Electrical and Optical Simulation of Organic Light-Emitting Diodes and White Organic Light-Emitting Diodes

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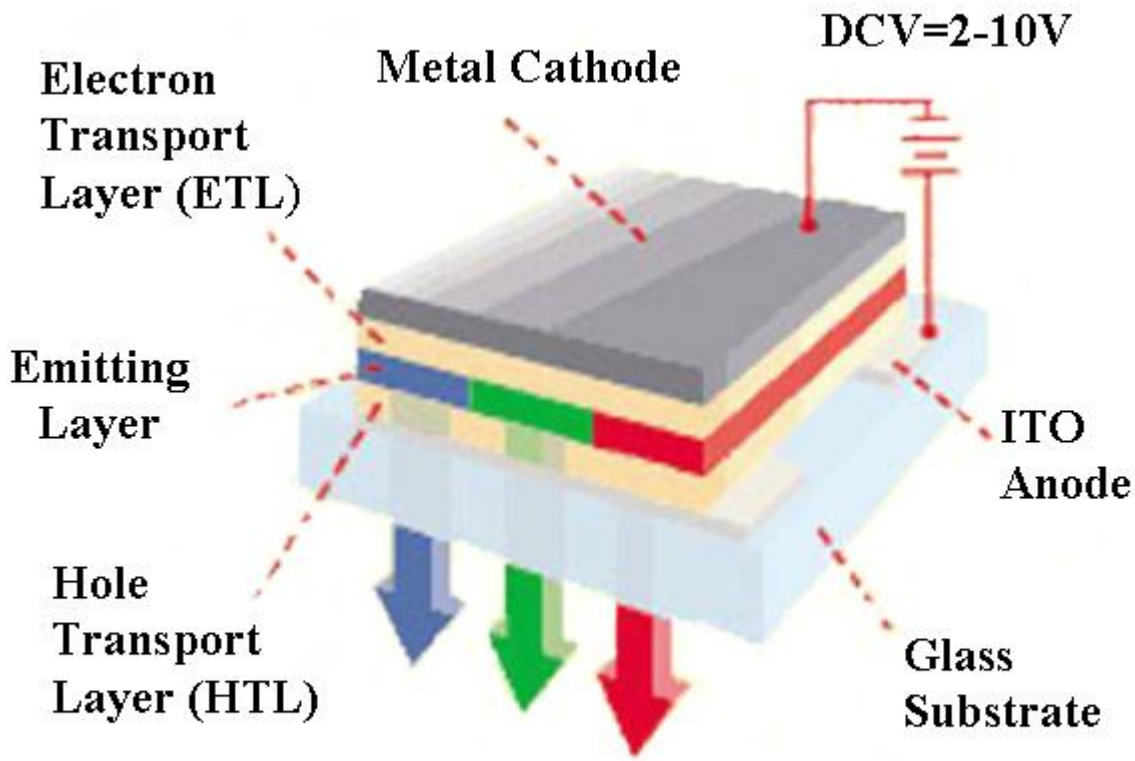
Contents

- **Treating organic semiconductor with quantum drift-diffusion model.**
- **Light extraction using 3D ray-tracing (non-microcavity model).**
- **Electroluminescent spectra modeling with Frenkel exciton.**
- **Inclusion of microcavity effects.**
- **Application for AMOLED.**
- **Triplet diffusion and WOLED**
- **Low-voltage PIN structures**
- **Tandem OLED**

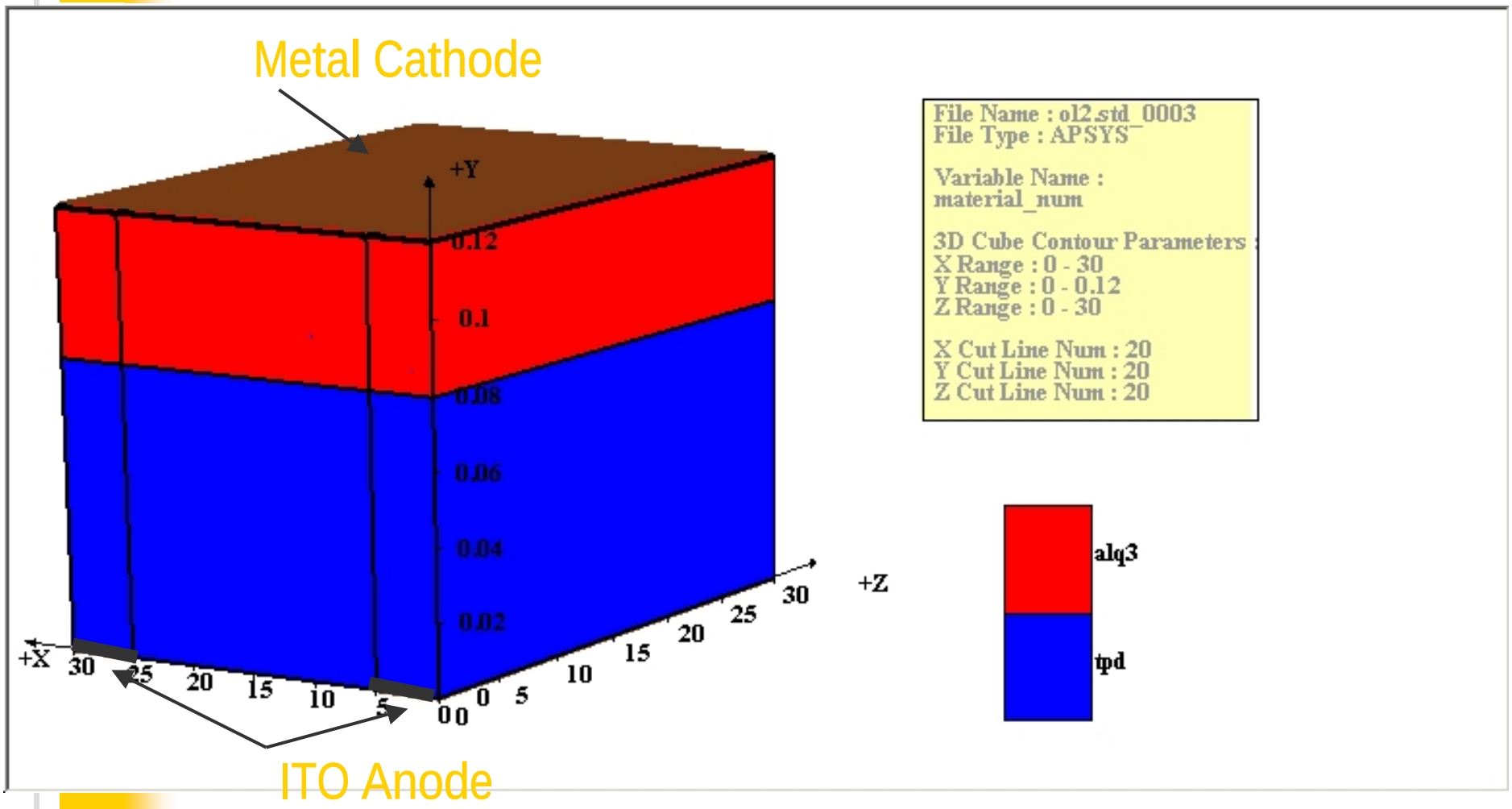
Quantum drift-diffusion for OLED

- Quantum drift-diffusion model for current flow/spreading.
- Poole-Frenkel field dependent mobility model.
- Mobility-dependent bi-molecular radiative recombination model.
- Quantum tunneling model for Schottky barriers and heterojunctions.
- Self-heating model.
- Quantum wells?

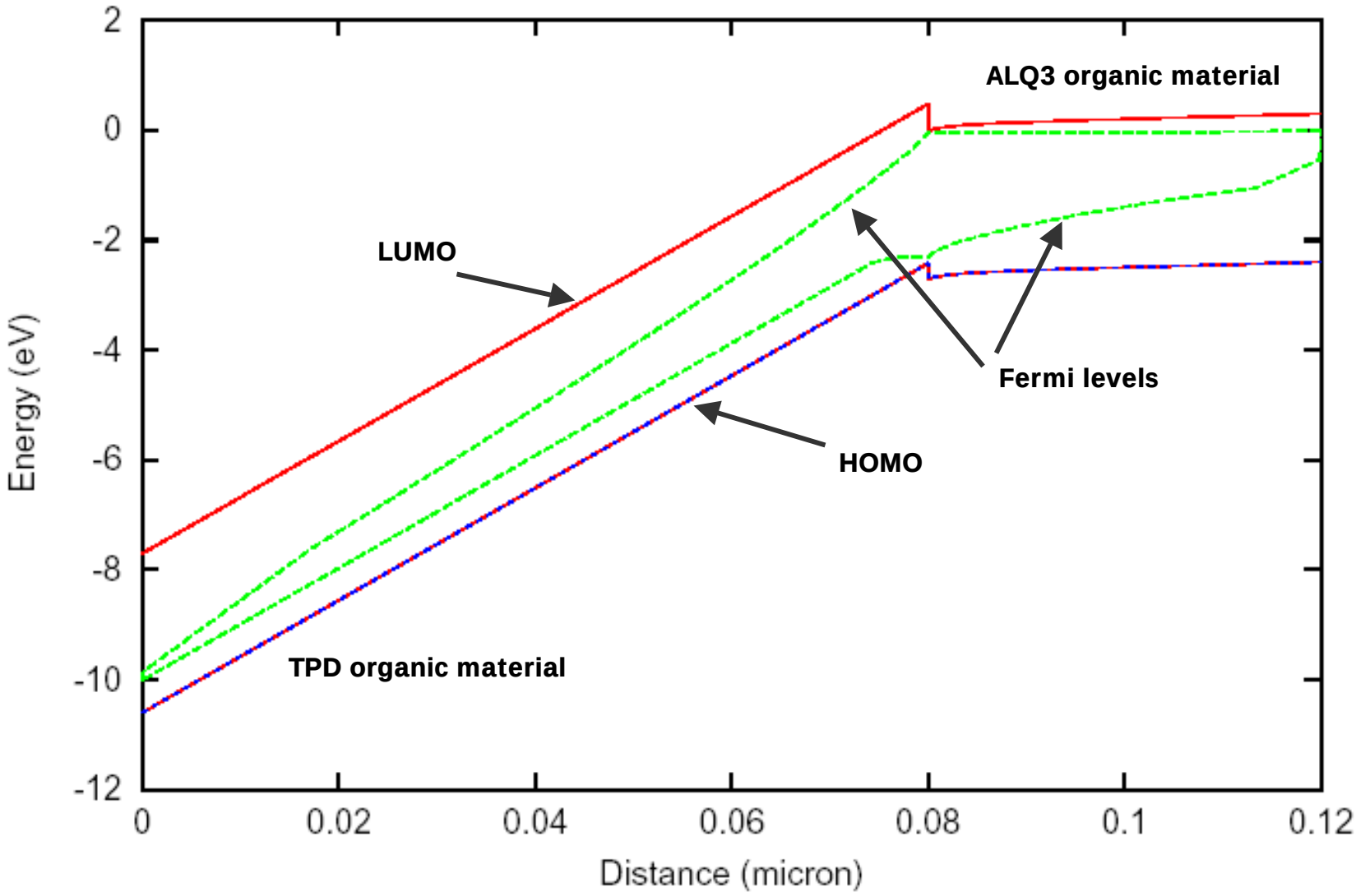
An Example


DCV=2-10V
Metal Cathode
Electron
Transport
Layer (ETL)
Emitting
Layer
ITO
Anode
Hole
Transport
Layer (HTL)
Glass
Substrate

Coordinates

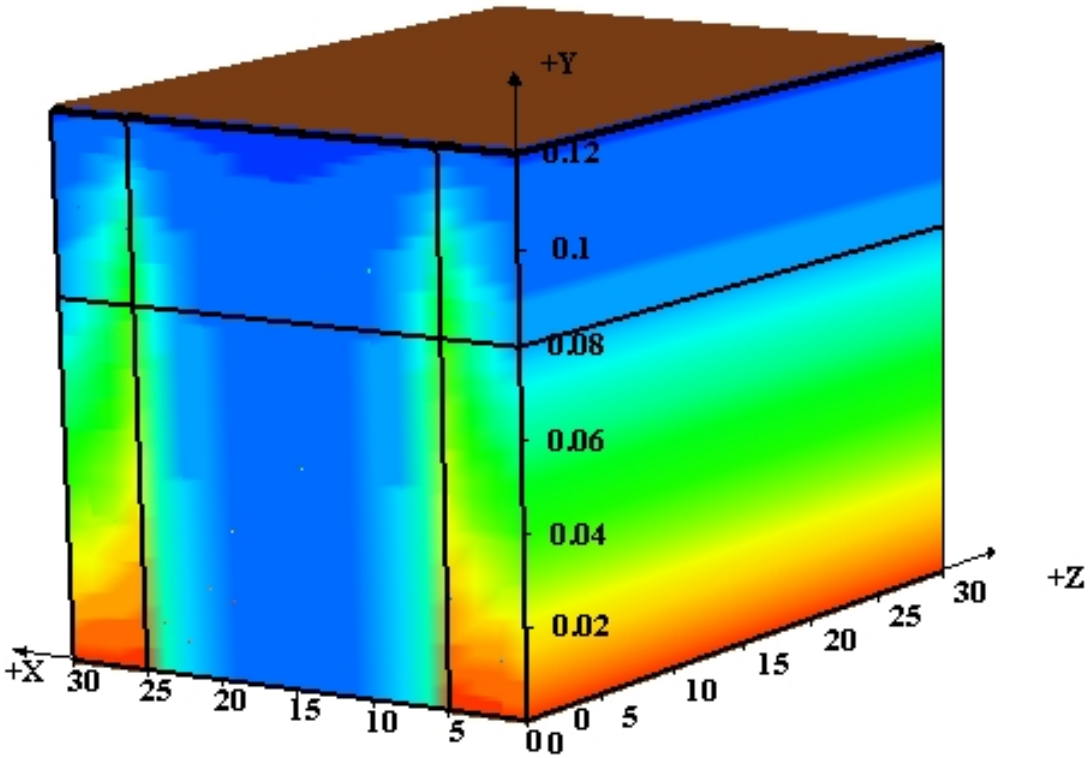


Band Diagram



Energy band diagram of a OLED of TPD / ALQ3

3D Potential Distribution

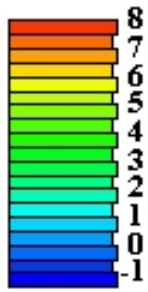


File Name : ol2.std_0003
File Type : APSYS

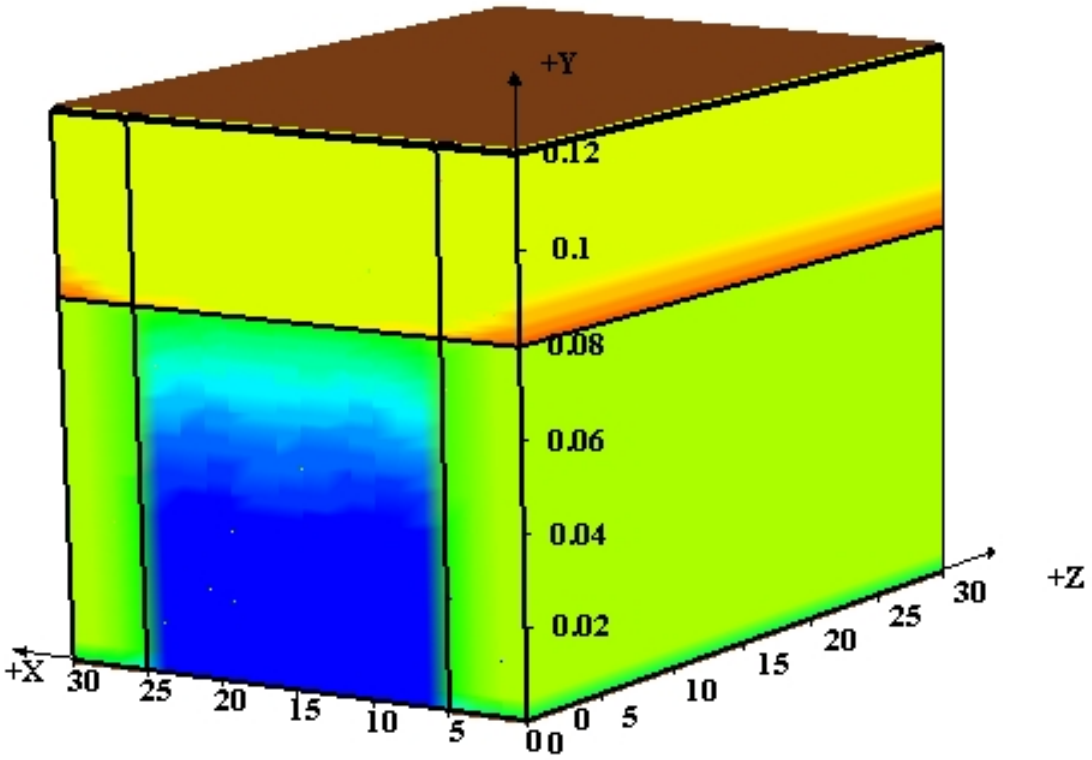
Variable Name :
potential

3D Cube Contour Parameters :
X Range : 0 - 30
Y Range : 0 - 0.12
Z Range : 0 - 30

