HL 40: Perovskite and Hybrid Photovolatics I (joint session HL/CPP)

Time: Thursday 9:30–13:00   Location: H36

HL 40.1   Thu 9:30   H36

Effects of Masking on Open-Circuit Voltage and Fill Factor in Solar Cells — •Kristofer Tvingstedt[1], David Kiermasch[1], Lidón Gil-Escrig[2], and Henk J. Bolink[2] — [1]Experimentelle Physik VI Julius Maximilians Universität Wûrzburg — [2]Instituto de Ciencia Molecular, Universidad de Valencia, C/Catedrático J. Beltrán 2, 46980 Paterna, Spain

Guidelines for the correct measurement protocol of novel photovoltaic technologies such as perovskites are now becoming more frequent in literature. This because, as will be confirmed in this talk, it is not straightforward to correctly measure the true efficiency parameters of these and many other novel solar cells. This is particularly the case for small area research devices which are prone to overestimate the short circuit current density, due to edge effects of various types. To reduce the inaccuracy of current density determination, the common recommended practice is to utilize masks with well-defined apertures, often smaller than the device active area. Herein we show both experimentally and theoretically that this common practice, however, leads to erroneous determination of both open-circuit voltage and fill factor, which are figures of merit of equal importance to the short-circuit current density. Although the errors induced in voltage and fill factor by using a mask are generally smaller than what the errors in current can amount to when not using a mask, they are on the other hand omnipresent and can be quite well described.

HL 40.2   Thu 9:45   H36

Enhanced stability and optical properties of perovskite nanocrystals encapsulated in block copolymer micelles — •Hyowon Jeong, Carola Lampe, Moritz Gramlich, and Alexander S. Urban — Nanospectroscopy Group, Department of Physics, Ludwig-Maximilians-Universität München (LMU), Amalienstraße 54, 80799, Munich, Germany

Recently, lead halide perovskites in the form of colloidal nanocrystals (NCs) have emerged as promising candidates for use in light-emitting and photovoltaic devices. High photoluminescence quantum yields as well as size- and consequently band gap- tuning are enabled by a facile synthesis and the natural defect tolerance of the material. However, to exploit these fascinating properties, long-term stability of the NCs under different conditions is necessary. Here, a novel synthesis is introduced, where methylammonium lead iodide (MAPbI3) NCs are grown inside of micelles formed by block copolymers, which protect the NCs from the environment and greatly enhance their lifetime. In addition, the optoelectronic properties of the encapsulated NCs will be discussed based on single NC spectroscopy results.

HL 40.3   Thu 10:00   H36

Energy transfer in films of thickness-tunable CsPbBr3 nanoplatelets — •Andreas Singldinger, Moritz Gramlich, Carola Lampe, and Alexander S. Urban — Nanospectroscopy Group, Department of Physics, Ludwig-Maximilians-Universität München (LMU), Amalienstrasse 54, 80799 Munich, Germany

In recent years, lead halide perovskites have rapidly attracted attention not only in their bulk but also in their nanocrystal form. Various synthetic routes yield highly luminescent nanocrystals of different shapes and sizes and consequently enable band gap tuning through quantum confinement effects. A novel synthesis now allows the growth of quantum-confined CsPbBr3 nanoplatelets (NPLs) with an adjustable thickness ranging from two to six unit cells. These NPLs show strong blue emission induced by a fast radiative decay of excitons. For efficient electroluminescence, however, insight into energy and charge transfer between these NPLs is crucial. Here, we look at two energy transfer processes in perovskite nanocrystal films. Firstly we study energy transfer between NPLs of different bandgaps, via time-resolved photoluminescence-spectroscopy. Secondly we investigate diffusion of photo-excited excitons in films comprising NPLs of a single thickness.

HL 40.4   Thu 10:15   H36

Ultrafast phonon dynamics in lead halide perovskite — •Hong-Guang Duan[1,2,3], Vandana Tiwari[1], Ajay Jha[1], Pabitra Nayak[4], Michael Thorwart[2,3], Henry Snaith[4], and R. J. Dwayne Miller[1,3,5] — [1]MPSD, Hamburg, Germany — [2]I. Institut für Theoretische Physik, UH, Germany — [3]CUI, Hamburg, Germany

— [4]University of Oxford, UK — [5]University of Toronto, Canada

Hybrid organic-inorganic perovskites has 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 two-dimensional spectroscopy in thin films at room temperature. We distinctly observe the ground and excited state vibrational modes corresponding to organic and inorganic sub-lattices. The interplay of strongly coupled dominant vibrational modes to ultrafast carrier generation process will be discussed. Our experiments also unravel the role of organic cations in the ultrafast dynamics after photoexcitation.

