



School of
Engineering

ICP Institute of
Computational Physics

International Conference on Simulation of Organic Electronics and Photovoltaics 2016

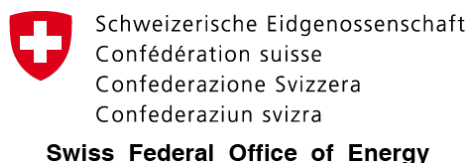


Zurich University of Applied Sciences

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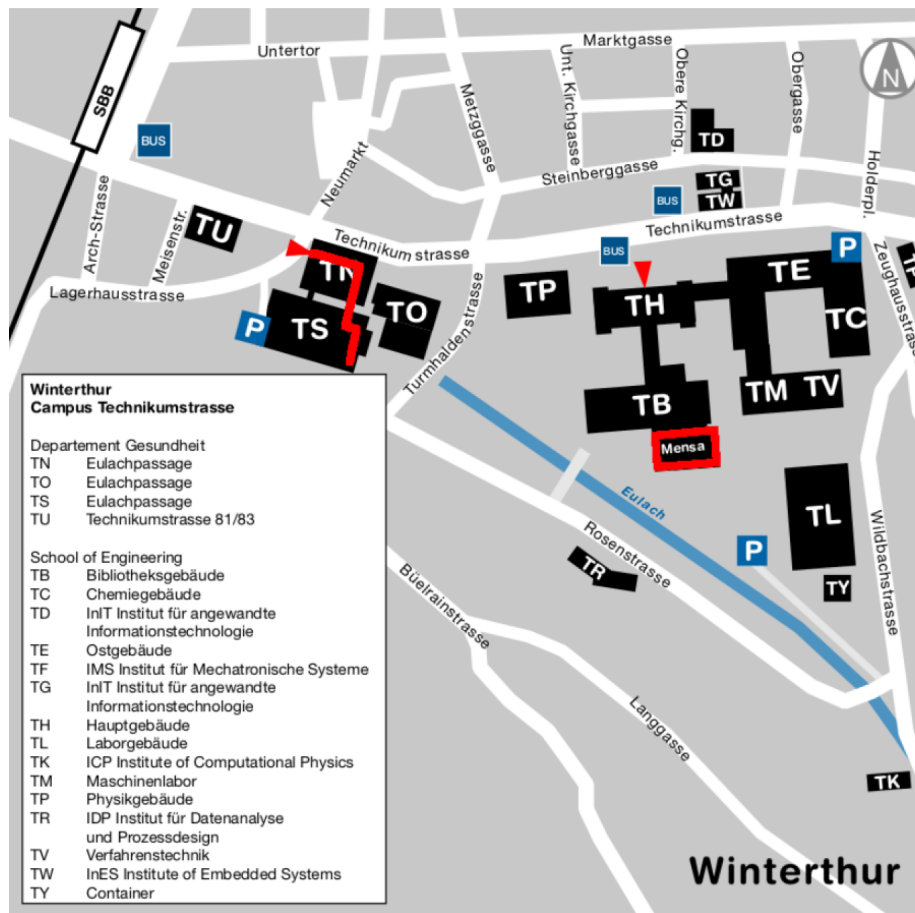


Site Plan

Conference: Building TS, Room 01.40. Please enter building TN through the doors at Technikumstrasse, walk up to the first floor and traverse to building TS to reach the conference lecture room 01.40. Coffee break refreshments and buffet lunch is served in the hallway in front of the lecture room 01.40 in building TS.

Fluxim workshops: Building TS, Room 02.44

Conference Dinner: Mensa



Scientific Program

Program SimOEP 2016 as of 29.8.2016

	14.09.16			15.09.16			16.09.16		
	OLED	PSC/OPV	TCE	OLED	PSC/OPV	TCE	OLED	PSC/OPV	TCE
08.45 - 09.00									
09.00 - 09.15				E. Mankel					Ch. Kirsch
09.15 - 09.30				V. Wissdorf					P. Losio
09.30 - 09.45				P. Africa				B. Niesen	
09.45 - 10.00				A. Hofacker				Z. Dong	
10.00 - 10.15		Fluxim Workshop: Large-Area PV and OLED Simulation (LAOSS)		E. Knapp					
10.15 - 10.30				S. Rizvi					
10.30 - 10.45				Coffee			Coffee		
10.45 - 11.00									
11.00 - 11.15					U. Würfel			Th. Lampe	
11.15 - 11.30				J. Mayer			A. Egel		
11.30 - 11.45				S. Züfle			L. Stepanova		
11.45 - 12.00				D. Andrienko			W. Tress		
12.00 - 12.15	REGISTRATION						A. Walker		
12.15 - 12.30									
12.30 - 12.45									
12.45 - 13.00									
13.00 - 13.15	OPENING			LUNCH			LUNCH		
13.15 - 13.30	W. Brütting								
13.30 - 13.45	T. Tsutsui						M. Neukom		
13.45 - 14.00	J. J. Kim				A. Opitz		Ph. Calado		
14.00 - 14.15	St. Altazin				Y. Leroy		J. Lehr		
14.15 - 14.30				Fluxim Workshop: Char. of OLEDs (PAIOS)	I. Cardinaletti		Fluxim Workshop: Simulation of OLEDs (SETFOS)	Fluxim Workshop: Simulation of PV (SETFOS)	
14.30 - 14.45					U. Palanchoke				
14.45 - 15.00					A. Spies				
15.00 - 15.15					P. Lopez				
15.15 - 15.30	Coffee			Coffee					
15.30 - 15.45									
15.45 - 16.00	H. van Eersel								
16.00 - 16.15	H. Kim					Ch. Ballif			
16.15 - 16.30	M. Regnat					Y. Romanyuk			
16.30 - 16.45	Ch. Hänisch					LAOSS			
16.45 - 17.00	S. Jenatsch								
17.00 - 17.15	M. Lindh								
17.15 - 17.30					Fluxim Workshop: Characterizati on of PV (PAIOS)				
17.30 - 17.45									
17.45 - 18.00	Apéro & Fluxim Product Demo								
18.00 - 18.15									
18.15 - 18.30									
18.30 - 18.45									
18.45 - 19.00									
19.00 - 20.00									
20.00 - 22.00				Dinner					

Wednesday, 14.9.2016

10.00-12.00	Fluxim Workshop <i>Large-Area PV and OLED Simulation (LAOSS)</i>	
12.00-13.00	Registration	
13.00-13.15	Welcome/Opening <i>Beat Ruhstaller, ICP ZHAW and Fluxim, Switzerland</i>	
13.15-13.45	OLED Roll-off and Degradation Analysis by Transient Optical and Electrical Methods <i>Wolfgang Brütting, University of Augsburg, Germany</i>	7
13.45-14.15	Analysis of Long-Term Degradation in OLEDs and its Application for Lifetime Prediction <i>Tetsuo Tsutsui, CEREBA, Japan</i>	8
14.15-14.45	Quantitative Analysis of the Efficiency of OLEDs <i>Jang-Joo Kim, Seoul National University, South Korea</i>	9
14.45-15.15	On the role of polar molecules and charge injection in OLEDs <i>Stéphane Altazin, Fluxim, Switzerland</i>	10
15.15-15.45	Coffee Break	
15.45-16.15	Study of transient phenomena in phosphorescent and TADF OLEDs: a Monte Carlo simulation approach <i>Harm van Eersel, Simbeyond, Netherlands</i>	12
16.15-16.30	Horizontal orientation of phosphorescent emitting dipoles for highly efficient organic light-emitting diodes <i>Kwon-Hyeon Kim, Seoul National University, South Korea</i>	13
16.30-16.45	Linking the OLED efficiency roll-off to the change of the emission zone by electro-optical device modelling <i>Markus Regnat, ICP ZHAW, Switzerland</i>	14
16.45-17.00	Precise determination of the molecular orientation in organic thin films by simulating the spectral radiant intensity of finite thickness emission layers <i>Christian Hänisch, IAPP Dresden, Germany</i>	16
17.00-17.30	Doping evolution and junction formation in stacked cyanine dye light-emitting electrochemical cells <i>Sandra Jenatsch, EMPA, Switzerland</i>	18
17.30-17.45	Emission characteristics of light-emitting electrochemical cells <i>Mattias Lindh, Umeå University, Sweden</i>	20
17.45-18.45	Apéro & Fluxim Product Demo	

Thursday Morning, 15.9.2016

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09.30-09.45	Simultaneous Extraction of DOS Width and Injection Barrier in OTFTs <i>Pasquale Africa, MOX Modeling and Scientific Computing, Milano, Italy</i>	22
09.45-10.00	Non-Equilibrium Charge Carrier Kinetics in a Drift-Diffusion Model of Organic Disordered Semiconductors <i>Andreas Hofacker, DC-IAPP Dresden, Germany</i>	24
10.00-10.15	Origins of Negative Capacitance in Organic Single Layer Devices <i>Evelyne Knapp, ICP ZHAW, Switzerland</i>	25
10.15-10.30	A Critical Look at the Mott-Schottky Analysis for Extraction of Background Doping in Organic Diodes <i>Syed Rizvi, Indian Institute of Technology Kanpur, India</i>	26
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11.00-11.30	How charge carrier transport and electrode selectivity influence the performance of (organic) solar cells <i>Uli Würfel, ISE Freiburg, Germany</i>	28
11.30-12.00	Diffraction Gratings for Enhanced All-Season Energy-Harvesting in OPV Devices <i>Jan Mayer, CSEM Muttenz, Switzerland</i>	30
12.00-12.30	Shedding light on the stability of organic solar cells <i>Simon Züfle, ICP ZHAW, Switzerland</i>	31
12.30-13.00	Molecular understanding of heterostructures of organic semiconductors <i>Denis Andrienko, MPIP Mainz, Germany</i>	33
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15.45-16.15	Coffee Break	
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17.30-18.45	Fluxim Workshop <i>Characterization of PV (PAIOS)</i>	
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09.00-09.15	Simulation of lateral charge transport in large-area optoelectronic semiconductor devices <i>Christoph Kirsch, ICP ZHAW, Switzerland</i>	46
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10.00-10.30	Modeling tandem perovskite/c-silicon solar cells <i>Dong Zhang, aECN-Solliance, Netherlands</i>	50
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11.00-11.15	Optical simulations of birefringent organic semiconductor devices <i>Thomas Lampe, University of Augsburg, Germany</i>	52
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13.45-14.15	Explanation for reduced IV-curve hysteresis in highly efficient perovskite solar cells <i>Martin Neukom, ICP ZHAW and Fluxim, Switzerland</i>	59
14.15-14.30	Simulating transient optoelectronic measurements on perovskite solar cells: Evidence for ion migration in devices with minimal hysteresis <i>Philip Calado, Imperial College London, United Kingdom</i>	60
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OLED Roll-off and Degradation Analysis by Transient Optical and Electrical Methods

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Organic light-emitting diodes (OLEDs) suffer from a loss of efficiency under electrical operation in two distinct ways: (i) First, even in pristine devices with well-balanced charge carrier injection and recombination, the quantum efficiency is a function of current density and particularly decreases in the application-relevant range of currents. This efficiency roll-off has been investigated in phosphorescent OLEDs by combining electrical and optical excitation in time-resolved spectroscopic experiments¹. We are able to correlate changes of the triplet lifetime with a decrease of the radiative emitter quantum efficiency and identify the dominant exciton quenching process. (ii) Second, upon long-term operation of OLEDs at drive currents required for lighting applications their efficiency degrades further in an irreversible manner. Using transient methods to analyze both electrical and optical changes during an accelerated aging protocol for phosphorescent OLEDs, we are able to identify different contributions to the drop of luminance, namely exciton quenching as well as non-radiative recombination due to trap formation and imbalanced carrier flow².

¹ S. Wehrmeister, L. Jäger, T. Wehler, A. F. Rausch, T. C. G. Reusch, T. D. Schmidt, and W. Brütting, “Combined electrical and optical analysis of the efficiency roll-off in phosphorescent organic light-emitting diodes”, *Phys. Rev. Appl.* 3, 024008 (2015)

² T. D. Schmidt, L. Jäger, Y. Noguchi, H. Ishii, and W. Brütting, “Analyzing degradation effects of organic light-emitting diodes via transient optical and electrical measurements”, *J. Appl. Phys.* 117, 211502 (2015)

Analysis of Long-Term Degradation in OLEDs and its Application for Lifetime Prediction

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Understanding of degradation mechanisms of OLEDs has been one of the most important subjects in terms of both basic science and application-oriented research and development of OLEDs. Even though some typical examples for the origins of device and material degradation, which have implications for device and materials design have been elucidated, comprehensive feature of the long-term degradation of OLEDs has not been shown yet. The analysis of the time dependency of luminous degradation under constant-current driving can be one of the simplest but most powerful methods for obtaining a comprehensive picture of OLED degradation. In 2002, Ishii and Taga showed that the shapes of the luminous degradation curves had little dependency on both the current density and temperature in fluorescent OLEDs by using a fitting function called stretched-exponential decay (SED).¹⁾ Fery et al. demonstrated that the SED function described the degradation curves of phosphorescent OLEDs and proposed a degradation model, which traced the curve of the SED function well.²⁾ Although the SED function has been used for the curve fitting of luminous fading in many publications, since then, little analysis and discussion regarding the physical meaning of fitting parameters have been added. Recently, Tsujimura et al. reported that the luminance degradation in their phosphorescent OLEDs was fitted with a linear combination of two simple exponential decay functions, and they discussed the meaning of the separated initial and normal degradation modes.

In this presentation, we first propose a systematic approach for phenomenological description of the luminous decay curves under continuous constant-current driving. We will show that systematic quantitative descriptions of luminous decay curves at both various current-density and environmental-temperature conditions are possible, when following conditions are satisfied.

1. The contribution of initial short-term degradations is carefully eliminated from experimental degradation curves.
2. A long-term degradation component is described with an appropriate decay functions, typically a stretched-exponential function.
3. The decay functions obtained at different temperature and current-density conditions are scalable, and a time-scale parameters are extracted from decay functions.
4. The extracted parameters are represented as a function of both temperature and current density.

We discuss about scientific background of our approach and represent some typical examples of analytic results based on our experimental long-term degradation curves.

In addition, we demonstrate how we can apply our approach of luminous-decay analysis to the prediction of lifetime of OLEDs using both temperature and current-density acceleration conditions, contributing to the test time reduction of a factor of up to 1/40.

1. M. Ishii and Y. Taga, *Appl. Phys. Lett.* 80, 3430 (2002).

2. C. Féry, B. Racine, D. Vaufrey, H. Doyeux, and S. Cinà, *Appl. Phys. Lett.* 87, 213502 (2005).

Quantitative Analysis of the Efficiency of OLEDs

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Department of Materials Science and Engineering, Seoul National University, Seoul 151-744,
South Korea

A comprehensive model is presented for the quantitative analysis of the factors influencing the efficiency of organic light emitting diodes (OLEDs) as a function of current density. It accounts for the contributions of charge carrier imbalance, quenching processes, and the optical design loss of the device coming from optical effect including the cavity structure, location and profile of the exciton, the effective radiative quantum efficiency, and the out-coupling efficiency. The quantitative analysis of the efficiency can be performed with the optical simulation using material parameters and experimental measurements of exciton profile in the emission layer and lifetime of the exciton as functions of current density. The analysis method is applied to three different phosphorescent OLEDs (PhOLEDs) based single host, mixed host, and exciplex-forming co-host. The three factors (charge carrier imbalance, quenching processes, and optical design loss) are found to influence in different ways for different devices. This model has the potential to be utilized for analyzing the physical processes and optimizing the structures of OLEDs.