X Cut Line Num : 20
Y Cut Line Num : 20
Z Cut Line Num : 20



3D Electron Density

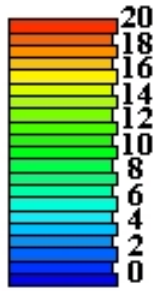


File Name : ol2.std_0003
File Type : APSYS

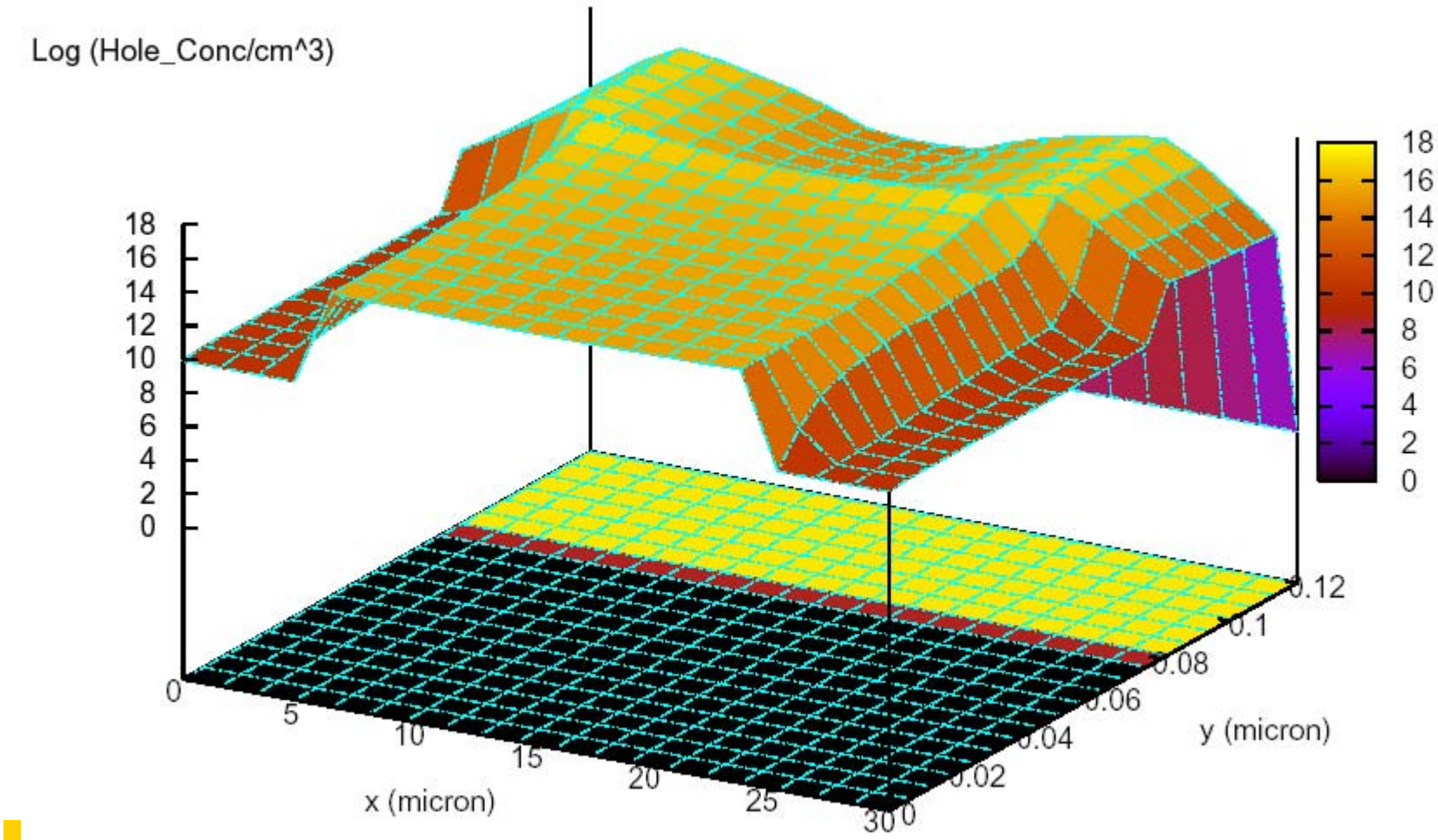
Variable Name :
elec_conc

3D Cube Contour Parameters :
X Range : 0 - 30
Y Range : 0 - 0.12
Z Range : 0 - 30

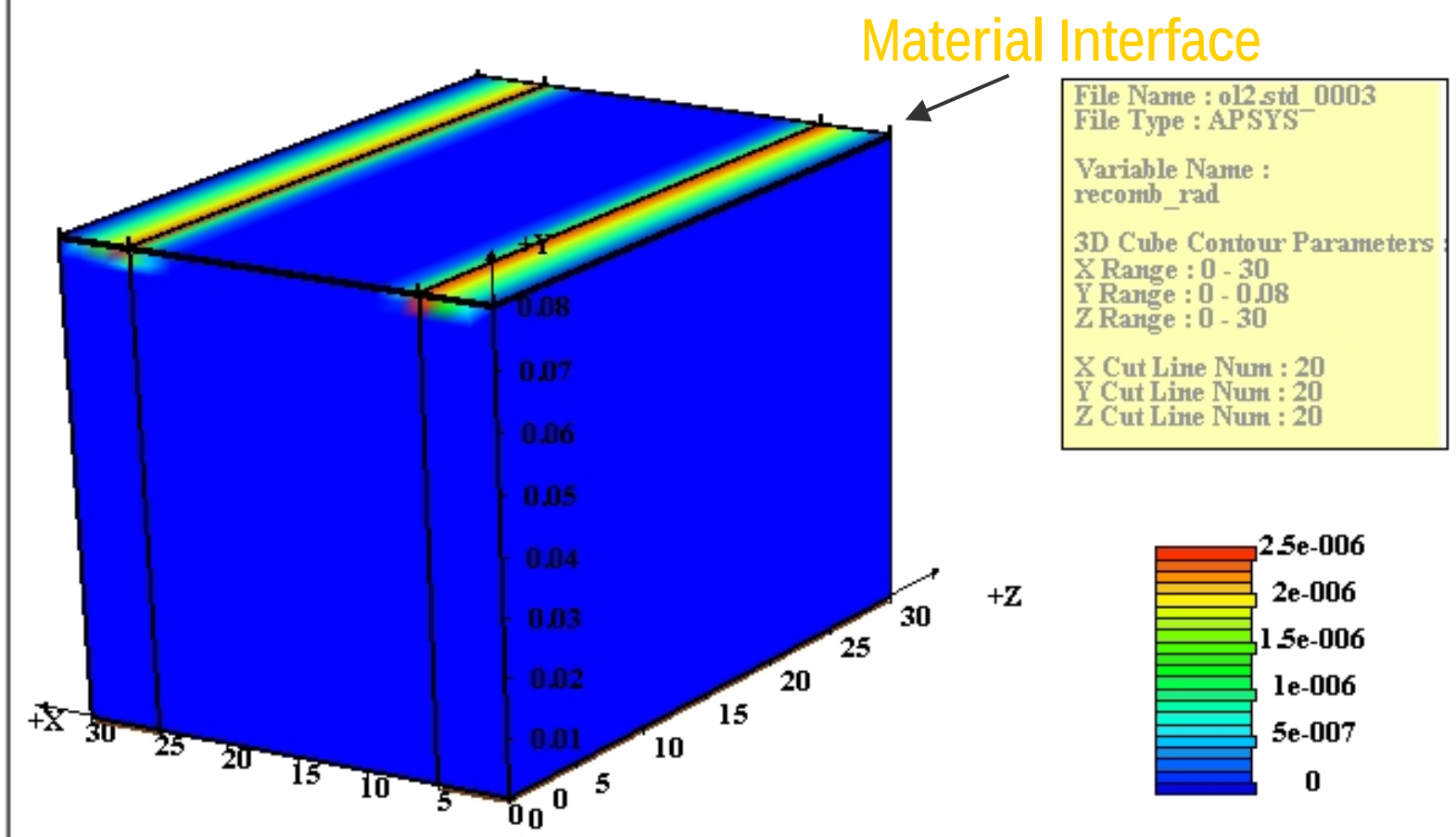
X Cut Line Num : 20
Y Cut Line Num : 20
Z Cut Line Num : 20



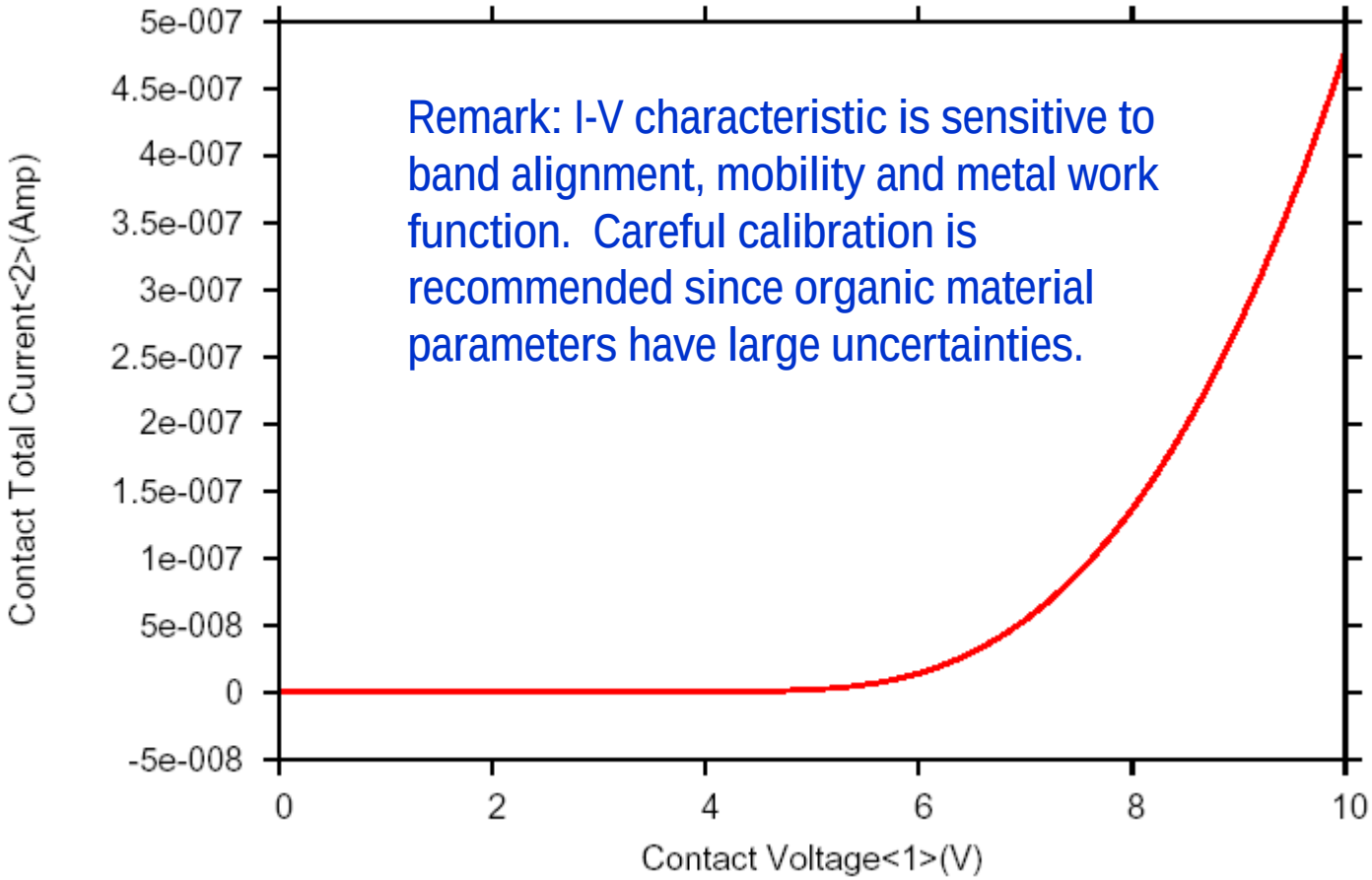
Hole Density



3D Radiative Light Source



I-V Curve


Contact Total Current<2>(Amp)
5e-007
4.5e-007
4e-007
3.5e-007
3e-007
2.5e-007
2e-007
1.5e-007
1e-007
5e-008
0
-5e-008
0
2
4
6
8
10
Contact Voltage<1>(V)
Remark: I-V characteristic is sensitive to band alignment, mobility and metal work function. Careful calibration is recommended since organic material parameters have large uncertainties.

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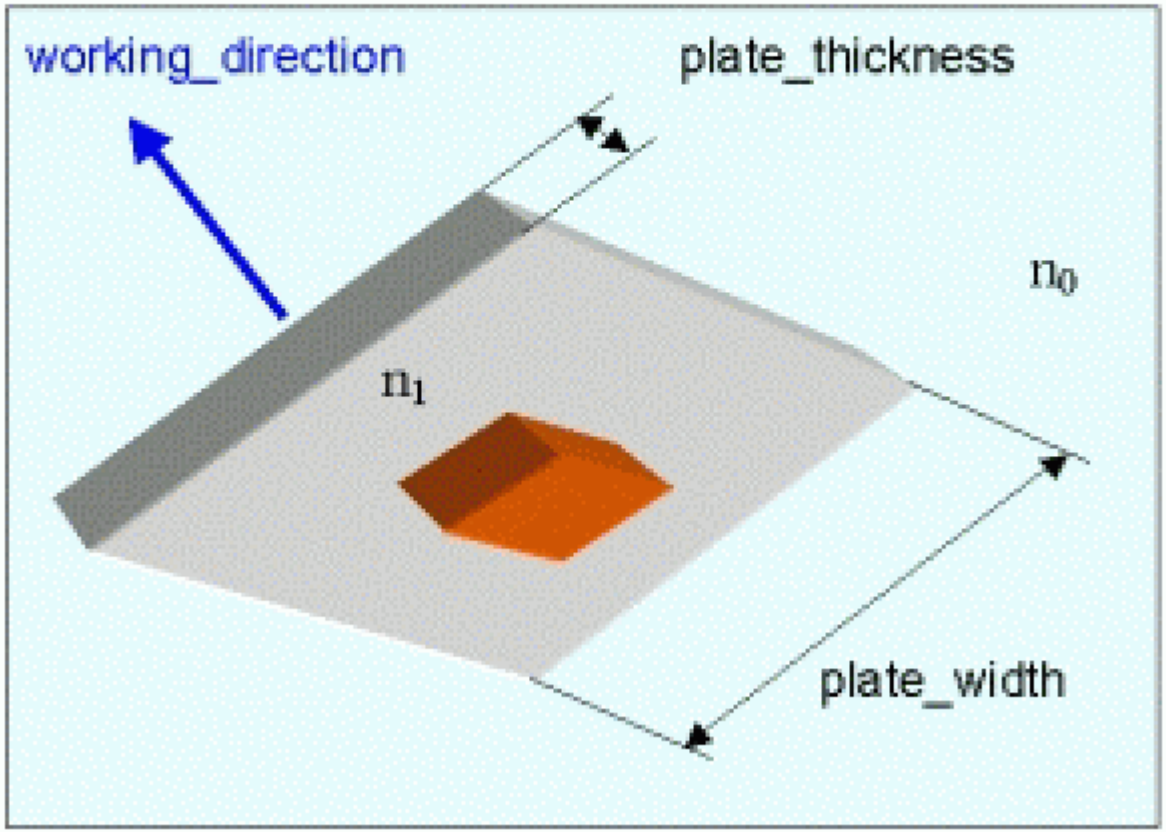
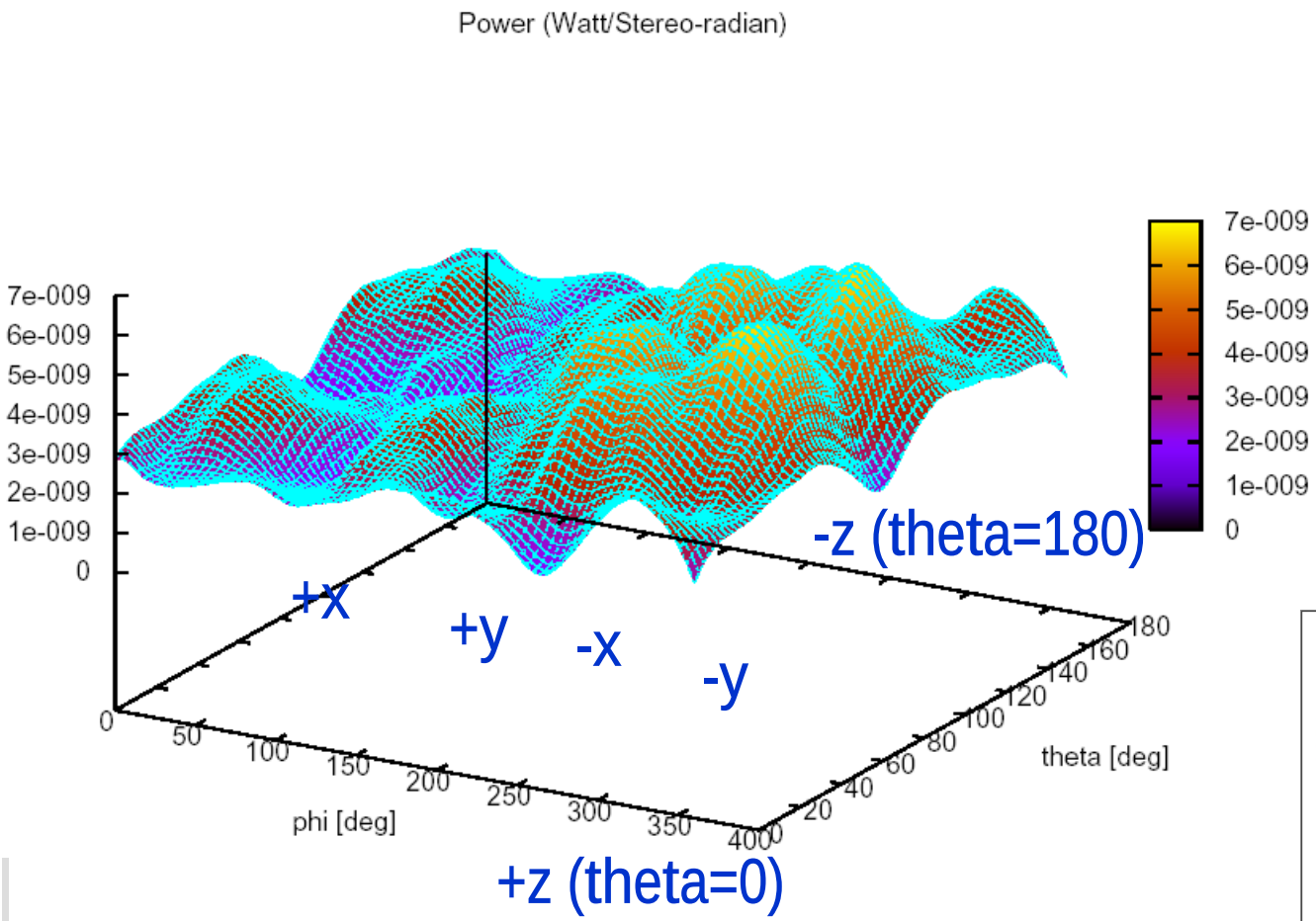
3D Ray-tracing with glass plate

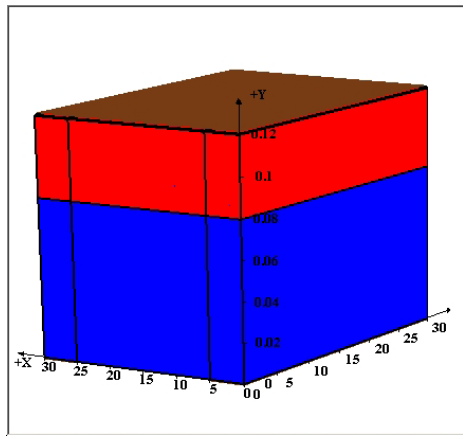
external_package=**plate**

n_0 – space_refr_index; n_1 – package_refr_index



3D Raytrace Power





Angular distribution of emitted power from the OLED with glass plate at the bottom in a 3D simulation ➔ -y emission is clearly favored.

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Simulation statistics

TRANSMITTED POWER in absolute and relative to total (%) units THROUGH

-y side:

1.189041490400453E-008 26.159221035142593

+y side:

8.326234960594208E-010 1.831793276207384

-x side:

8.134143692337187E-009 17.895326992147012

+x side:

8.066635157261732E-009 17.746806464894195

-z side:

8.265096296359655E-009 18.183426115803123

+z side:

8.265096296359703E-009 18.183426115803226

➔Glass substrate helps extract power

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Frenkel Exciton

- Organic semiconductor emits light via Frenkel exciton recombination.
- Conventional semiconductor theories based on free-carrier / many-body interband transition no longer valid.
- To establish optical spectrum model based on exciton-phonon interaction within an organic crystal.