HL 40.5   Thu 10:30   H36

Effect of the organic cation in halide perovskites on vibrations in the far-infrared region: a combined theoretical-experimental study — •Christian Gehrmann[1], Michael Sendner[2,3], Sebastian Beck[2,3,4], Robert Lovrincic[2,4], and David A. Egger[1] — [1]Institute of Theoretical Physics, University of Regensburg, 93040 Regensburg, Germany — [2]InnovationLab, 69115 Heidelberg, Germany — [3]Kirchhoff Institute for Physics, Heidelberg University, 69120 Heidelberg, Germany — [4]Institute for High Frequency Technology, TU Braunschweig, 38106 Braunschweig, Germany

Halide perovskites (HaPs) are intriguing optoelectronic materials. In particular, hybrid organic-inorganic HaPs have attracted much interest as possible solar-cell materials. Since phonons might be the dominant source of scattering for charge carriers at room temperature, special efforts should be made to understand lattice dynamics in HaPs. While the internal vibrations of the organic cation are mainly in the mid-infrared region, we study the impact of the cation on vibrations in the far-infrared region by comparing the hybrid MAPbBr3 to the all-inorganic CsPbBr3. To this end, we present lattice dynamics calculations, based on density functional theory. The theoretical results are compared to experimental data obtained from far-infrared spectroscopy. Using reflectance measurements, we can even present experimental results related to the LO phonons which are not infrared active, but inherently contained in our calculations.

HL 40.6   Thu 10:45   H36

Density-functional-theory modeling of point defects in halide-perovskite alloys — •Li Jingrui and Patrick Rinke — Department of Applied Physics, Aalto University, Finland

Perovskite solar cells (PSCs) are a promising emergent technology, because their photo-conversion-efficiency has been increasing rapidly in recent years. In these cells, the photoabsorbing material is a hybrid (organic-inorganic) halide perovskites (ABX$_3$), that is usually grown with low-temperature solution-based synthesis. This synthesis method introduces many point defects, that may critically affect the electronic and structural properties of the PSCs. To gain microscopic insight, we used density-functional-theory to study a series of point defects of halide-perovskite alloys with ion-mixing at A (methylammonium, Cs and Rb), B (Pb and Sn) and X (I, Br and Cl) sites. The considered defects include A- and X-site vacancies and native interstitials in different charge states, as well as K and H impurities. For each model system, a $4 \times 4 \times 4$ perovskite supercell was relaxed using the PBEsol exchange-correlation functional. Then the electronic structure and defect formation energy were calculated using the hybrid HSE06 functional. Our results indicate that the defect formation energy sensitively depends on the phase and composition of halide perovskites. For halogen vacancies, the +1 state that has very small impact on the electronic structure is the most stable in a large Fermi-energy range. Only within the strongly n-type doped region, neutral or negatively-charged halogen vacancies will be formed, which introduce states in the band gap and significantly limit the PSC efficiency.

15 min. break

HL 40.7   Thu 11:15   H36

Calculating structural properties of halide perovskites: Influence of the computational approach — •Hubert Beck, Christian Gehrmann, and David A. Egger — Institute of Theoretical Physics, University of Regensburg, 93040 Regensburg, Germany

Hybrid organic-inorganic halide perovskites (HaPs) have shown

promising results in the development of new optoelectronic devices. Density functional theory (DFT) based first-principles calculations can address several of the open questions for these systems. For many of those calculations it is of vital importance to understand, which of the various microscopic effects in different DFT-related approximations play an important role. Here, we present an investigation of the importance of various theoretical aspects in the DFT calculations of the structural properties for the prototypical case of $MAPbI_3$. The main focus is on a comparison of calculations varying the DFT functional, the account of dispersion forces as well as the inclusion of spin-orbit coupling. The relative effect for the calculations of structural properties of $MAPbI_3$ is evaluated by comparing our computed unit-cell volumes and bulk moduli to results of experiments. Finally, we also present results on the impact of temperature-induced structural fluctuations on calculating the structural properties of $MAPbI_3$.

