2024 Quantsol Winter Workshop

European Society for Quantum Solar Energy Conversion

March 17-21, 2024

Welcome Address

OLD TEXT!

Welcome to the 33rd Workshop on Quantum Solar Energy Conversion

Our workshop continues a long tradition of successful meetings, which were only interrupted during the Covid crisis. What makes the Quantsol workshops so special is a view on solar energy conversion from different aspects and technologies with time and space for fruitful and intense discussions.

This year again, the program covers solar cells, electrochemical cells, and a range of established and emerging materials. I personally look forward to the talks and discussions that touch on the fundamental limitations of solar energy conversion.

A very warm thank you to Ute and Omer for taking so well care of the organization and putting a great program together! And thanks to all of you for contributing your latest results and sharing your thoughts on solar energy conversion.

We can also look forward to the warm hospitality of Rauriser Hof and the great landscape of the Rauris valley. I look forward to a week of inspiring talks, intense discussions, and meeting old and new friends.

Susanne Siebentritt

Contents

Relationship between exciton and charge dynamics in organic blends through nano-morphology

Day, 00:00

Ardalan Armin
Swansea University

Exciton diffusion plays a vital role in determining the power conversion efficiency in organic semiconductor based solar cells through controlling the efficiency of exciton splitting [1]. However, measurements of diffusion length in organic semiconductors requires specialized equipment and expertise [2, 3]. Measurements of exciton splitting efficiencies rely upon quenching experiments prone to erratic errors and large uncertainties. In this presentation I will introduce a quasi-steady state technique to measure exciton diffusion lengths in organic semiconductors, named pulsed-PLQY [4]. Further, I will show how this technique can be utilized in bulk heterojunctions to measure the efficiency of exciton splitting and, also the difficult-to-measure-domain-size. Finally, I will discuss the relationships between nanoscale exciton dynamics and the enhanced charge carrier dynamics seen in state-or-the-art non-fullerene organic solar cells.

The long diffusion lengths measured in non-fullerene acceptor based organic solar cells [3, 4] support large domain sizes while maintaining high exciton splitting efficiencies. These increased domain sizes can lead to large reductions in bimolecular recombination [5, 6], further impacting the efficiency of devices. Lastly, I will discuss the relationship between the enhanced charge carrier dynamics seen in state-of-the art non-fullerene organic solar cells [7] and improved exciton dynamics, enabled by the nano-morphology of the bulk heterojunction.

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YYY, 00:00

Materials Theory of Halide Perovskites: Insights from Atomistic and Molecular Modeling

Tao Shuxia

Materials Simulations and Modelling, Applied Physics, Eindhoven University of Technology, the Netherlands

Metal halide perovskites, known for their unique chemical and physical properties, have attracted considerable attention over the past decade for their potential in optoelectronic applications such as solar cells, LEDs, and photodetectors. Our research approach leverages a sophisticated blend of computational methods, including electronic structure calculations (DFT and tight binding) and reactive molecular dynamics simulations with ReaxFF and machine learning potentials. This comprehensive, multiscale approach enables us to thoroughly investigate the electronic and dynamical properties of halide perovskites, leading to significant insights and advancements.

One of our research focuses is on the analysis of defects in halide perovskites. Understanding and mitigating these defects is crucial for enhancing the efficiency and longevity of optoelectronic devices. Our approach includes the detailed determination of electronic energy levels and thorough examination of the dynamic properties of these materials. By identifying defects responsible for recombination losses and chemical degradation, we develop targeted strategies for their mitigation. These strategies involve engineering perovskite compositions and interfaces, using passivation agents, and optimizing the quality of perovskite films through precise control of synthetic chemistry and processing parameters.

