

# International Conference on Simulation of Organic Electronics and Photovoltaics 2022



# **Zurich University of Applied Sciences**

# 7.-9. September 2022

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# Scientific Program

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#### Perovskite solar cells - reducing the defect density

Emilia Schütz, Muhammad Irfan Haider, Azhar Fakharuddin, Tobias Seewald, Daniele Cuzzupé, <u>Lukas Schmidt-Mende</u>

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DE-78457 Constance, Germany

In perovskite optoelectronic devices, we find defects in the perovskite layer itself as well as at the interfaces. In this presentation, we will give examples to reduce the defects within the perovskite bulk as well as at the interfaces between perovskite and charge transport layers. We will demonstrate how perovskite films on NiO<sub>x</sub> interfacial layers strongly depend on NiO<sub>x</sub> treatment. The role of oxygen stoichiometry towards the surface properties of NiO<sub>x</sub> and its influence over perovskite film morphology, band alignment, charge extraction at NiO<sub>x</sub> / MAPbI<sub>3</sub> interface and long-term shelf-life stability of the inverted perovskite solar cells will be analyzed (see Fig.1).<sup>1</sup>



**Fig. 1** Long term measurement demonstrate that perovskite solar cells prepared on r-NiOx films are more stable (only 5% drop after 200 days). We even observe an initial increase in performance. However, higher Ni<sup>3+</sup> containing films seem detrimental for interface stability in perovskite devices.

<sup>&</sup>lt;sup>1</sup> M. I. Haider, A. Fakharuddin, S. Ahmed, M. Sultan, L. Schmidt-Mende, Modulating defect density of NiO hole transport layer via tuning interfacial oxygen stoichiometry in perovskite solar cells, *Solar Energy* **2022**, 233, 326.



**Fig. 2** Recrystallization of a perovskite film. After intercalation of methylamine gas into the perovskite crystal structure, the film is getting liquid and transparent. If the partial pressure of the methylamine gas is reduced and the temperature is increased the film starts to recrystallize again. This process is observed in this figure, starting at random nucleation points, which then grow until the film is completely transferred to a perovskite film again with large crystals.

In another example, we demonstrate how the reduction of bulk defects such as those related to grain boundaries can be achieved. The reaction between a preexisting perovskite film and methylamine (MA) gas has been shown to possess the striking ability to both improved morphology and increased grain size drastically (see Fig. 2), boosting performance in photodetectors. This promising post-deposition treatment could provide the means to decouple quality from the initial deposition process, thus promising to facilitate upscaling and lowering production costs.<sup>2,3</sup> We demonstrate how with such an MA treatment the film morphology can be tuned from small to large grain sizes and how this improves the performance of photodetectors based on such treated perovskite films with large grain size.

<sup>&</sup>lt;sup>2</sup> T. Seewald, E. R. Schutz, C. Ebenhoch, L. Schmidt-Mende, Curing perovskites-a way towards control of crystallinity and improved stability, *Journal of Physics-Energy* **2020**, 2.

<sup>&</sup>lt;sup>3</sup> E.R. Schütz, A. Fakharuddin, Y. Yalcinkaya, E. Ochoa-Martinez, S. Bijani, A. Rahsid bin Mohd Yusoff, M. Vasilopoulou, T. Seewald, U. Steiner, S.A.L. Weber, L. Schmidt-Mende, Reduced defect density in crystalline halide perovskite films via methylamine treatment for the application in photodetectors, submitted **2022**.

#### **Device and Energy Yield Modelling of Perovskite Solar Cells**

<u>Pilar López-Varo<sup>1</sup></u>, Jean-Baptiste Puel<sup>1,2</sup>, Davide Regaldo<sup>1</sup>, Philip Schulz<sup>1,3</sup> <sup>1</sup>IPVF, Institut Photovoltaïque d'Île-de-France, Palaiseau, 91120, France <sup>2</sup>EDF R&D, Palaiseau, 91120, France. <sup>3</sup>CNRS, IPVF, UMR 9006, Palaiseau, 91120, France

Device modelling and energy yield (EY) calculations are essential tools to optimize solar cell architectures, and in particular, perovskite solar cells (PSC). In this work, we present our latest progress on device modelling and EY calculations. Device modelling through coupled optical and electrical simulations allows to analyze device performance under standard conditions. It is well known that the energy alignment at the interfaces, ion migration and potential distribution along the device have an impact on device performance. To investigate them, we have characterized horizontal microstructures, which are a flipped version of the PSC vertical sandwich structure. We were able to draw the potential distribution along the solar cell structure by using X-ray photoelectron spectroscopy (XPS)<sup>1</sup>. Furthermore, in this work we took into account the role of ion migration for the analysis of the band device structure. EY calculations estimate the total output generated energy of a solar cell after one year in a specific place. EY calculations allow not only a device optimization in terms of maximization of the predicted output power, but also the analysis of device stability<sup>2,3</sup>. Device stability can be affected by the variation of external ambient conditions, such as light intensity and temperature. Thus, it is crucial to determine accurately the device temperature<sup>3</sup>. To estimate cell temperature, we propose a thermal model which is a function of device parameters, environmental variables, and is strongly linked with the experimental optical-electrical-thermal performance<sup>3</sup>. We studied the effect of realistic temperature conditions on the performance of PSCs and their transient response to environmental external changes using a theoreticalexperimental combined approach. Linking the experimental results and our model, we were able to evaluate the most sensible device layers that increment device temperature affecting device stability.

<sup>&</sup>lt;sup>1</sup> D. Regaldo, A. Bojar, S. P. Dunfield, P. Lopez-Varo, M. Frégnaux, V. Dufoulon, S.-T. Zhang, J. Alvarez, J. J. Berry, J.-B. Puel, P. Schulz, J.-P. Kleider. Prog Photovolt Res Appl. 1–9 (2021)

<sup>&</sup>lt;sup>2</sup> J.-P. Bastos, G. Uytterhoeven, W. Qiu, U. W. Paetzold, D. Cheyns, S. Surana, J. Rivas, M. Jaysankar, W. Song, T. Aernouts, J. Poortmans, and R. Gehlhaar. ACS Appl. Mater. Interfaces. 11, 16517–16526 (2019)

<sup>&</sup>lt;sup>3</sup> P. López-Varo, M. Amara, S. Cacovich, A. Julien, A. Yaïche, M. Jouhari, J. Rousset, P. Schulz, J.-F. Guillemoles, and J.-B. Puel. Sustainable Energy Fuels, 5, 5523-5534 (2021)

### Importance Of Light Outcoupling for Organic Photovoltaics Films Characterization: Exciton Diffusion Length and Charge Transfer State Energy.

<u>Julien Gorenflot</u>,<sup>1</sup> Valentina Belova,<sup>2</sup> Maryam Alquatani,<sup>1</sup> Ali AlMarhoun,<sup>1</sup> Aleksandr Perevedentsev,<sup>2</sup> Frédéric Laquai,<sup>1</sup> Mariano Campoy-Quiles<sup>2</sup>

- KAUST Solar Center, Physical Science and Engineering Division, Material Science and Engineering Program, King Abdullah University of Science and Technology, 4700. 23955-6900 Thuwal, Saudi Arabia
- 2- Nanopto Group, Nanostructured Materials for Optoelectronics and Energy Harvesting Research Unit, Material Science Institute of Barcelona, ICMAB-CSIC, 08193 Bellaterra, Spain

Emission spectroscopy enables to access the excited states property in the heart of organic solar cells. This includes quantities as critical as excitons diffusion length  $L_D$  and charge transfer state energy  $E_{CT}$ . However, optical interferences between the multiple layers of this sample affect considerably the spectral shape and intensity of those emission and need to be accounted for. Here we present two examples of in-coupling and out-coupling simulation and correction for diffusion length and charge transfer state energy extraction, respectively.

Herein, a high-throughput approach to probe  $L_D$  in nonfullerene acceptors (NFAs) is reported, that builds upon the conventional photoluminescence (PL) surface quenching method using NFA layers with a graded thickness variation in combination with spectroscopic PL mapping. The method is explored for two archetypal NFAs, namely, ITIC and IT-4F, using PEDOT:PSS and the donor polymer PM6 as two distinct and practically relevant quencher materials. Interestingly, conventional analysis of quenching efficiency as a function of acceptor layer thickness results in a threefold difference in  $L_D$  values depending on the specific quencher. This discrepancy can be reconciled by accounting for the simulated differences in light in- and outcoupling efficiency for different multilayer architectures. In particular, it is shown that the analysis of glass/acceptor/PM6 structures results in a major overestimation of  $L_D$ , whereas glass/acceptor/PEDOT:PSS structures give no significant contribution to outcoupling, yielding  $L_D$  values of 6–12 and 8–18 nm for ITIC and IT-4F, respectively. Practical guidelines for quencher choice, sample geometries, and analysis approach for the accurate assessment of LD are provided.<sup>[1]</sup>

Similarly electroluminescence spectra emitted from PM6:Y6 solar cells are found to vary considerably with the active layer, seemingly indicating an evolution of the charge transfer state energy. We show that this discrepancy can be resolved by correcting for the out-coupling

spectra, determine the intrinsic emission spectrum of the active layer material and the corresponding emitting states energy. This spectra are in turn decomposed in charge transfer state emission and Y6 emission to finally access the PM6:Y6 interface state energetics.

[1] Belova, V., Perevedentsev, A., Gorenflot, J., De Castro, C.S.P., Casademont-Viñas, M., Paleti, S.H.K., Karuthedath, S., Baran, D., Laquai, F. and Campoy-Quiles, M. (2022), Effect of Quencher, Geometry, and Light Outcoupling on the Determination of Exciton Diffusion Length in Nonfullerene Acceptors. Sol. RRL 2100822. <u>https://doi.org/10.1002/solr.202100822</u>



**Figure 1:** The modulation effect of light in- and outcoupling on the photoluminescence from layered structures of widely employed organic photovoltaics materials, used for the assessment of the exciton diffusion length LD, is investigated. The effect is shown to depend on the optical constants of the layers and the sample architecture. A possible correction method is introduced. Source: [1].

#### Lateral Junctions for Organic Solar Cells

Masahiro Hiramoto

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Blended junctions are indispensable for organic solar cells. However, the fabrication of electron and hole transport routes in blended junction remains quite challenging. In this work, we proposed a novel concept of the junction structure of organic solar cell, namely, a lateral alternating multilayered junction (Fig. 1)<sup>1</sup>. An essential point is that the photogenerated holes and electrons are laterally transported and extracted to the respective electrodes.

Minimum units of proposed junction are hole highway and electron highway. At first, the lateral extraction of photogenerated holes and electrons of the order of 1 mm in the hole and electron highways using ultra-high mobility organic films was demonstrated. Observed macroscopic value of milli-meter order is surprising long compared to the conventional value below 1  $\mu$ m. Next, the successful operation of organic solar cell having a lateral alternating multilayered junction by combining the hole highway and electron highway was demonstrated. A total of 93% of the photogenerated electrons and holes are laterally collected over a surprising long distance (0.14 mm). The exciton-collection efficiency reaches 75% in a lateral alternating multilayered junction with a layer thickness of 10 nm.





Therefore, the lateral junction is proved to have an ability to collect both excitons and carriers almost completely. A lateral alternating multilayered junction can be regarded to be an alternative blended junction for organic solar cells.

Even lateral cells with L = 1.8 cm showed clear photovoltaic behavior<sup>2</sup>. The lateral tandem cells with carrier transport and generation layers can be successfully operated<sup>3</sup>. Advantage of a lateral cell is its unlimited thickness in the vertical direction. Therefore, lateral-tandem solar cells that can utilize the full solar spectrum can be freely designed.

<sup>2</sup> Jaseela P. I., A. Girault, M. Kikuchi, Y. Yabara, S. Izawa, M. Hiramoto, "Photovoltaic behavior of centimeterlong lateral organic junctions", Appl. Phys. Exp. 14, 094001, (2021).

<sup>3</sup> Jaseela P. I., Y. Yabara, S. Uto, S. Izawa, M. Hiramoto, "Lateral-tandem organic photovoltaic cells with carrier transport and generation layers, Appl. Phys. Exp. 14, 101003, (2021).

<sup>&</sup>lt;sup>1</sup> M. Kikuchi, M. Hirota, T. Kunawong, Y. Shinmura, M. Abe, Y. Sadamitsu, A. M. Moh, S. Izawa, M. Izaki, H. Naito, and M. Hiramoto, "Lateral alternating donor/acceptor multilayered junction for organic solar cells", ACS Appl. Energy Mater. 2, 2087, (2019).

### Thermal Degradation Mechanism in PM6: Y-series Acceptor Bulk Heterojunction for Organic Solar Cells by Device Simulation

Si Chen, Julien Gorenflot, Jianhua Han, Frédéric Laquai KAUST Solar Center, King Abdullah University of Science and Technology, 4700. Thuwal, Saudi Arabia

Organic solar cells (OSCs) have promising advantages such as low cost and flexibility, which have received great attention from the scientific and industrial community<sup>1</sup>. However, factors such as poor light and thermal stability still greatly limit the industrialization of organic solar cells, and the study of the degradation mechanism of high-efficient PM6:Y series systems is an urgent problem to be solved. Although we can feel some parameters or properties are degradated by experimental measurements, we cannot figure out quantitatively how they impact the J-V performance. However, simulation can help connect the experimental parameters and J-V performace quantitatively. We chose PM6:Y2, PM6:Y6, PM6:Y12 systems to test their thermal stability because of their typical difference on their chemical structures like the length of side chains and end groups, and finally we found their different effects of degradation. We measured time-resolved photoluminescence (TRPL), transient absorption (TA), time-delayed collection field (TDCF) before and after thermal degradation(85°C in N<sub>2</sub>), from which we extracted parameters like generation efficiency and recombination efficienty to simulate different loss channel. Then we got the contributions from each channel or parameter to the device performance from simulated JV curves with simulation software SETFOS 5.2. Additionally, with the help of simulation, we can conclude a quantitative current loss diagram before and after degradation. The diagram suggests a potential maximum current density up to 40mA/cm<sup>2</sup> for some systems like PM6:Y6 and PM6:Y12. Finally, we can not only conclude the different internal mechanism of thermal degradation in PM6:Y-series OSCs, but also connect the loss with the structures of acceptors, which can provide the design direction of future organic photovaltaic materials.

