron-phonon interactions. Here we present a minimal, coarse-grained, two-dimensional model of charge transport based on the interactions of charge carriers with freely rotating molecules embedded in the inorganic lattice. Our phase diagram analysis reveals the presence of four phases, characterized by the distinct ordering of the molecular dipole moments, which strongly depends on the tunneling anisotropy of the carriers as well as on the strength of carrier-molecule interactions. The most striking outcome is the coexistence of degenerate ferroelectric phases of reduced symmetry beyond a certain threshold of interaction strength, even in the case of isotropic tunneling. The coexistence of these phases motivates a symmetry breaking mechanism similar to the Jahn-Teller effect, which introduces transport anisotropy into an otherwise symmetric system. We relate these results to the proposed formation of ferroelectric large polarons as an explanation of the remarkable optoelectronic properties of HOIPs.

KFM 4.12 Mon 12:45 EW 203

Cavity-enhanced absorption measurements of perovskite nanocrystals — •Ines Amersdorffer[1], Andreas Singldinger[1], Thomas Hümmer[1,2], Alexander Urban[1], and David Hunger[3] — [1]Faculty of physics, LMU Munich, Germany — [2]Qlibri GmbH, Munich, Germany — [3]Physikalisches Institut, KIT, Germany

The search for optimal materials for optoelectronic devices such as solar cells or quantum light emitters has piqued significant interest in perovskite nanocrystals. This is due to their outstanding properties, which range from easy, scalable synthesis at room temperature to ultra-wide colour tunability and high quantum efficiency.

Spectroscopy on single perovskite crystals helps to reveal their excitonic fine structure, which in thin films is concealed by inter-particle phenomena. However, measuring the marginal absorption of such nanocrystals is challenging. One way to overcome this issue is to use an optical resonator in which the light passes through the nanoscale sample multiple times and thereby enhances its absorption to a measurable amount.

To this end, we use a high-finesse microcavity to perform wavelength dependent absorption measurements on perovskite nanocubes. These are complemented by low-temperature photoluminescence and scanning electron microscopy images. The combination of these techniques will provide unprecedented insight into the size- and morphology dependence of the energetic structure of individual nanocrystals. The results show progress towards routine hyperspectral absorption measurements at the nanoscale.