HL 40.8   Thu 11:30   H36

Identification of trap states by photo-induced transient spectroscopy in metal halide perovskites — •Mathias Fischer[1], Andreas Baumann[2], and Vladimir Dyakonov[1] — [1]Experimental Physics VI, Julius Maximilian University of Würzburg, 97074 Würzburg — [2]Bavarian Center for Applied Energy Research (ZAE Bayern), 97074 Würzburg

Defects in perovskite solar cells ar often related to a reduced device performance. A common method for the characterization of defects in conventional semiconductors is the deep level transient spectroscopy (DLTS). This technique is based on a modulation of the depletion layer width by injecting charge carriers, mostly by applying voltage pulses. Due to electrical field screening in the device, redistribution of mobile ions and relatively low conductivity of organic transport layers, particularly when studying the device at low temperatures, charge carrier injection via voltage pulses is often difficult. This limitation can be mostly overcome by charge carrier injection charges via optical pulses, which enables us to record a rich trap spectrum over the broad temperature range down to 30 K. We apply this technique, which is also known as photo induced-transient spectroscopy (PITS), to perovskites of various compositions and compare the results with complementary techniques, such as thermal admittance spectroscopy and TSC. We show that multiple additional trap states can be observed when optical pulses instead of voltage pulses are used for injection of charges into the device.

HL 40.9   Thu 11:45   H36

Identifying recombination dynamics in efficient perovskite solar cells with transient optoelectrical techniques via active layer thickness alteration — •David Kiermasch[1], Lidón Gil-Escrig[2,3], Andreas Baumann[4], Henk J. Bolink[2], Kristofer Tvingstedt[1], and Vladimir Dyakonov[1] — [1]Experimental Physics VI, Julius Maximilian University of Würzburg, 97074 Würzburg — [2]Instituto de Ciencia Molecular, Universidad de Valencia, 46980, Paterna, Spain — [3]Helmholtz-Zentrum Berlin für Materialien und Energie, 12489 Berlin — [4]Bavarian Center for Applied Energy Research (ZAE Bayern) Bayern, 97074 Würzburg

Quantifying charge carrier recombination processes leading to energy losses in perovskite solar cells is of crucial importance for further improvements. Usually time-resolved photoluminescence (PL) is used to study charge carrier recombination, since electrical techniques are often limited by large capacitance values due to thin film layers and mostly short charge carrier lifetimes. By optimizing both the active layer thickness and the surrounding transport layers, we identified bulk dynamics using the combination of Transient Photovoltage (TPV) and Charge Extraction (CE) in $MAPbI_3$ solar cells with V_{oc}'s above 1.1 V and efficiencies up to 18 %. This allows us to assign loss processes to be mainly of Shockley-Read-Hall and free-carrier recombination type in line with PL studies on pure films. Our work also shows that increasing the perovskite thickness, advantageous to achieve high photocurrents, does not affect the recombination dynamics significantly confirming the high quality of the vacuum processed solar cells studied herein.

HL 40.10   Thu 12:00   H36

Describing the Optoelectronic Properties of Halide Perovskites with a Tight Binding Approach — •Maximilian J. Schilcher[1], Matthew Z. Mayers[2], Liang Z. Tan[3], David R. Reichman[2], and David A. Egger[1] — [1]Institute of Theoretical Physics, University of Regensburg, 93040 Regensburg, Germany — [2]Department of Chemistry, Columbia University, New York, NY 10027, USA — [3]Molecular Foundry, Lawrence Berkeley National Laboratory, Berkeley, CA 94720, USA

The intriguing optoelectronic properties of halide perovskites (HaPs) have invoked the development of new solar cells with enormous power conversion efficiencies. However, it is still not understood how these optoelectronic properties can emerge despite the remarkably soft lattice of HaPs. Theoretical calculations reveal important structure-function relations, yet are often limited by computational costs. We aim for developing theoretical tools that enable the simulation of HaPs with low computational costs, since for the calculation of realistic properties large super cells are often required. Their simulation via more conventional theoretical tools, such as density functional theory (DFT), can be challenging or even impossible. An efficient approach to model large-scale system sizes is the tight binding (TB) formalism. Here, we develop a TB parametrization for the HaP methylammonium lead bromide by projecting the Bloch wave functions on the basis functions corresponding to specific orbital sites of the atoms. This allows us to examine the impact of ionic composition and the effect of dynamical structural distortions on the optoelectronic properties of HaPs.