Hamiltonian

- Hamiltonian established to include intra-molecular and inter-molecular electronic and vibronic interactions.
- A phonon cloud of several unit cells is used to represent exciton-phonon interaction.
- All excited states of the molecular crystal are solved and optical transition dipole moments computed between all states.
- Model similar to Refs: [1] M. Hoffmann and Z. G. Soos, Phys. Rev. B 66, 024305 (2002)[2]Vragovic, R. Scholz and M. Schreiber, Europhysics Letters,vol. 57 (2), pp. 288-294, 2002.

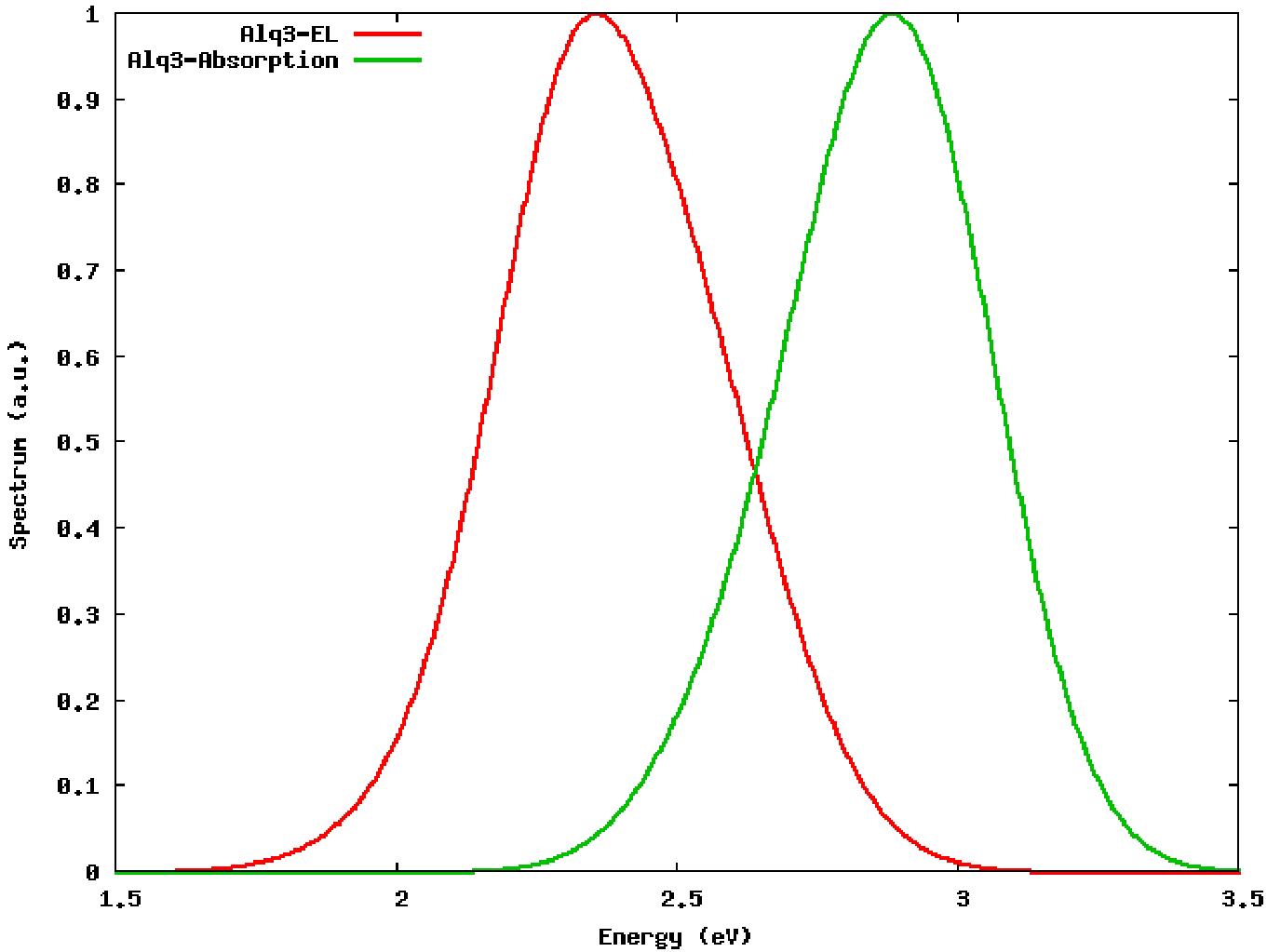
Modeling Procedures

- Although mechanisms involved in the Hamiltonian are complex, input parameters are few and fitting to experimental spectrum is easy.
- Typical input/adjustable parameters: exciton bandgap, molecular vibronic quanta, intra-molecular exciton-phonon interaction constant, and inter-molecular hopping parameter.
- Doped organic semiconductor may easily be modeled as combination of emission from the host and the dopant separately.

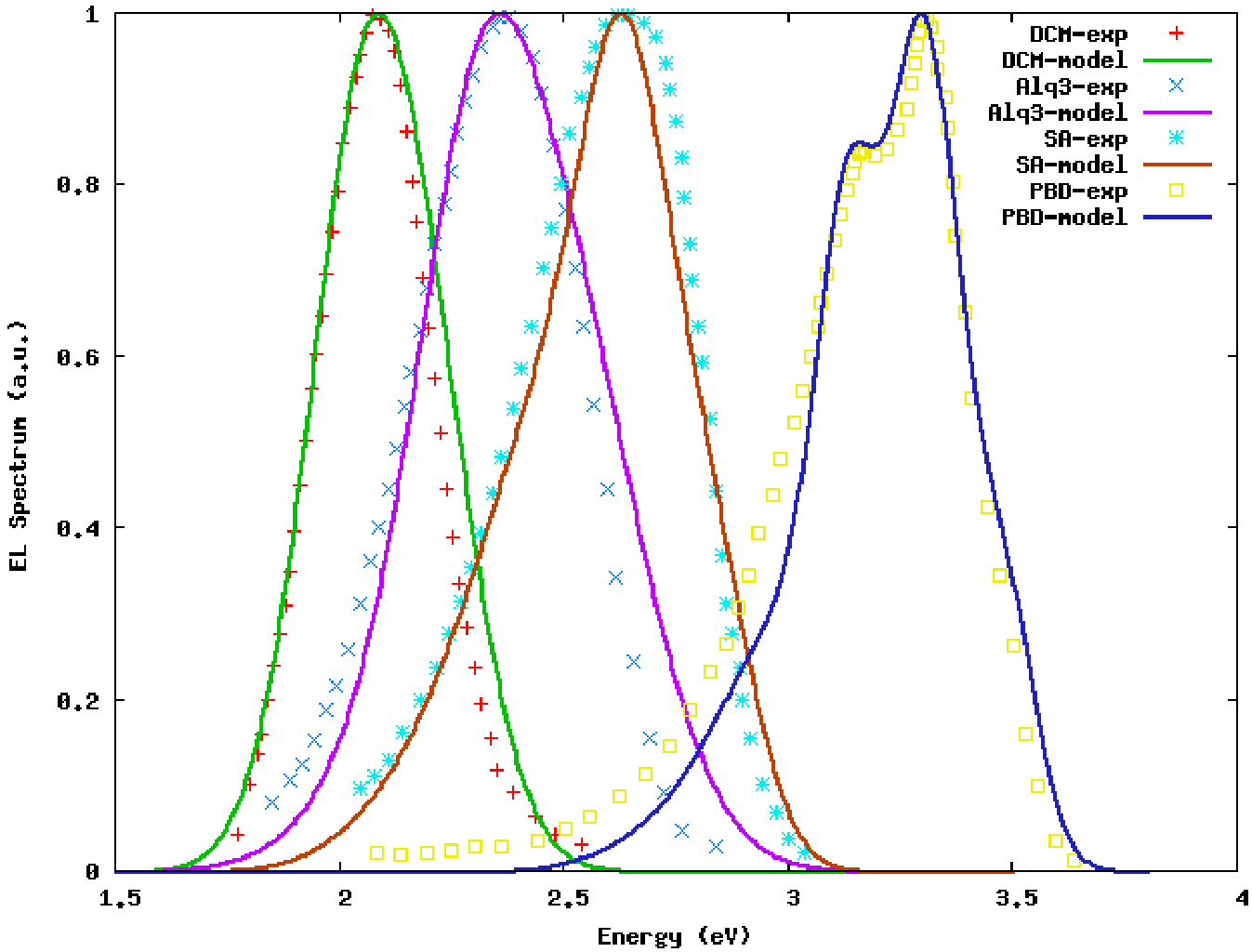
Numerics

- Numerically efficient: several minutes per spectrum.
- Integrated into APSYS-OLED option so that optical extraction may be calculated based on the spectrum.
- Bias and current injection dependent spectrum may be simulated to fine tune the color of the OLED.

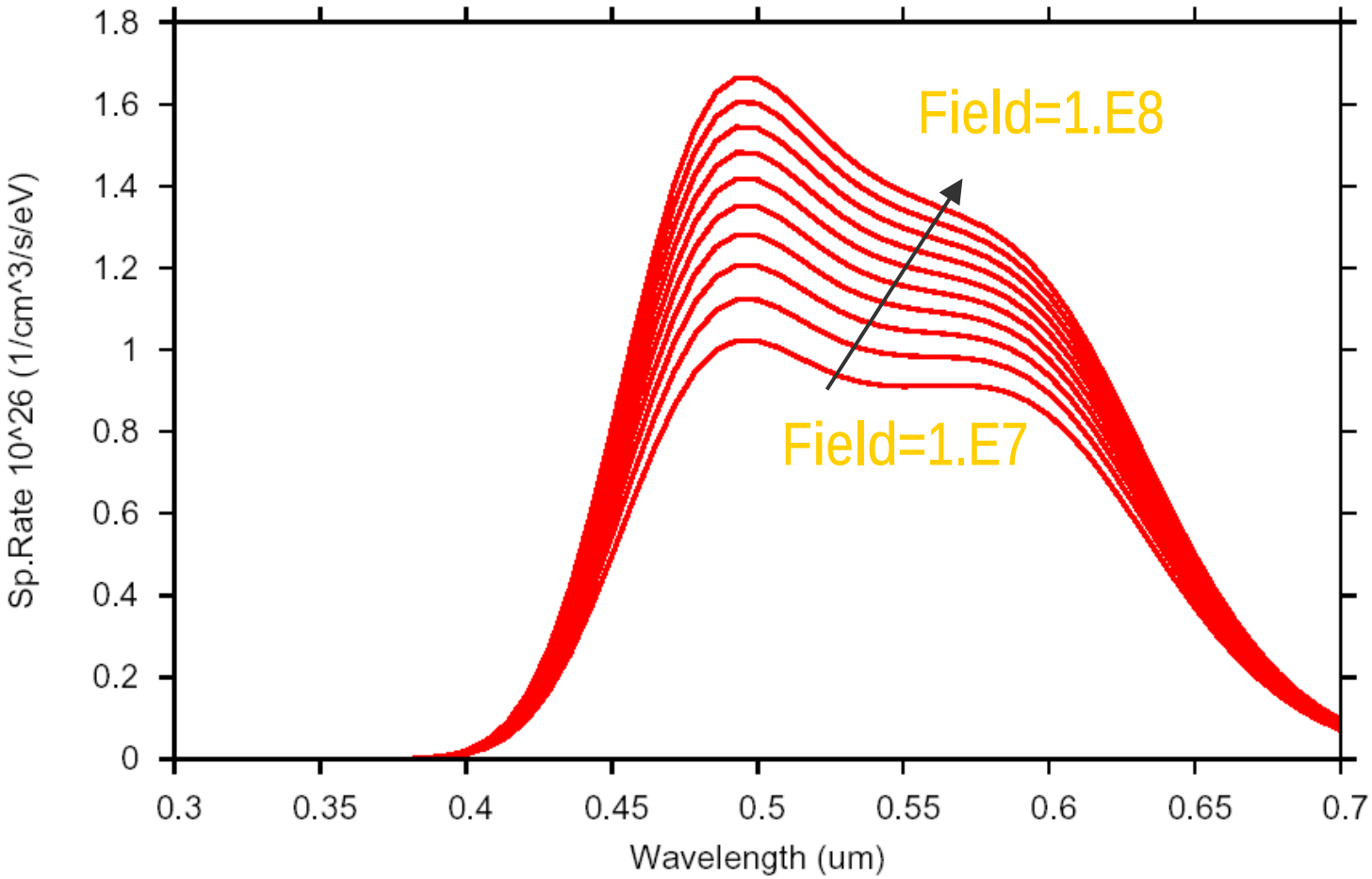
Typical EL/Absorption Spectra



Some Fitted EL Spectra



EL Spectrum Model of Alq3:DCM



Applied electric field ranges from
1.E7 to 1.E8 V/m

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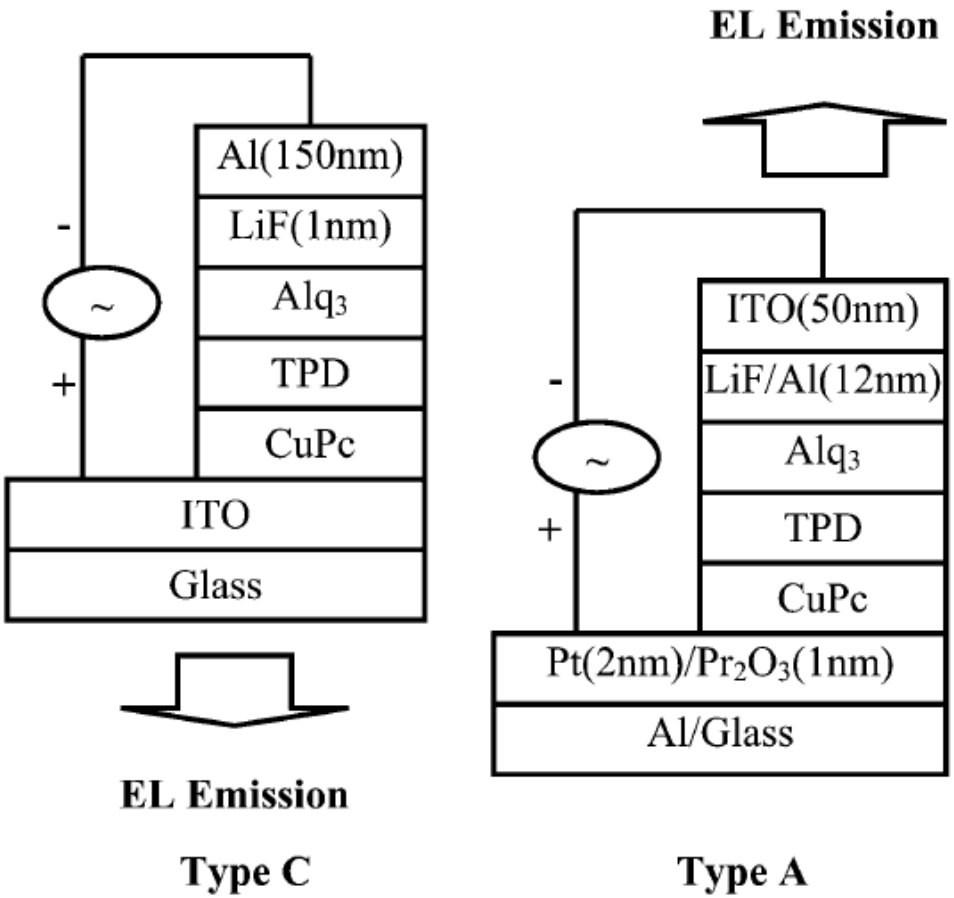
Theoretical Basis

- **Self-consistent calculation of material spontaneous emission rate based on rigorous Frenkel exciton theories coupled with 2/3D simulation of current injection from the Crosslight APSYS drift-diffusion solver.**
- **Coupling of spontaneous emission with microcavity modes based on theory of C. H. Henry (1986) [1].**
- **Henry's theory has been extended from waveguide to RC-OLED by proper accounting of mode densities in a quasi-2D/3D emission situation.**
- **Photon recycling effects taken into account by accurate determination of photon power density inside the RCLED and self-consistent model of material absorption of the active layers in OLED.**

[1] C. H. Henry, "Theory of spontaneous emission noise
in open resonators and its application to lasers and optical amplifiers,"
J. Lightwave Technol., vol. LT-4, pp. 288--297, March 1986.

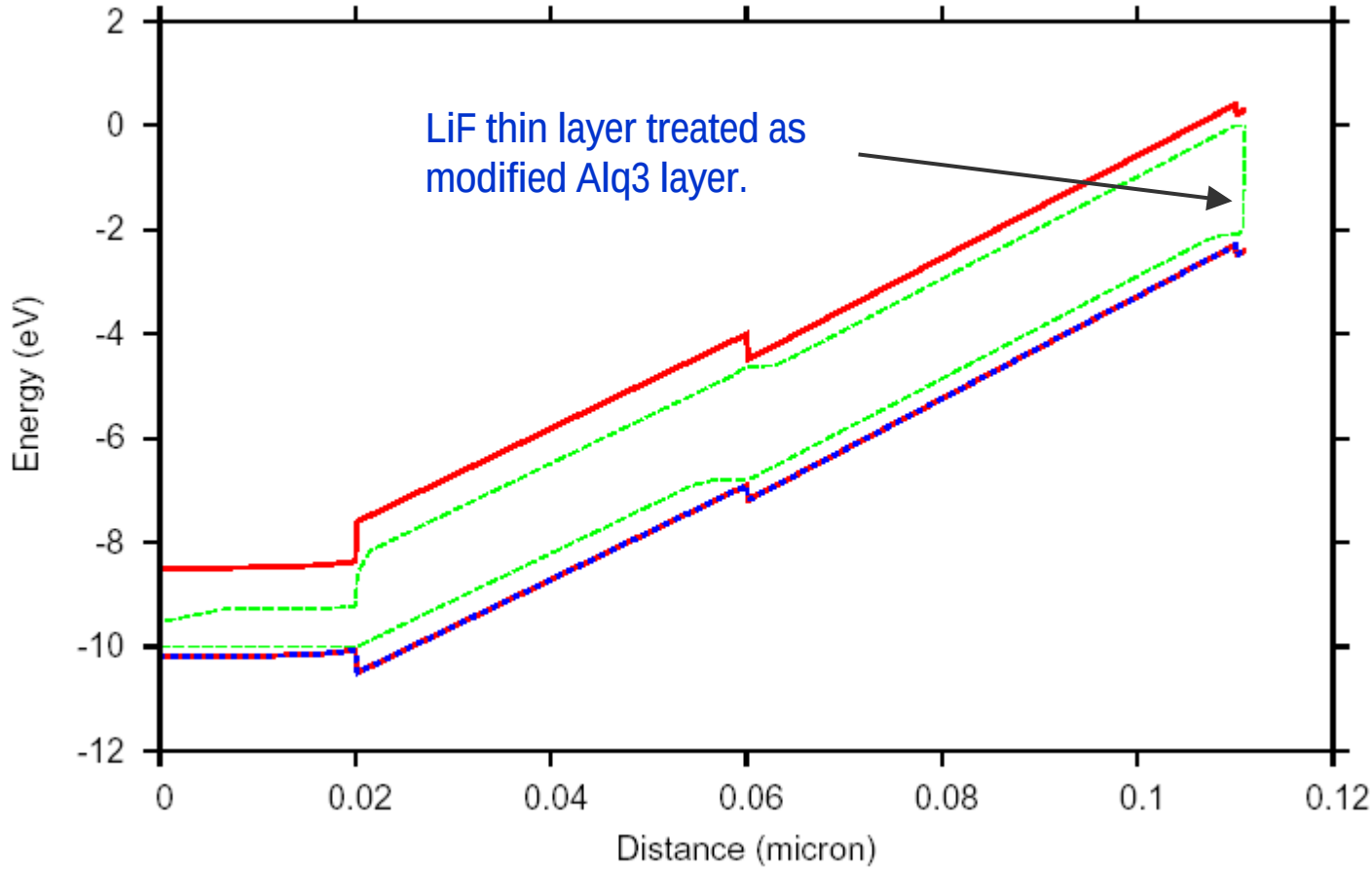
An Example of RC-OLED



Structure according to C. Qiu, et. al. IEEE Trans. on Elec. Device, vol. 51,2004 p. 1207.

