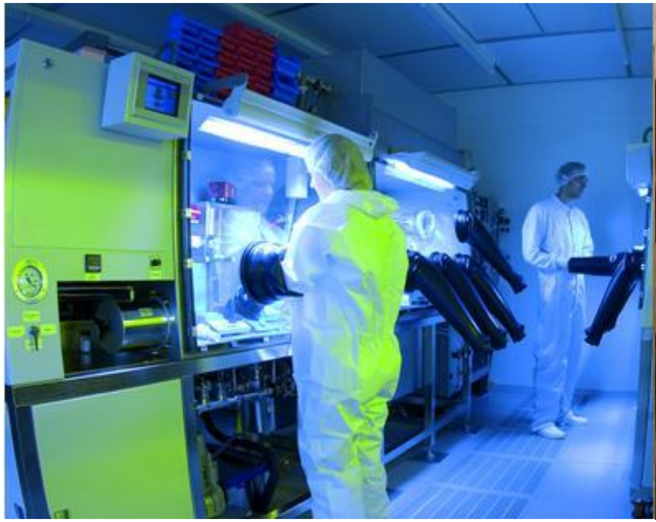
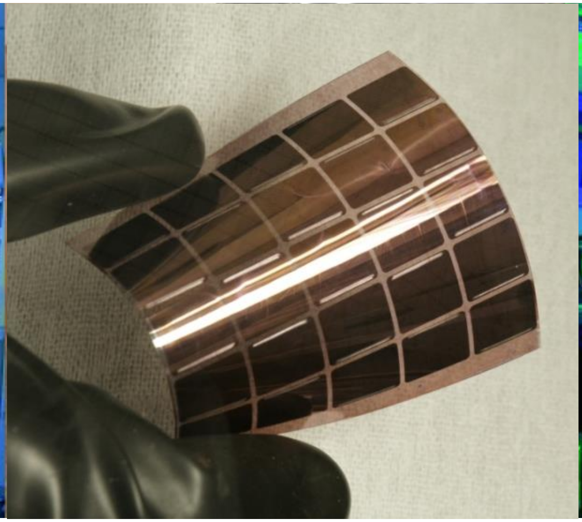
Karlsruhe Institute of Technology

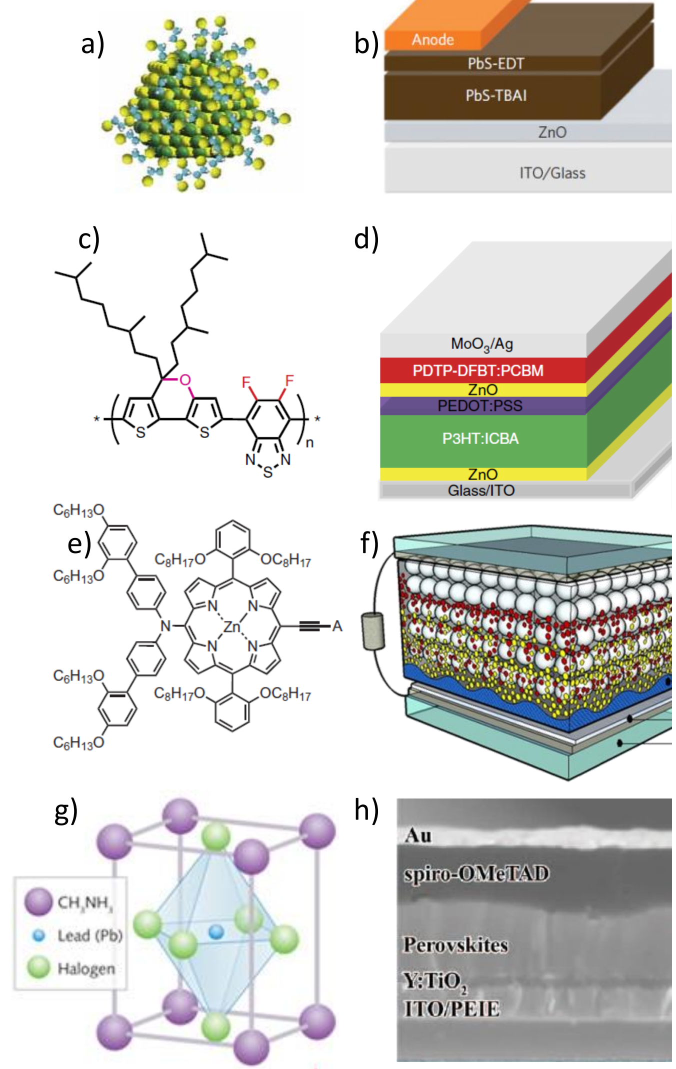
Excitonic Solar Cells Part 1


Excitonic Solar Cells

- Big picture goals of novel materials
  - low-cost, large-area processing
  - printing, spraying, roll-to-roll

- Candidate Systems
  - Quantum dots (PCE 8.6%)
  - Semiconducting polymers (PCE 11.1 %)
  - Dyes (PCE 13%)
  - Hybrids / Perovskites (PCE ~ 20%)
  - Metal Organic Frameworks (PCE < 1%)



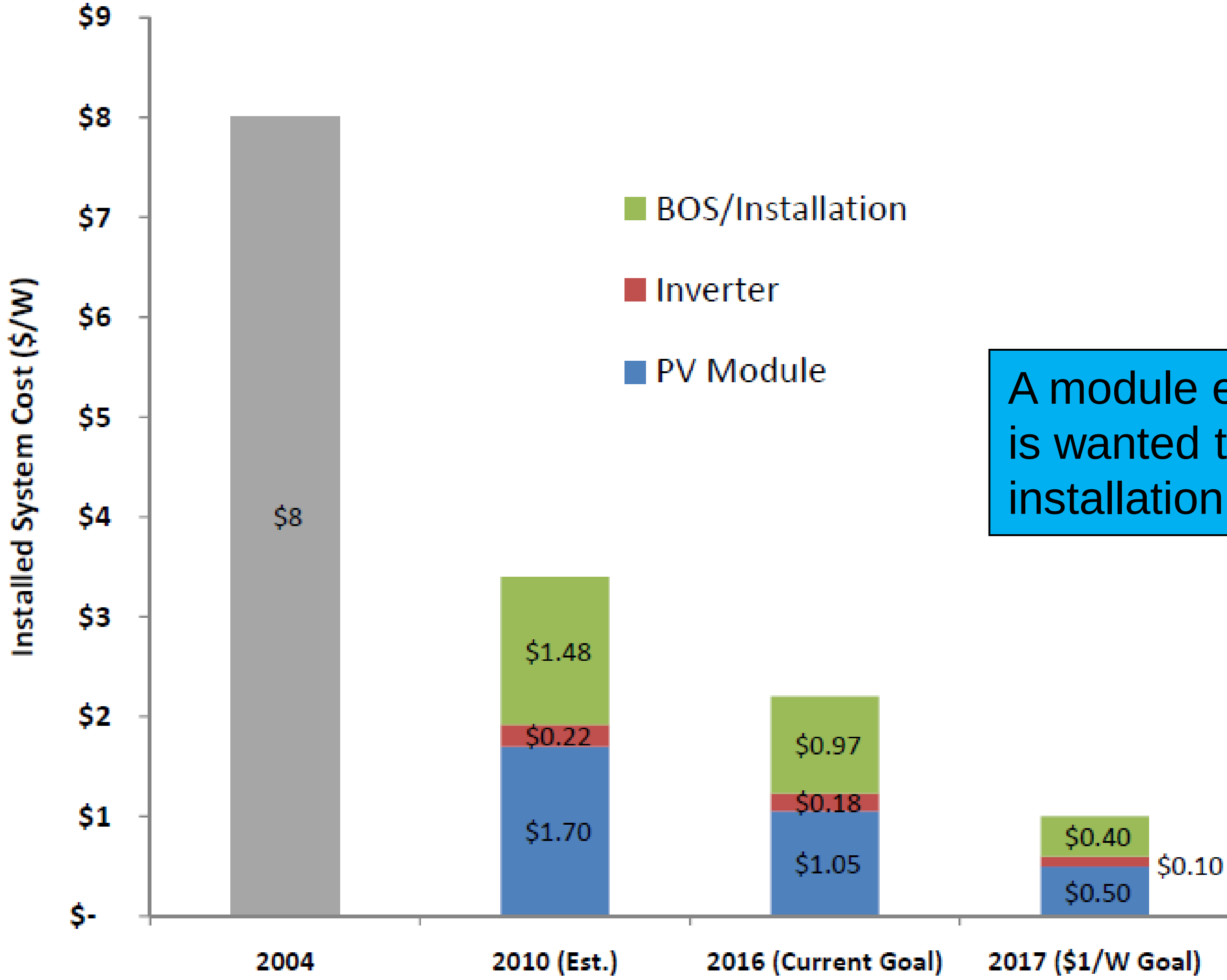
Figures adapted from: a) Tang, Nat Mater 2011, 10, 765 b) Chuang, Nat Mater 2014, 13, 3984. c) You, Nat Comm 2013, 4, 1466. d) also from ref. c) e) Mathew, Nat Chem 2014, 6, 242 f) https://schanze.chem.ufl.edu g) http://www.laserfocusworld.com h) Zhou, Science 2014, 345, 542

Outline

Part I: OPV and Dye Sensitized Cells

1. Introduction
2. Bulk Heterojunctions
3. Nanomorphology, Nanostructure, etc,
4. Low Bandgap Absorbers
5. Semitransparent , Tandem Solar cells, Electrodes
6. Nanophotonics for Organic Solar Cells
7. OPV Status and Industry

DOE's Sunshot Goal: $1/W by 2017



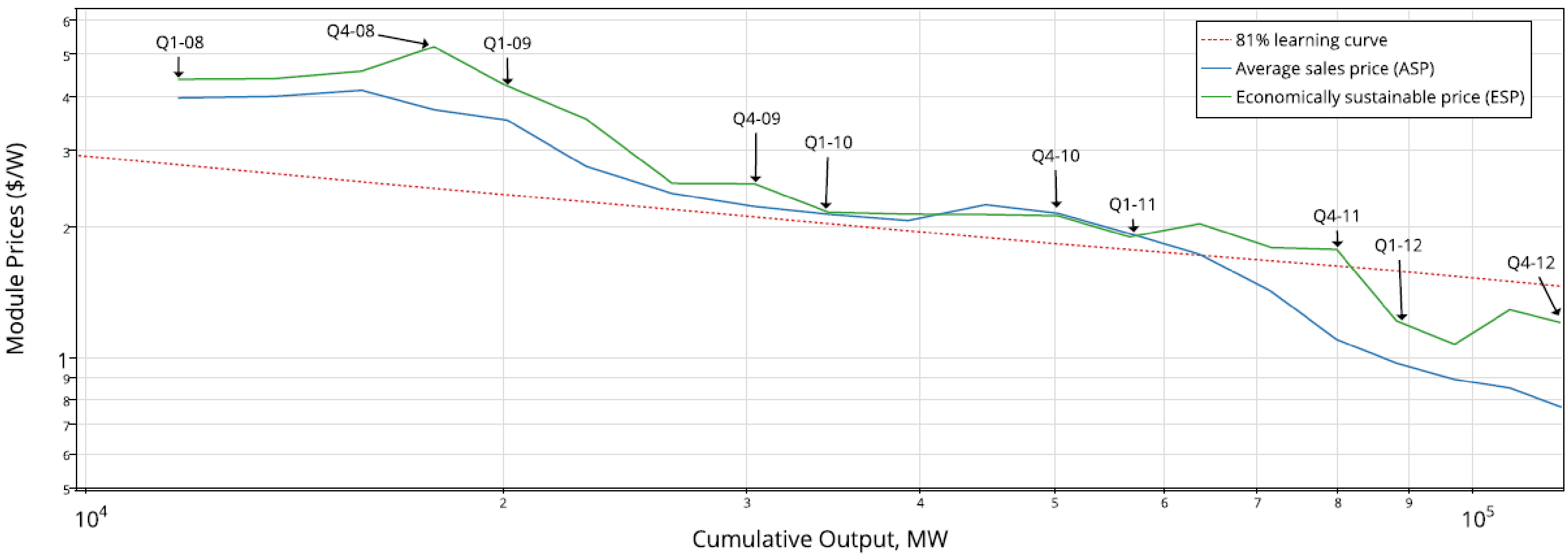
A module efficiency of 25 % is wanted to enable the installation cost reductions.

Source: US DOE report "$1/W Photovoltaic Systems," August 2010.

Last Week's Lecture by Anshu Sahoo and Stefan Reichelstein

Silicon is currently competitive in favorable locations with the current subsidies.

Average Sales Prices of Modules and Cumulative Module Output



In 2017 the cost of silicon cells will probably be $0.65/W.

Reminder: I-V-characteristics of solar cells

Maximum power in generator mode:

$$P_{\max} = U_{\max} I_{\max}$$

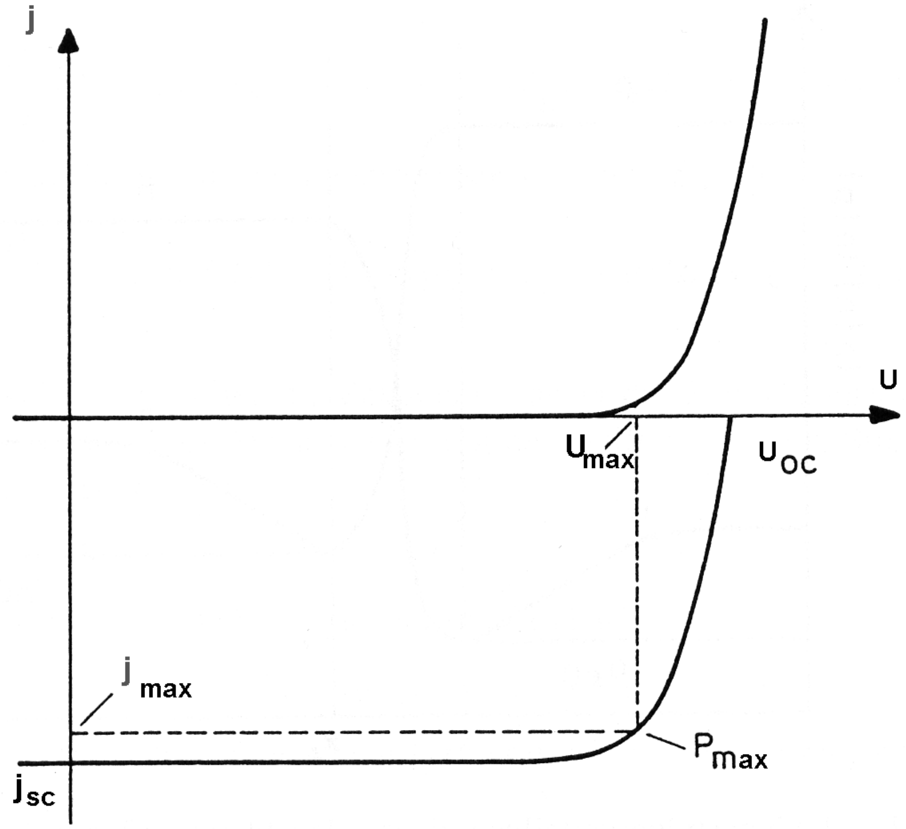
Efficiency of solar cell:

$$\eta = \frac{U_{\max} I_{\max}}{\phi}$$

$\phi =$ optical power on solar cell

Fill Factor:

$$FF = \frac{U_{\max} I_{\max}}{U_{oc} I_{sc}}$$



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Part I: Organic Photovoltaics

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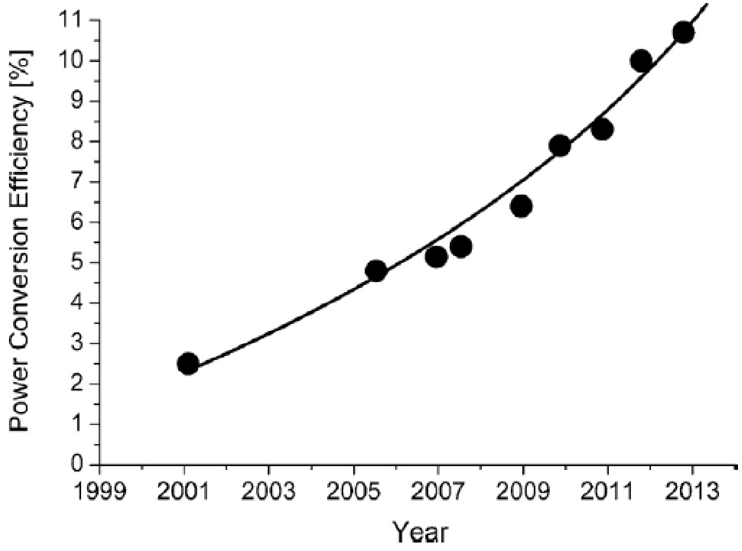
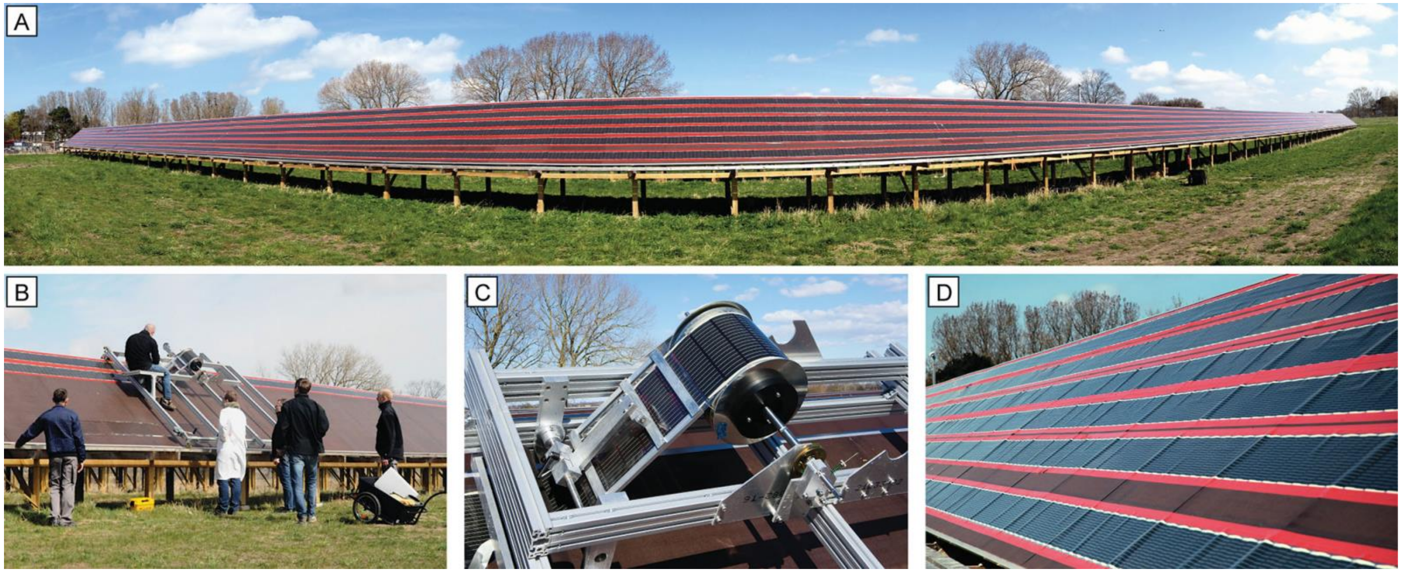
Status quo of organic solar cells



- Solution-processed photovoltaic devices
- Large-area processing by R2R printing
- Potential for low-cost production
- Mechanical flexibility
- Interesting photophysical properties ☺

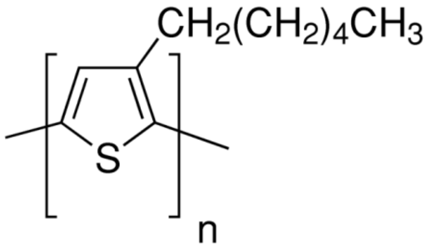
- Efficiency is an issue (commercialization)
- Lifetime / degradation problems



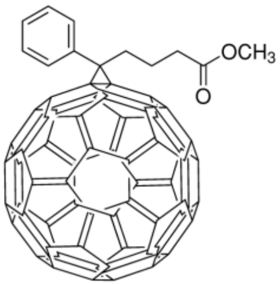
Roll mounting of organic solar cells in a prototype solar park in Denmark

Scharber, M. & Sariciftci, N., *Progress in Polymer Science* , **2013**, 10.1016/j.progpolymsci.2013.05.001
Krebs, F. et al, *Advanced Materials,* **2013**, 10.1002/adma.201302031

Interplay between structure, dynamics and function

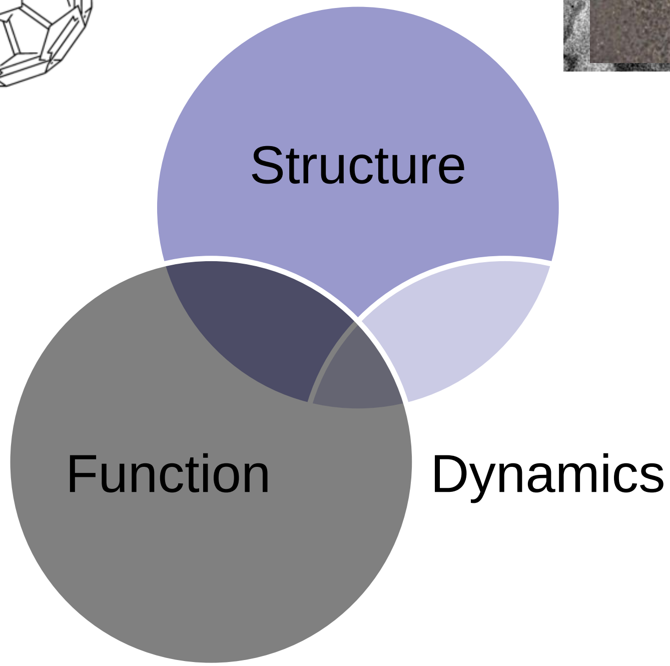


Chemical structure



Morphology

Processing



Context

- Organic materials have low dielectric constants

- Photon ⇒ Exciton (D*)

  e-h bound by Coulomb interaction

- Bulk heterojuntion concept

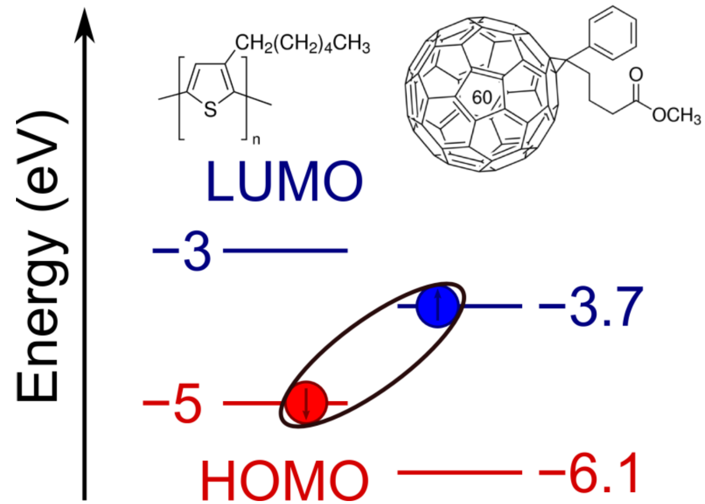
  $D^* \Rightarrow D^+ + A^-$

- But what about interfacial CT states?