Another key area of our research is exploring chirality in perovskites. Introducing chiral organic ligands into perovskite structures alters their mirror symmetry, leading to unique properties like chiral-induced spin selectivity (CISS) and chiroptical activity, both exhibiting temperature-dependent behaviors. We use first-principles methods and models for calculating circular dichroism (CD) and analyzing electron/spin transport. We study the impact of the dynamic microstructure of chiral perovskites under various temperature conditions, aiming to identify structural features that influence the optoelectronic responses. These insights provide valuable information for the design of new chiral perovskites with potential applications in novel optoelectronics, such as spin LEDs and chiral photodetectors.

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XXX, 00:00

Additional lighting effects for photovoltaic improvements in the performance of solar cells

Ivana Validzic

Department of Atomic Physics, Vinča Institute of Nuclear Sciences, National Institute of the Republic of Serbia, P.O. Box 522, 11001 Belgrade, University of Belgrade, Serbia

The basic function of the water flow lens (WFL) system is to cool, decrease, and increase light intensity with inevitable spectral oscillations, but in reality, that manipulation helps us better understand the possible additional optical and light effects and, thus, the nature of light itself, in the hopes of making significant progress toward the use of solar energy. According to our published research on a variety of solar devices, including commercial monocrystalline and amorphous Si-solar cells, differently designed Sb2S3-based solar cells made of synthesized undoped and doped semiconductors, and dye-sensitized solar cells (Dyesol/Greatcell Solar DSL 30 NRD-T) with varying sensitizers and co-sensitizers, PV performance using the WFL system can show significant improvements in all tested conditions. Based on all of our previous results on different solar devices, many potential explanations for demonstrating common extra-light effects for increases in the performance of solar cells were experimentally compared and discussed. The theoretical history of light nature was reviewed, and our findings were commented on along with new disclosure.

Performance and Stability of Quantum Dot-based Luminescent Solar Concentrators

day, 00:00

Celso de Mello Donega
Debye Institute for Nanomaterials Science, Utrecht University

Luminescent solar concentrators (LSCs) consist of transparent thin sheets in which luminescent species (luminophores) are dispersed. The luminophores absorb sunlight and reemit the energy as down-shifted photons that are partially trapped by total internal reflection and waveguided to the edges where they are absorbed by solar cells. LSCs are promising building integrated photovoltaic elements since they offer the prospect of adding energy-harvesting functionality to windows and façades, thus addressing the need to generate sustainable energy in urban environments. Their deployment has however been hindered by the lack of suitable luminophores. Colloidal semiconductor nanocrystals (quantum dots, QDs) are promising luminophores for LSCs, due to their very broad absorption spectra, large absorption cross-sections, narrow emission spectra, potentially high photoluminescence quantum yields and solution processability. In this talk, I will discuss the results of a long-term (2-year) outdoor test comparing the performance of three different large-area QD-based LSCs (CdSe/CdS/ZnS, InP/ZnSe/ZnS and CuInS2/ZnS QDs) [1,2]. An organic dye (Lumogen) -based LSC and a blank device with no luminophore were used as references. Our results show that the efficiency changes in the QD-based LSCs are related to an interplay between photodarkening and photobrightening processes in the QDs, which may be ascribed to, respectively, the creation and annihilation of traps for the photogenerated carriers. These results are relevant not only to LSCs but also to any application involving photoinduced processes in QDs.

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Day, 00:00

The influence of interface recombination on quasi-Fermi level splitting and open circuit voltage