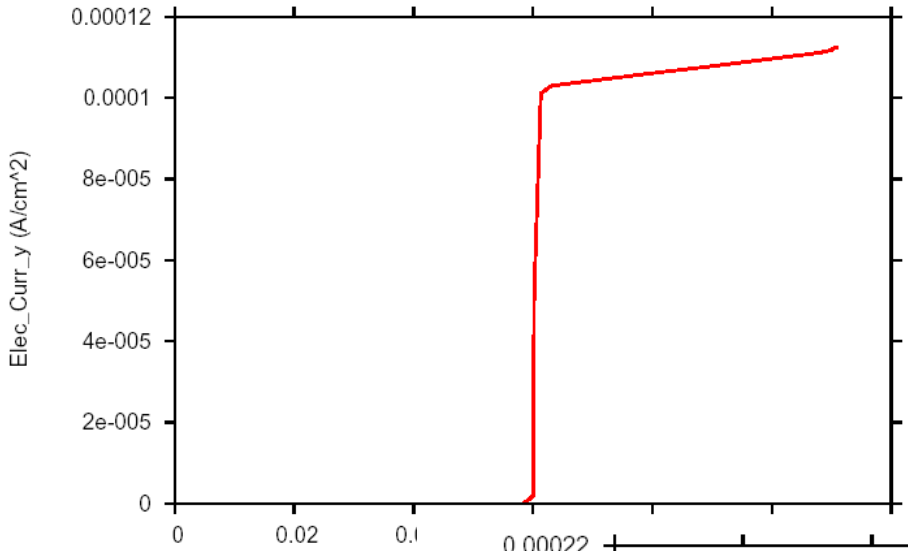
Strong microcavity effects reported for Type A.

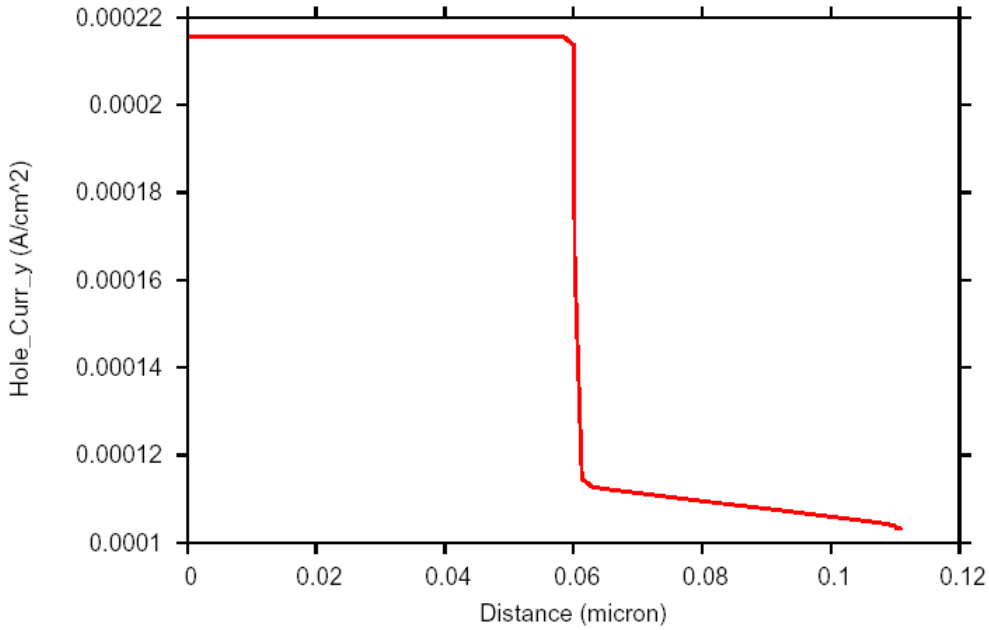
Simulated band diagram



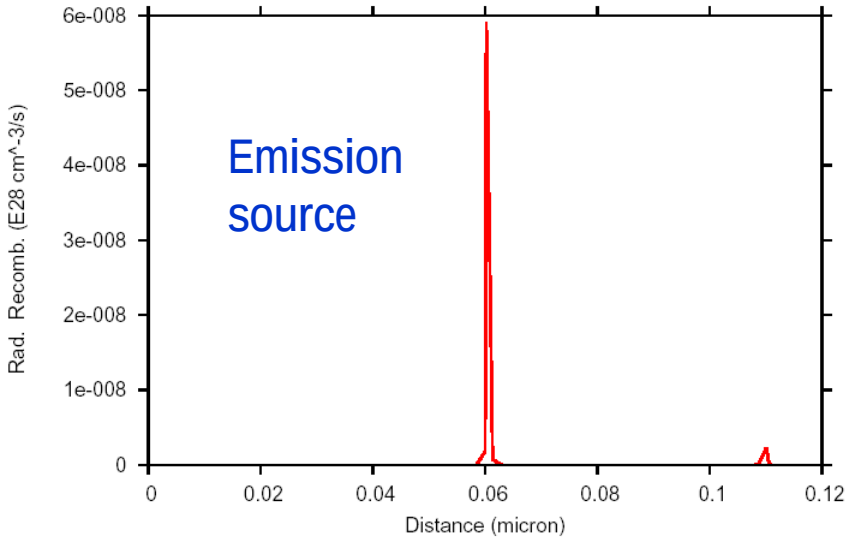
Contact layers are included in optical simulation but ignored in electrical simulation for simplicity.