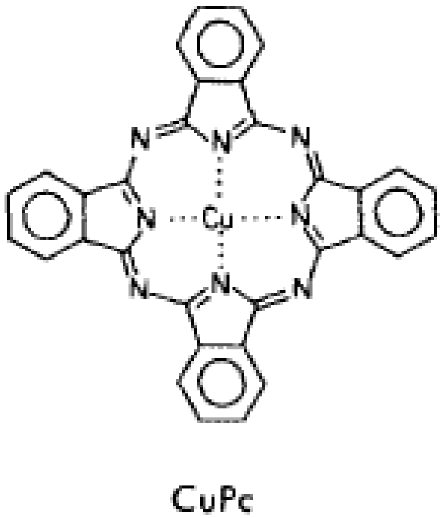
  $D^* \Rightarrow DA^* \Rightarrow D^+ + A^-$

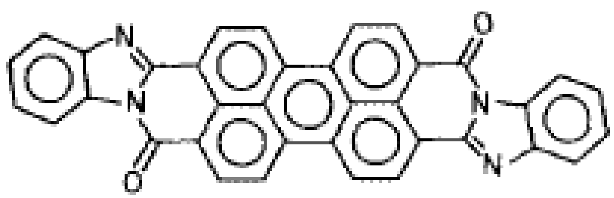
  $\Rightarrow D + A$

- CT states absorb, emit, and recombine.



The Tang-cell: First demonstration of efficient dissociation at an internal interface





C.W. Tang, Kodak, 1985
1 % at AM 2

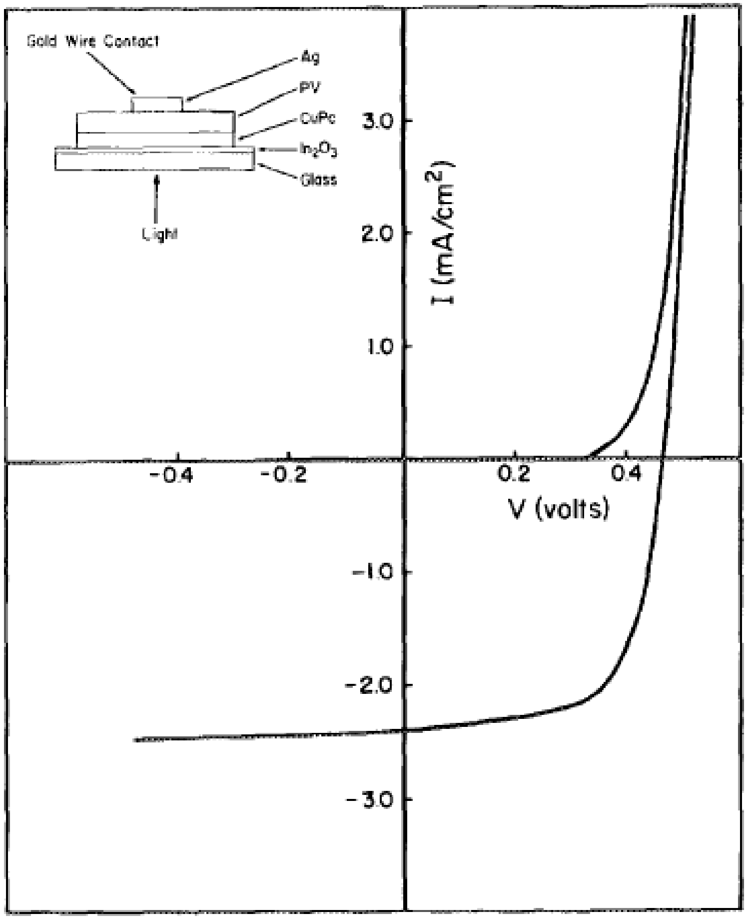
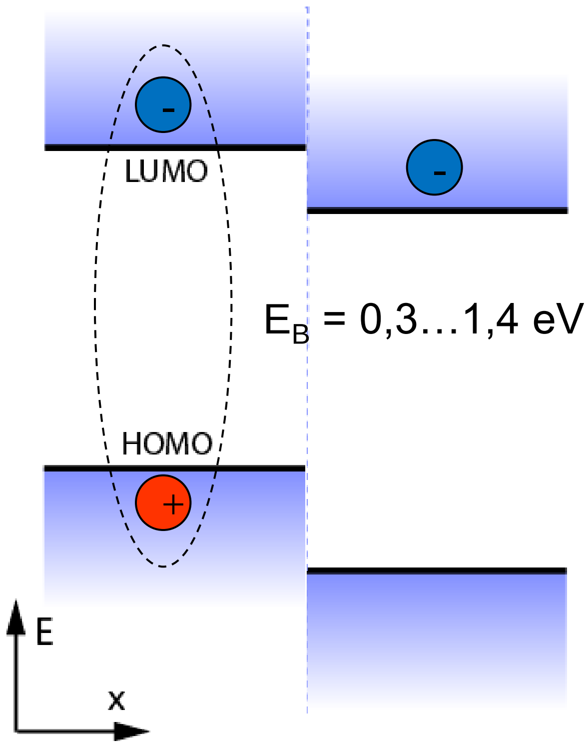


FIG. 1. Configuration and current-voltage characteristics of an ITO/CuPc (250 Å)/PV(450 Å)/Ag cell.

Tang, C. W. *"Two Layer Organic Photovoltaic Cell"* , Appl. Phys. Lett. 48, 183 (1986).

Organic Solar Cells



Bilayer Heterojunction

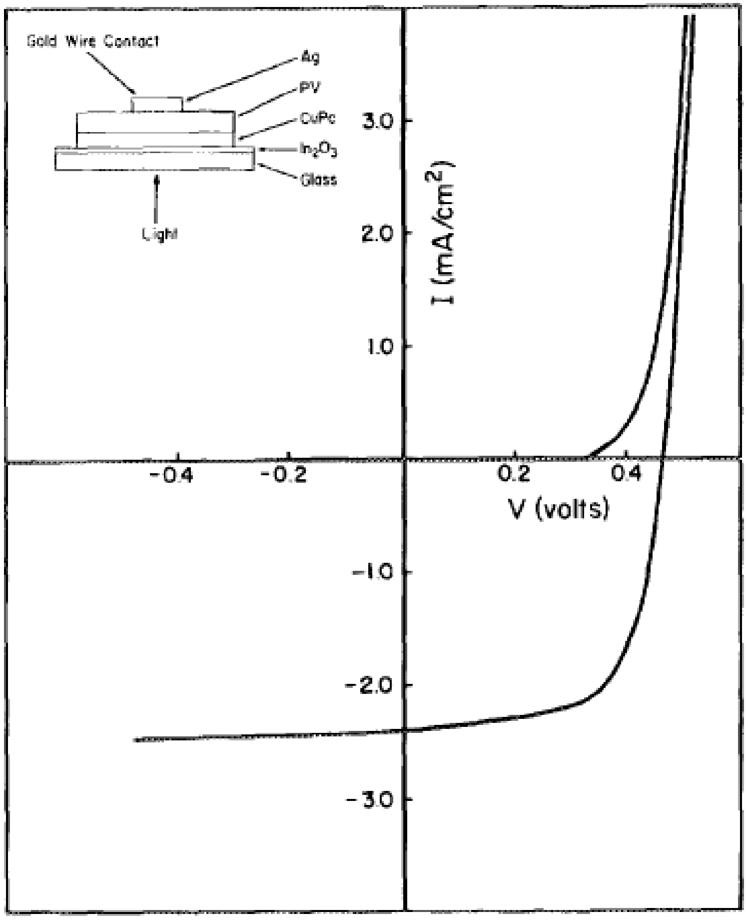
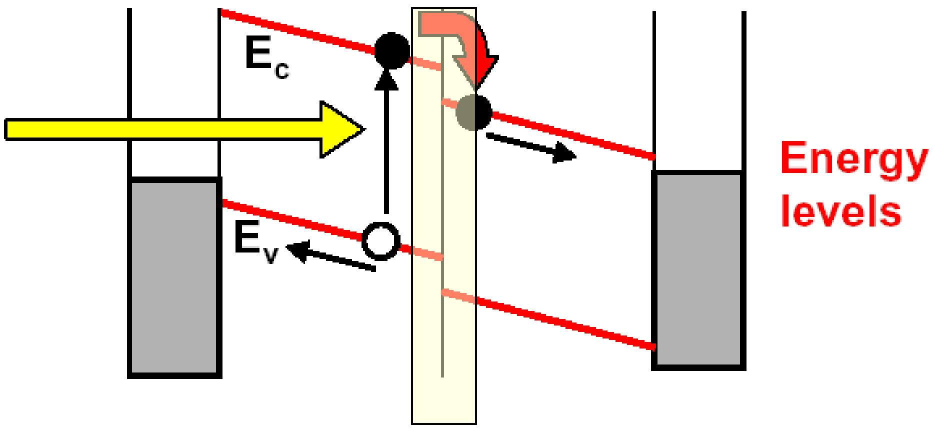


FIG. 1. Configuration and current-voltage characteristics of an ITO/CuPc (250 Å)/PV(450 Å)/Ag cell.

From a bilayer to a bulk heterojunction

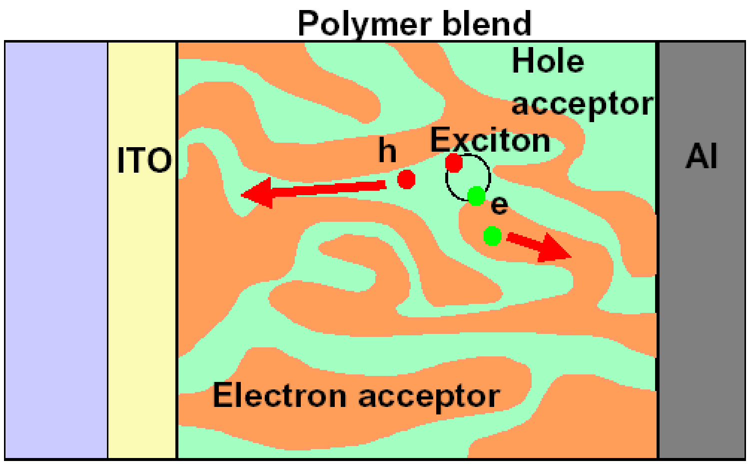
Bilayer:

-the active layer of the device is limited to the diffusion length (10-20 nm)
-only the interface is contributing to the quantum efficiency



„Bulk-Heterojunctions“
„Interpenetrating Networks“

The whole layer is contributing to the photocurrent generation.



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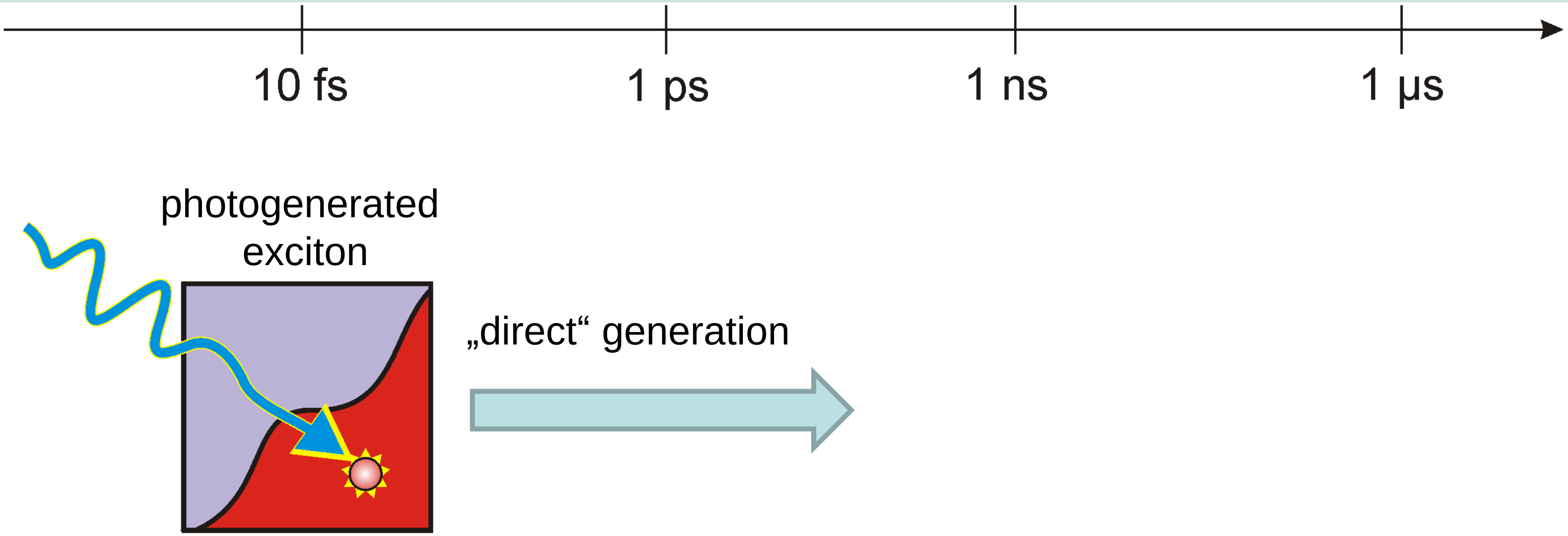
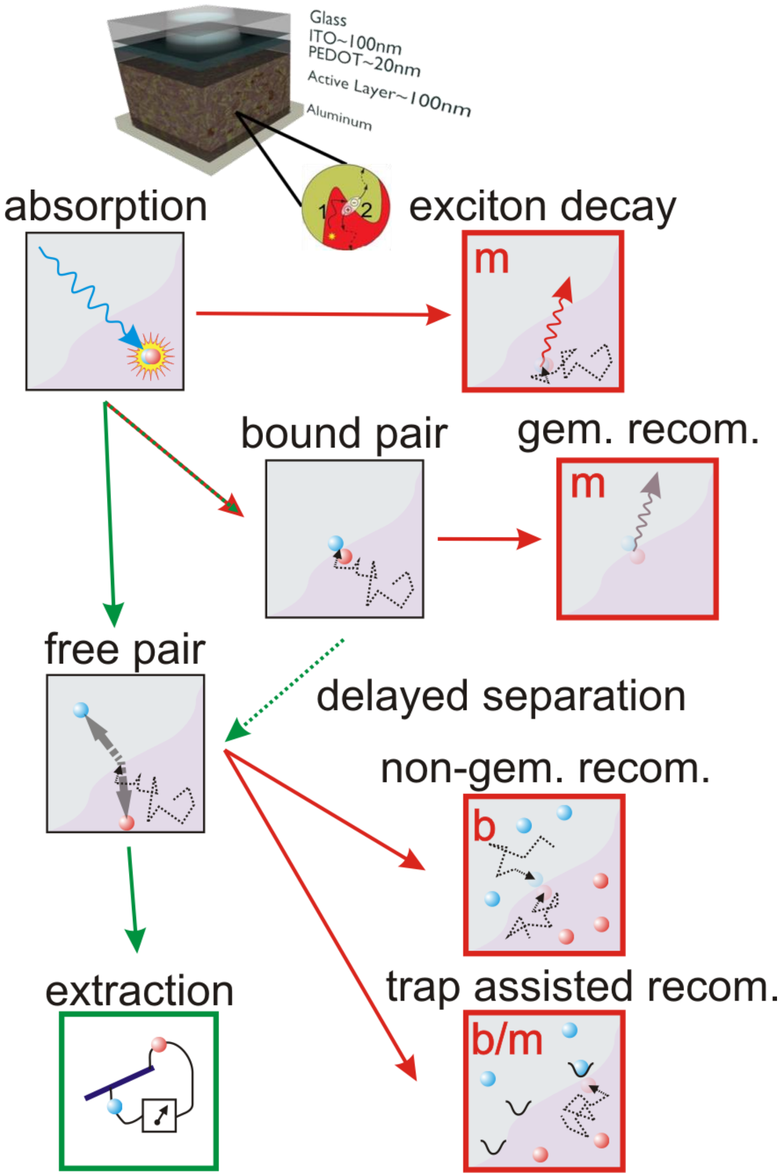
Photophysical processes and time scale



Efficiency-
limiting
processes

What recombination implies about generation



Geminate recombination: $$\frac{dn}{dt} \propto k\, n$$

→ Monomolecular
→ <u>concentration-independent</u> process.

Non-geminate recombination: $$\frac{dn}{dt} \propto \mu\, n^{2+x}$$

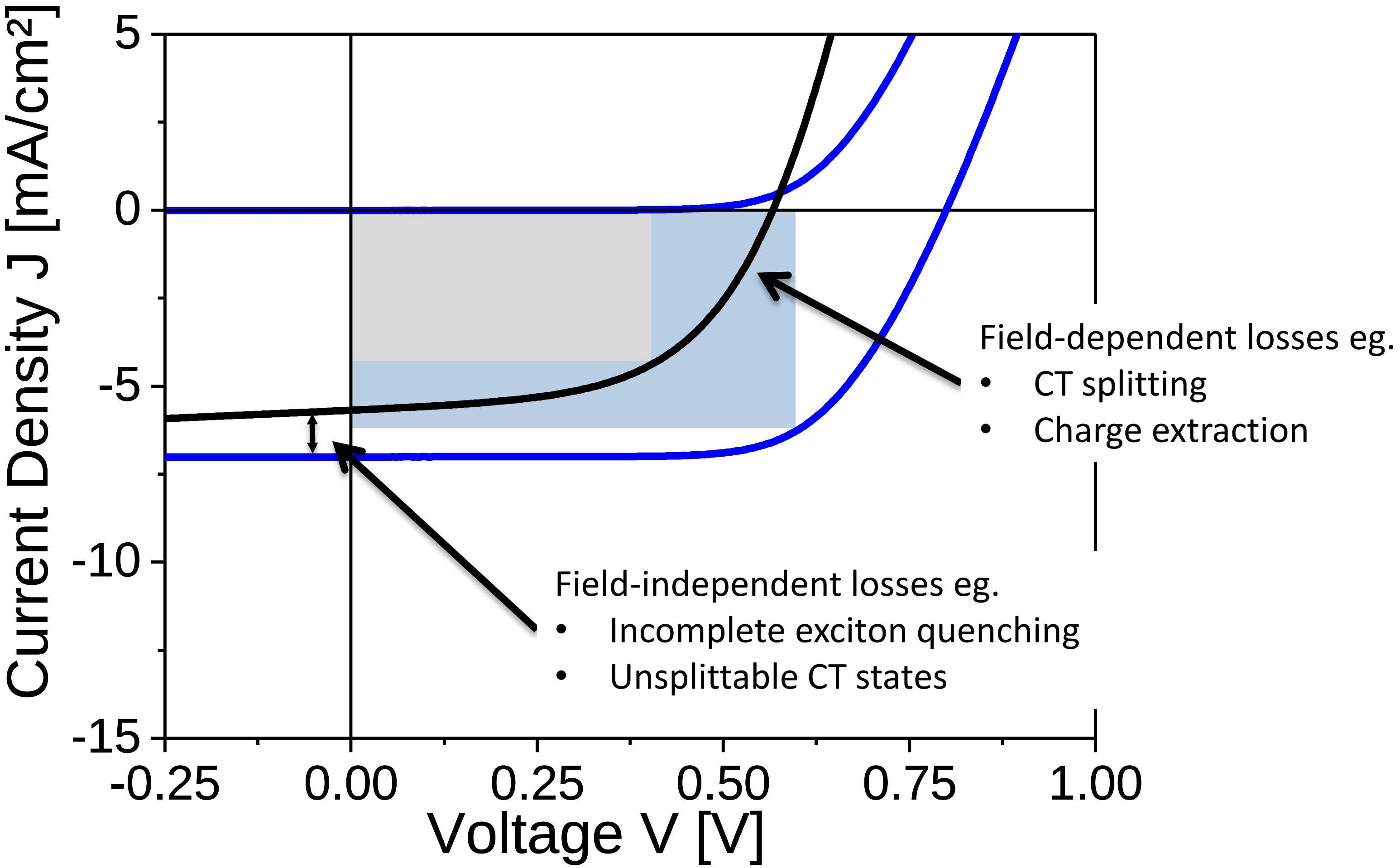
→ Higher order power law
→ <u>concentration-dependent</u> process.

