

CPP 77: Perovskite and photovoltaics IV (joint session HL/CPP)

Time: Wednesday 15:00–17:30

Location: POT 251

CPP 77.1 Wed 15:00 POT 251

Computational study of the effect of surfaces on the molecular orientation and optical properties of hybrid lead halogenide perovskites — ●GABRIELE SALEH, GIULIA BIFFI, and SERGEY ARTYUKHIN — Istituto Italiano di Tecnologia, Via Morego 30, Genova 16163 Italy

MAPbX₃ perovskites (MA = CH₃NH₃⁺, X = Cl, Br, I) are attracting intense interest due to their outstanding performance in optoelectronic devices. The efficiency of MAPbX₃ as photovoltaics is governed by the electronic excitation and de-excitation processes. It is widely recognized that MA molecules have an indirect yet strong influence on the band gap and on the optoelectronic properties in general. This effect is particularly intricate since it depends on the mutual orientations of neighboring molecules (configurations), which is continuously changing at T > 100 K [1]. Here we study the energetics of MA configurations near surfaces and the correlation between the dynamical behavior of MA molecules and the optical properties of MAPbX₃. Force field molecular dynamics (MD) and electronic structure simulations are performed. We find a linear dependence of the band gap on the octahedral tilt angle of the PbX₃ scaffold, which in turn is determined by the orientation of MA molecules. We show how the surface influences the dynamics of MA molecules, and how this affects the surface optical properties. Representative snapshots of MD simulations at different T are extracted and their electronic structure is analyzed in order to explain the measured MAPbBr₃ photoluminescence spectra. [1] O. Selig et al. (2017) J. Am. Chem. Soc., 139, 4068

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New insights into emerging lead-free double perovskite materials for optoelectronics — ●FABIAN SCHMITZ¹, JONAS HORN², TERESA GATTI¹, and DERCK SCHLETTWEIN² — ¹Justus Liebig University Gießen, Institute of Physical Chemistry — ²Justus Liebig University Gießen, Institute of Applied Physics

Lead-halide perovskites have become top-notch photoactive materials for application in solar cells, photodetectors and LEDs, due to their ease of synthesis, low-costs and excellent solution processability. For photovoltaics particularly, an impressive growth in power conversion efficiency (PCE) has been realized in few years, with current certified record at 25.2 %. Nevertheless, the most efficient perovskite solar cells still suffer from poor environmental stability and contain toxic elements. In this regard, there have been attempts to substitute lead by other metals such as tin or germanium, but no significant improvement in stability has been achieved. Good perspectives are emerging from the use of an all inorganic double perovskite, namely *Cs₂AgBiBr₆*. In this species, divalent lead is replaced by equal molar amounts of monovalent silver and trivalent bismuth. The material is characterized by an excellent environmental stability, but PCE of solar cells containing it are never higher than 2.5 %. Here we report on an optimized preparation of *Cs₂AgBiBr₆* films and on a method to estimate charge carrier diffusion lengths within them. We also provide insights into novel material design strategies for improving optoelectronic performance in this material, by resorting to 2D/3D engineering and doping/alloying.

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Costly efficient computational approach for calculating electronic structure of complex Organic/Inorganic Perovskites — MOHAMMAD MOADDELI, ●MANSOUR KANANI, and AMIR TANGESTANI — Department of Materials Science and Engineering, School of Engineering, Shiraz University

Predictive and costly efficient computational approaches are demanding for emerging perovskite structures including heavy elements. Despite large number of research productivity on this field, there is not a consensus of multiscale material design approach. One of the main bottlenecks comes from complexity of electronic interactions in such a structure which can only be analyzed using costly modified quantum mechanical methods. We introduce a hybrid quantum/molecular dynamics computational framework to deal with different diversities of synthesized perovskite layers. We use powerful reactive force field interatomic potentials for relaxing the organic part including the van der Waals effect and interactively get electronic structure using the DFT-1/2 approach. All the procedure is verified and may be modified by the accurate hybrid functional methods. Couple of potential inor-

ganic cations with various combination of halides are considered via this approach. This let us to find best candidate among large number of combinations. Furthermore we could predict a modified fractional distribution and doping elements for some introduced mixed cation perovskites in the literature.

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Spatially elucidating the role of defects in the photophysics of mixed halide perovskites — ●DAVID O. TIEDE¹, JUAN F. GALISTEO-LÓPEZ², MAURICIO E. CALVO², and HERNÁN MÍGUEZ² — ¹University of Münster, Institute of Physics, Münster, Germany — ²Instituto de Ciencia de Materiales de Sevilla (CSIC-US), Seville, Spain

Light-induced structural changes in mixed halide perovskites are among the most critical instabilities of this material that hamper their future commercialization. In particular, reversible light-induced phase segregation and its corresponding spectral changes in emission is a major drawback in precise bandgap tuning, which is crucial for both efficient light harvesting in tandem solar cells and modifying the spectral output of LEDs. Over the past years, various reports have pointed towards the critical role of defects in this process and a strong dependency on environmental effects. However, the exact mechanism of the migration of halides is still under debate.

In this work we have carried out a spectrally-resolved micro-photoluminescence experiments, employing a confocal microscope, to study the formation of iodine-rich domains in CH₃NH₃PbBrI_{3-x} thin films. With macroscopic measurements we verify that defects account for a dominant role on phase segregation and cause the formation of iodine-rich domains in minor parts of the material. By modifying the atmospheric conditions and changing the stoichiometry of halide components during the synthesis, we clarify the impact of vacancies and interstitials on the photophysics of these materials.