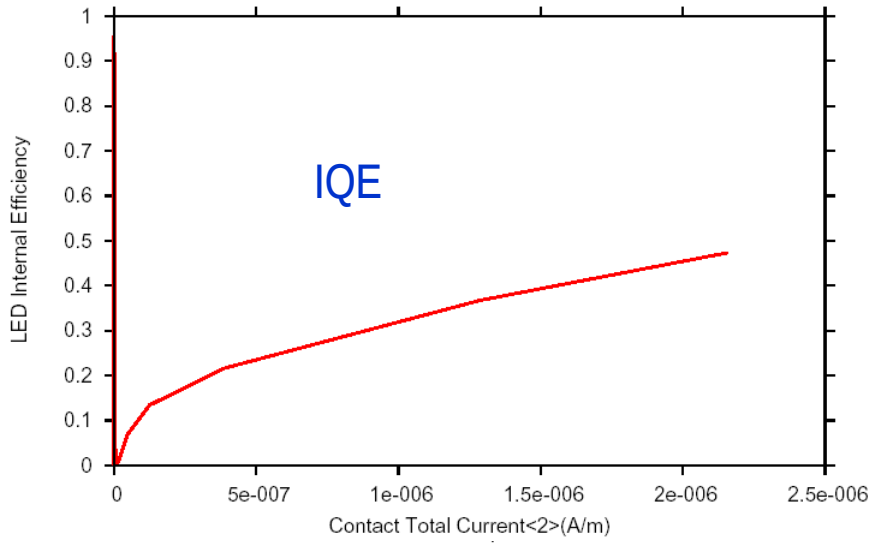
Leakage currents

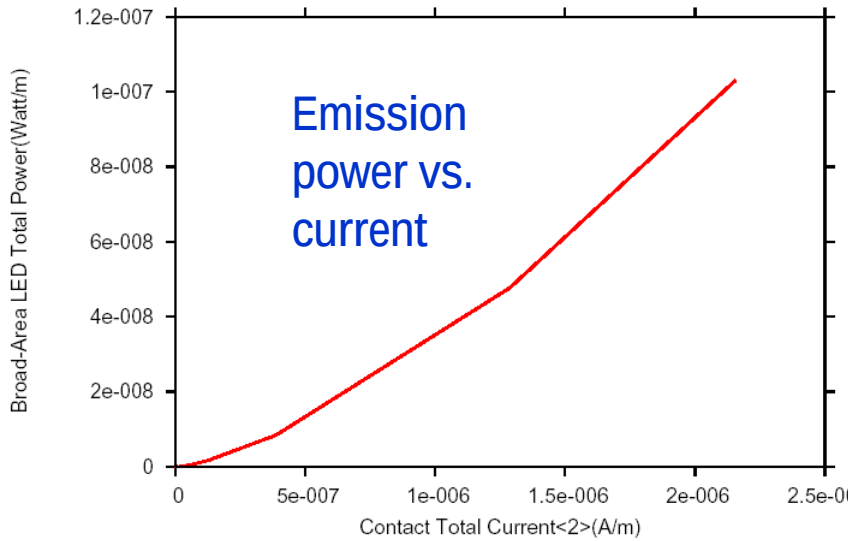


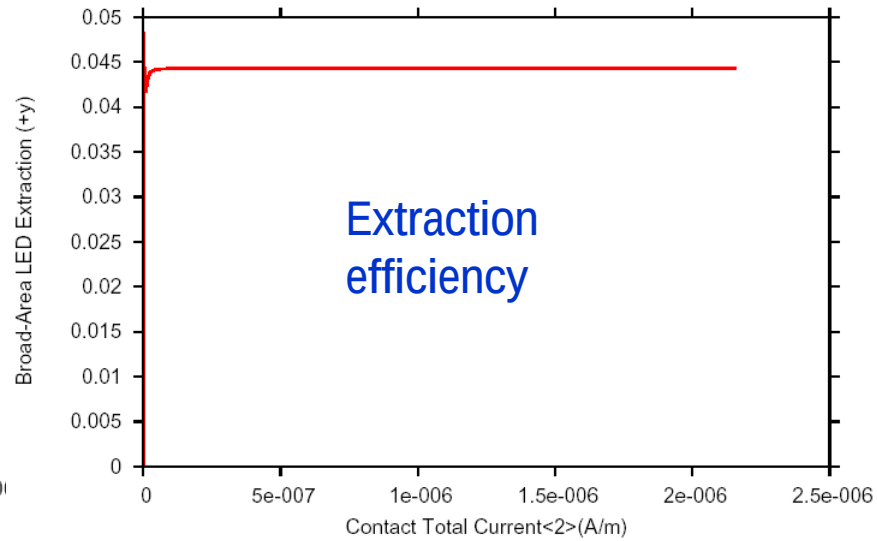


Performance

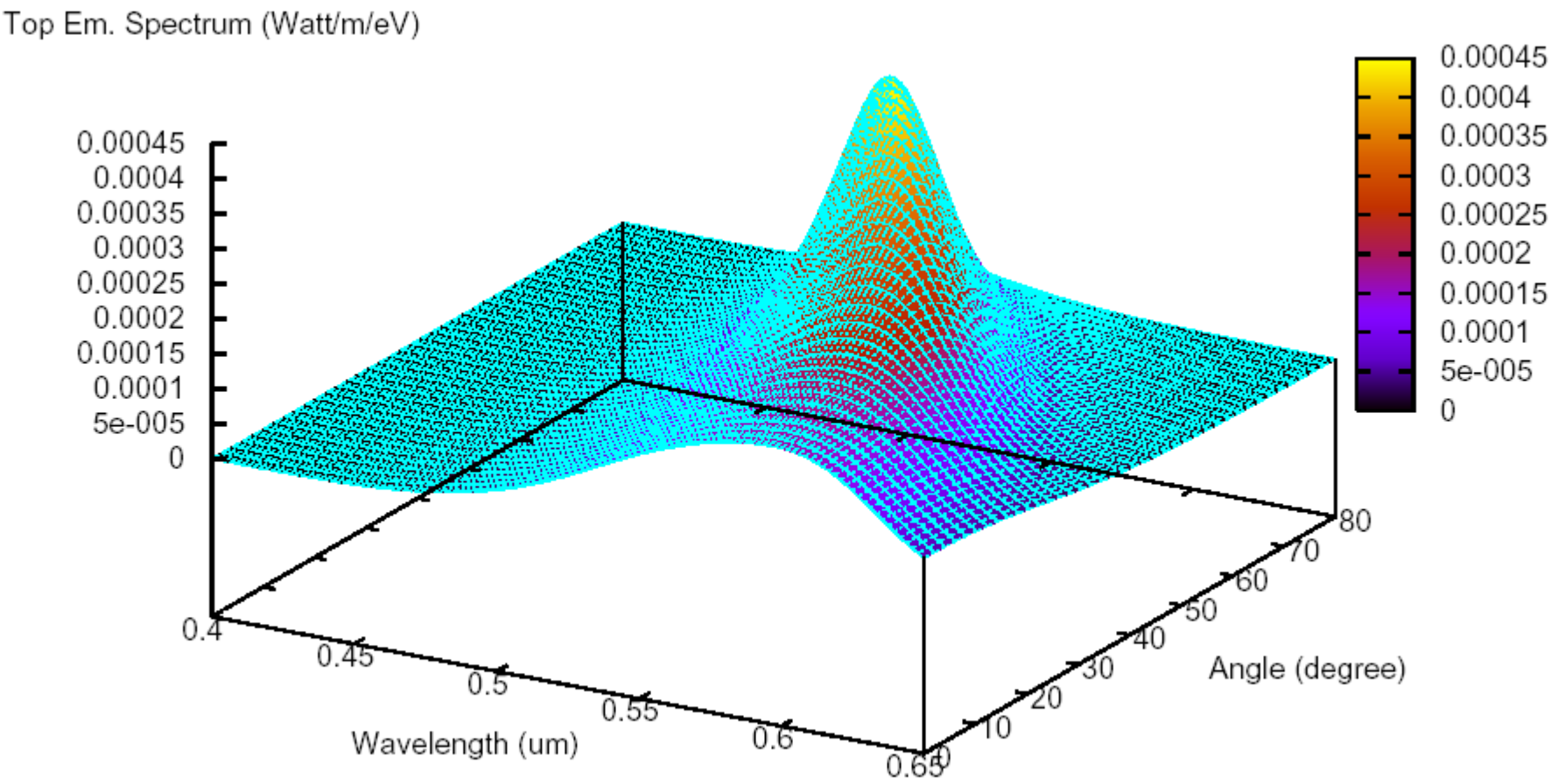






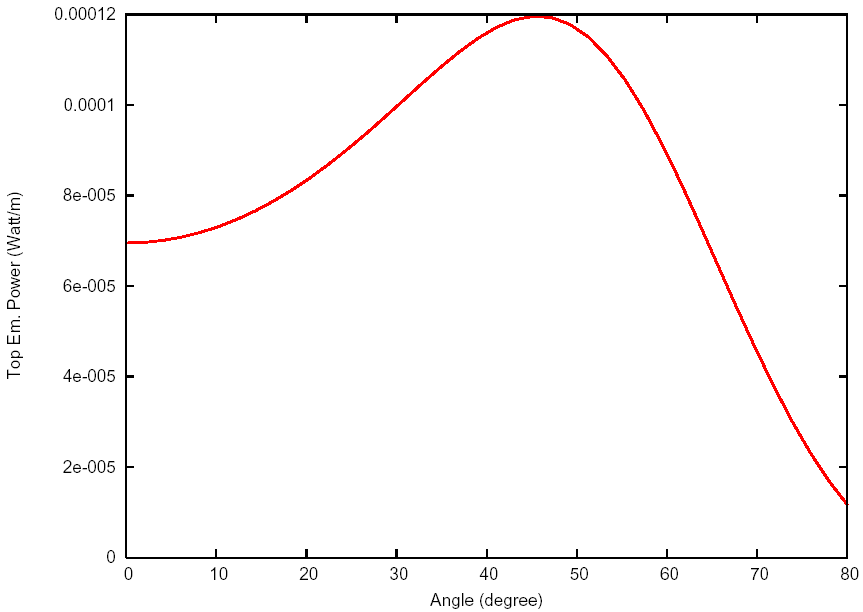


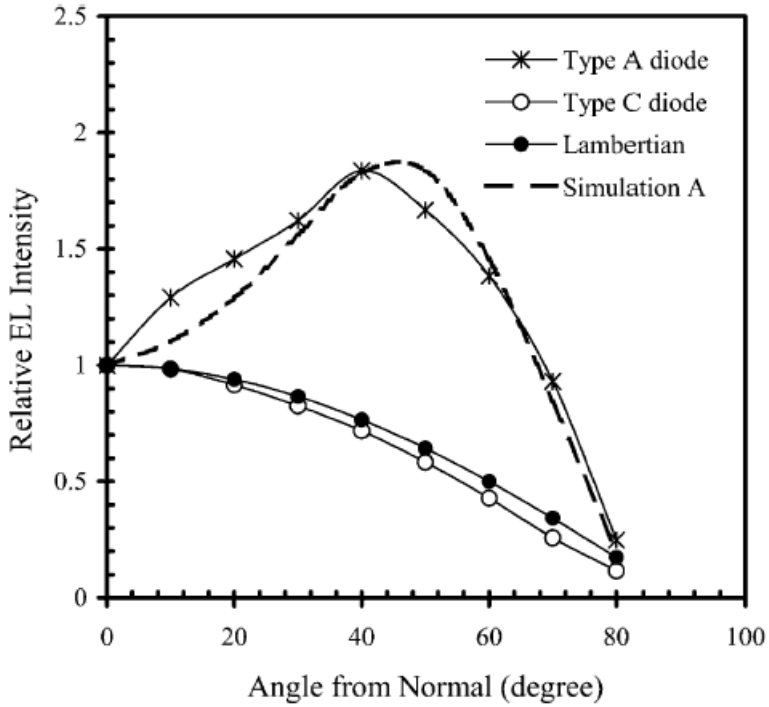
Spectrum vs. angle



➔ Color changes with angle due to microcavity effect

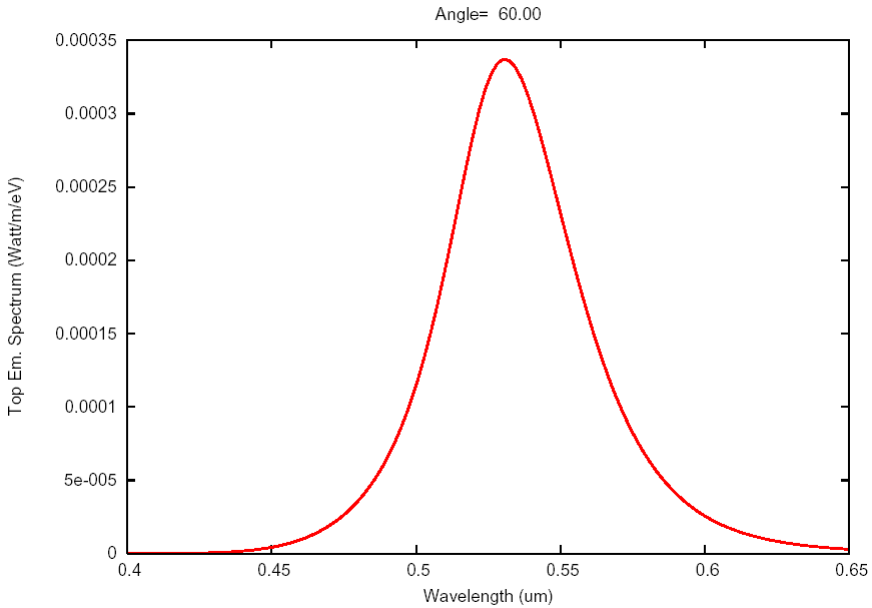
Power vs. angle

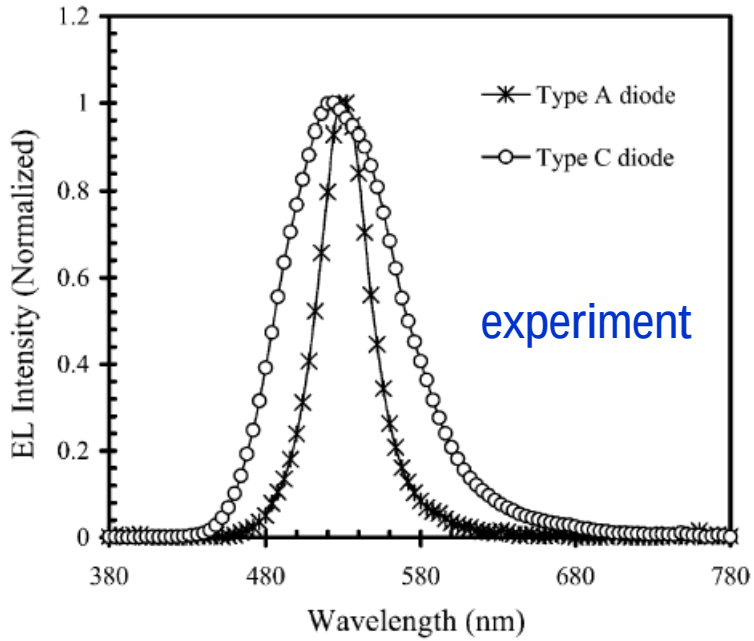




➔ Reasonable agreement with Type A experimental data

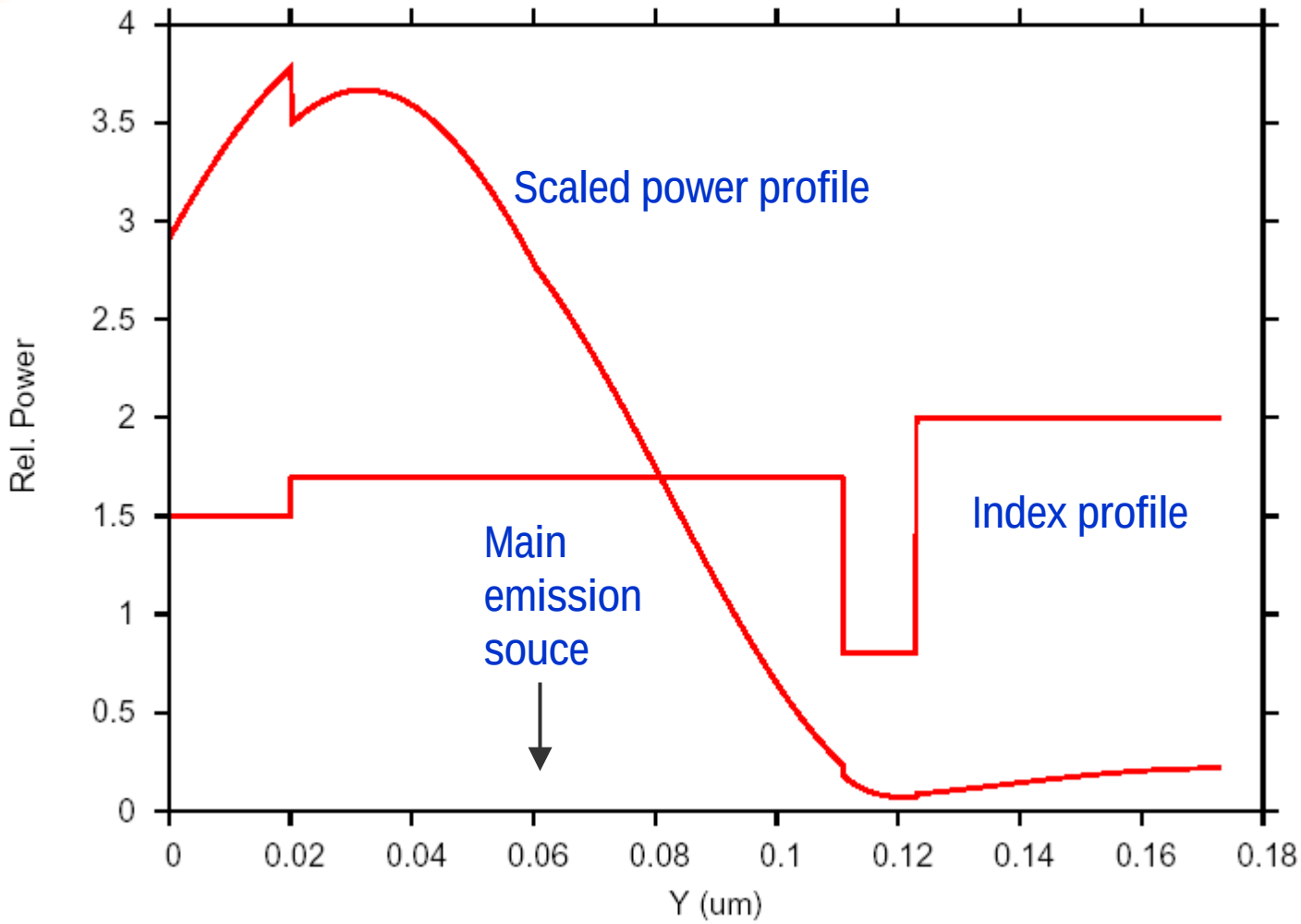
Spectrum





➔ Narrower spectrum means resonance

Can be better?



➔ Device may be optimized if position of emission source is adjusted to the peak of power.

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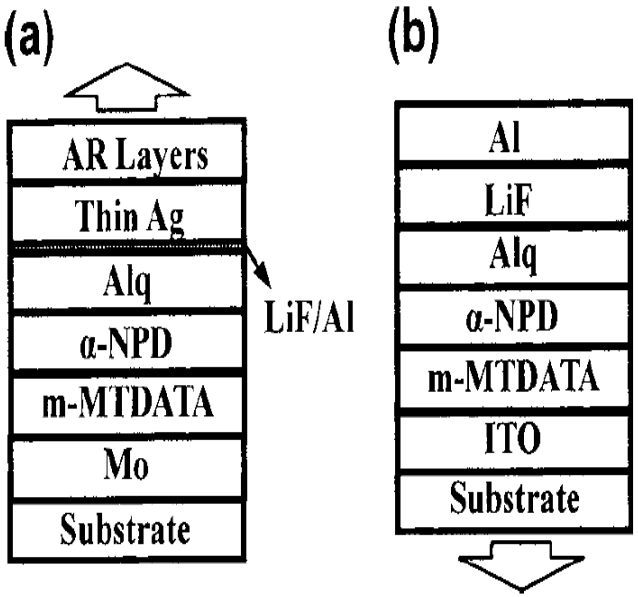
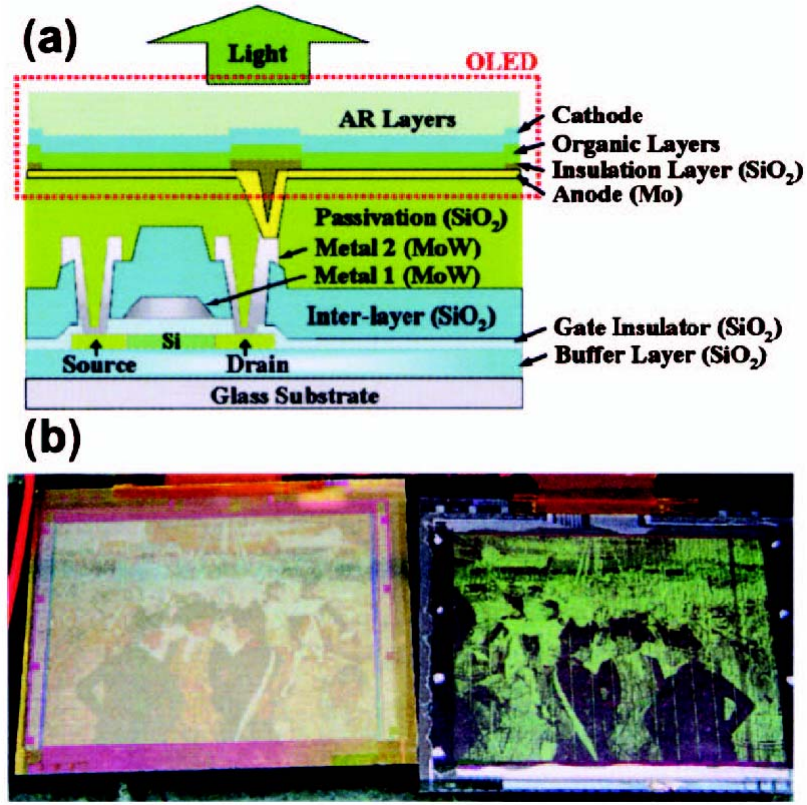
AMOLED Structure



(a) A more recent top-emitting OLED; (b) conventional bottom-emitting OLED.

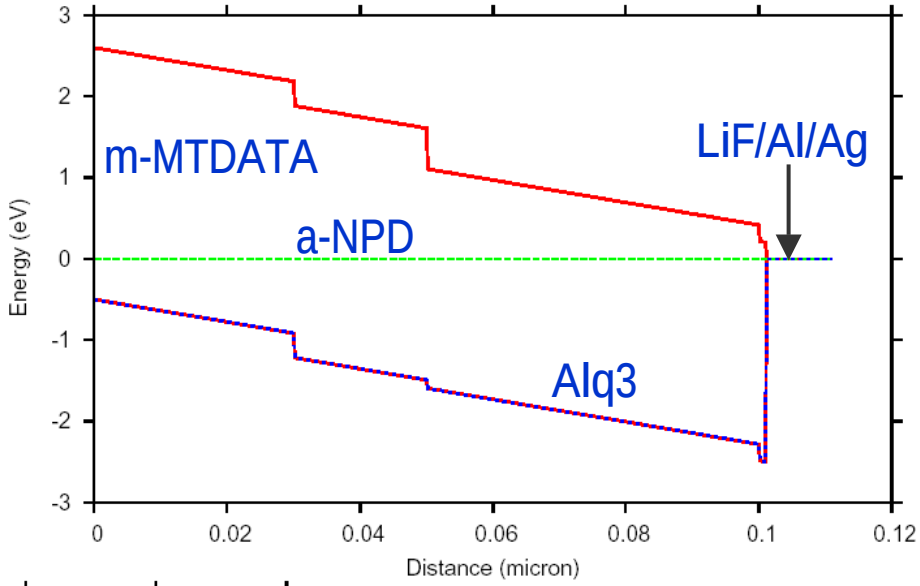
Structure taken from Yang et. al, APL 87, 143507 (2005). We shall simulate top-emitting OLED here.

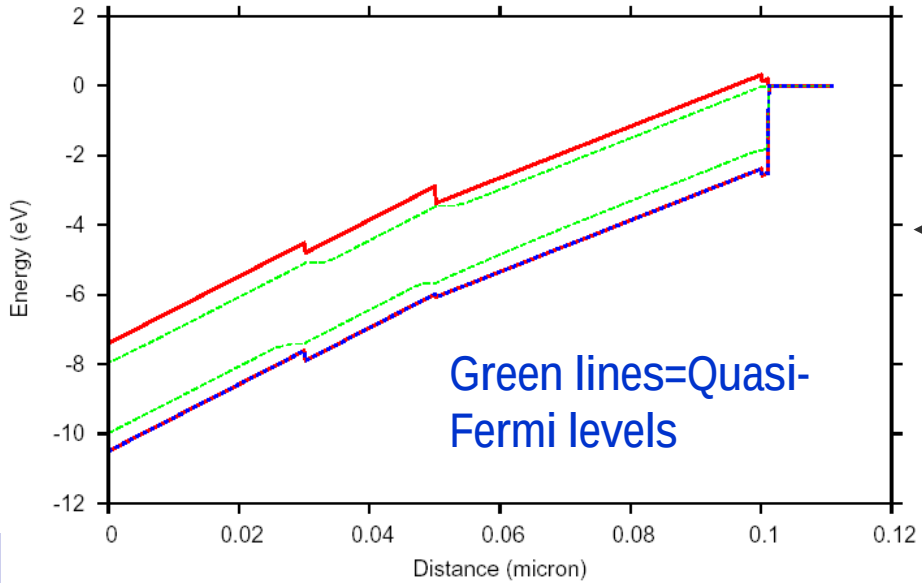


(a) Schematic of top-emitting OLED; (b) Photos of Left=top-emitting, right=bottom emitting OLED.

Band alignment

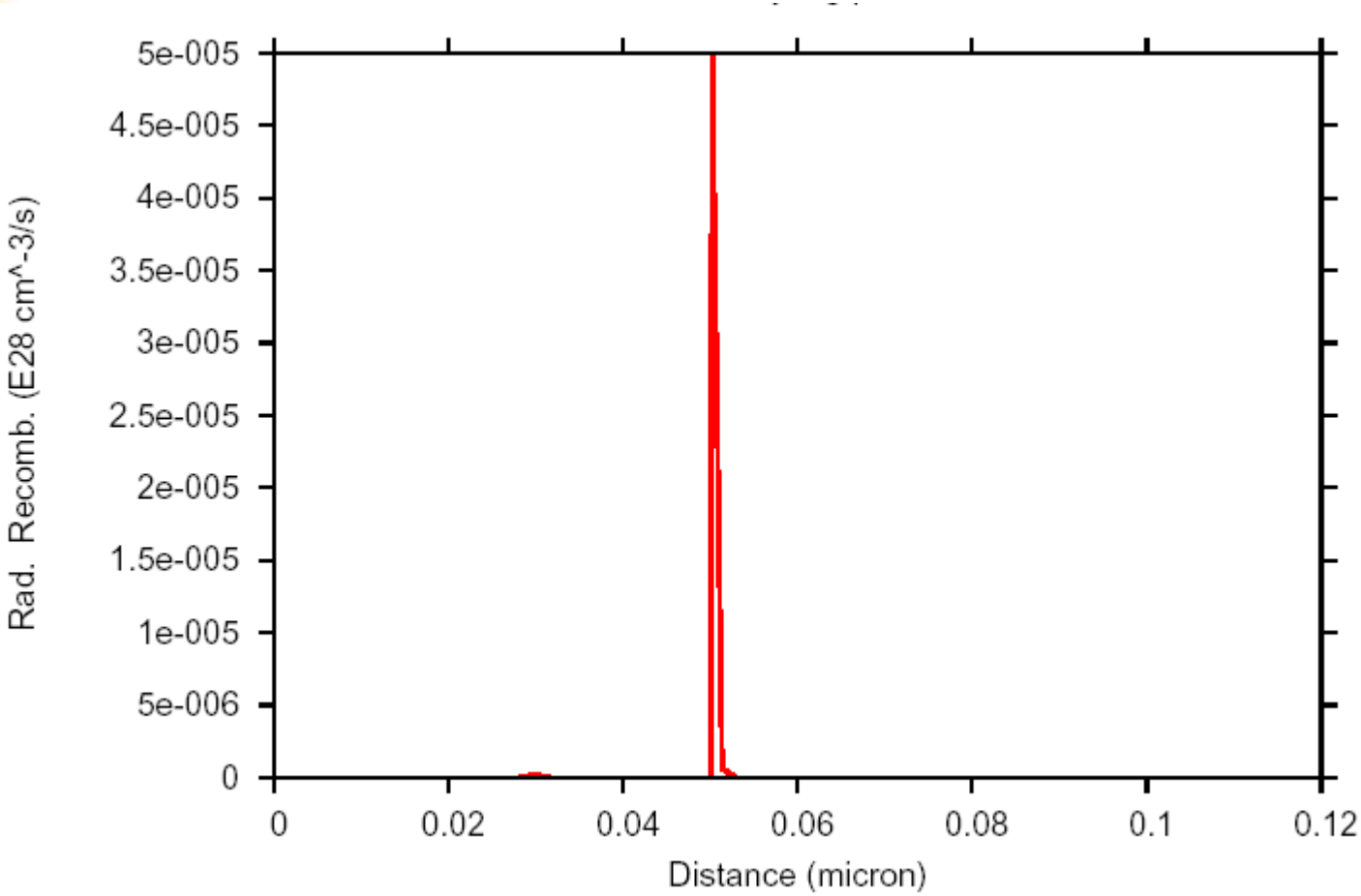
Band diagram at equilibrium.





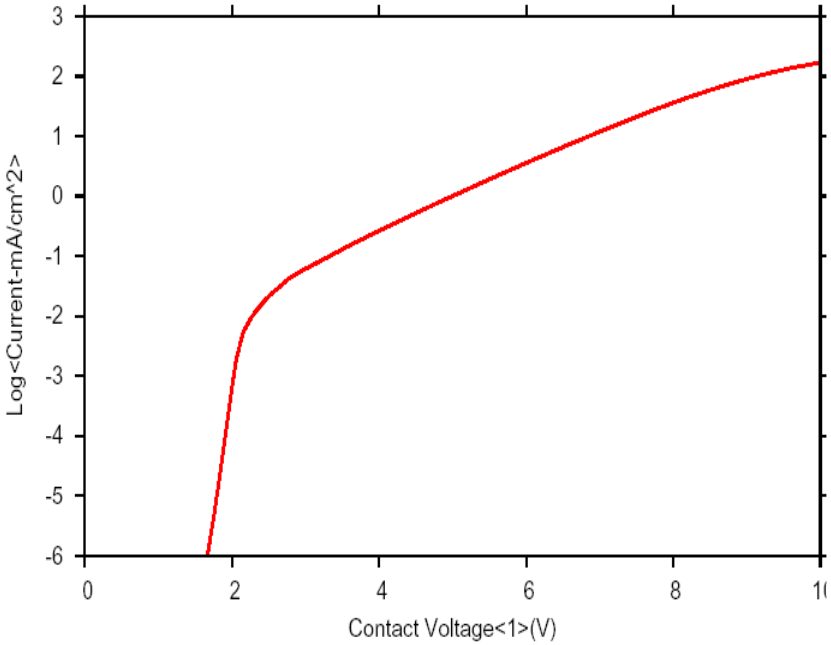
At a bias of 10 volts.

