HL 55: Perovskite and Photovoltaics III (joint session HL/KFM)

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

HL 55.1 Fri 9:30 EW 203

Data-efficient machine learning for perovksite alloys — •Jarno Laakso, Henrietta Homm, and Patrick Rinke — Department of Applied Physics, Aalto University, Espoo, Finland

Perovskite solar cells are highly efficient, but their commercialization has been hindered by toxicity and lack of stability. Compositional engineering can mitigate these problems [1], but the complexity of the perovskite materials space makes the search for an optimal solar cell material challenging. We previously demonstrated how machine learning (ML) can accelerate property predictions for the perovskite alloy $CsPb(Cl/Br)_3$ [2]. However, the extensive density functional theory (DFT) calculations required for model training prevent applications to more complex materials. Here, we facilitate model training with a data-efficient scheme, validated on $CsPb(Cl/Br)_3$ data and extended to the ternary alloy $CsSn(Cl/Br/I)_3$.

Our approach employs clustering to build a compact but diverse initial data set of atomic structures. We then apply a two-stage active learning approach to first improve the robustness of the ML-based structure relaxations and then fine-tune the accuracy near equilibrium structures. Tests for $CsPb(Cl/Br)_3$ reveal that our scheme reduces the number of required DFT calculations during model training by up to 50%. The fitted model for $CsSn(Cl/Br/I)_3$ is robust, with all ML-based structure relaxations converging in our tests. The relaxations are also highly accurate, having an average error of 0.5 meV/atom.

[1] *iScience* **23**, 101359 (2020). [2] *Phys. Rev. Mater.* **6**, 113801 (2022).

HL 55.2 Fri 9:45 EW 203

Passivation and Charge Carrier Selectivity Optimization for Narrow Bandgap Perovskite Solar Cells — •Athanasia Vicha, Georgios Loukeris, Markus Kohlstädt, Uli Würfel, and Andreas Bett — Fraunhofer ISE, Freiburg im Breisgau, Germany

This study aims to enhance narrow bandgap perovskite solar cells for all-perovskite tandem applications. Initial efforts focused on bulk passivation of the perovskite layer. The combination of two additives, Methylammonium chloride and Lead thiocyanate, increased Voc up to 0.82V and Jsc to 31mA/cm^2. Glycine hydrochloride improved result reproducibility leading to a favourable FF, and increasing Thiourea concentration simultaneously raised Jsc. Further optimizations and the effects of additive combinations will be presented. Concurrently, this study investigates the selectivity of the hole transport layer/perovskite interface. Thus, we examined substituting an aqueous PEDOT:PSS formulation for a solvent-based one to address water-induced degradations in the underlying perovskite layer in all-perovskite tandem devices. Preliminary assessments on single-junction narrow bandgap solar cells using solvent-based PEDOT yielded a notable 16% Voc increase compared to aqueous PEDOT:PSS. However, this rise in Voc results in a significant 23% Jsc reduction. In response, we explored strategies to boost Jsc by varying the PEDOT thickness. In thinner layers, Jsc exceeds the one of PEDOT:PSS-based cells, aligning with simulations. This research outcome provides valuable insights into optimizing passivation and enhancing selectivity in narrow bandgap perovskite solar cells.

HL 55.3 Fri 10:00 EW 203

Understanding and mitigating atomic oxygen-induced degradation of Perovskite solar cells for near-earth space applications — •Biruk Seid — Institute of Physics and Astronomy, University of Potsdam, D-14476 Potsdam-Golm, Germany

Atomic oxygen (AtOx) in low-Earth orbit is known to etch, corrode, and form metal oxide on the metal contact with PSC devices. To mitigate this issue, we report the applicability of thermally evaporated 0.7*m silicon oxide (SiOx) encapsulation as an AtOx barrier for triple-cation PSC. Moreover, the AtOx-induced degradation mechanism of phenethylammonium iodide (PEAI)-passivated and non-passivated devices are discussed and analyzed. We found that after a total exposure duration of 120 minutes, the SiOx-encapsulated cells maintained over 97% of their initial power conversion efficiency (PCE), regardless of the device type (passivated or non-passivated). In contrast, in the case of unencapsulated devices, PCEs for non-passivated and PEAI-passivated devices the PCE declined to a maximum value of 43% and 62%, respectively. In non-passivated and unencapsulated devices, AtOx has no impact on the short-circuit current density (JSC) but degrades the fill factor (FF) and open circuit voltage (VOC). In PEAI-passivated devices, the JSC additionally degrades by almost 35%. perovskite was not the origin of PCE degradation. Instead, inefficient charge extraction and mobile ions, due to a swiftly degrading PEAI interlayer are the primary causes of AtOx-induced device performance degradation in passivated devices, whereas a large ionic FF loss limits non-passivated devices.