On the role of polar molecules and the barrier for charge injection in OLEDs

S. Altazin¹, S. Züfle^{1,2}, E. Knapp², C. Kirsch², T. D. Schmidt³, L. Jäger³,
W. Brütting³, B. Ruhstaller^{1,2}

¹ Fluxim AG, Winterthur, Switzerland

² Zurich University of Applied Sciences, Institute of Computational Physics, Winterthur, Switzerland

³ Experimental Physics IV, Institute of Physics, University of Augsburg, Germany

Electron transport layers (ETLs) often have a non-zero molecular moment. When deposited in an OLED device, this effectively leads to a layer with a positive sheet charge density on one side and negative on the other. So far, only experimental studies have been performed to study the impact of the polarity on the device operation¹. For the first time we combine electrical characterizations with drift-diffusion simulations of such devices. In this contribution, we have studied a traditional bi-layer OLED where the Alq₃ was used as ETL (Fig. 1). In a first step, by comparing experimental capacitance versus voltage (C-V) (Fig. 2) and frequency (C-f) measurements with simulations, we demonstrate that the drift-diffusion solver of SETFOS² is able to simulate OLEDs fabricated with such materials; we were then able to extract the sheet charge density induced by the polar nature of the ETL. Based on this successful comparison with experiments, simulations have shown that the polarity of the ETL can be beneficial for the device efficiency if correctly oriented, as it can increase or decrease the current level in the device depending on its orientation. Finally, we made use of both temperature dependent C-f and charge extraction by linearly increased voltage (CELIV) characterizations. Combining these two techniques and comparing measurements with simulations we could extract the hole injection barrier in the device.

¹ Brütting, W., Berleb, S., Mückl, A. G., "Device physics of organic light-emitting diodes based on molecular materials" *Org. Elec.*, **2** (1), 1-36 (2001).

² Simulation software SETFOS version 4.3 by Fluxim AG, www.fluxim.com.

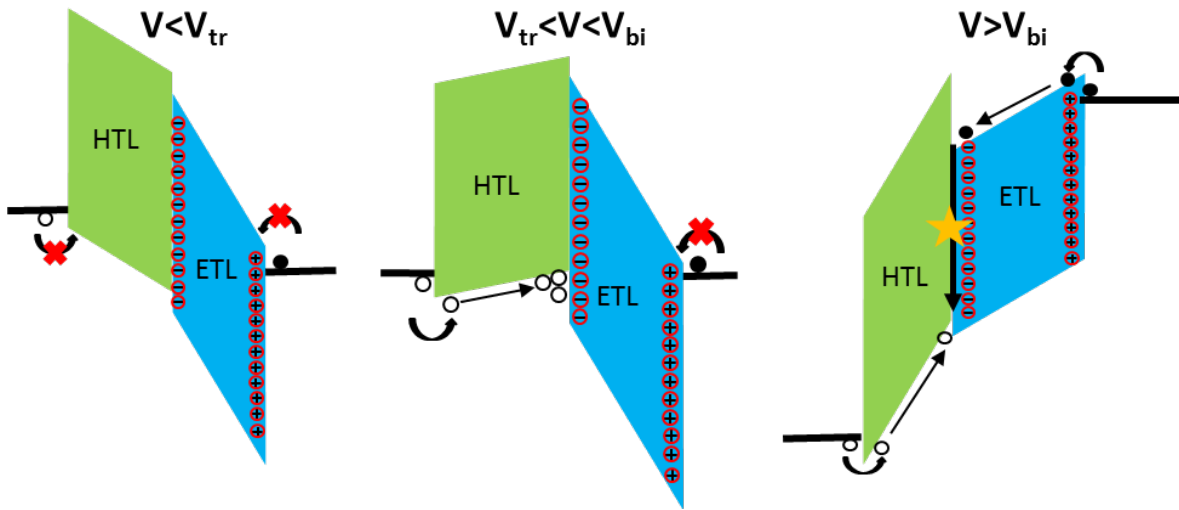


Fig. 1: Schematic band diagram of the bi-layer OLED used in this study. The ETL (Alq_3) layer is supposed to have a molecular polar moment that is not randomly oriented, resulting in sheet charge densities on either side of the layer. This sheet charge density modifies the band diagram.

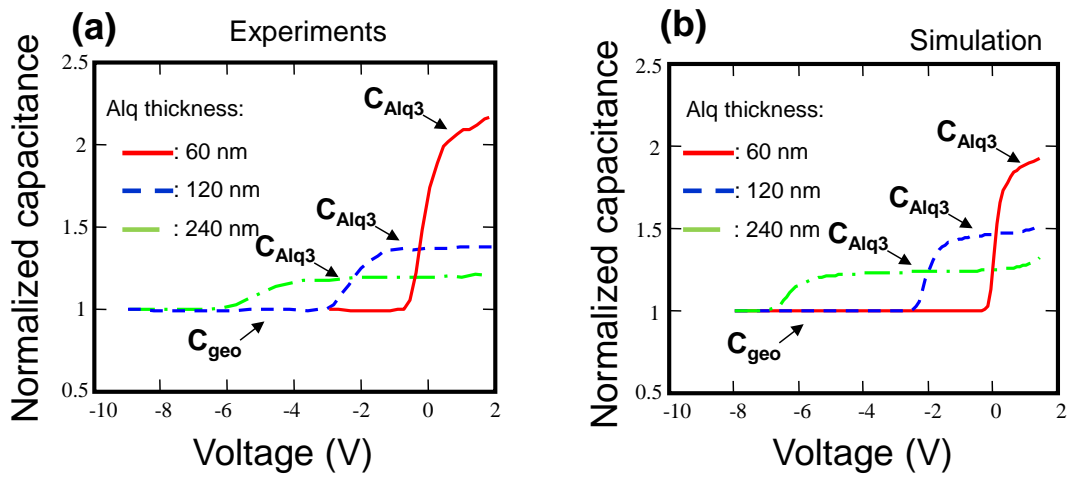


Fig. 2: Experiments from [1] (a), and simulations (b) using setfos [2] of a bilayer polar OLED

Study of transient phenomena in phosphorescent and TADF OLEDs: a Monte Carlo simulation approach

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From an extensive study, we have shown how molecular-scale kinetic Monte Carlo (kMC) simulations can be used to understand the device physics of a hybrid (phosphorescent red and green, fluorescent blue) white OLED stack.¹ The $J(V)$ characteristics and color point were surprisingly well reproduced, starting only from physical parameters obtained from experiment. More recently, we have shown how such simulations can be used to study the cause of roll-off in prototypical phosphorescent monochrome red and green devices,² how it can be used to understand the concentration-dependence of triplet-triplet annihilation in phosphorescent dyes,³ and how roll-off and degradation can depend on different material parameters.⁴ In this contribution, we show how kMC simulations can be used to study transient electroluminescence in OLEDs, to facilitate the study of quenching processes,⁵ and to understand phenomena such as the sometimes observed overshoot in emission⁶ and delayed emission⁷ as function of the voltage, dye concentration and host material. The simulations include exciton quenching processes (exciton-polaron quenching, exciton-exciton annihilation), field-induced dissociation, Förster and Dexter transfer between dye molecules, and delayed fluorescence due to triplet-triplet annihilation. As an outlook, we show how these techniques can also be applied to 3.5th generation OLEDs containing both a thermally activated delayed fluorescence (TADF) sensitizer and a fluorescent guest.⁸

¹ M. Mesta *et al.* Nature Materials 12, 652 (2013); M. Mesta *et al.* Appl. Phys. Lett. 108, 13301 (2016)

² H. van Eersel *et al.* Appl. Phys. Lett. 105, 143303 (2014); H. van Eersel *et al.* J. Appl. Phys. 119, 163102 (2016)

³ H. van Eersel *et al.* J. Appl. Phys. 117, 115502 (2015); L. Zhang *et al.* Chem. Phys. Lett. 652, 142 (2016)

⁴ R. Coehoorn *et al.* Adv. Func. Mat. 25, 2024 (2015)

⁵ Song *et al.* Appl. Phys. Lett. 97, 243304 (2010)

⁶ Murawski *et al.* Advanced Materials 17, 6801 (2013)

⁷ Reineke *et al.* Nature 459, 234 (2009)

⁸ Nakanotani *et al.* Nature Communications 5, 4016 (2014)

Horizontal orientation of phosphorescent emitting dipoles for highly efficient organic light-emitting diodes

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Recently, the emitting dipoles with perfect horizontal orientation parallel to the substrate result in the theoretical external quantum efficiency (EQE) limit over 45% without any extra light extraction structures compared to 25~30% for randomly oriented emitting dipoles. Practically, an EQE of over 40% is possible with PL quantum yield = 95% and horizontal emitting dipoles ratio (Θ) = 95%. But, EQEs of OLEDs over 30% have not been much reported even though horizontally oriented transition dipoles can result in efficiencies of over 30%. Here, we analyzed emitting dipole orientation of phosphorescent emitters using angle dependent PL measurement and discussed how to increase the Θ of phosphorescent emitter in terms of emitter molecular structure. Based on our strategy, we developed emitting layer having high Θ using Ir and Pt complexes and demonstrated EQE of OLEDs closed to 40%.^{1,2,3,4}

[1] K.-H. Kim, S. Lee, C.-K. Moon, S.-Y. Kim, Y.-S. Park, J.-H. Lee, J. W. Lee, J. Huh, Y. You, J. J. Kim, *Nat. Commun.* 5, 4769 (2014).

[2] K.-H. Kim, C.-K. Moon, J.-H. Lee, S.-Y. Kim, J.-J. Kim, *Adv. Mater.* 26, 3844-3847 (2014).

[3] K.-H. Kim, J.-Y. Ma, C.-K. Moon, J.-H. Lee, J. Y. Baek, Y.-H. Kim, J.-J. Kim, *Adv. Optical Mater.* 3, 1191-1196 (2015).

[4] K.-H. Kim, J.-L. Liao, S. W. Lee, B. Sim, C.-K. Moon, G.-H. Lee, H. J. Kim, Y. Chi, J.-J. Kim, *Adv. Mater.* 28, 2526-2532 (2016)

Linking the OLED efficiency roll-off to the change of the emission zone by electro-optical device modelling

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² FLUXIM AG, Winterthur, Switzerland

We demonstrate the determination of the emission zone in a phosphorescent, three-layer OLED with a 35 nm thin emissive layer (EML) and how the emission zone evolves with increasing current densities. Electro-optical device simulations^{1,2,3} revealed a reduction of the charge balance factor γ due to the emission zone change, which nicely explains the measured efficiency roll-off at increased current densities.

For the determination of the emission zone and its change, we have measured the angular, polarized electroluminescence (EL) spectra, and employed a purely optical fit algorithm with Setfos¹ to determine the number and the position of the emissive dipoles inside the 35 nm EML^{1,2,3}. Fig. 1 shows the extracted density of emissive dipoles constituting a split emission zone in the EML with high dipole densities on either side of the EML. At low current densities the majority of emissive dipoles are formed at the EML/ETL interface as it is expressed by the ratio of the two emission zone peak intensities given in Fig. 1. Interestingly at high current densities the ratio shifts to the HTL/EML interface.

The electro-optical device model nicely reproduces the observations of the double peak in the emission zone, as well as the dependence on current density. With this model it is possible to show, that the shift of the emission zone causes an increase of the light out-coupling factor χ , and at the same time a reduction of the charge balance factor γ . The simulated product $\gamma * \chi(0^\circ)$ describes with good accuracy the measured current efficiency at 0° over the entire measurement range (see Fig. 2), without considering additional effects such as triplet-triplet annihilation (TTA) and triplet-polaron quenching (TPQ).

The same methodology can also be applied to study long-term degradation effects in OLEDs.

¹ Simulation software SETFOS by Fluxim AG, www.fluxim.com, Switzerland

² B. Perucco, N.A. Reinke, D. Rezzonico, E. Knapp, S. Harkema, B. Ruhstaller, *Organic Electronics* 13 (10), 1827ff, (2012)

³ B. Perucco, N. A. Reinke, D. Rezzonico, M. Moos and B. Ruhstaller, *Optics Express* 18 (S2), 246ff, (2010).

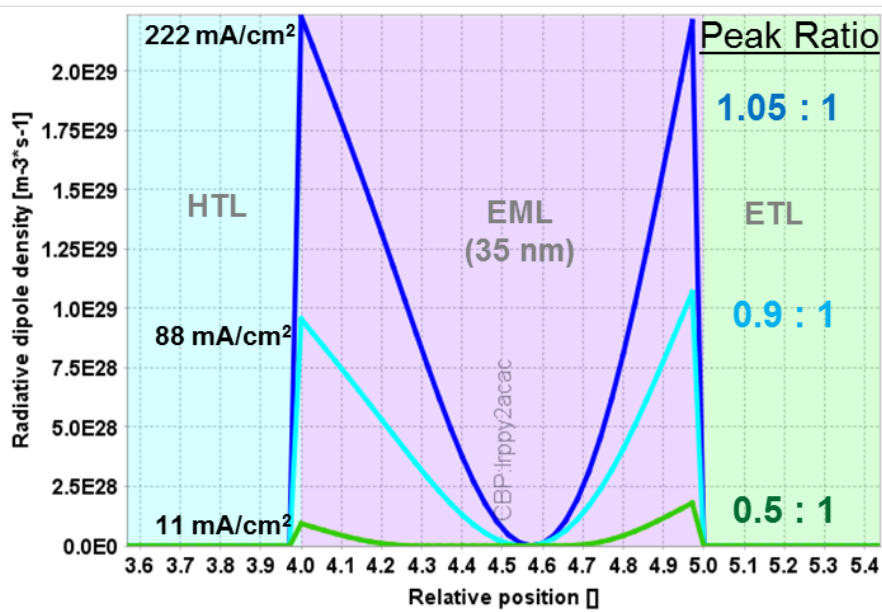


Fig. 1: Extracted dipole density distribution for three different current densities. At low currents, the maximum dipole density is located at the EML/ETL interface, whereas for the high currents it shifts to the HTL/EML interface, as expressed with the peak ratio.

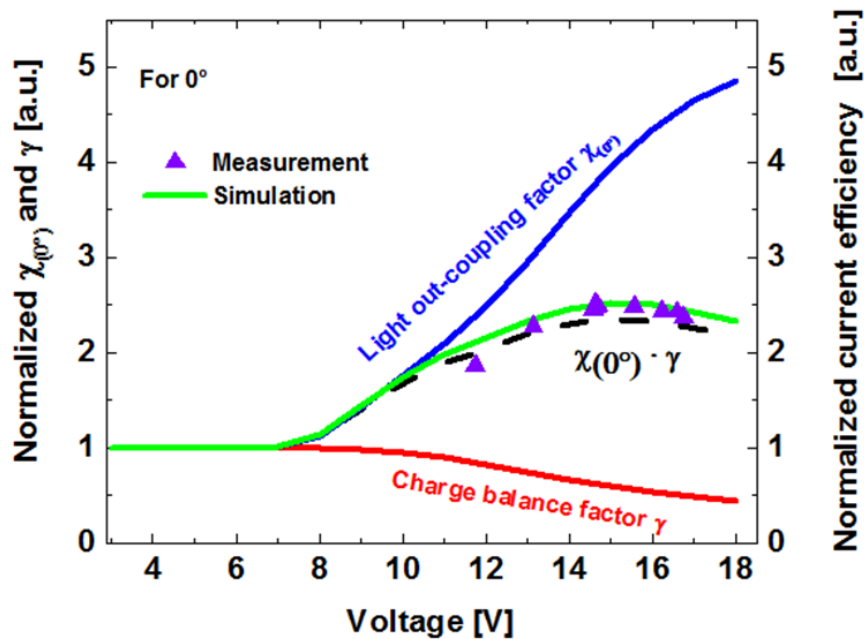


Fig. 2: Simulated product $\gamma * \chi(0^\circ)$ of the light out-coupling factor increase and charge balance factor reduction, is in very nice agreement to the measured current efficiency over the entire measurement range.

Precise determination of the molecular orientation in organic thin films by simulating the spectral radiant intensity of finite thickness emission layers

Christian Hänisch, Cornelius Fuchs, Christoph Wellm, Markas Sudzius,
Simone Lenk, Sebastian Reineke

Dresden Integrated Center for Applied Physics and Photonic Materials (DC-IAPP) and
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Organic light-emitting diodes (OLEDs) belong to the most recent generation of light sources and are already versatily used in display applications¹. The overall efficiency of such devices is limited by the organic material's high refractive index causing a trapping of large portions of the initially emitted light. One approach reducing this loss channel is to use emitter molecules whose transition dipole moment is aligned parallel to the interface planes of the OLED's multi-layer geometry and, hence, decreasing the total internal reflection at the interfaces.