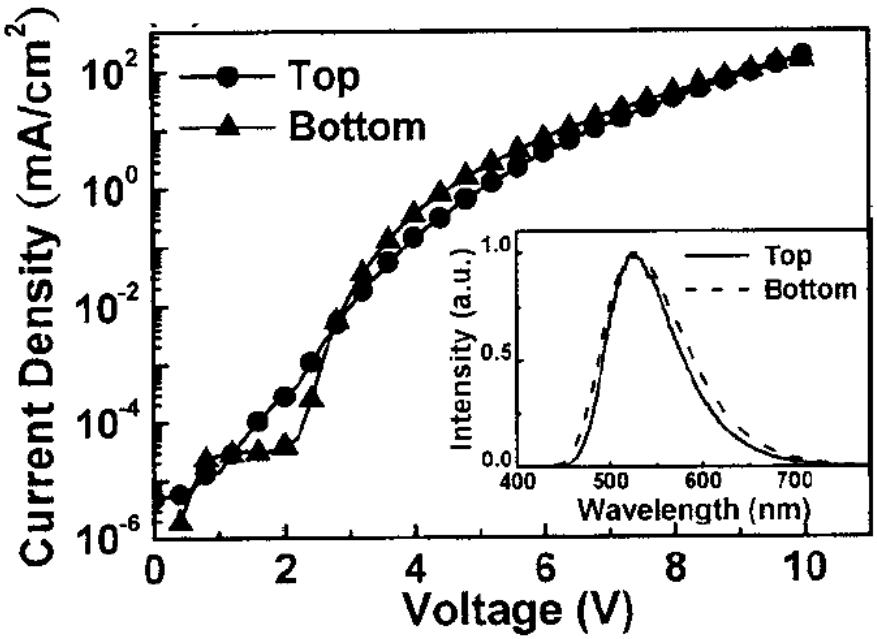
Radiative emitting source



Remark: simulation shows that only under some band alignment conditions will the emitting source be located at Alq3/alpha-NPD interface.

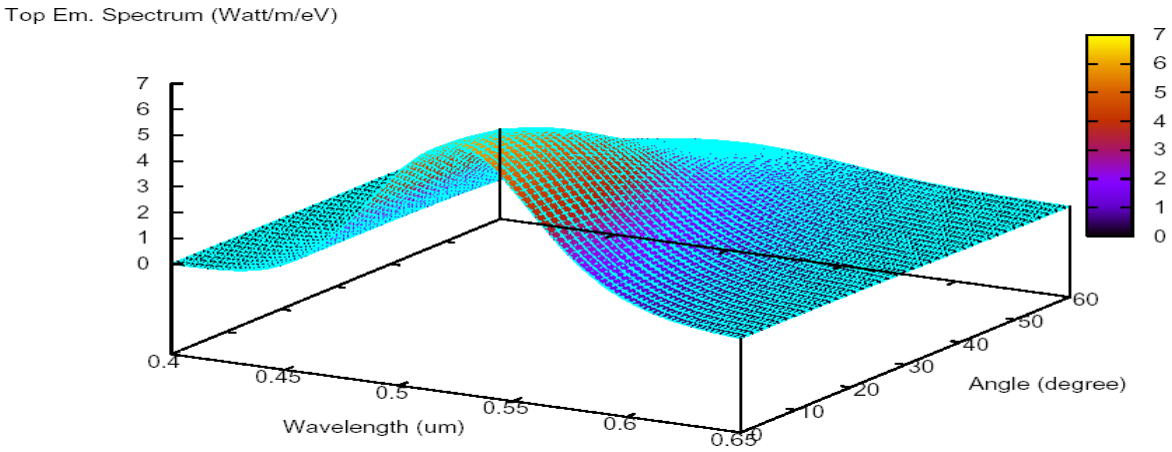
Simulated I-V versus experiment

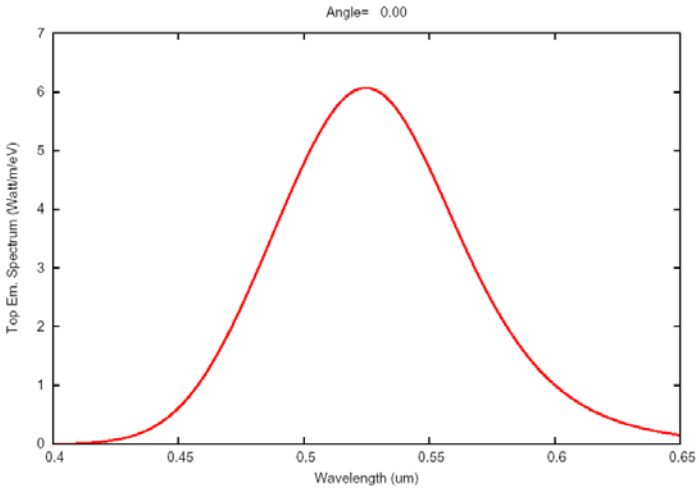


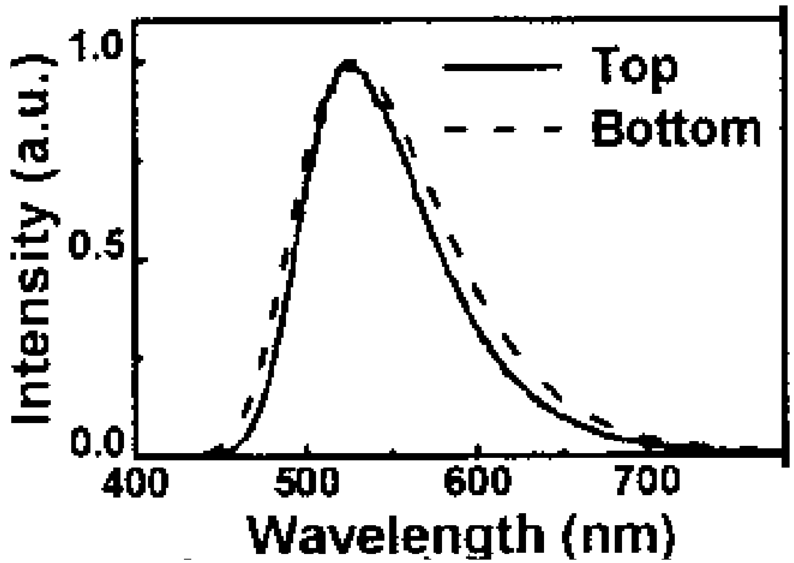


Remark: by adjusting carrier lifetime, mobility, band alignment and metal work functions, reasonable agreement can be achieved over a few decades of current magnitude at higher voltage bias.

Simulated microcavity spectrum

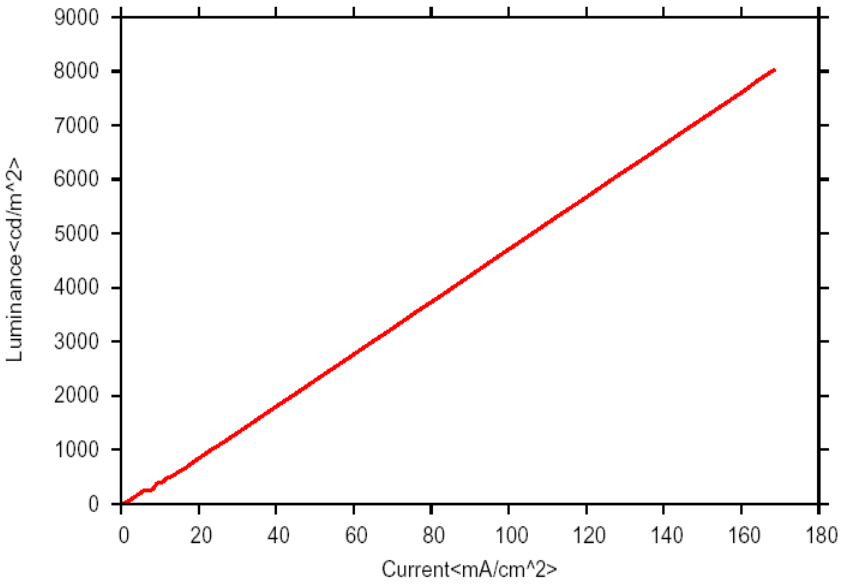


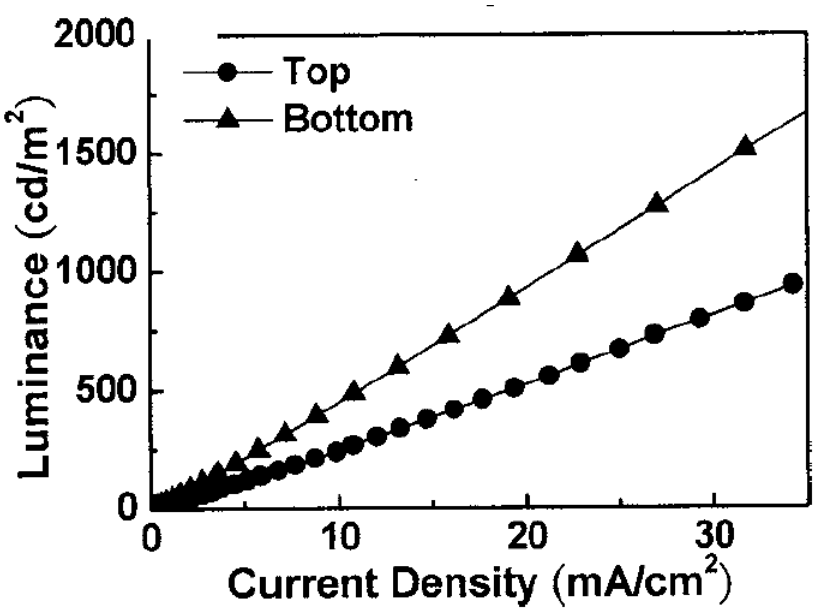




➔ Reasonable agreement with peak position and line width.

Simulated luminance





Agreement on luminance calculation is reasonable considering no fitting effort is made after I-V curve is calibrated. ➔ We have a good IQE/extraction model for RC-OLED.

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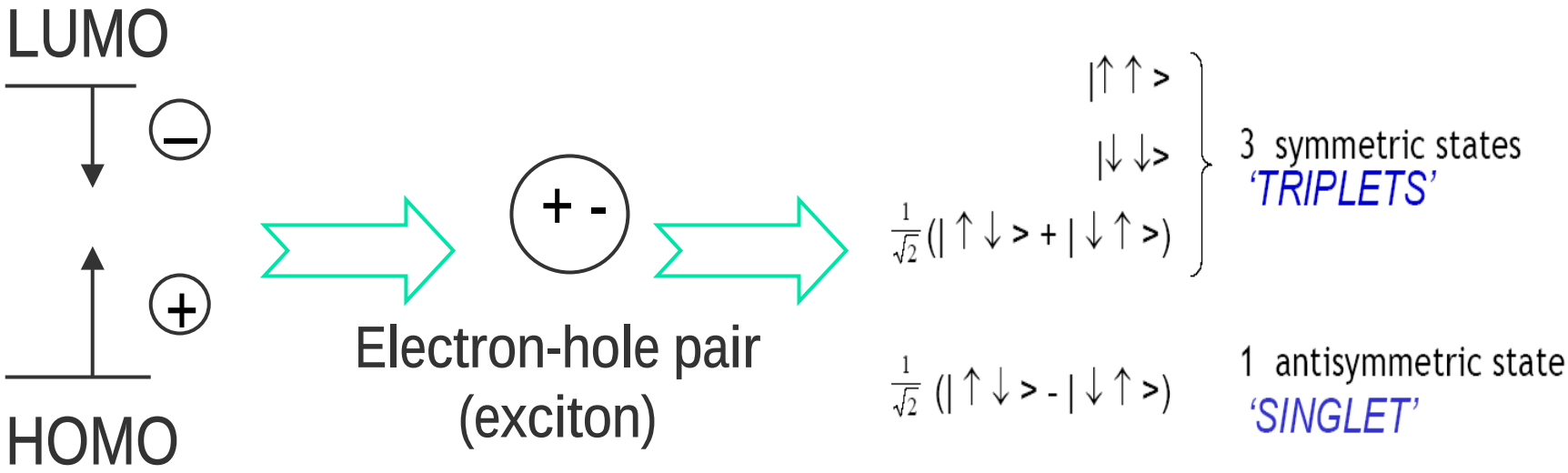
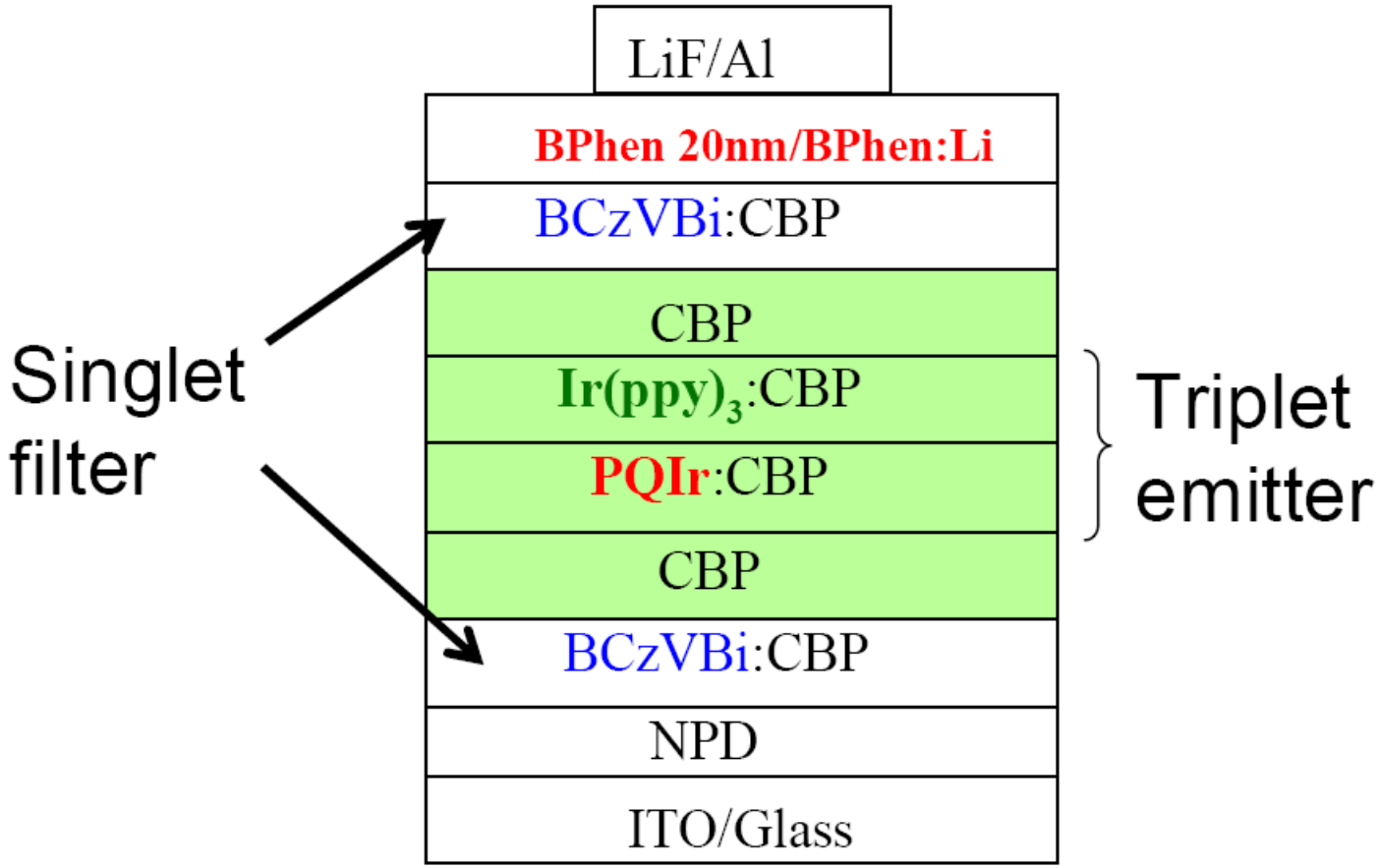
- Tandem OLED

Making use of triplets



- ✓ Singlets may recombine to emit light
- ✓ Triplets are normally wasted unless harvested by phosphorescent dopants

A WOLED using both singlets and triplets



Y. Sun, et. al. "Management of singlet and triplet excitons for efficient white organic light-emitting devices," Nature, Vol 440,13 April 2006, p. 908

Our model

Basic exciton diffusion equations for both singlet: and triplets [1]:

$$\frac{\partial S(x)}{\partial t} = \gamma \cdot r(x) \cdot n(x) \cdot p(x) + D_S \cdot \frac{\partial^2 S(x)}{\partial x^2} - \frac{S(x)}{\tau}$$

— quenching_terms

Exciton quenching may include bulk/interface quenching and triplet-triplet biexciton quenching [2]

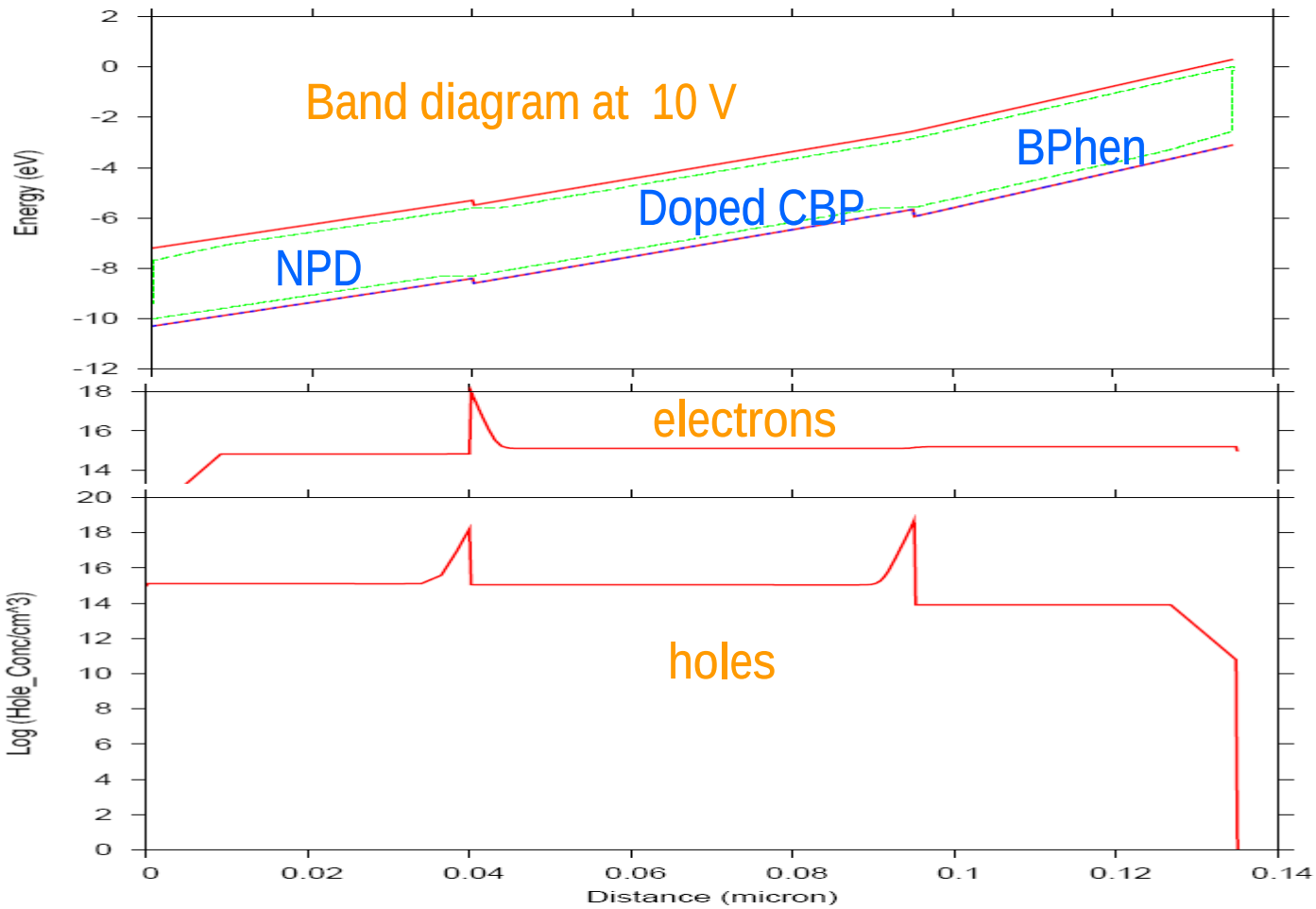
[1]B. Ruhstaller, et.al.,“Simulating Electronic and Optical Processes in Multilayer Organic Light-Emitting Devices,” IEEE J. SEL. TOPICS IN QUANTUM ELECTRONICS, VOL. 9,2003,p. 723.