HL 55.4 Fri 10:15 EW 203

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HL 55.5 Fri 10:30 EW 203

Passivating Inorganic Interlayers at the Perovskite/C_{60} Interface in Monolithic Perovskite Silicon Tandem Solar Cells — •Johanna Modes[1,2], Patricia S.C. Schulze[1], Carl Eric Hartwig[4], Stefan Lange[4], Armin Richter[1], Juliane Borchert[1,3], and Andreas Bett[1,2] — [1]Fraunhofer ISE — [2]Albert-Ludwigs-Universität — [3]INATECH — [4]Fraunhofer CSP

Metal halide perovskites emerged in recent years as promising absorber materials for solar cells that can combine high power conversion efficiency with low production costs. Significant non-radiative charge carrier recombination occurs at the perovskite interface with the electron contact C_{60}, which prevents the full exploitation of the solar cell's potential. C_{60} induces states within the band gap, lowers the quasi-Fermi level splitting, and limits the open circuit voltage (V_{OC}). Ultra-thin passivation layers at the perovskite/C_{60} interface are used to reduce those losses. To enable industrial upscaling, our focus lies on inorganic passivation layers deposited via atomic layer deposition. An AlO_x interlayer was developed that increases the iV_{OC} up to 50 mV and improves the V_{OC} for single-junction and tandem devices. To better understand the effects involved in this passivation, photoluminescence quantum yield (PLQY), angle-resolved X-ray photoelectron spectroscopy (ARXPS) and surface photovoltage (SPV) measurements were carried out. State-of-the-art perovskite solar cells using a LiF_x passivation layer suffer from device degradation over time. Initial stability testing indicates that a thin AlO_x passivation layer can improve device stability and can serve as an alternative to LiF_x.

HL 55.6 Fri 10:45 EW 203

Analyzing defect thermodynamics of $(Ag,Cu)GaSe_2$ solar cell absorbers using a machine-learning interatomic potential — •Vasilios Karanikolas, Delwin Perera, and Karsten Albe — Institut für Materialwissenschaft, Technische Universität Darmstadt, Germany

One of the most widely used absorber materials for thin-film solar cells is $Cu(In,Ga)Se_2$ (CIGS). Currently, CIGS yields the highest efficiencies within this technology and the addition of Ag has been found to further improve the efficiency. The performance of the CIGS absorber, however, is also governed by defects, especially by the type and density of grain boundaries (GBs) [1].

In this work, we investigate the thermodynamic properties of GBs for $(\mathrm{Ag}_{1-x}\mathrm{Cu}_x)\mathrm{GaSe}_2$ structures based on a machine learning interatomic potential (MLIP)[2]. The training dataset for the regression machine learning model is created by density functional theory (DFT) calculations. The MLIP allows us to perform molecular dynamics simulations for structurally complex GBs that are inaccessible by conventional electronic structure methods. In particular, we investigate the thermodynamic properties of symmetric GBs beyond $\Sigma 3$ and include an analysis of silver segregation at the interfaces.

[1] D. Abou-Ras *et al.*, Acta Materialia **118**, 244-252 (2016).
[2] Y. Lysogorskiy *et al.*, npj Computational Materials **7**, 1 (2021).