Finding methods and techniques to actively control the molecular orientation requires a stable and reliable measurement of this very quantity. Comparing the reported orientation values reveals in some cases discrepancies for one and the same material^{2,3,4}. Clearly, a more standardized measurement procedure is needed in order to enable a more systematic investigation of materials and processing techniques.

In the presented work, a specific orientation determination method initially proposed by Frischeisen *et al.*⁵ is investigated in detail. It is based on the measurement of the thin film's angular resolved photoluminescent emission spectrum denoted as spectral radiant intensity (SRI) and a numerical simulation of the latter. The so-called anisotropy coefficient serves as fitting parameter and represents the orientation of the emitter dipole.

¹ Y. Cho, T. U. Daim and P. Sklar, "Forecasting OLED TV technology using bibliometrics and Fisher-Pry diffusion model", 2015 Portland International Conference on Management of Engineering and Technology (PICMET), Portland, OR, 2015, pp. 2167-2176.

² A. Graf, P. Liehm, C. Murawski, S. Hofmann, K. Leo and M.C. Gather, "Correlating the transition dipole moment orientation of phosphorescent emitter molecules in oleds with basic material properties", Journal of Materials Chemistry C: Materials for optical and electronic devices, 2:10298 10304, 2014.

³ T. Lampe, T.D.Schmidt, M.J. Jurow, P.I. Djurovich, M.E. Thompson, and W. Brütting, "Dependence of phosphorescent emitter orientation on deposition technique in doped organic films", Chemistry of Materials, 2016.

⁴ K.H. Kim, S. Lee, C.K. Moon, S.Y. Kim, Y.S. Park, J.H. Lee, J.W. Lee, J. Huh, Y. You, and J.J. Kim. "Phosphorescent dye-based supramolecules for high-efficiency organic light-emitting diodes", Nature Communications, 5:4769, 2014

⁵ Frischeisen, Jörg, et al. "Determination of molecular dipole orientation in doped fluorescent organic thin films by photoluminescence measurements", Applied Physics Letters 96.7 (2010): 073302.

The optical simulation is very sensitive to both the thicknesses of the involved organic layers and the distribution of the emitter dipoles within the emission layer. A homogeneous distribution of dipoles is compared to an exponentially decreasing dipole strength considering the absorption of the excitation light. Furthermore, the simulation of the whole emission spectrum is compared to the exclusive consideration of the emission peak wavelength.

The presented approach delivers anisotropy coefficients with a high statistical stability for emission layers with different thicknesses and on varying sub-layers.

Doping evolution and junction formation in stacked cyanine dye light-emitting electrochemical cells

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Light-emitting electrochemical cells (LECs) are receiving interest because of their peculiar functional principles that allow for low driving voltages, the usage of air-stable contacts and process-tolerant device fabrication. After a long-standing debate consensus has been reached that the operational principle of LECs can best be described by an electrochemical doping model (ECD). In this model ionic charges drift to the respective electrodes upon applying a driving voltage where they facilitate electronic charge injection. Injected charges cause p- and n-doped regions next to the anode and cathode, respectively, which are locally compensated by ionic charges. In the central device intrinsic region charges recombine radiatively and light emission occurs. The fundamental requirement for the active material is the ability to carry both ionic and electronic charges. This has been realized by admixing a salt to a light-emitting polymer or by using ionic transition metal complexes. Recently, the use of small molecules as the active component has been demonstrated as well^{1,2}.

Here, we studied LECs based on cyanine dyes. Cyanine dyes are fluorescent and charged semiconducting molecules that are accompanied by a counter anion. Therefore, cyanines have intrinsic built-in ionic and electronic charge conductivity. We demonstrate that cyanine LECs follow the predictions of the ECD model and use electro- and photoluminance, attenuation and capacitance measurements to determine the intrinsic layer thickness and doping concentrations. We present a new method based on photocurrent spectral response measurements and optical simulation to determine the position of the intrinsic junction region in an operating device. We suggest that the high reactivity of neutral radicals formed in the n-type doped region results in irreversible consumption of the active material during operation which currently limits the long-term device stability.

A drawback of using cyanine dyes as emissive material is their low photoluminescence quantum efficiency (PLQE) resulting in poor LEC performance. To overcome this intrinsic limitation we use a host-guest approach successfully presented for OLEDs and LECs^{3,4}. By varying the host material and optimizing the guest concentration in cyanine-cyanine blends we find promising PLQE of >30%. LECs comprising this blend approach their theoretical external quantum efficiencies if operated at constant current bias. This work demonstrates the possibilities and the limitations of using cyanine dyes as active layer material in LECs.

¹ Tang, S.; Tan, W.-Y.; Zhu, X.-H.; Edman, L. *Chemical Communications* 2013, 49, 4926.

² Pertegas, A.; Tordera, D.; Serrano-Perez, J. J.; Orti, E.; Bolink, H. J. *Journal of the American Chemical Society* 2013, 135, 18008

³ Pertegas, A.; Yin Wong, M.; Sessolo, M.; Zysman-Colman, E.; Bolink, H. J. *ECS Journal of Solid State Science and Technology* 2016, 5 (1), R3160-R3163

⁴ Tang, S.; Buchholz, H. A.; Edman, L. *Journal of Materials Chemistry C* 2015, 3, 8114

Emission characteristics of light-emitting electrochemical cells

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The luminous power conversion efficacy (PCE), i.e. the conversion efficiency from input electric power to output visible light, is one of the most important figures of merit for light sources. Ideally, an integrating sphere is used for its measurement, but often the simpler experimental procedure of a forward-direction luminance measurement combined with an assumed angular emission profile is employed instead. Organic surface-emitting devices such as light-emitting diodes and light-emitting electrochemical cells are commonly assumed to emit with a Lambertian emission profile, i.e. the luminance is constant with respect to the viewing angle¹. This results in a conversion factor of π /voltage between the forward luminous current efficacy and PCE. However, the validity of the Lambertian assumption is seldom verified, despite that several studies on organic light-emitting diodes have reported that it can be incorrect².

By using an in-house developed spectroscopic goniometer and software, we examine the angular dependence of the luminance and the electroluminescence spectrum of a range of light-emitting electrochemical cells, and show that their emission-characteristics strongly depend on the choice of components and configurations, e.g., the thickness of the emitting layer, and the choice of electrode and substrate. Our results indicate that the Lambertian assumption is reasonable for standard bottom-emitting light-emitting electrochemical cells featuring a flat glass substrate, an ITO anode and a reflective top cathode. However, it often results in a significant underestimation of the PCE of devices that, e.g., comprise nanowire electrodes or structured substrates. Specifically, we find that the PCE value of such devices can be underestimated with up to 40% by using the Lambertian assumption. Therefore, we urge researchers interested in the study of the efficiency of organic light-emitting devices to either employ an integrating sphere setup, or, if that it is not possible, to investigate the emission profile of the device before calculating and reporting the important PCE value.

¹ S.R. Forrest, D.D.C. Bradley, M.E. Thompson, "Measuring the Efficiency of Organic Light-Emitting Devices", *Adv. Mat.*, 15(13), 1043–1048, (2003)

² T. Tsutsui, K. Yamamoto, "Evaluation of true power luminous efficiency from experimental luminance values", *Jap. J. of Appl. Phys.*, 38(5A), 2799–2803, (1999)

The role of in-operando energy band diagrams for a consistent drift-diffusion description of organic semiconducting layers

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The drift-diffusion approach is a widely-used procedure to calculate current-voltage (IV) as well as capacitance-voltage (CV) characteristics of organic semiconducting devices. Compared to other approaches drift-diffusion reveals less time-consuming calculations and therefore allows data fitting of electrical measurements followed by extraction of material parameters. However, the simple drift-diffusion approach oftentimes leads to unsatisfying fitting results. A consequence of this is the more complex description of some specific model parameters like carrier mobility (for bulk properties) or barriers (for injection properties). Especially in the steady-state case the introduction of more complex parameters oftentimes leads to a better description of the electrical measurements. In contrast to this for transient measurements the introduction of additional features (e.g. time dependent trap dynamics) seems to be inevitable for a curve shape closer to measured data.

Instead of considering more detailed material specific influences on carrier mobility or injection barriers in this presentation we follow a different approach reflecting the device properties of typically measured samples. The core of the presented ideas is the consistency of the respective in-operando energy band diagrams in every operational state. The consistency is represented by the continuity of the electrochemical potential in the device and the validity of the first law of thermodynamics. We apply the ideas to a simple unipolar single layer device structure. We show how the idea of energy band diagram consistency influences the IV behavior of the device in comparison with other widely-used drift-diffusion approaches which are frequently discussed in literature. Furthermore, a deeper insight into the potential and field distributions of the in-operando device structure is given. Finally, we exemplarily show how the consistency ideas influence the transient CV measurements.

Simultaneous Extraction of DOS Width and Injection Barrier in OTFTs

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Introduction

Recently it has been shown that Capacitance–Voltage measurements can be exploited to assess the width of the Density of States, in the framework of a Gaussian-shaped DOS, in Metal–Insulator–Semiconductor structures for a given value of the injection barrier [1]. Performing the fitting of experimental data to static numerical simulations enables to solve the entanglement between the Density of States and the mobility: while the latter is deduced from Capacitance–Voltage measurements and simulations, the former is fitted by comparison of numerical and experimental Current–Voltage (I–V) transcharacteristics of OTFTs in the linear operation regime. In

[1] it was also shown that the extracted width of the DOS is dependent on the correct knowledge of the metal–semiconductor injection barrier. Here we first show, by extending the numerical model to the non quasi–static regime, that values of the injection barrier deduced by simply comparing the nominal metal work function and the semiconductor electron affinity lead to inconsistencies between the numerical model and experimental measurements of Capacitance–Voltage characteristics at medium frequency. We then show that a more accurate modeling of charge injection and the field dependence of the metal–semiconductor Schottky barrier enables to improve the fitting of measured linear OTFT transcharacteristics. Using the barrier value corresponding to the best fit of the I–V curves allows to extend the validity of the non–stationary model to a wider range of frequency. Contact resistance computations are also used as a further validation of the extracted barrier height.

Results

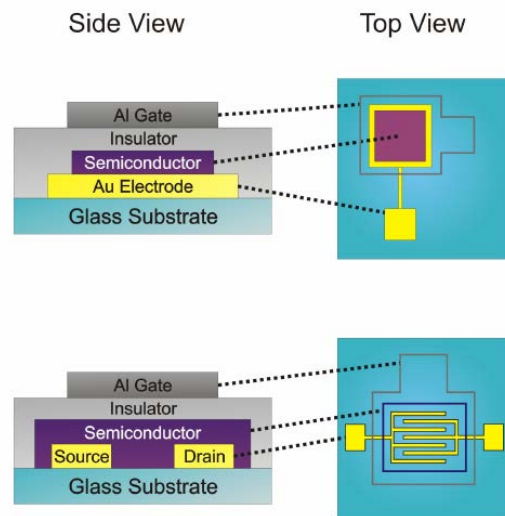


Figure 1: Top and side view of the devices used: the MIS capacitor at the top and the OTFT at the bottom. Reprinted with permission from ref. [1]. Copyright Elsevier 2015.

The devices being considered in this study are shown in figure 1, the MIS capacitor at the top is used for CV measurements and simulations, while the OTFT at the bottom is used for IV measurements and simulations. Notice that in both cases 1D simulations are carried out, being performed along a cross-section normal to the semiconductor-insulator interface for the MIS capacitor and in a cross-section along the source-drain direction for the OTFT. Figure 2 shows Capacitance-Frequency curves for the MIS device, assuming different values of the nominal barrier height Φ_B . Notably, for values of Φ_B close to what could be deduced by simply comparing the nominal metal work function and semiconductor electron affinity, the model predictions appear to be inconsistent with experiments, while for sufficiently small barrier values CF curves become almost indistinguishable and show good agreement with experimental data. Within this range an optimal value can be found by assessing the quality of the fit of simulated IV curves with respect to experimental ones: figure 3 shows the residual of the least-squares fit, which has a well defined optimum corresponding to $\Phi_B = 0.53$ eV.

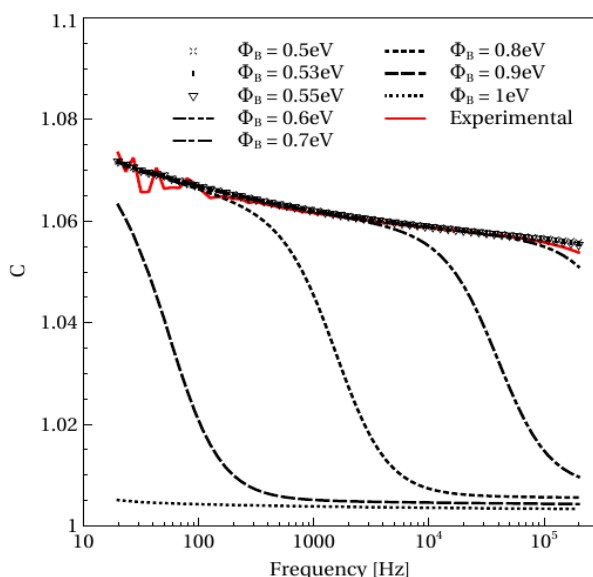


Figure 2: Capacitance-Frequency characteristics of the MIS device in the high accumulation regime computed for various values of the injection barrier Φ_B . Measured accumulation Capacitance-Frequency characteristics shown for comparison.

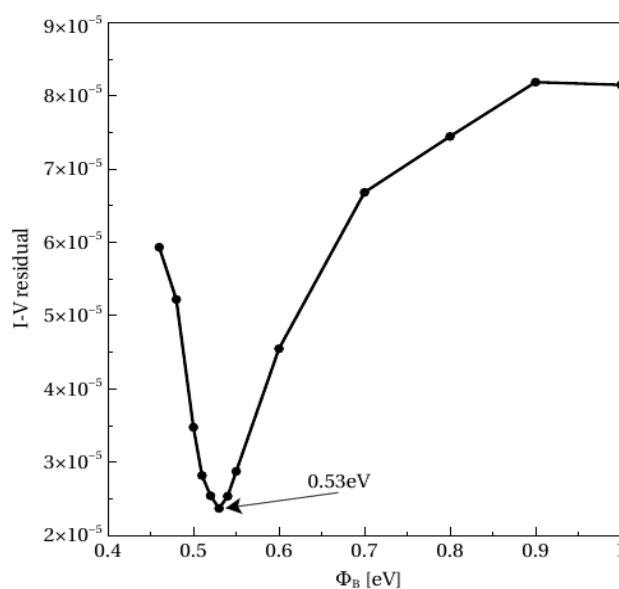


Figure 3: Residual of the least-squares fit of the IV transcharacteristics at various values of the injection barrier Φ_B .

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Non-Equilibrium Charge Carrier Kinetics in a Drift-Diffusion Model of Organic Disordered Semiconductors

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Charge carrier relaxation within the density of states is a common process for energetically disordered materials and can happen on timescales from nano- to microseconds. The relaxation kinetics have an influence on charge transport and carrier recombination for many experimental paradigms as for example transient current measurements. They are, however, not captured by standard drift-diffusion simulations widely used for semiconductor device modeling. This shortcoming limits the range of experiments that can be correctly modeled with the drift-diffusion approach to those that are conducted strictly in thermal equilibrium. We implemented the description of carrier relaxation into a drift-diffusion model and simulated time-of-flight transients and photodetector response currents in the time domain. In this contribution, we describe our algorithm and report a comparison of two approaches to describing relaxation: the explicit transport energy model¹ and a method using an effective energy of localized carriers ϵ^* ². We show that the full transport energy model can be replaced by the effective energy approach when no continuous carrier generation is present, the effective energy approach requiring much lower computational cost. For both models, the experimental data is reproduced accurately. In conclusion, our algorithm enables fast drift-diffusion simulation of time-dependent electronic measurements for systems in and out of equilibrium. This capability makes the prediction of experimental results from techniques like transient charge extraction possible and contributes to a more clear interpretation of these measurements.