[2] M. A. BALDO, et.al., "Transient analysis of organic electrophosphorescence. II. Transient analysis of triplet-triplet annihilation,"p. 10 967 PRB vol. 62, 2000

Some model details

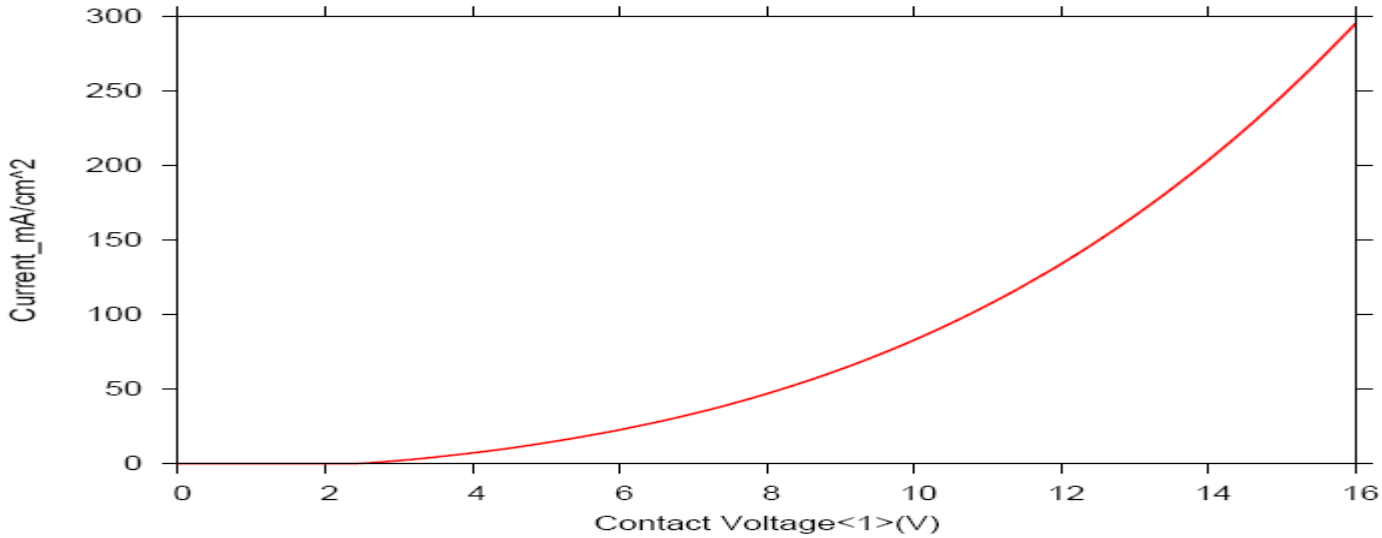
- Biexciton quenching rate takes the form k_TT*S_T*S_T where S_T is triplet exciton density.
- Dopant dependent exciton lifetimes for different layers are adjusted to tune the color of the EL spectrum.
- Linear field dependent EL energy transfer from one dopant to another. This is used to model energy transfer from green dopant to red dopant.

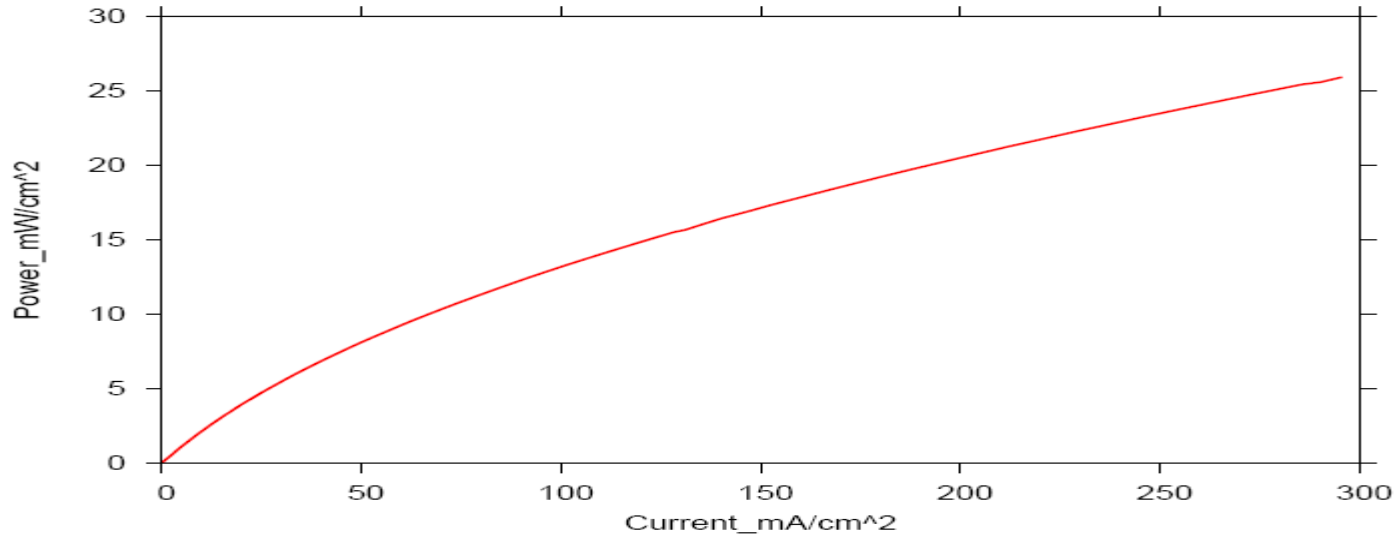
Band alignment and carrier profile



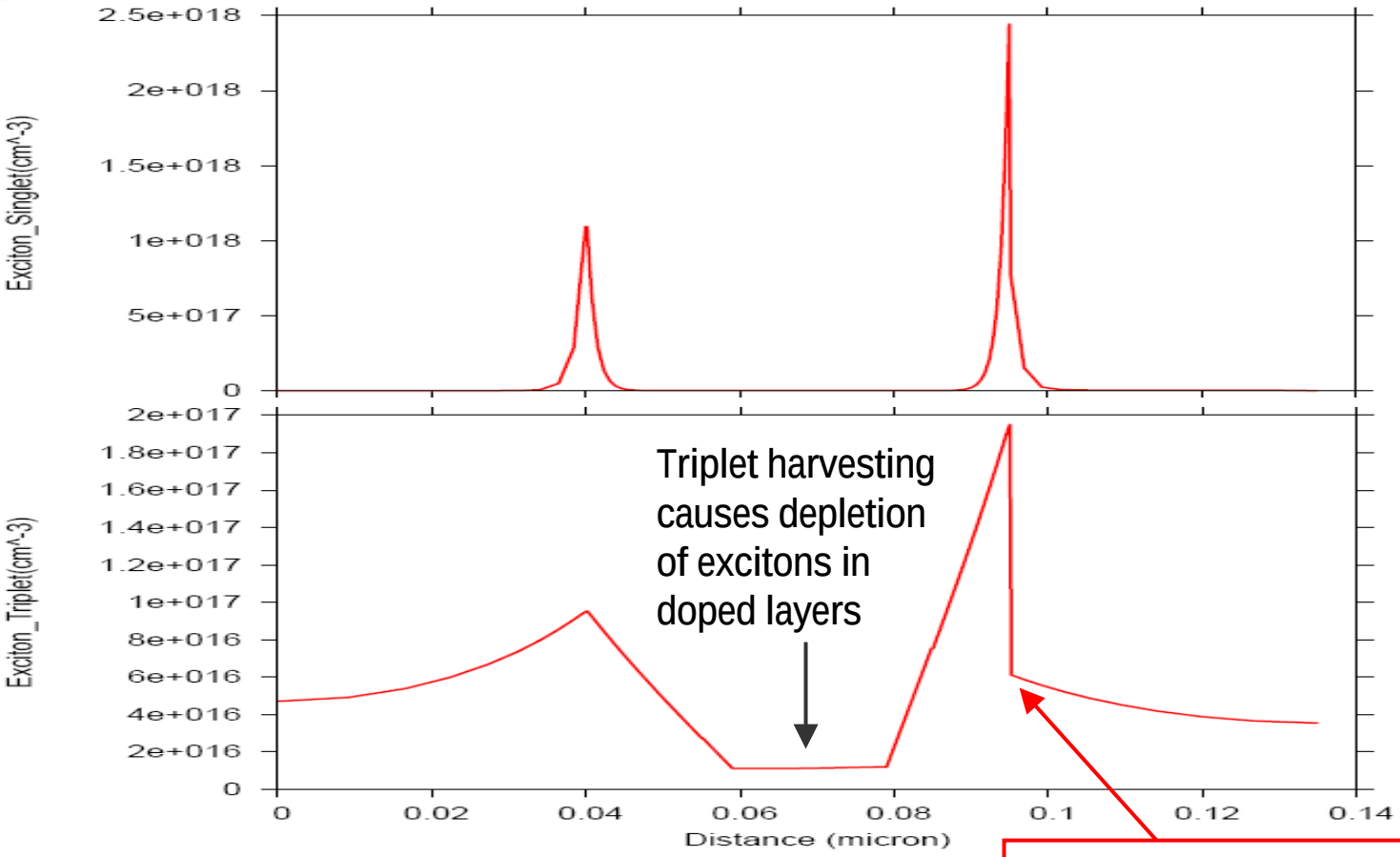
Carrier profiles are sensitive to band alignments

Simulated I-V and P-I



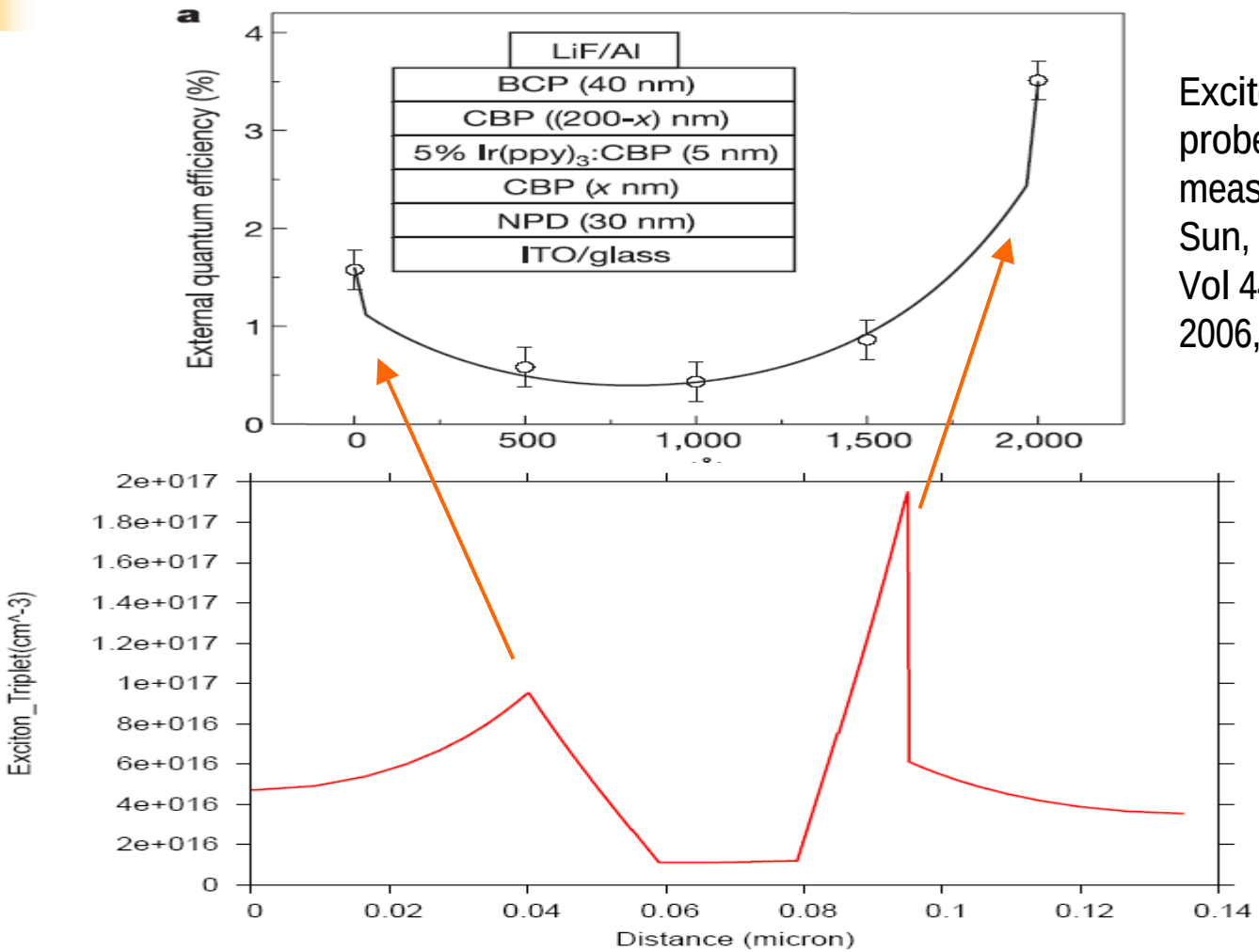


Exciton profiles



Remark: Tripelets have a much longer diffusion length (46nm) than singlets (1nm).

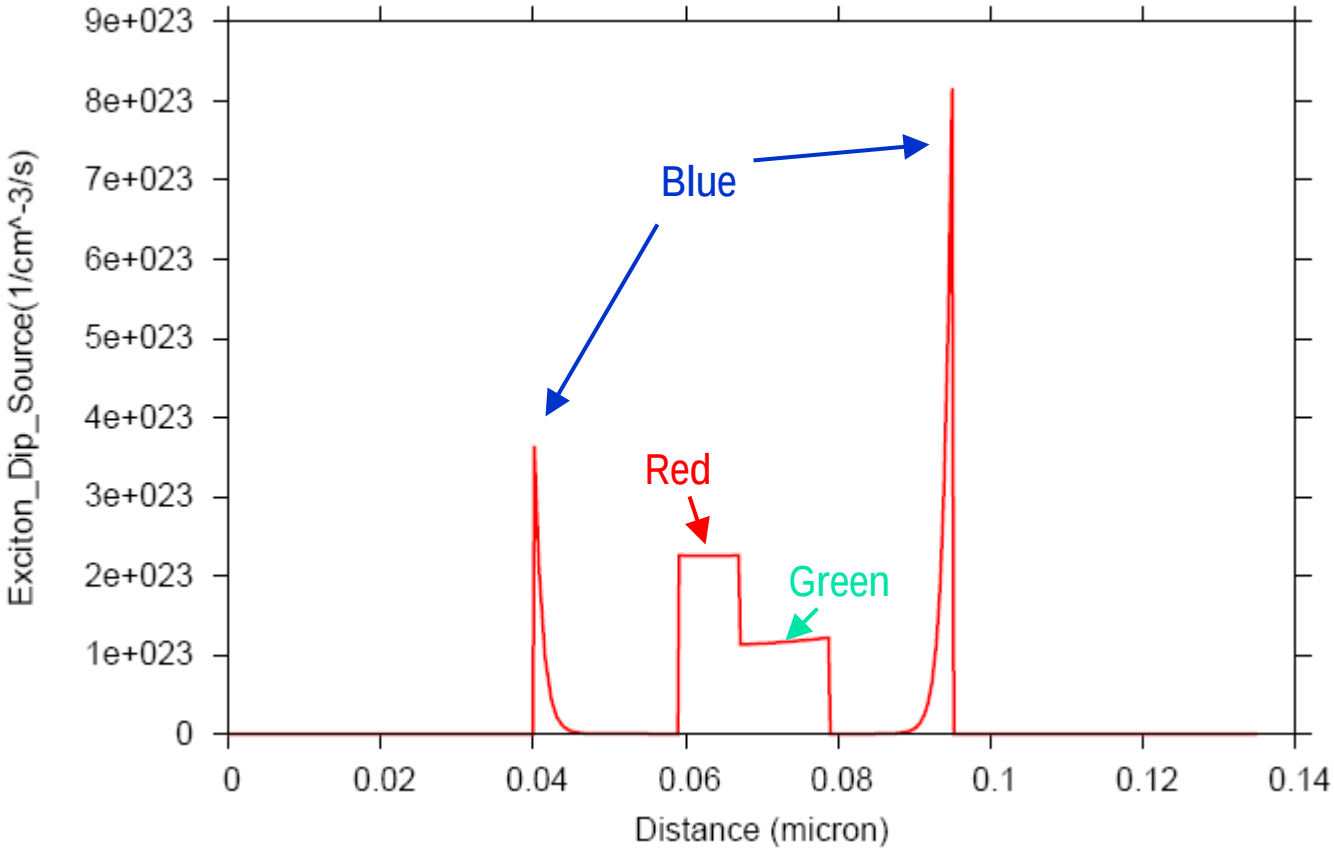
Triplet behavior



Exciton profile probed by EQE measurement by Y. Sun, et. al. , Nature, Vol 440,13 April 2006, p. 908

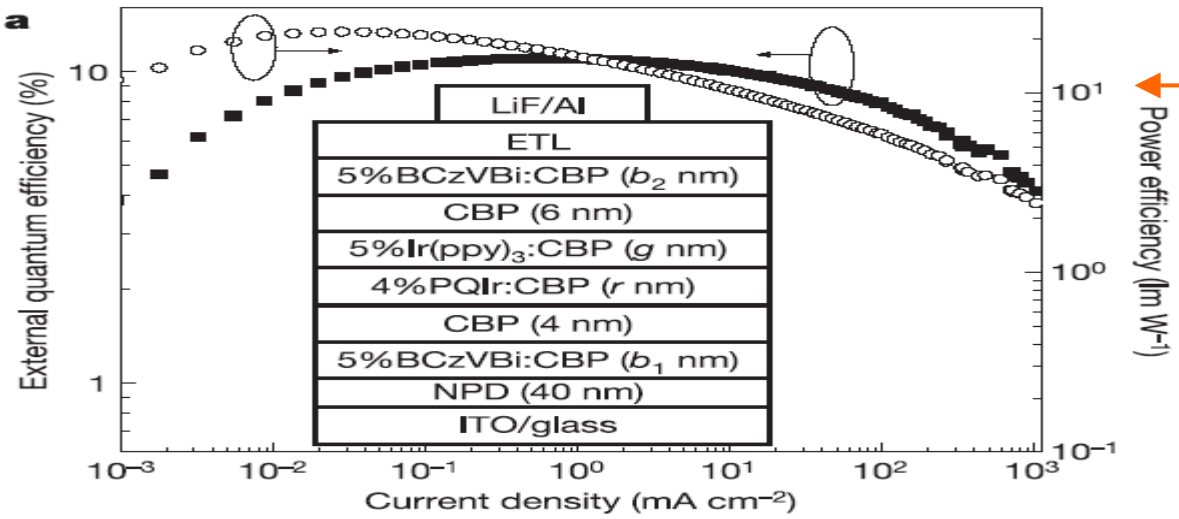
✓Simulated triplet profile consistent with experimental data reported in literature.

