

CPP 78: Hybrid and Perovskite Photovoltaics II

Time: Friday 9:30–12:45

Location: C 264

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Trap State Dynamics and Phase Transition in MAPbI₃ — TOBIAS SEEWALD, CAROLA EBENHOCH, SUSANNE BIRKHOLO, and •LUKAS SCHMIDT-MENDE — University of Konstanz

Perovskite semiconductors are a new class of semiconductors, significantly different from organic and also inorganic semiconductors. For efficient charge carrier generation, it is essential to minimize non-radiative recombination that can be initiated by energetic trap states. For this purpose, we have studied polycrystalline methylammonium lead iodide perovskite films with varying crystal sizes. Morphology and crystallinity have been modified by means of methylamine gas induced defect healing. Time-resolved photoluminescence as well as pump-probe spectroscopy have been applied in order to relate trap state distribution with crystal sizes and grain boundaries. In this study we can demonstrate the importance of large crystal grains, which allow high carrier mobilities and reduced non-radiative losses that are crucial for efficient solar cell devices. Furthermore, we have investigated the mechanism of the orthorhombic to tetragonal phase transition in MAPbI₃ and the resulting changes in material properties. We present experimental evidence for a transient intermediate phase in MAPbI₃ that occurs prior to the phase transition into the tetragonal phase, which might be induced by an increasing liberation/polarization of the organic cation within the orthorhombic phase at low temperatures. Our results demonstrate that this phase transition has strong impact on solar cell performance parameters.

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Characterization and phase identification in cesium lead bromide thin films by correlative scanning electron microscopy — •SEBASTIÁN CAICEDO-DÁVILA¹, HANNAH FUNK¹, CHRISTIAN MÜLLER², MICHAEL SENDNER³, ROBERT LOVRINCIC², and DANIEL ABOU-RAS¹ — ¹Helmholtz Zentrum Berlin for Materials and Energy, Berlin, Germany — ²Institute for High-Frequency Technology, InnovationLab, TU Braunschweig, Heidelberg, Germany — ³Kirchhoff Institute of Physics, Heidelberg University, Heidelberg, Germany

Designing high-efficiency solar cells based on wide-gap, halide-perovskite absorbers is challenging, and understanding what limits the performance of such devices is still an open issue in the field. One of the wide-gap materials of interest is cesium lead bromide. If deposited as thin film, various Cs-Pb-Br phases may be present. Identification of these phases, their distribution at a microscopic level, as well as the correlation between microscopic, optoelectronic properties with macroscopic materials characteristics are fundamental tasks in research and development. In the present contribution, we report on local compositions and optoelectronic properties of Cs-Pb-Br thin films synthesized by spin-coating and coevaporation, which were analyzed by using SEM imaging, energy-dispersive X-ray spectrometry, cathodoluminescence, XRD, and photoluminescence spectroscopy. We discuss the different phase distribution and its dependence on synthesis method, and microscopic structuring of the film, the challenges concerning specimen preparation for more advanced SEM techniques, and the issue of damaging the sample by the electron beam.

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Ingression of Moisture into Hybrid Perovskite Thin Films Probed with In-Situ GISANS — •JOHANNES SCHLIFF¹, LORENZ BIESSMANN¹, LUKAS OESINGHAUS², EZZELDIN METWALLI¹, LIONEL PORCAR³, and PETER MÜLLER-BUSCHBAUM¹ — ¹TU München, Physik-Department, LS Funktionelle Materialien, 85748 Garching — ²TU München, Physik-Department and ZNN, LS Physics of Synthetic Biological Systems, 85748 Garching — ³Institut Laue-Langevin, 38042 Grenoble, France

Solution-processed hybrid perovskites are of great interest for photovoltaics. However, polycrystalline perovskite thin films show strong degradation in humid atmosphere which poses an important challenge for large-scale market introduction. With in-situ grazing incidence neutron scattering (GISANS) we analyze water content, degradation products and morphological changes during prolonged exposure to several humidity levels. At the highest humidity levels, formation of metastable hydrate phases is accompanied by domain swelling and transforms the faceted crystals to a pebble-like form. The films incorporate much more water than can be integrated in the hydrates.

In low ambient humidity, around 10 vol% of water is adsorbed on the crystals without formation of crystalline degradation products. Thus, although production in ambient atmosphere is of interest for industrial production it might lead to long-term degradation without appropriate countermeasures.