Susanne Siebentritt
University of Luxembourg

Interface recombination represents a non-radiative loss and reduces the open-circuit voltage of any solar cell. However, its influence on the quasi-Fermi level splitting qFls can be much less pronounced, particularly in thick absorbers with a somewhat short diffusion length. In this case, interface recombination leads to a gradient in the minority quasi-Fermi level. Thus, deep inside the absorber the qFls is higher than near the interface. We determine qFLs from luminescence measurements. The emission flux depends exponentially on the qFls, thus the emission from the region with the high qFls will always dominate the emission spectrum. VOC is given by the difference between the electron quasi-Fermi level at the n-contact and the hole quasi-Fermi level at the p-contact. Therefore, the gradient in the minority quasi-Fermi level reduces the VOC below the maximum qFls. In very thin absorbers, compared to the diffusion length, this difference can disappear, because a gradient in the quasi-Fermi level cannot be sustained. Often however, the difference between VOC and qFls can give an indication of the severity of interface recombination [1]. In chalcopyrite solar cells, we can compare rather similar cells with (Cu-rich) and without (Cu-poor) interface recombination and demonstrate the influence of interface recombination on the difference between VOC and qFls. A significant difference (¿ 10meV) is only found in cells which are dominated by interface recombination. The dominance of interface recombination is also demonstrated in the electrical behaviour: the activation energy of the saturation current is lower than the band gap. This behaviour can be caused by an interface band gap lower than the bulk band gap (i.e. unfavourable band alignment) or by a high density of defects, that cause Fermi level splitting [2]. A then surface layer with a high density of defects will also cause all signatures of interface recombination, including a large difference between qVOC and qFls [1].

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The dark saturation current density in organic solar cells

Day, 00:00

Carsten Deibel
Institut für Physik, Technische Universität Chemnitz, Chemnitz 09126, Germany

The dark saturation current density in organic solar cells

Carsten Deibel, Maria Saladina Institut für Physik, Technische Universität Chemnitz, Chemnitz 09126, Germany

The dark saturation current density is a critical parameter for diodes, as it not only characterises the recombination current in the absence of light at zero volts, but also provides insights into how this current changes under illumination for higher voltages [1]. The dark saturation current density can provide valuable information about the effective energy range where charge carrier losses occur. Recent studies on organic solar cells have proposed that this effective energy range corresponds to mid-gap states, which possess energies approximately half of the effective bandgap of the respective photodiode or solar cell material blends [2]. However, I will argue that this observation is inconsistent with the ideal diode equation. In addition, the determination of the effective energy gap from the temperature dependent dark saturation current density has to account for the dependence of the diode ideality factor on the open circuit voltage – which is the case in organic solar cells [3]. I will present experimental data and modelling results for various organic semiconductor blend devices to demonstrate that the dark saturation current is not dominated by mid-gap states.

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Day, 00:00

Negative Polaron in n-Doped Organic Semiconductors: Energy Levels and Hubbard U Interactions

Antoine Kahn
Princeton University

Doping organic semiconductors leads to the formation of polarons, positive in the case of p-doping and negative for n-doping. The released carrier polarizes neighboring molecular sites and induces intra- and inter-molecular relaxation. In the traditional picture, prevalent in the literature since the 1980's, the formation of a negative polaron results in two localized levels within the gap. The addition of the electron to the LUMO/CBM leads to a gain in energy equal to the electron affinity of the neutral molecule. The ensuing molecular relaxation stabilizes the excess charge, resulting in a singly occupied level below the LUMO/CBM, and a doubly occupied level which relaxes upwards above the HOMO/VBM. Conversely, the formation of a positive polaron results in an empty level below the LUMO/CBM and a half-occupied level above the HOMO/VBM. Recent experimental and theoretical studies have challenged this original interpretation. [1-3] In the case of an added electron (hole), the new picture points to a half-occupied (empty) level below the semiconductor LUMO/CBM, split from an unoccupied (half-occupied) level positioned above (below) the LUMO/CBM (HOMO/VBM). The split corresponds to the intra-molecular Coulomb interaction, also known as Hubbard interaction U. In this work, we investigate n-doping of two organic semiconductors, the polymer P(NDI2OD-T2) and the small molecule C60, with the n-dopants [RhCp*Cp]2, [N-DMBI]2 and Cs for P(NDI2OD-T2) and [N-DMBI]2 for C60. We use ultra-violet and inverse photoemission spectroscopy (UPS, IPES) aided by EPR measurements and DFT computations. The UPS data show that the n-dopants induce the growth of a (partially) occupied DOS in the former empty gap of the organic material, while IPES shows an additional empty DOS in the unoccupied band above the LUMO. In agreement with the above-mentioned new interpretation, the additional DOS in the gap and above the LUMO are assigned to the formation of the singly occupied and unoccupied levels, respectively. These experimentally determined features allow a direct estimation of the Hubbard U, equal to 1.1-1.3 eV in the polymer, in good agreement with theory. [4] Initial experimental results lead to a Hubbard U value of 1.4 eV in C60. [5]