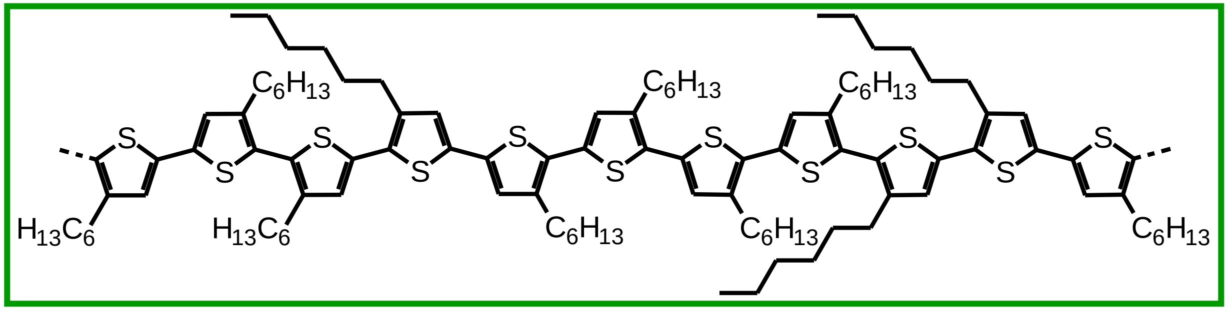
green: desired processes

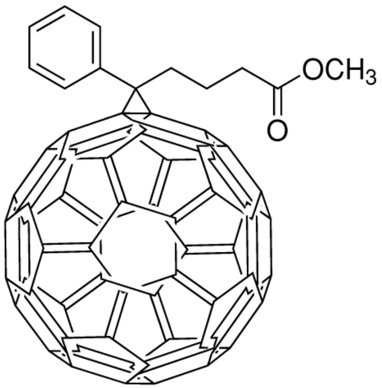
red: loss processes

Context

- How CT states affect PV efficiency

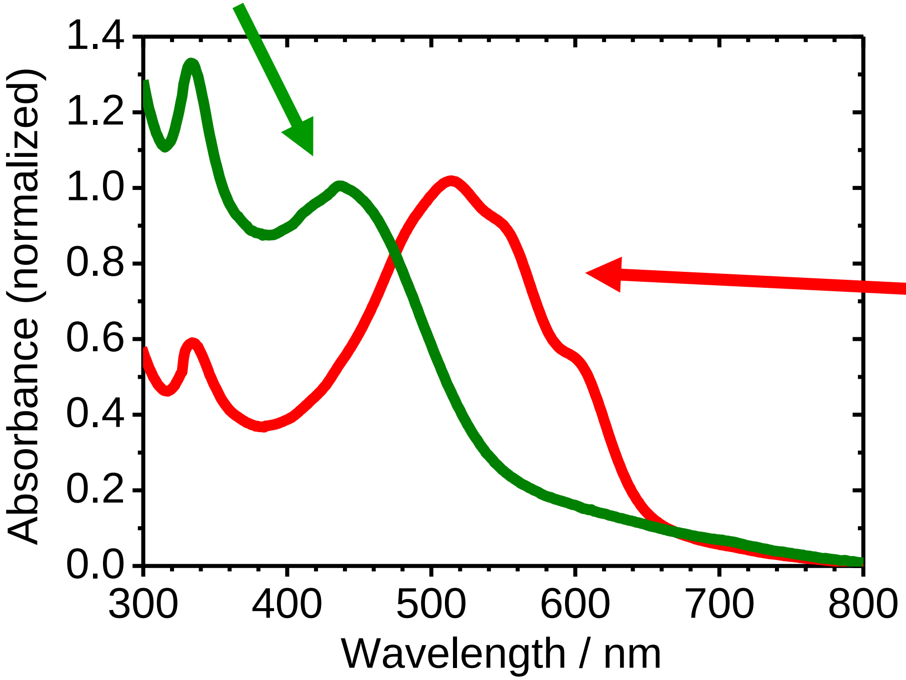


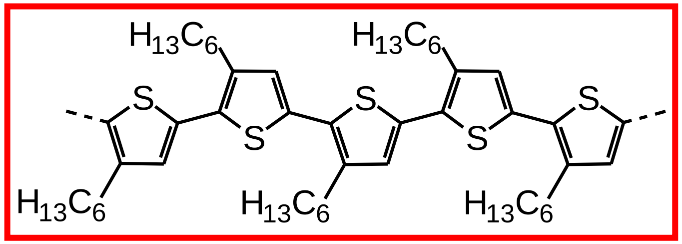




$PC_{61}BM$

PCE = 0.03 %

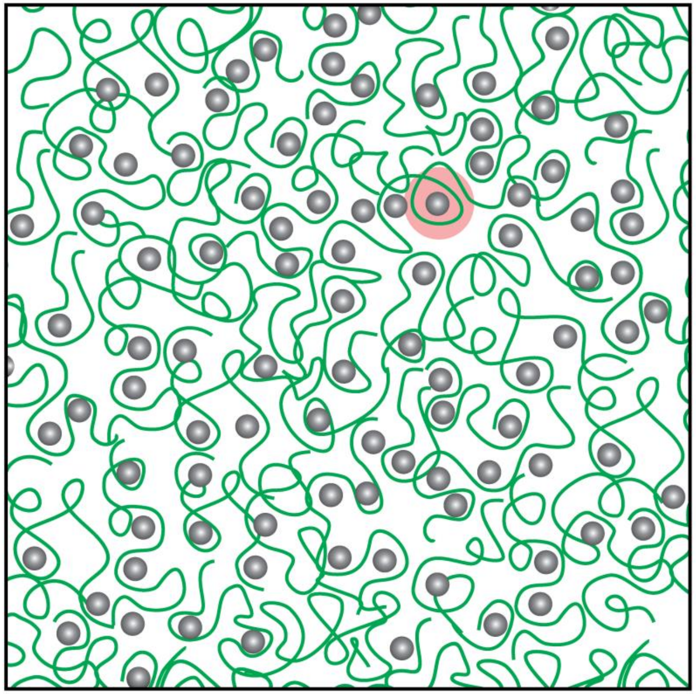




PCE = 3.1 %

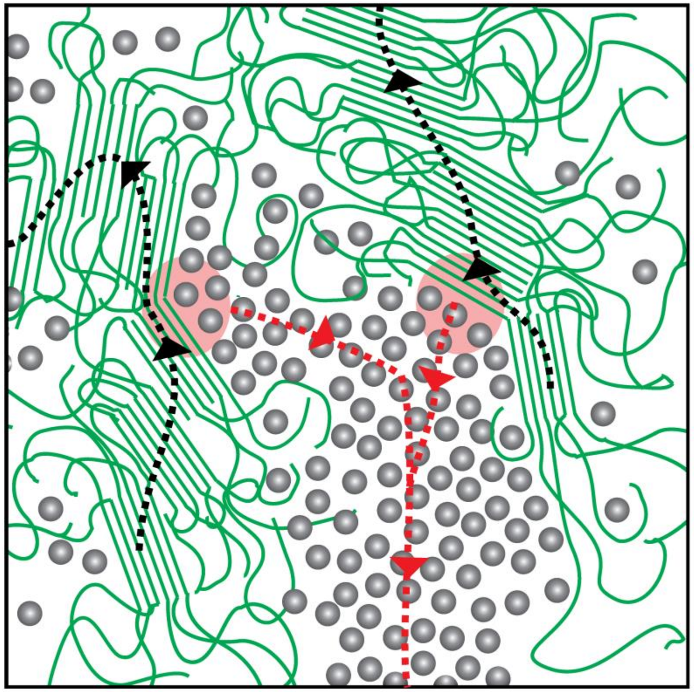
Transient absorption spectra of P3HT:PCBM blends

RRa-P3HT:PCBM
(amorphous, disordered)



→ molecularly mixed blend

RR-P3HT:PCBM
(crystalline, ordered)

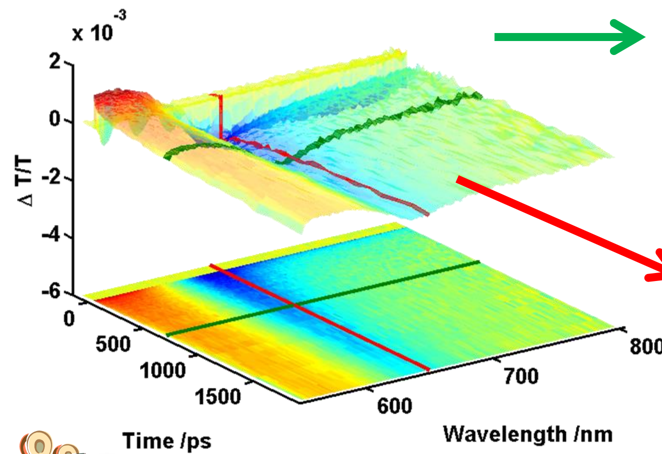


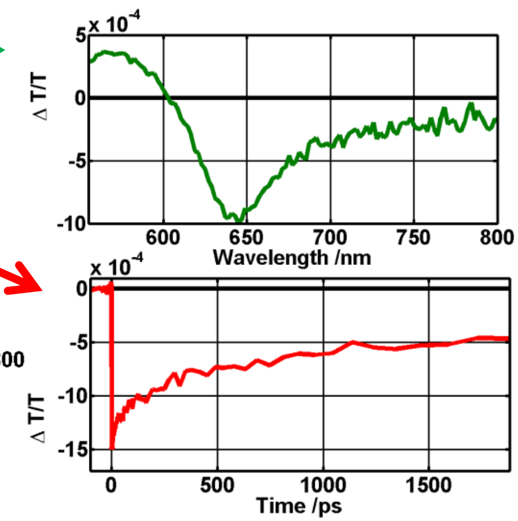
→ demixed blend, larger pure domains

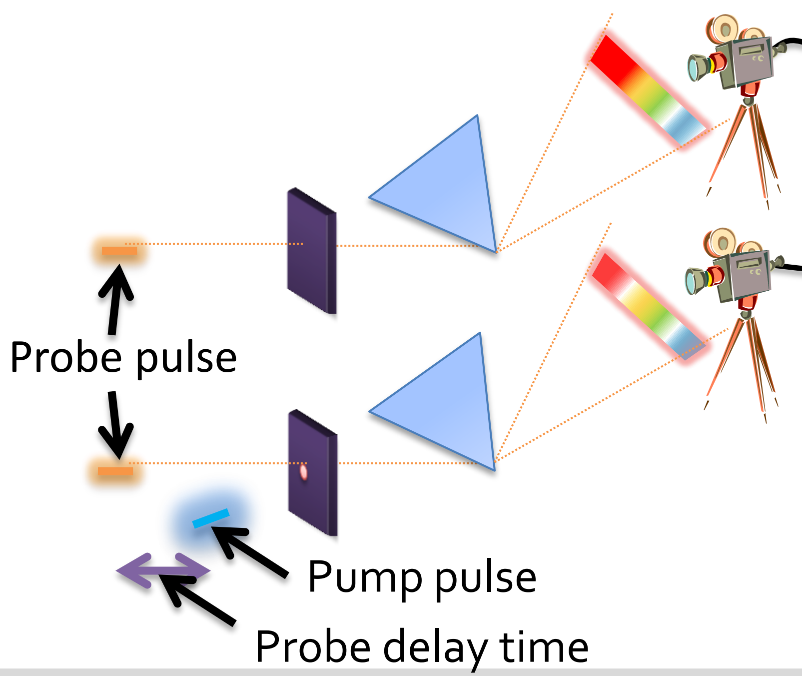
I.A. Howard, R. Mauer, M. Meister, F. Laquai, *J. Am. Chem. Soc.* **2010**, *132* (42), 14866-14876.

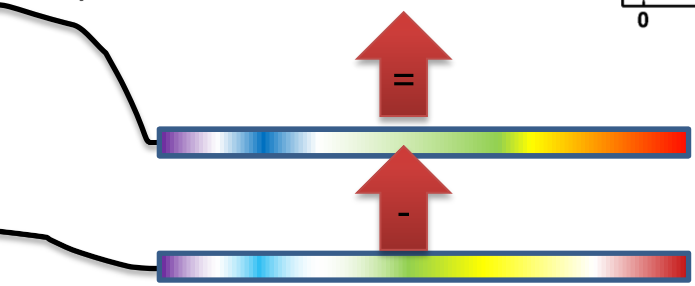
Transient Absorption Spectroscopy

- TAS is a pump-probe technique
- Measures the change in transmission induced by excited-states at given delay after photoexcitation









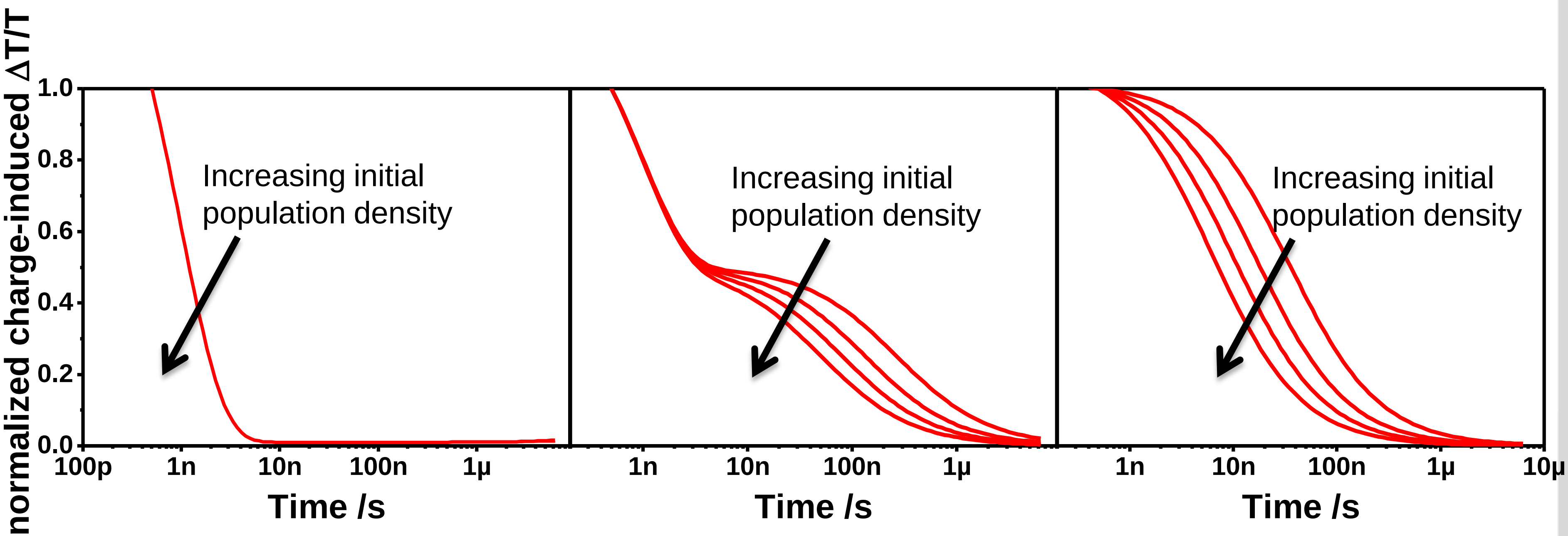
- Modeling of the TAS surface reveals population dynamics
- And more …

Intensity dependence of charge population

Only CT states

50% CT states
50% free charges

Only free charges



Test case: charge generation and morphology

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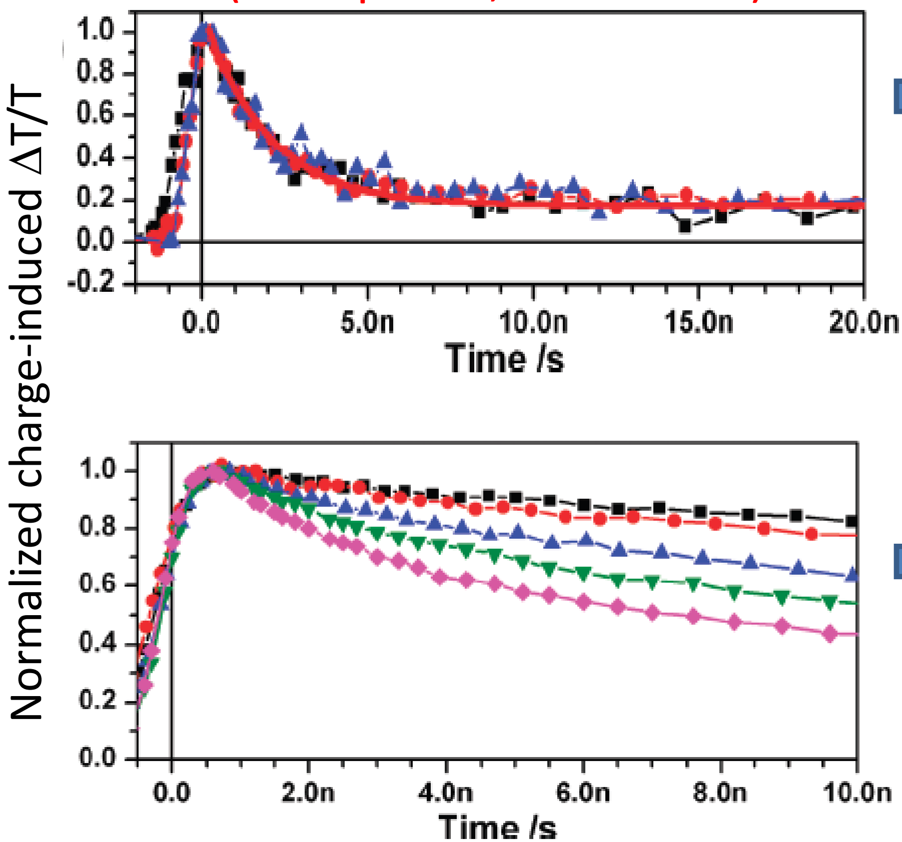
RRa-P3HT:PCBM
(amorphous, disordered)



RR-P3HT:PCBM
(crystalline, ordered)

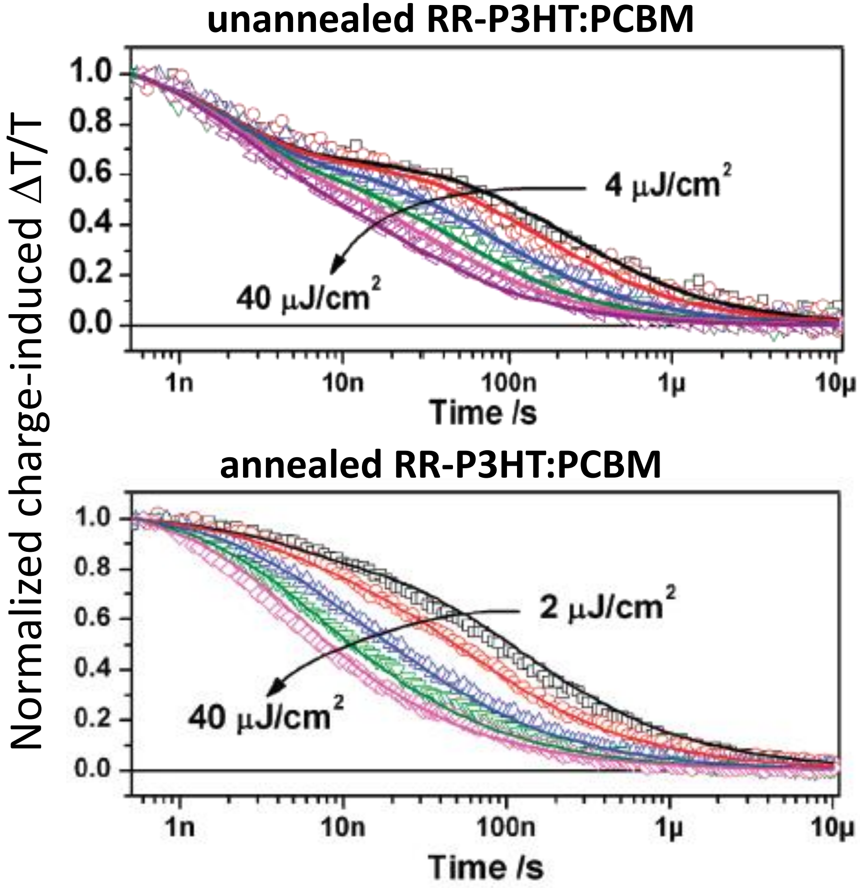
$$\frac{dn}{dt} \sim k\, n$$

geminate recombination of trapped interfacial **charge-transfer (CT) states**

$$\frac{dn}{dt} \sim \gamma\, n^{\lambda+1}$$

non-geminate recombination of mobile and **free charge carriers (SSC)**

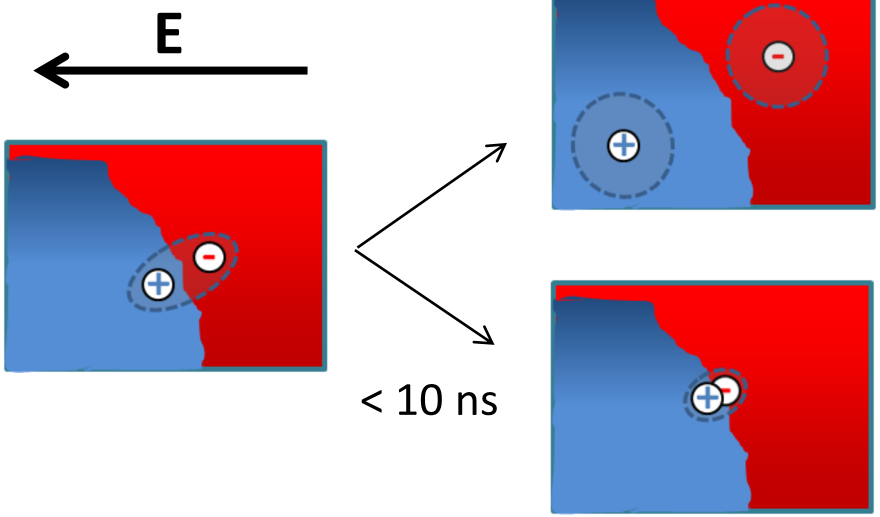
Test case: charge generation and morphology

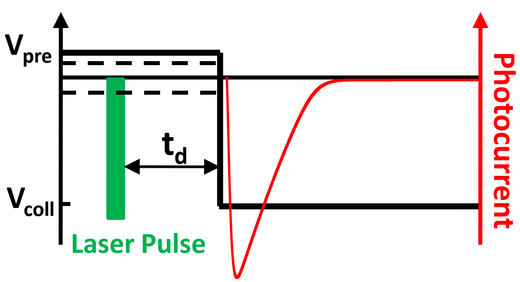


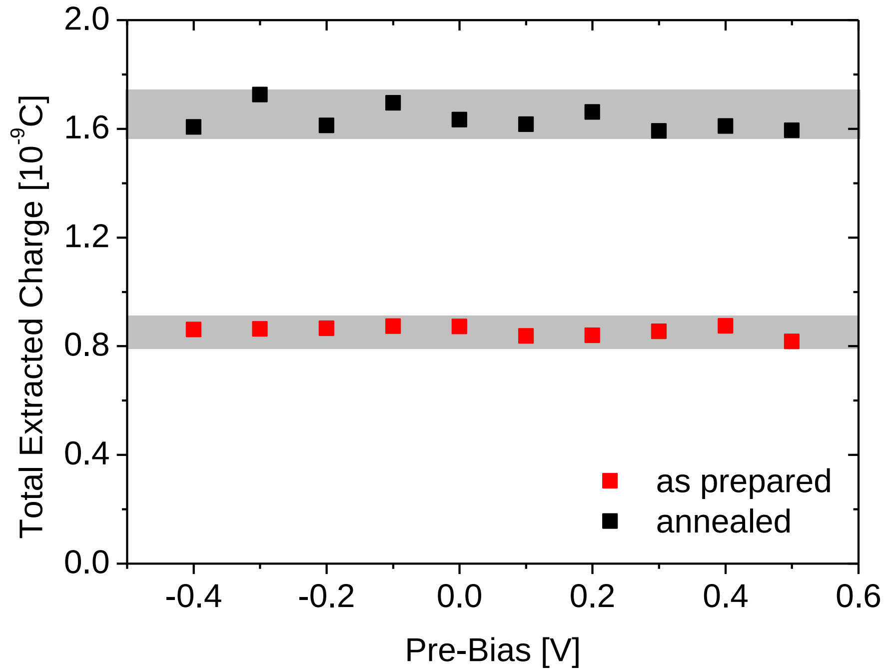
~40% relaxed CT state recombination
~60% recombination of free charges

< 20% relaxed CT state recombination
~ 80% recombination of free charges

Time-delayed collection field experiments – D. Neher







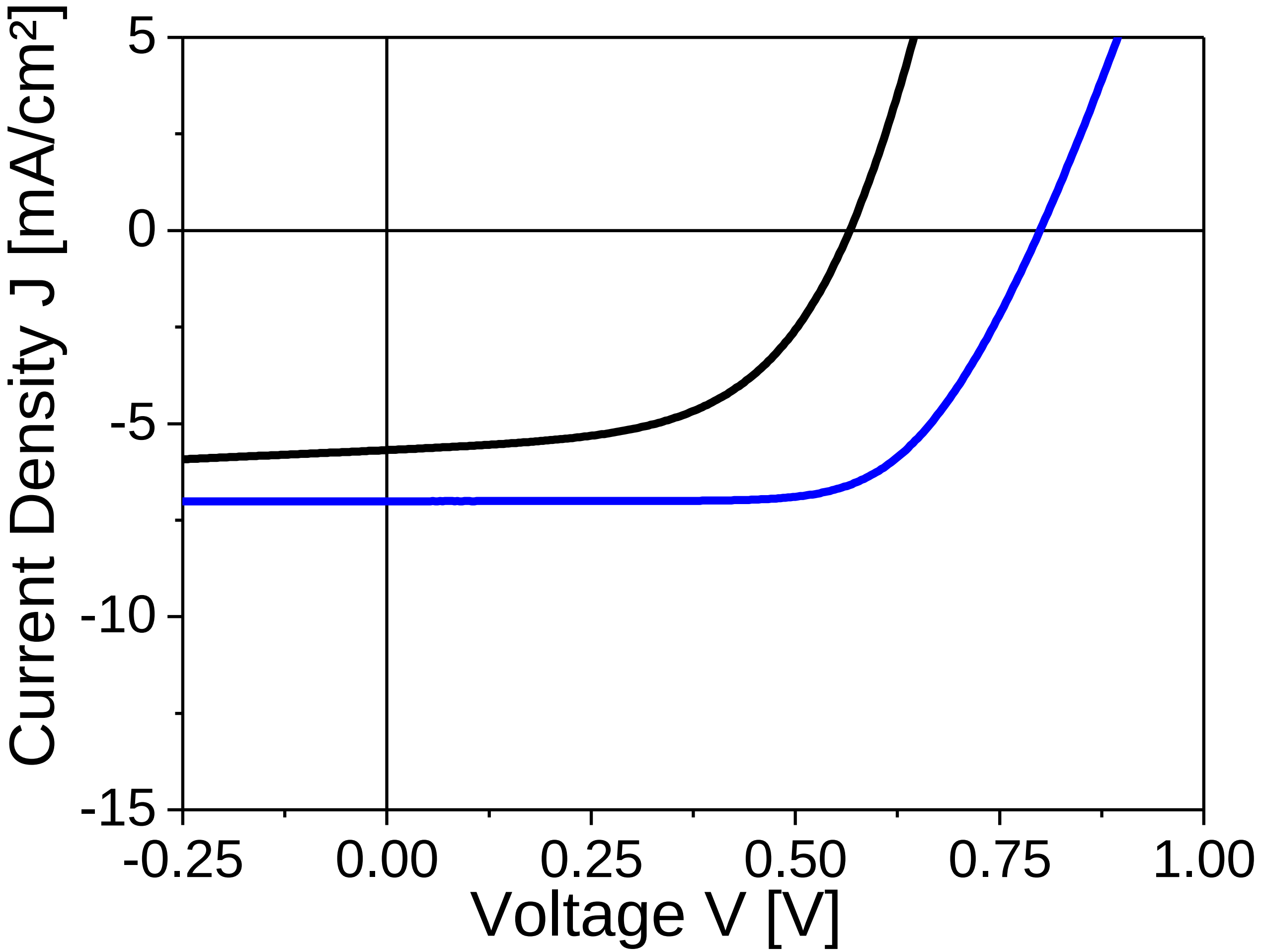
t_d = 20 ns
V_{coll} = -5 V
Pulse fluence: 0.7µJ/cm²

Extracted charge independent of pre-bias

→ free charge generation independent of electric field
→ FF must be limited by non-geminate recombination

Juliane Kniepert, Marcel Schubert, James C. Blakesley, and Dieter Neher, *J. Phys. Chem. Lett.* **2011**, *2*, 700–705.

