

# Small Molecule Organic Solar Cells – Status and Perspectives

R. Schueppel, K. Schulze, C. Urich, D. Wynands,  
B. Männig\*, M. Pfeiffer\*, M.K. Riede, K. Leo

*Institut für Angewandte Photophysik,  
Technische Universität Dresden, 01062 Dresden, Germany, [www.iapp.de](http://www.iapp.de)*

E. Brier, E. Reinold, P. Bäuerle

*Institut für Organische Chemie II und Neue Materialien, Universität Ulm*

*\* Heliatek GmbH, Dresden*

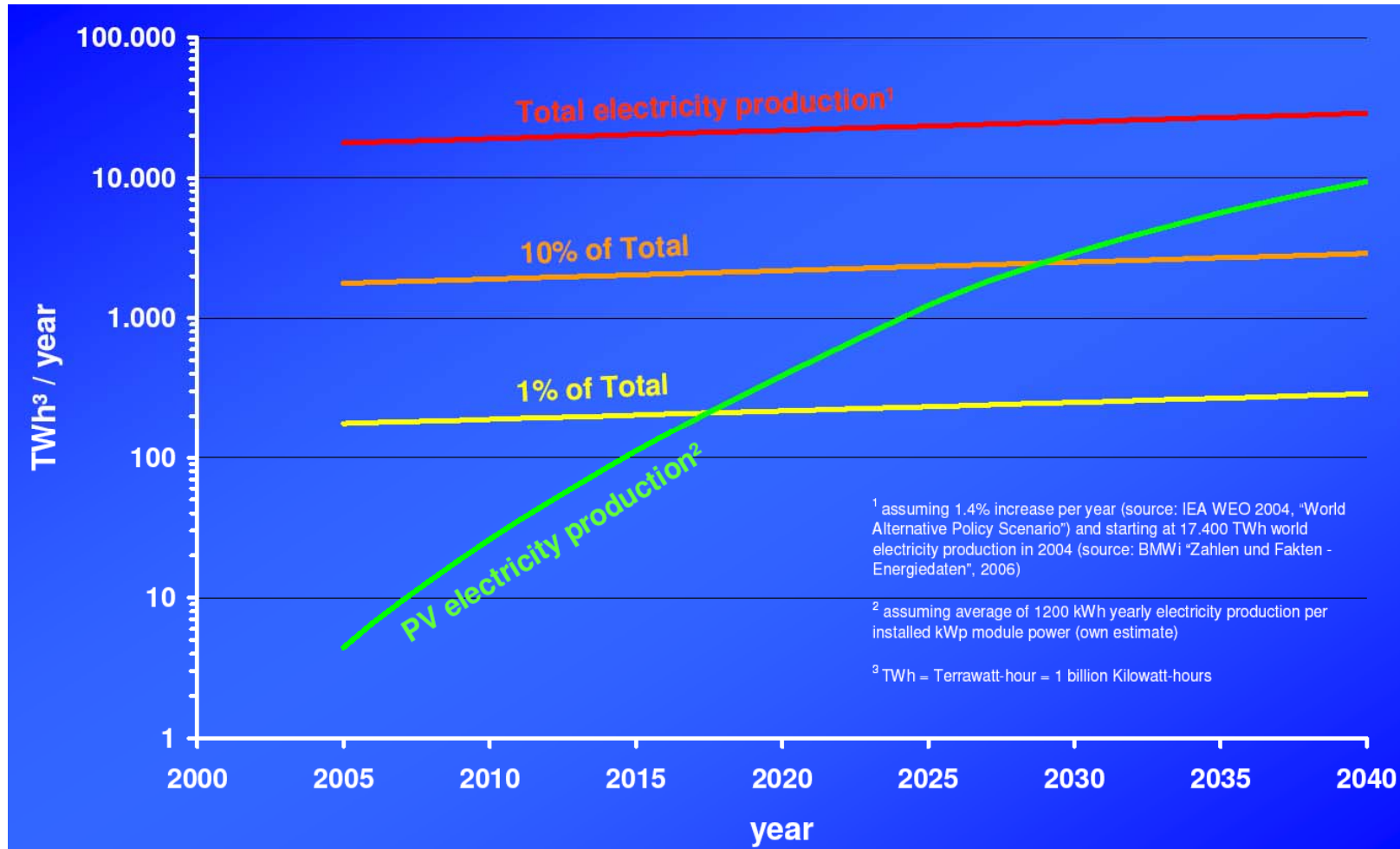
Linz, February 7, 2008



# Outline

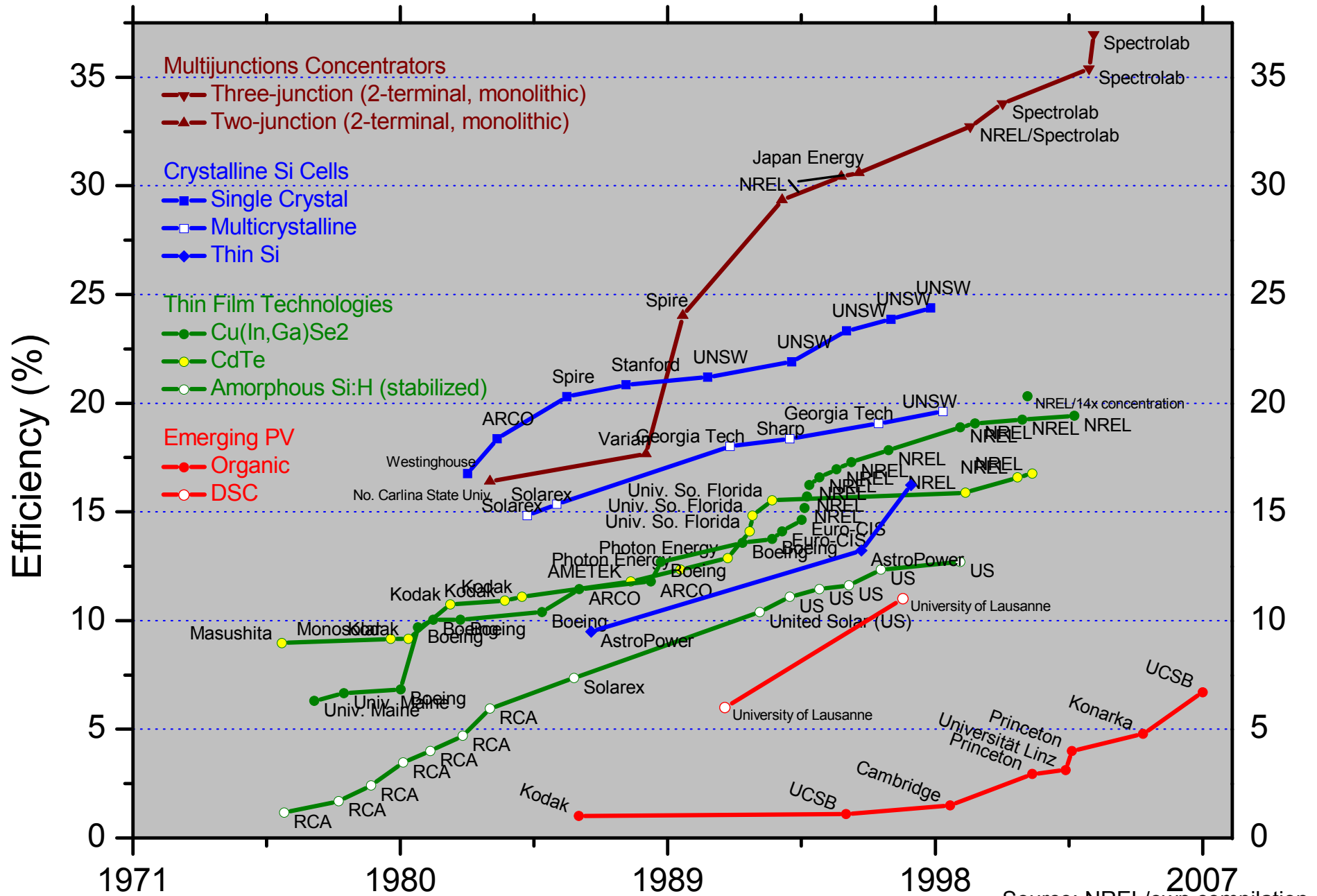
- Some thoughts about Organic PV in general
- Status of small-molecule organic solar cells:  
key innovations in the past
- Future challenges:
  - understanding and increasing the voltage
  - covering the entire solar spectrum: IR & tandem cells
  - stability
- Low-cost manufacturing

# Photovoltaics: ... just taking off



PV „power generation“ market will grow by another factor of 5000!

# Status: Efficiencies

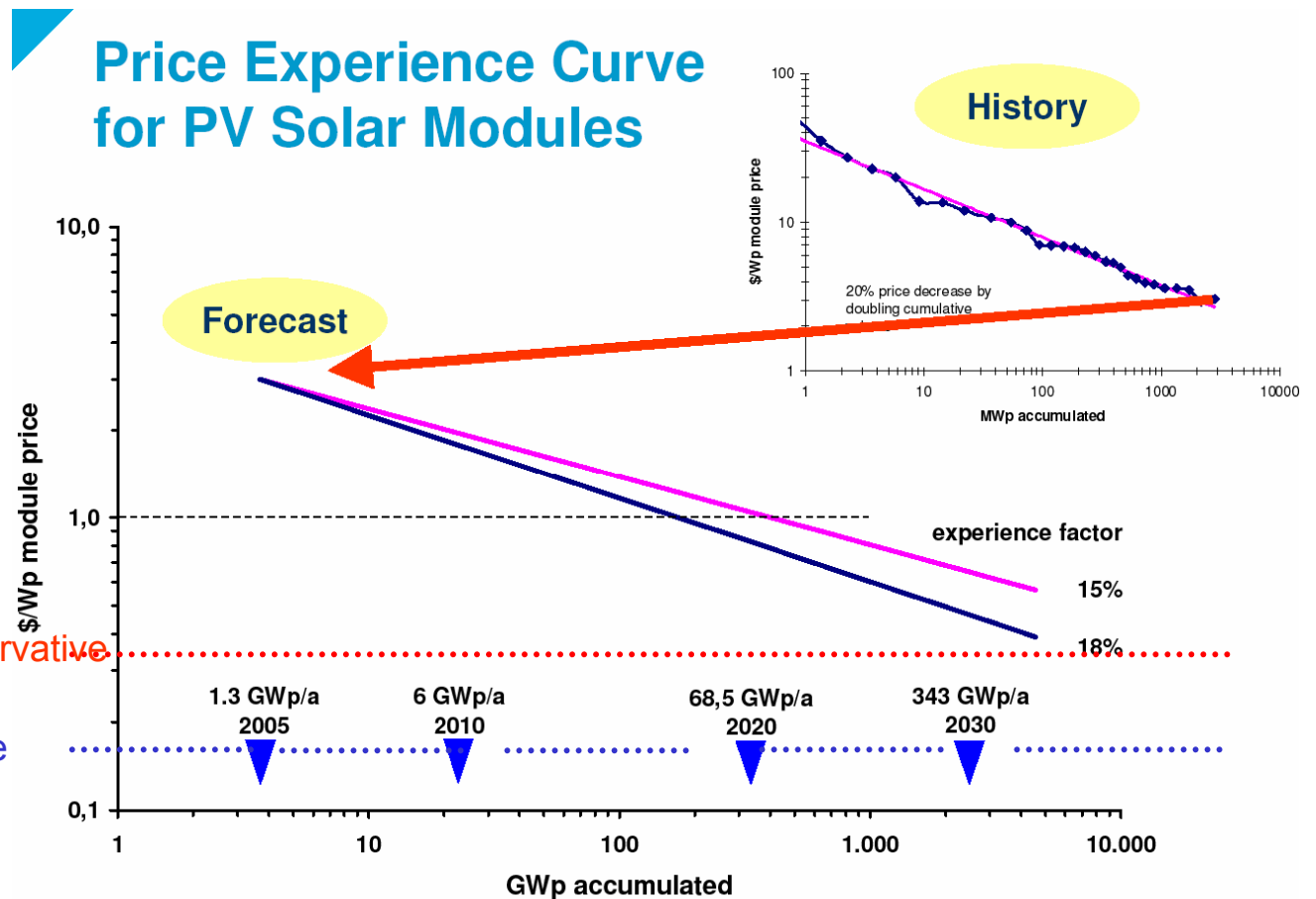


Source: NREL/own compilation

# Cost of organic solar cells

- Factor of 2-3 below Si/thin film seems possible
- What is needed: 8-10% in module = 13-15% in lab
- Lifetime at least 10 years

## Price Experience Curve for PV Solar Modules



OPV conservative

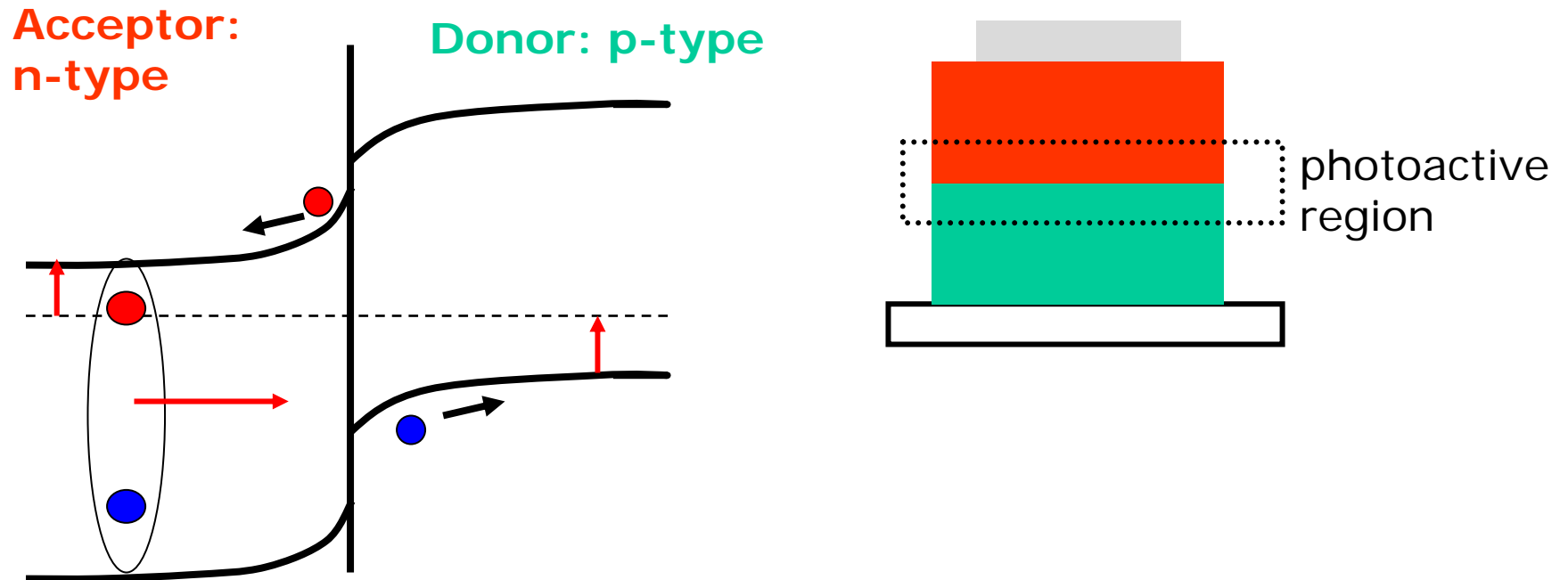
aggressive

ref: European Photovoltaic Industries Association (EPIA) and W. Hoffmann personal estimates

# Status of small molecule organic solar cells

- Started in 1986 with Ching Tang's work at Kodak
- Looked for Solar Cell and also stumbled into OLED
- Big progress, but still immature compared to OLED
- Many contributions I cannot cover: Yase, Saito, Fostiropoulos, Schlettwein, Lemmer, Kowalsky, ....

# Small molecule organic solar cells

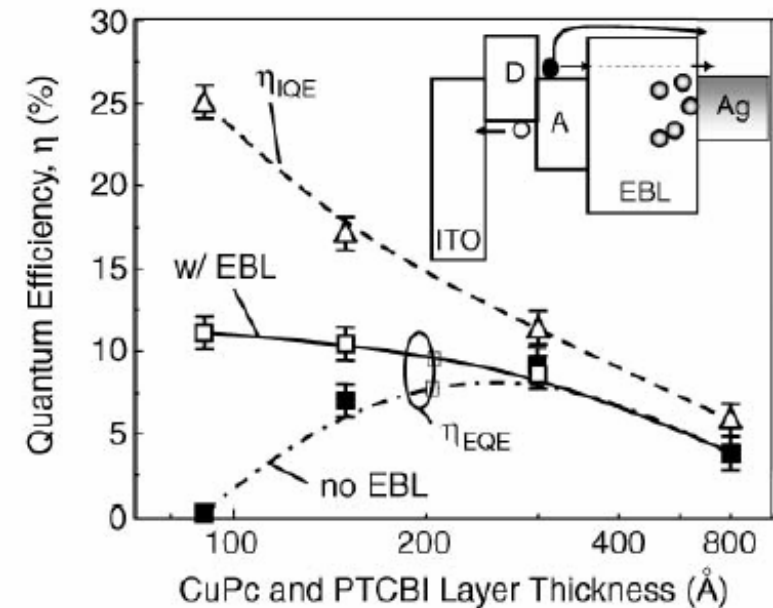


*C.W. Tang (Kodak)  
(1986)*

- Materials: CuPc/PTCBI
- efficiency 1%
- exciton diffusion length too low  
=> active layer very thin

# Exciton blocker layer

- P. Peumans, V. Bulovic, and S.R. Forrest, Appl. Phys. Lett. 76 (2000) 2650
- Excitons are reflected
- Optical standing wave is optimized





# pin-Structure

- M. Hiramoto et al., J. Appl. Phys. 72 (1992) 3781
- Pin-structure, but no active Fermi level control in p- and n-layer
- Bulk heterojunction!

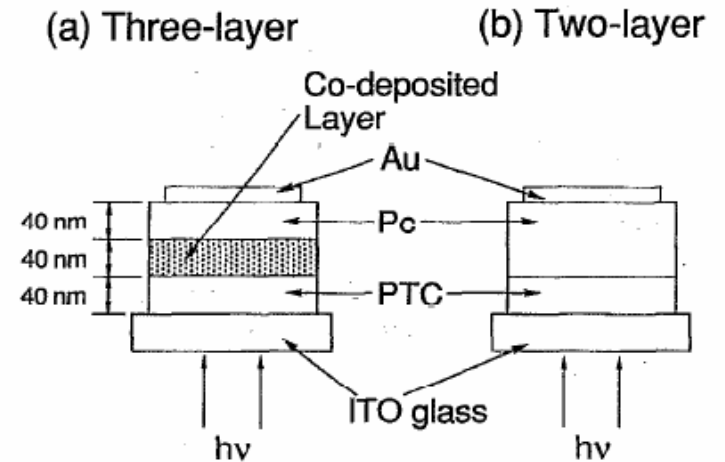
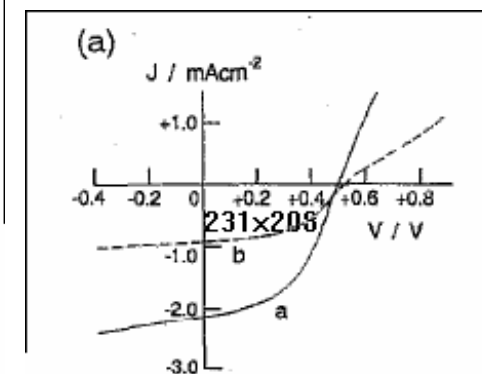
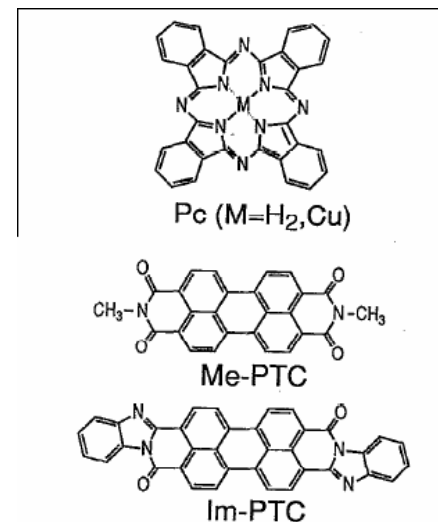


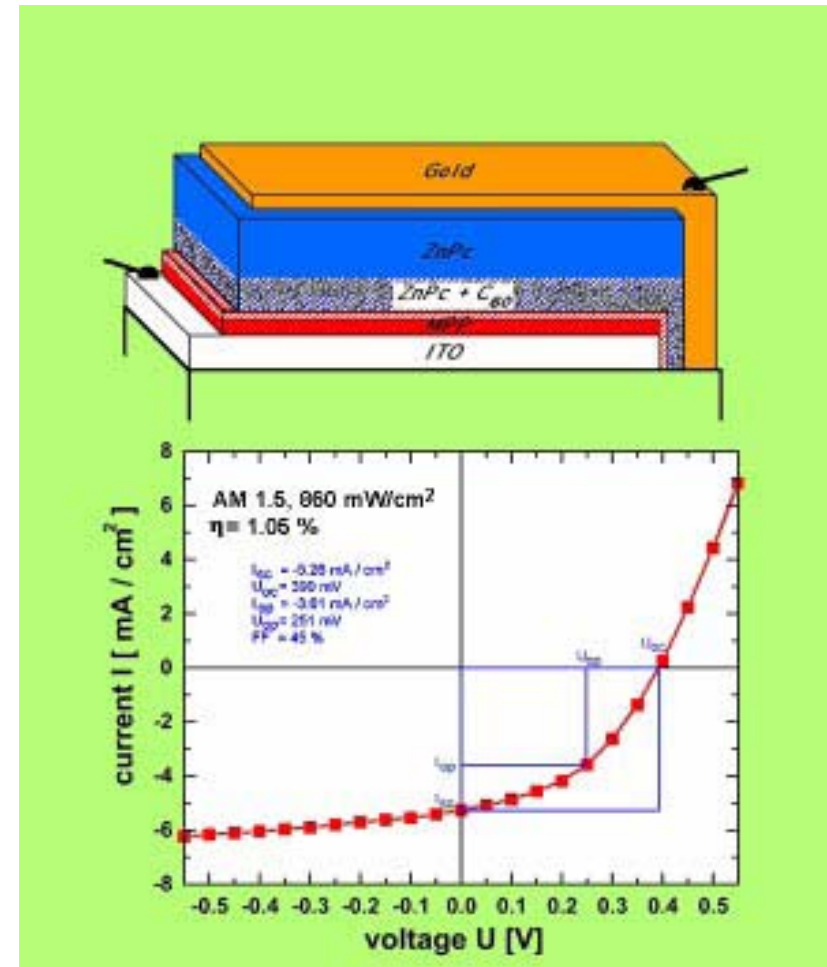
FIG. 2. Schematic representation of the configurations of (a) three-layered and (b) two-layered organic solar cells.