30 min. break

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Study of photon recycling in perovskite optoelectronics — ●CHANGSOON CHO^{1,2,3,4}, BAODAN ZHAO¹, GREGORY TAINTER¹, FREDERIK NEHM², KARL LEO², JUNG-YONG LEE³, RICHARD FRIEND¹, DAWEI DI^{1,4}, FELIX DESCHLER¹, and NEIL GREENHAM¹ — ¹Cavendish Laboratory, Department of Physics, University of Cambridge, J.J. Thomson Avenue, Cambridge CB3 0HE, UK — ²Dresden Integrated Center for Applied Physics and Photonic Materials (IAPP), Technische Universität Dresden, Dresden 01187, Germany — ³School of Electrical Engineering, Korea Advanced Institute of Science and Technology (KAIST), Daejeon 34141, Republic of Korea — ⁴State Key Laboratory of Morden Optical Instrumentation, College of Optical Science and Engineering, Zhejiang University, Hangzhou 310027, China

Photon recycling (PR), consisting of successive light re-absorption and re-emission processes, is a key mechanism to understand the distinct optical properties of perovskite light-emitting diodes (PeLEDs). Here we verify the existence of PR effect via the measurement of spatially-resolved photoluminescence (PL) and electroluminescence (EL). To quantify the PR effect in PeLEDs, we propose an optical modelling method taking photon re-absorption and re-emission into account. According to the optical analysis of currently reported state-of-the-art devices, PR is shown to be able to contribute to more than 70% of the EQEs. Finally, we introduce various optical designs of PeLEDs, to maximize PR effect and enhance the quantity and property of light emission.

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On the Radiative Recombination Efficiency and Carrier Lifetime in Halide Perovskite Solar Cell Materials — ●THOMAS UNOLD¹, MARTIN STOLTERFOHT², JOSE MARQUEZ-PRIETO¹, DIETER NEHER², and THOMAS KIRCHARTZ³ — ¹Helmholtz-Zentrum Berlin — ²Institute of Physics and Astronomy, Universität Potsdam — ³Forschungszentrum Juelich GmbH

The open-circuit voltage, which is currently considered the main performance limitation of halide perovskite solar cells, is determined by bulk and interface recombination processes in the solar cell.[1] Under

ideal conditions the open-circuit voltage approaches the internal quasi-Fermi level splitting (QFLS), which may be estimated by the measurement of the external photoluminescence quantum yield (PLQY) and the absorption properties.[2] The photoluminescence quantum yield and quasi-Fermi level splitting can also be estimated by measurement of the time-resolved photoluminescence (TRPL), if the radiative recombination constants and photon recycling are taken into account properly.[3] A survey of the literature shows that the reported PLQY for measured open-circuit voltages sometimes vary by orders of magnitude, which is difficult to understand from the point of theory. In this contribution we show that careful consideration of the above points leads to a consistent picture of the interrelation of the QFLS, PLQY and TRPL lifetime, and discusses possible sources of error in the analysis. [1] Stolterfoht et al., *Nature Energy* 3 (2018) 847 [2] Liu et al., *ACS Energy Lett.* 4 (2019) 110 [3] Staub et al., *Phys. Rev. Appl.* 6 (2016) 044017

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Carrier Diffusion in Bulk and Nanocrystalline Halide Perovskites — ●MICHAEL LICHTENEGGER and ALEXANDER URBAN — Nanospectroscopy Group, Nano-Institute Munich, Department of Physics, Ludwig-Maximilians-Universität München, Königinstr.10, 80539 Munich, Germany

Carrier injection, transport and extraction are crucial physical properties for optoelectronic applications such as solar cells and light-emitting diodes (LEDs). Depending on the magnitude of the exciton binding energy in a material, electron-hole pairs can exist as free carriers or can form a bound state, so-called excitons. Large binding energies appear especially for systems where at least one spatial dimension is confined, e.g. in nanocubes, 2D nanoplatelets 1D nanowires or 0D quantum dots. Consequently, carrier transport will be very different depending on the nature of the material.

Our goal is to investigate and understand carrier diffusion in large

perovskite crystals and assemblies of nanocrystals. For this we have realized a contactless photoluminescence (PL) and confocal time-correlated single photon counting (TCSPC) setup. We use this to measure the diffusion length for bulk perovskite films, and thin films of nanocrystals. We modify the perovskite compositions, geometries and passivating ligands to investigate the nature of the transport processes and determine critical parameters such as diffusion lengths and diffusion coefficients.

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Temperature-dependent high spatial-resolution spectroscopy to investigate NCs in single micelles — ●MARKUS SCHÖGER, CAROLA LAMPE, MORITZ GRAMLICH, and ALEXANDER URBAN — Nanospectroscopy Group, Department of Physics, Ludwig-Maximilians-Universität, Munich, Germany

Halide perovskite nanocrystals (NCs) have shown outstanding potential for light-emitting applications with high quantum yields and an emission wavelength tunable throughout the visible spectrum. Despite these impressive advances, they still suffer from several issues, which currently impede widespread commercialization. To enhance their stability, we recently introduced a colloidal synthesis, wherein the NCs are grown inside diblock copolymer micelles.¹ These nanoreactors additionally provide a protective shell, limiting degradation and ion migration. In order to maximize their potential and enable optoelectronic integration, an in-depth understanding of the nanostructure structure and resulting properties on a single micelle level is critical. Here, we present temperature-dependent high spatial-resolution spectroscopy to investigate single micelle-encapsulated perovskite NCs in temperature intervals from 4 K up to room temperature to gain insight into their fundamental properties.

¹ V. A. Hintermayr, C. Lampe, M. Low, J. Roemer, W. Vanderlinden, M. Gramlich, A. X. Bohm, C. Sattler, B. Nickel, T. Lohmüller, and A. S. Urban, *Nano Lett.* 19 (8), 4928 (2019).