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Origins of Negative Capacitance in Organic Single Layer Devices

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In order to characterize an organic semiconductor device, a number of different techniques are available and commonly performed. In admittance spectroscopy, negative capacitance values are often observed at high bias and low frequency as shown in Fig. 1 and have started a controversial debate. A wide range of origins for the negative capacitance have been brought forward for organic semiconductor devices¹⁻⁹.

In this contribution we give an overview of possible origins and investigate them with the aid of numerical simulations. For the analysis we employ a 1D drift-diffusion model and consider single-layer devices in the uni- and bipolar case. Moreover, we show that the presence of charge trapping impacts the occurrence of negative capacitance.

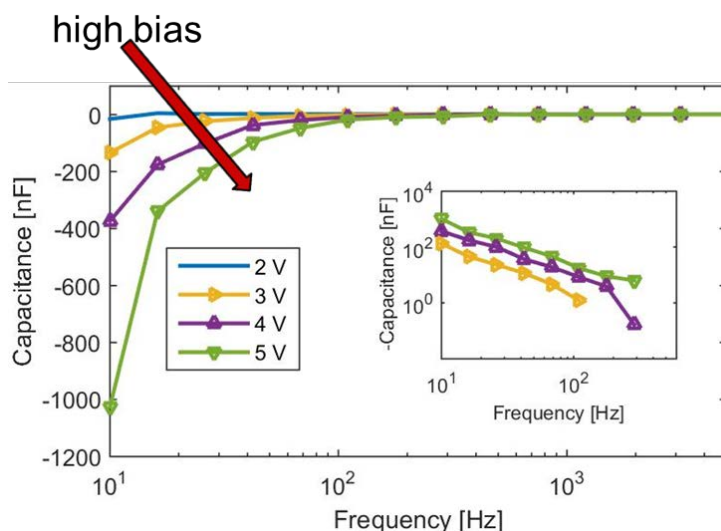


Fig. 1 Negative capacitance values of a hole-only device at low frequency. With increasing bias the effect becomes more distinct.

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A Critical Look at the Mott-Schottky Analysis for Extraction of Background Doping in Organic Diodes

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Background doping in organic semiconducting films can have significant impact on device performance especially thin film transistors. Capacitance-Voltage ($C-V$) characteristics is commonly used to estimate this concentration of unintentional doping through Mott-Schottky (M-S) ($1/C^2 - V$) plot¹. M-S analysis, originally derived for conventional inorganic semiconductor diodes, is based on the depletion approximation near Schottky contact, shallow impurities which readily ionize, absence of traps etc which if violated can give rise to an erroneous estimate. In the present work, we propose a simple consistency check between the estimated doping level and peak capacitance (C_{peak}) magnitude to determine the validity of Mott Schottky analysis.

Drift-diffusion simulations were employed to emphasize that C_{peak} is affected quite considerably with energetic depth of doping in a device. To illustrate the proposed idea we now assume that no a priori information is available about the nature of doping levels. Corresponding M-S analysis of Fig. 1(a) shows that extracted doping concentration for one $C-V$ curve is $3.96 \times 10^{17} \text{ cm}^{-3}$ whereas for other it is $1.75 \times 10^{17} \text{ cm}^{-3}$. A consistency check between extracted values of doping and expected C_{peak} value can be done through the numerically simulated universal curve, shown in Fig. 1(b), which tells that (normalized) C_{peak} is independent of thickness variations and is only dependent on (normalized) doping density as long as inherent assumption of shallow doping level in M-S analysis is true. It should be noted from universal curve that for extracted doping density of $3.96 \times 10^{17} \text{ cm}^{-3}$ expected C_{peak} turns out to be nearly seven times geometric capacitance (C_{geo}) and matches fairly with the simulated results for shallow energy level of doping where input density is $4 \times 10^{17} \text{ cm}^{-3}$, see Fig. 1(a), which is an indication of the validity of M-S analysis. Similarly, for extracted density of $1.75 \times 10^{17} \text{ cm}^{-3}$ universal curve shows expected C_{peak} to be nearly 4.5 times of

C_{geo} for M-S analysis to be valid. But this prediction doesn't match either of the C_{peak} values of Fig. 1(a) which questions validity of M-S analysis where deep energy level of doping is present. With reference to these observations a self-consistent method is proposed to analyze experimental $C-V$ results in conjunction with proposed universal curve so that elevation in C_{peak} can be truly comprehended in terms of doping density. Furthermore, possibility of violations in proposed protocol is also addressed through a more practical scenario where both doping levels and traps are present in the device.

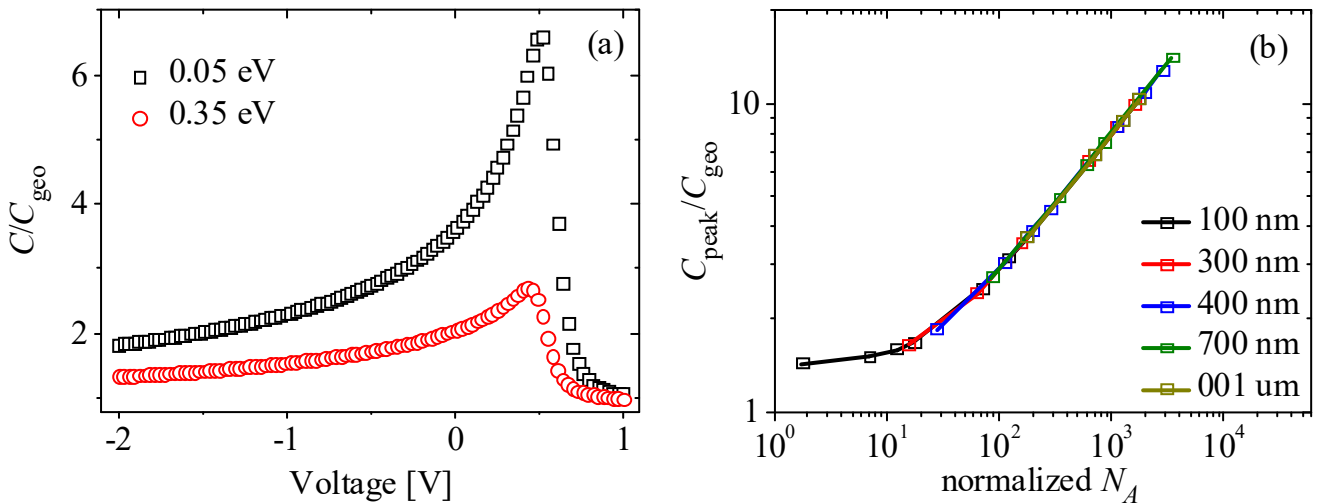


Figure 1. (a) Ratio of capacitance to geometrical capacitance ($C_{geom} = \epsilon A / L$) against voltage for different energetic levels with acceptor concentration $N_A = 4 \times 10^{17} \text{ cm}^{-3}$ for film thickness $L=100 \text{ nm}$ and (b) Ratio of capacitance peak to geometrical capacitance versus acceptor concentration at 300 K; normalized with $\epsilon k_B T / q^2 L^2$ where ϵ is the permittivity, k_B is the Boltzmann constant, T is temperature, q is elementary charge and L is the organic layer thickness; with shallow doping level of 50 meV from highest occupied molecular orbital. These results are simulated for hole only diodes where cathode barrier for holes at 0.9 eV and anode barrier is at 0 eV.

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How charge carrier transport and electrode selectivity influence the performance of (organic) solar cells

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I) Organic semiconductors used in the photoactive layer of organic solar cells usually comprise rather low charge carrier mobilities. Under typical operation conditions this can lead to considerable accumulation of charge carriers causing enhanced recombination. An important consequence is that the voltage applied at the contacts does not equal the voltage inside the photoactive layer. For this reason the current-voltage characteristics of such a transport-limited solar cell differs significantly from the well-known Shockley diode equation. An analytical model will be presented which considers the effect of limited mobility explicitly and which allows determining the correct value for the voltage required for the application of the Shockley equation¹. In addition, the model can be used to evaluate efficiency potentials in a more realistic manner.

II) Surface recombination is a loss mechanism in addition to the recombination (radiative and non-radiative) in the bulk of the photoactive layer². To minimize it in organic solar cells charge carrier selective layers are used between the photoactive layer and the electrode. These layers (PEDOT:PSS, TiO_x, ZnO, WO_x, MoO_x, etc.) often have a rather large band gap and thus can block the “wrong” type of charge carrier quite efficiently. However, surface states within the band gap of these materials at the interface with the photoactive layer can still act as recombination centers. Recently, there are numerous examples of polar (organic) molecules providing a high degree of charge carrier selectivity, one prominent example being the work of He et al. on PFN³. Some of these materials are however rather expensive. We used simple, harmless, extremely cheap and easy-to-process organic molecules with permanent dipole moments. These molecules alter the effective work function of the electrode as confirmed by scanning Kelvin probe force microscopy and ultraviolet photoelectron spectroscopy (UPS) which revealed a corresponding shift of the surface potential. This leads to a strong increase (decrease) of the electron (hole) concentration in the adjacent photoactive layer. It will be shown in detail that this is the main reason for the enhanced selectivity causing an increase of the open-circuit voltage (and fill factor) for a variety of different photoactive materials. A theoretical model was set up and the results of the numerical simulations are in full accord-

ance with the experimental data. Interestingly, DFT-calculations prove that the energy levels of the dipole molecules used are not suited to conduct charge carriers from the photoactive layer to the electrode but that a tunneling mechanism can be expected to be involved. Implementing this into our model, it is found that there is no necessity to assume preferential tunneling for electrons. Their accumulation and the depletion of holes due to the altered work function are sufficient to explain the observed behavior⁴.

III) This model of different electrode selectivities can - without the dipole molecules - also be applied to explain the often observed hysteresis in perovskite solar cells as ion migration reduces or increases the contact selectivity depending on the direction of their movement.

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Diffraction Gratings for Enhanced All-Season Energy-Harvesting in OPV Devices

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Already for standard solar test conditions, the application of diffraction gratings as light incoupling structure can enhance the optical absorption of organic solar cells, yielding a 12% increase in device efficiency.¹ However, the use of diffraction gratings as light-management films for OPV reveal their full potential even more, when solar movement and various operating conditions are taken into account.

In this work we present a simulation procedure based on the software SETFOS from Fluxim AG, which combines the diffractive optics of the grating with the wavelength and angle dependent field distribution in the organic solar cell to first predict the enhancement profile of the EQE with respect to a pristine device. The nice agreement at measurable lab conditions further suggests the calculation of the current generated with the grating for different conditions throughout the year, resulting in an increase of about 12% in the yearly harvested power with respect to a reference device.

Moreover, we show that the grating properties can be tailored by several parameters to perfectly take into account the illumination conditions of various applications (automotive, façade, consumer electronics, shading) resulting in optimized yearly-integrated energy harvesting.

Finally, we will consider the integration of our light harvesting solution with conductive grid electrodes in large area devices and we will present first preliminary simulation results obtained with the software LAOSS from Fluxim AG.

¹ Mayer et al., Opt. Express 24, A358-A373 (2016)

Shedding light on the stability of organic solar cells

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Stability is one of the major challenges for research in organic solar cells today. These devices can degrade due to effects in the organic absorber layer itself but also due to processes happening in the transport layers and electrodes. The different processes can be triggered by light, by light combined with oxygen (and humidity), by device current, or can happen even in the dark in nitrogen atmosphere. It is therefore crucial to set controlled experimental conditions for investigating OPV degradation. Furthermore it is a necessity to perform systematic and controlled measurements.

In order to learn about device and degradation physics in organic solar cells several analytical models for parameter extraction as well as numerical simulations have been employed. However it has been found that extracted parameters can be inaccurate and unreliable, when using only one single measurement technique like IV-curves¹. We have shown an approach that combines a multitude of electrical characterization techniques in the steady-state, transient and frequency domain. Performing a global and synoptic analysis allows to extract parameters and draw conclusions more reliably².

For our studies we have developed and employed the measurement platform Paios³ which is able to perform steady-state, transient and impedance experiments in automated and systematic measurement routines has been developed and employed. We further use the fully coupled optoelectrical drift-diffusion simulation tool Setfos⁴ which allows to model all experiments with a single set of parameters.

Here we present that by comparing experiment with simulation we are able to extract valuable information about the physics behind degradation of organic solar cells, without the need for further expensive or destructive measurement techniques. This is possible as we relate specific degradation processes to changes in specific parameters. The different mechanisms all lead

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⁴ Setfos 4.3, www.fluxim.com, 2016

to degradation in the IV-curve and device efficiency, but can be identified by their specific signatures in the various measurement techniques. This combinatorial approach thus allows to distinguish between processes that have the same signature in the IV-curve.

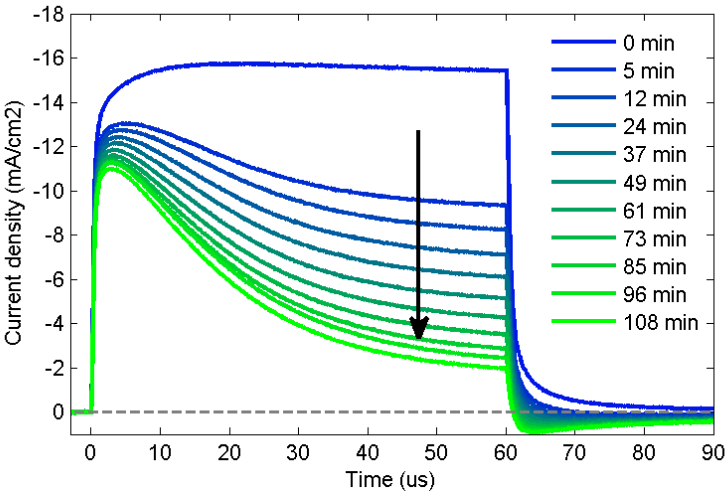


Fig. 1 Transient photocurrent response to a light pulse cell during degradation.

Molecular understanding of heterostructures of organic semiconductors

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We will discuss the role of mesoscale order, electrostatic effects, defects, and roughness for charge splitting and detrapping at donor-acceptor interfaces. We will show how inclusion of mesoscale order resolves the controversy between experimental and theoretical results for the energy-level profile and alignment in a variety of photovoltaic systems, with direct experimental validation [1,2]. We predict open-circuit voltages of planar heterojunction solar cells in excellent agreement with experimental data, based only on crystal structures and interfacial orientation. We show how long-range molecular order and interfacial mixing generate homogeneous electrostatic forces that can drive charge separation and prevent minority carrier trapping across a donor-acceptor interphase [2]. Comparing a variety of small-molecule donor-fullerene combinations, we illustrate how tuning of molecular orientation and interfacial mixing leads to a trade-off between photovoltaic gap and charge-splitting and detrapping forces, with consequences for the design of efficient photovoltaic devices. By accounting for long-range mesoscale fields, we obtain the ionization energies in both crystalline and mesoscopically amorphous systems with high accuracy [4,5].