15 min. break

HL 55.7 Fri 11:15 EW 203

Halide segregation and ionic motion in lead-halide perovskites — •Francisco Peña-Camargo, Felix Lang, and Martin Stolterfoht — Institute of Physics and Astronomy, University of Potsdam, Karl-Liebknecht-Str. 24-25, 14476 Potsdam, Germany

Halide segregation is a photoinduced phenomenon observed in perovskites with multiple halides at the X site of the ABX3 lattice. The evidence shows that the photoluminescence spectrum of the mixed phase red-shifts and increases its intensity in a timescale of minutes. On the other hand, it has been proven that mobile ions are present in cells in densities around 1016 cm-3, which is sufficient to screen the internal electric field, generating, among other effects, the hysteresis in the forward and reverse JV scans. The scan-speed dependence of hysteresis suggests that mobile ions have an effect on the cell performance mostly at a determined condition when the voltage sweep matches the ionic timescales. Here, by measuring the JV characteristic at different speeds with a novel technique called fast hysteresis (FH) from 10-2 to 104 V s-1, we quantify the ionic diffusion coefficient and conclude that the ionic motion happens in the seconds to minutes timescales. Moreover, using bias-assisted charge extraction (BACE) we are able to quantify the ionic volumetric density and confirm the ionic transient timescales. Based on that experimental evidence, it is plausible to link the ionic motion to halide segregation, particularly when it comes to the timescales which appear to match very well. This is also supported by the correlation of the ionic losses with the increasing Br/I ratio in triple-cation lead-based perovskite solar cells.

HL 55.8 Fri 11:30 EW 203

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HL 55.9 Fri 11:45 EW 203

Assessing machine-learning force fields for defect calculations in halide perovskites — •Frederico Delgado[1], Frederico Simões[1], Leeor Kronik[2], and David A. Egger[1] — [1]Physics Department, TUM School of Natural Sciences, Technical University of Munich, Germany — [2]Department of Molecular Chemistry and Materials Science, Weizmann Institute of Science, Israel

The excellent optoelectronic properties exhibited by bulk halide perovskites (HaPs) are important for their photovoltaic performance and have been extensively investigated. Despite this, the ubiquity of both point defects and extended ones like surfaces requires further careful examination of their impact on such properties. Precise investigations of dynamic effects in this context via ab-initio molecular dynamics imply large computational costs, especially when the need for accurate exchange correlation functionals arises. Therefore, in order to sample sufficiently long time scales and sufficiently large supercells, usage of on-the-fly machine learning (ML) forces fields appears to be particularly appealing. In this study, we investigate such defects and their dynamical properties in CsPbBr_3 using ML force fields. Our results can aid rationalizing the correlations between local structural dynamics and the observed optoelectronic behavior.

HL 55.10 Fri 12:00 EW 203

Nanospectroscopy and ultrafast out-of-plane carrier dynamics in metal halide perovskites — •Martin Zizlsperger[1], Svenja Nerreter[1], Qimu Yuan[2], Kilian B. Lohmann[2], Fabian Sandner[1], Felix Schiegl[1], Christian Meineke[1], Yaroslav Gerasimenko[1], Laura M. Herz[2], Thomas Siday[1,2], Markus A. Huber[1], Michael B. Johnston[2], and Rupert Huber[1] — [1]Department of Physics and Regensburg Center for Ultrafast Nanoscopy (RUN), University of Regensburg, 93040 Regensburg, Germany — [2]Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford, OX1 3PU, UK

The high efficiencies of metal halide perovskite solar cells have been associated with effective charge-carrier diffusion. Still, the characteristic nanograins and crystallographic disorder entailed in perovskite films hindered unraveling details of the vertical transport, calling for simultaneous experimental access to chemical composition, structural phase and ultrafast out-of-plane dynamics. Here, we promote depth-sensitive terahertz near-field nanospectroscopy to extreme subcycle time scales. In $\mathrm{FA}_{0.83}\mathrm{Cs}_{0.17}\mathrm{Pb}(\mathrm{I}_{1-\mathrm{x}}\mathrm{Cl}_\mathrm{x})_3$ films, we discern domains of the α-phase from δ-phase and PbI_2 nano-islands. Based on deep-subcycle time shifts of the scattered terahertz waveform after photoexcitation, we access the single-grain vertical diffusion. Despite topographic irregularities, diffusion is surprisingly homogeneous on sub-micrometer length scales, only varying between mesoscopic regions. Our approach linking nano-morphology with carrier dynamics may benefit all future optoelectronic devices using nanocrystalline materials.