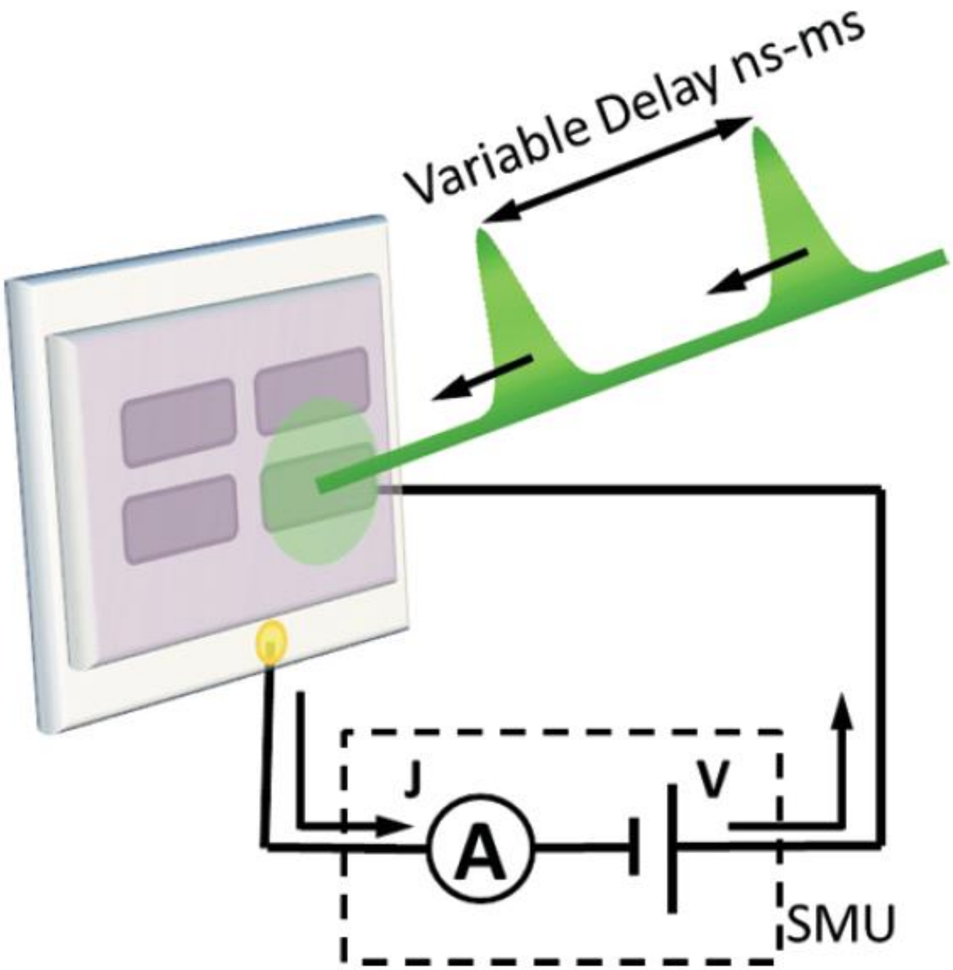
Free charges made in absence of field! Then what explains FF?



If CT separation does not determine fill factor what does?

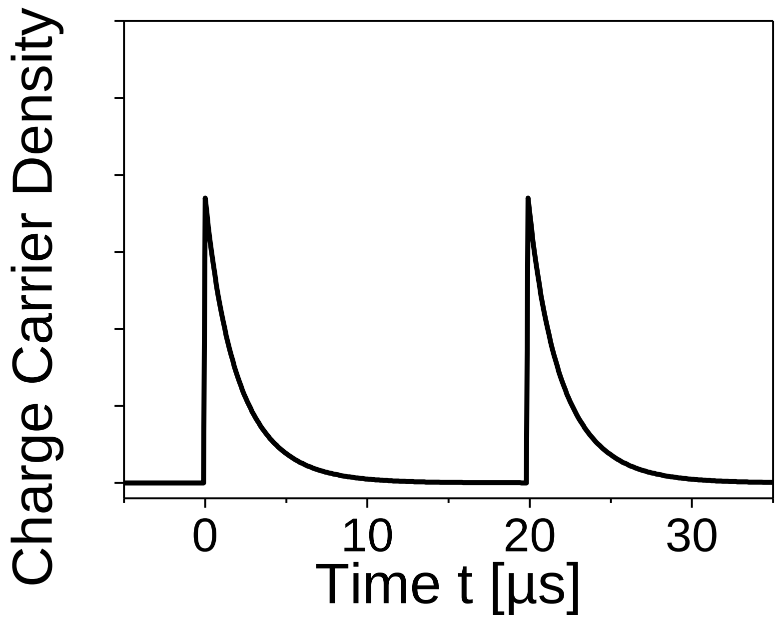
Electrical readout
- **Sensitive (measure devices easily under 1 sun conditions)**
- Poor time resolution

Optical readout
- Insensitive (challenge to measure devices under 1 sun short-circuit charge densities)
- **Excellent time resolution**



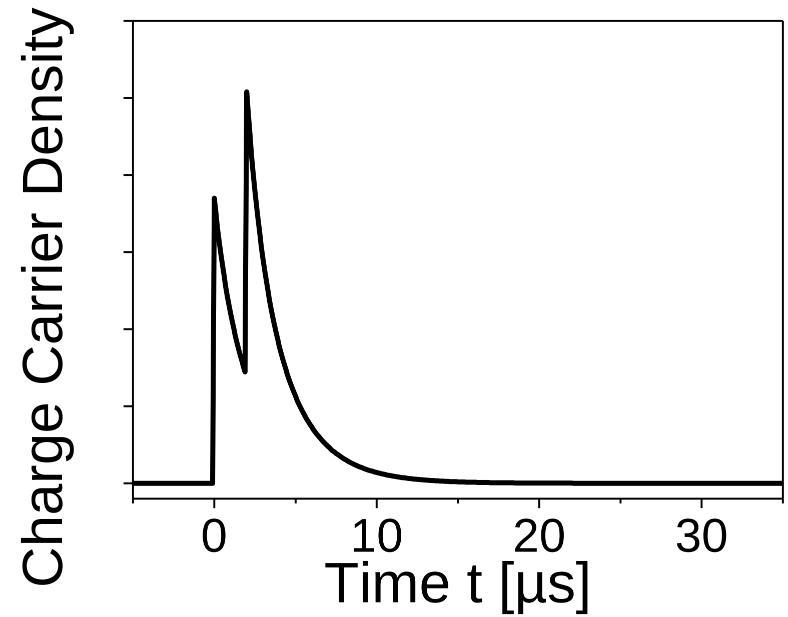
Double pump – photocurrent measurement

$n_{extr} = n_1 + n_2$

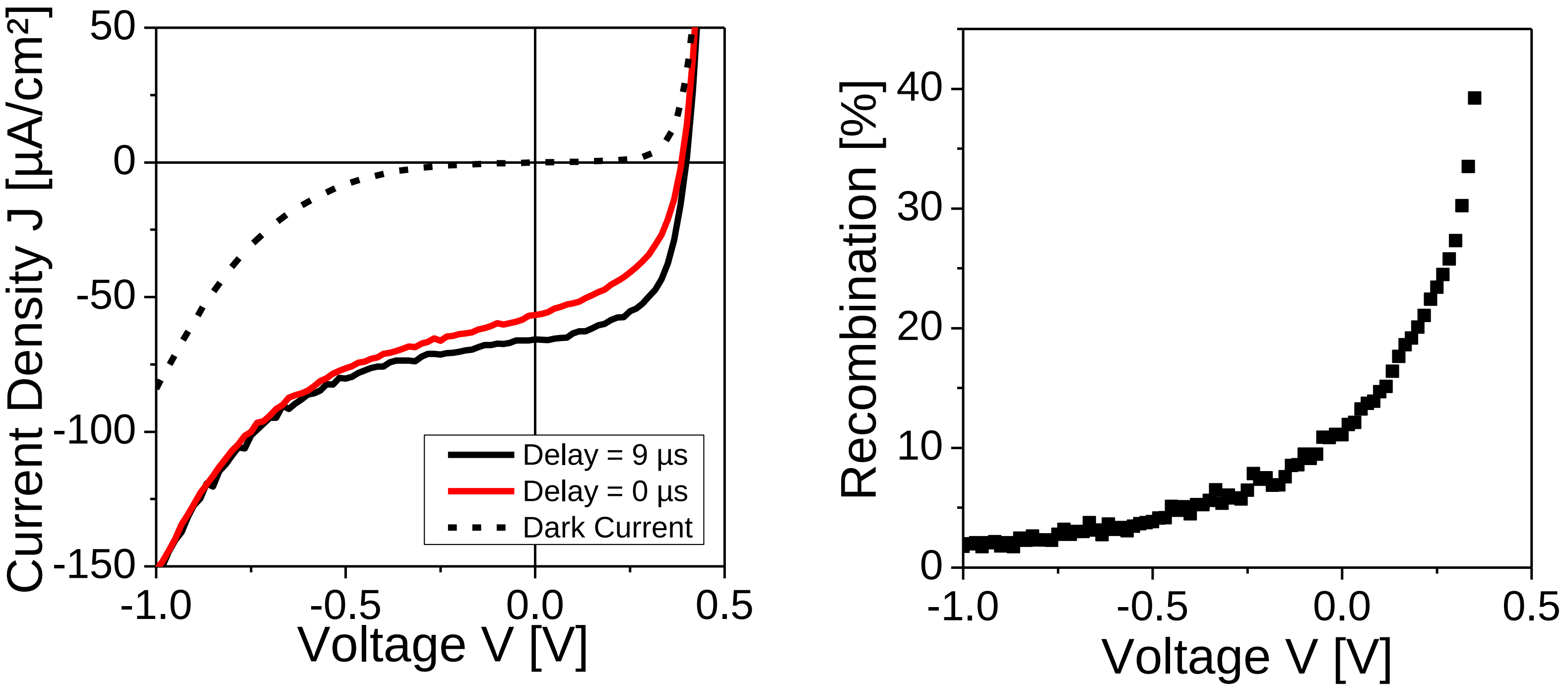


$n_{extr} = n_1 + n_2 - X$

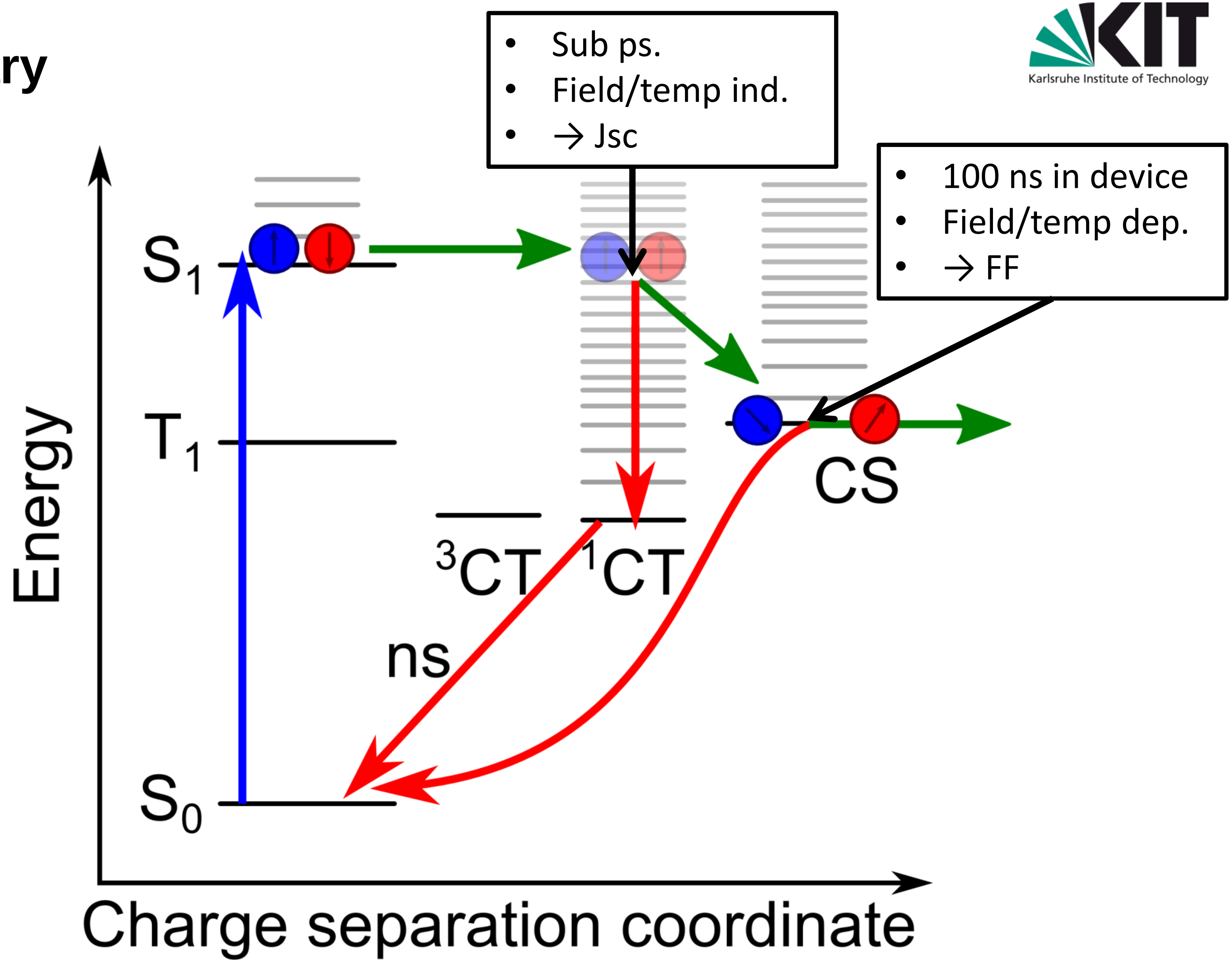
X= additional recombination



Field dependence of extraction determines FF



Summary

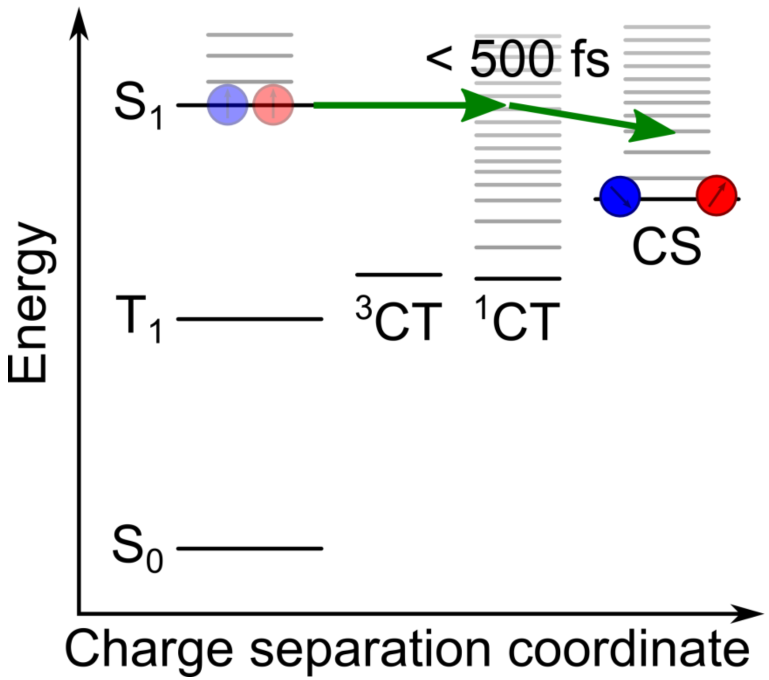


• Sub ps.
• Field/temp ind.
• → Jsc
• 100 ns in device
• Field/temp dep.
• → FF
Energy
S_1
T_1
S_0
^{3}CT
^{1}CT
CS
ns
Charge separation coordinate

Summary

Voc → contacts, HOMO/LUMO, photocurrent
(diffusion current in special cases)
Jsc → exciton quenching, CT separation
FF → competition between extraction and recombination (in poor systems can be affected by CT splitting)

Many blends show excellent charge separation now. Perhaps low dielectric organic semiconductors are better suited for solar cells than one would think!



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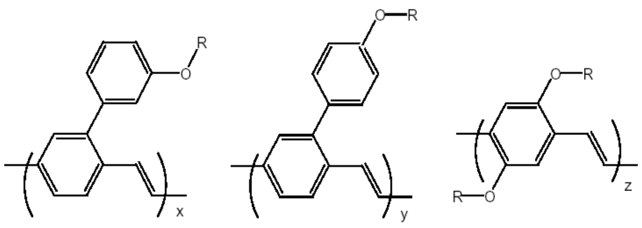
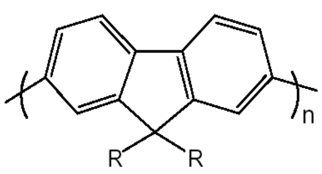
Materials for Organic Electronics
Two approaches lead to similar electronic properties

Conjugated polymers



R = $(CH_2)_3CH(Me)(CH_2)_2CHMe_2$

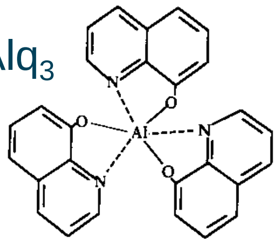
PPV co-polymers

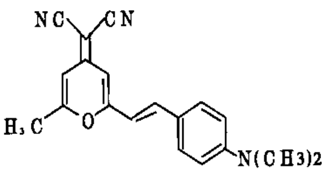


Polyfluorene

Small evaporated molecules

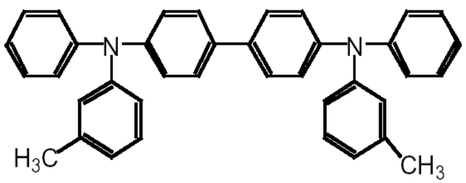
Alq_3



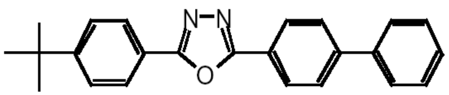


(DCM)

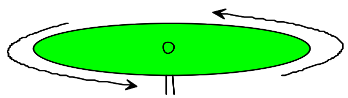
TPD



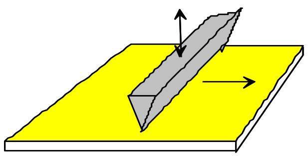
PBD



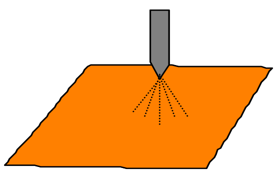
Organic Semiconductor Deposition



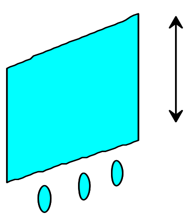
Spin Coating



Rakeln

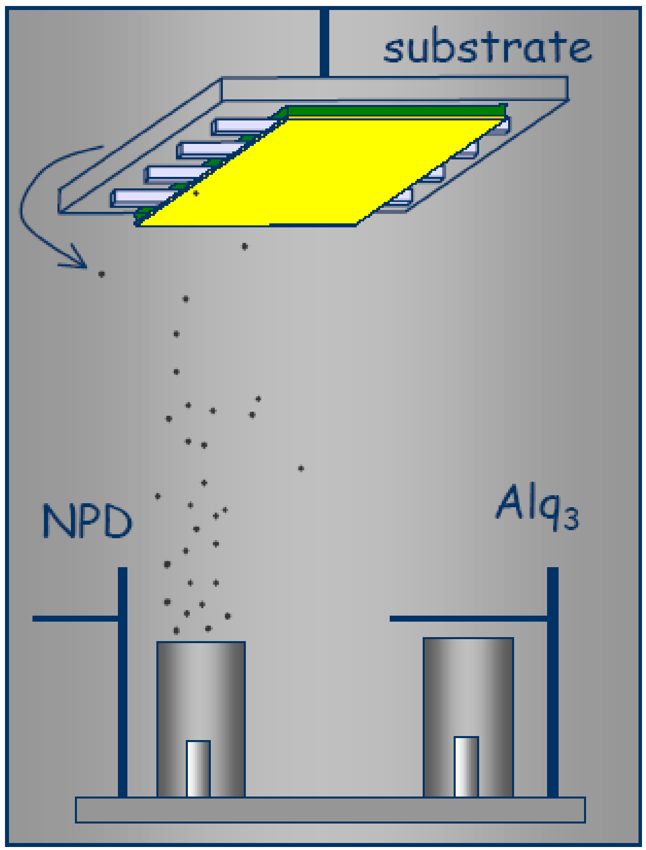


Ink Jet Printing



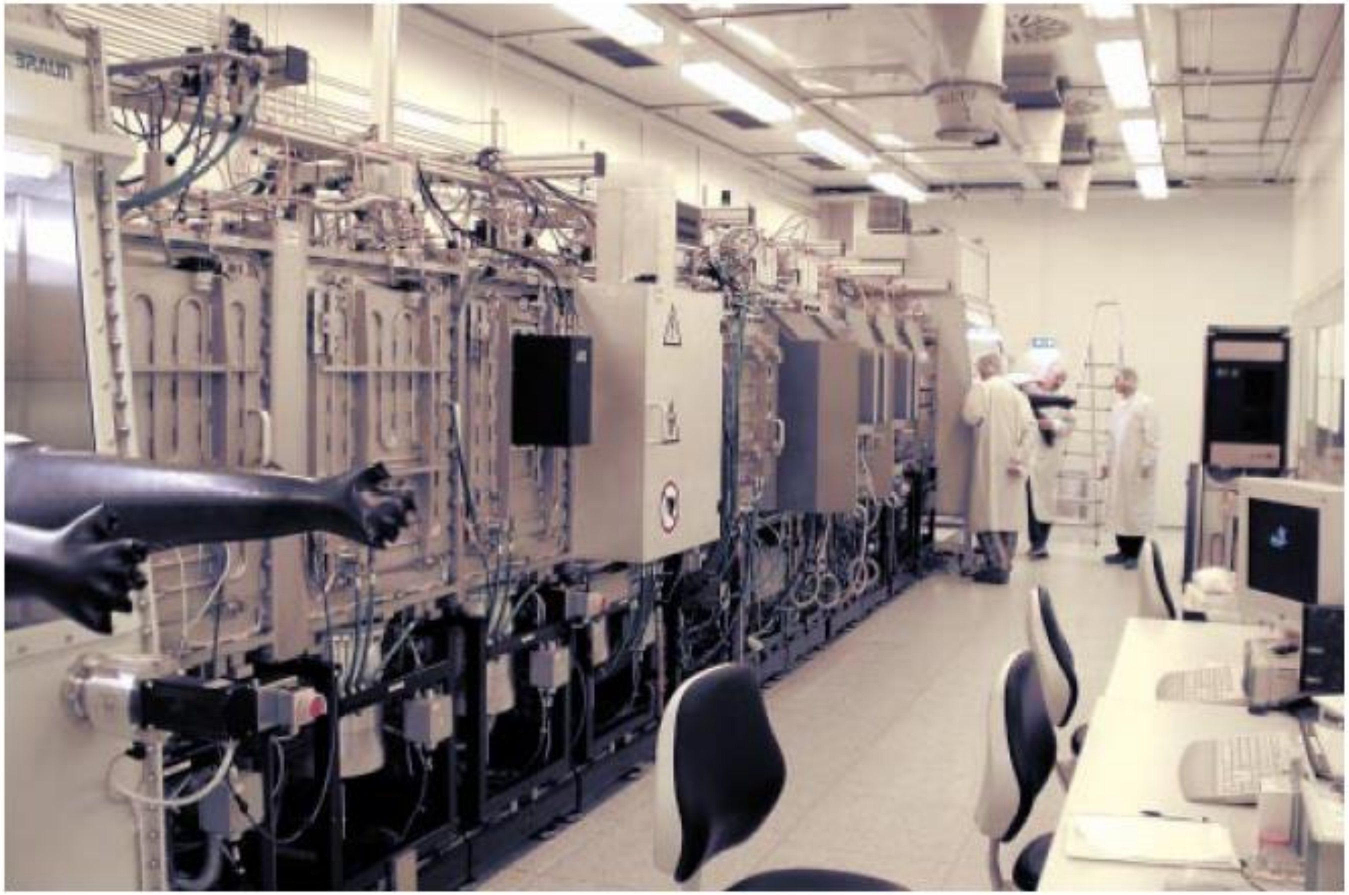
Dipping

Solution processing/Coating/Printing



Evaporation of small molecules

Evaporation: in-line equipment



11m long in-line evaporation equipment at Fraunhofer IPMS Dresden

Competence Center Printing & Device Physics @ InnovationLab in Heidelberg

Mission

Lab-to-Fab

Partners BASF SE, Merck KGaA, KIT, TU Darmstadt

1 - Proof of concept

2 - Prototype

3 - Scale Up

Proof of concept

- Fluid < 10 ml
- Low material usage
- Direct feedback for material design and formulation development

