

CPP 13: Molecular Electronics, Hybrid and Perovskite Photovoltaics

Time: Friday 10:00–12:45

Location: H3

Invited Talk

CPP 13.1 Fri 10:00 H3

Electron-lattice relaxation effects in halide perovskites —

•DAVID A. EGGER — Technical University of Munich, Germany

Halide perovskites (HaPs) have shown great promise as materials for applications in energy and optoelectronic devices owing to their fascinating microscopic properties. Of particular scientific interest is the coupling of electronic to lattice-dynamical properties of HaPs, because a comprehensive understanding of it is key to predicting and further improving charge-carrier and optical transport characteristics. In this talk, I will present our recent theoretical findings on electron-lattice relaxation effects in HaPs. Specifically, using molecular dynamics in conjunction with electronic-structure theory, it will be shown that the soft, polar lattice of paradigmatic HaPs leads to a variety of very interesting electron-lattice relaxation phenomena. These include structural anharmonicities, nonlinear electron-phonon couplings and short-range correlated disorder potentials. It will be demonstrated that these mechanisms are key to a microscopic picture of charge-carrier mobilities and optical absorption properties of HaPs.

CPP 13.2 Fri 10:30 H3

In situ GIXS study on the crystallization and mesoscale film formation of lead-free MBI perovskite on mesoporous titania during spray deposition from a green solvent —

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Hybrid halide perovskites are highly promising materials in photovoltaics, due to high efficiencies and industrial favorable processability. However, most of the studied systems are based on water-soluble lead and toxic solvents, which lower the broad application within European standards of hazardous materials. Bismuth-based alternatives like methylammonium bismuth iodide (MBI) can offer a green route to lead-free perovskite solar cells on a large scale when synthesized in the comparably non-toxic solvent methyl acetate and processed by industrially applicable spray deposition. In this work, we investigate with in situ synchrotron GIXS the crystallization and mesoscale film formation of MBI perovskite on mesoporous titania during spray deposition, to understand MBI formation on a typical solar cell interface.

CPP 13.3 Fri 10:45 H3

In situ GIWAXS phase and texture tracking of 2-step slot-die coated perovskite —

•MANUEL ANDREE SCHEEL¹, LENNART KLAUS REB¹, RENJUN GUO¹, MATTHIAS SCHWARTZKOPF², STEPHAN VOLKHER ROTH^{2,3}, and PETER MÜLLER-BUSCHBAUM^{1,4} — ¹Lehrstuhl für Funktionelle Materialien, Physik-Department, Technische Universität München, James-Frank-Str. 1, 85748 Garching, Germany — ²DESY, 22607 Hamburg, Germany — ³KTH, Department of Fibre and Polymer Technology, SE-100 44 Stockholm, Sweden — ⁴Heinz Maier-Leibnitz Zentrum (MLZ), Technische Universität München, Lichtenbergstr. 1, 85748 Garching, Germany

Perovskite slot-die coating is a promising thin-film deposition technique for organic-inorganic perovskite materials and might lead the way towards commercial high efficient solar cells. Roll-to-roll compatible deposition techniques offer the possibility to combine high production throughput with minimal waste, and offer high customizability at the same time. We investigate the conversion of slot-die coated lead iodide and slot-die coated methylammonium iodide to perovskite by in situ grazing-incidence wide-angle X-ray scattering (GIWAXS). Here, we study the thin-film morphology and texture of the resulting thin film during the annealing process. We track the phase evolution and their respective crystal orientations over time. We find, that the precursor-solvent phase influences the final crystal orientation in the thin-film by acting as a precursor source that controls the level of available precursor material. We also investigate deposition-technique depending differences that can influence thin-film characteristics.

15 min. break

CPP 13.4 Fri 11:15 H3

Perovskite and Organic Solar Cells Generate Power in Space —

•LENNART KLAUS REB¹, MICHAEL BÖHMER¹, BENJAMIN PREDESCHLY¹, SEBASTIAN GROTT¹, CHRISTIAN LUDWIG WEINDL¹, GORAN IVKOVIC IVANDEKIC¹, RENJUN GUO¹, CHRISTOPH DREISSIGACKER², JÖRG DRESCHER², ROMAN GERNHÄUSER¹, ANDREAS MEYER², and PETER MÜLLER-BUSCHBAUM^{1,3} — ¹TU München, Garching, DE — ²Deutsches Zentrum für Luft- und Raumfahrt (DLR), Köln, DE — ³Heinz Maier-Leibnitz-Zentrum, Garching, DE

Perovskite and organic solar cells have become a hot research topic in the last few years. The lightweight thin-film solar cells are of particular interest for space applications due to their exceptional power per mass, exceeding their inorganic counterparts by magnitudes.

Here, we present the Organic and Hybrid Solar Cells In Space experiment (OHSCIS) and the launch of perovskite and organic solar cells to space for the first time [1, 2]. The mechanical and electronic design of the experiment aims at maximizing the data collection rate and precise measurements. We show that the solar cells operate in space conditions and produce reasonable power per area of up to 14 and 7 mW cm⁻², respectively. Also, during a phase being turned away from the Sun, the solar cells produce power from collecting faint Sun-light scattered from Earth. Our results highlight the potential for near-Earth applications and deep space missions of these technologies.