References

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- [2] C. Poelking, D. Andrienko, *J. Am. Chem. Soc.*, 137, 6320, 2015
- [3] M. Schwarze, W. Tress, B. Beyer, F. Gao, R. Scholz, C. Poelking, K. Ortstein, A. A. Guenther, D. Kasemann, D. Andrienko, K. Leo, *Science*, 352, 1446, 2016
- [4] P. Kordt, J. J. M. van der Holst, M. Al Helwi, W. Kowalsky, F. May, A. Badinski, C. Lennartz, and D. Andrienko, *Adv. Funct. Mater.* 25, 1955, 2015
- [5] C. Poelking, D. Andrienko *J. Chem. Theory Comput.*, DOI: 10.1021/acs.jctc.6b00599

Organic heterojunctions: Contact-induced molecular reorientation, interface states, and charge re-distribution

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The planar heterojunction formed between the hydrogen and fluorine terminated copper phthalocyanines has been found to behave as charge generation layer¹. Therefore, this interface was investigated by ultraviolet photoelectron and X-ray absorption spectroscopy². Pinning at the Fermi level of the underlying electrode is observed for both materials—one p-type and the other one n-type. This results in a sheet charge density at the organic/organic interface due to interfacial charge transfer. An interlayer with co-facial intermolecular arrangement, which differs from the respective bulk structures, at the interface was found by both spectroscopy techniques; this interlayer, noteworthy, is unpinned.

The combined experimental approach results in a comprehensive model for the electronic and morphological characteristics of the interface between the two investigated organic semiconductors. Additionally, electrostatic simulations confirm the effects of charge accumulation and vacuum level shifts. The presence of a π -orbital stacking between different molecules at a heterojunction is also of interest for photovoltaic active interfaces or for ground-state charge-transfer.

¹ A. Opitz, B. Ecker, J. Wagner, A. Hinderhofer, F. Schreiber, J. Manara, J. Pflaum, and W. Brütting, “Mixed crystalline films of co-evaporated hydrogen- and fluorine-terminated phthalocyanines and their application in photovoltaic devices”, *Org. Electron.* 10, 1259–1267, (2009)

² A. Opitz, A. Wilke, P. Amsalem, M. Oehzelt, R.-P. Blum, J. P. Rabe, T. Mizokuro, U. Hörmann, R. Hansson, E. Moons, and N. Koch, “Organic heterojunctions: Contact-induced molecular reorientation, interface states, and charge re-distribution,” *Sci. Rep.* 6, 21291, (2016)

2D Model of Bulk Heterojunction Organic Solar Cells: Importance of the Donor-Acceptor Interface Length

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In this communication, we present a study conducted with our two-dimensional model of an active layer of a bulk heterojunction (BHJ) organic solar cell¹. The model is based on an elementary unit composed of a donor and an acceptor domains separated by a non-planar interface (see Fig. 1a). The mechanism considered in the model takes explicitly into account the existence of charge transfer (CT) states pinned at this interface and restrains the motion of electrons and holes to their respective domains only (see Fig. 1).

The elementary unit, with its non-planar interface, is assumed to reproduce the mean effect of complex morphologies encountered in real BHJ active layers. In order to check this assumption and since such morphologies are quite complicated to extract, we generate artificial morphologies by considering a sinusoidal interface of amplitude A and periodicity N_{per} and choosing various values for these parameters. Then the different electrical responses, each being associated with an artificial unit, can be combined to obtain the global response of more realistic structures.

Finally, the study shows that taking an elementary unit with adequate geometrical parameters allows to reproduce, with excellent agreement, the electrical response of a complex morphology. The interface length of the elementary unit is found to correspond to the average value of the interface length of the realistic active layer.

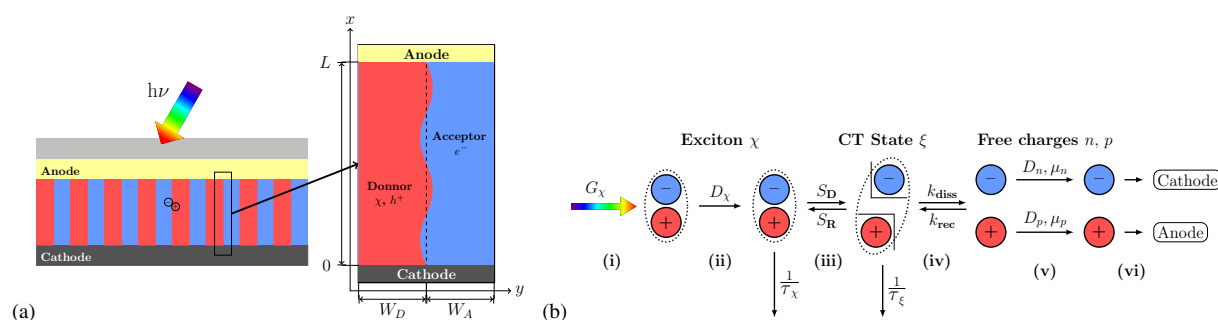


Fig. 1 (a) Geometry of the elementary unit used to model an ideal BHJ organic solar cell. (b) Balanced equations summarizing the mechanism of charge generation in a realistic BHJ active layer.

¹A. Raba, Y. Leroy, and A.-S. Cordan, "Organic Solar Cells: a Rigorous Model of the Donor-Acceptor Interface for Various Bulk Heterojunction Morphologies" J. Appl. Phys. 115, 054508, (2014).

The impact of recombination on the Fill Factor of organic-based solar cells

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Organic-based solar cells have attracted increasing interest over the last few decades, due to their promisingly low fabrication costs, and their innovative flexibility and esthetical possibilities. The mechanisms behind the performance limits of these devices have been investigated, and the major responsables for low open circuit voltage and short circuit current could be identified. However, the Fill Factor of these solar cells has still a more controversial origin.

Some groups have proposed the link between this performance parameter and recombination losses^[1], or low charge carriers' mobilities^[2].

Here, we propose an alternative look into the effect of recombination on the determination of organic solar cells' Fill Factor, by comparing the influence of different recombination pathways (namely bimolecular, trap-assisted, and geminate) through the use of a simulation program developed in the house, *Simiconductor*^[3], which solves the drift-diffusion equations for an equilibrium condition. Alongside the introduction of the various losses, we take care of considering the impact that residual doping may have on the relation between Fill Factor and recombination.

The proposed simulations can contribute towards a better understanding of the mechanisms responsible for the low Fill Factors, which often characterize organic photovoltaic devices. This understanding is of paramount importance towards the design of strategic roadmaps for more efficient solar cells.

[1] D. Bartesaghi, I. D. C. Pérez, J. Kniepert, S. Roland, M. Turbiez, D. Neher, L. J. A. Koster, *Nat. Commun.* **2015**, *6*, 7083.

[2] J. a. Bartelt, D. Lam, T. M. Burke, S. M. Sweetnam, M. D. McGehee, *Adv. Energy Mater.* **2015**, n/a–n/a.

[3] J. Liesenborgs, "Simiconductor," can be found under <http://research.edm.uhasselt.be/~jori/simiwebtest/>

Opto-Electrical Properties of Rectifying Antenna

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The conversion of solar energy to electrical energy has been focused on photo voltaic effect using silicon and other semiconductor materials. Using such effect the upper limit of photo-conversion efficiency is predicted to be around 30% for single junction silicon solar cells. The photo voltaic effect utilizes the photonic nature of solar radiation. However, one can also exploit the electromagnetic nature of radiation to directly convert it to electrical energy. In this work, we study and develop 3rd generation solar cells composed of plasmonic nano-antenna associated with self assembled rectifying molecular diodes (rectenna) to directly convert light to electric energy. The efficiency limit imposed by band gaps in photo voltaic effect could be eliminated using rectenna as it is not based on the use of semi conducting materials. Studies have been made in rectenna based on Metal-Insulator-Metal tunneling diodes¹. However, there is lack of study in opto-electrical properties of rectenna structure. Here, using an opto-electrical model, we study whether molecular diodes associated with plasmonic nano-antennas can be used as rectennas to convert light into electricity in visible and infrared regime.

First, the geometry of plasmonic nano-antennas has been optimized using FDTD method. We systematically study the different antenna structures for example, dipole antenna with integrated waveguide, pyramidal Metal-Insulator-Metal antennas. We also study the effect of different antenna parameters in coupling of incident light into waveguide structure consisting rectifying elements. Then, using barrier tunnel model we optimize the design parameters of the molecular rectifiers^{2,3} such as their size and the energy position of their molecular orbitals.

The study of the physical phenomena that govern the electrical properties of the Metal-SAM-Insulator-Metal diodes is crucial to evaluate the conversion efficiency of the molecular rectennas. A 1D transfer matrix code (TMM) has been developed and serve to optimize the rectification ratios of the diodes. Ferrocenyl-containing alkenathiol have been consider in the simulations. We compare the calculated I(V) characteristics of the diodes under darkness with experimental measurements and we give an insight into the physical phenomena occurring in ferrocenyl-alkanethiols based molecular diodes. Especially, we investigate through simulation how bound and quasi-bound states

of the electronic levels of the molecules can participate to the charge transport.

Finally, we developed a model integrating the optical and the electrical simulations to evaluate the conversion efficiency of the rectennas. This model is based on a semi-classical tunneling model to take into account a photo-assisted transportation of charges within the molecular diodes.

¹S. Grover et al., “Travelling Wave Metal/Insulator/Metal Diodes for Improved Infrared Bandwidth and Efficiency of Antenna coupled Rectifiers”, IEEE transaction on nanotechnology, vol.9 no. 6, 2010

²Li Yuan et al., “Controlling the direction of rectification in molecular diode”, Nature communication, 2015

³Christian A. Nijhuis et al., “Mechanism of Rectification in Tunneling Junctions Based on Molecules with Asymmetric Potential Drops”, J. AM. CHEM. SOC. 132, 18386–18401, 2010

Charge Carrier Selectivity of Contacts for Organic Solar Cells

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The selectivity of electrodes in photovoltaic device is a crucial factor that can limit the solar cell's performance. If a contact is not selective, electrons and holes from the bulk arrive at the same contact where they recombine non-radiatively. As a matter of fact, this recombination mechanism is an additional loss channel for charge carriers. The surface recombination current under open-circuit conditions is accompanied by a gradient of the quasi-Fermi energies required as driving force for the current flow, as shown in Figure 1.

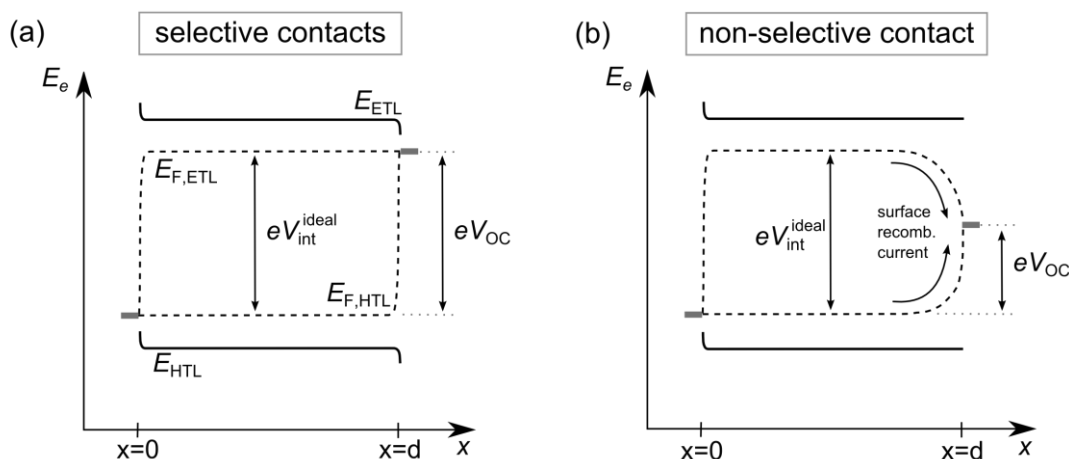


Fig. 1: Scheme of the energy level diagrams under open-circuit conditions for (a) selective contacts and (b) a non-selective electron contact. The photoactive layer is mimicked by using an effective semiconductor model (from $x=0$ to $x=d$) where only the electron transport level ETL and the hole transport level HTL of the free charges are taken into consideration. The hole contact is located at $x=0$ and the electron contact at $x=d$.

This results in a reduction of the open-circuit voltage and in a lower efficiency of the solar cell. Several authors have already reported that there is an optimum finite mobility for non-selective electrodes where the efficiency reaches a maximum.¹⁻³ We further investigate this interpretation by thorough simulation data of an effective semiconductor model which is ac-

¹ Deibel, C., Wagenpfahl, A. & Dyakonov, V. Influence of charge carrier mobility on the performance of organic solar cells. *Phys. Status Solidi RRL* 2, 175–177 (2008).

² Wagenpfahl, A., Deibel, C. & Dyakonov, V. Organic Solar Cell Efficiencies Under the Aspect of Reduced Surface Recombination Velocities. *IEEE J. Sel. Topics Quantum Electron.* 16, 1759–1763 (2010).

accompanied by experimental results from charge extraction, electroluminescence and photoluminescence. With these results at hand we demonstrate that two contributions which strongly depend on the transport properties of the photovoltaic device are responsible for the lower open-circuit voltage:

- i) a reduction due to an effective injection barrier and
- ii) a reduction of the bulk charge carrier density.

For significantly low mobilities, the lower open-circuit voltage is a phenomenon which is solely restricted to the close proximity of the electrode's surface whereas the recombination of charge carriers in the bulk is hardly affected. The importance of mobility for the contact selectivity is further examined with respect to the imbalance of mobilities. In the simulation we see that in the case of a hole contact, a higher electron than hole mobility leads to an electrode which is more prone to surface recombination. The reverse holds true for the electron contact. This is especially important for organic solar cells where the donor and acceptor materials commonly possess distinct imbalanced mobilities. In addition, we reveal design guidelines to further improve contact selectivity.

³ Tress, W., Leo, K. & Riede, M. Optimum mobility, contact properties, and open-circuit voltage of organic solar cells: A drift-diffusion simulation study. *Phys. Rev. B: Condens. Matter Mater. Phys.* 85 (2012).

Boundary Value for the free Charge Density in the Modeling of Organic Photovoltaic Device

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Organic solar cells (OSCs) are a viable technology to capture the solar radiation because of many advantages including light weight, flexibility and low manufacturing costs¹. However, for large scale manufacturing of OSCs with predictable performance, accurate physics-based engineering models are needed. In this paper, we analyze the effect of the boundary values of the free charge density employed in the modeling of OSCs. In order to determine the current-voltage characteristics of photovoltaic devices, the set of transport, Poisson and continuity equations must be solved. The application of this set of differential equations (DEs) to OSCs is made by the inclusion of the particular physics parameters of the organic semiconductors and considering the role of the metal-organic (MO) interfaces. The value of the free charge density at the MO interface can be considered as the balance of different physical-chemical mechanisms that take place in it. The main processes are given in Fig 1 and can be classified in two groups: those that favor the injection of charge and those that favor the extraction^{2,3}. Different models for the extraction and injection of charge in the MO interfaces have been developed in order to find proper boundary conditions for the free charge density². Nevertheless, due to the numerical complexity, these models are not used and approximations are preferred in order to obtain a quick solution. For this purpose, Boltzmann approximation or linear relations between the current density and the electron and hole charge densities, $J-n$, $J-p$, respectively, are usually coupled to the DEs. The existence of different types of recombination at the MO interfaces can result in non-linear $J-n$ and $J-p$ relations. In this work, we focus on the effect of the metal-organic interfaces on the boundary values of

¹ M.J. Deen, "Organic Semiconductor Devices," Wiley Encyclopedia of Electrical and Electronics Engineering, Editor, J.G. Webster, John Wiley and Sons, Inc., 17 pp (Published on-line 15 Dec 2014).

² A. Petersen, T. Kirchartz, and T. A. Wagner, "Charge extraction and photocurrent in organic bulk heterojunction solar cells", Phys. Rev. 85, 045208, (2012)

³ P. López-Varo, J.A. Jiménez Tejada, J.A. López Villanueva, M J. Deen, "Space-charge and injection limited current in organic diodes: A unified model" Org. Electron., 15, 2526 (2014).

the free charge density and propose a model accordingly. Recently, we have suggested a power-law relation between the charge density and the current density⁴: $n, p = K_1 J_{n,p}^m + K_2$ for MO contacts. This expression also includes Boltzmann approximation and linear relations $n, p - J_{n,p}$ with $m=0$ and $m=1$, respectively. In previous works, we demonstrated that this relation is required for the interpretation of current-voltage $J-V$ curves in single-carrier metal-organic contacts. Among other effects, this boundary condition for the charge density keeps information about the limited recombination velocity at the contacts and the contribution from space charge limited conduction (SCLC) in the bulk. In diffusion-dominated transport, at low bias close to the diode's built-in voltage, the charge density at the contact is almost constant with the current. The values of m , K_1 and K_2 depend on the specific metal-organic contacts. The final relation between charge density and current density for injecting electrodes, extracted from the analysis of single-carrier diodes, can be used as a boundary condition in bipolar devices. In this work, we have incorporated our power law relation in the modeling of OSCs. We have reproduced experimental $J-V$ characteristics for OSCs in dark and under illumination. Finally, we have checked that the use of non-constant values for the free charge density at the interfaces is especially important close to the open-circuit voltage and in the high-voltage region.