Large-Area-Prototyping

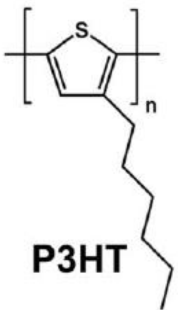
- Fluid < 100 ml
- Lab-scale
- Up to letter-size substrates
- Targeted lateral resolution: < 10 µm

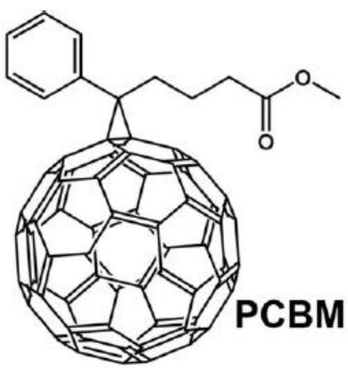
R2R R&D platform

- Fluid < 1 l
- R&D pilot line for production research
- Web width: 330 mm
- Web speed: up to 100 m/min


PEP3

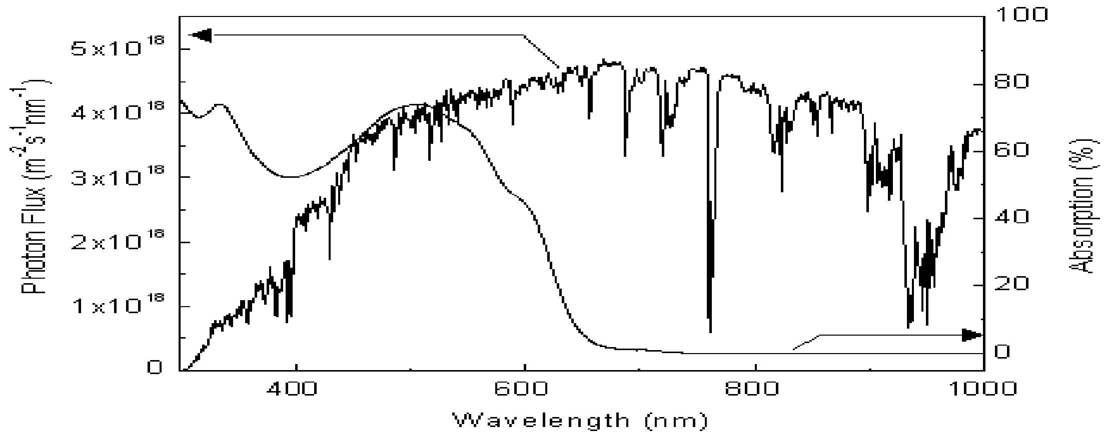
The need for lower bandgap absorbers

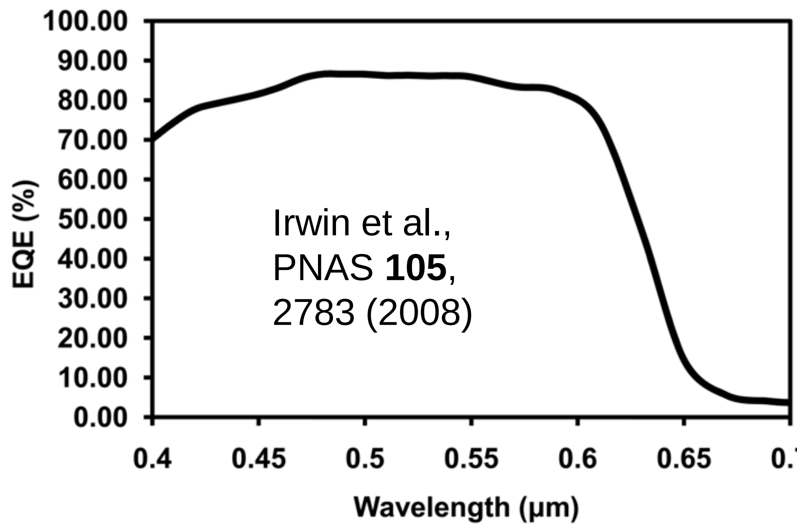




P3HT

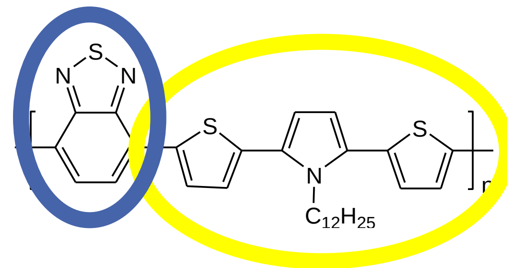
PCBM



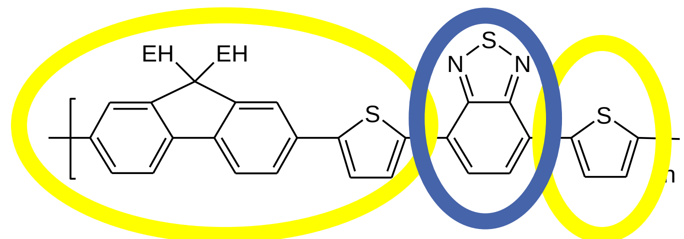


Irwin et al., PNAS **105**, 2783 (2008)

Low bandgap polymers

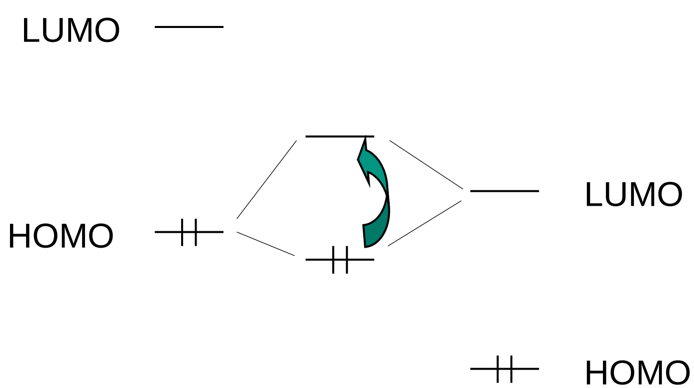


PTPTB



PFDTBT

Donor + Acceptor → internal charge transfer complex in which the **whole** polymer acts as an electron donor



Low bandgap polymers

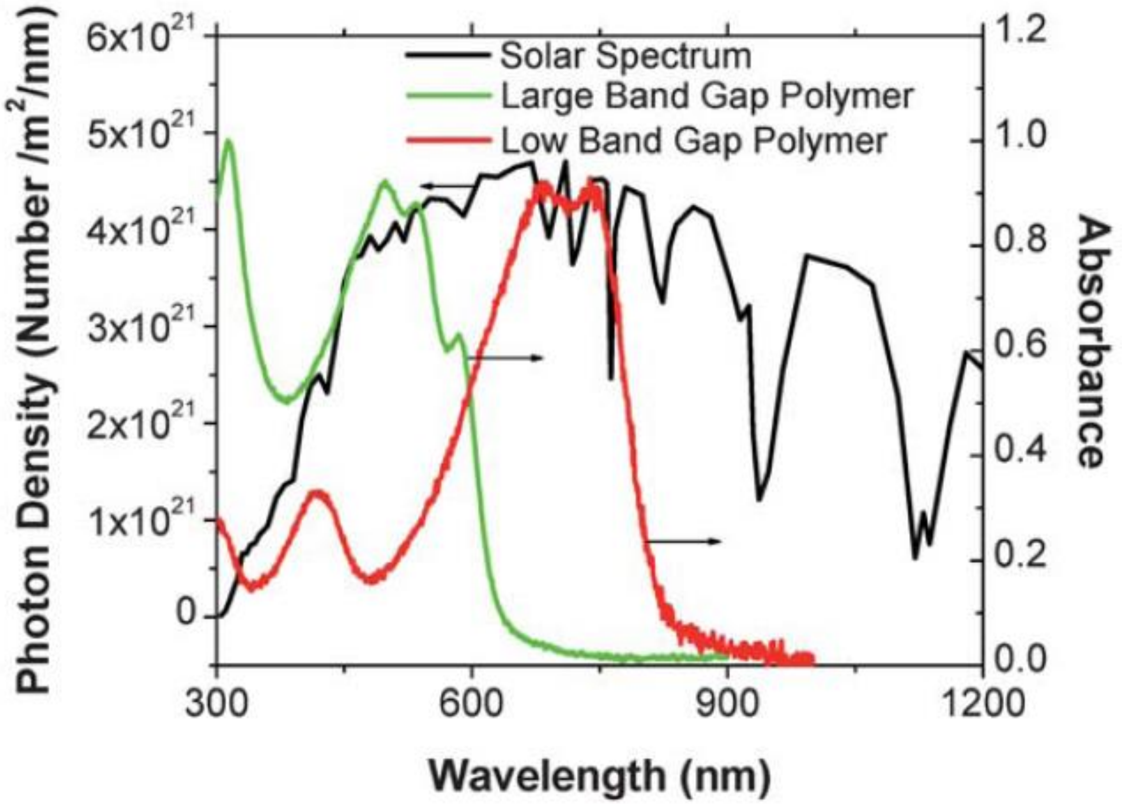
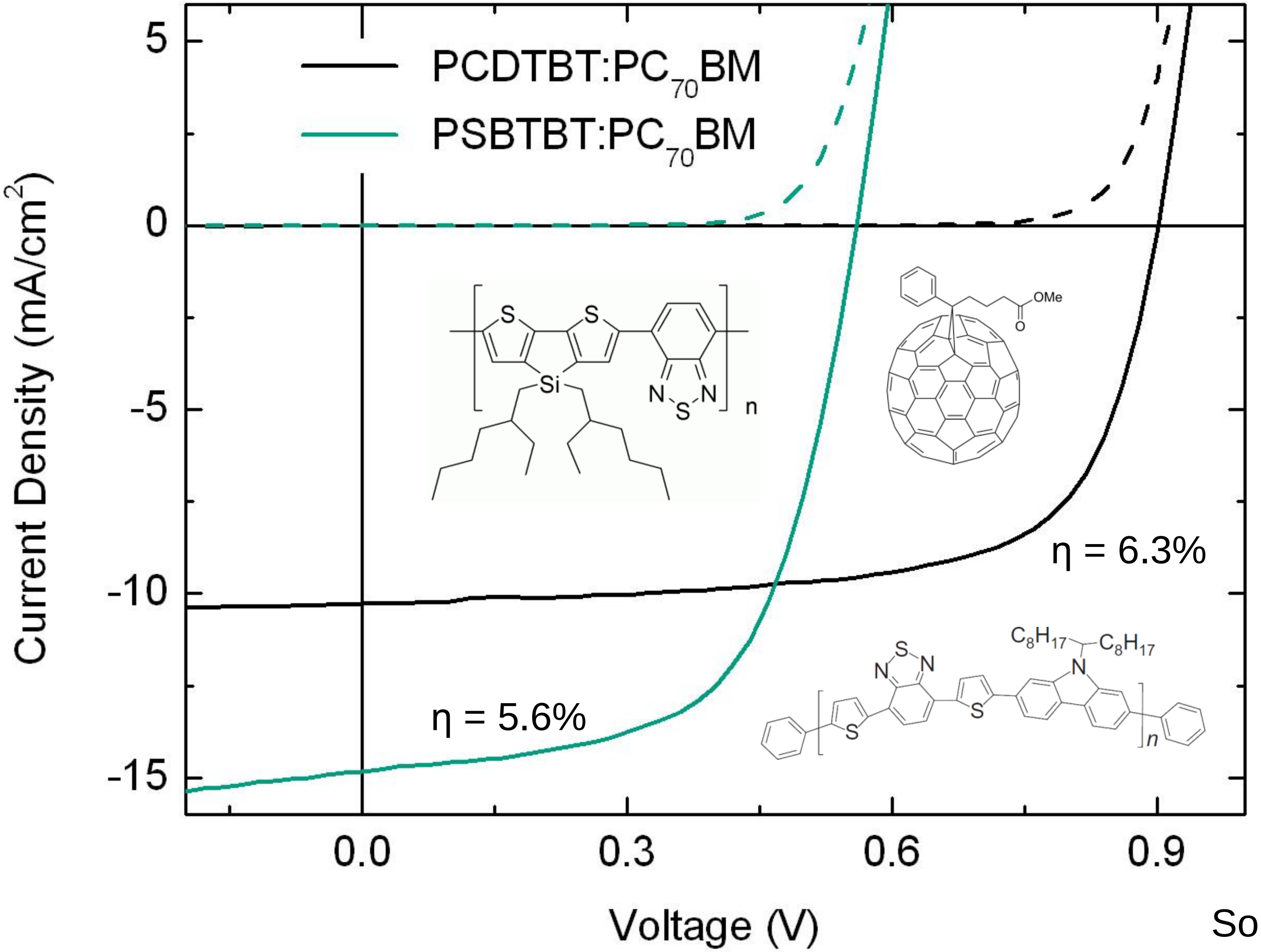


Fig. 1 Absorbance of P3HT a large band gap polymer (green) and PSBTBT a low band gap polymer (red) is compared to the solar spectrum which spans from 300–1200 nm and even beyond.

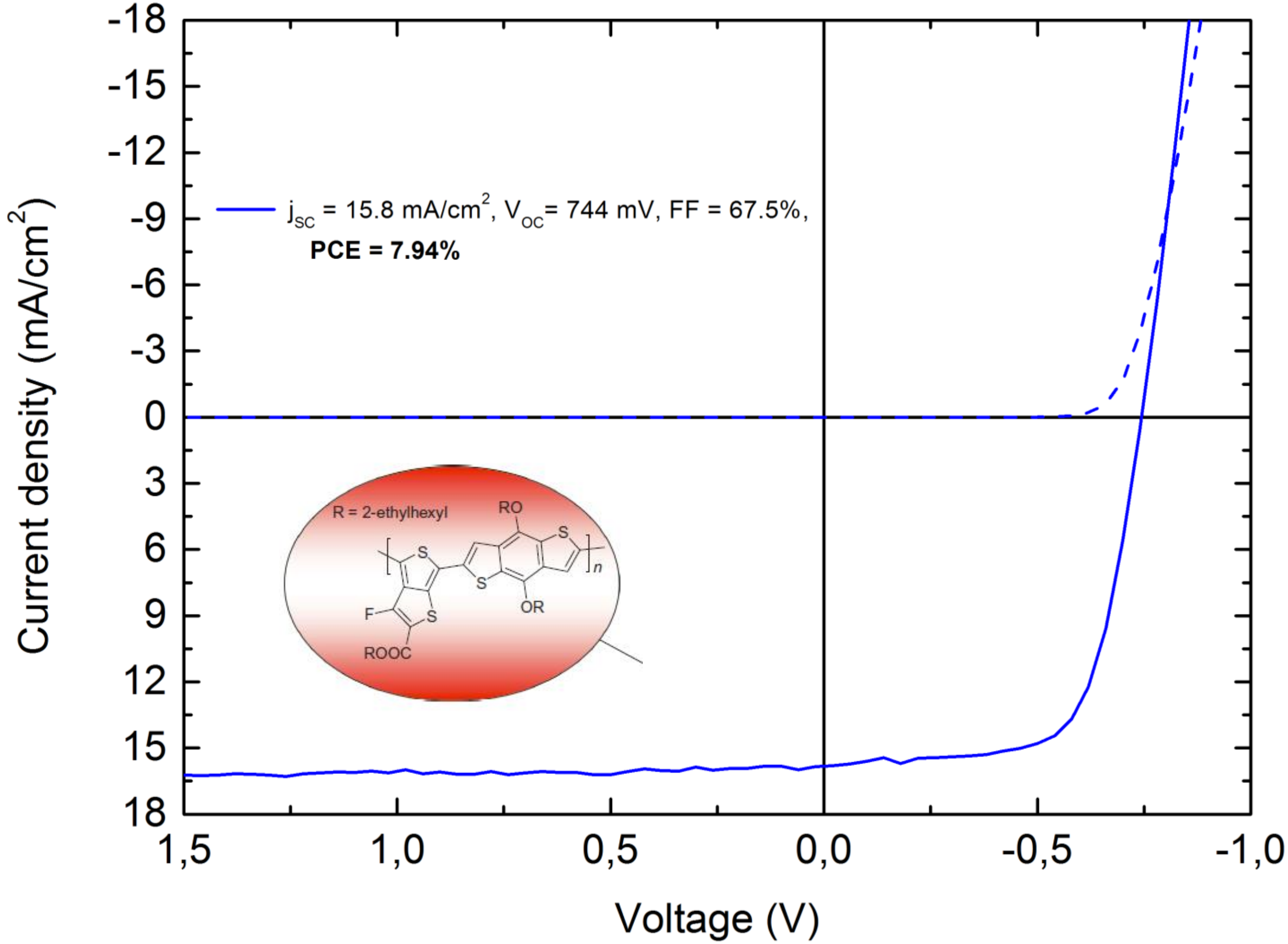
Source: Sista et al., Energy Environ. Sci. 4, 1606 (2011)

Highly efficient single layer devices in 2011



Source: LTI

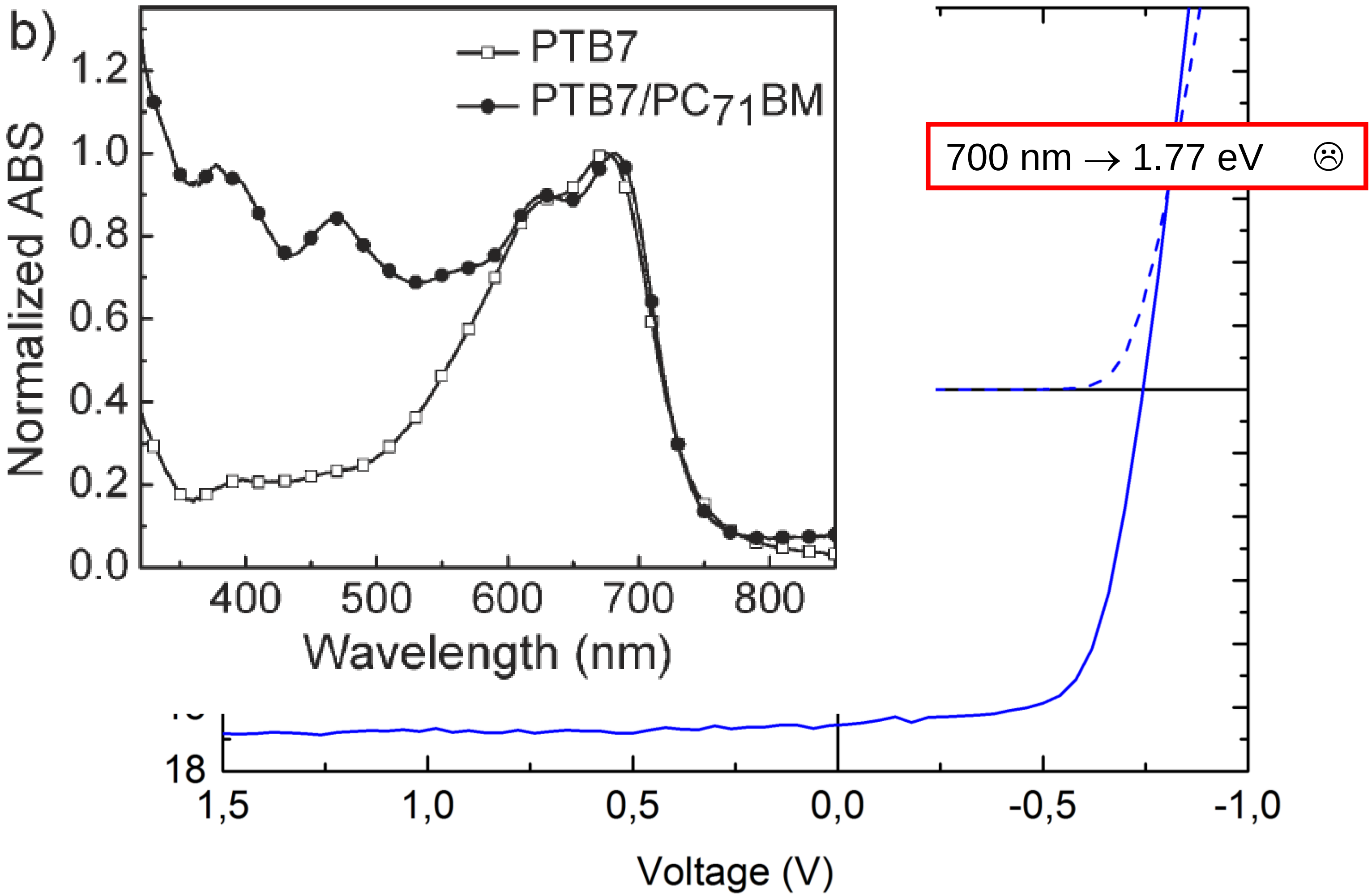
Highly efficient single layer devices in 2013