[1] L. Reb et al., *Joule* 4,1880-1892 (2020), doi.org/10.1016/j.joule.2020.07.004[2] L. Reb et al., *Rev. Sci. Instrum.* 92 (2021), doi.org/10.1063/5.0047346.

CPP 13.5 Fri 11:30 H3

Optimally-tuned range-separated hybrid functionals for accurate molecular excited-state geometries —

•BERNHARD KRETZ and DAVID A. EGGER — Technical University of Munich, Germany

Molecular excited-state (ES) potential energy surfaces (PES) obtained by computational methods can shed light on reaction mechanisms and pathways in photocatalytic reactions. These ES PES can be calculated using either time-dependent density functional theory (TD-DFT) or high-level wave-function methods. TD-DFT based calculations are computationally very efficient, but often lack the accuracy achieved by computationally more expensive wave-function methods[1]. Recently, the class of optimally-tuned range-separated hybrid (OT-RSH) functionals[2] was developed which promises to reduce the gap in accuracy.

In our recent work[3], by comparison with high accuracy wave-function data from literature, we assessed the accuracy of TD-DFT and OT-RSH for the excited-state geometries for a selection of organic molecules with varying complexity of their ES PES. We mainly focused on the structural parameters of the lowest-excited singlet states. Our results show that OT-RSH maintains the accuracy of conventional functionals for small molecules, while it improves the description of more complex ES PESs involving charge-transfer states.

[1] C. Azarias, *J. Phys. Chem. A*, 121, 32, 6122 (2017)[2] L. Kronik et al., *J. Chem. Theory Comput.*, 8, 5, 1515 (2012)[3] B. Kretz and D. A. Egger, *J. Chem. Theory Comput.*, 17, 1, 357 (2021)**Invited Talk**

CPP 13.6 Fri 11:45 H3

Light-actuated colloidal nano- and microparticles —

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Propelled colloidal particles constitute biomimetic analogues to natural microswimmers and represent ideal agents for responsive and adaptive soft matter. Among the numerous propulsion mechanisms developed recently to self-propel such nano- and microparticles, light is a most promising stimulus, since it enables a natural spatio-temporal control of the motion, especially for large numbers of the particles, leading to swarming-like behaviour or novel active materials with exceptional properties.

We introduce on the one hand a novel class of such particles that are light-actuated based on symmetry-broken refraction, fabricated by

3d laser printing based on two-photon polymerization. By numerical simulations, we will discuss collective, light-actuated effects including unexpected patterns and transport effects.

On the other hand, we demonstrate that light actuation of a large number of colloidal particles in optical feedback leads to self-organized pattern formation including the superposition of symmetric patterns as hexagons, rolls and squares. We discuss the stability of these patterns and their control.

CPP 13.7 Fri 12:15 H3

Laser-Induced Electronic and Vibronic Dynamics in the Pyrene Molecule and its Cation — •KATHERINE R. HERPERGER^{1,2}, JANNIS KRUMLAND², and CATERINA COCCHI^{2,3} — ¹Department of Physics, University of Ottawa, Ottawa, Ontario K1N 6N5, Canada — ²Humboldt-Universität zu Berlin, Physics Department and IRIS Adlershof, 12489 Berlin, Germany — ³Carl von Ossietzky Universität Oldenburg, Institute of Physics, 26129 Oldenburg, Germany

Among polycyclic aromatic hydrocarbons (PAHs), pyrene (C₁₆H₁₀) is widely used as an optical probe thanks to its unique ultraviolet absorption and infrared emission features. In this first-principles study based on real-time time-dependent density-functional theory coupled with the Ehrenfest molecular dynamics scheme, we investigate the sub-picosecond electronic and vibronic response of a pyrene molecule and its cation excited by a coherent ultraviolet Gaussian pulse. The response of both the species is analyzed in terms of electronic population, absorption spectra, and vibrational activity. Combining this

knowledge with the symmetry properties of pyrene, we gain insight into the transient response of this molecule to laser perturbation, setting the stage for future studies on larger and more complex PAHs.

CPP 13.8 Fri 12:30 H3

Charge transfer dynamics across the Au/Ferrocene (aq) interface studied by a two photon pump - photovoltage probe scheme — •MANUEL BRIDGER, ZHIPENG HUANG, YUJIN TONG, OSCAR NARANJO, ALEXANDER TARASEVITCH, RICHARD KRAMER CAMPEN, and UWE BOVENSIEPEN — University Duisburg/Essen

Photo-excitation at interfaces redistributes electrons between bulk phases on microscopic spatial and ultrafast time scales. Here we employ a two photon pump - photovoltage probe scheme to quantify the relationship of particular resonant optical transitions to interfacial charge redistribution for the case of charge transfer between a gold electrode and ferrocene self assembled monolayer in an electrochemical cell. We access vibrational and electronic excited states of ferrocene with femtosecond pulses of center wavelength 3 μm and 400 nm respectively. Two signals are apparent: a large amplitude voltage transient at 120 ± 80 fs and a small at 4.0 ± 2.7 ps. Control experiments and comparison to similar systems probed in vacuum by 2PPE spectroscopy suggest the former is the result of charge transfer via vibronically coupled ferrocene CH vibration and hybrid ferrocene/Au electronic states and the later charge transfer via vibrationally mediated interfacial polarization. These results imply that stabilising such vibronically coupled states is a useful target for devices that rely on efficient charge transfer at solid/liquid interfaces.