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First-principles modelling of excitons in metal-halide perovskites and beyond

Day, 00:00

Linn Leppert
University of Twente

Metal-halide perovskites are a structurally, chemically, and electronically diverse class of semiconductors with applications ranging from photovoltaics to radiation detectors and sensors. Understanding neutral electron-hole excitations (excitons) is key for predicting and improving the efficiency of energy-conversion processes in these materials. First-principles calculations have played an important role in this context, allowing for detailed insight into the formation of excitons in many different types of perovskites [1]. Such calculations have demonstrated that excitons in some perovskites significantly deviate from canonical models due to the chemical and structural heterogeneity of these materials [2-5]. In this presentation, I will provide an overview of our current understanding of excitons in metal-halide perovskites. I will focus on results based on Green's function-based many-body perturbation theory in the GW+Bethe-Salpeter Equation approach, the prevalent method for calculating excitons in extended solids [6, 7]. This approach readily considers the anisotropic electronic structures and dielectric screening present in many perovskites and other heterogeneous semiconductors, and important effects such as spin-orbit coupling. However, the complex and diverse electronic structure of these materials, and its intricate coupling to pronounced and anharmonic structural dynamics [8 - 10], pose challenges which are currently not fully addressed within the GW+Bethe-Salpeter Equation approach, and which I will also briefly discuss in this contribution.

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Use of Autoencoders to speed up molecular dynamic simulations

Day, 00:00

Alessio Gagliardi
Technische Universität München

With the advent of nanotechnology and the perspective of atomic-precision engineering the holy grail of material scientists has become the ability to design, in silico, materials with target properties for functionality objectives. Recently, the improvement in deep learning and generative methods has open the possibility to boost the discovery of new materials [1]. This is possible by a direct structure-to-property prediction, by coupling with experimental characterization data or by improving numerical models making them more efficient, especially when they involve expensive dynamical simulations. When the dynamics of the interested phenomenon is in some sense collective, the biasing can be performed along the slowest modes of this collective dynamics, the modes termed as collective variables (CVs). There are several methods developed over the past decades in this direction in the framework of molecular dynamic simulations [2, 3]. Despite the availability of this variety of methods, the prerequisite for efficient exploration of the configurational space is the availability of good CVs [4]. To select a number of hand-picked CVs require deep chemical intuition about the system dynamics and becomes increasingly difficult for complex systems [5]. One the other hand, one can choose as many general coordinates of the system as necessary and consider the collective variables a function of them. The task then is to come up with the transformation between the general coordinates and the collective variables. Recently, the focus of research has been either development of powerful Bayesian framework methods [6] or using a variation of an auto-associative artificial neural network or Autoencoder [7]. An Autoencoder is a nonlinear compression-decompression architecture which aims to embed input data to a relatively low dimensional space. This property is associated with a class of mathematical models called latent space models. Recent application of deep Autoencoder networks to different types of data demonstrate their ability as dimensionality reduction tool [8]. Recently, we used this method and demonstrated its utility on a large molecular system to study binding modes for drug-target interactions [9].