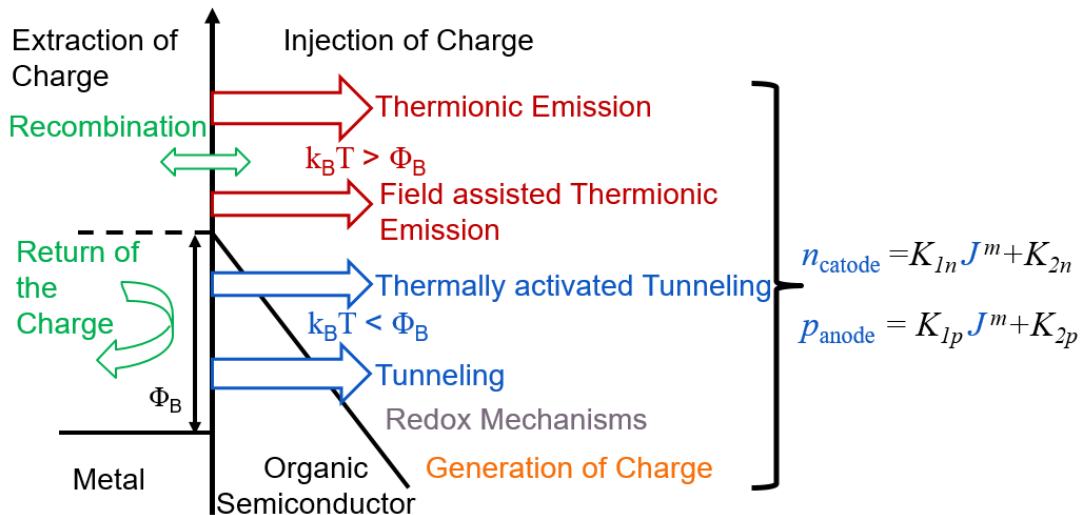


Fig. 1 Extraction and injection mechanisms that take place at the electrode-organic semiconductor interface and the relation that we proposed to enclose all these effects. The metal-organic energy barrier is denoted Φ_B and the thermal energy is given by the product of Boltzmann constant k_B and the temperature, T , $k_B T$. When the thermal voltage is higher than the energy barrier, thermionic emission is given. In the opposite case the tunnelling is dominant.

⁴ P. López-Varo, J.A. Jiménez-Tejada, O. Marinov, C.H. Chen, M.J. Deen, "Charge density at the contacts of symmetric and asymmetric organic diodes" *Org. Electron.*, 35, 74 (2016).

Advanced transparent conductive electrodes for solar cells and OLEDs

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In this presentation, we'll first review the requirements for realising high performance transparent conductive electrodes (TCE) for devices requiring high current density such as OLED or solar cells. We'll show in particular how critical is the level of transparency of the electrodes and how a perfect control of carrier concentration and mobility in transparent conductive oxides (TCO) can allow the achievement of quasi perfect electrodes, possibly in combination with metal grids. We'll comment briefly on the potential of other materials such as metallic nanowires in transparent matrix, or graphene layers and we'll show that these alternative approaches will normally lead to performance losses compared to approaches with optimised TCO. In a second part we'll review some of the latest research performed on the use of both indium based and indium free TCO for application in OLED and Solar cells, demonstrating various kind of single junction and multiple junction devices with state-of-the efficiencies.

Transparent conductive oxides by soft deposition methods

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High-quality transparent conducting oxides (TCOs) such as $\text{In}_2\text{O}_3:\text{Sn}$ (ITO) and doped ZnO are typically deposited by magnetron sputtering, often with additional substrate heating. Plasma-induced damage during sputtering can deteriorate the underlying functional layers of thin film solar cells, whereas the annealing step that is incompatible with flexible, plastic-based substrates desired for organic photovoltaics.

Two examples of “soft” deposition methods of TCOs are presented, which offer a reduced (or absent) plasma damage and do not require substrate heating. First, a solution approach in combination with a UV-annealing treatment is used to deposit conductive Al-doped ZnO (AZO) with resistivity down to 25 Ohm sq and a visible transmission above 90%.¹ The solution-grown AZO layers are implemented as front contacts into inverted PTB7/PC71BM polymer solar cells on PET substrates² (Figure 1). A conversion efficiency of 6.4% and 6.9% is achieved for the indium-free solar cells on PET and glass substrates, respectively. The devices are relatively stable in air whereby an initial efficiency loss in the order of 15% after storage for 15 days can be fully recovered by light soaking.

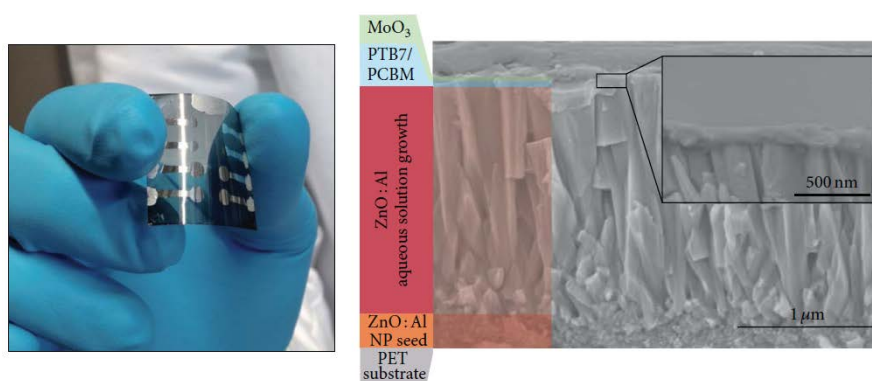


Fig. 1 (left) Photograph of solar cells with solution-processed AZO on PET polymer foil substrate. (right) SEM cross section of the device on PET foil.

¹ H. Hagendorfer et al., “Highly Transparent and Conductive ZnO:Al Thin Films from a Low Temperature Aqueous Solution Approach”, *Adv. Mater.* 26, 632 (2014.)

² P. Fuchs, A. Paracchino, H. Hagendorfer, L. Kranz, T. Geiger, Y. E. Romanyuk, A. N. Tiwari, F. Nüesch, „Indium-Free PTB7/PC71BM Polymer Solar Cells with Solution-Processed Al:ZnO Electrodes on PET Substrates”, *Int. J. Photoenergy*, 2047591 (2016).

Second example is H-doped In_2O_3 (IOH) layers that are deposited at room-temperature by off-axis sputtering in confocal configuration at room temperature. Thanks for the high carrier mobility in as-deposited amorphous IOH, the optical absorption in the visible and near IR range can be reduced, and the 330 nm-thick IOH electrode helps to improve the conversion efficiency of a $\text{Cu}(\text{In,Ga})\text{Se}_2$ thin film solar cell from 15.7% to 16.2% as compared to a reference AZO electrode, also increasing the open circuit voltage by 20 mV.³

³ T. Jäger, Y. E. Romanyuk, S. Nishiwaki, B. Bissig, F. Pianezzi, P. Fuchs, C. Gretener, M. Döbeli, A. N. Tiwari, "Hydrogenated indium oxide window layers for high-efficiency $\text{Cu}(\text{In,Ga})\text{Se}_2$ solar cells", *J. Appl. Phys.* 117, 205301 (2015).

Simulation of lateral charge transport in large-area optoelectronic semiconductor devices

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Summary A one-dimensional mathematical model for the current-voltage characteristics of stacked semiconductor materials is combined with a two-dimensional model for the charge transport in the lateral direction within the thin-film electrodes. Numerical simulation with the finite element method yields the current-voltage characteristic of a large-area device, which takes into account the sheet resistance of the electrodes. The two-dimensional simulation domain can have an arbitrary shape, and it may consist of multiple subdomains with different electrical properties. The numerical simulation may thus facilitate the process of device design.

Problem description The lateral charge transport in thin-film semiconductor devices is affected by the sheet resistance of the various layers. This may lead to a non-uniform current distribution across a large-area device resulting in an inhomogeneous luminance, for example, as observed in organic light-emitting diodes¹. Numerical simulation of this current distribution may assist in the evaluation of various device designs or material combinations, and it may help to reduce the number of expensive trial-and-error stages during device fabrication.

Mathematical model We assume that the steady-state electric current in the stacked semiconductor materials is purely vertical, whereas in the electrodes it is purely lateral. These assumptions allow us to combine a one-dimensional mathematical model for the stack with two-dimensional models for the electrodes. The advantage of such a coupled 1D-2D modeling approach is a much faster simulation than with a full three-dimensional model.

¹ K. Neyts, M. Marescaux, A. U. Nieto, A. Elschner, W. Lövenich, K. Fehse, Q. Huang, K. Walzer, and K. Leo, "Inhomogeneous luminance in organic light emitting diodes related to electrode resistivity", J. Appl. Phys. 100, 114513 (2006)

This model can be further reduced to one electrode by assuming negligible sheet resistance in the other electrode. In this case, we obtain the following semilinear elliptic partial differential equation (PDE) for the electric potential distribution ψ [V] in the electrode:

$$\operatorname{div}(R_{\square}^{-1}\nabla\psi) = -j(\psi). \quad (1)$$

Here, R_{\square} [Ω/\square] denotes the effective sheet resistance of the electrode, which may also vary in the domain. The electric current density j [Am^{-2}] on the right-hand side of (1) is computed from the current-voltage characteristic of the semiconductor material stack.

Numerical simulation We solve the PDE (1) numerically with the finite element method using linear Lagrange elements. To complete (1), several types of boundary conditions can be imposed on the boundary segments. Newton iterations are used to solve the nonlinear system of equations obtained after discretization.

The numerical simulation yields the electric potential distribution on the given geometry and for given boundary conditions – an example is shown in Fig. 1:

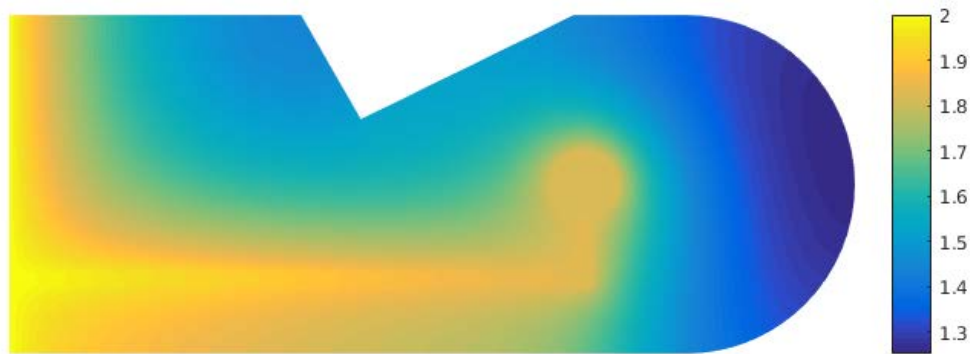


Fig. 1 Simulated electric potential distribution in a thin-film electrode consisting of two subdomains with different sheet resistances. An electric potential of 2 Volts is prescribed on the left boundary segment, and no electric current is flowing through the remaining boundary segments.

By post-processing of the numerical solution quantities such as the (lateral) electric current density distribution or the total device current may be computed. Parameter studies allow us to investigate the influence of geometry or material parameters on the device performance. These simulations are rather fast due to the coupled 1D-2D modeling approach – therefore, more elaborate tasks such as parameter estimation or optimization also become feasible.

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Evolutionary Optimization of TCO/Mesh Electrical Contacts

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The optimization of contacts based on a combination of TCO with a metallic mesh is complex due to the different conductivities of the materials and due to the counteracting effects of adding a metallic mesh: shadowing and enhancement of conductivity. An approach based on evolutionary optimization of a metallic mesh combined with 2D+1D FEM analysis of the ohmic losses is proposed. The FEM modelling subdivides the TCO and the mesh into two 2D layers as shown in Fig. 1 and allows for an accurate calculation of the voltage distribution and ohmic losses on extended electrodes. An evolutionary optimization algorithm¹ reduces the overall losses by iteratively adding metal on top of the TCO areas with highest losses. In the end, this approach allows to automatically optimize the shape of metallic contacting meshes. As an example, the predicted performance of two automatically designed contacts is compared with experimental results of CIGS solar cells².

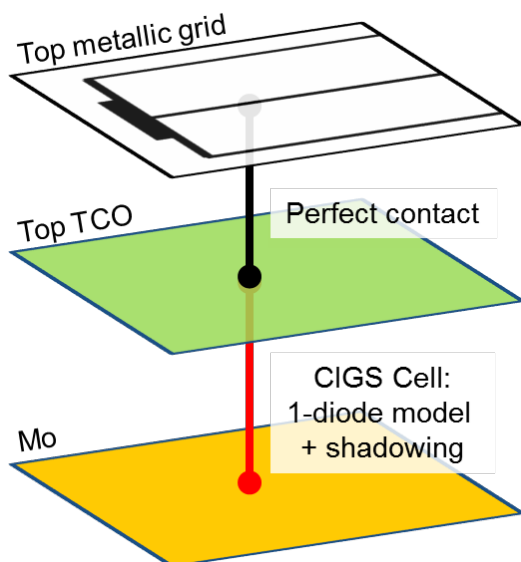


Fig. 1 Schematic representation of the model geometry, dimensionality and couplings

¹ G.P. Steven, Q. Li, Y.M. Xie, Evolutionary topology and shape design for general physical field problems, *Comput. Mech.* 26 (2000) 129–139. DOI:10.1007/s004660000160.

² P.A. Losio, T. Feurer, S. Buecheler, B. Ruhstaller, 32nd EUPVSEC, München (2016), DOI:10.4229/EUPVSEC20162016-3CV.4.3

Perovskite/crystalline silicon tandem solar cells

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Perovskite solar cells have recently emerged as attractive candidates to boost the performance of wafer-based silicon solar cells in perovskite/silicon tandem devices. This is especially interesting as single-junction silicon solar cells approach their practical efficiency limit. We present perovskite solar cells with high near-infrared transparency suitable for tandem integration, reaching efficiencies of up to 16.4%, and perovskite/silicon tandem cells in the mechanically stacked 4-terminal configuration with efficiencies of up to 25%. These efficiencies were enabled by the development of broadband transparent electrodes and uniform, pinhole-free perovskite absorber layers. In the monolithic configuration, we obtain tandem efficiencies of up to 21.2%. This tandem configuration is experimentally more challenging to realize, as the perovskite top cell is directly processed onto the silicon bottom cell. We identify photocurrent losses due to reflection and parasitic absorption in the perovskite top cell and show how performance can be optimized by adjusting layer thicknesses to tune interference. In addition, the application of a random-pyramid texture at the rear side of the silicon bottom cell was found to result in a strong quantum efficiency enhancement in the near-infrared by improved light trapping. Experimental results are compared to numerical simulations based on the Fluxim SETFOS 4.4 software package with absorption and light-scattering modules, showing excellent agreement. Results from these simulations are also employed to show pathways for further performance enhancement and to discuss the interplay between the rear-side texture and a micro-textured antireflective foil applied at the front side of the tandem cells.