3782 J. Appl. Phys., Vol. 72, No. 8, 15 October 1992

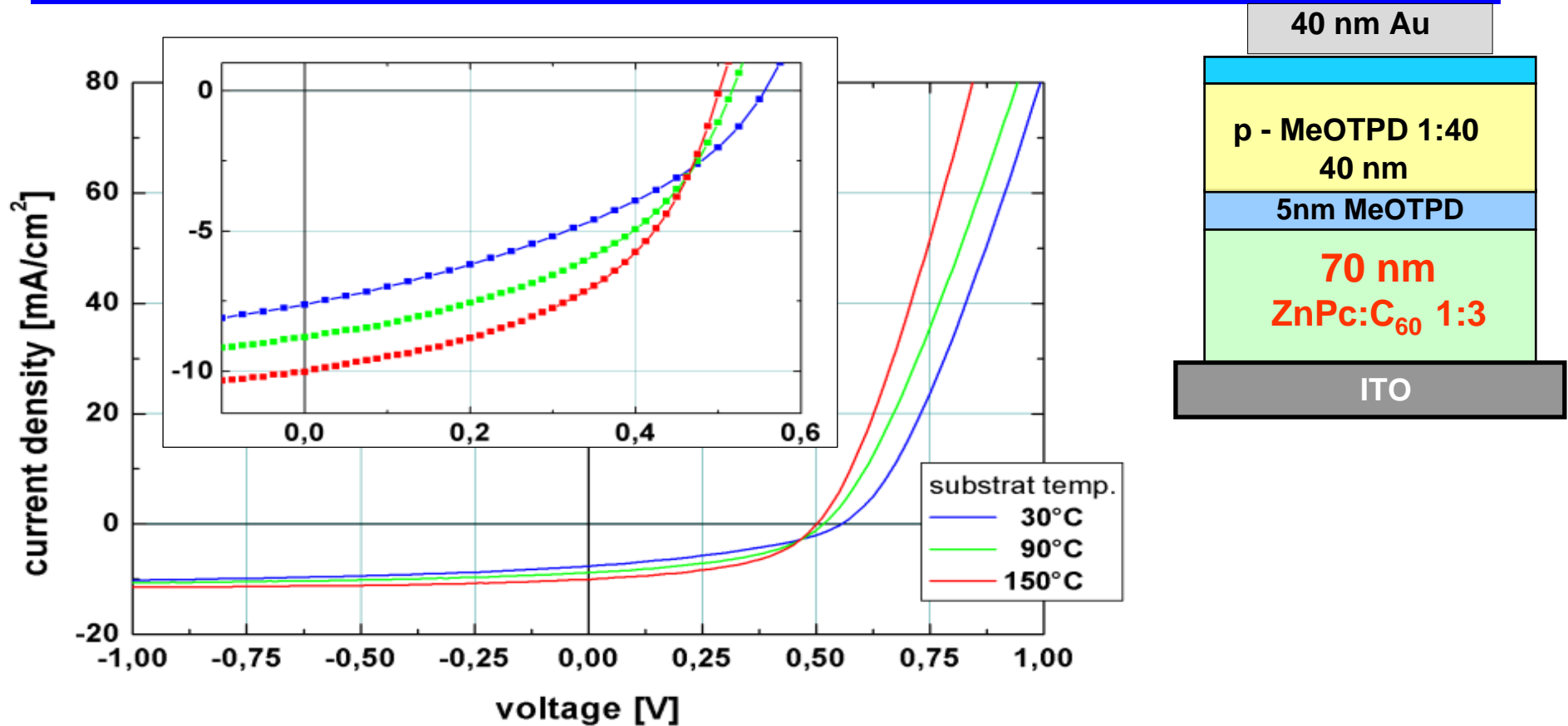


# Bulk heterojunction with C<sub>60</sub>

- D. Meissner et al.  
Photon **2** (1999), 34 – 37
- Prepared by coevaporation
- Problem: morphology control



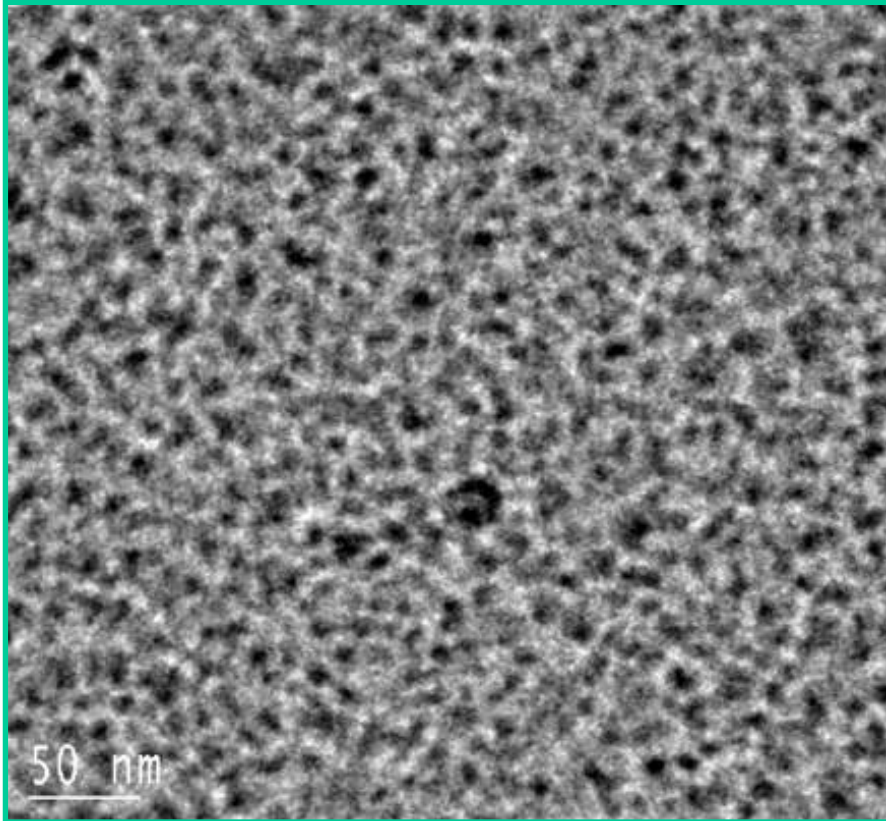
# Influence of blend layer morphology



temp	I <sub>sc</sub> [mA]	U <sub>oc</sub> [V]	FF [%]	eff [%]	reverse slope [mA/cm²V]
30°C	7,63	0,56	37,6	1,61	2,4
90°C	8,79	0,52	44,9	2,05	1,7
150°C	10,03	0,50	49,0	2,44	1,3

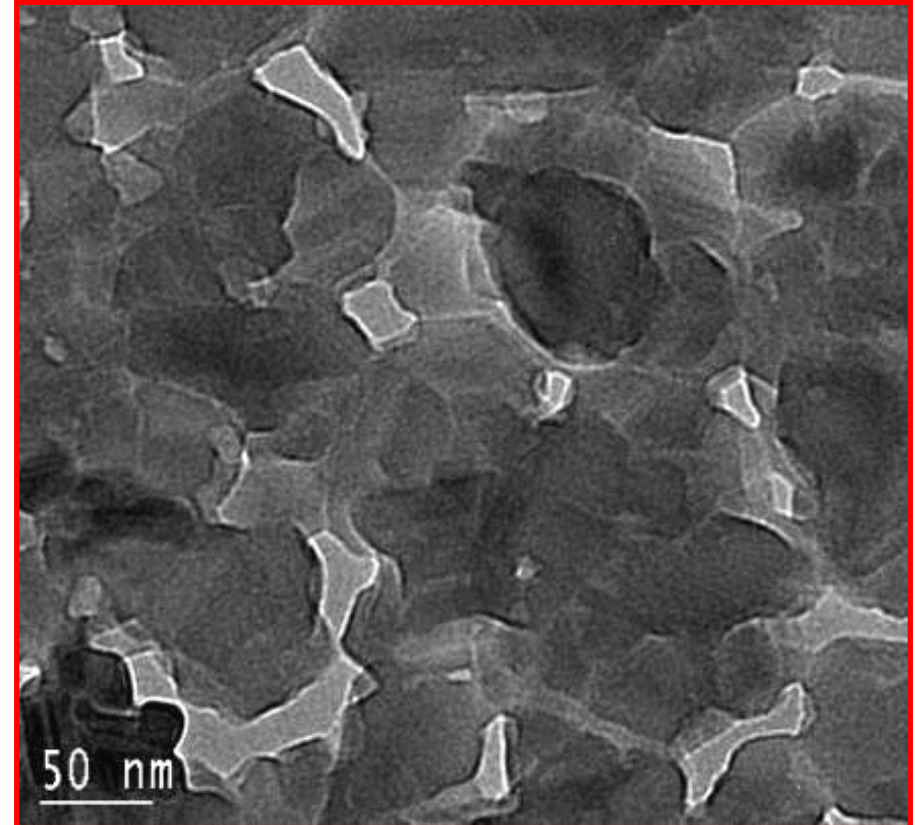
Substrate temperature  $\leftrightarrow$  blend layer morphology  
REM pictures for ZnPc\* $C_{60}$

30°C



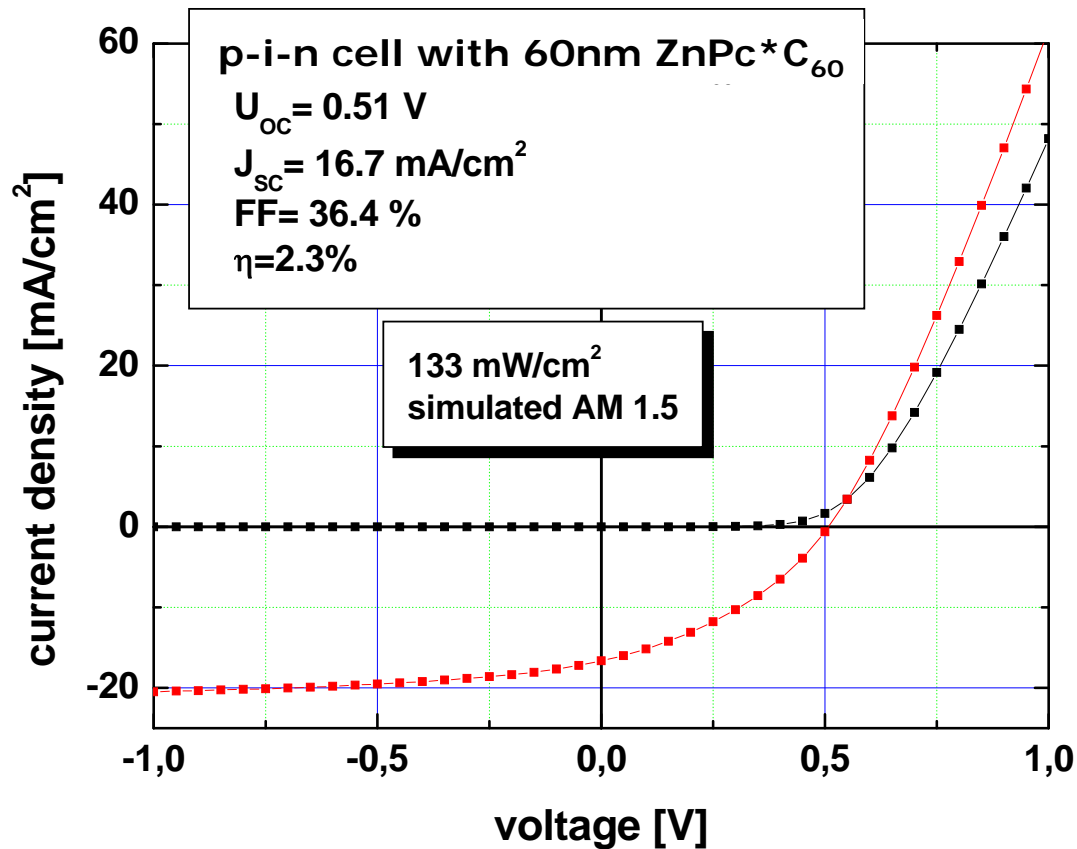
- no domains
- amorphous structure

150°C



- domains of 20-100nm
- 120° angles => hexagonal structure of  $C_{60}$  nanocrystallites

# ZnPc-C<sub>60</sub>-BH cells: the benchmark



- Current status:  $\eta \approx 3.5\%$  (Hiramoto, Forrest, IAPP)
- Clearly behind polymer solar cells
- Limits: insufficient absorption; low mobility in blend; high voltage loss

Unpublished data (Hiramoto): 1  $\mu$ m absorber!,  $\eta \approx 5\%$  with ultrapure C<sub>60</sub>

# Tandem cells

- M. Hiramoto et al., Chem. Lett. **1990** (1990) 327
- First efficient realization:  
A. Yakimov & S.R. Forrest, Appl. Phys. Lett. **80** (2002) 1667
- Au Clusters recombination centers

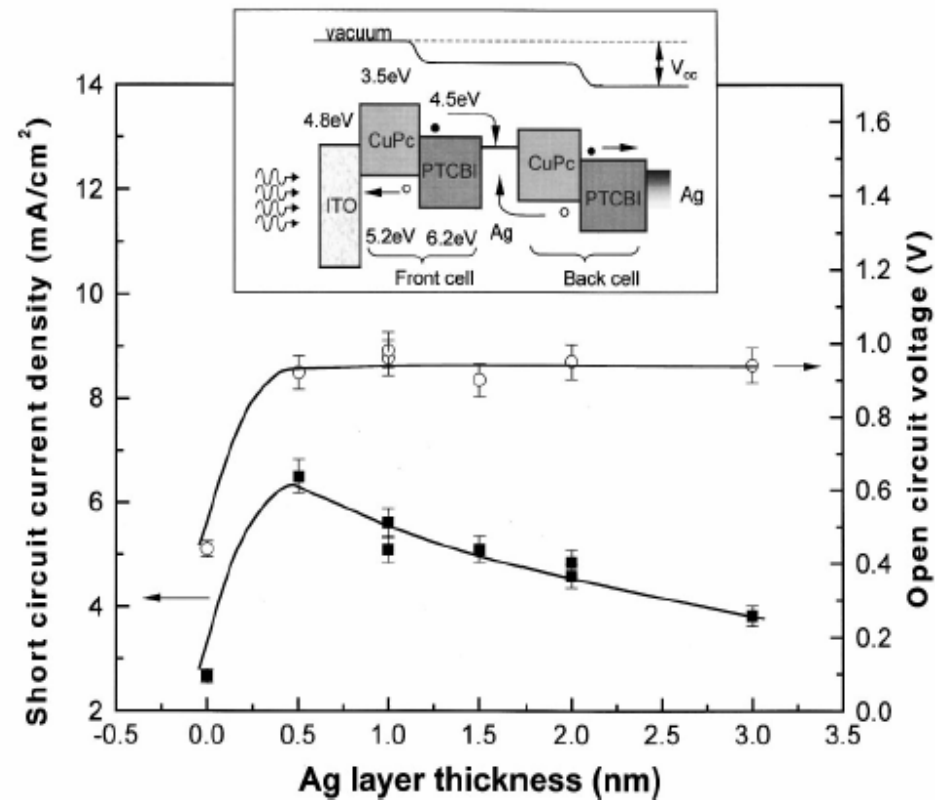
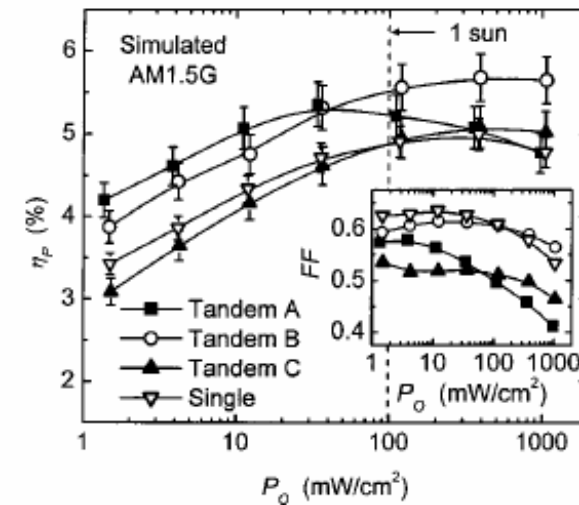
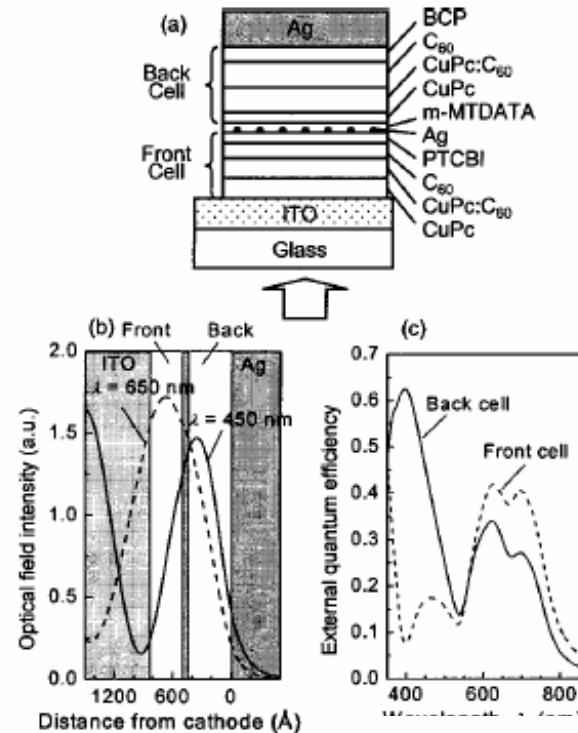


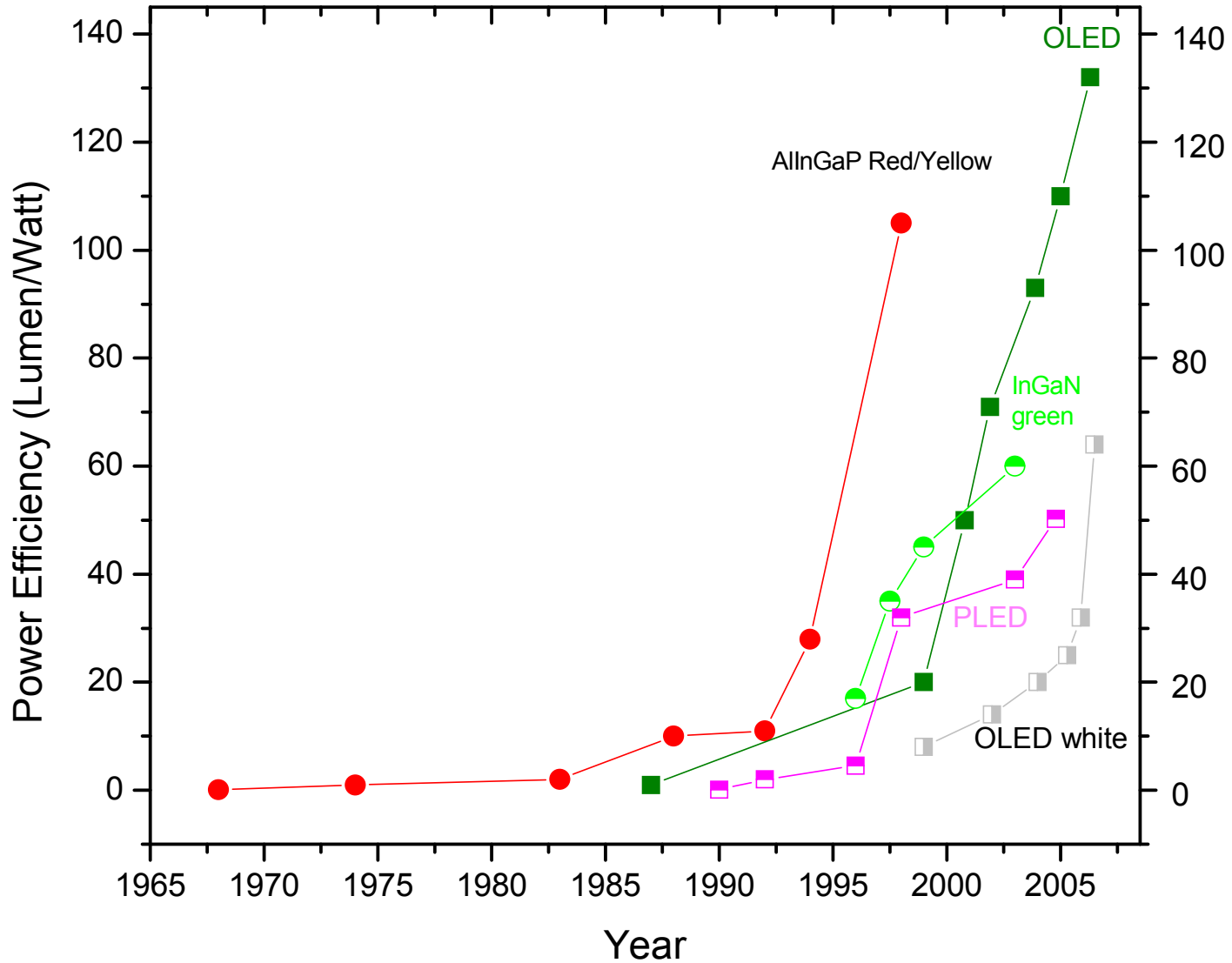
FIG. 1. Short circuit current density (closed squares, left axis) and open circuit voltage (open circles, right axis) for dual cells having Ag interlayers of different average thicknesses. The measurements were performed under AM 1.5, 100 mW/cm<sup>2</sup> (1 sun) illumination. The inset shows the proposed energy level diagram of the dual-HJ device.