<sup>&</sup>lt;sup>1</sup> Cheng, Pei, and Xiaowei Zhan. "Stability of organic solar cells: challenges and strategies." Chemical Society Reviews 45.9 (2016): 2544-2582.



Figure 1: The impact of each parameter on thermal degradation for PM6:Y2 devices

### A SPICE Simulation for Optimised Perovskite-based PV Modules for Integrated Applications

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Thin film PV modules are promising for low weight to power ratio installations, such as BIPV and VIPV or for indoor applications. However, in a residential landscape, modules are often misoriented and mispositioned and buildings, street lamps, or other urban constructions partially shade the solar device. Despite the increased temperatures and potential generation of hotspots, silicon PV is barely affected by reverse bias behaviors. On the contrary, perovskites have shown low reverse breakdown voltages and potential irreversible degradation when subject to reverse biases<sup>1</sup>.

To address the stressful conditions induced by inhomogeneous illumination for cells connected in series, we improved our previously presented SPICE simulation<sup>2</sup> to evaluate the performances of alternative PV module configurations under the effects of partial shading. The module is modelled as a mesh of 1-diode equivalent circuits in parallel to recreate solar cells and properly interconnected through a modelled P1P2P3. In this presentation, we present a sensitivity analysis on how different material's and layout's parameters impact shaded devices and propose optimal configurations for shade tolerant perovskite based modules for residential applications.

<sup>&</sup>lt;sup>1</sup>Najafi, L.; Bellani, S.; Gabatel, L.; Zappia, M. I.; Di Carlo, A.; Bonaccorso, F. Reverse-Bias and Temperature Behaviors of Perovskite Solar Cells at Extended Voltage Range, ACS Applied Energy Materials 2022 5 (2), 1378-1384

<sup>&</sup>lt;sup>2</sup> Carolus, J.; Purohit, Z.; Vandenbergh, T.; Meuris, M.; Tripathi, B.; Daenen, M. Proposing an Electro-Thermal SPICE Model to Investigate the Effect of Partial Shading on CIGS PV Modules, 35th European Photovoltaic Solar Energy Conference and Exhibition, Session 5CV.3.35, Pages 1343 - 1345, ISBN 3-936338-50-7.

### Electro-Thermal Small-Signal Analysis of Defects in Large-Area Perovskite Solar Cells

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The production of uniform layers without defects is crucial for the efficient upscaling of perovskite solar cells. To understand the origin of defects and their impact on efficiency, we compare steady-state and alternating current (AC) measurements with simulations from a 2D+1D finite element method (FEM) tool called Laoss. The software supports the upscaling process from small- to large-area devices by solving for the potential and temperature distribution in 2D top and bottom electrode domains, which are connected by a vertical 1D coupling law.

We measured and accordingly modelled a screen printed carbon-based hole-transporter-free perovskite solar cell with a pinhole-like defect. Current-voltage characteristics and electroluminescence (EL) images of the cell were measured in steady-state and then compared with simulations for detailed insights into the device. The pinhole-like defect can be seen in the EL-image, among other noticeable features, e.g. efficiency limiting areas originating from the printing process.

The software can not only perform steady-state simulations but also determine the influence of nonideal electrodes on potential and temperature distributions in the frequency domain. Therefore, we also introduce the small-signal dark lock-in thermography (SS-DLIT) method to measure and simulate electro-thermal effects in perovskite solar cells in the dark with high accuracy thanks to the use of a small, periodic voltage modulation at a chosen offset voltage. This adapted DLIT method can be simulated with the thermal AC module of our software tool and allowed a successful quantification of the pinhole-like defect by manually fitting the simulated AC temperature amplitude to the SS-DLIT measurements.

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Fig. 1 SS-DLIT measurement and simulation of a carbon-based hole-transporter-free PSC.

#### Bandgap and temperature dependent modelling of perovskite solar cells

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Abstract: Perovskite solar cells (PSCs) have shown extraordinary advancement and could be placed along with established photovoltaic technologies under laboratory conditions. Nevertheless, control environment induced in laboratory measurements do not entirely relate with real-operating conditions where devices are subject to different spectra, irradiance, and operating temperatures. Comprehensive temperature-dependent optical-electrical modeling is essential to quantify optoelectrical losses and energy yield of state-of-the-art single junction perovskite and perovskite-based tandem devices under real-world operating conditions. Such models require accurate bandgap and temperature-dependent optical constants of perovskites. Here, we report on the temperature-dependent optical constants, empirically determined via the spectroscopic ellipsometry technique, of triple-cation perovskites with bandgaps ranging between 1.58 eV to 1.77 eV. In addition, we have developed a simple empirical model to calculate the optical constants of with bandgaps ranging between 1.58 eV to 1.77 eV between the temperature range of 25°C - 95°C. Finally, we conducted measurements under various temperatures to show that the empirical model can extract the optical constants of various perovskite absorbers of different bandgaps at a range of temperatures suitable for modeling in typical solar cell operating conditions.



Fig. 1: (a) Temperature-dependent measured (symbol) and modeled (line) optical constants (n and k) at different temperatures (25 and 75°C) for the 1.66 eV perovskite film (b) Modelled (line) k data at higher temperature (75°C) near the band edge extracted using room temperature (25°C) measured k data. Mean Square Error (*MSE*) map of optical constants (c) n and (d) k, between measured and modeled as function of temperature and bandgap of the perovskite.

# Modelling the evolution of IV characteristics in perovskite solar cells under different operating conditions

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The perovskite photovoltaic technology is unique when compared to other technologies. There are different cell architectures (P-I-N, N-I-P planar or in mesoporous) that are being researched. Although some of the architectures may resemble a PN or P-I-N architecture of c-Si or CIGS technology, the charge creation and collection mechanism in perovskites are significantly different as the cell structure is organic or organic/inorganic hybrid. Perovskites also exhibit unique behaviours like light-soaking effect, hysteresis, reversible degradation, etc.

Multiple researchers have linked the anomalous device behaviour to the ionic movement within the perovskite layer<sup>1–3</sup>. Ion dynamics in perovskites are very sensitive to external conditions like temperature, illumination, and biasing conditions, thus varying the IV performance of the device. Through measurements, we quantified the individual effects of irradiance, temperature, and bias on the ion dynamics, thereby it effects on IV characteristics. The performance of the device has also been known to degrade during illumination and recover in dark<sup>4,5</sup>. We also study the evolution of the IV performance over time to identify and model the degradation and recovery of the device. We will present an analytical model to simulate the device phenomenon under different stressors. With the model, we will be able to estimate the instantaneous performance, energy yield and lifetime of the device.

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### Influence of Phase Segregation on Charge Transport and Recombination in CH<sub>3</sub>NH<sub>3</sub>Pb(I<sub>1-x</sub>Br<sub>x</sub>)<sub>3</sub> based-Perovskite Solar Cells

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Abstract: Mixed halide perovskites enable tunable bandgap engineering for tandem solar cells and light emitting diode applications. However, the mixed halide perovskites suffer from phase segregation (I<sup>-</sup> rich and Br<sup>-</sup> rich regions) under visible light illumination. The photoinduced phase segregation are well established in mixed halide perovskite thin films & single crystals and characterized by various optical techniques such as two photon Photoluminescence microscopy.<sup>i</sup> The effect of phase segregation on the charge transport properties in devices is not well addressed in the literature. In this study, we fabricated a series of inverted (p*i-n*) heterojunction perovskite solar cells (PSCs) with active layer  $CH_3NH_3Pb(I_{1-x}Br_x)_3$ , where x = 0, 0.25, 0.50, 0.75, 1.0. Intensity modulated photocurrent spectroscopy (IMPS) and intensity modulated voltage spectroscopy (IMVS) are used to estimate charge transport and recombination lifetime ( $t_{CT}$  and  $t_R$ ), respectively.<sup>ii</sup> The  $t_{CT}$  and  $t_R$  are highly influenced by I<sup>-</sup>rich and Br rich regions and shows two lifetimes at different timescales. However, for pure MAPbI<sub>3</sub> and MAPbBr<sub>3</sub> devices, the charge transport and recombination processes are characterized by a single lifetime. Furthermore, IMPS and IMVS are correlated with other optoelectronic studies such as photoluminescence and photoelectron spectroscopy to validate the results. In addition, the effect of time dependent phase segregation on the charge transport and recombination dynamics is discussed and correlated with the device performance.

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### How Selective Contacts Influence Capacitance Measurements of Perovskite Solar Cells

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Capacitance measurements as a function of voltage, frequency and temperature are a useful tool to gain deep insight into the electronic properties of solar cells. In the case of perovskite solar cells, techniques such as capacitance-voltage, Mott-Schottky analysis or thermal admittance spectroscopy measurements are frequently employed in order to obtain relevant parameters of the perovskite absorber. The interpretative framework for these analyses always assumes that the entire capacitance signal is dominated by the perovskite absorber, ignoring any contributions of the selective contacts. Such an assumption is erroneous since the selective contacts are often quite resistive in nature, which implies that their resistance will significantly contribute to the total device impedance and thereby also affect the overall capacitance of the device. Based on this premise, we develop a simple electrical model of the perovskite solar cell that explicitly accounts for the impedance and capacitance of the selective contacts.<sup>1</sup> Analysis of this model supplemented by full drift-diffusion simulations, yields fundamental limits to the resolution of spatial doping profiles and minimum values of doping/trap densities,<sup>2</sup> built-in voltages and activation energies. We observe that most of the experimental capacitance-voltage-frequency-temperature data, calculated doping/defect densities and activation energies reported in literature are within the derived cut-off values, indicating that the capacitance response of the perovskite solar cell is indeed strongly affected by the capacitance of its selective contacts. In addition, we extend this analysis to the interpretation of the frequency-domain small-perturbation techniques - namely impedance spectroscopy (IS), intensity-modulated photocurrent spectroscopy (IMPS) and intensity-modulated photovoltage spectroscopy (IMVS), by explicitly accounting for the selective contact resistance using appropriate boundary conditions in the diffusion-recombination model. Our developed analyses are valid for the interpretation of capacitance measurements of any photovoltaic technology that employs low-conductivity charge transport layers in addition to the absorber layer.

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<sup>&</sup>lt;sup>2</sup> Ravishankar, S., Unold, T. and Kirchartz, T., 2021. Comment on "Resolving spatial and energetic distributions of trap states in metal halide perovskite solar cells". *Science*, *371*(6532), pp. eabd8014.

### $C_{\text{tot}}$ (V, f) depends on R||C of three layers



**Fig. 1** Simple electrical model to model the impedance and capacitance response of perovskite solar cells, while accounting for the influence of the selective contact layers to the total measured capacitance. Analysis of this model yields resolution limits for the measurement of doping/defect densities, built-in voltages and activation energies.

#### Determining the mobile ion density in perovskite solar cells

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Mobile ions in perovskite solar cells cause performance losses and degradation by screening the built-in-field and impeding charge extraction.<sup>1,2</sup> In order to characterize and mitigate these losses, it is crucial to obtain the mobile ion density in these cells. Using Drift-Diffusion simulations, we discuss how to interpret commonly employed measurement techniques<sup>3,4</sup> and evaluate them based on their ability to accurately reproduce correct densities. We find that the measurement technique has to be adapted to the cell's parameters and that implicit assumptions need to be checked when analyzing measurement data. If the ionic density is low compared to the electrode charge, charge extraction experiments may be used to accurately determine ionic density. For higher ionic densities, space charge layers form, which can be measured using low-frequency capacitance measurements. By combining the two methods, we can obtain ionic density over a broader range of parameters than previously accessible.

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#### **Overcoming the Limitations of the Transient Ion Drift Method for Extracting Material Parameters of Perovskite Solar Cells**

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Modelling the charge dynamics of perovskite solar cells can significantly accelerate the device optimization process and provides in-depth understanding of the mixed electronic-ionic charge transport. Reliable device models require knowledge of the material parameters, which are typically obtained from measurements. However, obtaining reliable material parameters can be challenging. Properties of bulk material might not apply once the material is incorporated in a thin-film device. And when measuring complete devices, the same apparent device characteristics can be described by multiple parameter sets, i.e. parameters can be correlated<sup>1</sup>.

At Fluxim we combine numerical device models with complementary measurements of complete devices in order to reliably extract relevant material parameters. Simultaneously involving steady-state, transient and AC measurement techniques can significantly improve the accuracy of the extracted parameters due to a reduction of parameter correlations, as successfully demonstrated for organic and perovskite solar cells <sup>2-3</sup>.

Here we focus on the extraction of the charge concentration and the mobility of ionic charge carriers in perovskite devices, which is particularly challenging because most experimental techniques are only sensitive to the product of concentration and mobility but not to the individual parameters themselves<sup>3</sup>. M.H. Futscher et al. and L. McGovern et al.<sup>4-5</sup> recently applied a measurement technique called transient ion drift to perovskite solar cells. Originally developed for inorganic p-n junctions, this technique allows to individually and quantitatively

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measure both concentration and mobility of slowly moving mobile charges<sup>6</sup>. However, the analytical model describing the capacitance over time limits the validity of the extraction method to materials with low concentrations of mobile charges and comparably high doping densities<sup>4-6</sup>. We use the revised drift-diffusion solver in Setfos 5.3 to model the transient ion drift experiment, by combining transient simulations with a small-signal analysis approach to compute the transient capacitance. We show that the limitations of the analytical model can be overcome by combining measurement and simulations, allowing the application of the method to a broader set of devices.

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### Ion migration in perovskite solar cells: modelling and impedance characterization

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Hybrid metal halide perovskites are mixed ionic-electronic semiconductors with exceptional optoelectronic properties, ideal for applications in photovoltaics, along with many others like lighting, lasing and x-ray detection. In all these applications, robustness and stability of the material is crucial. Bearing in mind that perovskites are ionic materials, it is expected that ion migration should play a significant role in all stability issues under operational conditions, which often are triggered by irreversible ionic displacements.