Source: LTI

Highly efficient single layer devices in 2013

Source: Liang et al., Adv. Mater. 2010, 22, E135–E138m



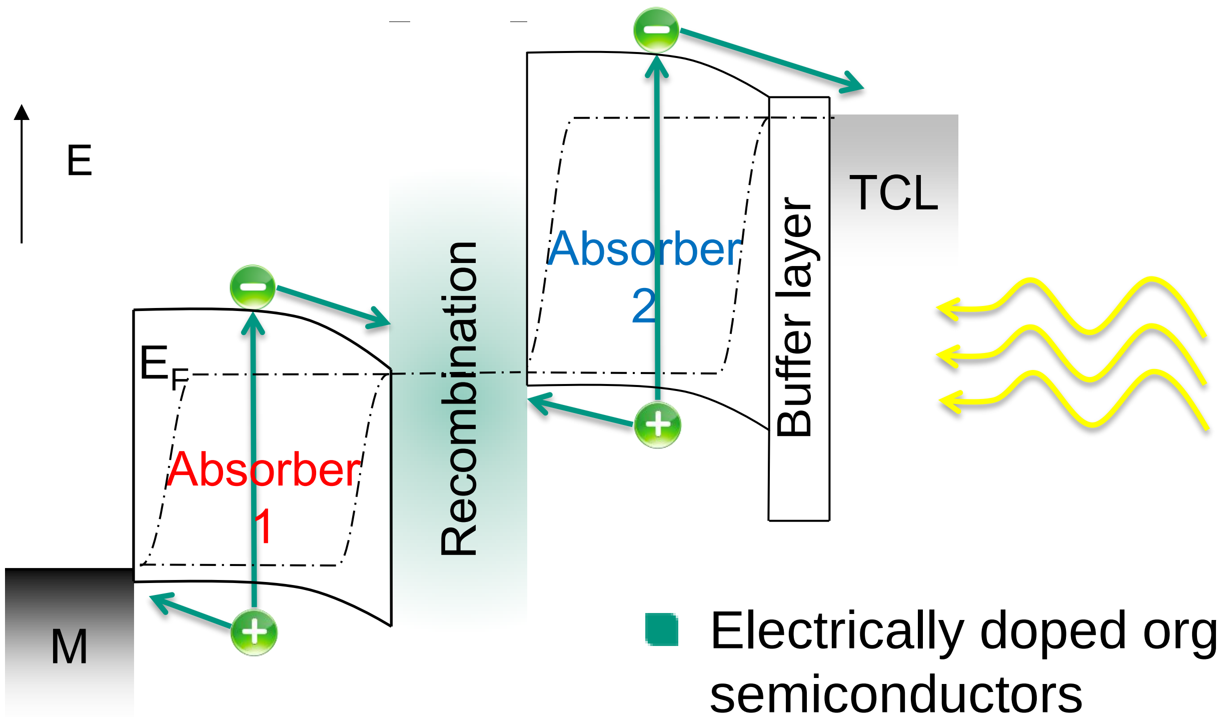
Outline

Karlsruher Institut für Technologie

Part I: Organic Photovoltaics

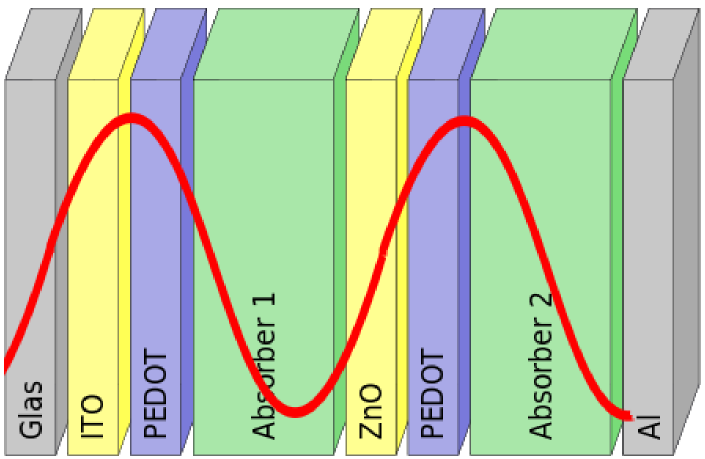
1. Introduction
2. Bulk Heterojunctions
3. Excitons → Free Charge
4. Low Bandgap Absorbers
5. Semitransparent, Tandem Solar cells, Electrodes
6. Nanophotonics for Organic Solar Cells

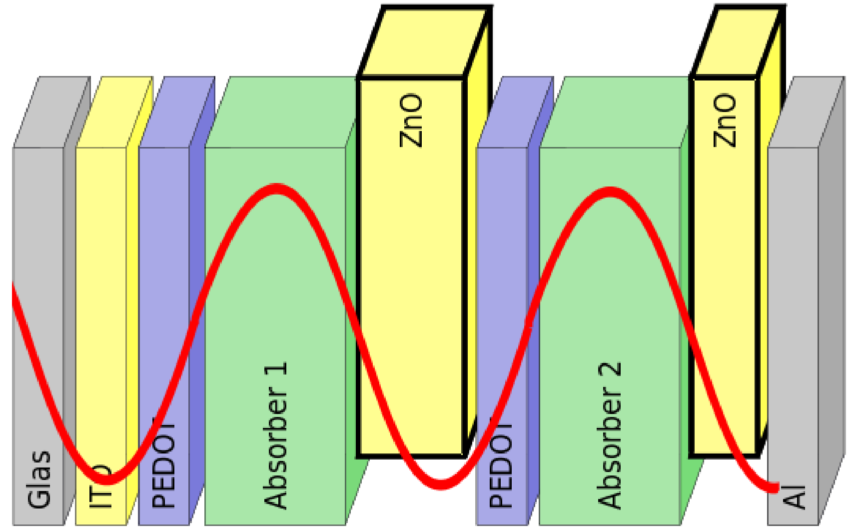
Tandem solar cells



- Electrically doped organic semiconductors
- Metal oxides from precursors
- Metal oxide nanoparticles

Recombination layer



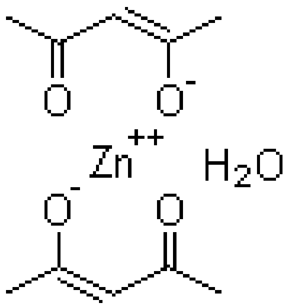


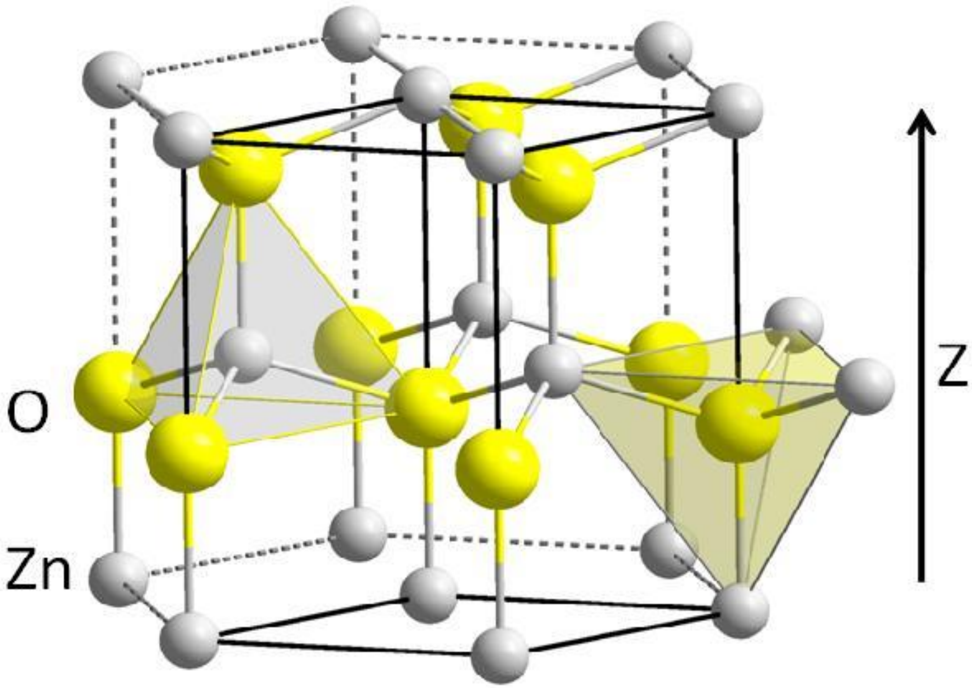
Requirements:

- Low absorption
- Good (selective) conductivity
- Easy processing

Zincoxide from precursor solution

- Zinc acetylacetonate hydrate
  $Zn(acac)_2$

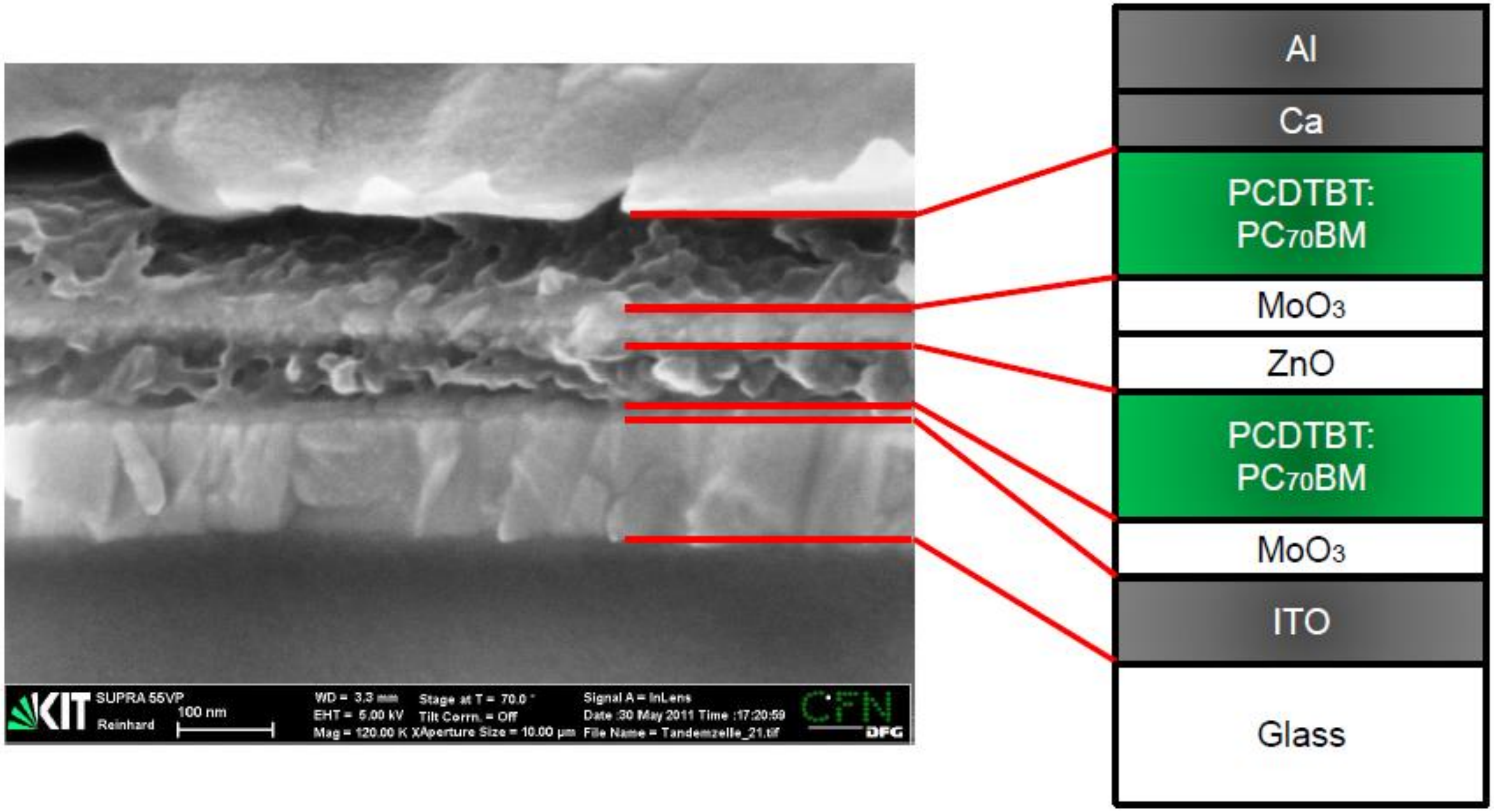




P. de Bruyn et al. , Organic Electronics 11 (2010) 1419–1422

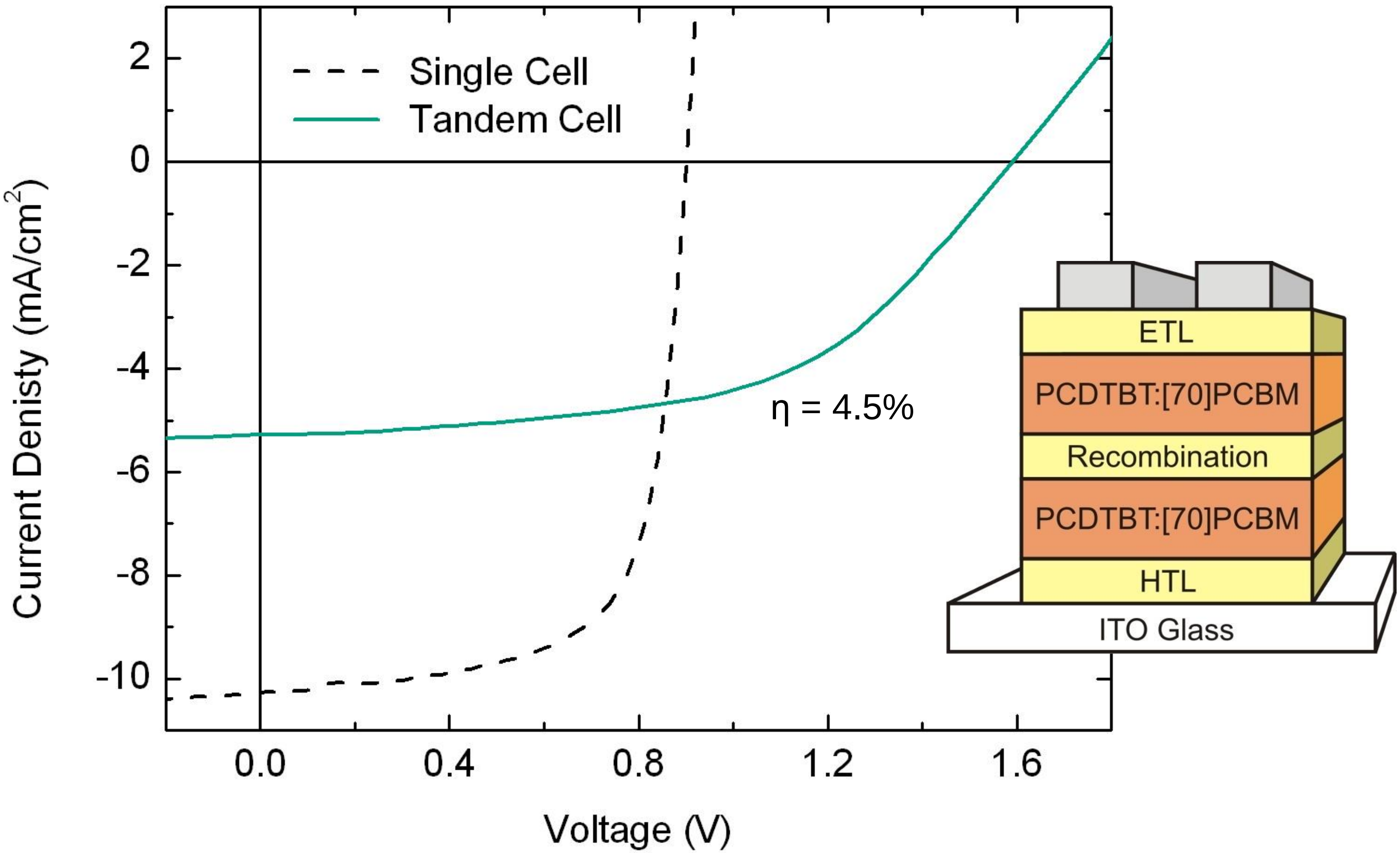
*Source: Wikipedia and ChemBlink

(Almost) Fully Solution Processed Tandem Solar Cell



A. Pütz et al. Organic Electronics **13**, 2696 (2012).

Tandem solar cells



A. Pütz et al. Organic Electronics **13**, 2696 (2012).

ARTICLE

NATURE COMMUNICATIONS | DOI: 10.1038/ncomms2411

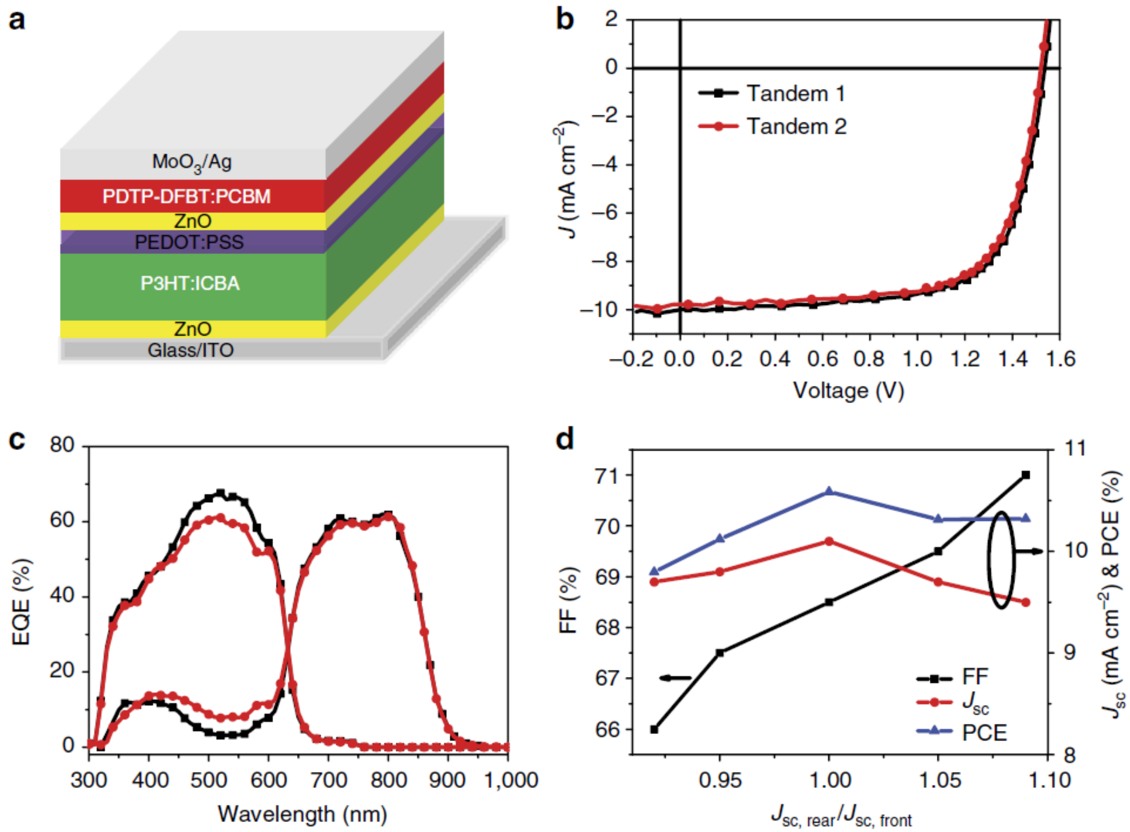
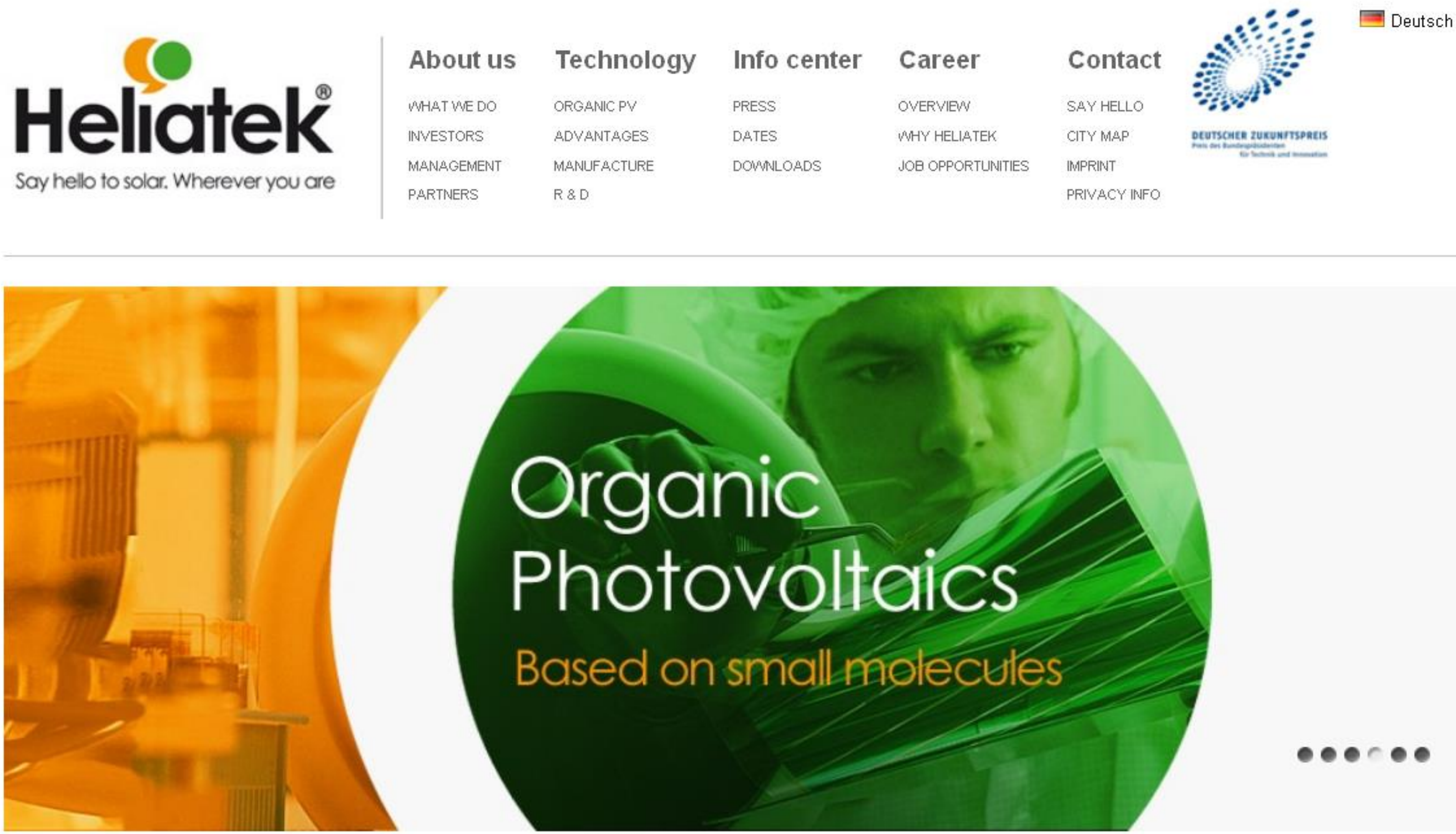


Figure 6 | Tandem devices structure and performance. (**a**) Device structure of the tandem solar cell (Glass/ITO/ZnO/P3HT:ICBA/PEDOT:PSS/ZnO/PDTP-DFBT:PCBM/MoO_3/Ag). (**b**) *J–V* curve of P3HT:ICBA/PDTP-DFBT:PC_{61}BM combination (Tandem 1) and P3HT:ICBA/PDTP-DFBT:PC_{71}BM combination (Tandem 2) under AM1.5G illumination from a calibrated solar simulator with an irradiation intensity of 100 $mWcm^2$ (one Sun). (**c**) EQE of the tandem 1(black line) and 2 (red line) devices. A 700 and 550 nm light bias are used to get front and rear cell EQE, respectively. (**d**) The relationship of tandem cell FF and short circuit current (J_{SC}) versus rear and front cell current ratio ($J_{SC,\ rear}/J_{SC,front}$).



www.heliatek.de

Heliatek

Say hello to solar. Wherever you are

Heliatek sets new world record efficiency of 10.7% for its organic tandem cell

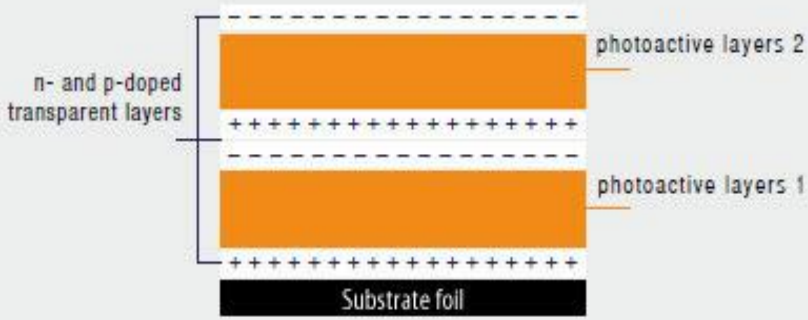
April 27, 2012

Heliatek GmbH, technology leader in the field of organic solar films, continues to set new world records for organic solar cells. The company has commissioned SGS, an accredited and independent testing facility, with a measurement campaign of its latest organic photovoltaic (OPV) cells. The result of this campaign is a new world record for OPV with 10.7 % cell efficiency on 1.1 cm2. It also confirms the superior low light and high temperature performances of OPV compared to traditional solar technologies.