# Best literature value for small-molecule cell

- J. Xue et al. Appl. Phys. Lett. 85 (2004) 5757
- CuPc/C60:CuPc/PTCDI tandem cell
- Au Clusters and p-doped interlayer
- 5.7% at 1 sun



# OLED: Polymer is behind small molecule



← Fluorescent Lamp

← Tungsten Bulb (unfiltered)

← Red Filtered





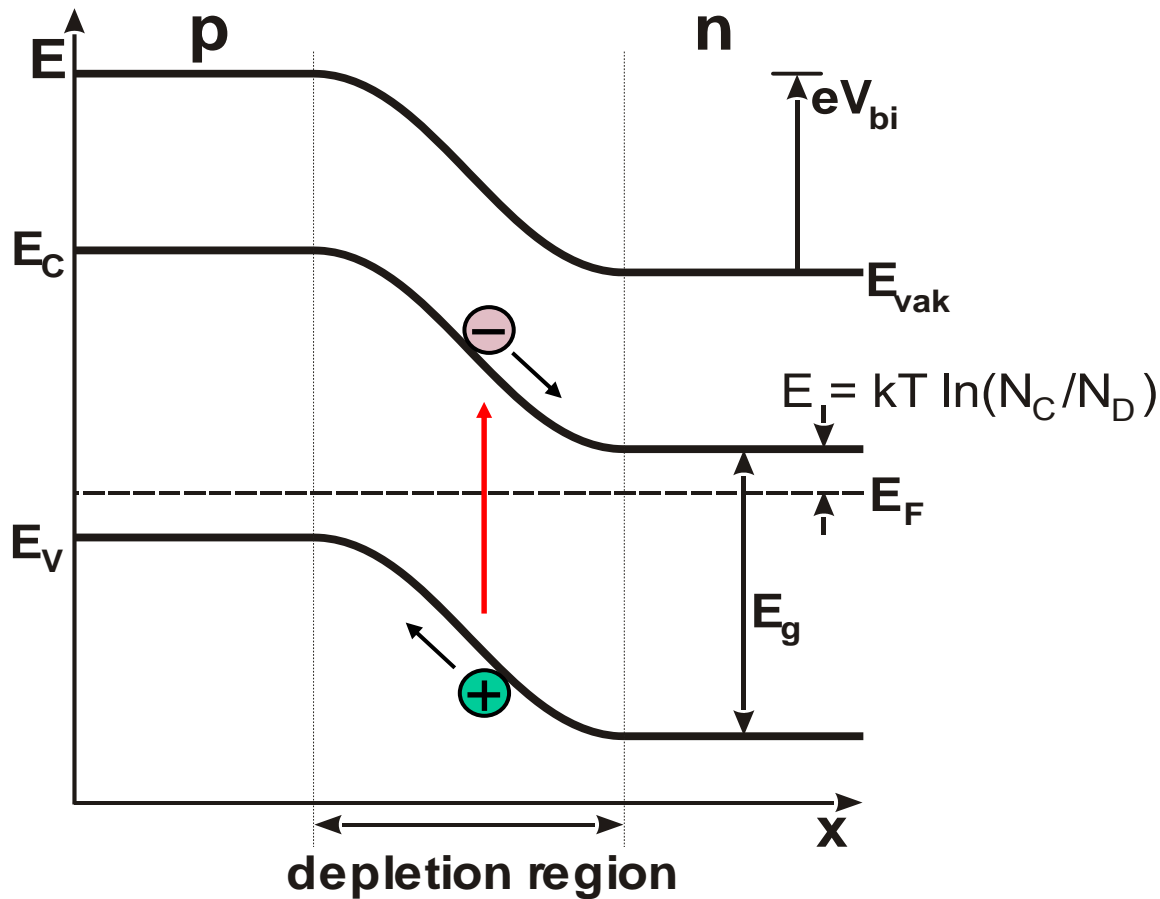
# Why are polymer solar cells better than small-molecule solar cells?

- Bulk heterojunction morphology control: many more “handles” for polymers:
  - solvent
  - temperature
  - concentration
- Polymer might have a basic mobility advantage in bulk heterojunction
- Materials basis broader

# Outline

- Some thoughts about Organic PV in general
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key innovations in the past
- **Future challenges:**
  - **understanding and increasing the voltage**
  - covering the entire solar spectrum: tandem cells
  - stability
- Low-cost manufacturing

# Origin of the open-circuit voltage



- Inorganic solar cell: Built-in voltage by Fermi-level difference of doped transport layers

$$eV_{bi} = E_g - kT \ln [N_C \cdot N_V / N_D \cdot N_A]$$

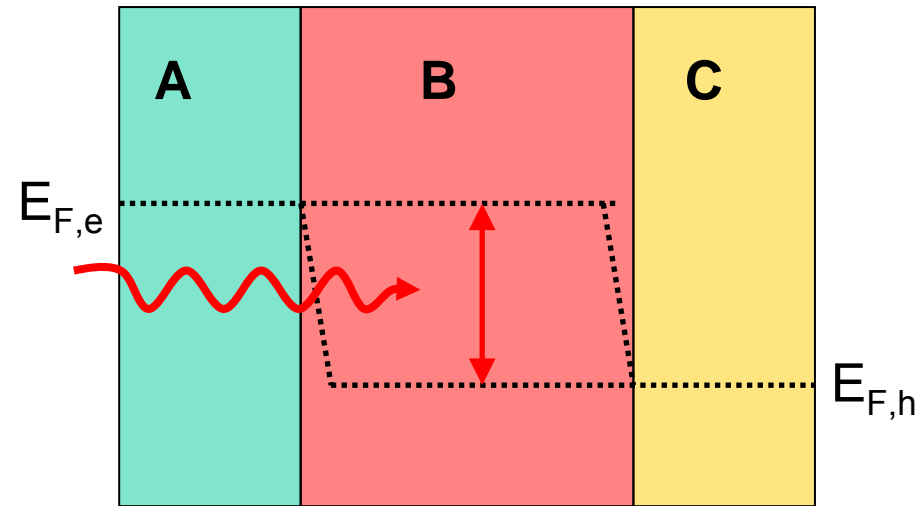
- Organic solar cells: built-in voltage by contacts?

**Open-circuit voltage cannot exceed  $V_{bi}$  !**

# The Würfel picture

Ideal solar cell structure to reach maximum  $V_{OC}$ :

- Photoactive material **B** between highly conductive non-absorbing materials **A** and **C**
  - interfaces **A/B** and **B/C** perfectly semipermeable membranes: only one type of carrier can pass
- >  $V_{OC}$  is independent from  $V_{bi}$   
->  $V_{OC}$  equals splitting of quasi-Fermi levels

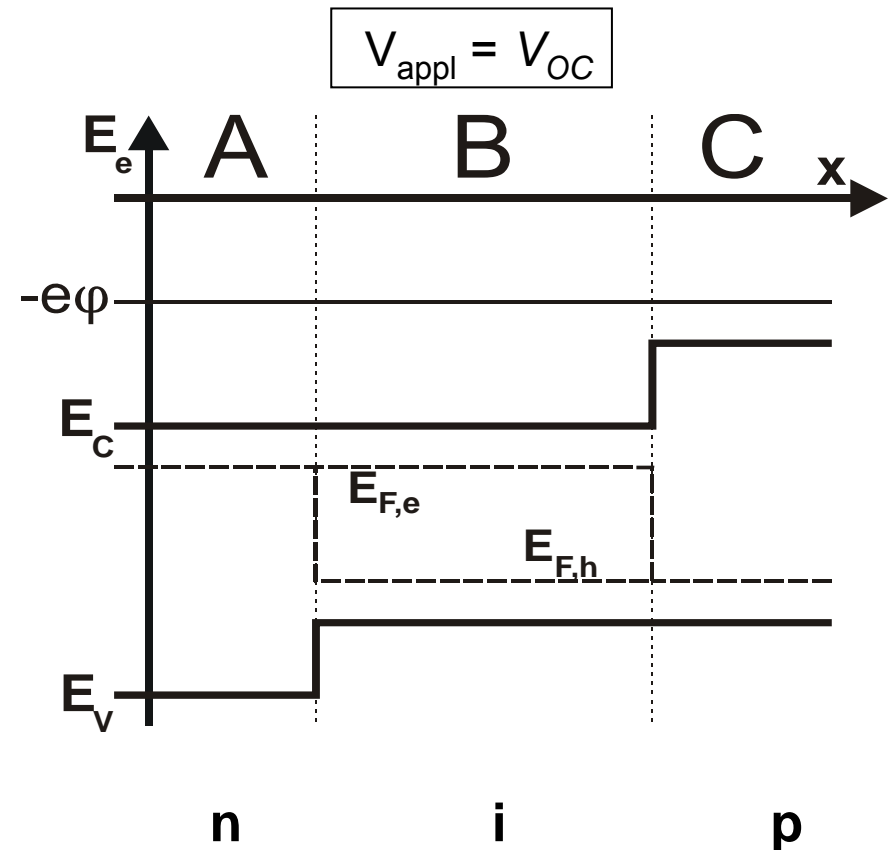


# The Würfel picture realized in an organic solar cell

- Window materials with suitable hetero-offset work as selective membranes
- Carriers driven by drift **and** diffusion

$$J_{\pm}(x) = \frac{\sigma_n}{e} \text{grad } E_{F,n} + \frac{\sigma_p}{e} \text{grad } E_{F,p}$$

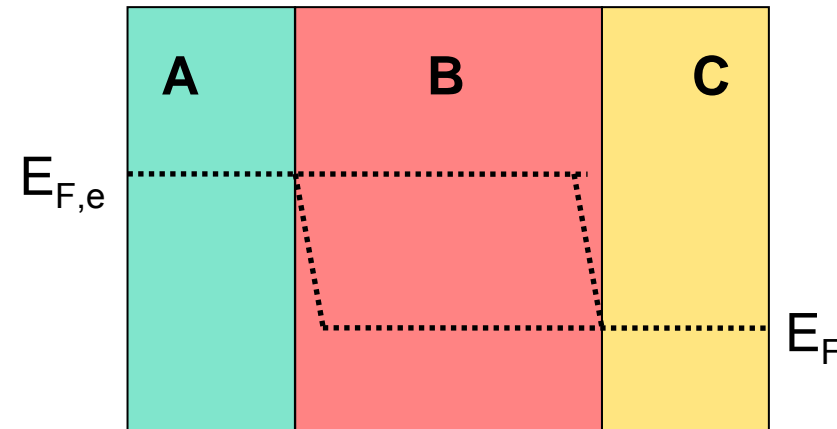
- Quasi-Fermi levels need to be „picked up“ by transport materials:  
 $E_{F,e}$  in A needs to be high,  
 $E_{F,h}$  in C needs to be low:  
**pin-structure with doped layers**



$$V_{\text{OC}} \leq E_{F,n} - E_{F,h} = E_g - kT \cdot \ln \frac{N_c N_v}{n_n n_p}$$

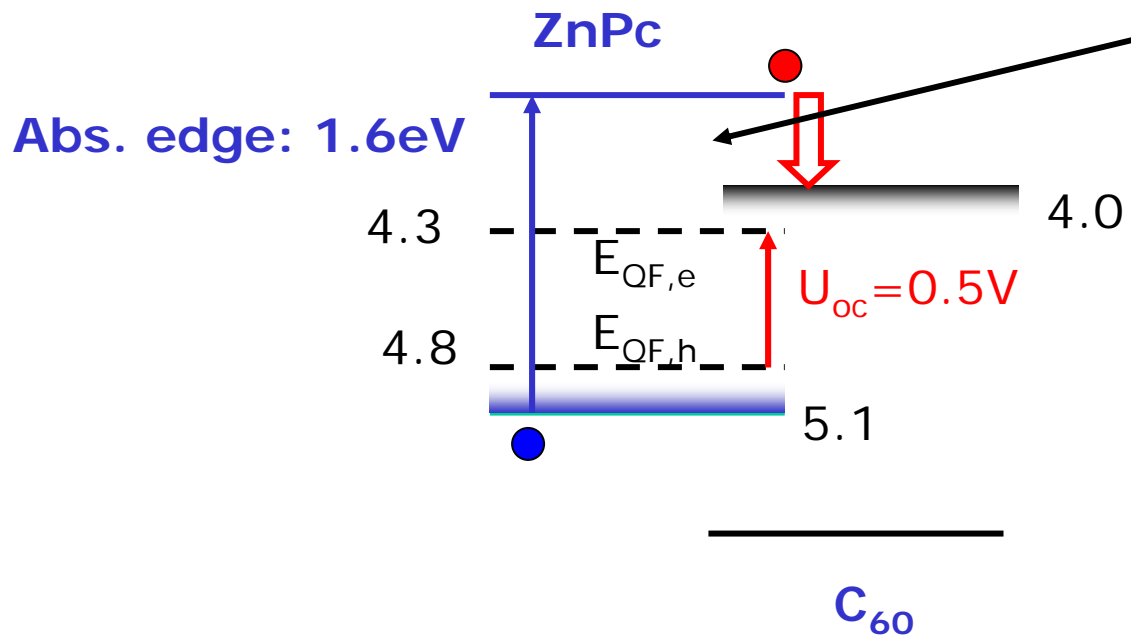
# The 3 steps to high voltage according to the Würfel picture

1. Quasi-Fermi-levels  $E_{F,e}$  and  $E_{F,h}$  must be well separated in absorber  
 $\Rightarrow$  excitons must be efficiently separated in absorber with little energy loss
2. Quasi-Fermi-levels must be „picked up“ well by transport layers A and C
3. Energy loss at electrodes must be avoided



# Step 1: Optimize the exciton separation, but with little energy loss

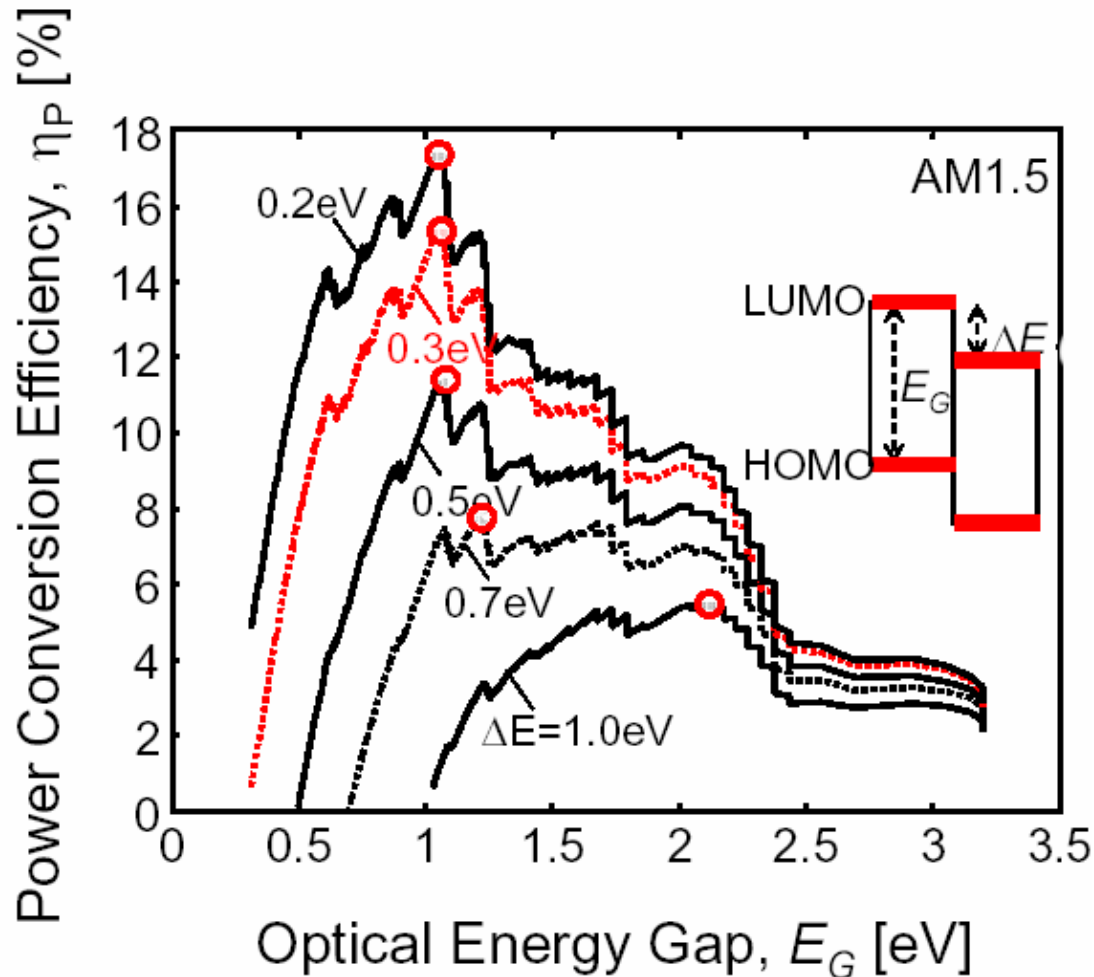
Example system: ZnPc/C<sub>60</sub>



Minimum energy loss upon charge separation: 0.2....0.7 eV?

(recent Results in polymers: Durrant et al., JACS, in press)

# Efficiency Outlook: Peumans data



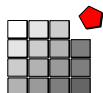
- Low-offset is critical, in particular for low gap
- Optimum gap around 1 eV
- Efficiencies  $\approx 10\%$  feasible: **tandem concepts needed!**



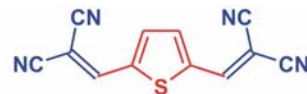
# New low gap thiophene oligomers



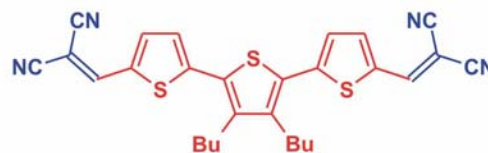
University of Ulm  
Department Organic  
Chemistry II



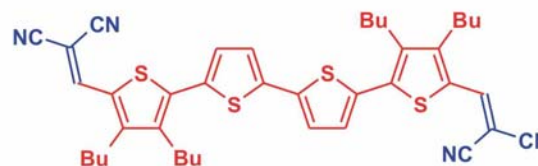
E. Brier,  
E. Reinold,  
P. Kilickiran,  
P. Bäuerle