It is well-known that most experimental advances and record performances are not obtained with pure perovskites but with mixed compositions, empirically devised. In addition, many key features in photovoltaic perovskites occur at relatively long-time scales due to the relatively slow ionic motion. Description of perovskites using first-principles techniques such as density functional theory (DFT) are limited by their high computational demands, which restricts their use to small systems (hence only pure materials can be studied) and short-time scales (hence ionic motion cannot be properly described). The Holy Grail in photovoltaics would be to be able to predict the expected performance, i.e. the current-voltage curve, starting from materials properties and low-level first-principles atomistic calculations.

In this work we combine atomistic calculations with drift-diffusion (DD) modelling to simulate the current-voltage curve of perovskite solar cells. In order to get around the limitations of DFT, we use classical molecular dynamics (CMD), which is less expensive and enables simulations for larger systems and longer times. We have developed the required force field for CMD by means of a genetic algorithm.<sup>1</sup> The algorithm finds the best parameter set that simultaneously fits the DFT energies obtained for several crystalline structures with moderate degrees of distortion with respect to the equilibrium configuration. Using this strategy we have found a transferable classical force field to describe the mixed hybrid perovskite  $MA_xFA_{1-x}Pb(Br_yI_{1-y})_3$  for variable composition ( $\forall x, y \in [0, 1]$ ). The model includes Lennard-Jones and Buckingham potentials to describe the interactions between the atoms of the inorganic lattice and the organic molecule, and the AMBER model to describe intramolecular

atomic interactions. The resulting model reproduces correctly the XRD patterns, the expansion of the lattice upon I/Br substitution and the thermal expansion coefficients. From the simulations we have extracted the ion diffusion coefficients of the pure and mixed perovskites, presenting for the first time these values obtained by a fully dynamical method using a transferable model fitted to first principles calculations.

The ion diffusion coefficients obtained are then transferred to a DD model<sup>2,3</sup> including transport and recombination of charge carriers (electrons and holes) in the perovskite, transport of a single positive ionic species (iodine vacancies in the perovskite), and electron and hole transport in the electron transport layer and hole transport layer, respectively. The charge carriers and ions are fully coupled through including the electron and hole charge densities with the ion charge density in the Poisson's equation. The DD calculation allows to simulate the current-voltage curve both in the steady state and in a transient situation for different scan rates. The ionic parameters derived from the atomistic simulations are proven to lead to the correct prediction of the ion-induced hysteresis and the low frequency features of the impedance spectrum for a variety of perovskite compositions.



**Fig. 1** Drift-diffusion modelling transient current-voltage results near the maximum power point for several choices of the ionic parameters.

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Affect Perovskite Solar Cell Performance: Insights from a Coupled Charge Transport/Ion Migration Model. Energy Environ. Sci. 2018, 12, 396–409.

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#### **Over 24% Flexible Monolithic All-perovskite Tandem Solar Cells**

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Perovskite solar cells (PSCs) have received vigorous attention from academia and industry due to their high power conversion efficiency, solution processability, and low cost. Stacking two perovskite solar cells with complementary bandgaps in a monolithic tandem solar cell configuration, the highest certified efficiency of 26.4% has been achieved on glass substrate.<sup>1</sup> In addition to high-efficiency potential, all-perovskite tandems can be fabricated on flexible and lightweight substrates with very high power-to-weight ratios leveraging low-temperature solution-based coating methods and high throughput roll-to-roll manufacturing, thus significantly lowering the manufacturing cost and CO<sub>2</sub> footprint. Besides high efficiencies at low costs and low CO<sub>2</sub> emission, all-perovskite tandem photovoltaic technology also requires low resource consumption compared to c-Si solar cells, which makes them very attractive for lowcost TW-scale PV applications. So far, the efficiency of flexible all-perovskite tandems is lagging far behind their rigid counterparts. In this contribution, we report wide-bandgap (~ 1.77 eV) and narrow-bandgap (~1.24 eV) flexible perovskite solar cells with the highest efficiency enabled by interface engineering. We demonstrate over 24% monolithic all-perovskite tandem solar cells on flexible polymer foil. We will discuss the key challenges and pathways toward developing over 30% flexible all-perovskite tandem solar cells.



Fig. 1 FIB-SEM image of flexible all-perovskite tandem solar cells on flexible polymer foil.

<sup>&</sup>lt;sup>1</sup> Lin, R., Xu, J., Wei, M. et al. All-perovskite tandem solar cells with improved grain surface passivation. Nature 603, 73–78 (2022)

#### Bayesian parameter estimation for halide perovskite solar cells

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In the field of halide perovskites, a vast number of different optical and optoelectronic characterization techniques are used to better understand the key electronic properties of these materials such as mobilities, lifetimes, defect and doping densities as well as band offsets or other features of the band diagram. These techniques include steady state methods such as current or photoluminescence vs. voltage measurements, transient methods (transient photoluminescence, absorption or photoconductivity) or frequency domain methods (Impedance or intensity modulated photocurrent or photovoltage spectroscopy). Each of these methods cannot typically be analyzed by using one analytical equation that provides a single parameter. In fact, the analysis of each of these methods would require solving at least three coupled partial differential equations in time and space using numerical methods. The forward problem, i.e. going from the material and interface parameters to the outcome of an experiment (e.g. current-voltage curve) is typically simple and can be done by a variety of commercial or open access simulation tools. The inverse problem of going from the experimental data to the material parameters is challenging as it would involve doing many forward simulations until a suitable parameter set is found to represent the data. Often the inverse problem is solved by an attempt to fit the data, which has however many downsides such as the lack of insight into other solutions.

Here, we want to briefly mention two strategies to overcome the problem. The first strategy would be to highly simplify the mathematical problem and develop novel analytical equations that are not based on the equations used for pn-junction solar cells but that are adapted to the specific properties of halide perovskites. The second approach is based on Bayesian statistics.<sup>1-</sup> <sup>2</sup> The general workflow is as follows:

First a surrogate model is created that is trained with numerical simulation data that was created using a sensible range of material parameters. The numerical simulations will take a long time to perform, but for a set of similar samples one only has to perform these simulations once. Then they are stored in a database and used to train the neural network. The neural network will then store the information contained in the database in the weights of its nodes and will be able to release the information quite quickly. Subsequently, an algorithm has to sample the multidimensional parameter space that the neural network (surrogate model) has access to, to search for a sensible material parameter set to describe the experimental data. The typically used algorithm is a Markov Chain Monte Carlo (MCMC) algorithm that consists of a large number of

walkers that sample the parameter space. Each time a walker tests a particular set of parameters, the neural network will provide the resulting simulated experimental data and then calculate the likelihood that the material parameters correctly describe the experimental data. From this parameter maps can be created that show the correlation between parameters and show how good the confidence in the result is. A typical parameter map for a halide perovskite solar cell is shown in Figure 1.



**Fig. 1** Posterior probability distributions of conduction band offset, surface recombination velocity of ETL/perovskite interface, valence band offset and HTL mobility obtained from BPE. Also, the joint probability distribution of every two-parameter combination is shown to highlight how they correlate with each other. The brighter regions in these plots show that the two parameters are most likely to be in this region of the parameter space. The joint probability distribution of e.g. conduction band offset and surface recombination velocity at the perovskite/ETL interface shows a high correlation.

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Brandt, R. E.; Kurchin, R. C.; Steinmann, V.; Kitchaev, D.; Roat, C.; Levcenco, S.; Ceder, G.; Unold, T.; Buonassisi, T., Rapid Photovoltaic Device Characterization through Bayesian Parameter Estimation. *Joule* **2017**, *1* (4), 843-856.

### First Perovskite solar cell in consumer electronic devices Abstract for the SimOEP 2022 Conference

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Perovskia Solar is a Swiss-based cleantech startup. It is a spin-off of Empa, Dübendorf, providing digitally printed custom design solar cells for electronic, IoT devices & sensors.

#### ``Our vision is to enable a solar cell on every device``

Today we are surrounded by billions of power-hungry electronic devices needing constant charging or battery change. This pain is going to increase with the advent of industry 4.0 where we will have much more interconnected devices.

With our patented perovskite solar technology, we can simply print solar cells into any shape and size facilitating mass solar adoption into everyday electronic devices. Our technology enables energy-self-sufficient devices and makes them battery-free or extends battery life considerably.

Our solar cells are tuned to work even in low lighting conditions of homes and offices while having a 10x lower carbon footprint compared to the existing silicon-based technology. Moreover, they can be fully recycled or even re-furbished.

Last year we launched the first fully perovskite solar-powered consumer electronic device for a customer.





Fig. 1 First fully perovskite solar cell-powered consumer electronic device and custom design solar products from Perovskia Solar

Currently, we are working with 10+ market-leading global companies and have a robust pipeline of 4M plus solar cell price inquiries. Last year we launched the first consumer electronic device powered fully by a custom design perovskite solar cell. We are on the path to revolutionizing how we power our day-to-day life. We will be generating a billion watts with a billion beautiful custom-printed solar cells. Performance boosting polymeric finish layer for perovskite solar cells

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Polymeric finish layer (PFL) has been employed as a post treatment in perovskite solar cells to improve efficiency and stability. Polymeric finish layer as a post treatment consists of deposition of a polymer on top of the cell using nearly-saturated polymer solutions through a simple spin coating method without further treatments such as heat treatment. Using PFL treatment, it is possible to heal old cells and increase efficiency to even more than the initial efficiency through voltage and current density improvement. We were able to achieve 21.48% and 19.05% efficiency from cells with the gold and carbon as a back contact and initial efficiency of 16% and 14% respectively. Besides efficiency improvement, PFL treatment is able to improve stability. Not only polymeric finish layer on top of the cell is able to improve stability through being a weak encapsulation layer but also efficiency increases as a results of initial stability improvement. Also stability under reverse bias, which is a huge concern for perovskite solar cells, gets improved by PFL treatment allowing the treated cell to fully recover after stressing at -10 V. The effect of PFL treatment is investigated by transient electrical measurements, impedance spectroscopy and photo- and electroluminescence.

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2. H. Zhang, X. Ren, X. Chen, J. Mao, J. Cheng, Y. Zhao, Y. Liu, J. Milic, W.-J. Yin, M. Grätzel and W. C. H. Choy, *Energy & Environmental Science*, 2018, 11, 2253-2262.

3. T. Duong, J. Peng, D. Walter, J. Xiang, H. Shen, D. Chugh, M. Lockrey, D. Zhong, J. Li, K. Weber, T. P. White and K. R. Catchpole, *ACS Energy Letters*, 2018, 3, 2441-2448.

#### Physical pictures of the anomalous ionic-electronic phenomenology in perovskite solar cells by using electrical strategies and advanced mathematical tools

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Understanding the peculiar and puzzling phenomenology of perovskite-based solar cells is one of the most elusive issues prevalent in perovskite's community in the last years (e.g., complex transient behavior at long time scales, rate-dependent J-V hysteresis and/or non-ideal memory-based capacitive effects).<sup>1,2</sup> Here, we reevaluate the basic conceptual picture of anomalous ionic-electronic processes underlying the heterogeneous environment of perovskite solar cells. In this sense, fractional calculus emerges as the most adequate tool to model the cooperative relaxation phenomena at long time scales, which have a significant impact on the appearance of hysteresis (slow and anomalous memory-based hysteretic currents) and, more extensively, on the photovoltaic performance.<sup>3,4</sup> We analyze perovskite solar cells with different architectures, both in the dark and under illumination, using stepwise-JV measurements and Impedance Spectroscopy (IS). Our fresh interpretation establishes a key connection between the dynamic normal/inverted hysteresis, fractional-order capacitive/inductive responses, and device physics. Finally, we provide a discussion on the optimal configuration parameters in order to suppress reversible effects of hysteresis in device performance assessment (J-V modelling),<sup>5</sup> with the aim of improving reproducibility and comparability between research laboratories. Our study outlines key guidelines for further dedicated works to adequately interpret the characteristic ionic-electronic phenomenology and evaluate the device performance.

<sup>&</sup>lt;sup>1</sup> P. López-Varo, J.A. Jiménez-Tejada, M. García-Rosell, S. Ravishankar, G. Garcia-Belmonte, J. Bisquert, O. Almora, "Device physics of hybrid perovskite solar cells: Theory and experiment", Adv. Energy Mater. 8, 1702772, (2018).

<sup>&</sup>lt;sup>2</sup> W. Tress, N. Marinova, T. Moehl, S.M. Zakeeruddin, M.K. Nazeeruddin, M. Grätzel, "Understanding the rate-dependent J-V hysteresis, slow time component, and aging in CH<sub>3</sub>NH<sub>3</sub>PbI<sub>3</sub> perovskite solar cells: the role of a compensated electric field", Energy Environ. Sci. 8, 995, (2015).

<sup>&</sup>lt;sup>3</sup> E. Hernández-Balaguera, B. Romero, B. Arredondo, G. del Pozo, M. Najafi, Y. Galagan, "The dominant role of memory-based capacitive hysteretic currents in operation of photovoltaic perovskites", Nano Energy 78, 105398, (2020).

<sup>&</sup>lt;sup>4</sup> E. Hernández-Balaguera, G. del Pozo, B. Arredondo, B. Romero, C. Pereyra, H. Xie, M. Lira-Cantú, "Unravelling the key relationship between perovskite capacitive memory, long timescale cooperative relaxation phenomena, and anomalous *J-V* hysteresis", Solar RRL 5, 2000707, (2021).

<sup>&</sup>lt;sup>5</sup> E. Hernández-Balaguera, L. Muñoz-Díaz, C. Pereyra, M. Lira-Cantú, M. Najafi, Y. Galagan, "Universal control strategy for anomalous ionic-electronic phenomenology in perovskite solar cells efficiency measurements", Mater. Today Energy 101031, (2022).