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Exciton-dominated core-level absorption spectra of hybrid organic-inorganic lead halide perovskites — •CHRISTIAN VORWERK^{1,2}, CLAUDIA HARTMANN³, CATERINA COCCHI^{1,2}, GOLNAZ SADOUGHI⁴, SEVERIN HABISREUTINGER⁴, ROBERTO FÉLIX³, REGAN WILKS^{3,5}, HENRY SNAITH⁴, MARCUS BÄR^{3,5}, and CLAUDIA DRAXL^{1,2} — ¹Institut für Physik and IRIS Adlershof, Humboldt-Universität zu Berlin, 12489 Berlin, Germany — ²European Theoretical Spectroscopy Facility — ³Renewable Energy, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 14109 Berlin, Germany — ⁴Clarendon Laboratory, Department of Physics, University of Oxford, Oxford OX1 3PU, United Kingdom — ⁵Energy Materials In-Situ Laboratory Berlin (EMIL), Helmholtz-Zentrum Berlin für Materialien und Energie GmbH, 12489 Berlin, Germany

In a combined theoretical and experimental work, we investigate x-ray absorption near-edge spectra (XANES) from the iodine *L*₃ and the lead *M*₅ edge of the methyl-ammonium lead iodide (MAPbI₃) hybrid inorganic-organic perovskite and its binary phase PbI₂. We find that the absorption onsets are dominated by bound excitons with sizable binding energies of a few hundred meV and pronounced anisotropy. The spectra of both materials exhibit remarkable similarities, suggesting that the fingerprints of core excitations in MAPbI₃ are essentially given by its inorganic component, with negligible influence from the organic groups. Our theoretical analysis allows for an insightful characterization of experimental observations in this complex material.

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Assessing density functionals using many body theory for hybrid perovskites — •MENNO BOKDAM, JONATHAN LAHNSTEINER, BENJAMIN RAMBERGER, TOBIAS SCHÄFER, and GEORG KRESSE — University of Vienna, Faculty of Physics and Center for Computational Materials Sciences, Vienna, Austria

Which density functional is the "best" for structure simulations of a particular material? A concise, first principles, approach to answer this question is presented. The random phase approximation (RPA)—an accurate many body theory—is used to evaluate various density functionals.[1] To demonstrate and verify the method, we apply it to the hybrid perovskite MAPbI₃, a promising new solar cell material. The evaluation is done by first creating finite temperature ensembles for small supercells using RPA molecular dynamics, and then evaluating the variance between the RPA and various approximate density functionals for these ensembles. We find that, contrary to recent suggestions, van der Waals functionals do not improve the description of the material, whereas hybrid functionals and the SCAN (strongly constrained appropriately normed) density functional yield very good agreement with the RPA. Finally, our study shows that in the room temperature tetragonal phase of MAPbI₃, the molecules are preferentially parallel to the shorter lattice vectors but reorientation on ps time scales is still possible.

[1] Bokdam et. al., Phys. Rev. Lett. 119, 145501 (2017)

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The Finite Temperature Structure of the MAPbI₃ Perovskite: Comparing Exchange-Correlation Potentials, Force Fields and Model Hamiltonians — •JONATHAN LAHNSTEINER¹, GEORG KRESSE¹, JURN HEINEN², and MENNO BOKDAM¹ — ¹University of Vienna, Faculty of Physics and Center of Computational Materials Science, Vienna Austria — ²Van 't Hoff Institute for Molecular Sciences, University of Amsterdam

Determining the finite temperature structure of the hybrid perovskite MAPbI₃ at high accuracy is a challenge for both experimental and theoretical methods. A very powerful computational method is *ab-initio* molecular dynamics. It comes with the necessity of choosing a density functional approximation which directly influences the atomic structure. The complexity of the system has led to the development of more approximate methods, such as classical force fields and model

hamiltonians. Here we compare the different computational methods by analysis of the MA molecular ordering patterns in one consistent manner. For those methods that also describe the PbI_3 framework its symmetry is analyzed. Available experimental data in literature is discussed and compared to the outcomes of the different methods. The spread in these data is still too large to uniquely determine a method that 'best' describes the perovskite. However, the comparison on equal footing between the computational methods does allow to evaluate the applicability of the different approximations.