HL 40.11   Thu 12:15   H36

A completely solvent free route for hybrid perovskite film processing based on pressure treatment of perovskite powders - decoupling material synthesis and film formation — Nico Leupold[2], Maximilian Schulz[1], Konstantin Schötz[1], Ralf Moos[2], and •Fabian Panzer[1] — [1]Soft Matter Optoelectronics — [2]Department of Functional Materials, all University of Bayreuth, Bayreuth, 95440, Germany

Even though hybrid perovskites have undergone a remarkable development within the last years, state of the art processing approaches such as solution processing or evaporation suffer from an intrinsically high complexity, as the perovskite crystallization and its film processing happen simultaneously and are inextricably interconnected. Here we present an alternative, entirely dry processing approach, decoupling perovskite crystallization and film formation, by using readily prepared perovskite powders and produce films by appropriate mechanical pressure treatment. We show how a mechanochemical synthesis by ball milling allows to produce a wide range of phase pure and highly stable perovskite powders with a high flexibility in processing and we address the impact of milling parameters on the powder properties. Using these powders, we demonstrate how the used pressure and the powder microstructure, i.e. particle size and stoichiometry affect the mechanical stability, compactness and surface roughness of the pressed layers. We further address how specific temperature treatment during the pressing step can improve the properties of the pressed layer, and show their capability to be used in perovskite based optoelectronic devices.

HL 40.12   Thu 12:30   H36

Ultrafast two-dimensional electronic spectroscopy of CsPbBr3 perovskite crystals — •Xuan Trung Nguyen[1], Daniel Timmer[1], Yevgeny Rakita[2], David Cahen[2], Antonietta De Sio[1] und Chirstoph Lienau[1] — [1]Institut für Physik, Carl von Ossietzky Universität Oldenburg, Germany — [2]Weizmann Institute of Science, Israel

Halide perovskites are attractive as cost-effective materials for developing highly efficient solar cells. Many research discussed about generation of excitons and free charges in halide perovskites under optical excitation. However, the role of excitons and their interplay with free charges for the optoelectronic and transport properties is not yet clear. Here we use two-dimensional electronic spectroscopy (2DES), with 10-fs-time resolution, to study the initial dynamics of optical excitations in single crystals of CsPbBr3. Upon excitation, we observe bleaching of the exciton resonance as diagonal peak and a cross-peak suggesting their interaction with free charges. Interestingly, the exciton peak vanishes within only ~20 fs and an intense elongated cross-peak persists for several ps. These results suggest efficient screening of exciton by free charges and highlight the importance of many-body interactions in the ultrafast dynamics of halide perovskites crystals.

HL 40.13   Thu 12:45   H36

Exploring the stability of halide perovskite alloys by combining density-functional theory and machine learning — •Guo-Xu Zhang[1,2], Lauri Himanen[1], Jingrui Li[1], and Patrick Rinke[1] — [1]Department of Applied Physics, Aalto University, Finland — [2]School of Chemistry and Chemical Engineering, Harbin Institute of Technology, China

Halide perovskites (ABX$_3$) have attracted considerable attention in recent years due to their breakthrough performance as photovoltaic materials in perovskite solar cells (PSCs). We here consider the materials space of perovskites spanned by A = Cs and Rb, B = Sn and Pb, and X = Cl, Br, and I. Since this space is too large to peruse with density-functional theory (DFT) alone, we combine DFT with machine learning. We use the recently proposed many-body tensor representation (MBTR) [1] as structural descriptor. We then train a kernel ridge regression (KRR) model for fast energy prediction with DFT energies for 2×2×2 and 4×4×4 perovskite supercell models of varying composition.

We analyse the effect of MBTR parameters on the KRR learning quality and then use KRR to explore the vast alloy space. We compute the convex-hull of several binary alloy series, for example $Cs_xRb_{1-x}PbI_3$, $CsPb_xSn_{1-x}I_3$, and $Cs_xRb_{1-x}Pb_ySn_{1-y}Cl_{z_1}Br_{z_2}I_{3-z_1-z_2}$. Our prediction accuracy for the cohesive energy of different alloys is as low as few meV/atom. This suggests that KRR in combination with the MBTR can be used to speed up the discovery of stable halide perovskite alloys.

[1] Huo and Rupp, arXiv 1704.06439.