### Simulation of the Perovskite Material Hysteretic Behavior during Capacitance-Voltage Analysis using SETFOS

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When it comes to capacitance techniques for perovskite semiconductor devices characterization, some important elements have to be looked at. One is the ionic nature of those devices and how it can translate into a hysteretic behavior when a sweep voltage is applied to it. This phenomenon is mainly studied in the voltage-current characterization but is also present during capacitance-voltage analysis and depend on different factors as voltage sweep rate and direction as well as pre-bias duration and voltage<sup>1</sup>. Therefore, Mott-Schottky method is hardly suitable for perovskite device information extraction as it is strongly prone to error<sup>2</sup>. To help overcome this issue, numerical tools can be used to achieve better interpretation of C-V data. However, unlike current-voltage measurements, it is necessary to consider both time and frequency domain.



**Fig. 1** Example of the hysteretic behavior that can be reproduced for capacitance measurement simulations. **(a)** Capacitance-Voltage and (b) Mott-Schottky plots simulated for three different voltage sweep rate.

<sup>&</sup>lt;sup>1</sup> Fischer et al., "Doping Profile in Planar Hybrid Perovskite Solar Cells Identifying Mobile Ions".

<sup>&</sup>lt;sup>2</sup> Almora et al., "On Mott-Schottky Analysis Interpretation of Capacitance Measurements in Organometal Perovskite Solar Cells".
In this communication we present how SETFOS software can be used to replicate the C-V hysteretic behavior of a perovskite MIS capacitor as displayed in Figure 1. This particular structure is designed to ensure parameter extraction as free charge carrier density. Such an approach allows approximating the level of ionic charge and their impacts on transient C-V characterizations.

#### Wave optics in differential absorption spectroscopy for thin-film systems

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Differential absorption spectroscopy techniques (such as transient absorption spectroscopy, photoinduced spectroscopy, and charge modulation spectroscopy etc.) serve as powerful techniques to study the excited species in semiconductors<sup>1,2</sup>. However, it has always been challenging to employ these techniques for characterizing relatively thick thin-film systems, especially when a reflective top contact is involved, like solar cells. In this work, a systematic study on how a combination of interference effect and non-uniform excited species distribution profile severely manipulates the experimental spectra and measured decay dynamics is presented based on experimental and simulation data. Furthermore, a practical methodology to correct these optical artifacts in differential absorption spectroscopies based on inverse transfer matrix is proposed and benchmarked. The results and proposed correction method generally apply for all kinds of differential absorption spectroscopy techniques and various thin film systems (organics, perovskites, kesterites, etc.). Strong spectral distortion due to optical artifacts are found as the active layer thickness is higher than 150 nm. This is nowadays additionally meaningful as this thickness matches the thickness range of industrial desired organic solar cells and most perovskite solar cells. In addition, while interference and cavity effects are considered to be fluence independent, we showed clearly how in certain conditions, such optical considerations can have an impact on measured decay dynamics. With the help of proposed correction formalism, the differential spectra and the decay dynamics can be characterized on the full device of thin film solar cells in transmission mode and yield accurate and reliable results to provide design rules for further progress.

<sup>&</sup>lt;sup>1</sup> A.Y.B. Meneau, Y. Olivier, T. Backlund, M. James, D.W. Breiby, J.W. Andreasen, H. Sirringhaus, Temperature dependence of charge localization in high-mobility, solution-crystallized small molecule semiconductors studied by charge modulation spectroscopy, Adv. Funct. Mater. 26 (2016) 2326–2333.



**Fig. 1** Example of optical artifact distorted differential absorption spectrum (left panel) and the corresponding change in refractive index and extinction coefficient with optical artifacts corrected (right panel).

<sup>&</sup>lt;sup>2</sup> N.F. Zorn, F. Scuratti, F.J. Berger, A. Perinot, D. Heimfarth, M. Caironi, J. Zaumseil, Probing Mobile Charge Carriers in Semiconducting Carbon Nanotube Networks by Charge Modulation Spectroscopy, ACS Nano. 14 (2020) 2412–2423.

### Optical Coupling of Internal Radiation in Perovskite-Silicon Tandem Cells and its Impact on Optoelectronic Device Characteristics

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In the recent years, metal halide perovskite materials have emerged as promising candidates for single-junction solar cells with efficiencies exceeding 25% power conversion efficiency (PCE) [1]. To go even further, combining different absorber materials in a single tandem solar cell yields the advantage of a more efficient utilization of the energy in solar radiation. In this regard, metal halide perovskites proved furthermore useful as thin-film top cells combined with well-established silicon solar cells as bottom cells, yielding PCEs of >29% [2].

While this type of tandem cell is already intensively being assessed experimentally, and even efforts for commercialization are being made, the theoretical treatment has been mainly focused on the optical modelling of the absorption of the incident solar radiation to achieve good current matching. In order to improve the understanding and optimize the design process, the electronic characteristics as well as the fate of internally emitted light from the perovskite absorber should furthermore be taken into account as well. The latter consists of two aspects, reabsorption in the perovskite absorber itself (photon recycling) on the one hand, and reabsorption in the silicon bottom cell (luminescent coupling) on the other. One difficulty in modelling such a system are the vastly different length scales of the thin top and thick bottom cell, such that the optics of the top cell require a rigorous wave optical treatment, while optical coherence is lost in the bottom cell and a ray-optical treatment is more appropriate. To this end, we present an extension of our recently presented dyadic Green's function model [3] to handle also thick and optically incoherent layers as present in perovskite-silicon tandem cells, going beyond the application on single-junction perovskite devices [4].

As an example system, in which the role of photon recycling and luminescent coupling on device performance is assessed, we use a tandem structure consisting of a 420 nm thick CH3NH3PbI3 (MAPI) top cell and a silicon heterojunction solar cell as bottom cell, consisting of a 280  $\mu$ m thick n-doped crystalline silicon (c-Si) wafer, similar to a structure recently analysed with a theoretical PCE of 30.4% [5]. The optical dyadic Green's function based

model is coupled to a well established drift-diffusion-Poisson solver [6] for a self-consistent treatment of optical and electronic characteristics, including non-idealities such as non-radiative recombination in the sub cells and a lossy interconnection through a recombination junction. Such a fully coupled model allows for an in-depth evaluation of the impact of photon recycling and luminescent coupling on the optoelectronic device characteristics.



Fig. 1: Left: Electrical and optical stack of the perovskite-silicon tandem cell. Right: Change in JV characteristics due to internal coupling.

- J. Jeong et al. Pseudo-halide anion engineering for α-FAPbI3 perovskite solar cells. Nature 592, 381 (2021).
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## Impact of photon recycling on the light extraction from metal halide perovskite light emitting diodes

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In contrast to OLED emitter materials, many of the metal halide perovskite materials used in the emerging field of perovskite LEDs (PeLEDs) exhibit steep band edge and small Stokes shift resulting in sizable re-absorption of the internal emission. While light re-absorbed in outcoupled modes represents a loss process, re-emission following re-absorption in guided modes can be beneficial for light extraction, as light is re-distributed to out-coupled modes<sup>1</sup>. Being an internal process, such photon recycling (PR) cannot directly be assessed via experiments. Quantification of the potential gain in external quantum efficiency due to reabsorption therefore needs to rely on realistic models of internal and external emission that take into account any re-absorption events, including parasitic absorption losses and non-radiative recombination.



**Fig. 1** (a) Level alignment of the PeLED stack used in the electrical simulation. (b) Effect of PR on current-voltage characteristics and emitted flux. (c) Spatial evolution of the photon flux @1.45 V with and w/o PR.

In our contribution, we present a simulation analysis of published PeLED structures based on methylammonium lead iodide (MAPI)<sup>2</sup> (stack shown in Fig.1a) that builds on a recently de-

<sup>&</sup>lt;sup>1</sup> C. Cho et al., "The role of photon recycling in perovskite light-emitting diodes", Nat. Commun., vol. 11, p. 611 (2020).

<sup>&</sup>lt;sup>2</sup> K. Elkhouly et al., "Operationally Stable Perovskite Light Emitting Diodes with High Radiance", Adv. Optical Mater., vol. 9, art. no. 2100586 (2021).

veloped model for dipole emission in media with strong self-absorption<sup>3</sup>. The local rates for emission and re-absorption are computed based on a Green dyad model that considers the local values of photon density of states and carrier densities, and are then used in the full opto-electronic simulation with SETFOS<sup>4</sup> that considers the actual level alignment as well as charge carrier mobilities and electrode configuration<sup>5</sup>. This provides a fingerprint of PR in both, current-voltage characteristics and radiance (Fig.1b), revealing a sizable enhancement of the emitted photon flux (Fig. 1c).

<sup>&</sup>lt;sup>3</sup> U. Aeberhard, S. Zeder, B. Ruhstaller, "Reconciliation of dipole emission with detailed balance rates for the simulation of luminescence and photon recycling in perovskite solar cells", Opt. Express, vol. 29, p. 14773 (2021).

<sup>&</sup>lt;sup>4</sup> Setfos by Fluxim AG, version 5.2, www.fluxim.com/setfos.

<sup>&</sup>lt;sup>5</sup> S. Zeder, B. Ruhstaller, U. Aeberhard, "Assessment of Photon Recycling in Perovskite Solar Cells by Fully Coupled Optoelectronic Simulation", Phys. Rev. Appl., vol. 17, p. 014023 (2022).

# Simulating the transient luminescence of perovskite light-emitting diodes under pulsed operation

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In perovskite light-emitting diodes (PeLEDs), the slow response of mobile ions can have an important effect under pulsed operation. In a study by Kumawat et al.<sup>1</sup>, the operation of near infra-red (NIR) PeLEDs was investigated under pulsed operation in the frequency range of 10 Hz up to 20 kHz. The transient electroluminescence (TrEL) of highly efficient devices was measured with varying pulse widths (duty cycle). The TrEL signal was found to show pulse-width dependent features attributed to the characteristic response times of mobile ions. It showed a constant plateau value after turn-on, with lower intensity for lower duty cycles. A large TrEL overshoot was observed at the end of the voltage pulse – which decreased and eventually disappeared with increasing duty cycle. Such TrEL overshoot was previously reported for perovskite solar cells and attributed to ionic accumulation effects<sup>2</sup>.

In the follow-up simulation study presented in this talk, a model built in the drift-diffusion simulation software Setfos 5.2 (Fluxim) has been used to better understand the role of mobile ions in the TrEL response of PeLEDs under pulsed operation. The results obtained can explain the duty-cycle dependance of the TrEL signal and confirm the slow response of mobile ions as a governing factor, which is important to consider for the development of PeLED-based technologies.

<sup>&</sup>lt;sup>1</sup> N. K. Kumawat, W. Tress, and F. Gao, 'Mobile ions determine the luminescence yield of perovskite lightemitting diodes under pulsed operation', *Nat. Commun.*, vol. 12, no. 1, p. 4899, Aug. 2021, doi: 10.1038/s41467-021-25016-5.

<sup>&</sup>lt;sup>2</sup> R. Gegevičius, M. Franckevičius, J. Chmeliov, W. Tress, and V. Gulbinas, 'Electroluminescence Dynamics in Perovskite Solar Cells Reveals Giant Overshoot Effect', *J. Phys. Chem. Lett.*, vol. 10, no. 8, pp. 1779–1783, Apr. 2019, doi: 10.1021/acs.jpclett.9b00618.

#### **Dynamics Due to Ions and Traps in Perovskite LEDs**

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After an extraordinary rise in power conversion efficiency, perovskite thin film technology is reaching the performance of silicon photovoltaics, and similarly the external quantum efficiency (EQE) of perovskite light-emitting diodes (PeLEDs) has surpassed 20% for green PeLEDs and longer wavelengths<sup>1</sup>. However, in view of the commercialisation of both technologies, it is imperative to increase the operational stability. Especially for PeLEDs, it is rather low with best performances of a few days until the EQE drops below 50% of its maximum<sup>1</sup>. Ionic point defects - mobilised by high temperature, irradiation and external electric fields - contribute to the intrinsic device instability. Dynamic processes due to mobile ions and electronic charge traps within the perovskite layer and at the interfaces are well studied individually, but their interplay has still to be further unravelled. Here, transient optoelectronic measurements of a CsPbBr<sub>3</sub> based PeLED reveal an emission feature within the time range of ionic mobility, related to an often-observed initial rise of EQE during operation<sup>2</sup>. Transient drift-diffusion simulation is able to describe the observed emission increase as a result of ionic movement. The subsequent emission drop is not reproduced by the simulation, and the missing model part is discussed.

Furthermore, an optical model based on changes of the spontaneous emission due to optical field surrounding the excited state is investigated. Theoretically, the outcoupling efficiency for a given Langevin recombination current drastically changes for different emitter positions within the active layer as shown in Fig. 1. It is very relevant to answer the question whether the model is as applicable to PeLEDs as it is to organic emitters<sup>3,4</sup>, because essentially every process that alters the space charge distribution, such as ion movement or trapping, can have

<sup>&</sup>lt;sup>1</sup> A. Fakharuddin, M. K. Gangishetty, M. Abdi-Jalebi, S.-H. Chin, A. R. bin Mohd Yusoff, D. N. Congreve, W. Tress, F. Deschler, M. Vasilopoulou, H. J. Bolink, Nat. Electron. 2022, 5, 203.

<sup>&</sup>lt;sup>2</sup> N. Li, Y. Jia, Y. Guo, N. Zhao, Adv. Mater. 2022, 34, 2108102.

<sup>&</sup>lt;sup>3</sup> R. R. Chance, A. Prock, R. Silbey, Advances in Chemical Physics (Eds.: Prigogine, I.; Rice, S. A.), Vol. 162, John Wiley & Sons, Inc., Hoboken, NJ, USA, 1978.