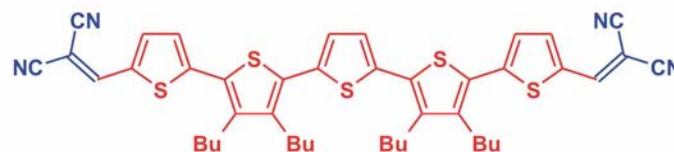
DCV1T



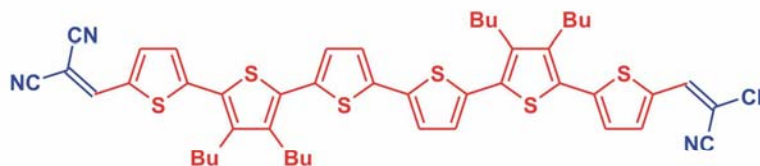
DCV3T



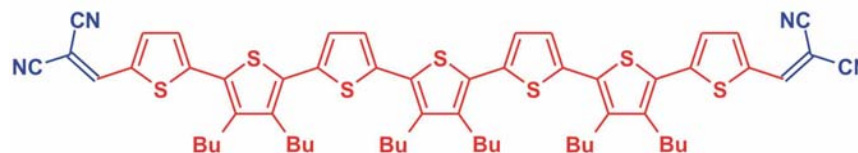
DCV4T



DCV5T

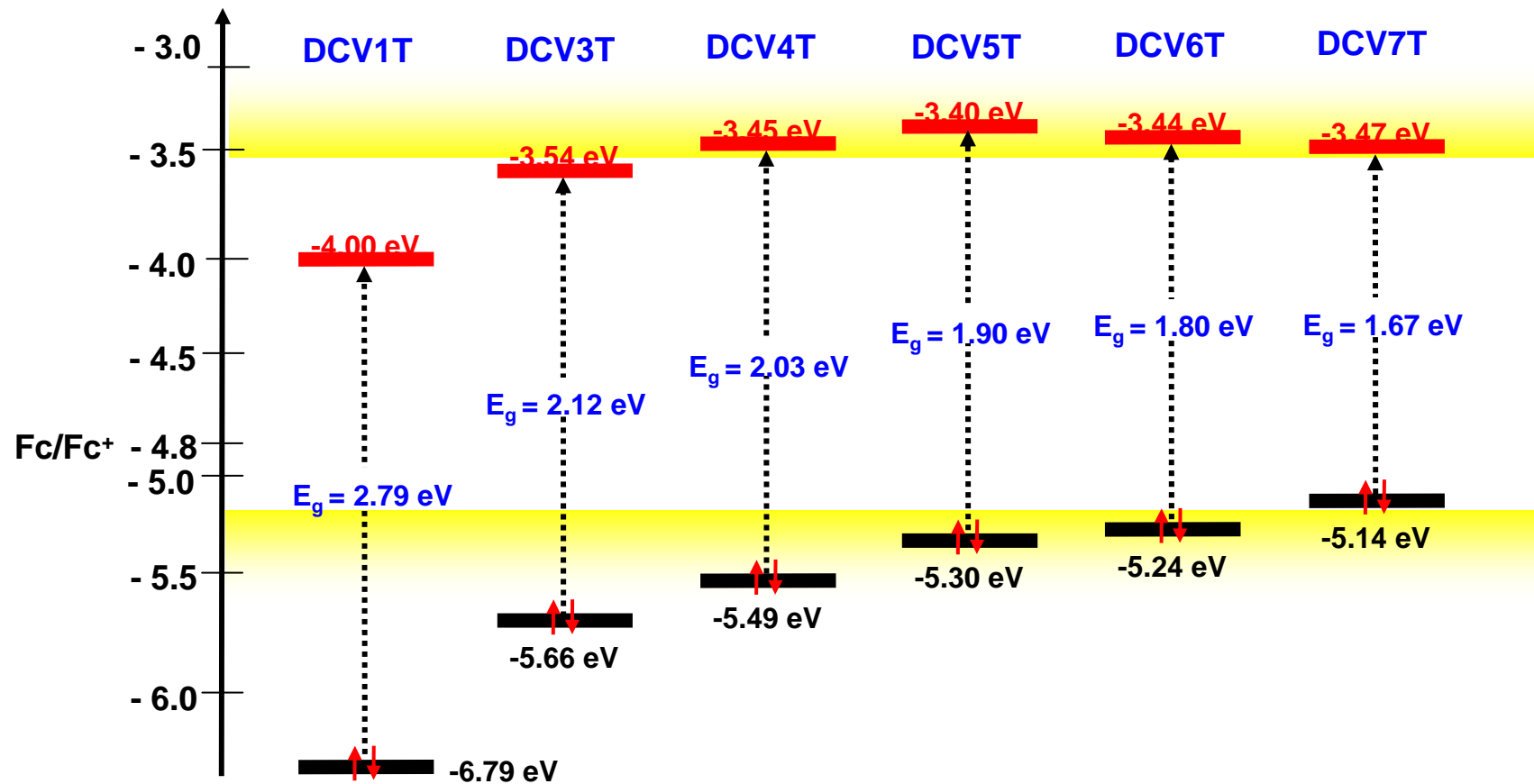


DCV6T

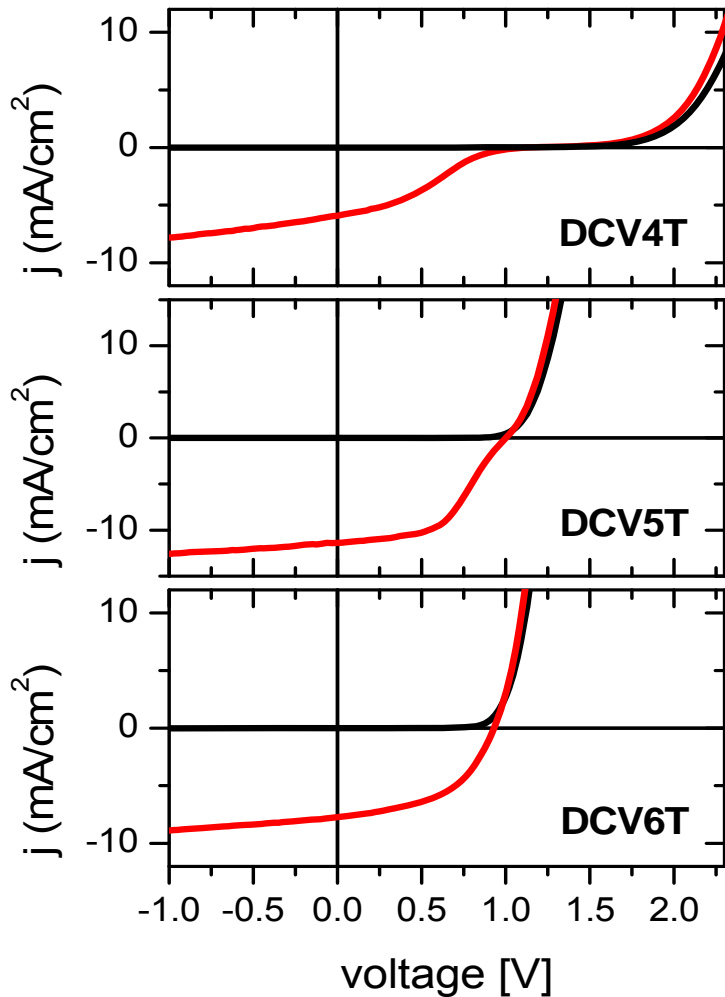


DCV7T

# New low gap thiophene oligomers: energy gaps



# Solar Cells with DCVnT



Open circuit voltage

$$V_{oc} = 1.13 \text{ V}$$

$$V_{oc} = 1.00 \text{ V}$$

$$V_{oc} = 0.93 \text{ V}$$

*decreases*

Charge carrier separation efficiency

fill factor FF

saturation factor  $j_{(-1V)}/j_{sc}$

$$FF = 27.6\%$$

$$j_{(-1V)}/j_{sc} = 1.32$$

$$FF = 50.4\%$$

$$j_{(-1V)}/j_{sc} = 1.10$$

$$FF = 49.7\%$$

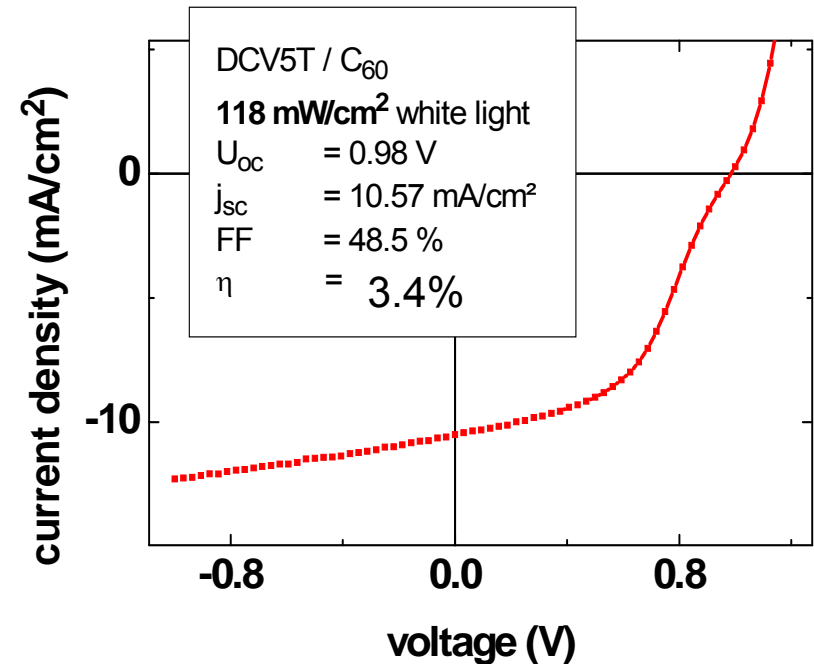
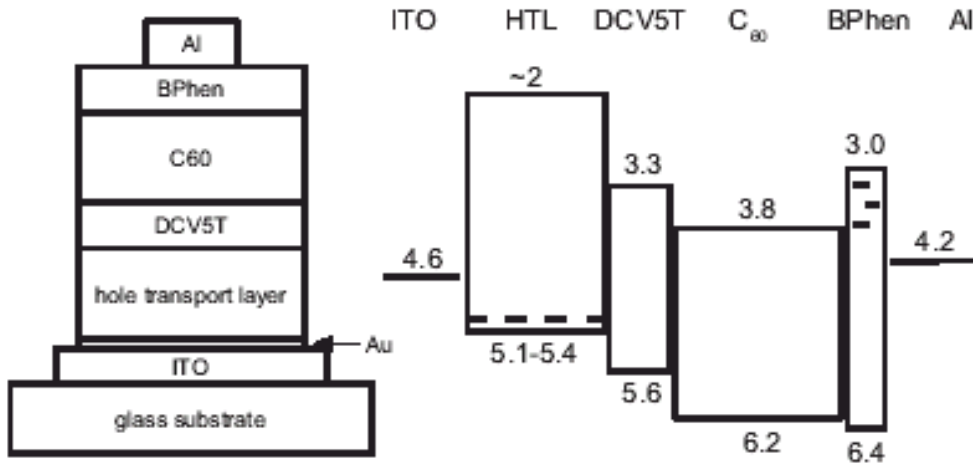
$$j_{(-1V)}/j_{sc} = 1.15$$

*increases*

with **increasing** chain length

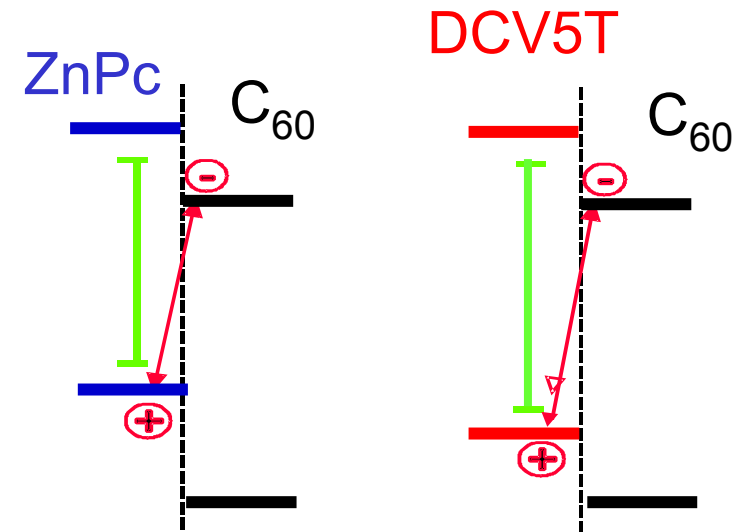
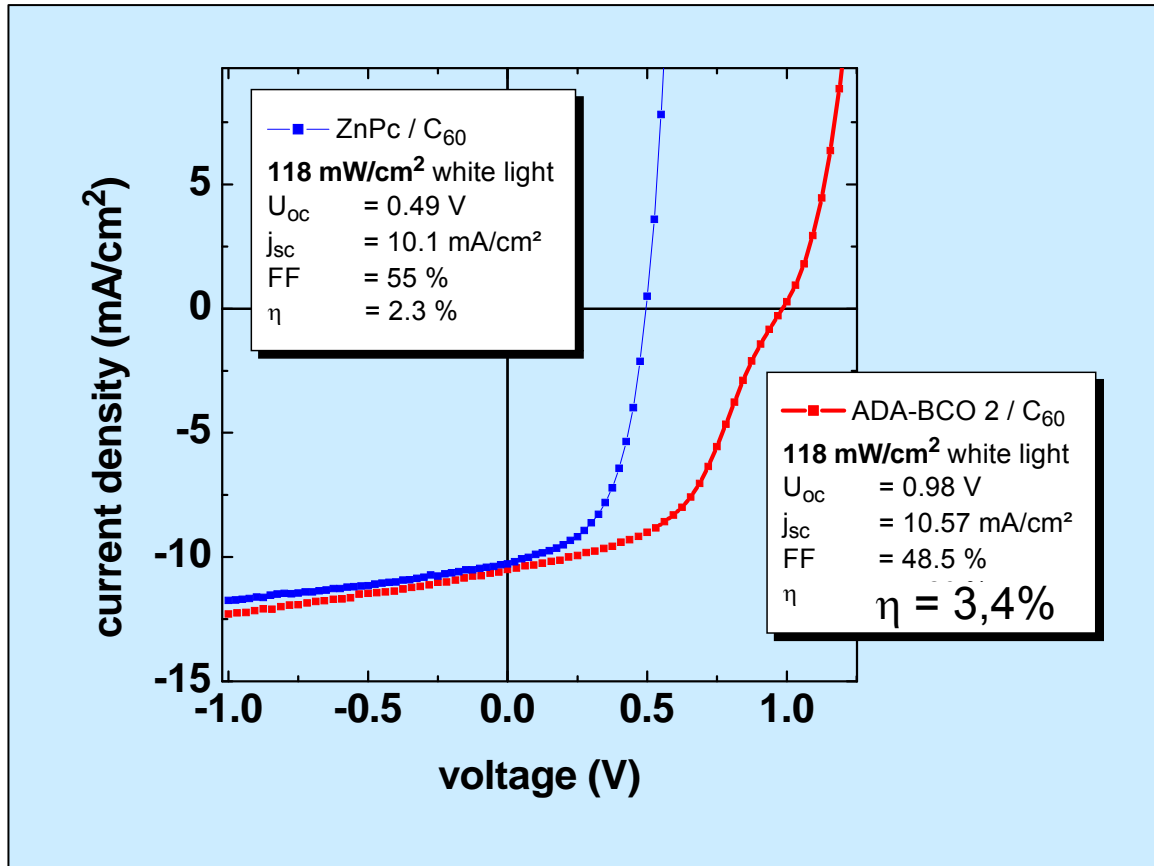
# Cells based on DCV5T / C<sub>60</sub> flat heterojunction

- V<sub>oc</sub> up to 1V for material with optical gap 1.77eV
- Single cell with up to ~ 4% efficiency @ 7nm active layer



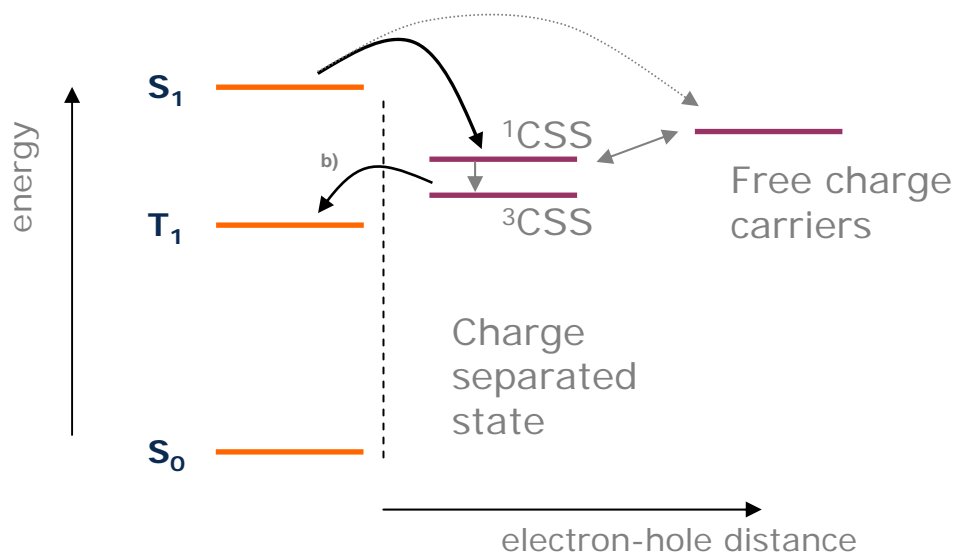
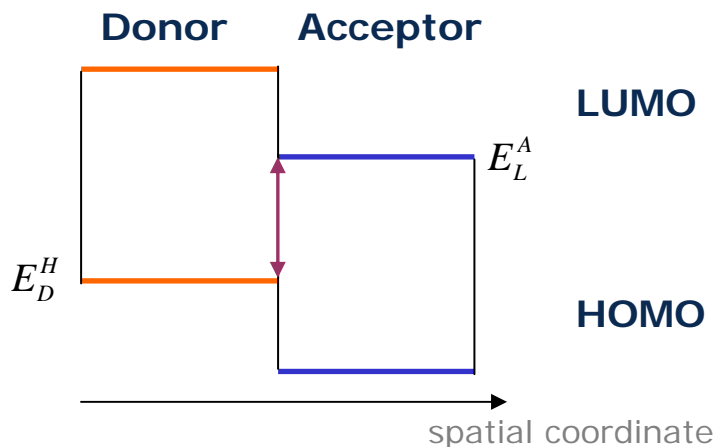
Thicker absorber layers: low fill factor -> transport problem

# Comparison DCV5T vs. ZnPc



# Study of Exciton Separation

R. Schueppel et al., ChemPhysChem 8, 1497-1503 (2007)



High open circuit voltage:

$$eV_{oc} \propto E_L^A - E_H^D$$

Increasing energetic gap between LUMO of acceptor and HOMO of donor

Impact on charge separation:

- minimized loss of free energy
- dissociation of geminate pair necessary, excess energy of „hot exciton“ is minimized
- recombination into the triplet state becomes possible

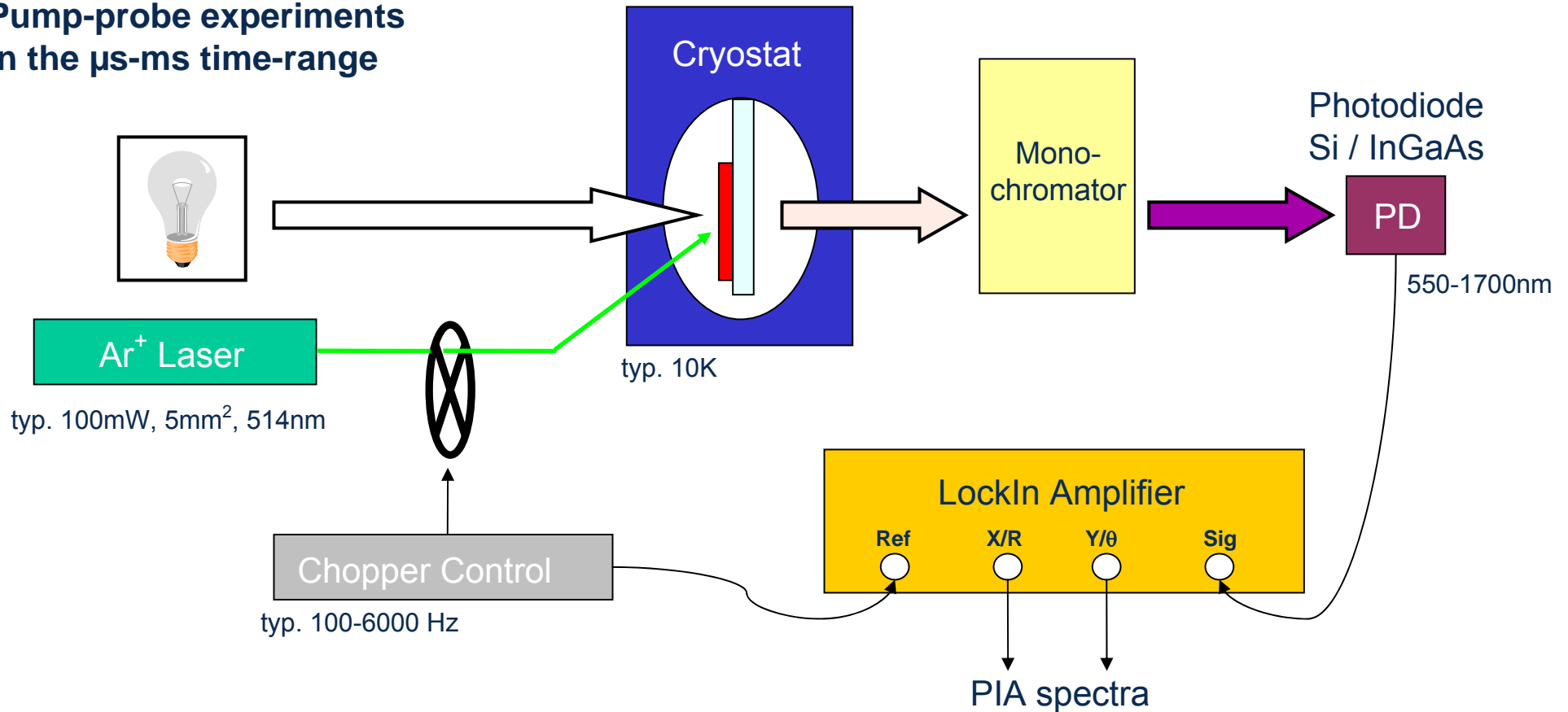
=> introducing a loss mechanism

Veldman *et al.*, Thin Film Solids 511, 333 (2006)

b) pathway suggested by Ford *et al.*, PRB 71, 125212 (2005)

# Photoinduced Absorption Spectroscopy

## Pump-probe experiments in the $\mu\text{s}$ -ms time-range

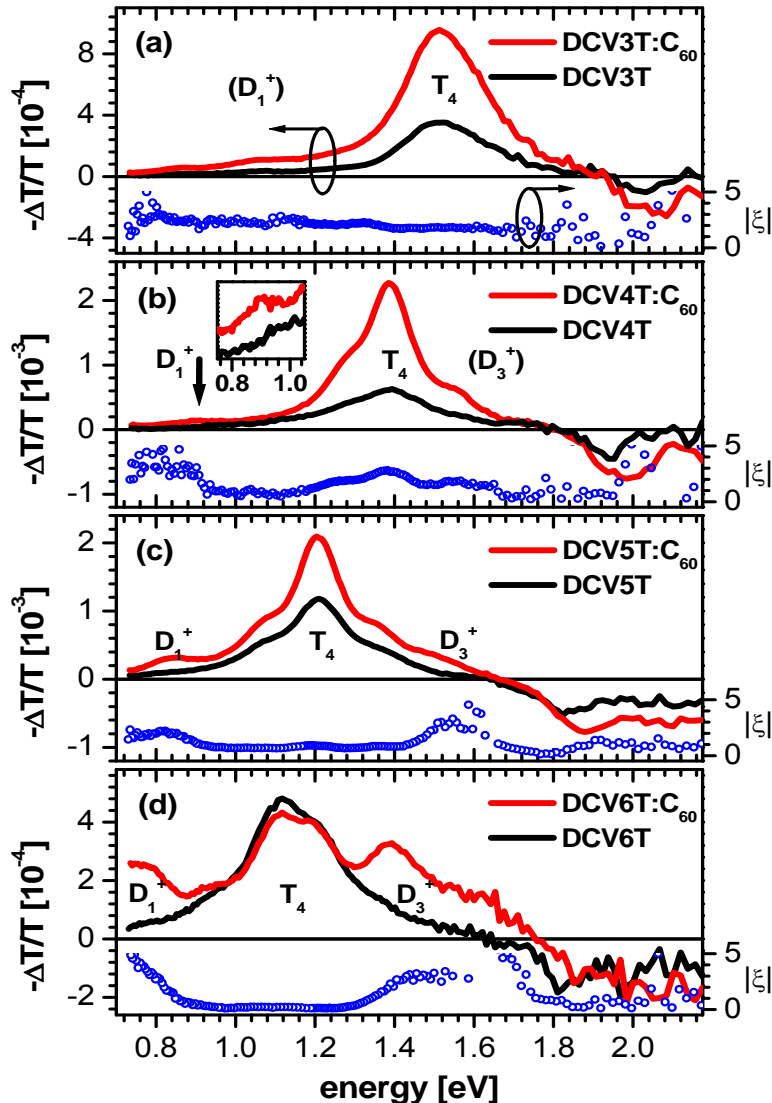


Parameters in photoinduced absorption experiment:

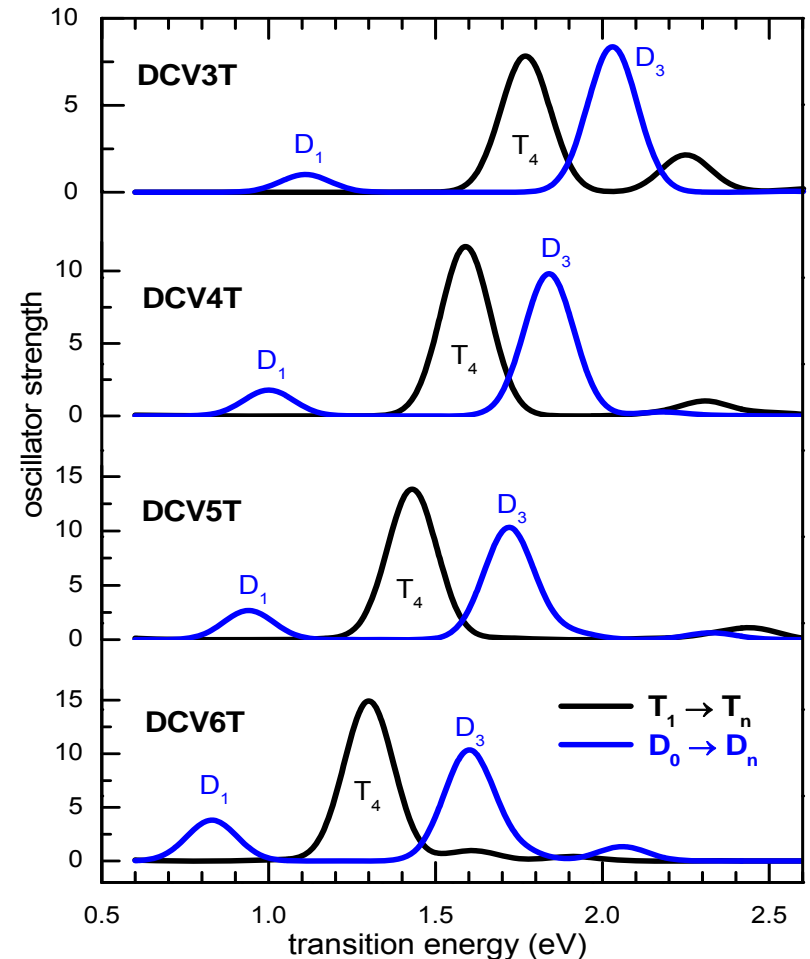
**wavelength**, modulation frequency, temperature, pump intensity, bias voltage

# Photoinduced Absorption Spectroscopy: Results

PIA spectra of DCVnT:C60 at 10K



Triplet ( $T_1 \rightarrow T_n$ ) and cation ( $D_0 \rightarrow D_n$ ) transitions of DCVnT; TD-DFT calculations by Karin Schmidt (B3LYP, unrestricted, relaxed geometries)



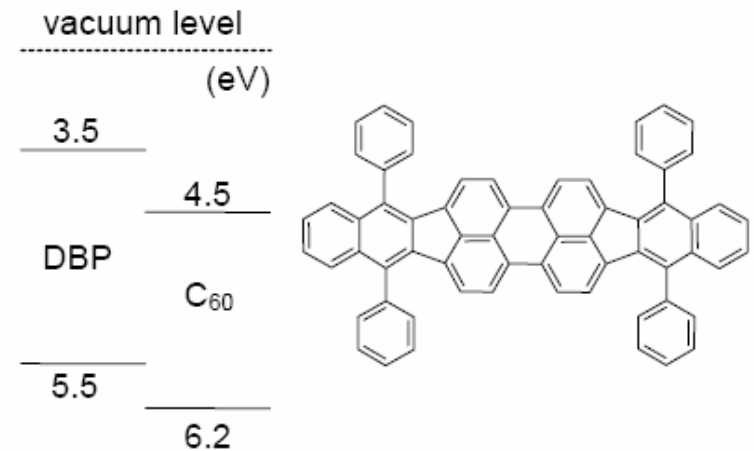


# Another new Donor Material

## ORGANIC THIN-FILM SOLAR CELL EMPLOYING A NOVEL ELECTRON-DONOR MATERIAL

Hiroshi Kanno<sup>1\*</sup>, Daisuke Fujishima<sup>1</sup>, Makoto Shirakawa<sup>2</sup>,  
Toshihiro Kinoshita<sup>1</sup>, Eiji Maruyama<sup>1</sup>, Kenichi Shibata<sup>2</sup>, and Makoto Tanaka<sup>1</sup>  
R&D H.Q., Advanced Energy Research Center,  
<sup>1</sup> Solar Energy Research Department,  
<sup>2</sup> Energy Device Research Department  
Sanyo Electric Co., Ltd.  
7-3-2 Ibukidai-higashimachi, Nishi-ku, Kobe, Hyogo 651-2242, Japan  
\* Corresponding address: hiroshi.kanno@sanyo.com

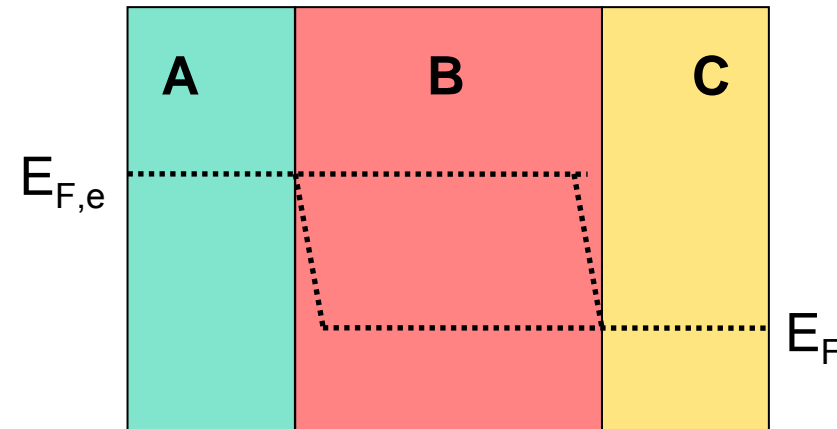
- $V_{OC}$  of 0.92V
- Efficiency: 3.56%



**Fig. 1** Left: The proposed energy diagram of DBP and C<sub>60</sub>. Right: The molecular structure of DBP

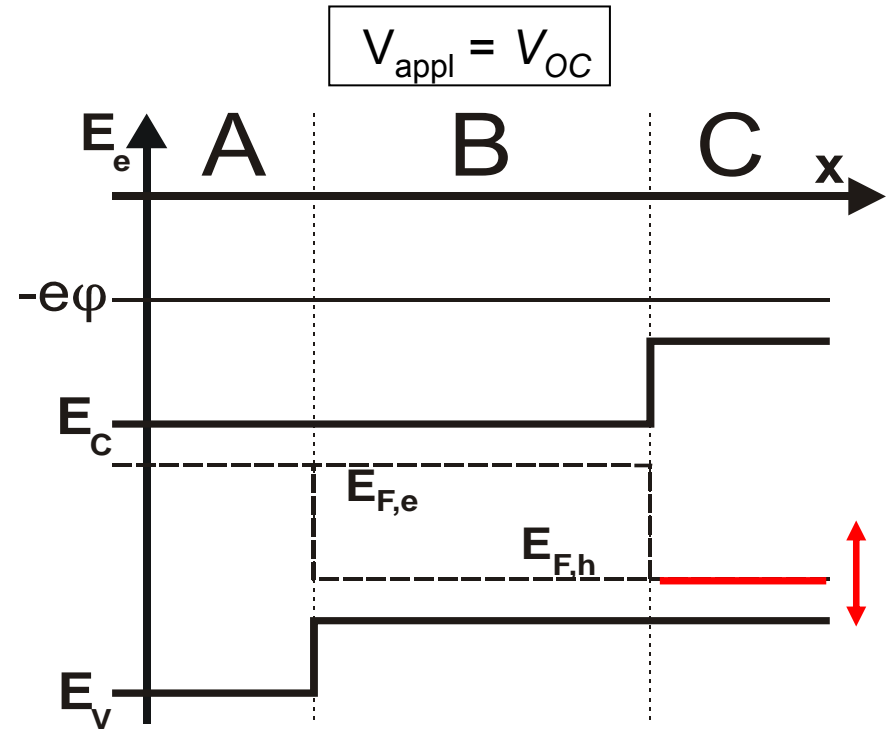
# The 3 steps to high voltage according to the Würfel picture

1. Quasi-Fermi-levels  $E_{F,e}$  and  $E_{F,h}$  must be well separated in absorber  
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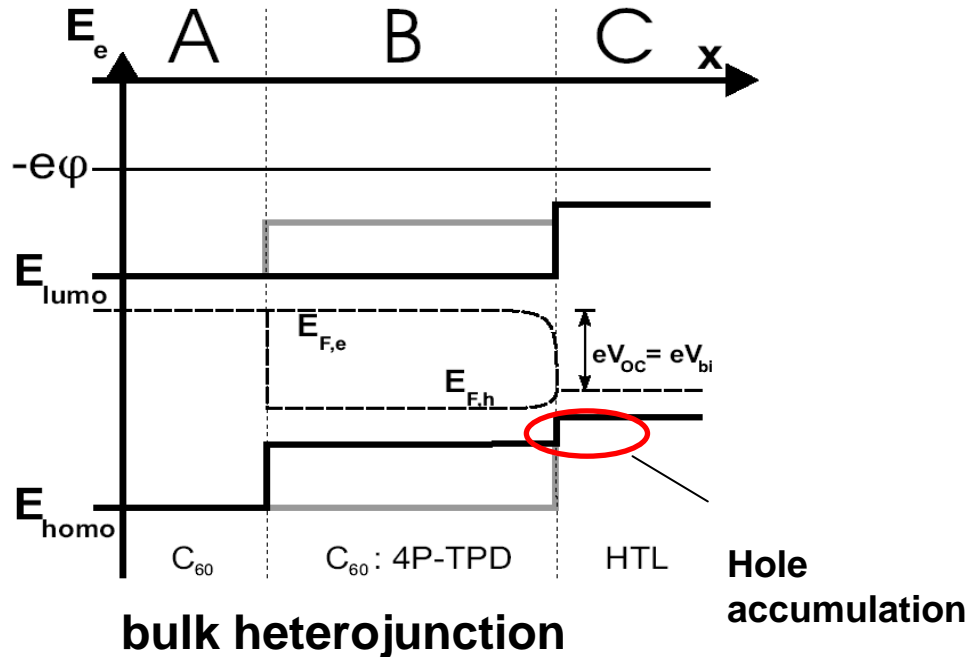
# Systematic study of quasi-Fermi level „pick-up“

- C. Urich et al., Adv. Functional Materials **17**, 2991 (2007)
- Shift of the Fermi level in the hole transport layer C
- Comparison of flat and bulk heterojunction



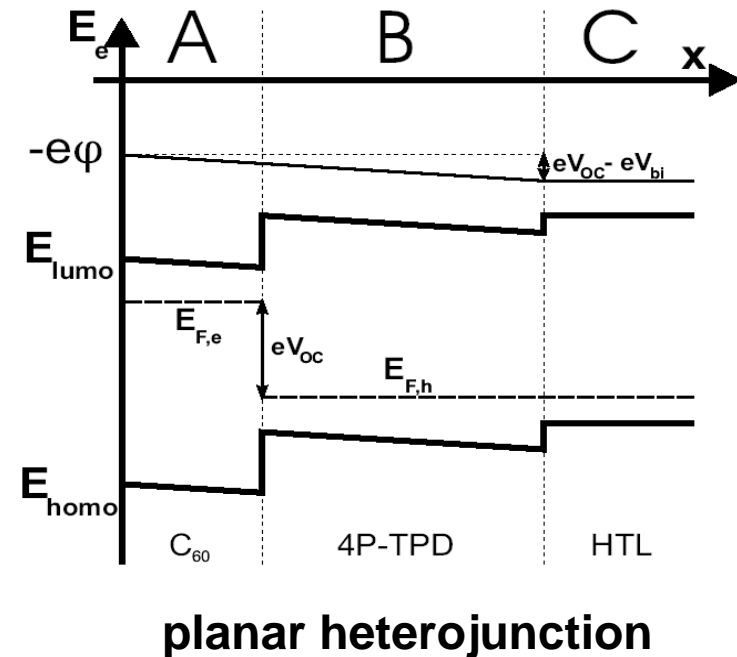
# Comparison bulk vs. planar heterojunction

Potential curves under open circuit condition:



- charge carrier generation in **B**
- HOMO offset betw. 4P-TPD and HTL: enhanced recombination at **B/C**

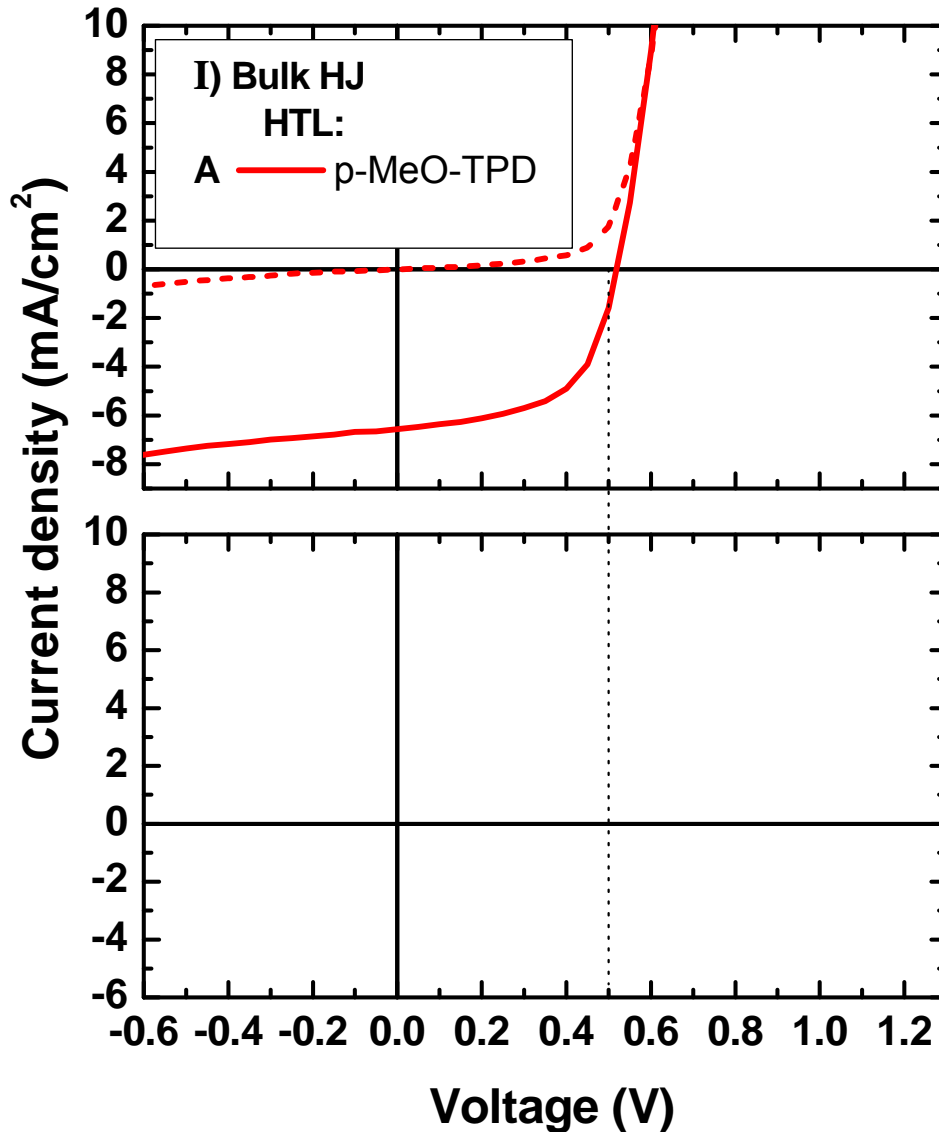
->  $V_{OC}$  cannot exceed  $V_{bi}$  significantly



- charge carrier generation at interface **A/B**
- $C_{60}$  and HTL are spatially separated
- quasi-Fermi levels are constant
- charge carriers are driven against electric field by diffusion ( $V_{OC} > V_{appl} > V_{bi}$ )

->  $V_{OC}$  equals splitting of QFL and exceeds  $V_{bi}$

# Influence of hole transporter Fermi level on $V_{OC}$

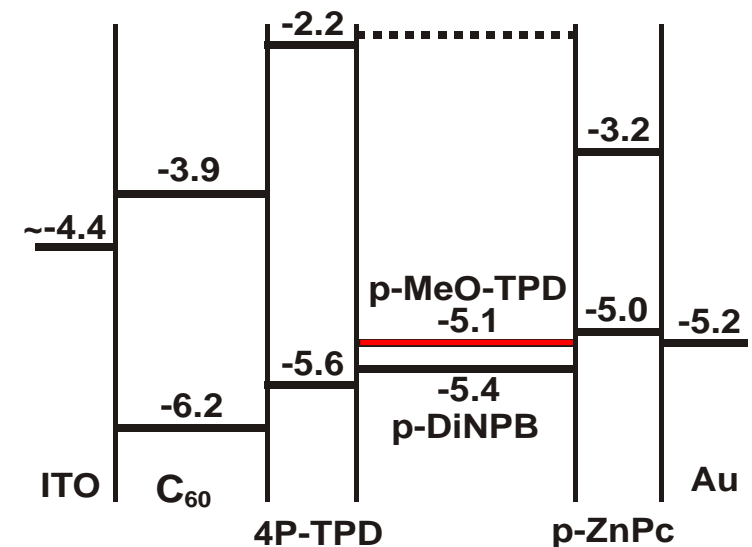


## I) bulk HJ

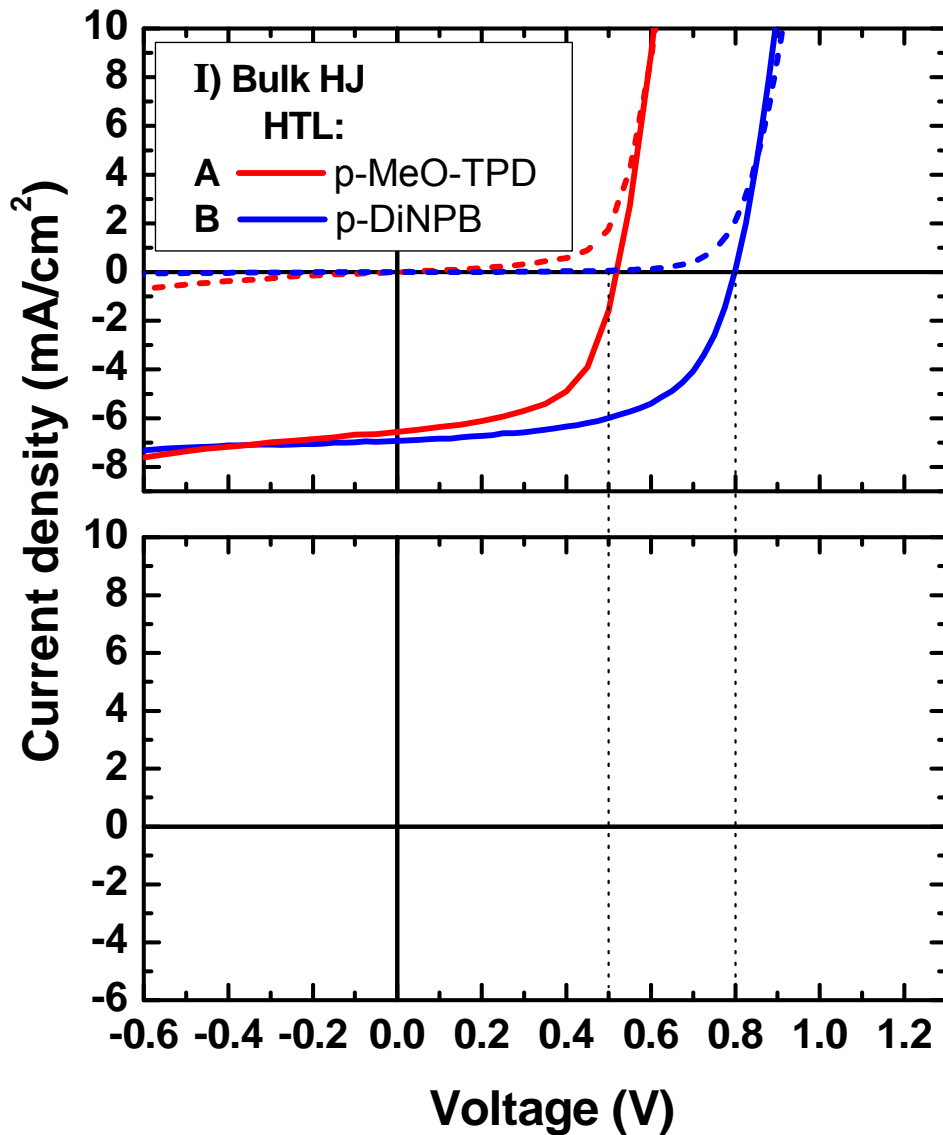
$V_{OC}$  cannot exceed  $V_{bi}$  significantly

bulk HJ with p-MeO-TPD:

$$V_{bi} \approx V_{OC} = 0.5V$$



# Influence of hole transporter Fermi level on $V_{OC}$



## I) bulk HJ

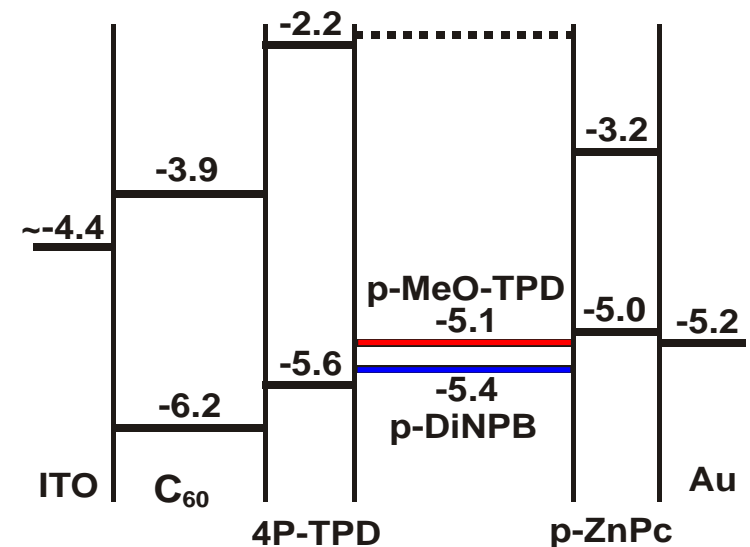
$V_{OC}$  cannot exceed  $V_{bi}$  significantly

bulk HJ with p-MeO-TPD:

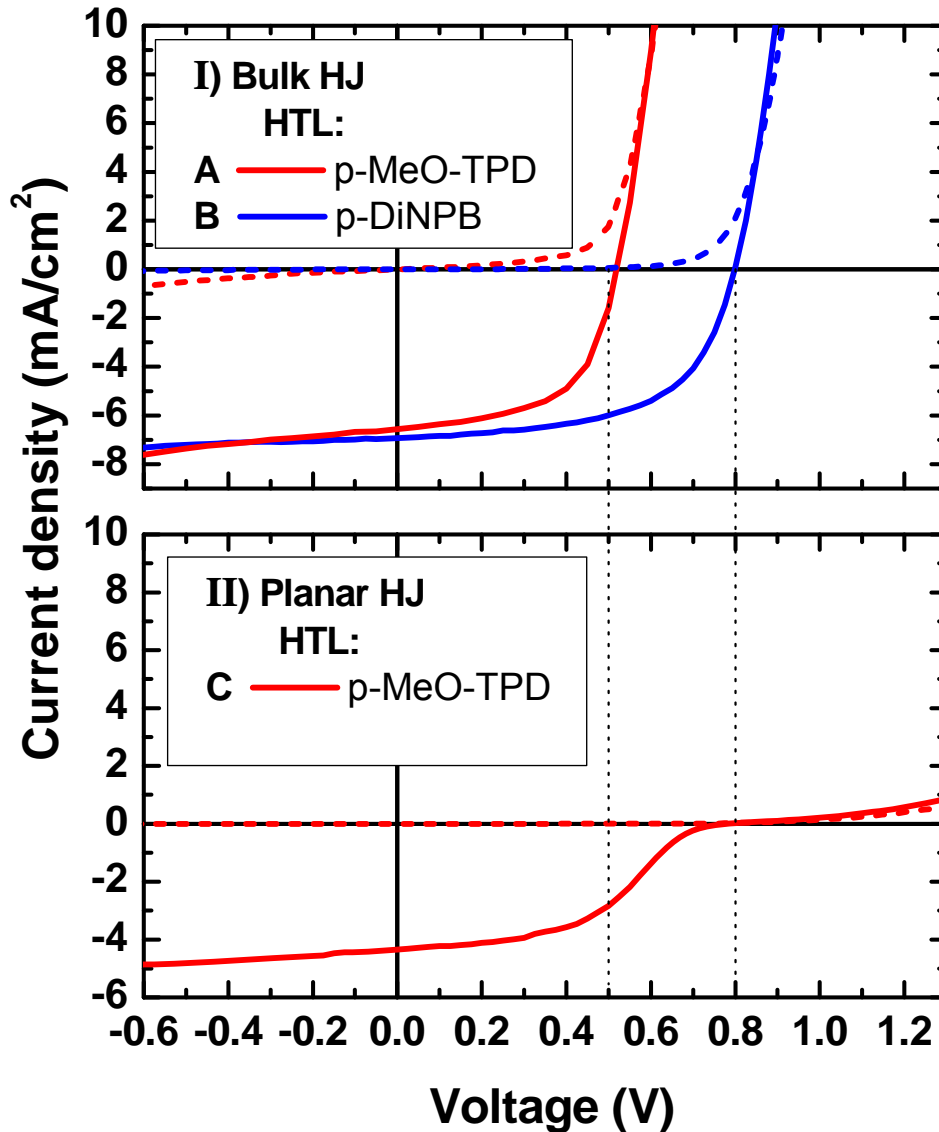
$$V_{bi} \approx V_{OC} = 0.5V$$

bulk HJ with p-DiNPB:

$$V_{bi} \approx V_{OC} = 0.8V$$



# Comparison bulk vs. Planar heterojunction



## I) bulk HJ

$V_{OC}$  cannot exceed  $V_{bi}$  significantly

bulk HJ with p-MeO-TPD:

$$V_{bi} \approx V_{OC} = 0.5V$$

bulk HJ with p-DiNPB:

$$V_{bi} \approx V_{OC} = 0.8V$$

## I) planar HJ

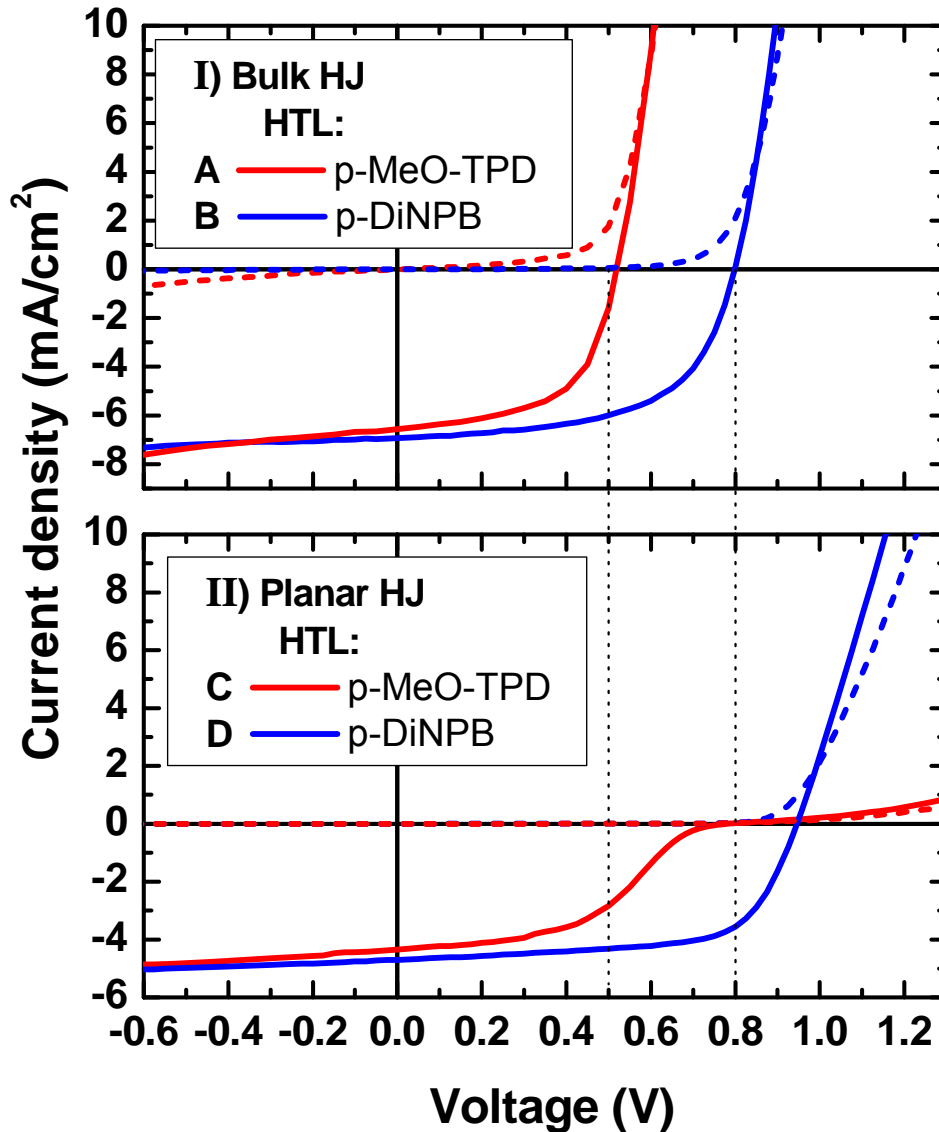
$$V_{OC} = E_{F,n} - E_{F,h} = E_g - kT \cdot \ln \frac{N_C N_V}{n_n n_p}$$

$V_{OC}$  is predominantly determined by  $E_g$

$$E_g = E_{\text{acceptor}}^{\text{LUMO}} - E_{\text{donor}}^{\text{HOMO}}$$

S-shape due to comp. low  $V_{bi}$  and barrier (HTL/4P-TPD)

# Influence of hole transporter Fermi level on $V_{OC}$



## I) bulk HJ

$V_{OC}$  cannot exceed  $V_{bi}$  significantly

bulk HJ with p-MeO-TPD:

$$V_{bi} \approx V_{OC} = 0.5V$$

bulk HJ with p-DiNPB:

$$V_{bi} \approx V_{OC} = 0.8V$$

## I) planar HJ

$$V_{OC} = E_{F,n} - E_{F,h} = E_g - kT \cdot \ln \frac{N_C N_V}{n_n n_p}$$

$V_{OC}$  is predominantly determined by  $E_g$

$$E_g = E_{\text{acceptor}}^{\text{LUMO}} - E_{\text{donor}}^{\text{HOMO}}$$

S-shape due to comp. low  $V_{bi}$  and barrier (HTL/4P-TPD)

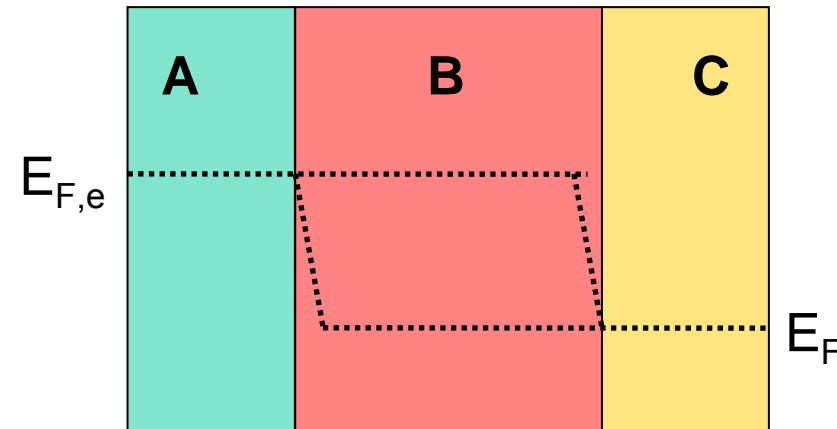
increased  $V_{bi}$

-> no S-shape, high FF (66%), high  $V_{OC}$  (0.95V)

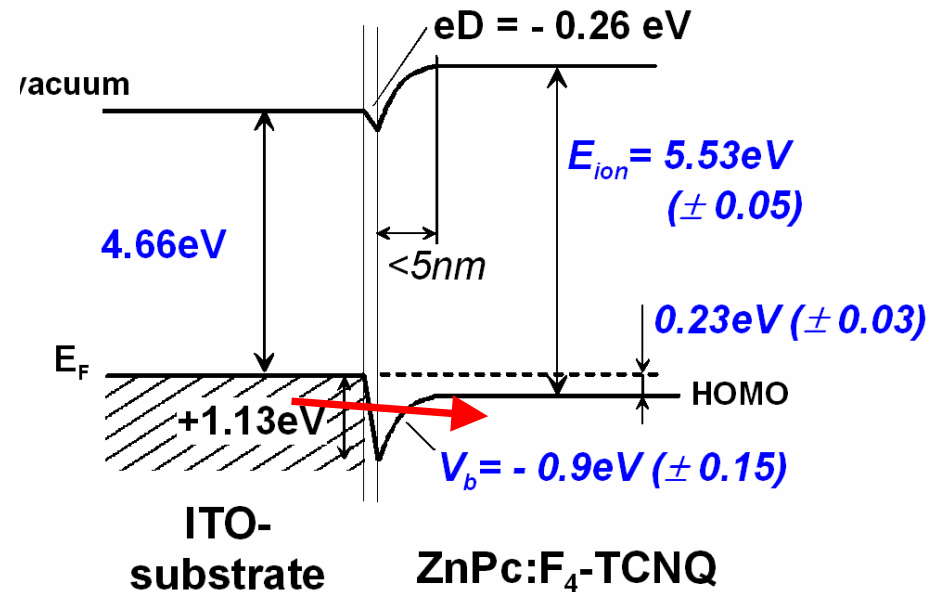
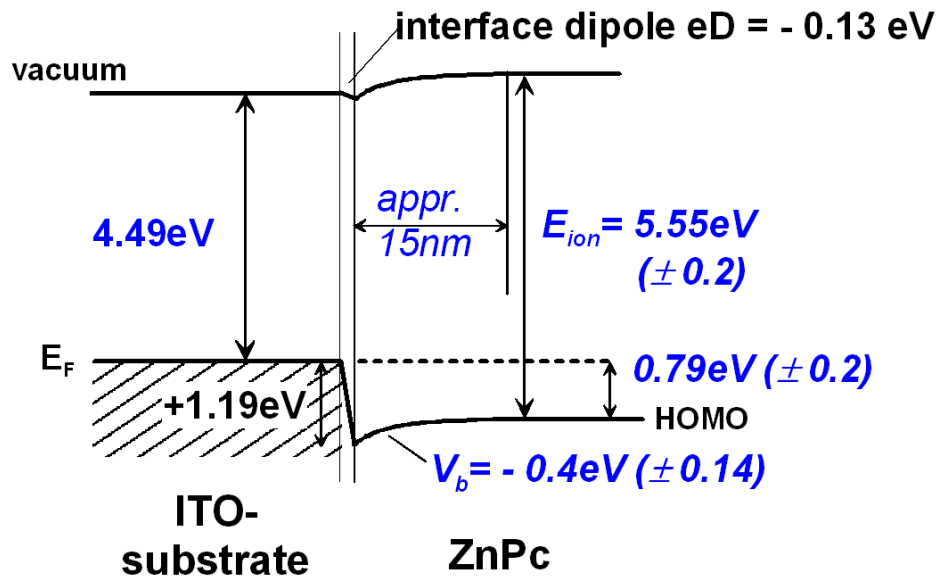


# The 3 steps to high voltage according to the Würfel picture

1. Quasi-Fermi-levels  $E_{F,e}$  and  $E_{F,h}$  must be well separated in absorber  
 $\Rightarrow$  excitons must be efficiently separated in absorber with little energy loss
2. Quasi-Fermi-levels must be „picked up“ well by transport layers A and C
3. Energy loss at contact to electrodes must be avoided



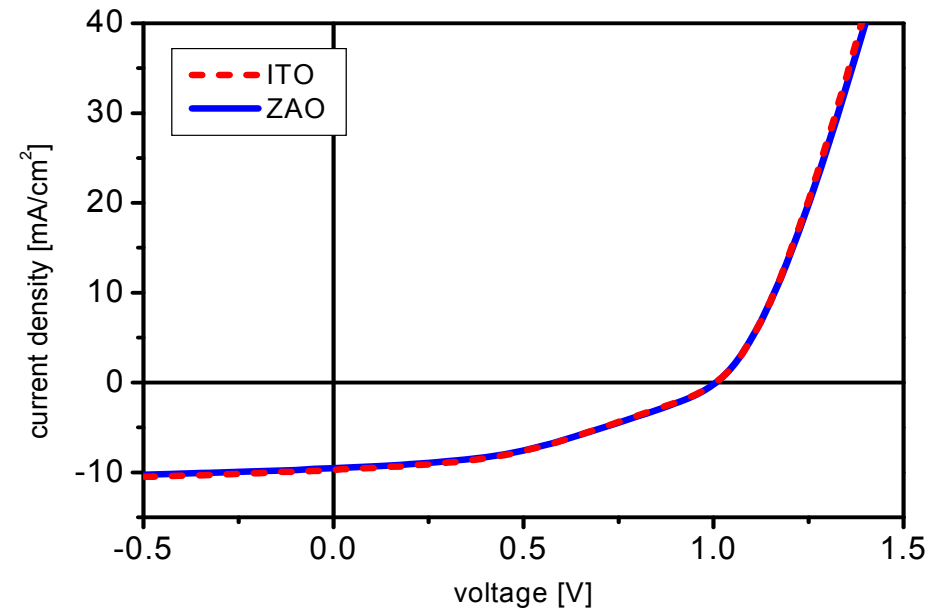
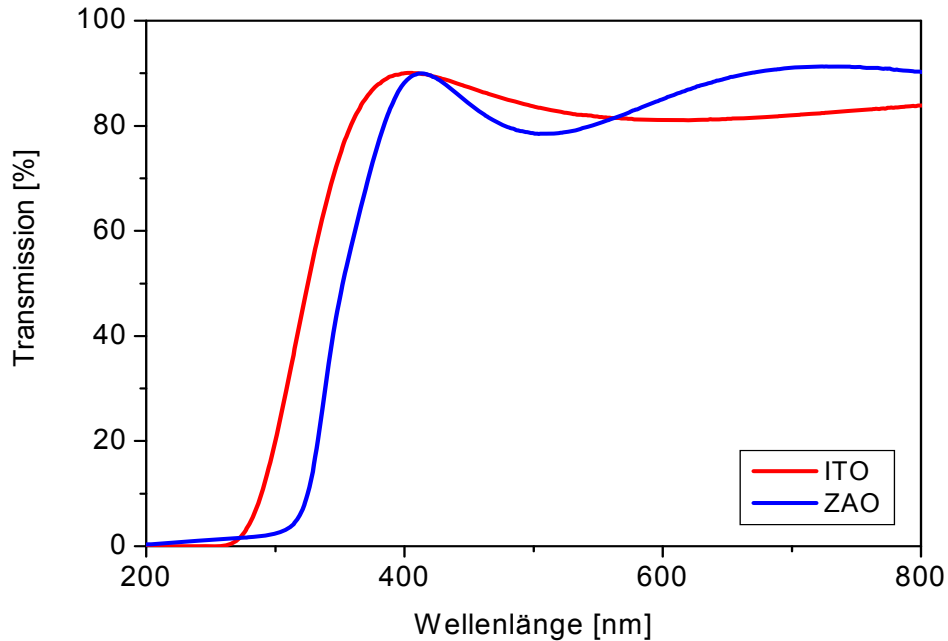
# Creation of ohmic contacts by doping



undoped: blocking

doped: ohmic

# Example: Replace ITO by ZnO



- IV-characteristic of both devices comparable

- same open circuit voltage  $V_{oc}$ :  
work function of TCO does not influence the  $V_{oc}$

	$j_{sc}$ [mA/cm <sup>2</sup> ]	$V_{oc}$ [V]	FF	$\eta^*$ [%]
ITO	6.35	1.01	0.4	2.6
ZAO	6.06	1.01	0.41	2.5

\*a spectral mismatch between sun simulator and AM 1.5 spectra was taken into account

## Conclusions on the voltage

- Optimized energy step at heterojunction is crucial
- $V_{oc}$  can exceed  $V_{bi}$  in organic solar cells
- However, there are limits: diffusion requires large carrier gradient:  
excessive recombination near the photoactive zone
- Doped transport layers allow high  $V_{bi}$  and virtually any contact material

# Outline

- Some thoughts about Organic PV in general
- Status of small-molecule organic solar cells:  
key innovations in the past
- **Future challenges:**
  - understanding and increasing the voltage
  - **covering the entire solar spectrum: IR & tandem cells**
  - stability
- Low-cost manufacturing

# Extending absorption to the infrared: SnPc

APPLIED PHYSICS LETTERS 87, 233508 (2005)

## Organic solar cells with sensitivity extending into the near infrared

Barry P. Rand, Jiangeng Xue,<sup>a)</sup> Fan Yang, and Stephen R. Forrest<sup>b)</sup>

*Department of Electrical Engineering and Princeton Institute for the Science and Technology of Materials (PRISM), Princeton University, Princeton, New Jersey 08544*

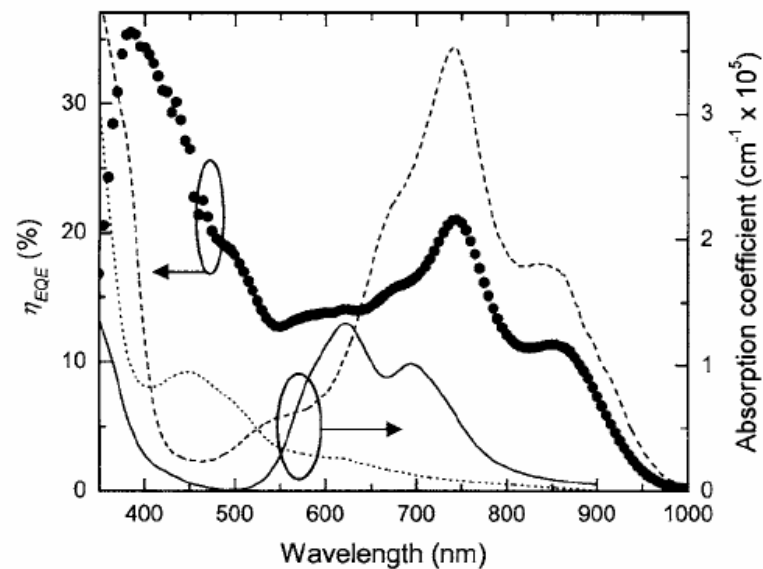


FIG. 3. Measured external quantum efficiency ( $\eta_{EQE}$ ) spectrum (filled circles) for the device of Fig. 1. The absorption coefficients of CuPc (solid line), a 50-Å-thick film on SnPc (dashed line), and C<sub>60</sub> (dotted line) are also shown.