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Unveiling the Working Mechanisms and Limitations of Photoconversion Devices through Modulated Techniques

Day, 00:00

Agustin Alvarez
AMOLF

In recent decades, the field of photoconversion devices has witnessed remarkable progress. However, promising technologies like perovskite solar cells and photoelectrochemical cells for hydrogen generation still grapple with limitations, particularly in stability and performance, hindering their widespread commercialization. Addressing these challenges necessitates a profound understanding of the internal mechanisms governing the operation of these devices and a systematic identification of their key limitations. In this presentation, I will elucidate the potential of modulated techniques, also known as small perturbation techniques, to unveil these internal mechanisms during the operation of photoconversion devices. Starting with an exploration of the most widely adopted modulated technique, impedance spectroscopy (IS), I will delve into the intricate relationship between IS and current versus voltage curves. Specifically, I will present the correlation between the observation of negative capacitances in IS responses and the manifestation of inverted hysteresis in cyclic voltammetry.[1] Experimental results for perovskite-based devices will be showcased, highlighting their inductive-like behavior, an essential aspect for memristors. Despite the considerable power of IS for in-operando device analysis, this technique has its limitations. Consequently, the talk will delve into how these limitations can be effectively addressed by combining IS with two additional modulated techniques: intensity-modulated photocurrent spectroscopy (IMPS) and intensity-modulated photovoltage spectroscopy (IMVS). While the analysis of IMPS and IMVS spectra traditionally focuses on characteristic times, the analysis of an IS spectrum is usually made through an equivalent circuit (EC) model, but the selection of the proper EC can be challenging. A novel procedure for jointly analyzing IS, IMPS, and IMVS responses will be presented. To underscore the efficacy of this combined analysis, two practical applications will be showcased, a silicon photodiode[2] and an electrochemical hydrogen generation cell with a Zr:BiVO4 photoanode[3]. Our findings demonstrate the possibility of analyzing all three techniques with the same equivalent circuit, selected with enhanced precision. Furthermore, the combined analysis yields more accurate parameters compared to individual analyses, and it allows the acquisition of additional parameters, including the separation efficiency.

References:

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Lead-Free Perovskites for Photocatalytic H2 Production: (In-)stability and Reaction Mechanism from First-Principles Simulations

Day, 00:00

Waldemar Kaiser
Technical University of Munich

Metal-halide perovskites (MHPs) are promising materials for a variety of energy conversion applications due to their outstanding optoelectronic properties. In principle, MHPs bring along favorable properties for heterogeneous photocatalysis due to their tunable electronic band gap and electronic energy levels, but the poor stability hampered the development of MHP photocatalysts. In the last few years, to our surprise, water-stable and photocatalytically active tin-halide perovskites (THPs) and double perovskites [1, 2] have been demonstrated, paving the way towards the design of lead-free photocatalysts for hydrogen production. In this talk, I will present recent results from first-principles simulations that unveil the atomistic origins of the (in-)stability of THPs in water and compare these to their lead-based counterparts. In detail, ab initio molecular dynamics simulations on THP/water interfaces demonstrate the origin of degradation of MASnI3 in water and shed light on the surprising water-stability of DMASnBr3 [3]. Furthermore, high-level density functional theory (DFT) calculations on the key steps of the hydrogen production mechanism at the perovskite/water interface are presented [4]. The peculiar importance of surface defects and localized charge carriers at the perovskite surface are discussed for both THPs and Cs2AgBiX6 (X=Br, Cl) double perovskites.

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Day, 00:00

Interface Design and Characterization of Halide Perovskite Based Semiconductor Devices

Philip Schulz

CNRS, Ecole Polytechnique, ENSCP Institut Photovoltaïque d'Île de France
(IPVF) UMR 9006, Palaiseau 91120, France