<sup>&</sup>lt;sup>4</sup> M. Diethelm, A. Schiller, M. Kawecki, A. Devižis, B. Blülle, S. Jenatsch, E. Knapp, Q. Grossmann, B. Ruhstaller, F. Nüesch, R. Hany, Adv. Funct. Mater. 2020, 30, 1906803.

additionally a dynamic component in the emission characteristic due to the movement of the emitter position. So far, there is little literature investigating this effect in PeLEDs<sup>5</sup>. Here, thickness variation of the evaporated TPBi transport layer of a CsPbBr<sub>3</sub> based PeLED indicates the validity of the optical model, but also shows the experimental complexity to unravel this purely optical effect.



**Fig. 1** Optical simulations of the current efficiency at a 0° observer angle for a perovskite LED. The dependence of out-coupled emission on active layer thickness and emitter position is shown for a constant recombination current.

<sup>&</sup>lt;sup>5</sup> L. Zhao, K. M. Lee, K. Roh, S. U. Z. Khan, B. P. Rand, Adv. Mater. 2019, 31, 1805836.

### Controlling Spontaneous Orientation Polarization in Organic Semiconductors

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Molecular orientation in organic semiconductors has many different facets, such as optical anisotropy leading to birefringence, alignment of their optical transition dipole moments (TDMs) and/or their permanent electrical dipole moments (PDMs)<sup>1</sup>. While TDM alignment is well established and readily exploited to improve light outcoupling in organic light-emitting diodes (OLEDs)<sup>2</sup>, PDM alignment and the resultant spontaneous orientation polarization (SOP) is by far less investigated, and it is not fully clear yet if it is beneficial or detrimental for device application<sup>3</sup>.

SOP is commonly observed in evaporated neat films of polar organic molecules and leads to a so-called giant surface potential (GSP), which can be measured by Kelvin probe, impedance spectroscopy or the displacement current method<sup>4</sup>. In general, the GSP slope is roughly proportional to the magnitude of the molecule's PDM (see Fig. 1). However, there are some points to be kept in mind: First, despite their very high PDM, some molecules exhibit vanishing SOP because they aggregate in pairs with antiparallel alignment, effectively cancelling out their dipole moments. Second, most of the known materials exhibit positive GSP slope with few exceptions only. And third, the order parameter (i.e., the degree of net PDM alignment) is typically less than 10%, with some recent exceptions, like BCPO having a record-high 30% PDM alignment<sup>5</sup>.

In this presentation, we review the current understanding of molecular orientation mechanisms, including factors like molecular shape, surface equilibration and intermolecular interactions. We report on the effects that substrate temperature during film growth, dilution in host matrices and details of the chemical structure have on SOP. Experimental results are

<sup>&</sup>lt;sup>1</sup> A.J. Hofmann, M. Schmid, W. Brütting; Advanced Optical Materials (2021); DOI: 10.1002/adom.202101004 <sup>2</sup> T. D. Schmidt, T. Lampe, D. Sylvinson M. R., P. I. Djurovich, M. E. Thompson, W. Brütting; Phys. Rev. Applied 8 (2017) 037001; DOI: 10.1103/PhysRevApplied.8.037001

<sup>&</sup>lt;sup>3</sup> Y. Noguchi, Y. Tanaka, H. Ishii, W. Brütting; Synth. Metals (2022) accepted

<sup>&</sup>lt;sup>4</sup> Y. Noguchi, W. Brütting, H. Ishii; Jap. J. Appl. Phys. 58, SF0801 (2019); DOI: 10.7567/1347-4065/ab0de8

<sup>&</sup>lt;sup>5</sup> B.A. Naqvi, M. Schmid, E. Crovini, P. Sahay, T. Naujoks, F. Rodella, Z. Zhang, P. Strohriegl, S. Bräse, E. Zysman-Colman, W. Brütting; Frontiers in Chemistry 8, 00750 (2020); DOI: 10.3389/fchem.2020.00750

linked to quantum chemical calculations to identify the impact of molecular conformers and non-bonding interactions on molecular alignment and the resulting SOP of a film.



**Fig. 1** GSP slope of evaporated neat films of some prototypical organic semiconductors (taken from Ref. 1, with BCPO added from Ref. 5).

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# Key optical properties of hyperfluorescent TADF OLEDs based on the v-DABNA fluorescent emitter.

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The photophysics of the multiple resonance TADF molecule v-DABNA<sup>1</sup> will be described. We show coupling of a 285 cm<sup>-1</sup> stretching/scissoring vibrational mode of the peripheral phenyl rings to the S<sub>1</sub> state. This leads to hot vibrational states emitting at room temperature that dictates the ultimate emission linewidth. However, a separate high amplitude mode, 945 cm<sup>-1</sup> of the N-biphenyl units mediates the rISC mechanism. A monotonic increase of the exciton emission linewidth with temperature indicates the role of hot transitions from vibrational excited states at RT, and combined with an observed temperature dependency of  $\Delta E_{ST}$  we deduce that the rISC mechanism is thermally activated rIC of T<sub>1</sub> to T<sub>N</sub> (n  $\geq$  2) followed by rapid rISC of T<sub>N</sub> to S<sub>1</sub>, i.e. upper state triplet crossing mediates rISC not second order vibrational coupling as in D-A TADF systems<sup>2</sup>. Thus, the rISC rate, between 3- $6x10^5$  s<sup>-1</sup> measured in both different solvents and solid hosts, is independent of environment, relatively slow leading to triplet polaron quenching at moderate drive currents and strongly temperature dependent even though the measured  $\Delta E_{ST}$  is effectively zero at RT.<sup>3</sup>

Concentration dependence studies in solution and solid state reveals a second emission band that increases nonlinearly with concentration, independent of environment assigned to excimer emission. Even at concentrations well below those used in devices, the excimer contribution affects performance. At 20K over the first 10 ns, we observe a broad Gaussian excimer emission band with energy on-set above the S<sub>1</sub> exciton band. An optical singlet triplet gap ( $\Delta E_{ST}$ ) of 70 meV is measured, agreeing with previous thermal estimates, however the triplet energy is also found to be temperature dependent. We discuss the trade-off between narrowing the emission linewidth through further molecular rigidification and removal of free periphery against the need for vibrionic coupling to mediate the rIC mechanism and prevent excimer formation.

TADF sensitised OLEDs with v-DABNA as a hyperfluorescent emitter (0.5 wt% and 1 wt%) exhibit an increase of maximum EQE, reaching 27.5% for the lower v-DABNA concentration. On the contrary, a Förster radius analysis indicated that the energy transfer ratio is smaller due to higher donor-acceptor separation (>2.4 nm) with weak sensitizer emission observed in the electroluminescence. This indicates excimer quenching in the 1 wt% devices. We also noticed that when we used DMAC TRZ as a sensitizer for v-DABNA, we managed to generate pure blue light, from DMAC TRZ green emission. After looking hard we found something rather strange, a second high energy emitting state in DMAC TRZ, which is a weak charge transfer state, and looks very much like an axial conformer, i.e. the donor is folded out of the plane of the acceptor compared to the more usually rotated through 90° twisted equatorial conformer that gives rise to the strong green CT state. So, can these high energy conformers in DMAC TRZ sensitization v-DABNA to give efficient pure blue emission. Maybe other systems can do this too?

#### Acknowledgment

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# Kinetic Monte Carlo Simulations of Degradation Scenarios in Thermally Activated Delayed Fluorescence Organic Light Emitting Diodes

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The lifetime of organic light-emitting diodes (OLEDs) is a limiting factor in the expansion of this technology to new applications, even with the use of a relatively new type of emitter materials that utilize thermally activated delayed fluorescence (TADF) to enhance efficiency. Device degradation is typically characterized by the time-dependence of the luminance decay and the voltage shift when operated at constant current. However, such measurements do not reveal the underlying mechanisms responsible for the degradation, nor where in the stack the degradation occurs, which degradation products are forming and how they impact the device operation on a microscopic scale.

In this work,<sup>1</sup> we present a workflow using three-dimensional kinetic Monte Carlo (KMC) simulations<sup>2</sup> to explicitly simulate the operational lifetime of OLEDs for different degradation scenarios. The simulations include an equilibration phase, an equilibrated pristine state phase and finally the degradation phase. The explicit simulation of the relevant time scales within a feasible simulation time is made possible by (i) the continuation of the simulation from the same equilibrated state assuming different degradation scenarios, (ii) the use of accelerated degradation as a result of higher current densities  $J_0$  and the subsequent extrapolation of the lifetime to the desired, lower values of  $J_0$  and (iii) the acceleration of degradation by assuming a higher degradation probability per degradation-triggering event in the simulation than is observed in experiment.<sup>3</sup>

<sup>&</sup>lt;sup>1</sup> C. Hauenstein, S. Gottardi, E. Torun, R. Coehoorn, H. van Eersel, "Identification of OLED Degradation Scenarios by Kinetic Monte Carlo Simulations of Lifetime Experiments", Front. Chem. 9, 823210 (2022) <sup>2</sup> We use the KMC simulation software tool Bumblebee from Simbeyond B.V. (https://simbeyond.com/)

<sup>&</sup>lt;sup>3</sup> N. C. Giebink, B. W. D'Andrade, M. S. Weaver, P. B. Mackenzie, J. J. Brown, M. E. Thompson, S. R. Forrest, "Intrinsic luminance loss in phosphorescent small-molecule organic light-emitting devices due to bimolecular annihilation reactions", J. Appl. Phys. 103, 044509 (2008)

We demonstrate our approach for an experimentally well-characterized green-emitting TADF OLED, for which the functioning in the pristine state was studied in an earlier KMC study.<sup>4</sup> The degradation scenarios we investigate vary with regards to the materials that are taken to be degrading, and with regards to the assumed properties of the degradation products resulting from those materials. Degradation is triggered by exciton-polaron quenching and exciton-exciton annihilation. Fig. 1 shows for four such scenarios a comparison of the experimental luminance and the simulated radiative decay rate *R*, including the operational lifetime "LT50", after which *R* has decreased to half its initial value. The comparison provides a prediction for  $P_{deg}$ , the probability that a degradation-triggering event leads to the formation of a degraded molecule.  $P_{deg}$  is for all scenarios found to be  $(0.2-0.7) \times 10^{-9}$ , which is consistent with the experimental approximation for other OLED materials in Ref. [3]. Further analysis of the different scenarios reveals that degradation of the emitter dominates the luminance decay, while transport layer degradation at the interfaces with the emitting layer is responsible for the observed voltage increase.

The workflow can be extended to a variety of device architectures and other degradation scenarios that might be envisioned and is expected to enable systematic *in silico* studies of the operational lifetime and its sensitivity to the material composition, layer structure, charge carrier balance, and the use of refined device principles such as hyperfluorescence.



**Fig. 1** a) Simulated decay of the radiative rate *R* for four degradation scenarios, with black crosses marking R = 0.5 ("LT50"). b) Voltage shift for a constant current density of  $J_0 = 19$  Am<sup>-2</sup>.

<sup>&</sup>lt;sup>4</sup> S. Gottardi, M. Barbry, R. Coehoorn, H. van Eersel, "Efficiency loss processes in hyperfluorescent OLEDs: A kinetic Monte Carlo study", Appl. Phys. Lett. 114, 109901 (2019)

# Influence of spontaneous orientation polarization in TADF organic light emitting diodes

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The efficiency of Organic semiconductor - based Light Emitting Diodes (OLEDs) is highly dependent on molecular orientation and the dielectric properties of organic films. Spontaneous Orientation Polarization (SOP) plays an important role in governing the charge injection and accumulation properties in an OLED with evaporated organic materials<sup>1</sup> with Permanent Dipole Moment (PDM). The SOP induces a polarization charge at the hetero interfaces of the stacked multilayer devices<sup>2</sup>. SOP is commonly observed in host materials for Thermally Activated Delayed Fluorescence (TADF) OLEDs and strongly correlates with device degradation<sup>2</sup> thus making it crucial to investigate further.

We investigate TADF OLEDs with host materials having considerable PDM and study the effect of doping a TADF emitter in these polar hosts thus analysing the effect of SOP on TADF OLEDs. Varying doping percentages of emitter in the guest host system attributes to steady dilution of the host films with increasing TADF emitter concentration thus resulting in a change of the SOP. Photoluminescence Quantum Efficiency (PLQE) is measured using combined electrical and optical experiments and external quantum efficiency is determined from Angular Dependant Electroluminescence Spectroscopy (ADEL) to analyse the device performance upon varying the SOP. We observe a reduction in SOP of the host materials with increasing doping concentration of our TADF emitter. The EQE and the PLQE of the device also reduces with higher doping percentage corresponding to the lower SOP making it an important parameter for TADF OLEDs.

<sup>&</sup>lt;sup>1</sup> Kohei Osada et al 2018 Org. Electronics. Vol 58, July 2018, Pg 313-317

<sup>&</sup>lt;sup>2</sup> Yutaka Noguchi et al 2019 Jpn. J. Appl. Phys. 58 SF0801

# Rotational model for description of the photophysics of thermally activated delayed fluorescence emitters with donor-acceptor structure

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Thermally activated delayed fluorescence (TADF) phenomenon has gained great attention lately thanks to its high potential for application in all-organic OLEDs. The most efficient TADF materials known to date, have donor-acceptor structure which is beneficial for both charge recombination in OLEDs and TADF efficiency. Taking into account rather short period of extensive investigation of TADF materials, their mechanism of triplet harvesting remains poorly understood.

Among several theoretical models developed recently to explain mainly reverse intersystem crossing (rISC) under specific conditions, we proposed a rotational model which describes ISC, rISC and the rate of radiative deactivation in media of different polarity and viscosity<sup>1,2</sup>. Minor deviations from the optimal geometry, namely rotations along the  $\sigma$ -bond connecting donor and acceptor, provide various rotamers with substantially different rISC and fluorescence rates, which explains complex spectral behavior of TADF emitters. This model was successfully tested on various emitters including blue and red ones, correctly describes TADF dependence on polarity of liquid solutions and photophysics in amorphous films. Due to the ease of visualization of specific molecular rotations or vibrations responsible for rISC enhancement, this model emphasizes new directions for improvement of TADF materials.