Heliatek‘s Technology

KIT
Karlsruher Institut für Technologie

Build of a tandem cell



- patented tandem cell technology
- complementary absorber systems
  > optimum harvesting of the complete sun spectrum
- increased open circuit voltage
- loss-free recombination contact between individual cells within tandem cell
- n- and p-doped transparent layers allow for the loss-free charge transport to the electrodes.

Part of the solution to both the worlds energy and environmental issues

Affordable energy for emerging nations

Light-weight, flexible energy on the move

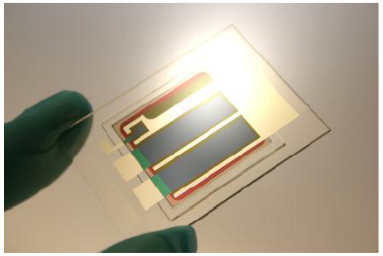
Heliatek®

Say hello to solar. Wherever you are

Heliatek consolidates its technology leadership by establishing a new world record for organic solar technology with a cell efficiency of 12%

Dresden, Germany, January 16, 2013 – Heliatek GmbH, the leader in organic solar films, today announced a record breaking 12.0% cell efficiency for its organic solar cells. This world record, established in cooperation with the University of Ulm and TU Dresden, was measured by the accredited testing facility SGS. The measurement campaign at SGS also validated the superior low light and high temperature performances of organic photovoltaics (OPV) compared to traditional solar technologies.

The 12.0% record cell on a standard size of 1.1 cm² combines two patented absorber materials, which convert light of different wavelengths. Using two different absorber materials creates a stronger absorption of photons and improves energetic utilization through a higher photovoltage. Thanks to OPV's unique behavior at high temperatures and low light conditions, this 12% efficiency is comparable to about 14% to 15% efficiency for traditional solar technologies like crystalline silicon and thin film PV. Whereas those technologies significantly lose cell efficiency with rising temperatures and decreasing solar irradiation, organic cells increase their efficiency in these conditions leading to a much higher energy harvesting in real life environments.

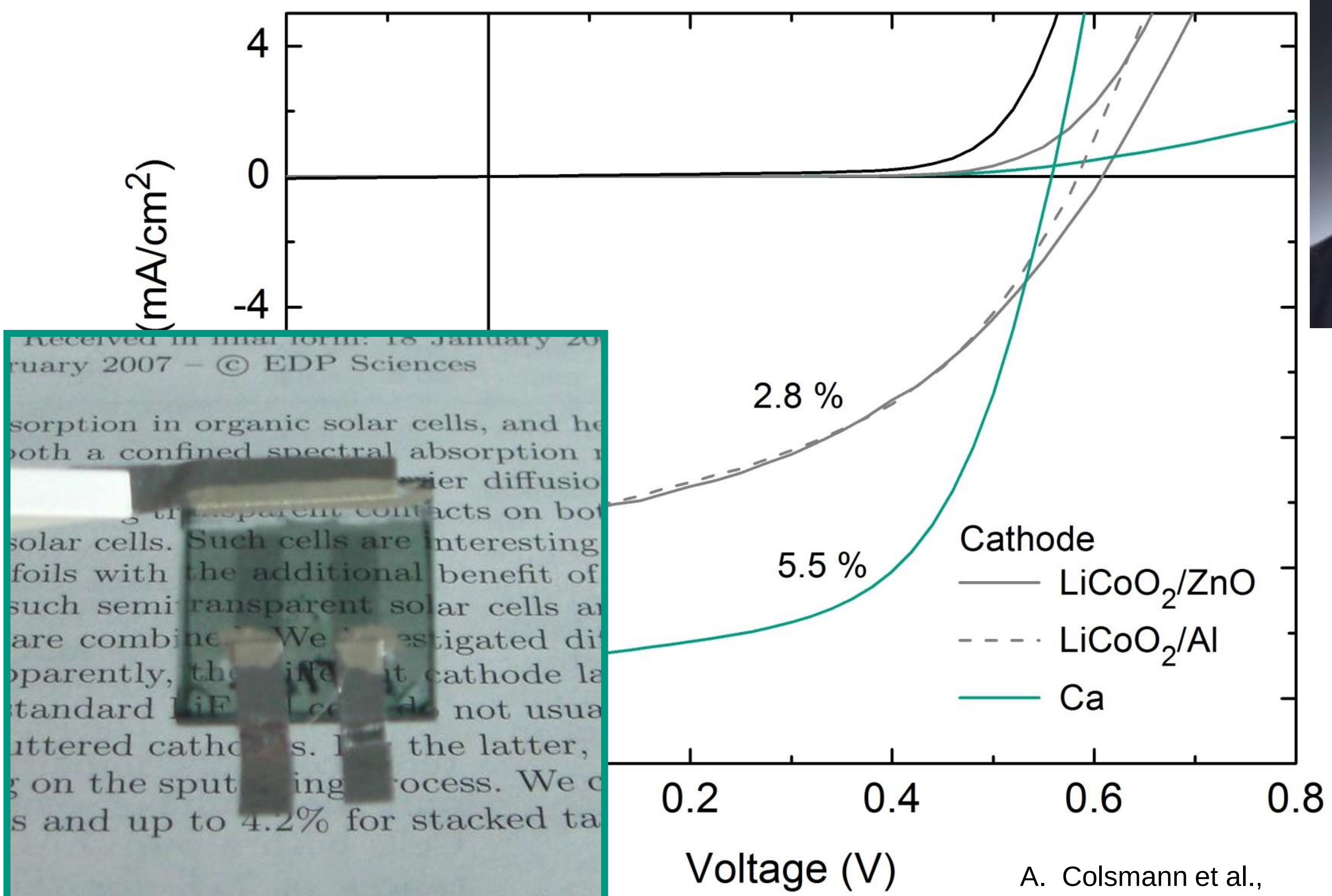


Heliatek world record cells with 12.0% efficiency on an active area of 1.1 cm².

Semitransparent solar cells

Source: Heliatek

Semi-transparent solar cells



A. Colsmann et al.,
Adv. Energy Mat. **1**, 599 (2011)

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Nanophotonics for organic solar cells ?

Some reasons why light management will not help that much:

-refractive index n of organic semiconductors is fairly low (1.6-1.9)
→reflection loss are low compared to inorganics

-absorption coefficient is very high ($10^5 cm^{-1}$)
→only thin films (100 nm) are needed

-organic solar cells are excitonic rather than photocarrier based
→quenching into plasmons has to be avoided

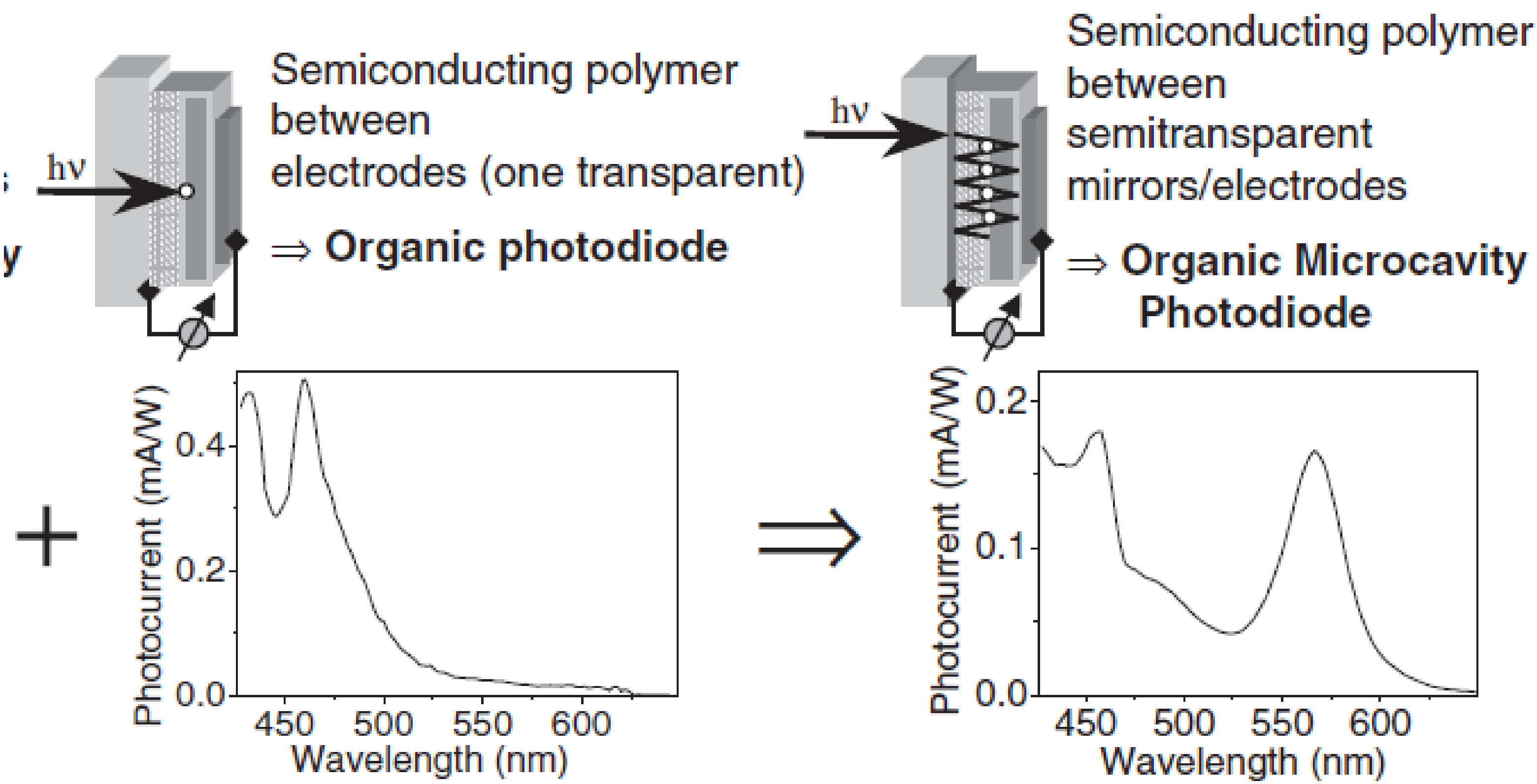
Some reasons why light management might be helpful:

-organic semiconductors can be coated into/onto any shape

-organic semiconductors have absorption tails

-some materials only work as very thin layers

Light trapping in organic photodiodes



R. Koeppe et al. Appl. Phys. Lett., 82, 2601 (2003)
J. Lupton et al., Adv. Mat. 15, 1471 (2003)

The role of plasmons in organic solar cells ?

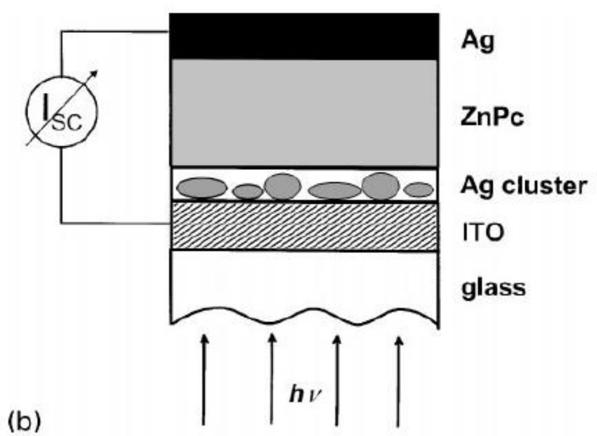
Solar Energy Materials & Solar Cells
ELSEVIER Solar Energy Materials & Solar Cells 61 (2000) 97–105
www.elsevier.com/locate/solmat

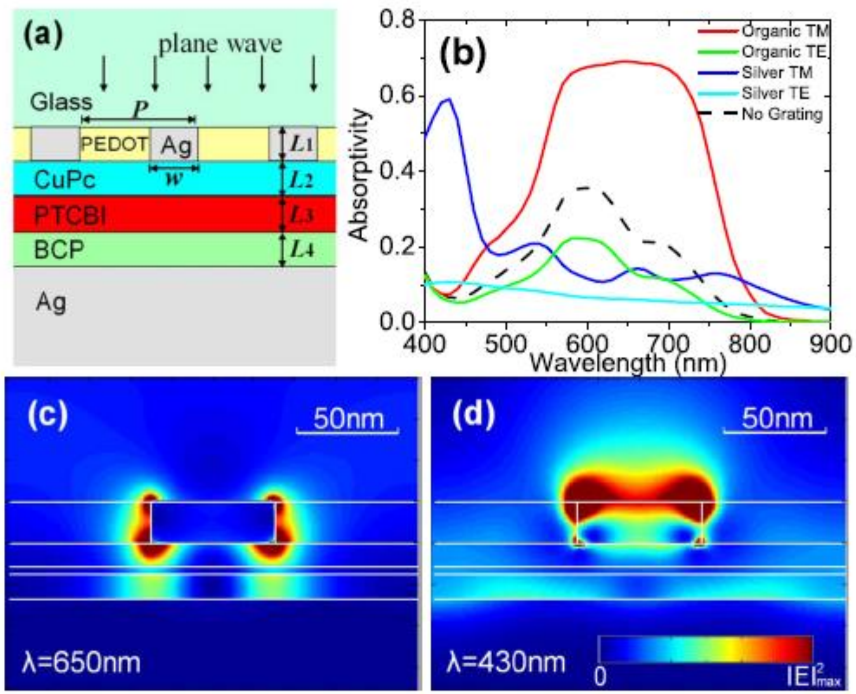
Metal cluster enhanced organic solar cells

M. Westphalen[a], U. Kreibig[a], J. Rostalski[b], H. Lüth[b], D. Meissner[c,*]

[a]Technical University Aachen, I. Physical Institute, 51056 Aachen, Germany
[b]Research Center Juelich, 52425 Juelich, Germany
[c]AQR, Forschungszentrum Juelich GmbH, Wendelinusstro.85, D-52428 Juelich, Germany

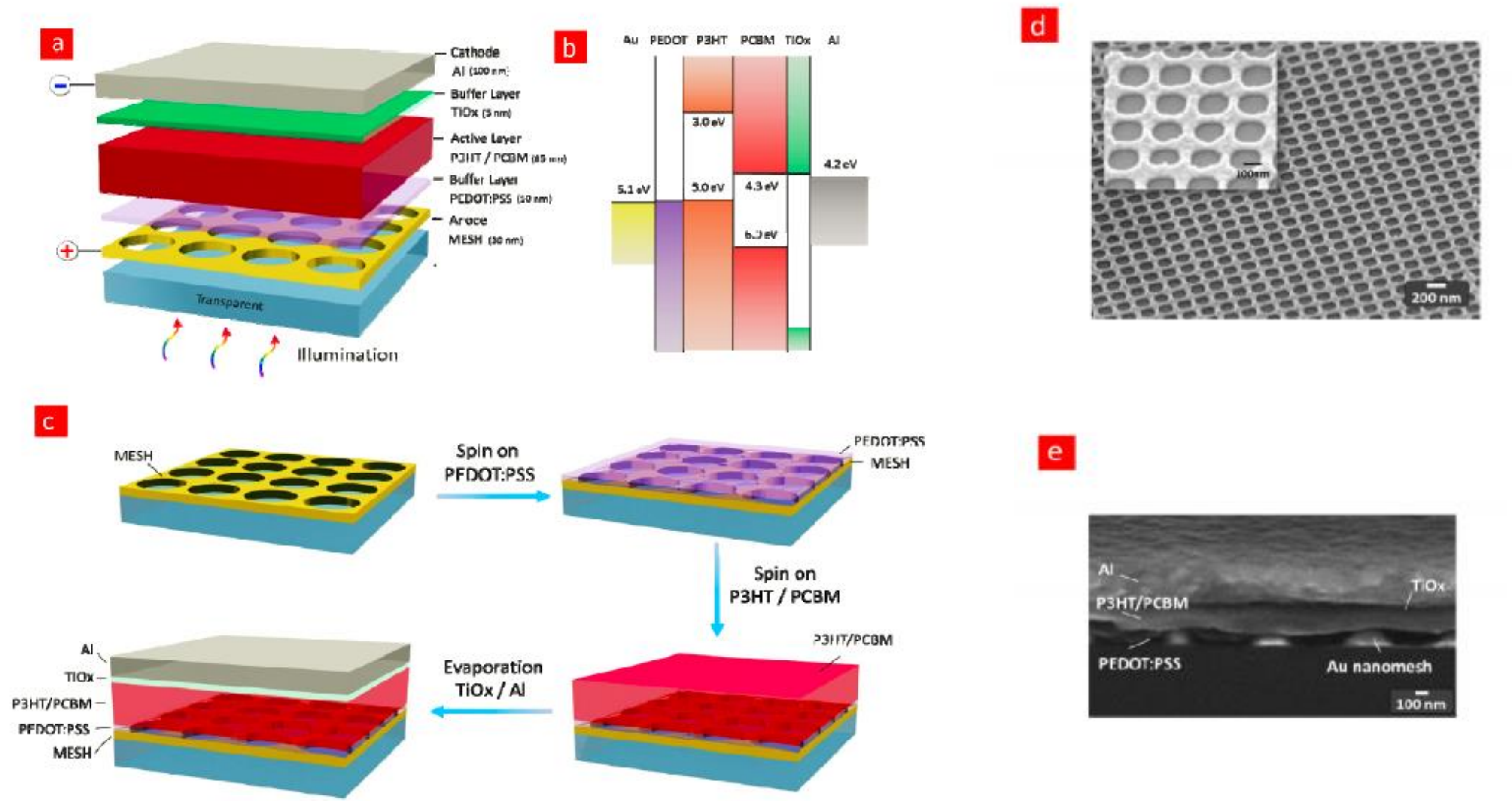
Accepted 25 May 1999





Min et al. Appl. Phys. Lett. 96, 133302 (2010)

Plasmonic Cavity with Subwavelength Hole-array (PlaCSH) Solar Cell (SC)



S. Chou et al., 14 January 2013 / Vol. 21, No. S1 / OPTICS EXPRESS A76

Outline

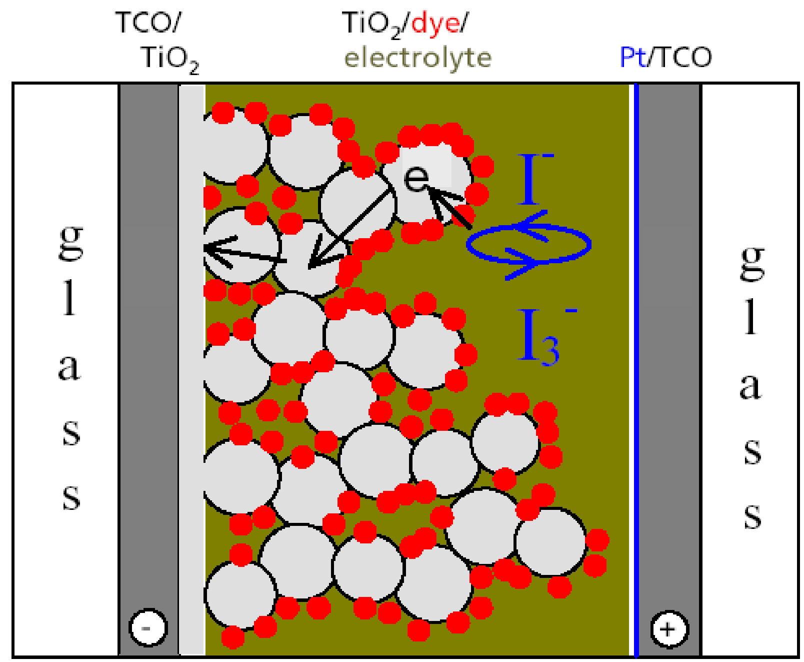
Part 1B: Dye-Sensitized Solar Cells

1. Liquid Electrolyte Cells
2. Solid state DSSCs
3. The road towards Perovskites

The dye sensitized solar cell (Grätzel-cell)

Layer sequence:

-TiO_2 nanoparticles form porous n-contact
-adsorbed monolayer of dye leads to absorption (ca. 1000-fold enhanced versus flat monolayer)
-p-contact via liquid electrolyte (I^-/I_3^-)



Reactions:

$S + \hbar\omega \rightarrow S^*$ (optical excitation)

$S^* \rightarrow S^+ + e^-_{CB}$ (electron transer, oxidation of dye)

$2S^+ + 3I^- \rightarrow 2S + I_3^-$ (reduction of dye back to neutral ground state)

Diffusion of I_3^- to counter electrode

$I_3^- + 2e^- \rightarrow 3I^-$ (reduction from triiodide to iodide)

A low-cost, high-efficiency solar cell based on dye-sensitized colloidal TiO_2 films

Brian O'Regan* & Michael Grätzel†

Institute of Physical Chemistry, Swiss Federal Institute of Technology, CH-1015 Lausanne, Switzerland

THE large-scale use of photovoltaic devices for electricity generation is prohibitively expensive at present: generation from existing commercial devices costs about ten times more than conventional methods[1]. Here we describe a photovoltaic cell, created from low- to medium-purity materials through low-cost processes, which exhibits a commercially realistic energy-conversion efficiency. The device is based on a 10-μm-thick, optically transparent film of titanium dioxide particles a few nanometres in size, coated with a monolayer of a charge-transfer dye to sensitize the film for light harvesting. Because of the high surface area of the semiconductor film and the ideal spectral characteristics of the dye, the device harvests a high proportion of the incident solar energy flux (46%) and shows exceptionally high efficiencies for the conversion of incident photons to electrical current (more than 80%). The overall light-to-electric energy conversion yield is 7.1–7.9% in simulated solar light and 12% in diffuse daylight. The large current densities (greater than 12 mA cm^{-2}) and exceptional stability (sustaining at least five million turnovers without decomposition), as well as the low cost, make practical applications feasible.