Hybrid organic inorganic metal halide perovskites (MHPs) denote a family of compound semiconductors, which established a novel class of optoelectronics, most prominently known for the perovskite solar cell. While the power conversion efficiency of these photovoltaic devices saw a steep rise in the past decade, tailoring the interfaces between the MHP film and charge transport layer became the major control lever to enhance performance. The use of photoemission spectroscopy to analyze the chemical and electronic properties of these interfaces has been challenging due to many possible chemical reactions at the buried interfaces.1,2 For instance, it has been well documented that the optoelectronic properties of the MHP can be altered by the substrate (or selective contact) underneath,3 however so far, we do not dispose of any conclusive picture explaining this effect. We hence fabricated a functional lateral heterojunction device, which consists of a substrate with two laterally arranged selective contacts, onto which we deposited MHP films. We then used a series of photoemission measurements along with complementary experiments such as X-ray absorption spectroscopy, Kelvin probe force microscopy, and hyperspectral imaging, to measure how substrate selectivity is affecting the optoelectronic properties at the perovskite surface. We find evidence suggesting that the contact selectivity is inducing a carrier concentration gradient in the perovskite layer across the junction connected to the functionality of the lateral device opening an avenue for fundamental materials studies in operando approaches but also new device architectures including buried cells terminals.4,5 Furthermore, I will present synchrotron- and lab-based hard X-ray photoelectron spectroscopy (HAXPES) experiments to address the particular chemistry of MHPs and oxide overlayers grown by atomic layer deposition on top. We find evidence for the formation of new chemical species (PbO, N and halide-containing compounds) and changes in the energy level alignment at the MHP/oxide interface. We also used HAXPES measurements to evaluate lead-free halide perovskite films based on formamidinium tin iodide (FASnI3), for which tin fluoride (SnF2) is a commonly used additive enabling a retardation of tin oxidation and a reduction of tin vacancies. We targeted films deposited on the organic hole transport layer PEDOT:PSS for which we find the formation of an ultrathin SnS interlayer. This adds a new aspect to the discussion of high-efficiency Sn-based perovskite solar cells which still commonly make use of PEDOT:PSS as HTL material in contrast to Pb-based solar cells.6 I will conclude my talk with a general discussion about the use of PES methods for the analysis of MHP layers and in particular the effect of irradiation-induced beam damage via synchrotron and lab-based X-ray sources.7 By using complementary photoluminescence measurements we are able to investigate beam-induced changes to the optoelectronic properties and track unique physicochemical phenomena such as stimulated self-healing in formamidinium lead bromide (FAPbBr3).8,9

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Day, 00:00

Carrier recombination in different halide perovskites dimensionalities and on different length scales

Sam Stranks
University of Cambridge

Halide perovskites are generating enormous excitement for next-generation optoelectronic devices including photovoltaics, light-emitting diodes (LEDs) and detectors. Here, I will outline recent work in our group towards the development and understanding different length scales of new halide perovskite semiconductors.

I will first cover topics around recombination in bulk 3D absorbers, delving into the extraction of rate constants from optical spectroscopy data – including the pitfalls with existing approaches. I will propose methods to reliability extract these constants using only luminescence techniques, and robust fitting algorithms.

I will then show results around the microscale behaviour of recombination in films tailored for either photovoltaics or LEDs. There is an interplay between local carrier traps and radiative recombination. I will show results extended to full operating devices, and how this provides rich information about performance and stability of the optoelectronic devices.

Finally, I will show results on new 2D perovskite systems with optically or electro-active 2D molecules. Through optical spectroscopy, we can track charge transfer between the active 2D space and the inorganic backbone. These results open up new degrees of freedom whereby the spacer layer can be actively involved in charge carrier separation and transport.

Energy level alignment at 2D/3D perovskite interfaces and challenges in photoemission studies of perovskites in general

Day, 00:00

Norbert Koch
Humboldt-Universität zu Berlin

Inter alia, the application of 2D phenylethylammonium lead quaternary iodide (PEA2PbI4)/three-dimensional (3D) metal halide perovskite (MHP) interfaces has improved various optoelectronic devices, where a staggered type-II energy level alignment was often assumed. However, a type-II heterojunction seems to contradict the enhanced photoluminescence observed for 2D PEA2PbI4/3D MHP interfaces, which raises fundamental questions about the electronic properties of such junctions. Using direct and inverse photoelectron spectroscopy, we reveal that a straddling type-I energy level alignment is present at 2D PEA2PbI4/3D methylammonium lead triiodide (MAPbI3) interfaces, thus explaining that the photoluminescence enhancement of the 3D perovskite is induced by energy transfer from the 2D perovskite. On another note, a comprehensive overview of the challenges in obtaining reliable energy levels from interfaces with perovskites from photoemission spectroscopy will be given. One key obstacle is that the UV or X-ray flux used to create the photoelectrons can already induce operando-type level re-alignment. The aim of this part of the presentation is to enable the audience making own critical assessment of published photoemission data and their interpretation.