The described model reveals that due to the existence of such rotamers, the direct spinflip between the excited states of the same charge transfer nature but different multiplicity

<sup>&</sup>lt;sup>1</sup> I. E. Serdiuk, M. Mońka, K. Kozakiewicz, B. Liberek, P. Bojarski, S. Y. Park. Vibrationally Assisted Direct Intersystem Crossing between the Same Charge-Transfer States for Thermally Activated Delayed Fluorescence: Analysis by Marcus–Hush Theory Including Reorganization Energy. J. Phys. Chem. B, 2021, 125, 10, 2696.
<sup>2</sup> M. Mońka, I.E. Serdiuk, K. Kozakiewicz, E. Hoffman, J. Szumilas, A. Kubicki, S.Y. Park, P. Bojarski. Understanding the internal heavy-atom effect on thermally activated delayed fluorescence: application of Arrhenius and Marcus theories for spin–orbit coupling analysis, J. Mater. Chem. C, 2022, 10, 7925.

 $({}^{1}CT \leftrightarrow {}^{3}CT)$  can be highly efficient. In spite of being "forbidden" by the selection rules, in some rotamers such a transition can occur even in sub-nanosecond regime. On the other hand, more "allowed" interaction of singlet  ${}^{1}CT$  state with a triplet localized on a donor or acceptor fragment ( ${}^{3}LE$ ) was found to be almost insensitive to the rotational isomerism, but responsible mainly for the ISC rate.

This presentation summarizes experimental evidences providing to the rotational model, its theoretical bases, examples of emitters used for its verification.

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# Temperature dependent dynamics of reversible electron traps in polymer light-emitting diodes

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Semiconducting polymers are intensively studied for opto-electronic device applications, including solution-processed polymer light-emitting diodes (PLEDs). Although the first PLED has been already introduced in 1990, there are remaining problems to be solved for PLEDs in terms of device stability and efficiency. One of the challenges is posed by the presence of ubiquitous deep electron trap states in conjugated polymers. Electron traps lower the electron mobility. This leads to a mismatch between the hole and electron carrier mobility and results in a shift of the emission zones closer to the cathode. Also, trap states act as non-radiative recombination centres which reduces the electrical efficiency.

Recently (Adv. Funct. Mater. 2022, doi:10.1002/adfm.202106185), we observed that filling of deep electron traps proceeds over many minutes, clearly in contrast to the general notion that trap filling occurs on a timescale of a few hundreds of microseconds or less, irrespective of the depth of the trap level. Furthermore, electrons traps decay when the PLED is turned off. As possible origin, water-oxygen complexes were proposed as a cause for trapping, but proof is missing. We believe that the observed slow electron trap formation is consistent with a diffusion process between water and oxygen precursor trap species. The neutral water-oxygen complex is weakly bound, but the complex is stable and forms a trap when it is reduced by an electron. After device turn-off, electrons de-trap thermally and water and oxygen separate again via diffusion: this explains why electron traps are reversible.

If this scenario of electron trap formation and decay is correct, it can be assumed that the trap dynamics is influenced by the measurement temperature, since the diffusion of water and oxygen is strongly temperature dependent. Here we drive PLEDs based on a polymer called superyellow (a PPV derivate) at different temperatures and current densities and investigate the formation and dissociation of the deep electron trap sites. In addition, we use drift-diffusion simulation to calculate the number of trap states present as a function of time. We find that the absolute increase in voltage over time for a given temperature – though indicative of trap formation – also strongly depends on the specific charge carrier mobilities and injection barriers. This leads to the largest voltage increase at 300 K over 5 minutes, while the simulated number of formed traps increases monotonously with temperature (see Figure). Below a current density of 20 mA/cm<sup>2</sup>, the limiting factor for the initial trap formation is the availability of free electrons.



Figure: Experimental voltage trends for constant current (10 mA/cm<sup>2</sup>) driven PLEDs at different temperatures (left). Simulated voltage trends (middle) and resulting electron trap densities (right).

# Using the weak microcavity to understand the light-emitting electrochemical cell

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The thin-film and planar architecture of electroluminescent devices such as the light-emitting electrochemical cell (LEC) and the organic light-emitting diodes (OLED) make them highly sensitive to microcavity effects. In OLEDs, the microcavity can be carefully designed to maximize the light outcoupling by fabricating a multilayered stack with the emission layer at its optical maximum position. This microcavity control is however more challenging in the LEC due to its in situ formation of an emissive p-n junction by electrochemical doping when a voltage is applied. In this presentation, we will show how to take advantage of microcavity effects to extract important parameters of a polymer-based LEC. First, we show that it is possible to shift the emissive p-n junction to a preferred position by the straightforward inclusion of an easily tuned additive in the active material<sup>1</sup>. Second, we show that the position of the emissive p-n junction can shift with time during LEC operation. Third, we discuss how the width and the dipole orientation of the emissive p-n junction can be determined and tracked over time with a device at its optical interference minimum. Finally, we will discuss how these measurements can be used to better understand the physics behind the LEC operation as well as to improve its performance.



**Fig 1.** Angular-resolved electroluminescence and emission profile determination of LECs devices. (a) Polar plot of the experimental angular radiant intensity of the LEC for the s-polarized light (solid circles) and the corresponding best fit (crosses). (b) Sketch of the simulated best-fit LEC doping profile and emission zone parameters (the position  $x_0$  and full width at half maximum, *FWHM*), as derived from the s-polarized data.

<sup>&</sup>lt;sup>1</sup> J. Ràfols-Ribé, X. Zhang, C. Larsen, P. Lundberg, E. M. Lindh, C. T. Mai, J. Mindemark, E. Gracia-Espino, L. Edman, Adv. Mater. 2021, DOI: 10.1002/adma.202107849e2107849.

#### Controlling the emission zone in light-emitting electrochemical cells

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The light-emitting electrochemical cell (LEC) is an emerging electroluminescent technology, which can feature bright area emission and which can be fabricated with low-cost printing and coating methods on flexible substrates. This makes the LEC suitable for a broad range of applications, including illumination, healthcare and signage.

The LEC consists of an active layer, comprising an organic semiconductor and an electrolyte, sandwiched between a transparent and a reflective electrode. During operation, electrochemical p- and n-type doping form in the active layer, and the light emission originates from the thin intrinsic p-n junction region. The position of this thin emission zone in the active layer directly influences the LEC efficiency because of constructive and destructive interference and exciton quenching by nearby dopants and electrodes. It is thus desirable and important to be able to control the position of the emission zone in LEC devices.

We have recently shown that the addition of designed additives into the active layer can shift the position of the emission zone through the concomitant introduction of electron and/or hole traps, which modify the mobility of the electrons and/or holes<sup>1</sup>. We have also recorded preliminary data that indicate that the driving current density also can have a strong influence on the emission zone position, and speculate that the origin could be a different field-dependent mobility for the electrons and holes.

<sup>&</sup>lt;sup>1</sup> Rafols-Ribe, J., Zhang, X., Larsen, C., Lundberg, P., Lindh, E. M., Mai, C. T., . . . Edman, L. (2022). Controlling the Emission Zone by Additives for Improved Light-Emitting Electrochemical Cells. Adv Mater, 34(8), e2107849. doi:10.1002/adma.202107849

# Charge carrier dynamics and exciton-polaron quenching studied by simultaneous measurement of displacement current and photoluminescence

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Charge accumulation at the organic hetero interfaces significantly affects the device properties of stacked multi-layer organic light-emitting diodes (OLEDs)<sup>1,2</sup>. Particularly, excitonpolaron quenching (EPQ) induced by the accumulated charge at the interfaces near the emission layer (EML) would be a main factor for the efficiency roll-off and device degradation<sup>3,4</sup>. In this study, we introduce a simultaneous measurement of displacement current and photoluminescence (PL) intensity (DCM-PL) to investigate the correlations between charge carrier dynamics and EPQ.

Fig. 1a shows the experimental setup for the DCM-PL. The current and PL intensity are simultaneously measured during application of a triangular wave voltage. A typical DCM-PL characteristics of a metal-insulator-semiconductor (MIS) device incorporating a partial structure of an Ir(ppy)<sub>3</sub>-based OLED (Fig. 1a) is shown in Fig. 1b. Hole accumulation behaviors and corresponding PL losses due to EPQ are clearly observed. The broad current curve in the backward sweep (the top panel in Fig. 1b) indicates the presence of a significant energy barrier for hole extraction at the EML/TCTA interface. Meanwhile, the PL intensity shows a hysteresis resulting from the delay of the hole evacuation from the EML. The DCM-PL combined with device simulation allows us to investigate not only the EPQ rate constant but also the detailed charge distribution around the EML. In this presentation, we will also discuss the influences of spontaneous orientation polarization (SOP)<sup>1,2</sup> on the charge carrier dynamics and EPQ (Fig. 1c).

<sup>&</sup>lt;sup>1</sup> Y. Noguchi, W. Brütting, and H. Ishii, "Spontaneous orientation polarization in organic light-emitting diodes", Japanese Journal of Applied Physics 58, SF0801, (2019)

<sup>&</sup>lt;sup>2</sup> Y. Noguchi, Y. Tanaka, H. Ishii, and W. Brütting, "Understanding spontaneous orientation polarization of amorphous organic semiconducting films and its application to devices", under review.

<sup>&</sup>lt;sup>3</sup> J. S. Bangsund, J. R. Van Sambeek, N. M. Concannon, and R. J. Holmes, "Sub-turn-on exciton quenching due to molecular orientation and polarization in organic light-emitting devices", Sci. Adv. 6, eabb2659, (2020).
<sup>4</sup> Y. Noguchi, H. Kim, R. Ishino, K. Goushi, C. Adachi, Y. Nakayama, and H. Ishii, "Charge Carrier Dynamics"

and Degradation Phenomena in Organic Light-Emitting Diodes Doped by a Thermally Activated Delayed Fluorescence Emitter", Org. Electron., 17, 184–191, (2015).



**Fig. 1** (a) Schematic illustration of the experimental setup for DCM-PL and the device structure under study, that is, a hole only MIS device with an emission layer (EML) of CBP or TPBi doped with Ir(ppy)<sub>3</sub>. (b) DCM-PL characteristics of the MIS device (TPBi host) at a sweep rate of 1 V/s. (c) Simulated DCM-PL characteristics with and without assuming SOP. The DCM curves (top panel) and charge distributions in the device were calculated by Setfos 5.2.

#### Multi-Scale Modeling of Charge Transport in Organic Electronic Materials

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The mobility of charge carriers in organic semiconductors, as in organic molecules or polymers, is an important metric for the performance of devices such as organic photovoltaics, light-emitting diodes, field-effect transistors, etc. The organic materials can be in semicrystalline or amorphous states, but generally transport involves thermally activated hops between molecular sites. To simulate charge transport and evaluate mobility in disordered organic materials, we need modelling that spans across different length scales. Specifically, it requires the combination of realistic models for the atomistic structure (arrangements, domains), the electronic interactions (site energies, transport parameters), the micro-electrostatic interactions (partial charges, self-consistent polarization screening), and stochastic processes (charge hopping). We have developed such a multiscale methodology, involving hierarchical simulations using density functional theory, molecular dynamics, and kinetic Monte Carlo, and applied it on several molecules typically of interest in organic photovoltaics to extract the mobility as a function of temperature and electric field<sup>1</sup> (see e.g., Fig. 1). This modelling activity is a smaller part of a more comprehensive simulation platform being developed by the EU project MUSICODE<sup>2,3</sup>, which includes also mesoscale and continuum simulations for microstructure evolution, printing and gas phase processing simulation, and device simulation. The overall structure and ambitions of the project MUSICODE are also going to be discussed at the end of this talk.



Fig. 1 Mobility calculation results for crystalline (top) and amorphous (bottom) IDIC.

<sup>1</sup>M. Andrea, K. Kordos, E. Lidorikis, D.G. Papageorgiou, Molecular description of charge transport in the IDIC non-fullerene acceptor for organic solar cells. Computational Materials Science 202, 110978 (2022). <sup>2</sup>musicode.eu

<sup>3</sup>The MUSICODE project has received funding from the European Union's HORIZON 2020 Research and Innovation Programme under Grant Agreement No 953187.

## Accelerated Lifetime Testing and Degradation Mechanisms of Three Organic Light-Emitting Diode Generations

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OLEDs are a widespread commercial display technology but they are still subject of ongoing research with the aim to expand their applications ranges through stack and material design. Beside the efficiency, the operational stability is one of the most important key properties that are steadily being improved. The long lifetimes of several thousands of hours of state-of-theart OLED structures at standard operating conditions demand for accelerated lifetime tests (ALTs) and the use of appropriate scaling laws for elevated temperatures T and current densities J. If potential fast burn-in effects are disregarded, the luminance decay follows a stretched exponential with an exponent  $\beta$ . The lifetime  $\tau$  typically follows the empirical law  $\tau = A \cdot J^{-\gamma} \cdot e^{\frac{k_a}{k_B T}}$ , where A,  $\gamma$  and  $E_a$  are fitting parameters and  $k_B$  is the Boltzmann constant.<sup>1</sup> We have tested three OLED generations (fluorescent, phosphorescent (Ph) and thermally activated delayed fluorescence) and ran stress test experiments at four temperatures and four current densities with the benchtop instrument Litos.<sup>2</sup> To analyze the decay curves we employed a global fitting algorithm to simultaneously determine the parameters  $\beta$ , A, y and E<sub>a</sub>. For all three types of OLEDs the data can be reproduced well with the empirical scaling law. It is found that the  $\gamma$  factor is about 1.5 for the PhOLED, while it is only about 1.1-1.2 for the other two stacks (Fig 1(b)). This can be interpreted as increased contribution of bimolecular degradation events in the PhOLED, such as exciton-polaron and exciton-exciton quenching, to the formation of degradation products. In order to exploit the full potential of the ALT method, we demonstrate that we can reduce the luminance decay targets but still keep the predictive capacity of LT70. This reduction can be done in two ways: by reducing the number

<sup>&</sup>lt;sup>1</sup> T. Yoshioka, K. Sugimoto, K. Katagi, Y. Kitago, M. Tajima, S. Miyaguchi, T. Tsutsui, R. Iwasaki, Y. Furukawa, "An Improved Method for Lifetime Prediction Based on Decoupling of the Joule Self-Heating Effect from Coulombic Degradation in Accelerated Aging Tests of OLEDs", SID Symp. Dig. Tech. Pap. 45, 642 (2014) <sup>2</sup> Litos measurement system by Fluxim AG, Switzerland, www.fluxim.com/litos

of stress conditions (<16, i.e. not all *T*, *J* pairs) or by considering only the luminance decay until for example LT90 (Fig 1(c)). We show that with both methods LT70 at normal operating conditions can be predicted accurately, with an error of < 5%. Finally, we performed additional characterization techniques at different states of degradation with the Paios measurement platform.<sup>3</sup> This allows us to gain further insight into the degradation mechanisms. In all three different OLEDs, a systematic shift in the capacitance-voltage onset is observed which may be assigned to localized formation of charge traps.