Modeling tandem perovskite/c-silicon solar cells

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The perovskite solar cell is considered a promising candidate as the top cell for high-efficiency tandem devices with crystalline silicon (c-Si) bottom cells, contributing to the cost reduction of photovoltaic energy. In this contribution, a simulation method, involving optical and electrical modelling, is established to calculate the performance of 4-terminal (4T) perovskite/c-Si tandem devices on a mini-module level. Optical and electrical characterization of perovskite and c-Si solar cells are carried out to verify the simulation parameters. With our method, the influence of transparent conductive oxide (TCO) layer thickness of perovskite top cells on the performance of tandem mini-modules is investigated in case of both tin-doped indium oxide (ITO) and hydrogen-doped indium oxide (IO:H). The investigation shows that optimization of TCO layer thickness and replacement of conventional ITO with highly transparent IO:H can lead to an absolute efficiency increase of about 1%. Finally, a practical assessment of the efficiency potential for the 4T perovskite/c-Si tandem mini-module is carried out, indicating that with a relatively simple 4T tandem module structure the efficiency of a single-junction c-Si mini-module (19.3%) can be improved by absolute 4.5%.

The method to analyze the performance of 4T hybrid tandem mini-modules is mainly based on advanced optical simulation combined with solving diode equations. Optical simulation is carried out with the GenPro4 program developed at Delft University of Technology, aiming to calculate the short-circuit current density (J_{sc}) of solar cells. As shown in Fig. 1 the accuracy of the optical models and input parameters is validated by optical measurements. In tandem devices, c-Si bottom cells operate at lower injection levels than the standard irradiance condition, which has direct impacts on the current density (I_{sc2}) and consequently on the open-circuit voltage (V_{oc2}) and fill factors (FF_2) as well. By introducing the injection-level-dependent diode parameters into the one-diode equation, the V_{oc2} and FF_2 of the c-Si mini-module at different injection levels can be accurately calculated as shown in Fig.2. Fig. 3 shows the layout of the designed 4T perovskite/c-Si tandem minimodule being analyzed and the power conversion efficiency (PCE) improvement from 4T tandem compared to standalone c-Si minimodules.

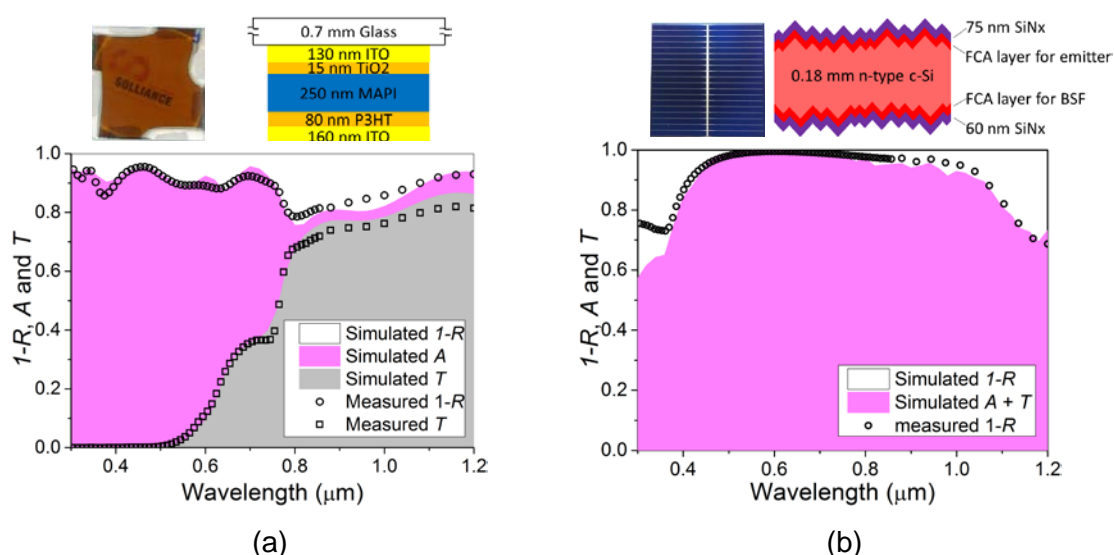


Fig. 1 Device picture, the optical structure and comparison between the simulated and measured reflectance, absorbance and transmittance spectra for (a) semi-transparent perovskite cells and (b) MWT c-Si cells

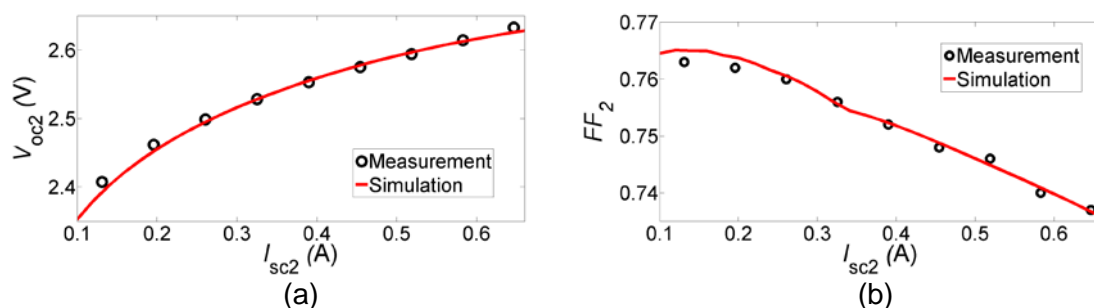


Fig. 2 Comparison of (a) V_{oc2} and (b) FF_2 calculated with modified one-diode equations for the c-Si mini-module to the results measured at different injection levels.

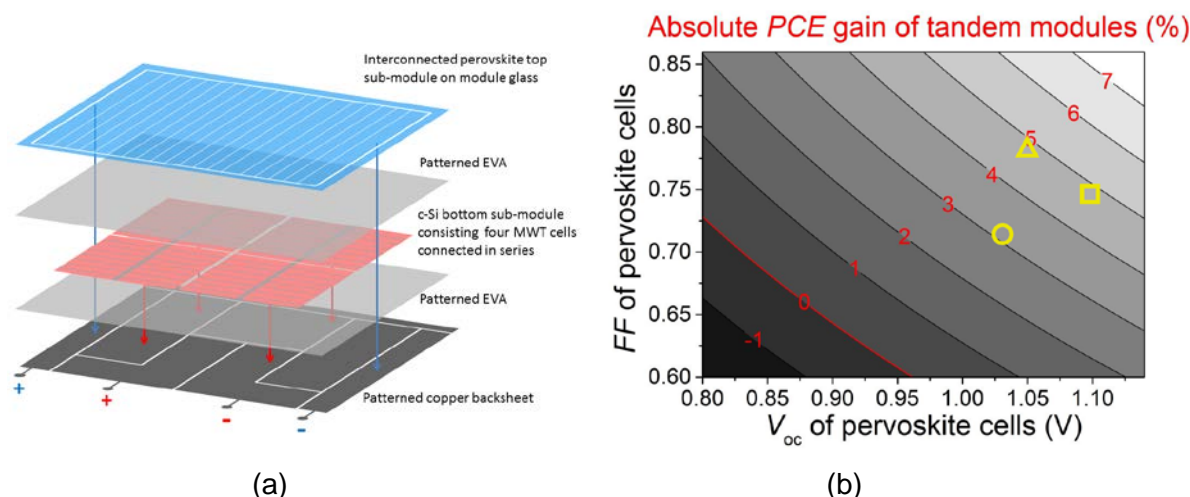


Fig. 3 (a) the sketch of designed 4T perovskite/c-Si tandem mini-modules and (b) absolute *PCE* gain of the designed tandem mini-module as a function of V_{oc} and FF of the perovskite top cell. The red line indicates the required V_{oc} and FF of perovskite cells to break even. The yellow circle indicates the V_{oc} and FF achieved in-house (1.03 V and 71.4%), and the yellow square and triangle correspond to the literature values: 1.104 V & 73.6%¹ and 1.034 V & 77.7%².

¹ F. Fu, T. Feurer, T. Jäger, E. Avancini, B. Bissig, S. Yoon, S. Buecheler & A. N. Tiwari, "Low-temperature-processed efficient semi-transparent planar perovskite solar cells for bifacial and tandem applications", Nat. Commun. 6, 1, (2015)

² J. Werner, C. Weng, A. Walter, L. Fesquet, J. Peter Seif, S. De Wolf, B. Niesen and C. Ballif, "Efficient Monolithic Perovskite/Silicon Tandem Solar Cell With Cell Area > 1 cm²", J. Phys. Chem. Lett. 7, 161, (2016)

Optical simulations of birefringent organic semiconductor devices

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Optical simulations of organic semiconductor devices are an established technique to optimize stack design and investigate intrinsic properties of such systems. The isotropic description of organic layers via the Jones Matrix-Transfer-Formalism has proven to be able to describe the optical response of most of the amorphous organic materials. Proper description of crystalline of highly ordered layers, however, is more challenging due to the inclusion of uni- or biaxial optical constants and the corresponding impact on the propagation of electromagnetic waves. We performed optical simulations of birefringent organic thin films, using formulas derived by Penninck et al.¹. The algorithm can be used for proper description of the anisotropic optical properties of materials like Diindenoperylene (DIP) or Dibenzotetraphenylperiflantene (DBP). Angular dependent measurements of the photocurrent in organic solar cells reveal differences between isotropic and birefringent descriptions of the optical constants and provide confirmation for the implemented simulation². Furthermore the investigated calculations offer the possibility to determine the alignment of the transition dipole moments via photoluminescence measurements and, thus, allow for simple measurements of the molecular orientation in such layers³. Although the inclusion of uniaxial anisotropy in optical modeling is not necessary for vertical light incidence in organic solar cells, the simulations offer interesting new methods for the investigation of the optical properties of organic thin films.

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Optical Simulations of Tunable Scattering Layers for Photon Management in Organic Light Emitting Diodes and Thin Film Solar Cells

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Scattering layers based on disordered photonic structures can be used for light extraction from organic light emitting diodes (OLEDs) or for light trapping in thin film solar cells. A versatile technique to fabricate such layers with a good control over the statistical properties is the polymer-blend lithography¹. This technique relies on the controlled phase separation of a mixture of two polymers. Selectively developing one of the materials, the resulting pattern can be transferred into a dielectric or a metallic layer, forming a compact scattering element within the thin film stack.

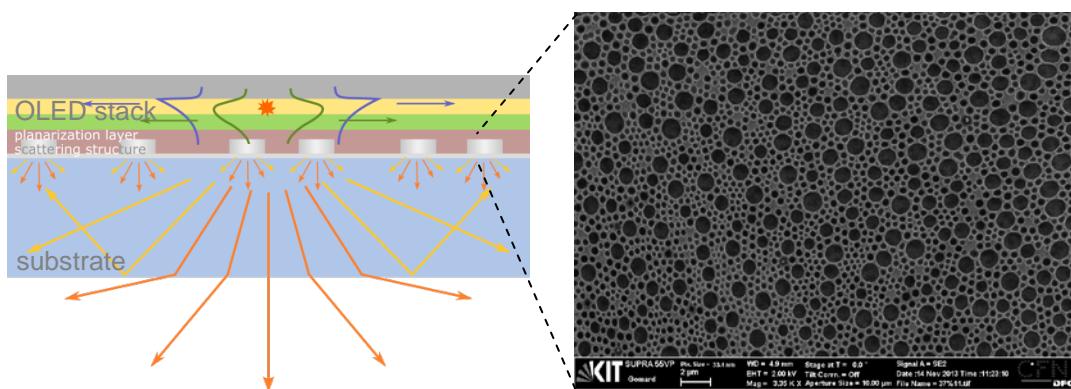


Fig. 1 Left: Scattering layer in an OLED for improved light outcoupling. Right: Structure formation by phase separation of a polymer blend.

We report on numerical studies that aim at a full optical simulation of optoelectronic thin film devices incorporating such a scattering layer. The simulations are based on two numerical methods: the finite element method (FEM) and the T-matrix formalism² for the description of the individual scattering centers. In the case of the T-matrix method, special care has to be taken regarding an overlap of the individual scattering centers' circumscribing sphere with the

¹ Lee, C. and Kim, J.-J., "Enhanced Light Out-Coupling of OLEDs with Low Haze by Inserting Randomly Dispersed Nanopillar Arrays Formed by Lateral Phase Separation of Polymer Blends". *Small*, 9: 3858–3863. (2013)

² A. Egel and U. Lemmer, "Dipole emission in stratified media with multiple spherical scatterers: Enhanced outcoupling from OLEDs," *J. Quantum Spectrosc. Radiat. Transfer* 148, 165–176 (2014).

layer interfaces³. In this context, we assess the suitability of spherical vector wave functions for scattering at flat discs touching a layer interface.

First simulation results based on the T-matrix method as well as on the finite element method are compared to optical measurements at the scattering structures.

³ A. Doicu, Y. Eremin, and T. Wriedt, “Convergence of the T-matrix method for light scattering from a particle on or near a surface,” *Opt. Commun.*, no. January, pp. 266–277, 1999.

Modelling of Light Scattering in Single Junction and Tandem Cells

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Achievement of the record efficiencies in photovoltaic devices require constant advancements in the structure of wafer-based silicon cells and tandem perovskite/silicon cells. In order to facilitate the process of finding an optimal cell configuration we introduce a simulation tool, which combines a 3D ray-tracing algorithm with thin films optics to model the light interaction with conformally coated textured silicon wafers and deposited anti-reflection foils. The two-scale framework¹ takes into account the interference in coherent layers coupled with the light propagation and scattering in incoherent textured wafers. In order to validate the simulation tool an example of the absorption spectra of the silicon heterojunction (SHJ) cell compared with the measurement² is presented (Fig 1). Furthermore the experimental data of the monolithic perovskite/silicon tandem solar cells³ with different configurations of the scattering interfaces is compared with the simulation results (Fig 2), modeling of conformally coated perovskite top cell on the textured silicon wafer is also analyzed.

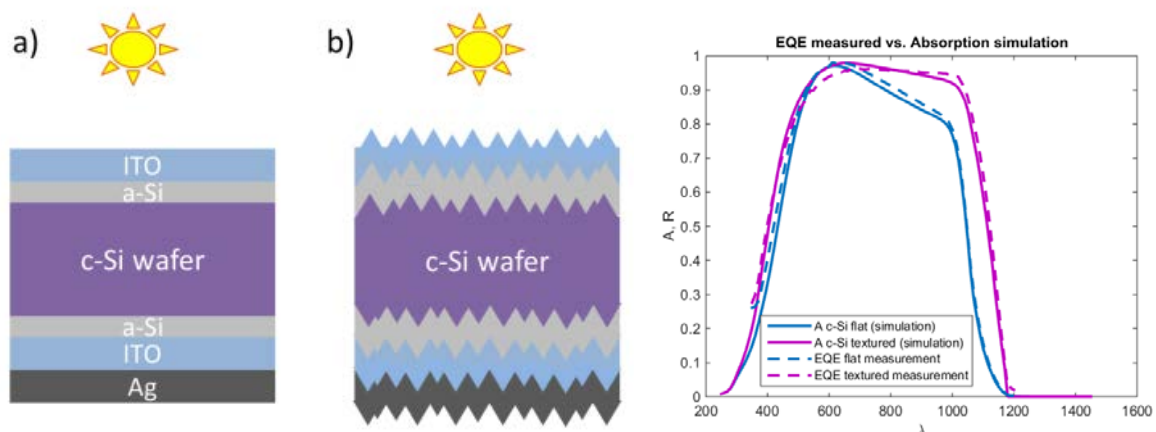


Fig. 1: SHJ planar and double-side textured case (left). Comparison of simulated absorption (full lines) and measured EQE spectrum (dashed lines) of the SHJ (right).