Day, 00:00

Performance-limiting processes in Cs2AgBiBr6 solar cells

Wolfgang Tress
Zurich University of Applied Sciences

Highly-efficient perovskite solar cells contain the heavy metal lead. One way to avoid lead consists of employing double perovskite materials. One of the most common representatives in photovoltaics research is Cs2AgBiBr6, so far commonly yielding power-conversion efficiencies below 5%. Although the potential of this material as an absorber in solar cells is limited due to an absorption onset above 2 eV, it is a highly interesting optoelectronic material. Understanding its properties might contribute to pave the way for the development of further lead-free perovskite(-similar) materials. In this contribution, the performance-limiting factors in Cs2AgBiBr6 solar cells are discussed based on a temperature-dependent study of the optoelectronic properties. The findings are very distinct from the lead-halide perovskites, which can be described as a semiconductor, where e.g. the common reciprocity relations are applicable. The large Stokes shift between absorption (peak at 2.8 eV) and emission (peak at 2.0 eV) of Cs2AgBiBr6 and the broad emission were early observations that gave rise to still ongoing debates. We find that it is indeed not straight-forward to employ reciprocity relations to deduce radiative open-circuit voltage (Voc) limits from sensitive measurements of the photocurrent onset. Fitting a Gaussian allows to calculate a radiative Voc limit of 1.95 V and roughly reproducing the photoluminescence (PL) spectra. However, using an exponential fit, a strongly temperature dependent Urbach energy with a value of ca. 50 meV at room temperature can be found. Furthermore, the PL, which increases by two to three orders of magnitude when decreasing the temperature to 100 K, can be separated into two peaks: One (630 nm), whose shape is hardly affected by temperature similarly to the absorption peak and a second one further in the red (670 nm) that shows a stronger temperature dependence. The first confirms earlier suggestions of a rather localized excitonic transition in contrast to a transition between a valence and conduction band of the crystal. Since the absorption peak remains mainly unaffected by temperature as well, this indicates the same origin of absorption and PL despite the large shift. Temperature-dependent photocurrent and -voltage that strongly drop for temperatures below 200 K indicate that charge separation requires temperature activation. From impedance spectroscopy, we deduce a dielectric constant of 21, which indicates efficient charge screening. Analyzing light- and temperature dependent open circuit voltage data in the range above 250 K allows for a linear extrapolation to values at 0 K, which are strongly dependent on the illumination intensity. The open-circuit voltage is limited by non-radiative recombination as electroluminescence yields of 1e-8 show, where interestingly the EL spectrum is further redshifted compared to the PL. One part of the rather low Voc of 1.1 V can be explained by poor contact selectivity as voltage-dependent external quantum efficiency measurements indicate. Also scan-rate dependent current-voltage curves unravel slow processes that

counteract efficient charge collection. Adding a 2D perovskite layer on top of the 3D one could slightly attenuate these losses. The overall goal of this contribution is to put above mentioned observations into perspective also in comparison to the lead-halide perovskites and stimulate discussions regarding their origins and consequences.