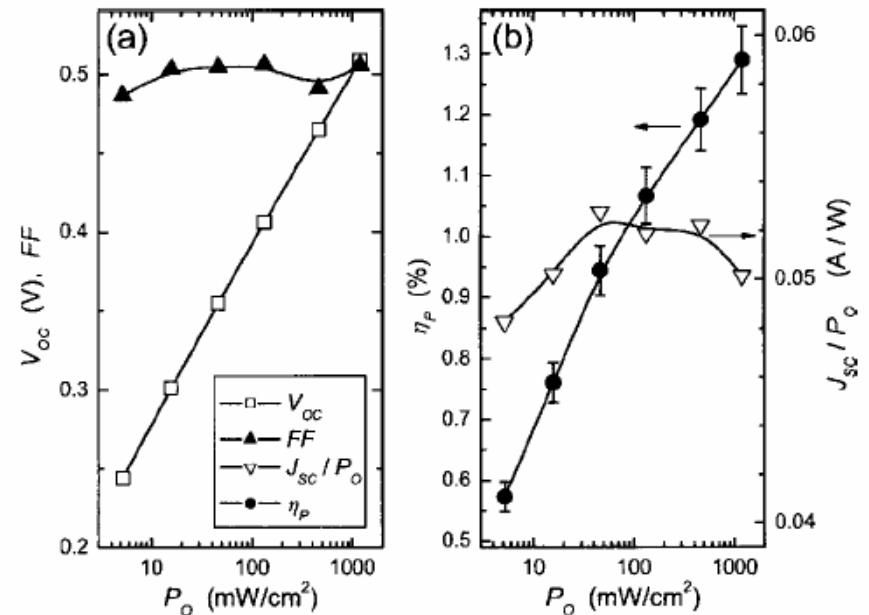
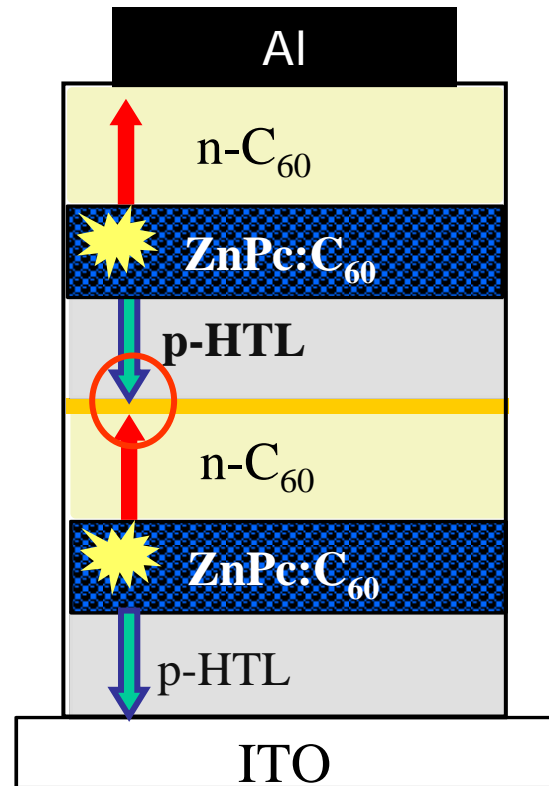
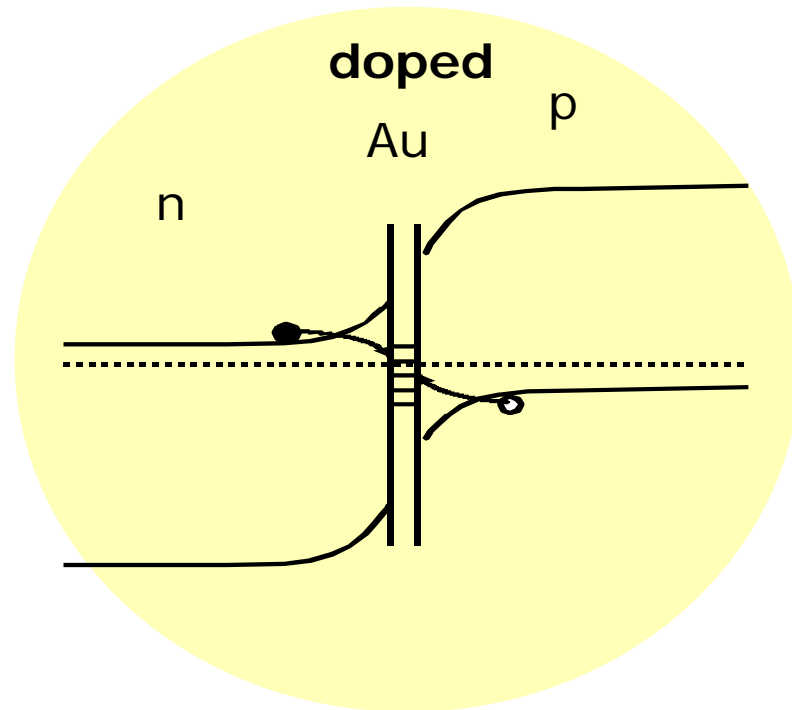
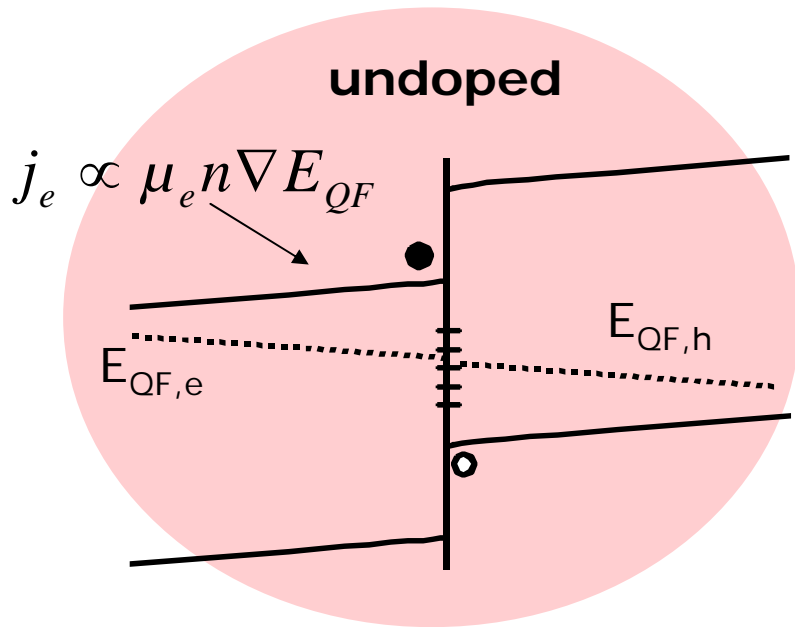
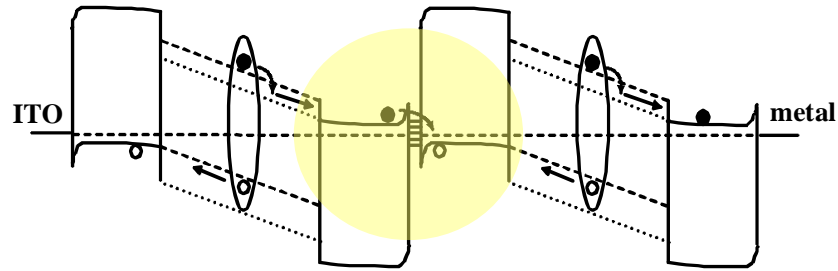


FIG. 2. (a) Fill factor (FF), open-circuit voltage ( $V_{OC}$ ), (b) responsivity ( $J_{SC}/P_0$ ), and power conversion efficiency ( $\eta_P$ ) of devices with the same structure as Fig. 1 under various AM1.5G standard solar illumination intensities,  $P_0$ . The solid lines serve as guides for the eyes.

# Pin-tandem cells: charge recombination

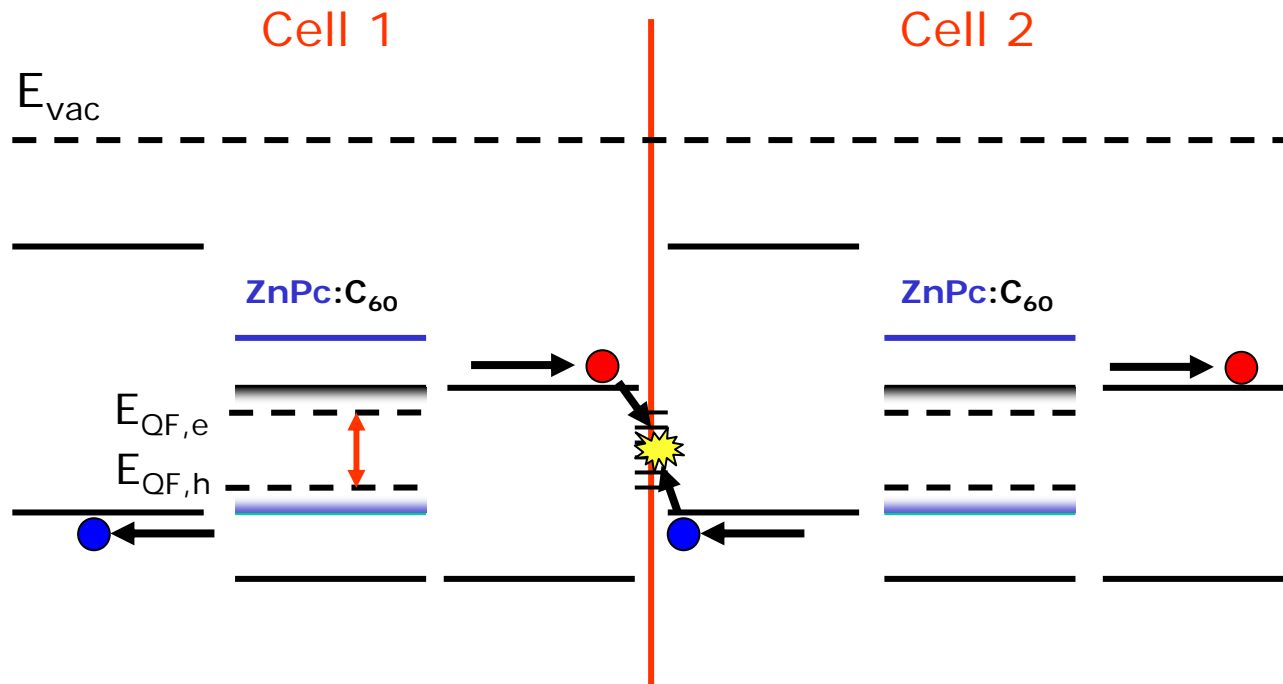


# Doping is crucial for tandem cells: Interface recombination with minimum free energy loss





# Recombination without doping I

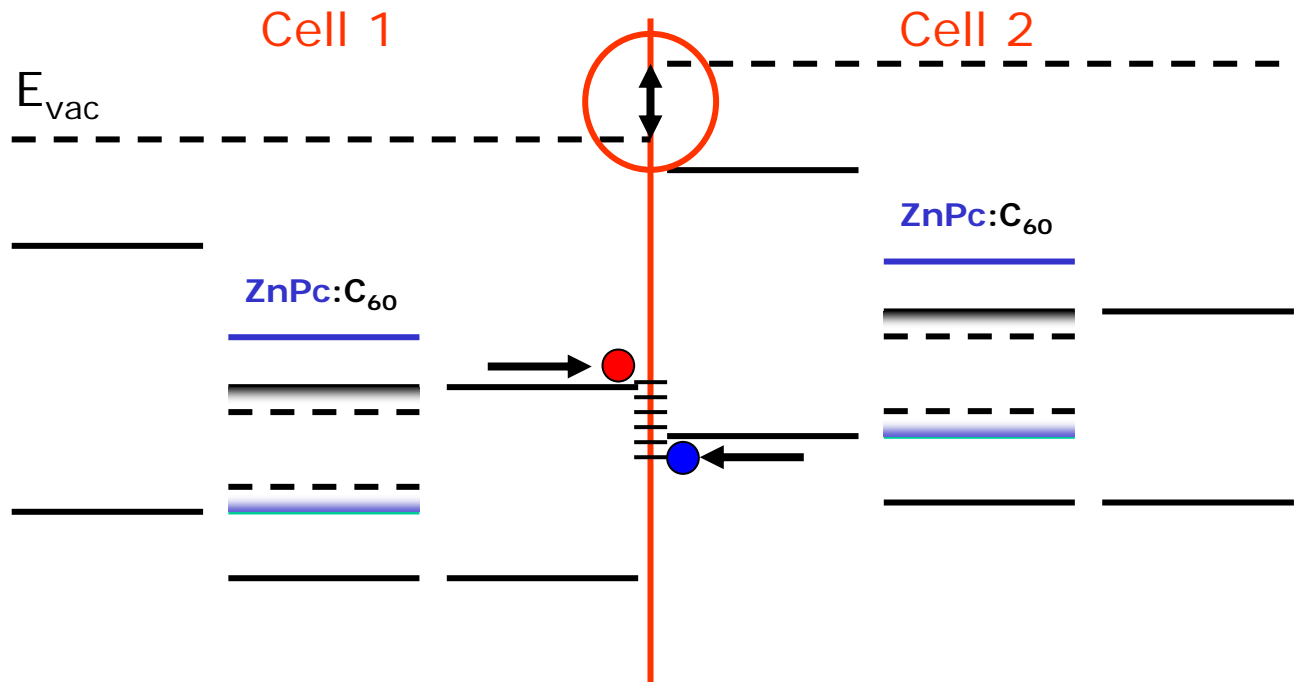


No doping, no interface dipole

→ Carrier pair transforms its complete energy into heat upon recombination

→ Recombination centers are not enough

# Recombination without doping II

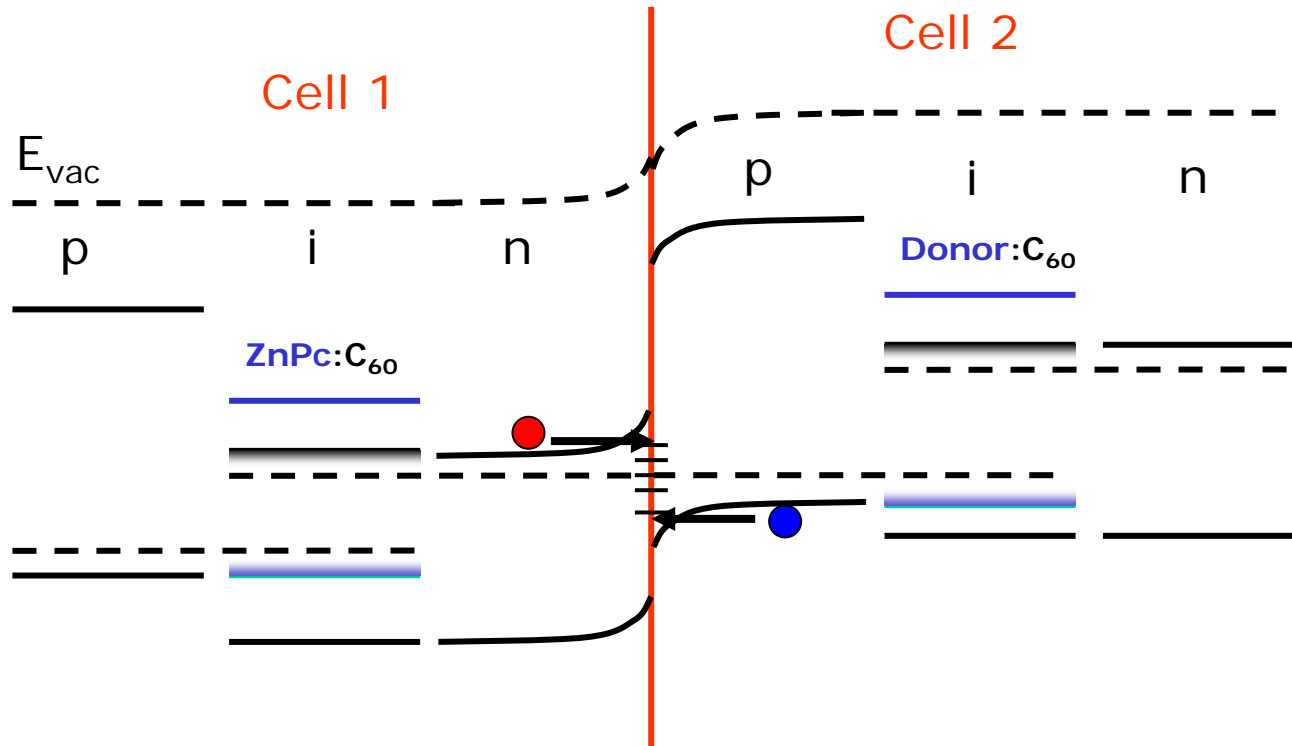


Lucky strike:

Formation of **suitable interface dipole** upon deposition of metal nanoclusters onto cell 1

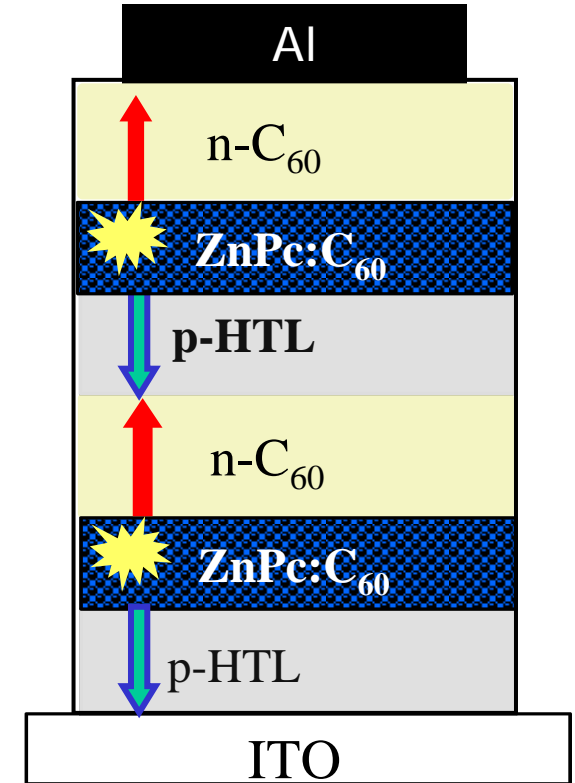
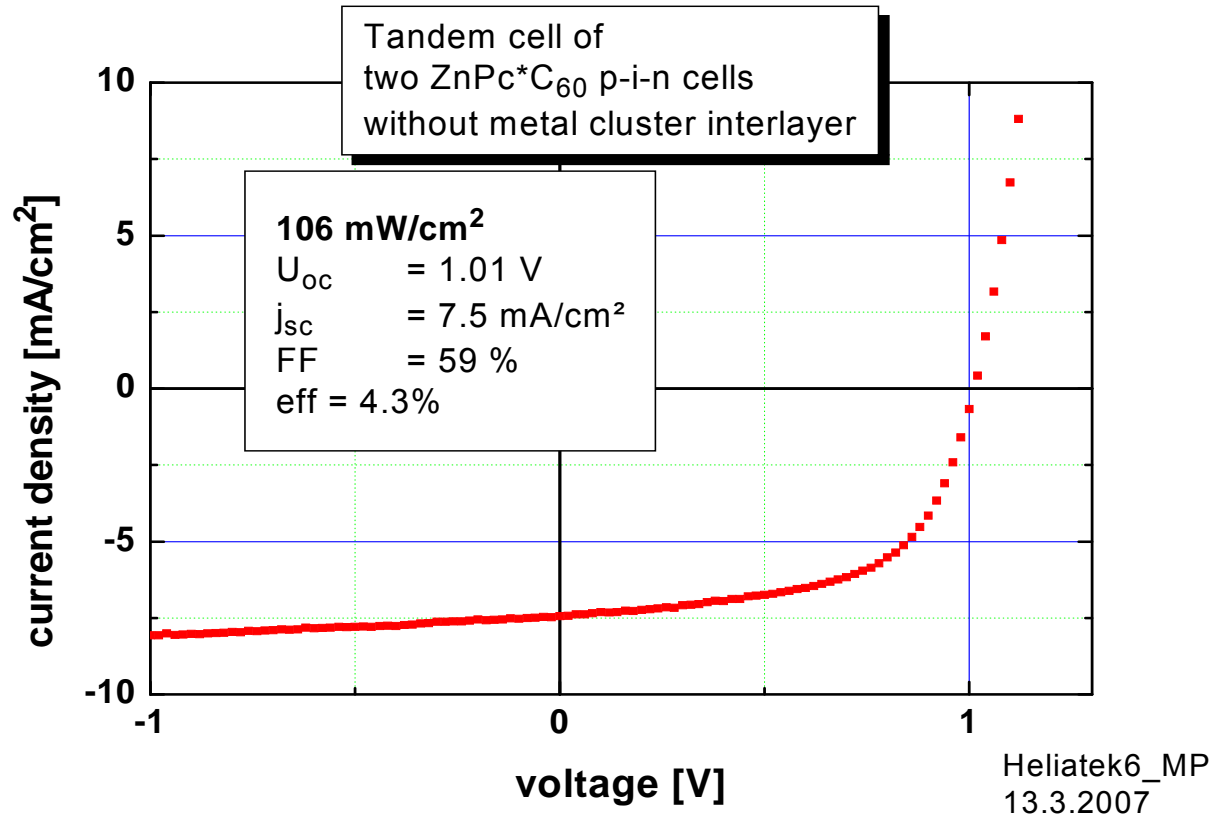
→ Recombination may be loss free even without doping  
cf. Yakimov, Peumans et al.