¹ SETFOS 4.4 software with Absorption and Light-scattering modules, Fluxim AG, Switzerland

² S. de Wolf, A. Descoedres, Z.C. Holman, C. Ballif, *Green* 2, pp. 7-24 (2012)

³ J. Werner, C.-H. Weng, A. Walter, L. Fesquet, J.P. Seif, S. de Wolf, B. Niesen, C. Ballif, *J. Phys. Chem. Lett.* 7, 161 (2016)

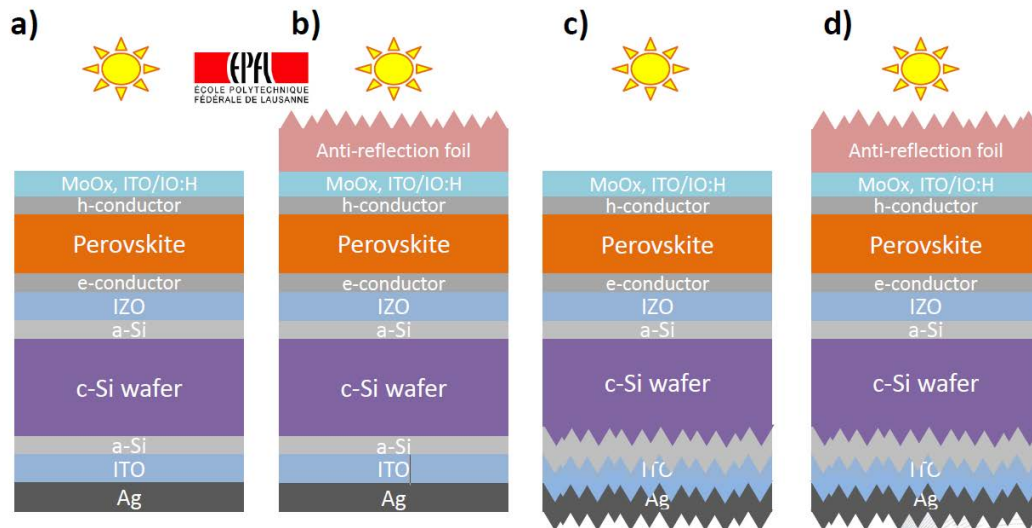


Fig. 2: Different monolithic perovskite/silicon tandem structures considered⁴: a) planar reference b) planar reference with external anti-reflection foil (ARF), c) planar perovskite solar cell on rear-textured silicon wafer, d) planar perovskite solar cell on rear-textured silicon wafer with ARF.

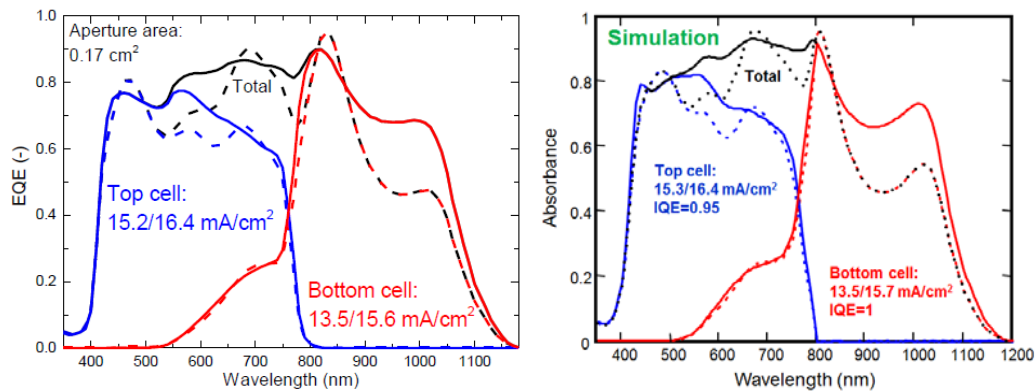


Fig. 3: Example of comparison of experiment⁴ and simulation by Setfos 4.4 of tandem absorption spectra for the planar perovskite/silicon tandem with an anti-reflection foil (ARF) (structure b)) and without ARF (structure a) of Fig. 2).

⁴ J. Werner, C.-H. Weng, A. Walter, L. Fesquet, J.P. Seif, S. de Wolf, B. Niesen, C. Ballif, J. Phys. Chem. Lett. 7, 161 (2016)

Inverted Current–Voltage Hysteresis in Mixed Perovskite Solar Cells: Polarization, Energy Barriers, and Defect Recombination

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Organic-inorganic metal halide perovskite solar cells show hysteresis in their current–voltage curve measured at a certain voltage sweep rate. Coinciding with a slow transient current response, the hysteresis is attributed to a slow voltage-driven (ionic) charge redistribution in the perovskite solar cell. Thus, the electric field profile and in turn the electron/hole collection efficiency become dependent on the biasing history. Commonly, a positive prebias is beneficial for a high power-conversion efficiency. Fill factor and open-circuit voltage increase because the prebias removes the driving force for charge to pile-up at the electrodes, which screen the electric field. Here, it is shown that the piled-up charge can also be beneficial. It increases the probability for electron extraction in case of extraction barriers due to an enhanced electric field allowing for tunneling or dipole formation at the perovskite/electrode interface. In that case, an inverted hysteresis is observed, resulting in higher performance metrics for a voltage sweep starting at low prebias. This inverted hysteresis is particularly pronounced in mixed-cation mixed-halide systems which comprise a new generation of perovskite solar cells that makes it possible to reach power-conversion efficiencies beyond 20%.

Understanding hysteresis in perovskite cells through simulations of coupled electron-ion motion

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In perovskite solar cells, a major concern is the occurrence of hysteresis in which the cell current–voltage characteristics are strongly dependent on the voltage scan rate, direction and preconditioning treatments. Here, I describe drift diffusion simulations of perovskite cell steady state and transient characteristics. Figure 1 is taken from ¹ in which we have identified the physical origin of the features seen in measured current-voltage, J-V, curves as mobile defects. Our model uses the method of matched asymptotic expansions to solve the charge transport equations, an approach that is widely applicable to other double layer problems occurring in electrochemical applications such as the evolution of transmembrane potentials in living cells. In this way we can account accurately for the Debye layers in which the ionic charge accumulates, that being a few nm in width, are much narrower than typical perovskite layer thicknesses of several hundred nm. Our model paves the way for the development of cells with improved and reproducible performance.

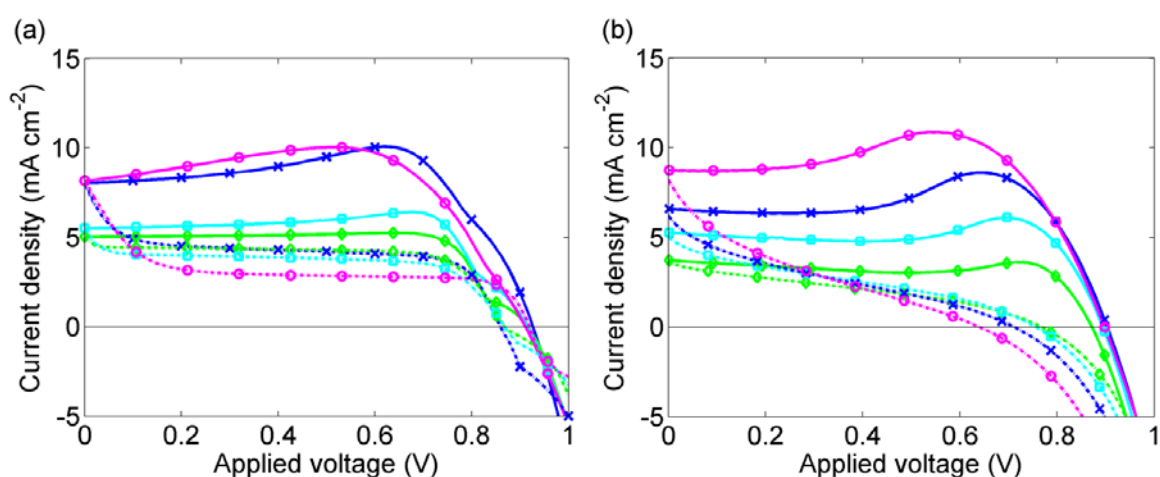


Fig. 1 (a) Calculated J–V curves for a perovskite cell without allowing for preconditioning; (b) measured J–V curves for this cell after an initial preconditioning at 1.2 V and two scan cycles. Solid lines show the 1.2 V to 0 V scan; broken lines show the 0 V to 1.2 V scan. Scan rates are 1 V s⁻¹ (magenta, circles), 500 mV s⁻¹ (blue, crosses), 250 mV s⁻¹ (cyan, filled squares), 100 mV s⁻¹ (green, diamonds).

¹ G Richardson, S E J O’Kane, R G Niemann, T A Peltola, J M Foster, P J Cameron, A B Walker “Can slow-moving ions explain hysteresis in the current-voltage curves of perovskite solar cells?” *Energy & Env Sci* **9** 1476 (2016),

Explanation for reduced IV-curve hysteresis in highly efficient perovskite solar cells

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There is increasing evidence for ion migration in methylammonium lead iodide perovskite solar cells. The electric field induced by the mobile ions affects the charge transport and is believed to be the origin of the hysteresis in IV-curves [1, 2]. The occurrence of hysteresis was also related to the contact layer materials [3]. Highly efficient devices generally show low hysteresis. Hereby the following question arises: If mobile ions in the bulk are responsible for the IV-curve hysteresis, why does the

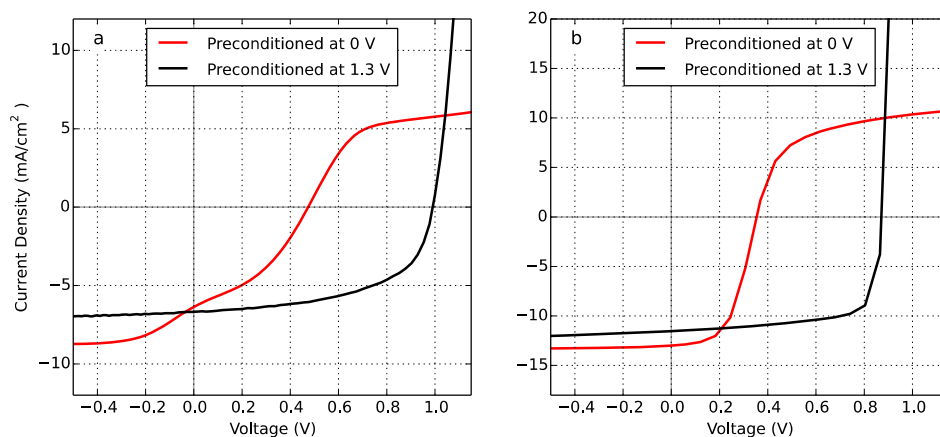


Figure 1 a) Measured Current-Voltage Characteristics with a fast ramp (30 ms) after 10 seconds preconditioning at 0 V and 1.3 V. b) Simulated IV-curves with different ion preconditioning.

We measure preconditioned IV-curves as proposed by Tress et al. [1] and use a numerical drift-diffusion model incorporating mobile ions to reproduce the measured effects (shown in Figure 1). Using the numerical model we demonstrate why the hysteresis vanishes almost completely if contacts with a low surface recombination are used: With good contacts electrons and holes can also be extracted without an electric field as they can “pile up” at the opposite interface and diffuse to the interface where they are extracted. This finding is consistent with the study of Philip Calado and Piers Barnes that find evidence for ion migration for devices with low hysteresis [4].

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Simulating transient optoelectronic measurements on perovskite solar cells: Evidence for ion migration in devices with minimal hysteresis

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Ionic migration has been proposed as a possible cause of photovoltaic current-voltage hysteresis in hybrid perovskite solar cells.¹²³⁴ A major objection to this hypothesis is that hysteresis can be reduced by changing the interfacial contact materials, which are unlikely to significantly influence the behaviour of mobile ionic charge within the perovskite phase.⁵⁶ Here we use transient optoelectronic measurements combined with drift diffusion simulations to show that the primary effects of ionic migration can in fact be observed in devices with 'hysteresis free' type contact materials, as well as those exhibiting hysteresis. The data indicate that electric-field screening, consistent with ionic migration, is similar in both high and low hysteresis CH₃NH₃PbI₃ cells. Transient photovoltage and photocurrent device simulations show that hysteresis requires the combination of both mobile ionic charge and recombination near the contacts.⁷ Low hysteresis is thus primarily due to high photogenerated carrier concentrations in the perovskite phase, capable of screening ionic charge, rather than necessarily an absence of mobile ions or higher ionic mobilities. In contrast, devices with high interfacial recombination, where electronic carrier concentrations are low, exhibit significant hysteresis.

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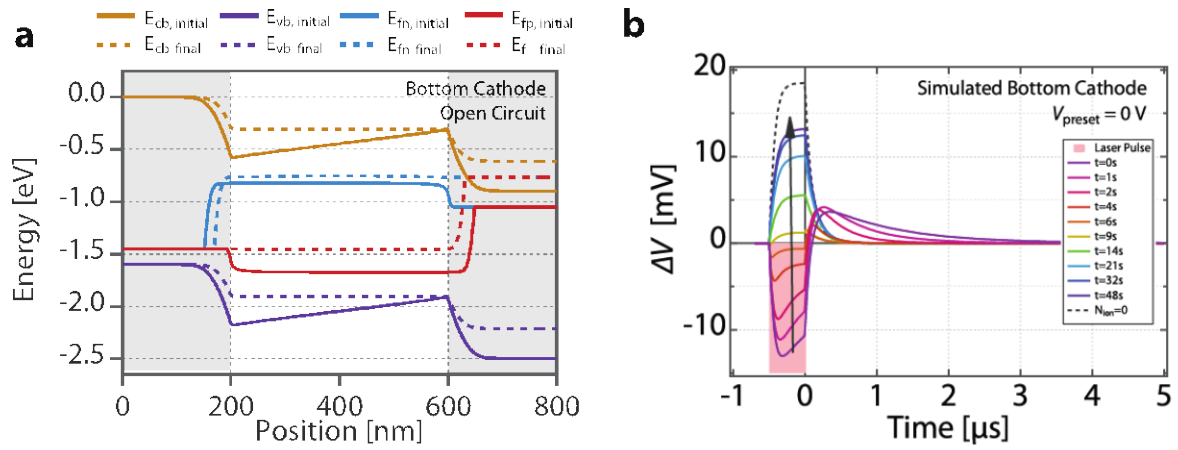


Fig 1 **a** Simulated energy level diagram for a bottom cathode perovskite solar cell at open circuit under illumination after equilibration at short circuit in the dark. Solid lines indicate initial states, dashed lines indicate the cell at steady state. Ionic migration to the p and n-type contact interfaces results in a reverse electric field in the device capable of driving charge carriers towards the wrong electrode. **b** Transient photovoltage evolution during the same simulation. The reverse field results in an initial negative deflection of the transient photovoltage signal, similarly observed in experimental measurements.

Laser pulsed transient photo currents on Perovskites to study charge carrier transport

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Transient photocurrent measurements are performed on perovskites to study bulk charge carrier dynamics. Charge excitation is executed by a nanosecond pulsed laser in order to analyze electron and hole transport in the semiconductor. Also slow effects e.g. ionic movement are induced within this method under electrical bias conditions and dominate transport at time-scales longer than 10^{th} of microseconds.

The transport of photo excited charges is analyzed at short times for different path lengths and electrical fields in first experiments at room temperature^{1,2}. We probe layers of perovskites for planar architecture and vary layer thickness. The observed photocurrent has dispersive character. In the case of transport limited currents charge extraction is determined by the layer with the lowest charge mobility. For simple devices based on organohalide perovskite without any organic ETL and HTL the observed charge processes would show intrinsic perovskite properties. As in time-of-flight experiments on organic systems, slow portions of charge release indicate a non-regular transport, which can be understood by a broadening of transport site energies for electrons induced by the organic cation of perovskites. The observed transients can be explained by thermal and field assisted hopping of charges out of states below the effective transport energy³.

¹ S. Valouch, M. Nintz, S. W. Kettlitz, N. S. Christ, and U. Lemmer, "Thickness-Dependent Transient Photocurrent Response of Organic Photodiodes", IEEE Photon. Technol. Lett. 24, 596, (2012)

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³ N. S. Christ, S. W. Kettlitz, J. Mescher, S. Valouch, and U. Lemmer, "Dispersive transport in the temperature dependent transient photoresponse of organic photodiodes and solar cells", J. Appl. Phys. 113, 234503, (2013)