Day, 00:00

Organic Semiconductor Heterojunctions for Solar Energy Conversion

Frédéric Laquai

King Abdullah University of Science & Technology (KAUST), KAUST Solar Center (KSC)

Organic semiconductor heterojunctions, consisting of polymeric and small molecule donor and acceptor materials with offset energy levels, have garnered significant attention as photoactive systems for photovoltaic (PV) and photocatalytic (PC) energy conversion processes. However, as molecular low dielectric constant semiconductors, their primary photoexcitations are strongly-bound electron-hole pairs (Frenkel-type excitons), which can only be separated into free charges at the cost of an ’energy penalty.’ Moreover, carrier recombination processes and inherent energy losses in disordered materials remain bottlenecks for further efficiency improvements. In the first part of the talk, I will discuss recent results on efficiency-limiting processes in state-of-the-art non-fullerene acceptor (NFA) photovoltaic devices obtained by using steady-state and advanced transient optical and electro-optical spectroscopies. While the mechanism leading to efficient charge photogeneration in fullerene-based blends has been extensively studied, processes occurring in NFA-based solar cells are still less clear. In many NFA-based systems, efficient energy transfer from the donor to the acceptor precedes charge transfer, causing a dependence of the devices’ internal quantum efficiency primarily on the heterojunction’s ionization energy (IE) offset. The required IE offset has been debated extensively, with values ranging from zero or even negative offsets to hundreds of meVs, depending on the experimental techniques used to determine the energetic landscape of organic semiconductors. A better understanding of such intrinsic limitations is a prerequisite for computational (in-silico) approaches to material design. Furthermore, I will demonstrate that spectroscopically determined parameters, including rate constants and process yields of fundamental photophysical processes, can accurately reproduce experimentally measured device characteristics. In the second part of the talk, I will present recent results on using organic semiconductors in bulk heterojunction nanoparticles decorated with noble metals as a cocatalyst for photocatalytic processes such as water splitting and carbon dioxide reduction in nanoparticle suspensions under visible light excitation. Recent work has shown that photocatalytic conversion efficiencies can exceed those obtained with inorganic photocatalysts in both hydrogen generation and carbon dioxide reduction. However, similar to photovoltaic devices, photocatalytic conversion efficiencies are limited by incomplete exciton quenching and limited carrier funneling to the catalytically-active sites, as well as nanosecond to microsecond charge carrier recombination. In addition, nanoparticle preparation and cocatalyst deposition conditions tremendously affect photocatalytic efficiencies and limit the yield of photocatalytically active nanoparticle batches. Despite these constraints, the study successfully demonstrates selective carbon dioxide to methane or carbon monoxide conversion, depending on the specific nanoparticle composition and cocatalyst used, while largely suppressing proton reduction, often occurring as an unwanted side reaction.

Day, 00:00

Halide perovskite degradation in contact to metal oxides

Selina Olthof
University of Cologne

For optoelectronic devices the function and performance depends crucially on the proper choice of charge transport layers. For halide perovskite-based devices, not only energetic barriers arising between the transport levels of the various layers in the device will affect the overall performance, it is becoming more and more obvious that interactions between the perovskite and the adjacent layers play a crucial role. In particular, the direct contact to metal oxides seems to trigger chemical reactions that can lead to a partial or even complete decomposition of the perovskite film. In this talk, I will summarize our work on a variety of organic and metal oxide transport layers in contact to perovskites, where we use photoelectron spectroscopy to analyze the degradation process. [1-7] To gain a deeper understanding, we investigated a variety of different perovskites (i.e. organic vs. inorganic ones, I vs. Br, etc.) as well as the individual perovskite precursors. We find that the reactivity strongly depends on the individual material combination and that different metal oxides show fundamentally different reaction/degradation pathways. Particular attention will be given to MoO_3, ZnO, and NiO_x, where in collaboration with the theory groups of Shuxia Toa (TU Eindhoven) [5,6] and Maytal Toroker (Technion) [7] we have developed a deep understanding of the underlying degradation processes. Based on these insights we were also able to explore possible strategies to mitigate the degradation, either by controlling the defect density on the metal oxide or by employing passivating agents. Overall, I will show how photoelectron spectroscopy measurements can help to probe and understand the processes going on at these various bottom contact interfaces to perovskit, which should ultimately help to improve the stability of perovskite related devices.

References:

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