**Fig. 1** (a) Selected luminance decay curves (b) Determined current scaling factor  $\gamma$  (c) Measured and fitted luminance decay curve (at *T* = 298 K / *J* = 20 mA/cm<sup>2</sup>). For the fit only the data until LT90 were considered.

<sup>&</sup>lt;sup>3</sup> Paios all-in-one measurement platform by Fluxim AG, Switzerland, <u>www.fluxim.com/paios</u>

# Detailed electro-optical modeling of thermally-activated delayed fluorescent OLEDs with different host-guest concentrations.<sup>1</sup>

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OLED device optimization often relies on time-consuming trial-and-error experiments. While the photoluminescence quantum yield can serve as a first indicator to find the best performing host-guest ratio, this quantity does not consider the impact of the latter on the charge transport in the full device. Herein, we analyse four thermally activated delayed fluorescence OLEDs with varied host-guest ratio in the emissive layer. These devices were characterized and modelled in steady-state, under transient conditions and in the frequency domain (experimental results shown in Fig. 1). In this set of devices charge injection into and transport inside the emissive layer plays a crucial role in the performance. Evidenced by a particular transient electroluminescence turn-off overshoot, we show that for the 5% guest concentration device electron and hole transport occurs mainly on the host molecules, with guest molecules acting as trap states. For the other devices with higher guest concentration, we find that transport occurs mainly on the guest molecules. The good agreement between simulations and all the experimental results allows us to extract useful parameters and information. In particular, from the fit of the luminance efficiency we can estimate the triplet-triplet annihilation and triplet-polaron annihilation rates. By comparing the extracted parameters, we found that they increase with increasing concentration of guest molecules. Moreover, we are able to identify triplet-polaron quenching from holes to be limiting the luminance efficiency at low current, while triplet-polaron quenching from electrons and triplet-triplet annihilation are the dominant non-radiative decay processes in the high current regime. Overall, we demonstrate that modelbased analysis of steady-state, transient and frequency domain data obtained for a thermally activated delayed fluorescence OLED allows to get a deeper understanding of the efficiency limiting factors for various host-guest concentrations and driving currents.

<sup>&</sup>lt;sup>1</sup> S. Sem, S. Jenatsch, P. Sahay, S. Züfle, M. Schmid, W. Brütting and B. Ruhstaller, "Detailed electro-optical modeling of thermally-activated delayed fluorescent OLEDs with different host-guest concentrations.". DOI: https://doi.org/10.1016/j.orgel.2022.106553



**Fig. 1** Experimental results of each device: (a) JV; (b) Luminance vs voltage; (c) injection-CELIV with Voffset=2V; (d) injection-CELIV with Voffset=4.5V; (e) C-f with Voffset=0V; (f) C-V with frequency=70Hz.

# Investigation of the orientation of polar organic TPBi molecules under film-growth temperature variation

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The preferential alignment of organic molecules is known to be an important aspect in further improvement of organic light emitting diodes (OLEDs). Horizontal transition dipole moment alignment in the emissive layer has been identified as key parameter for high light outcoupling and so an increase of the external quantum efficiency of OLEDs<sup>1</sup>. However, molecular alignment in the other layers of an organic device also influence the device properties and should not be neglected during the design of a device. For example, a preferred horizontal alignment can enhance the charge carrier mobility in an organic film due to an increase in  $\pi$ -electron overlap<sup>2</sup>. Additionally, some organic semiconductor materials can exhibit a permanent dipole moment (PDM) due to their asymmetric molecular structure. If the organic molecules with a PDM exhibit preferential alignment in a vapor-deposited thin film, the resulting build-up of a macroscopic film polarization, the so-called spontaneous orientation polarization (SOP), influences the charge injection and accumulation behavior in a device<sup>3</sup>. One possible way to tune the alignment of disc and rod-like molecules prepared by vapor-deposition is controlling the ratio between the substrate temperature and the specific glass temperature<sup>4</sup>. With further investigation of tuning the sample preparation conditions, we can gain more insight into the underlying mechanism of molecular alignment and SOP formation.

To this end, Kelvin-Probe, Ellipsometry (VASE) and Photoluminescence (ADPL) measurements on molecular thin films of polar TPBi at different film-growth temperatures were performed for their respective giant surface potential (GSP), birefringence and TDM orientation values. For more insight, the experimental results are complemented by DFT and timedependent (TD-)DFT calculations, to connect them with the corresponding microscopic molecular properties. The results show, that TPBi does not only exhibit a vanishing GSP and so nearly no net film polarization at elevated temperatures close to the glass temperature, but also vanishing anisotropy and a random transition dipole moment orientation. Additionally we observe, that the GSP and birefringence could be tuned quite linearly over a large temperature

<sup>&</sup>lt;sup>1</sup> T. Schmidt et. al., Physical Review Applied, 8, 037001 (2017)

<sup>&</sup>lt;sup>2</sup> D. Yokoyama, Journal of Materials Chemistry, 21, 19187 (2011)

<sup>&</sup>lt;sup>3</sup> Y. Noguchi et. al., Japanese Journal of Applied Physics, 58, SF0801 (2019)

<sup>&</sup>lt;sup>4</sup> D. Walters et. al., Journal of Physical Chemistry Letter, 8, 3380-3386 (2017)

range. The molecular simulations of TPBi resulted in disc-like conformers with their PDM standing perpendicular on the base plane, while the polarization eigenvectors and the TDM vectors are lying in the plane. The electrical and optical experimental results together with the molecular simulations hint to a preferred horizontal molecular alignment of the disc-like TPBi molecules, with an increasing degree of orientation at decreasing substrate temperatures. A similar behavior can possibly be seen in other polar organic thin films, too, and in the end could be used as a method to continuously tune the properties of films in organic devices.

# Master equation modeling of triplet-triplet annihilation in organic phosphorescent emission layers including correlations

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The annihilation of triplet excitons in phosphorescent emission layers of modern OLEDs compromises their performance and device lifetime. This triplet-triplet annihilation (TTA) process can occur by a Förster type interaction between two excitons, leading to a loss of one of them. TTA gives rise to correlations in the positions of the surviving excitons, which are generally ignored in the mean-field approximation of the master equation. These correlations can in principle be accounted for exactly in Kinetic Monte Carlo (KMC) simulations, but these are computationally relatively expensive. In this work, we present master equation modeling of TTA that accounts for correlations in a computationally efficient way. We present two approximations, the "pair approximation" (PA) and the "superposition approximation" (SA), which elucidate the important role of triplet-triplet correlations and allow for computationally efficient simulations. We consider systems without and with exciton diffusion, which partly washes out correlations, and study its influence in simulations of transient and steadystate photoluminescence (PL) experiments. A comparison between mean-field, PA, SA, and KMC simulation results reveals the effect of correlations on the rates of radiative decay and TTA. We compare the accuracy and computational efficiency of the mean-field, PA and SA approximations with KMC simulations and show that our master equation modeling with the PA and SA approximations is an accurate and computationally interesting alternative.<sup>1</sup>



**Fig. 1** Sketch of the effect of correlations on triplet positions in the presence of triplet-triplet annihilation (TTA). Left: triplet excitons with random positions (points), as occurring, for example, at the beginning of an experiment where triplets are generated by illumination. Right: triplet excitons with correlated positions, occurring as a result of TTA with no or weak diffusion. The circle diameter is a measure for the decay length of the triplet pair correlation function in the correlated case.

<sup>&</sup>lt;sup>1</sup> M. Taherpour, C. van Hoesel, R. Coehoorn, and P. A. Bobbert, Phys. Rev. B 105, 085202149 (2022).



**Fig. 2** a) Simulated transient PL intensity decay for various initial triplet densities  $T_0$  (from 0.001 to 0.02 nm<sup>-3</sup>) and  $R_{F,TTA}$  = 3.0 nm, without triplet diffusion. Comparison of master equation calculations (PA and SA) to simulations with the KMC software BUMBLEBEE.<sup>2</sup> b) Transient correlation function for different distances *r* on a cubic lattice.

<sup>&</sup>lt;sup>2</sup> https://simbeyond.com, the BUMBLEBEE software is provided by Simbeyond B.V.

# On the orientation mechanism of non-polar dyes in light-emitting guest-host systems

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After many years of development, OLEDs have continuously been optimized to reach higher efficiency, for which the horizontal orientation of the emitter molecules is one of the dominant factors<sup>1</sup>. For the purpose of studying intrinsic orientation without electrostatic interaction, our work considers non-polar dyes, namely TTPA, BSBCz, DIP and DBP. While the emitter orientation has been studied in neat film as a basic approach, their behavior in guest-host systems has not been studied widely. In this work we focus on the correlation between emitter concentration and its orientation. With the results from experimental and simulation study, we also discuss the possible orientation mechanism of emitter molecules on the substrate surface in terms of molecular structure, possibility to form crystallites and the effective glass transition temperature of the guest-host system. We observe that isotropic structure of the emitter as well as the crystallized and aggregated molecules are not favorable for horizon-tal orientation. However, molecules having a rod-like structure have the tendency to arrange horizontally to the substrate. These results contribute to a further understanding of orientation of non-polar emitter molecules.

<sup>&</sup>lt;sup>1</sup>Schmidt, T. D., Lampe, T., Djurovich, P. I., Thompson, M. E., & Brütting, "Emitter orientation as a key parameter in organic light-emitting diodes", Physical Review Applied, 8(3), 037001, (2007).

# Optical outcoupling efficiency of organic light-emitting diodes with a broad recombination zone

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#### Abstract

Although a broadened recombination zone is beneficial for the lifetime of organic lightemitting diodes (OLEDs), its effect on the optical outcoupling efficiency is unknown. Here, we present a numerical model to simulate the fraction of photons coupled to air for devices with a broad recombination zone. We demonstrate that the total outcoupling can be calculated as the integral of the outcoupling efficiency over different positions of emitting dipoles, weighted by the sum-normalized recombination profile, as obtained from electrical modeling.<sup>1</sup> We further present combined electrical and optical model calculations, demonstrating that the light-outcoupling efficiency in OLEDs is governed by the presence of electron traps and the anisotropy factor of optical dipoles. Electron trapping confines the recombination in a region close to cathode, resulting in strong optical trapping in surface plasmon polariton modes.<sup>23</sup> Trap-filling leads to a voltage dependent recombination profile and optical outcoupling efficiency. For conjugated polymers such as super yellow poly(p-phenylene vinylene) with the chains preferentially aligned in the plane of the film, the outcoupling efficiency can reach ~18% at 5 V. Elimination of electron trapping would allow for a further enhancement up to 24%.

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- Van der Zee, B., Li, Y., Wetzelaer, G. A. H. & Blom, P. W. M. Efficiency of Polymer Light-Emitting Diodes: A Perspective. *Adv. Mater.* 34, 2108887 (2022).
- Li, Y., Van der Zee, B., Wetzelaer, G. J. A. H. & Blom, P. W. M. Optical Outcoupling Efficiency in Polymer Light-Emitting Diodes. *Adv. Electron. Mater.* 7, 2100155 (2021).

#### On the Response Speed of Narrowband Organic Optical Upconversion Devices

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Organic upconversion devices (OUCs) consist of an organic infrared photodetector and an organic visible light-emitting diode (OLED), connected in series. OUCs directly convert photons from the infrared to the visible and are of use in applications such as process control or imaging. Many applications require a fast OUC response speed, namely the ability to accurately detect in the visible a rapidly changing infrared signal.

Here, high image-contrast, solution-processed, narrowband OUCs are reported that convert near-infrared (NIR) light at close to 1000 nm with a full width at half maximum of 130 nm into visible light. Transient photocurrent measurements show that the response speed decreases when lowering the NIR light intensity. This is contrary to conventional organic photodetectors that show the opposite speed versus light trend. It is further found that the response speed can even decrease when increasing the driving voltage. Again, this is an unexpected result because the charge drift velocity gets faster by an enhanced electric field. To understand this surprising response speed behavior, an analysis by numerical simulation is conducted. Results show that the response speed is primarily determined by the (low) electron mobility value in the OLED. Simulations confirm that the speed indeed decreases when lowering the NIR light intensity, and an increase of the applied voltage does not necessarily increase the response speed, as would intuitively be taken for granted. Our work is the first systematic study on the response speed of OUCs. A few reported single-parameter measurements show that the response speed of OUCs so far is limited to around 10 kHz. This is much lower than the speed of organic photodetectors, for which typical cut-off frequencies are above 100 kHz. We think that the main reason for this
discrepancy is the low electric field in the OUC emitter layer, which results in a low charge drift velocity, and in a lower effective electron mobility value because the mobility is electric field dependent. It thus seems that the specific device architecture sets a fundamental limit to the response speed of OUCs.

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**Fig. 1** (a,b) Speed of response under pulsed NIR illumination (950 nm) as a function of voltage and (c,d) as a function of light intensity. For the SY-OUC, the response speed was also measured during continuous device stressing over 10 h (b). For the Ir-OUC, the rise time clearly decreases with increasing light intensity, which indicates that the response speed slows down for lower light intensity (c).