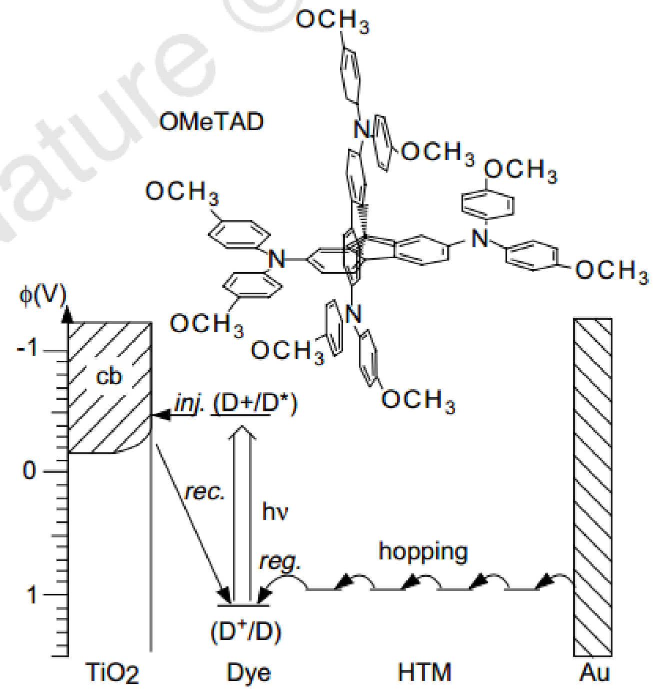
NATURE · VOL 353 · 24 OCTOBER 1991

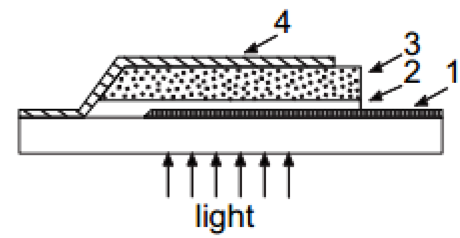
Solid-state dye-sensitized mesoporous TiO_2 solar cells with high photon-to-electron conversion efficiencies

U. Bach*, D. Lupo†‡, P. Comte*, J. E. Moser*, F. Weissörtel§, J. Salbeck§, H. Spreitzer† & M. Grätzel*

* *Institute of Photonics and Interfaces, Swiss Federal Institute of Technology, CH-1015 Lausanne, Switzerland*
† *Hoechst Research & Technology Deutschland GmbH & Co. KG, Industriepark Höchst, D-65926 Frankfurt, Germany*
§ *Max-Planck-Institut für Polymerforschung, D-55128 Mainz, Germany*

Solar cells based on dye-sensitized mesoporous films of TiO_2 are low-cost alternatives to conventional solid-state devices[1]. Impressive solar-to-electrical energy conversion efficiencies have been achieved with such films when used in conjunction with liquid electrolytes[2]. Practical advantages may be gained by the replacement of the liquid electrolyte with a solid charge-transport material. Inorganic p-type semiconductors[3,4] and organic materials[5–9] have been tested in this regard, but in all cases the incident monochromatic photon-to-electron conversion efficiency remained low. Here we describe a dye-sensitized heterojunction of TiO_2 with the amorphous organic hole-transport material 2,2′,7,7′-tetrakis(*N,N*-di-*p*-methoxyphenyl-amine)9,9′-spirobifluorene (OMeTAD; refs. 10 and 11). Photoinduced charge-carrier generation at the heterojunction is very efficient. A solar cell based on OMeTAD converts photons to electric current with a high yield of 33%.





Some DSC facts from Hagfeldt

See Chem. Rev. 2010, 110, 6595–6663

Outdoor performance - production cost per kWh an advantage for DSC:
a 10 % PCE rated DSSC module produces over one year the same amount of electricity as 14-15 % rated Si module (Sony).

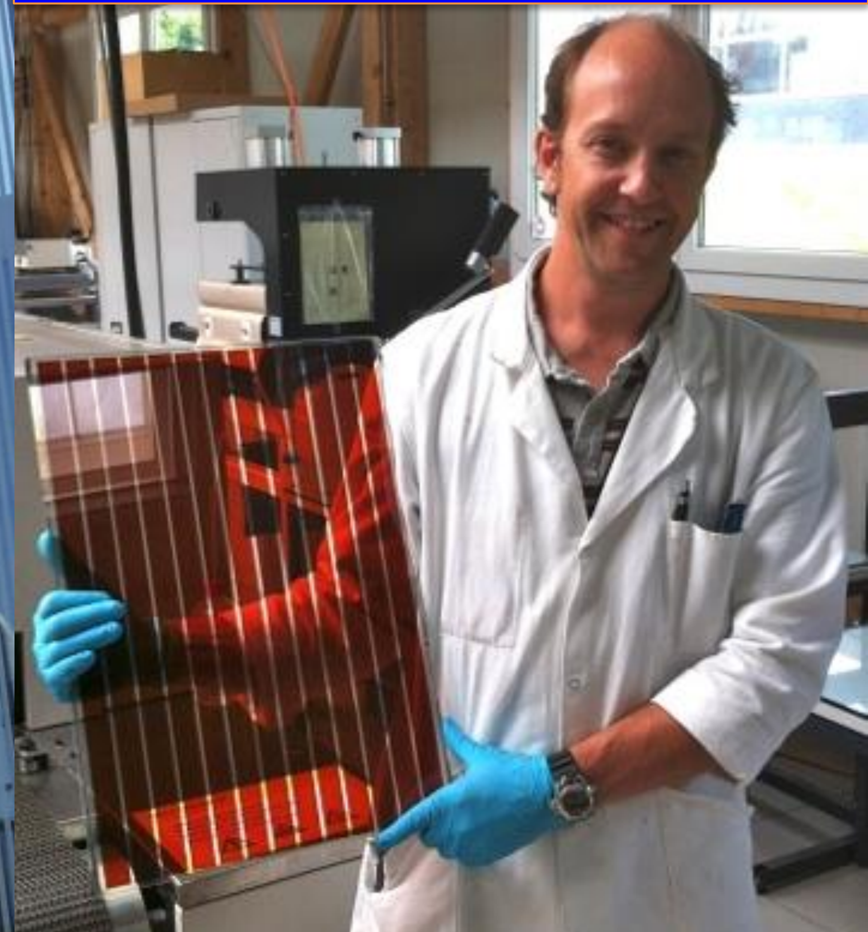
Electricity from ambient and indoor light:
DSC outperforms all competitors

stability
> 20 years outdoors accelerated testing (Dyesol, Fujikura …)

energy pay back time:
< 1 year (3GSolar and ECN life cycle analysis

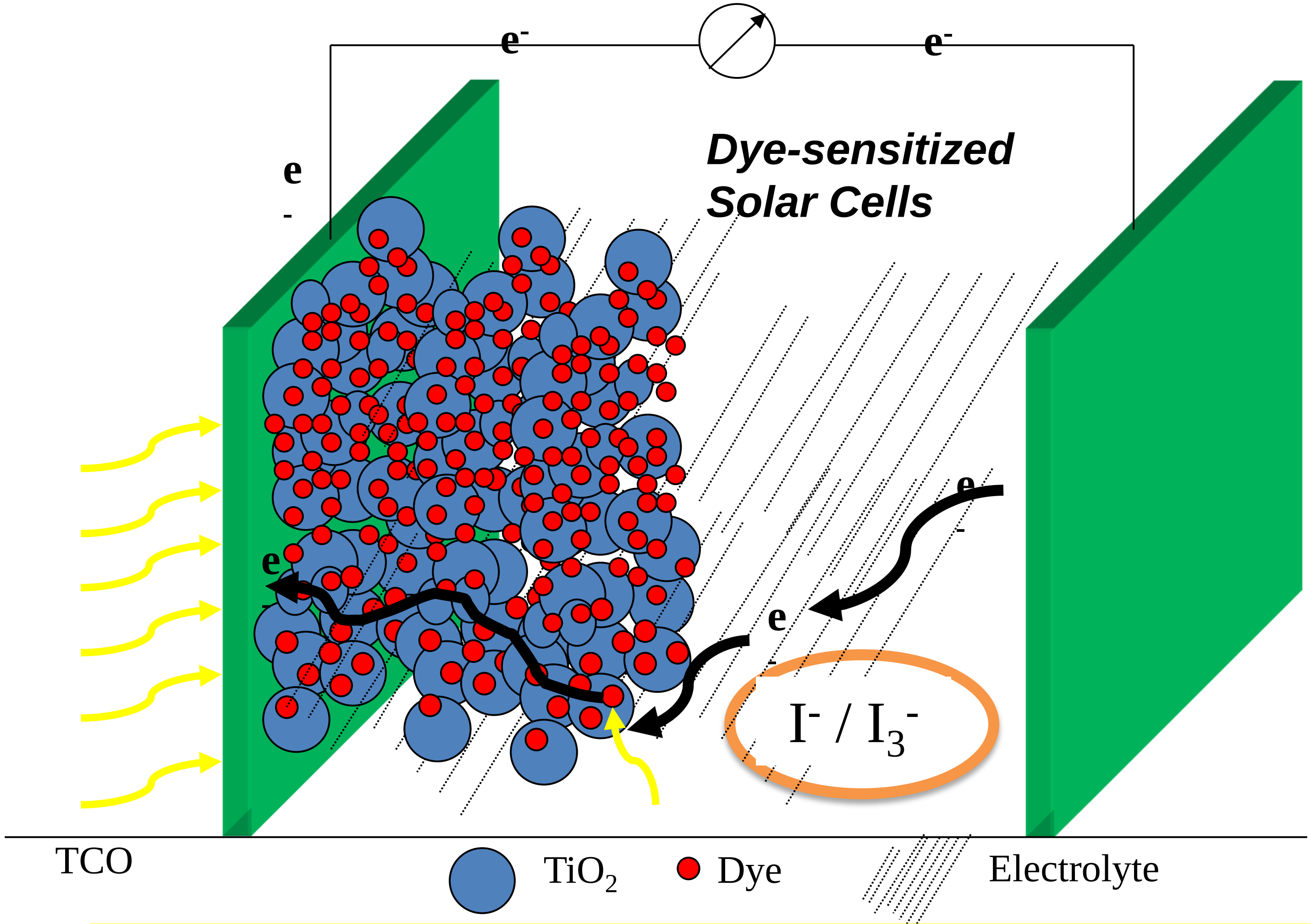

HANA AKARI
FLOWER LAMP
(SONY)
Design: Colours and Transparency
Product Integration

Façade for the new congress hall at EPFL, Lausanne
Building Integration



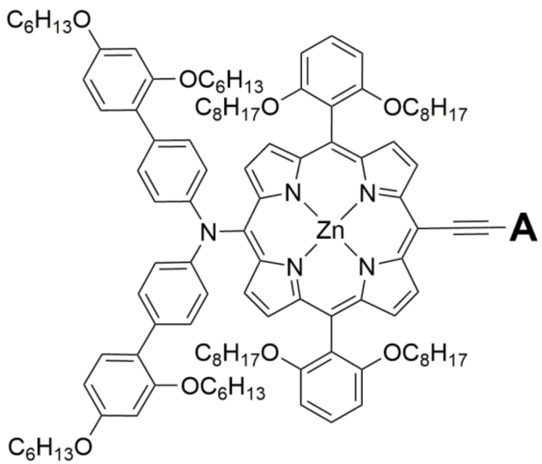
SOLARONIX
« efficiently innovative »

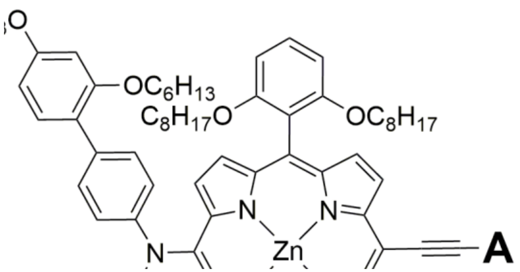
Where are the internal losses?
- the hunt for the half volt

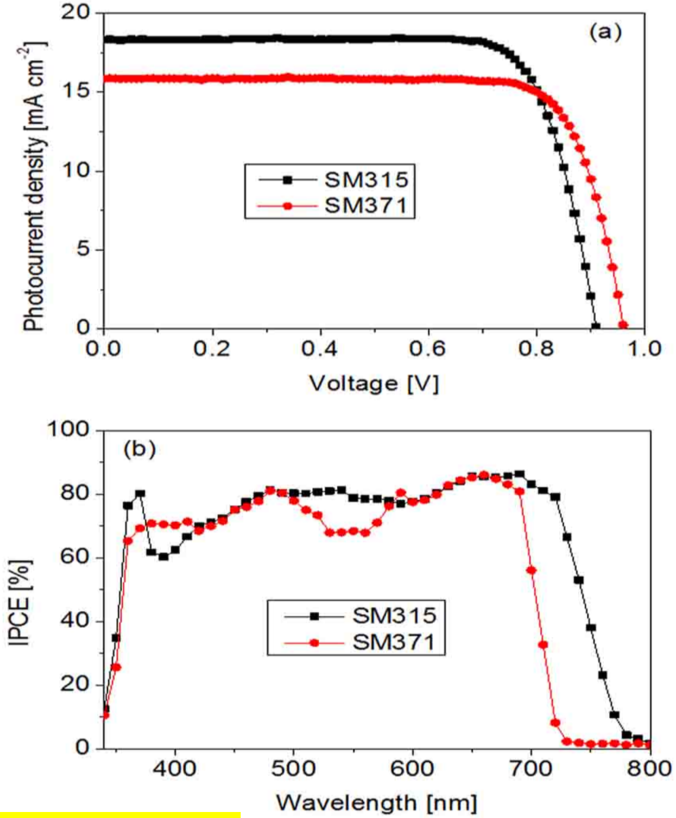


Can a 2-electron redox couple be replaced by a 1-electron couple?
A problem for almost 20 years

The World Record DSC is Based on Porphyrine Dye and Co-complex Redox Electrolyte

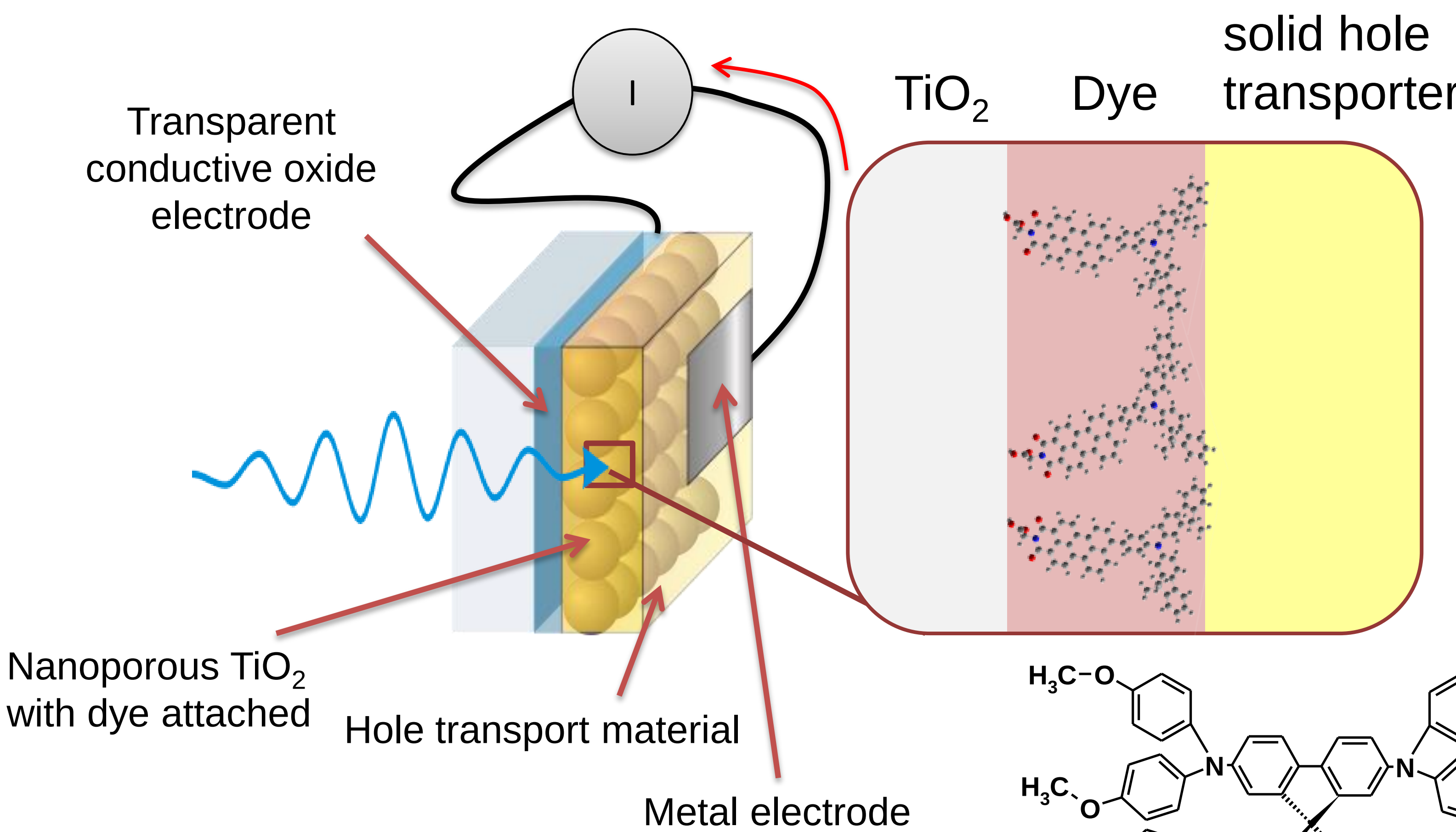






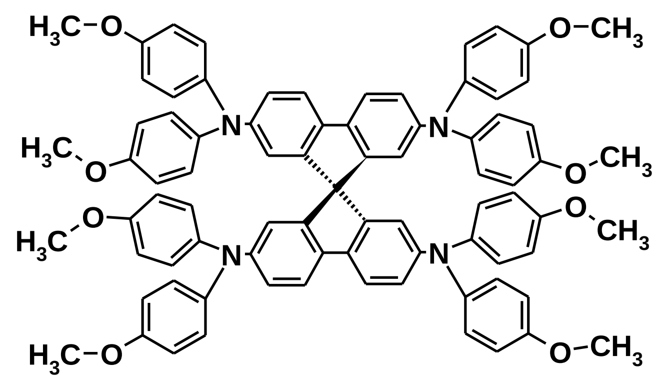
Grätzel and co-workers: The SM315 porphyrin reaches a record efficiency of 13% :

Solid State DSSC Efficiency is Still Lower



η ~ 7.2 % for solid state

η ~ 12.3 % for liquid electrolyte

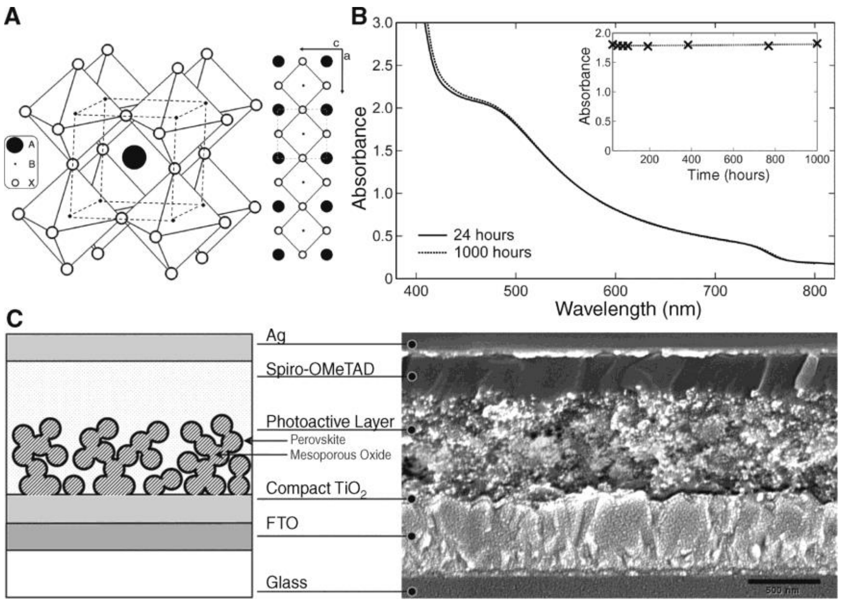


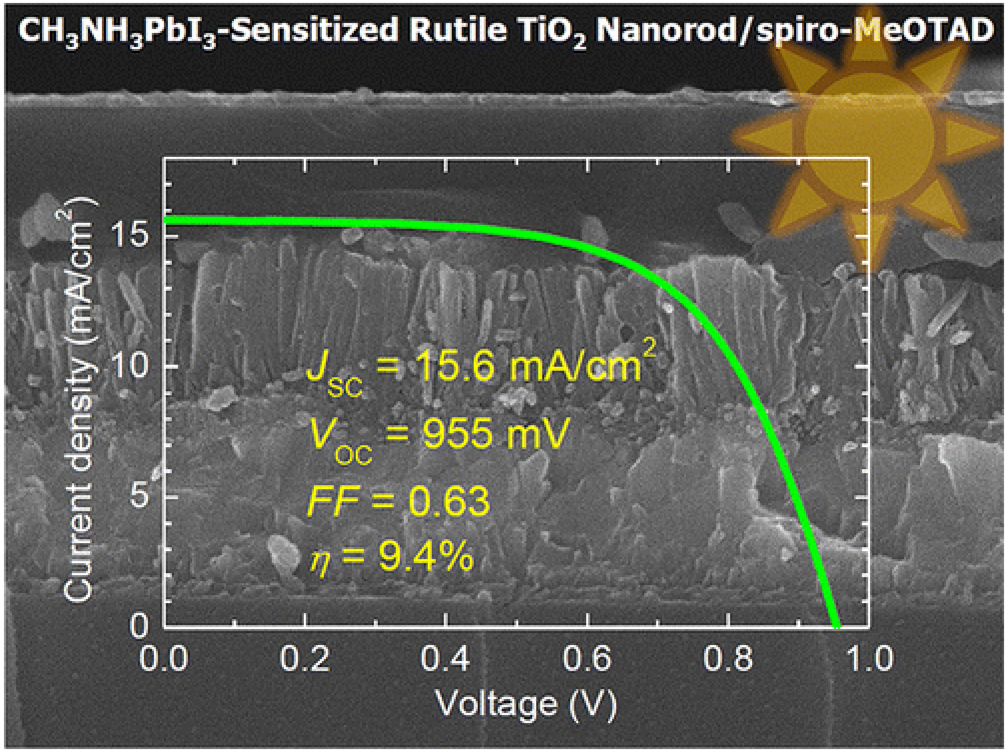
Hole transporter Spiro-MeOTAD

M. Meister, I.A. Howard, B. Baumeier, H. Wonneberger, N. Pschirer, R. Sens, I. Bruder, C. Li, K. Müllen, D. Andrienko and F. Laquai, *Advanced Energy Materials* **2013**, DOI: 10.1002/aenm.201300640.

Unless you count perovskites

10.2% efficiency





Science 2 November 2012:
Vol. 338 no. 6107 pp. 643-647
DOI: 10.1126/science.1228604

Nano Lett., **2013**, *13* (6), pp 2412–2417
DOI: 10.1021/nl400286w