HL 55.11 Fri 12:15 EW 203

Comparison of the application of the TRMC technique for the characterization of crystalline silicon and perovskite solar cell absorber material and efficient surface passivation — Arpana Singh[1], Biruk Alebachew Seid[2], Felix Lang[2], Marinus Kunst[3], and •Heinz-Christoph Neitzert[1] — [1]DIIN, Salerno University, 84084 Fisciano, Italy — [2]ROSI Freigeist Group, Institut für Physik und Astronomie, Universität Potsdam, 14476 Potsdam-Golm, Germany — [3]Helmholtz-Zentrum Berlin, 14109 Berlin, Germany

Time Resolved Microwave Conductivity (TRMC) as a contactless technique for the characterization of the charge carrier kinetics in photoconductive materials has been recently successfully applied for the characterization of perovskite thin films [1]. Charge carrier lifetimes depend strongly on excitation conditions, such as light intensity and wavelength. It is shown, how TRMC-measurements performed with excitation at low laser intensities and different wavelengths can give valuable information on the quality of the surface passivation of triple-cation, triple-anion Perovskite films by PEAI layers. A good correlation of the observed lifetime increase in the absorber films and the performance of the completed solar cells has been found. The results are compared to the characterization under identical measurement conditions of crystalline silicon wafers. Also in this case the influence of different passivation schemes on the charge carrier lifetime and on solar cell properties have been compared. [1] A. Marchioro, J. Teuscher, D. Friedrich, M. Kunst, R. van de Krol, T. Moehl, M. Grätzel and J. E. Moser, Nature Photonics 8, 250-256 (2014).

HL 55.12 Fri 12:30 EW 203

The detrimental role of PbI2 at the interface between absorber and electron-transport layer in halide-perovskite solar cells — •Dan Ralf Wargulski, Ke Xu, Steve Albrecht, and Daniel Abou-Ras — Helmholtz-Zentrum Berlin für Materialien und Energie, Berlin, Germany

There is an ongoing discussion in the halide-perovskite community about the beneficial or harmful effects of PbI2 in halide-perovskite solar cells (HPSCs). One school praises PbI2 for its passivating and stabilizing properties, while the other aims at getting rid of excess PbI2, because it leads to decreased cell efficiencies and degraded long-term stability. How can such a contradiction exist? The effect of PbI2 depends on its location in the solar-cell stack and on the specific HPSC architecture used. When applied as top cells in tandem devices, HPSCs do not exhibit a n-i-p structure but an p-i-n structure, often called *inverted* HPSC structure. As a result, the top surface where PbI2 forms is not situated at the interface between the absorber and the hole-transport layer anymore, but at the interface to the electron-transport layer. This modifies considerably the effects of PbI2 on the device performance. The present work discusses the formation of PbI2 in HPSCs with slot-die-coated, triple-halide perovskite absorbers, which was investigated by means of correlative electron microscopy. Studying the changing PbI2 coverage of the halide-perovskite absorbers with varying annealing temperature, a model will be presented that explains the effects of the PbI2 precipitates on the open-circuit voltages and the fill factors of the HPSCs with inverted cell architecture.

HL 55.13 Fri 12:45 EW 203

Stability Enhancement of Mixed-Cation Hybrid Halide Perovskites combining DFT-1/2 with SOC — •Mohammad Moaddeli[1], Mansour Kanani[1,2], and Anna Grunebohm[3] — [1]Department of Materials Science and Engineering, School of Engineering, Shiraz University, Shiraz, Iran — [2]Solar Energy Technology Development Center, Shiraz University, Shiraz, Iran — [3]Interdisciplinary Centre for Advanced Materials Simulation (ICAMS) and Center for Interface-Dominated High Performance Materials (ZGH), Ruhr-University Bochum, Germany

Tunable band gaps, easy processing, composition engineering, and enhanced performance make mixed-cation perovskites promising as light absorbers in third generation solar cells. However, stability challenges remain in understanding the underlying mechanisms. This study emphasizes the importance of accurately predicting structural and electronic properties. While density functional theory (DFT) is the preferred method for addressing these, standard exchange-correlation functionals often fail to reproduce the band structure. Therefore, we propose the DFT-1/2 method, which is applied to single- and mixed-cation systems. We explore how the choice of the A-cation modifies the Pb-I scaffold. We find that the addition of Cs and MA to FAPbI3 reduces the dispersion in the scaffold, which has a high potential to reduce the migration of iodide and thus improves the structural stability. Additionally, we compare Born effective charges with and without the SOC effect and the DFT-1/2 approach [1]. [1] M. Moaddeli et al, Phys. Chem. Chem. Phys. 25, 25511 (2023).