15 min. break

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Role of Förster Resonance Energy Transfer for Efficient Charge Generation at Hybrid Heterointerfaces — ●STEFAN SCHUPP, PHILIPP EHRENREICH, and LUKAS SCHMIDT-MENDE — Department of Physics, University of Konstanz, Universitätsstraße 10, 78457 Konstanz, Germany

During the last two decades, tremendous progress was made in the development of efficient dye-sensitized solar cells with power conversion efficiencies of more than 10%. In this context, performance improvements are commonly related with chemical engineering, i.e. the design of new chemical compounds with optimized absorption characteristics. So far, transparent hole transporting layers are used although polymers offer the conceptual advantage to act not only as hole transporting materials but also as an additional photoactive layer. In a recent study, we have demonstrated that Förster resonance energy transfer (FRET) is a promising strategy to enhance photocurrent contributions from the polymer, while loss channels have stayed unclear. In this contribution, we will generalize our findings and offer new design rules, which help to engineer highly efficient hybrid polymer-metal oxide interfaces. By a comparison of eight different dye-polymer combinations, we tune the FRET efficiency and energy landscape at the interface systematically. This combined analysis helps to understand fundamental loss channels of hybrid heterostructures and offers strategies to bring hybrid solar cells on a competitive level.

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LASER PATTERNING OF PEROVSKITE SOLAR CELLS FOR MINI-MODULE FABRICATION — ●CHRISTOF SCHULTZ¹, FELIX SCHNEIDER^{1,2}, MARKO JOST³, LUKAS KEGELMANN³, ANDREAS BARTELT¹, BERND RECH⁴, RUTGER SCHLATMANN^{1,2}, STEVE ALBRECHT³, and BERT STEGEMANN¹ — ¹HTW Berlin - University of Applied Sciences, Wilhelminenhofstr. 75A, D-12459 Berlin — ²Helmholtz-Zentrum Berlin für Materialien und Energie, PVcomB, Schwarzschildstr. 3, D-12489 Berlin — ³Helmholtz-Zentrum Berlin, Nachwuchsgruppe Perovskit Tandem-solarzellen, Kekuléstraße 5, D-12489 Berlin — ⁴Helmholtz-Zentrum Berlin, Institut für Silizium-Photovoltaik, Kekuléstraße 5, 12489 Berlin

Metal halide perovskite (MHP) solar cells have been rapidly improved over the last years. Successful up-scaling from lab-scales to large module sizes requires monolithical interconnection processes. Here we report on laser scribing processes of the P1, P2 and P3 patterning steps to interconnect perovskite solar cells for module fabrication. Particular emphasis is put on the P2 laser patterning step as it is known to induce detrimental material modifications of the absorber layer mostly at the bottom and in the vicinity of the scribed lines. By systematic variation of the pulse length (ns, ps), the wavelength (1064 / 532 / 355 nm) and the processing side (glass-/ layer-side) suitable process windows were established and optimized. Based on systematic analysis of the laser-matter interaction the selective layer removal, the proper morphology and the electrical functionality were elucidated allowing successful fabrication of a mini-module.

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In situ study of degradation in P3HT-titania based solid-state dye-sensitized solar cells — ●LIN SONG¹, WEIJIA WANG¹, STEPHAN PRÖLLER², DANIEL MOSEGUÍ GONZÁLEZ¹, JOHANNES SCHLIPP¹, CHRISTOPH J. SCHAFFER¹, KRISTINA PETERS³, EVA M. HERZIG², SIGRID BERNSTORFF⁴, THOMAS BEIN³, DINA FATTAKHOVA-ROHLFING³, and PETER MÜLLER-BUSCHBAUM¹ — ¹TU München, Physik-Department, LS Funktionelle Materialien, 85748 Garching — ²TU München, MSE, Herzig Group, 85748 Garching — ³LMU München, Department of Chemistry and CENS, 81377 München — ⁴Elettra-Sincrotrone Trieste 34149 Trieste, Italy

Solid state dye sensitized solar cells (ssDSSCs) have received great at-

tention over last decades due to low costs, ease of production and high efficiencies. However, the stability of these devices is still not verified. To address this issue, the degradation of ssDSSCs is studied in the present work. To make a comparison, mesoporous titania films with different pore size are used as photoanodes. The impact of pore size on temporal evolution of the P3HT crystallites under continuous solar illumination is compared using in situ grazing-incidence wide-angle x-ray scattering (GIWAXS) measurements, and the development of the photovoltaic characteristics is probed in parallel [1]. The findings of the in-situ study show that pore size plays an important role in the device performance and stability.