# Recombination with doping



- Minimum energy loss by alignment of Fermi levels
- Gold clusters not needed
- R. Timmreck et al. unpublished

# ZnPc/C<sub>60</sub> tandem cell (IAPP&Heliatek 2007)



- Interface between subcells without metal clusters
- direct contact between p-doped and n-doped layer

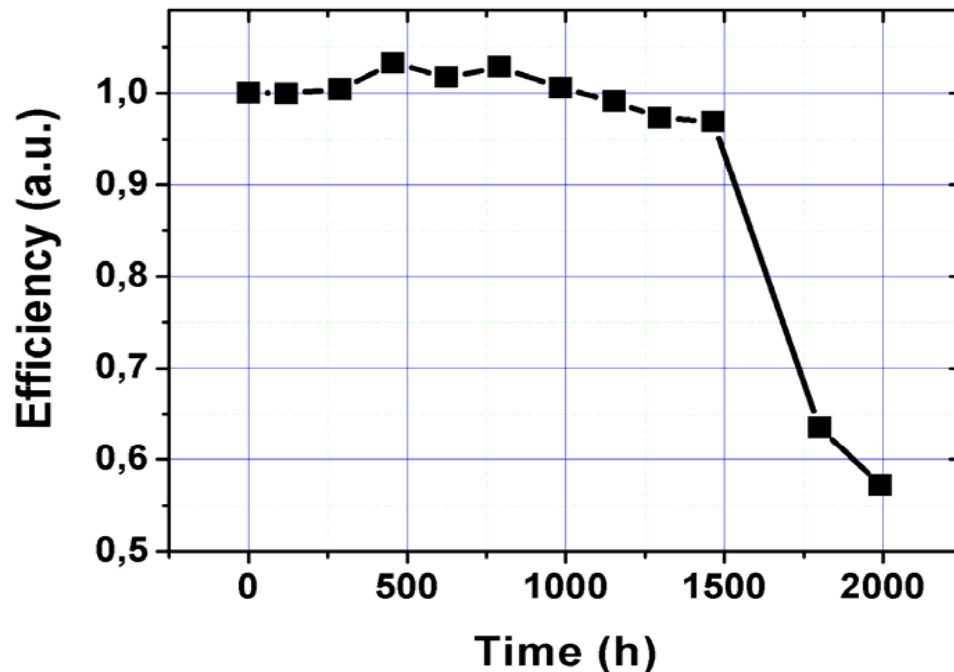
# Outline

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  - **stability**
- Low-cost manufacturing

# Long-term stable tandem cells

Encapsulated sample for **1000h** under halogene lamp at **50°C**;  
intensity corresponds to approx. **2 suns**:

- $V_{oc}$ ,  $j_{sc}$  and saturation factor ( $j(-1V)/j_{sc}$ ) perfectly stable
- FF reduced from 60% to 58%



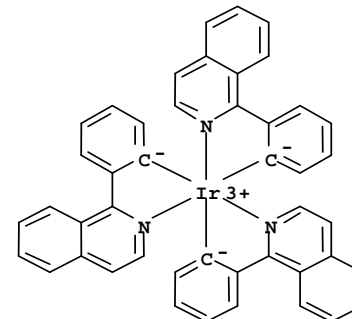
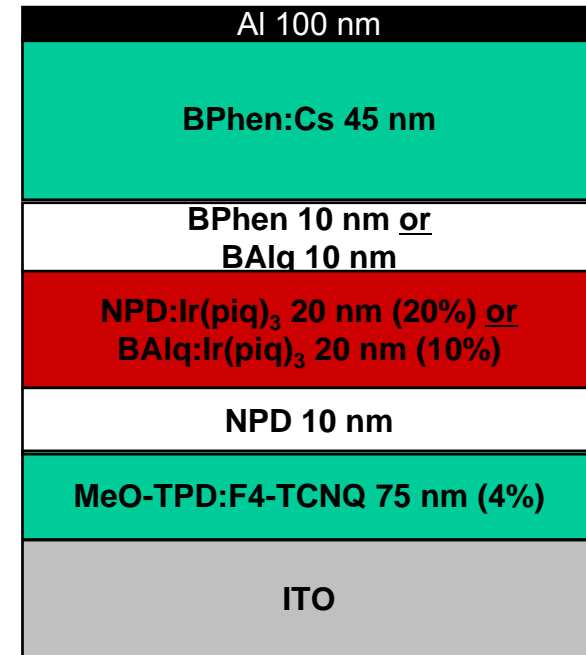
Extrapolated lifetime (80%):  
~ 10 000h  
at 100mW/cm<sup>2</sup>, 50°C

However:  
Rapid degradation beyond 1500h  
together with color change  
in epoxy resin  
→ probably breakdown of  
encapsulation

R. Franke et al. Solar Energy Materials & Solar Cells, in press

# Case Study: Ultra-Stable deep red pin OLED

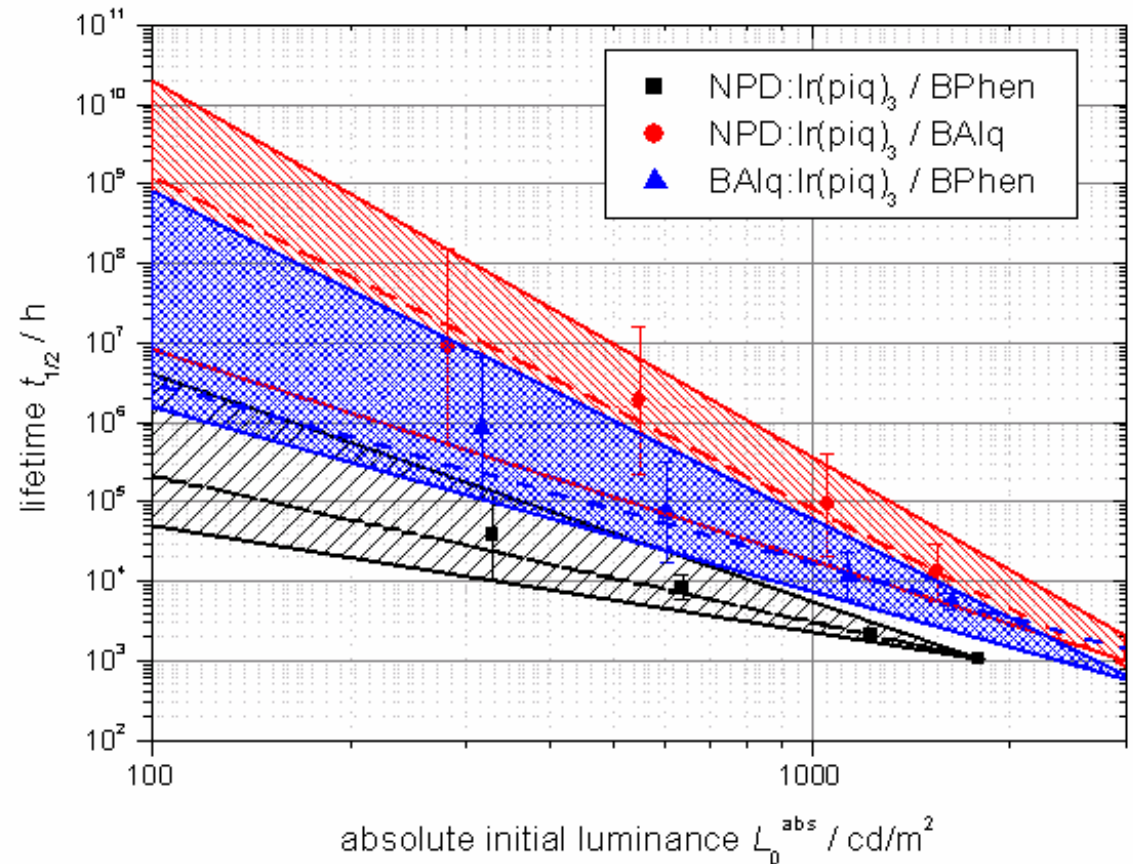
- Phosphorescent red pin OLED using „open“ materials
- Different hosts (NPD, Balq) and blockers (Balq, Bphen)
- All materials very carefully sublimed (Creaphys sublimator)



Ir(piq)<sub>3</sub> deep red  
CIE 0.68, 0.32

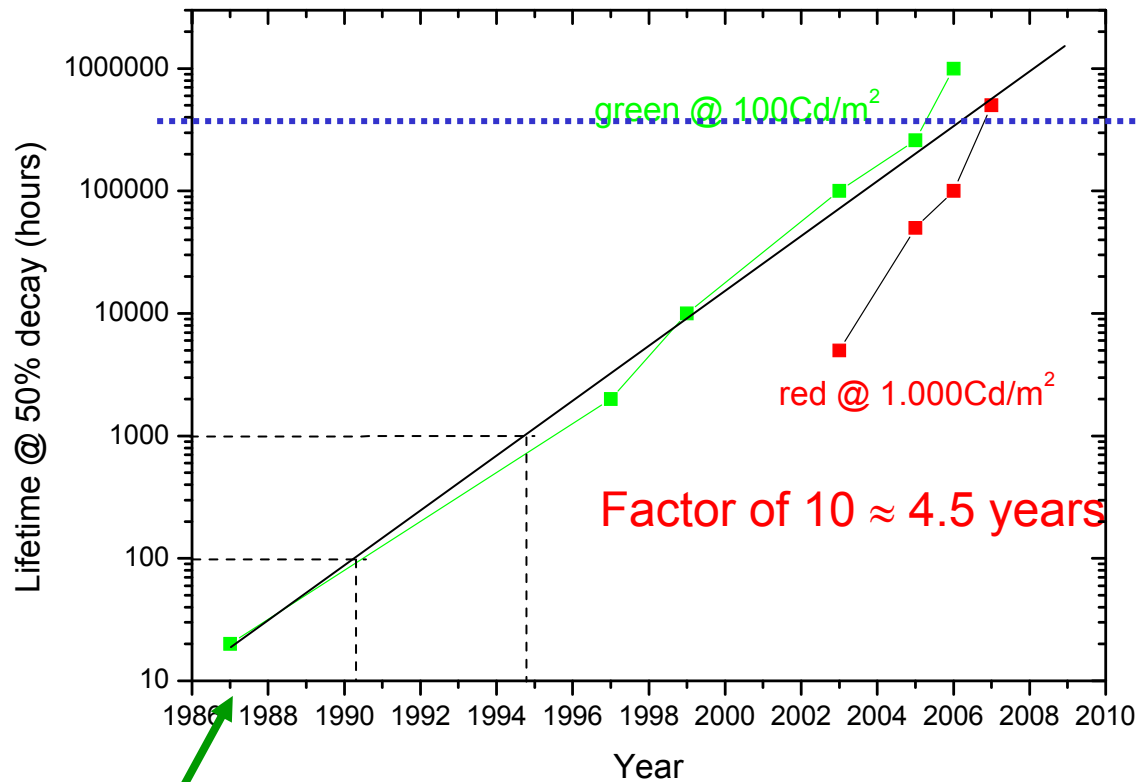
# Ultrastable red OLED: Lifetimes well beyond 10Mhrs @100Cd/m<sup>2</sup>

- Already the lower limits  $\delta \cdot t_{1/2}^{\text{SED}}$  gives **10 million hours** lifetime at 100 cd/m<sup>2</sup>
- One Emitter molecule runs through  $2.4 \cdot 10^{11}$  photocycles
- BAQq/BAQq devices „decay“ **even slower**





# OLED lifetime: It never stops to grow



- Solar cells should do better:
- Lower gaps
- Excitons are separated quickly

Tang & vanSlyke 1987

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  - stability
- **Low-cost manufacturing**

# Helpful: Comparison to Lighting



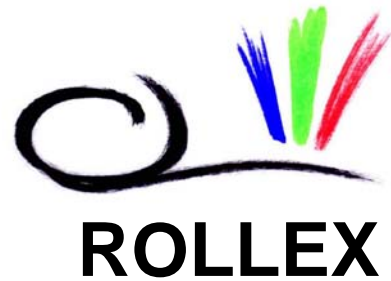
# OLED lighting: Technology Roadmap (USDC)

Property	Units	2004	2007	2010	2013
Energy efficiency	%	5	12.5	20	30
Efficacy	lm/W	20	50	80	120
Color rendering index	CRI	75	80	85	90
Life from 2000 cd/m <sup>2</sup>	hours	10K	20K	40K	50K
Panel width	in	14	40	40	>40
Panel thickness	mm	2.0	1.0	0.5	0.5
Panel weight	gm/cm <sup>2</sup>	0.5	0.25	0.1	0.1
Fabrication costs	\$/sq m	120	60	40	30

30 \$/m<sup>2</sup> = 0.3 Cent/cm<sup>2</sup> ≈ 30 Cent/pWatt !

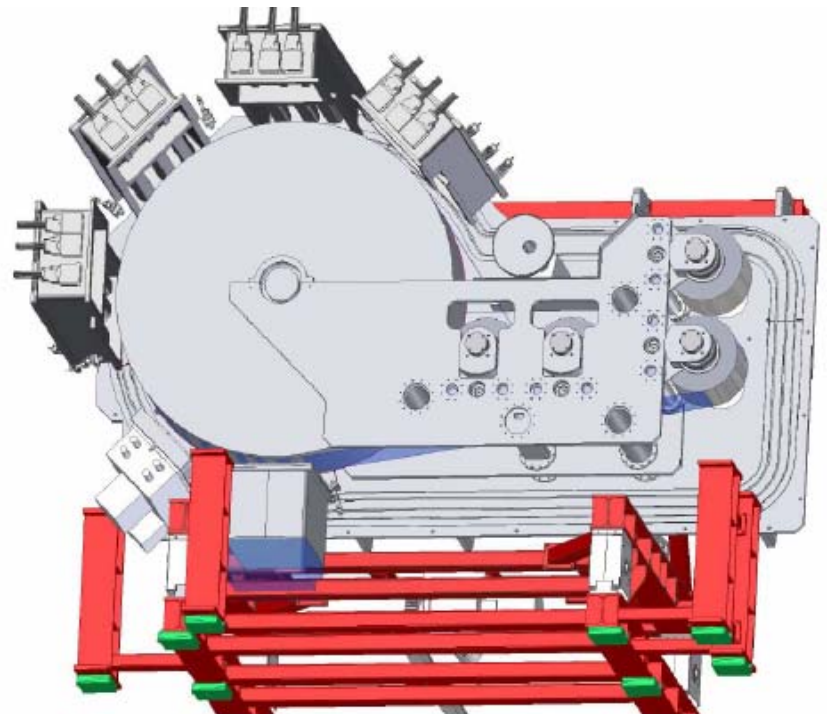
OLED lighting cost ≈ organic solar cell cost !

# New funding project: Roll-to-roll coating for small-molecule OLED

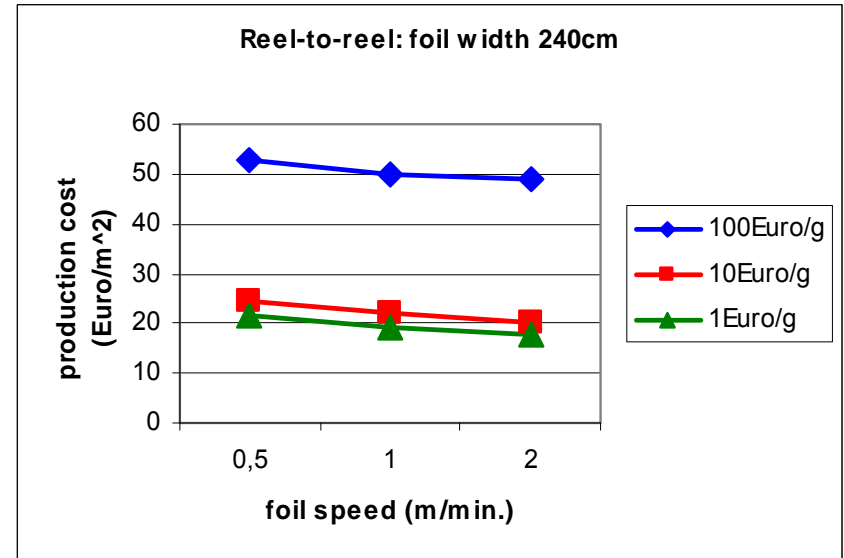
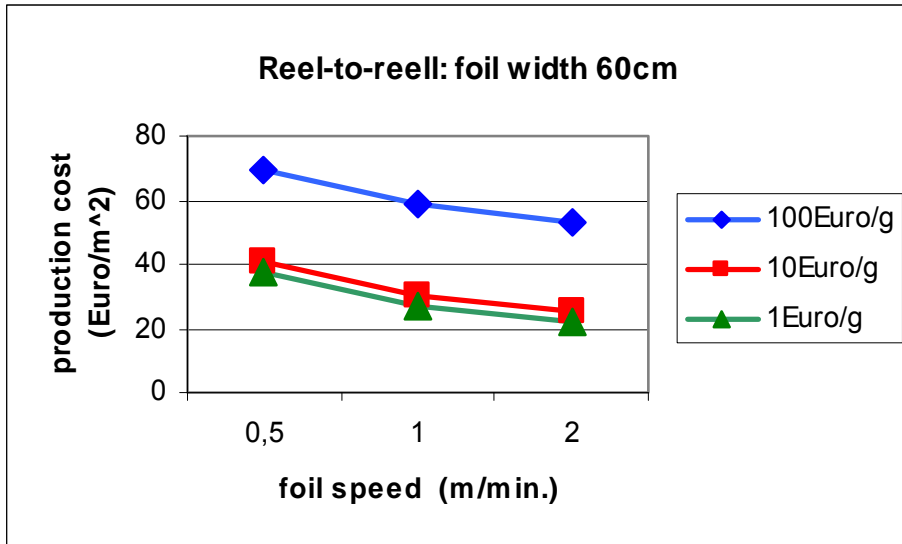


Partners:

- TU Dresden
- FhG-IPMS Dresden
- Novaled
- Laytec
- Von Ardenne
- Philips



# Coating cost for roll-to-roll tool



- Cost of below 20€/m<sup>2</sup> is achievable (materials limited)
- Organic Costs below 10€/g needed
- Cost for Encapsulation/Cathode is critical

# organic valley saxony



- ❖ Organic Valley
- ❖ Partners
- ❖ Organic Value Chain
- ❖ Organic Links
- ❖ Downloads
- ❖ Impressum

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Last update:  
January 07, 2008 17:40:44

## Organic Valley

The region of Dresden is Europe's largest cluster for organic semiconductor R&D and manufacturing. More than 10 companies with over 400 employees (2008) are active in this exciting and quickly growing new technology. This website is meant to give an overview about the topics. Further information may be obtained from the listed partners or directly from [Prof. Dr. Karl Leo](#).

Enjoy browsing this site!



# Organic Value Chain in Saxony

[www.organic-valley.org](http://www.organic-valley.org)

R&D



Materials  
Modeling

OLED-Technol.

Tools

Products

Industry



VON ARDENNE



More than 500 people in 2008!





# Conclusions

Small-molecule organic solar cells are a promising technology, but:

- Efficiency way too low to achieve broad application
- Materials basis still extremely narrow
- Bulk heterojunction morphology is a challenge
- Low-cost manufacturing technologies possible

# Acknowledgment

- G. Schwartz, K. Fehse, Q. Huang, S. Reineke, R. Schüppel, F. Lindner, K. Walzer, G. He, X. Zhou, J. Huang, A. Werner, J. Drechsel, F. Kozlowski, K. Schulze, C. Urich, R. Lessmann, T. Müller, J. Meiss, K. Harada, M.K. Riede, T. Fritz (IAPP)
- J. Amelung, C. May, M. Eritt, C. Kirchhof, M. Schreil, M. Toerker, Y. Tomita, M. Hoffmann, U. Todt (FhG-IPMS)
- J. Blochwitz-Nimoth, J. Birnstock, S. Murano, M. Vehse, M. Burghard, M. Hofmann, G. Sorin (Novaled)
- M. Pfeiffer, B. Männig (Heliatek)
  
- E. Brier, E. Reinold, P. Bäuerle (Ulm)
- U. Zokhavets, H. Hoppe, G. Gobsch
- D. Alloway, P.A. Lee, N. Armstrong, Tucson (XPS/UPS)
- N. Karl (Stuttgart)
- A. Hinsch, A. Gombert (ISE)
- D. Wöhrle (Bremen), J. Salbeck (Kassel), H. Hartmann (Merseburg/Dresden)
- C.J. Bloom, M. K. Elliott (CSU)
- W. Lövenich, A. Elschner (HCStarck)
- OLLA consortium
- BMBF, SMWA, SMWK, DFG, EC, FCI, NEDO

# Acknowledgment