### Characterization of mobile ions in metal halide perovskites and batteries

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als Science and Technology, Überlandstrasse 129, CH-8600 Dübendorf, Switzerland

Materials are often considered to be either purely ionic or electronic conductors. In reality, however, most materials are mixed ionic-electronic conductors, which has important implications for (opto)electronic measurement techniques and device operation.

In this talk, I will show how temperature-dependent current transient measurements can be used to quantify both ionic and electronic conductivities of mixed conductors. I will present some recent developments to reduce ionic conductivity in halide perovskites and to increase ionic conductivity in solid electrolytes, which contribute to a better understanding of ion migration in mixed conductors. Finally, I will show how the conductivities of some materials are affected by light and how photo-ionic effects can be used to improve the performance of batteries.

### Development of QD-LEDs breaking the light outcoupling efficiency limit

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In recent years, quantum dot light-emitting diodes (QD-LEDs) have arisen as ideal candidates to compete with organic light-emitting diodes (OLEDs) in display applications. The outstanding photophysical characteristics of colloidal semiconductor nanocrystals (NC), especially CdSe, InP and lead-halide perovskites (LHP), have led to a rapid surge in their use as active layers in high-efficiency devices, with multiple reports of external quantum efficiencies (EQE) surpassing 20%<sup>1,2,3,4</sup>. To date, the major steps in enhancing device efficiency have come from the development of passivation strategies to reduce NC defects and boost internal quantum efficiency (IQE). As IQE approaches unity, the major bottleneck to device efficiency remains light outcoupling. A well-known intrinsic strategy for improving light extraction in OLEDs is transition dipole moment (TDM) engineering<sup>5</sup>. Accordingly, EQEs approaching 40% have been demonstrated by designing efficient phosphorescent or thermally activated fluorescence (TADF) emitters with preferential orientation of their TDMs parallel to the device substrate.

At the same time, recent studies on few-monolayer-thick colloidal CdSe nanoplatelets (NPLs) have shown highly anisotropic TDM orientation<sup>6</sup>, suggesting that similar strategies might be extended to QD-LEDs. Inspired by these results, we wanted to tackle the following fundamental question: can we develop a model to understand and predict the TDM orientation in semiconductor NC films?

In general, in contrast with organic semiconductor molecules, inorganic materials tend to have highly symmetric crystal lattices, leading to isotropic electronic structure and strong degener-

<sup>&</sup>lt;sup>1</sup>Deng, Y. et al. Solution-processed green and blue quantum-dot light-emitting diodes with eliminated charge leakage. Nat. Photonics (2022)

<sup>&</sup>lt;sup>2</sup> Won, Y.-H. et al. Highly efficient and stable InP/ZnSe/ZnS quantum dot light-emitting diodes. Nature 575, 634–638 (2019).

<sup>&</sup>lt;sup>3</sup> Kim, Y.-H. et al. Comprehensive defect suppression in perovskite nanocrystals for high-efficiency lightemitting diodes. Nat. Photon 15, 148–155 (2021).

 <sup>&</sup>lt;sup>4</sup> Kumar, S., Marcato, T., Krumeich, F. et al. Anisotropic nanocrystal superlattices overcoming intrinsic light outcoupling efficiency limit in perovskite quantum dot light-emitting diodes. Nat Commun 13, 2106 (2022).
<sup>5</sup> K.-H. Kim, J.-J. Kim, Origin and Control of Orientation of Phosphorescent and TADF Dyes for High-

Efficiency OLEDs. Adv. Mater. 30, 1-1, (2018)

<sup>&</sup>lt;sup>6</sup> Scott, R., Heckmann, J., Prudnikau, A. et al. Directed emission of CdSe nanoplatelets originating from strongly anisotropic 2D electronic structure. Nature Nanotech 12, 1155–1160 (2017).

acy at the band-edge. However, nanostructuring offers the possibility of introducing favorable symmetry breaking and anisotropy by controlling the NC shape. In this talk, I will first present a multiscale model of the preferential horizontal TDM orientation in LHP NPLs that highlights three fundamental contributions (Fig. 1): (i) anisotropic conduction band Bloch states and large fine exciton splitting, (ii) anisotropic dielectric screening, (iii) ordered assembly in thin film.

This model identifies the NPL thickness and the film order parameter as the two fundamental degrees of freedom allowing for continuous tuning of the TDM orientation.

We showed that these criteria can already be satisfied in devices by inducing the formation of 2D ordered assemblies of LHP anisotropic NCs on a hole transport layer with low surface energy. This resulted in a ratio of horizontal dipoles up to 0.75, which allowed our optimized green QD-LEDs to achieve peak EQE up to 24.96%<sup>4</sup>.



**Fig. 1 Multiscale origin of PL emission directionality in LHP NPLs.** From left to right: the thickness dependence of NPL electronic structure and exciton TDM distribution; dielectric mismatch between NPL and ligand shell causing electric field rescaling; assembly of single-phase NPLs to form macroscopically different films characterized by their orientational distribution.

# Lessons learned from device modeling of solar cell current-voltage characteristics using Machine Learning techniques

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Device modeling is extensively used in solar cells research going from simple models such as the Shockley diode equation to more complex models like Monte-Carlo or drift-diffusion (DD). These models are mostly used in three ways: first as a predictive tool to assess a technology's ultimate efficiency. Secondly, as an investigation tool to understand the limitation of a given experimental technique to measure a certain physical quantity (mobility, defect density...) and to define the conditions under which the technique is accurate. Finally, as a diagnostic tool to understand and quantify the main losses for a given device by reproducing experimental results.

One of the main criticisms about using DD modeling as a means to quantify material properties is the many fitting (10 to 40) parameters that need to be estimated. Many would use the famous von Neumann argument: "With four parameters I can fit an elephant, and with five I can make him wiggle his trunk" and argue that with so many fitting parameters one could fit almost any model to the experimental data.

In this presentation, we will show that the use of DD modeling combined with machine learning (ML) tools to fit light-intensity-dependent current-voltage characteristics indeed leads to a unique solution. However, the uniqueness of this solution is not defined by a single set of parameters but by figures of merits (FOMs) that are a combination (product or ratio) of the material properties. This fact is reported for the first time and has been overlooked in the literature and means that the values reported are not necessarily accurate as in most cases only the FOMs can be estimated accurately and not the individual parameter values.

Therefore, we will also define under which conditions one can accurately quantify the materials parameters (mobility, defect density, density of states...).

Finally, we will show how DD modeling and ML can be combined to speed up the analysis of large datasets coming from degradation measurements or high-throughput experimentation.

### Solar Cell Parameter Extraction by Convolutional Neural Networks Trained on Simulated Imaging Data

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Physical models can help improve solar cell efficiency during the design phase as well as during quality control after the fabrication process. We present a data-driven approach to inverse modeling that can predict the underlying parameters of a finite element method (FEM) solar cell model based on imaging data of the solar cells. Thanks to the flexibility of convolutional neural networks (CNN), the approach can potentially be applied to predict any model parameters that are sufficiently well defined by a set of measurement image types. We demonstrate the validity of the proposed method based on a basic case in which the dark saturation current, the sheet resistance, the parallel conductivity and the applied voltage are estimated from electroluminescence (EL) images. 75,000 synthetic EL images were generated using LAOSS<sup>1</sup> with randomized parameters of the physical cell model. We combine 17 deep convolutional neural networks (CNNs) based on a modified VGG19 architecture into a deep ensemble to add uncertainty estimates<sup>2</sup>. The scheme of input and output data of the CNN model is shown in Figure 1.



Fig. 1 Structure of the CNN input and output data.

The trained network was tested on different physical solar cell samples for which the estimated parameters were used to create the corresponding model representations. The parameter prediction process and the resulting resimulations for two measurement images and one simulated validation image is shown in Fig. 2. The resimulations of the measurements resulted in relative deviations of the calculated from the measured junction voltage values of 0.2\% on average with a maximum of 10\%, demonstrating the validity of the approach. The results show inaccuracies in the prediction of the sheet resistance and the dark saturation current, which can be explained by the correlation of the two parameters, since the resulting EL image is mainly influenced by their product. We also note the well-known simulation-reality gap that arises when a CNN model fully trained on simulated data is applied to real measured data.



Fig. 2 Validation results of the CNN parameter prediction based on two measurement images and a simulated validation image.

Using the silicon solar cell model, we show that such a novel approach to data-driven statistical inverse modeling can help apply recent developments in deep learning to new engineering applications that require real-time parameterizations of physical models augmented by confidence intervals. The presented work paths the way for a future development in which we plan to combine data-driven inverse modelling with equation based inverse modelling by using physics informed neural networks (PINNs). The method will be integrated in an all-in-one imaging measurement device that aims to create a digital twin of the solar cell by predicting the parameters of a physical model based on visible spectrum image, EL image, dark lock-in (DLIT) image, photoluminescence (PL) image and illuminated lock-in (ILIT) image. ACKNOWLEDGMENT: We gratefully acknowledge financial support from Innosuisse (project number 37304.1 IP) as well as fruitful discussions with Thilo Stadelmann from the Center of Artificial Intelligence at ZHAW School of Engineering, Winterthur, Switzerland.

<sup>&</sup>lt;sup>1</sup>M. Diethelm, L. Penninck, M. Regnat, T. Offermans, B. Zimmermann, C. Kirsch, R. Hiestand, S. Altazin, B. Ruhstaller, 2020. Finite element modeling for analysis of electroluminescence and infrared images of thin-film solar cells. Solar Energy 209, 186 – 193.

<sup>&</sup>lt;sup>2</sup> Lakshminarayanan, B., Pritzel, A., Blundell, C., 2016. Simple and Scalable Predictive Uncertainty Estimation using Deep Ensembles. Advances in Neural Information Processing Systems 2017-December

## Material Parameter Extraction of Thin-Film-Devices with Machine Learning

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In most cases device modelling relies on the knowledge of material parameters. Some of them can be determined by dedicated experiments, others remain unknown or vary hugely in their reporting. Minimizing the error between the simulation and measurement by varying the unknown material parameters is a common approach to find a suitable parameter set that describes the measurement. The correlation of parameters can be reduced by combining multiple experiments<sup>1</sup>.

In this contribution, we explore machine learning methods instead of traditional least-squares approaches to find the underlying material parameters.

First, we generate a training data set of a pip device<sup>2</sup> with a semiconductor device simulator by varying the underlying parameters. A machine learning model is then trained with this data set and can predict the underlying material parameters from current-voltage curves and impedance spectroscopy spectra<sup>3</sup>.

In a more recent approach, we aim at integrating the physical laws into the machine learning approach and use physics-informed neural networks to estimate the model parameters<sup>45</sup>. With this approach, we take advantage of the machine learning framework and infrastructure for our task and do not require a large training data set. In Fig.1, we use a simplified large-area FEM semiconductor model for simulating the potential distribution of a solar cell in the dark. In a next step, a PINN is applied that incoperates the solar cell model to extract the model parameters. In a last step, we use the extracted parameters to compare the potential

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<sup>&</sup>lt;sup>2</sup> S. Jenatsch, S. Altazin, P.-A. Will, M. T. Neukom, E. Knapp, S. Züfle, "S. Lenk, S. Reineke, and B. Ruhstaller, "Quantitative analysis of charge transport in intrinsic and doped organic semiconductors combining steady-state and frequency-domain data," Journal of Applied Physics 124, 105501 (2018).

<sup>&</sup>lt;sup>3</sup> E. Knapp, M. Battaglia, T. Stadelmann, S. Jenatsch, B. Ruhstaller, "XGBoost trained on synthetic data to extract material parameters of organic semiconductors," Proceedings of the 8th SDS. 8th Swiss

<sup>&</sup>lt;sup>4</sup> G.E. Karniadakis, I.G. Kevrekidis, L. Lu et al. "Physics-informed machine learning," Nat Rev Phys 3, 422–440 (2021).

<sup>&</sup>lt;sup>5</sup> M. Raissi, P. Perdikaris, G.E. Karniadakis," Physics-informed neural networks: A deep learn-ing framework for solving forward and inverse problems involving nonlinear partial differen-tial equations," Journal of Computational Physics, Volume 378, 2019.

distribution with the original simuluation.



**Fig. 1** Parameters are extracted with the aid of a PINN from a potential distribution FEM simulation of a solar cell. To validate the result, we compare the reconstruction with the original image.

## Design of optical filters for high-efficiency and high-yield colored perovskite photovoltaics

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The implementation of photovoltaic systems in urban areas is not yet widespread due to aesthetic requirements. The possibility to choose the color of the photovoltaic panel is essential to increase its adoption on building facades. Optical filters provide color tunability to any solar cell technology without requiring modifications of the device stack. In this work, we considered notch (or cutoff) filters, which selectively reflect a portion of light due to their structure consisting of alternating layers of low and high refractive index materials. We alternated layers of SiO<sub>2</sub> (low index) and TiO<sub>2</sub> (high index), whereas the combination SiO<sub>2</sub>/TiO<sub>2</sub>/SiO<sub>2</sub> is identified as a repeat unit.

We carried out optical simulations with the software Setfos to analyze the influence of thickness variation of  $SiO_2$  and  $TiO_2$  individually and the number of repeat units on FWHM, peak wavelength, and intensity of the reflectance. It allowed the design of filters reflecting green, blue, and red colors with high color purity and reflectance at the desired wavelength.

We combined optical and drift-diffusion simulations to quantify the negative impact of the filter on the efficiency compared to a reference solar cell without the filter. Compared to the reference cell, the performance reduction goes from about 10% for the blue filter to 23% for the red filter for the perovskite solar cell with a bandgap of 1.61 eV. Different approaches to reduce the performance difference between a colored solar cell and the reference one, which include perovskite thickness, reflectance intensity, and reflectance bandwidth variations are evaluated. Moreover, the introduction of light scattering elements that reduce the angular shift of the reflectance color was exemplified.

The software Setfos gives the possibility to calculate the energy yield of a solar cell according to the geographical location. This allowed us to quantify the impact of the optical filter on the energy output of a solar cell with and without the optical filter during a year at different tilting positions giving an estimate of the power production of the facade of a house.