[1] ACS Energy Lett.2, 991-997 (2017)

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Increased Photovoltage by Use of Metal Complexes as Redox Couples in Dye-Sensitized Solar Cells based on Low-Temperature Electrodeposited ZnO — ●RAFFAEL RUESS, THI HAI QUYEN NGUYEN, SEBASTIAN HAAS, ANDREAS RINGLEB, and DERCK SCHLETTWEIN — IAP, JLU Giessen, Germany

Dye-sensitized solar cells (DSSCs) have received continued interest because of new record efficiencies (up to 14.3 %) and low energy pay-back times. This work is focused on DSSCs based on electrodeposited, porous ZnO which allows low preparation temperatures to further reduce energy consumption of cell production and to use various kinds of substrates such as flexible foils, threads, wires or fibers. One of the most promising strategies to reduce voltage losses in the cells consists in replacement of the I^-/I_3^- redox electrolyte by metal complex redox couples that show fast electron transfer kinetics at low driving force. Detailed photoelectrochemical studies are used to investigate charge transfer and transport in complete photovoltaic devices. Recombination at the ZnO/electrolyte interface as well as mass transport in the electrolyte have been identified as performance-limiting factors. Control of these factors was achieved by manipulating the electrolyte composition leading to promising power conversion efficiencies of 3.56 % or 3.85 % with cobalt-based or copper-based redox couples. This work shows that such metal complex redox couples can also be applied in cells based on ZnO and opens the door for further improvements in device efficiency of this type of DSSCs.

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macro- and nano-scale morphology evolution during in situ spray coating of titania films for perovskite solar cells — ●BO SU¹, HERBERT A. CALLER-GUZMAN¹, VOLKER KÖRSTGENS¹, YICHUAN RUI², YUAN YAO¹, NITIN SAXENA¹, GONZALO SANTORO³, STEPHAN V. ROTH³, and PETER MÜLLER-BUSCHBAUM¹ — ¹Technische Universität München, Physik-Department, Lehrstuhl für Funktionelle Materialien, 85748 Garching — ²College of Chemistry and Chemical Engineering, Shanghai University of Engineering Science, Shanghai 201620, China — ³DESY, 22607 Hamburg

To scale-up deposition processes, the use of spray deposition is advantageous, because it is easy to be controlled and has no limitation concerning substrate geometry. In this study, the structure evolution of mesoporous titania films, with a controllable pore size, is characterized by in situ grazing incidence small angle X-ray scattering (GISAXS) during the spray process. For the applied spray conditions, we observe micelle deposition, evaporation of the solvent and transformation of the morphology towards equilibrium. All processes influence the structure features on the nano-scale. On the macro-scale, the kinetics and thermodynamics of the spray depositions processes are simulated. We find a good agreement with the large-scale morphology of the spray-coated films obtained in the experiments. Hybrid perovskite (methylammonium lead iodide) solar cells based on the sprayed titania film are fabricated as a showcase of the developed spray-deposition for solar cell applications.

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Investigation of mobility controlled amphiphilic diblock copolymer templating for hybrid photovoltaic applications — ●NURI HOHN¹, STEFFEN SCHLOSSER¹, LORENZ BIESSMANN¹, SEBASTIAN GROTT¹, SENLIN XIA¹, KUN WANG¹, MATTHIAS SCHWARTZKOPF², STEPHAN ROTH³, and PETER MÜLLER-BUSCHBAUM¹ — ¹TU München, Physik-Department, LS Funktionelle Materialien, 85748 Garching — ²DESY, 22607 Hamburg — ³KTH, Department of Fibre and Polymer Technology, SE-100 44 Stockholm, Sweden

Current research in the field of hybrid photovoltaics aims to develop devices, which benefit from the advantages of both their organic and

inorganic nature. While the organic part can contribute flexibility, potential low cost and possible large scale production, the inorganic counterpart offers chemical and physical stability. Nevertheless, one major issue in device fabrication still consists of the backfilling process. Therefore, tuning of the pore size of the inorganic thin films is of importance. Based on an amphiphilic diblock copolymer templating

with polystyrene-*b*-polyethylene oxide and tuning of the dryness state of the casting solution different morphologies are achieved after blade coating. The introduced method is expected to facilitate the optimization process of high surface-to-volume ratio against sufficiently large pores for efficient backfilling. Surface and bulk analysis is carried out via SEM and grazing incidence small angle X-ray scattering.