Exciton dipole profile

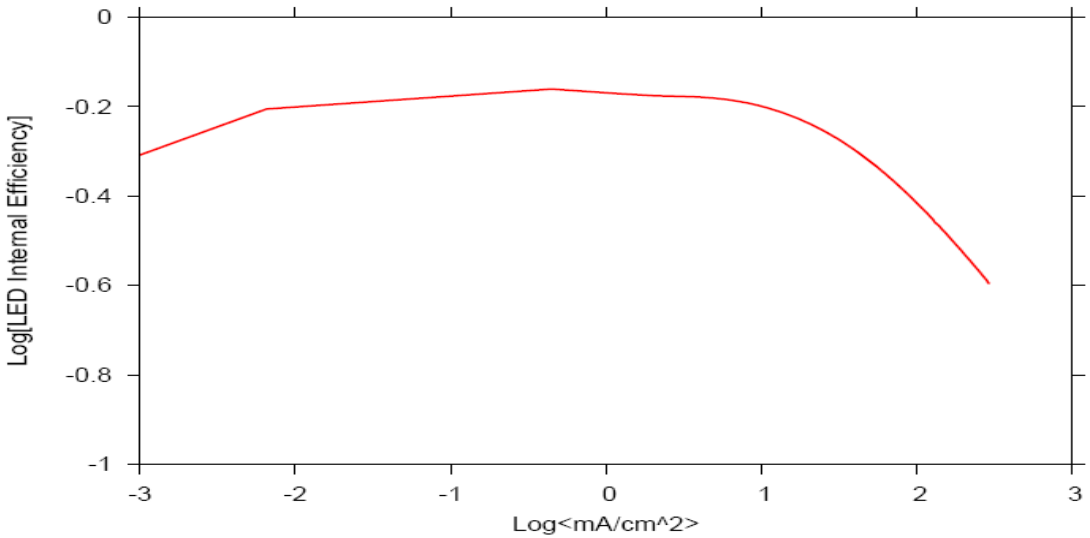


Remark: exciton radiation happens mainly in doped layers.

IQE behavior

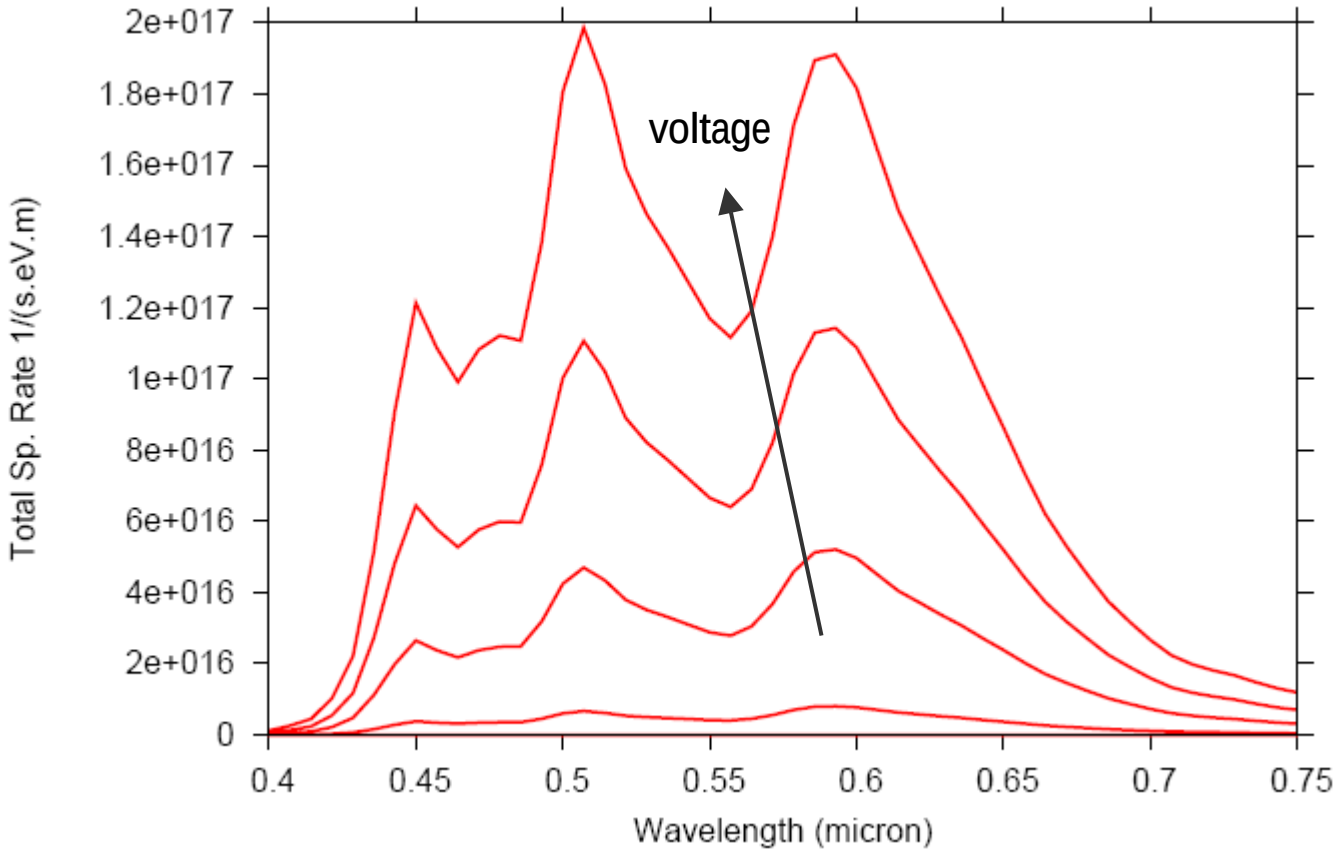


Y. Sun, et. al. , Nature, Vol 440,13 April 2006, p. 908



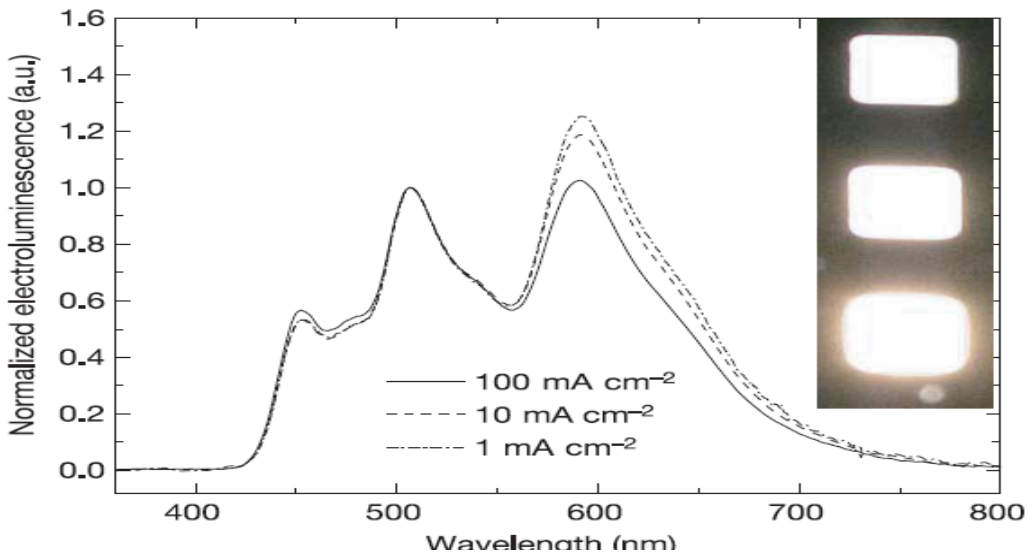
Similar physical trends are obtained for quantum efficiency. Better fit may be obtained if more quenching terms are included.

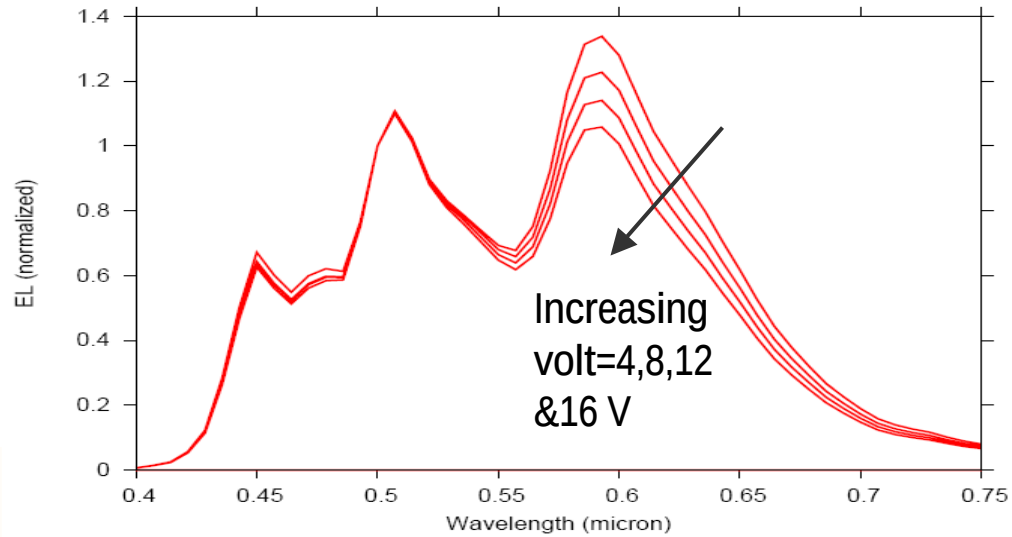
Stable white color obtained



Spectrum obtained at 4, 8, 12 and 16 volts of bias.

Normalized EL spectrum





Y. Sun, et. al. ,
Nature, Vol 440,13
April 2006, p. 908

Based on previous experiments from the same authors, exciton lifetimes were set at 1-100 of micro-seconds and adjusted to match experimental spectrum.

⇒ Reasonable agreement with experiment.

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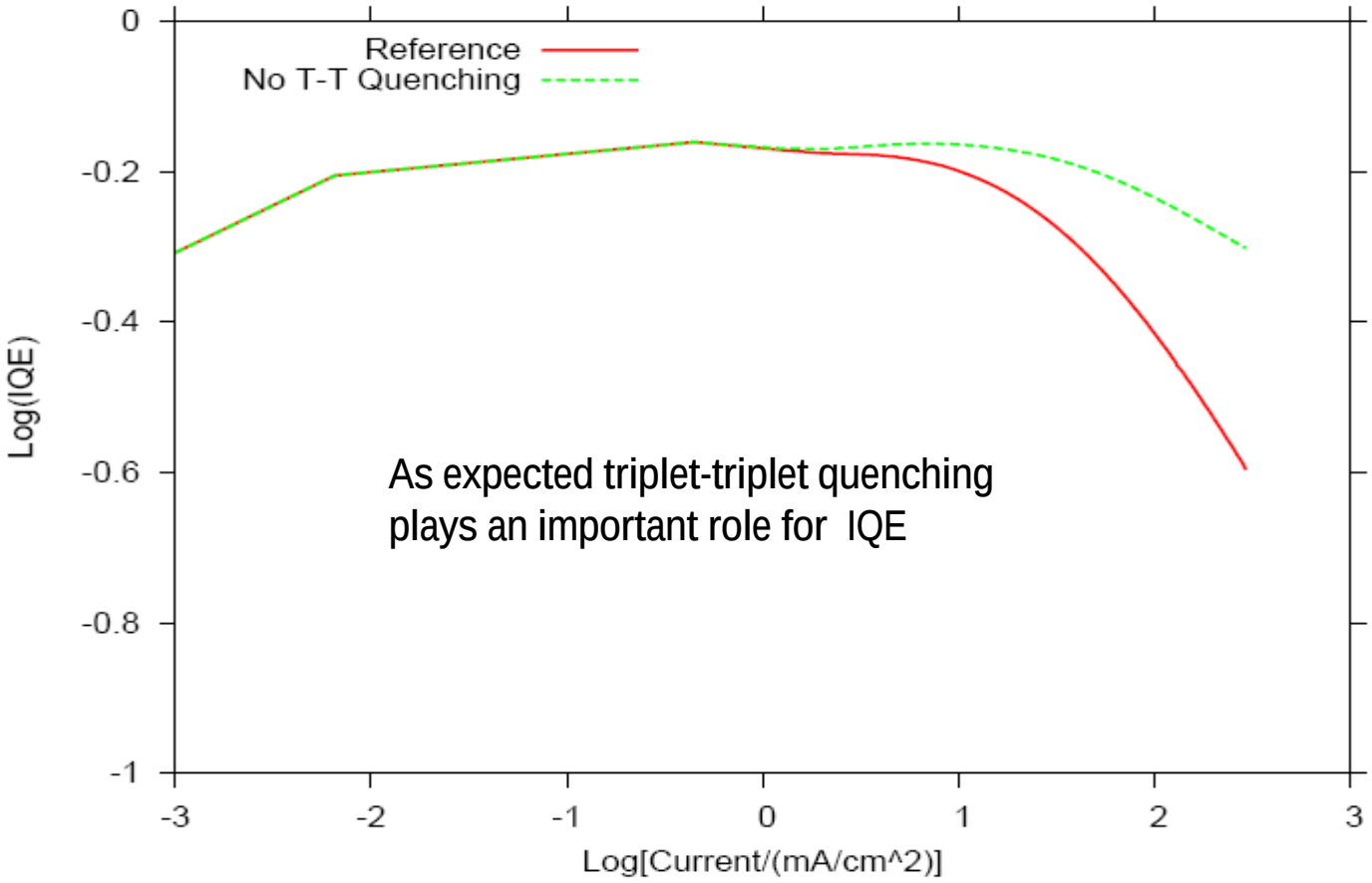
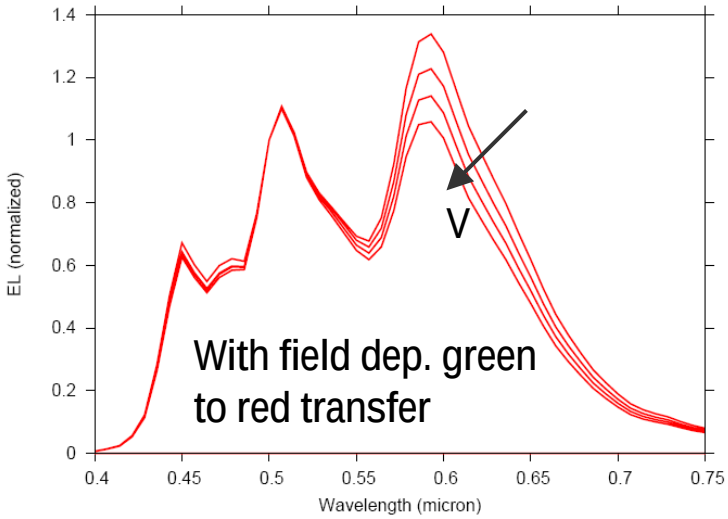
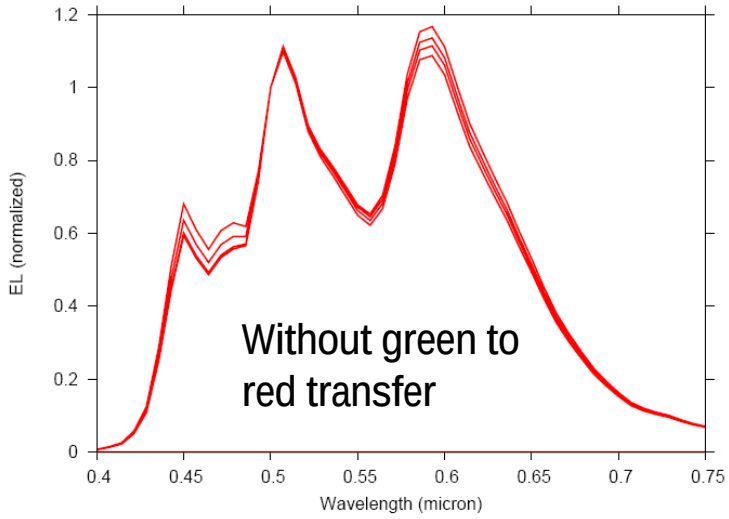
Biexciton quenching

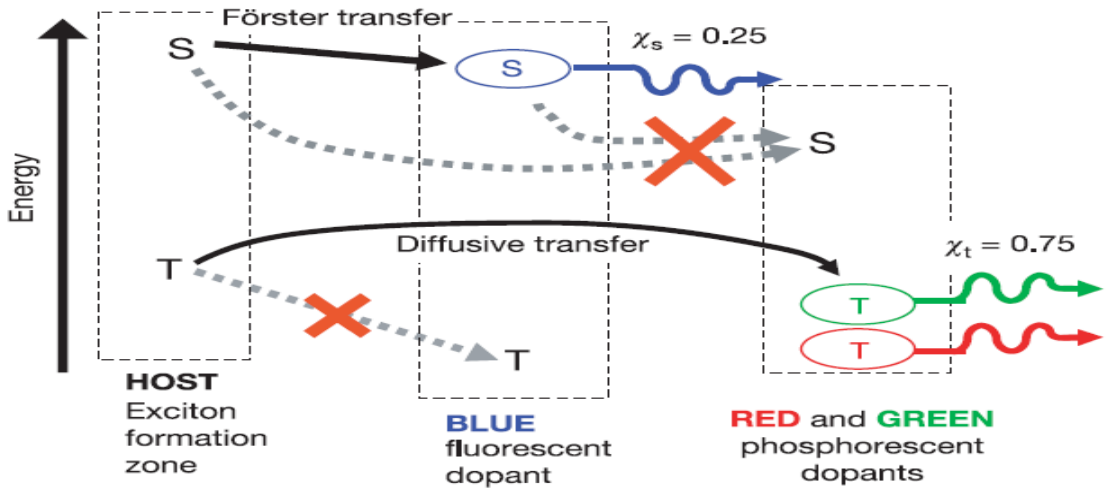


As expected triplet-triplet quenching plays an important role for IQE

Triplet dopant-dopant energy tranfer







Green to red energy transfer (loss) were proposed: Y. Sun, et. al. , Nature, Vol 440,13 April 2006, p. 908

Summary on WOLED simulation

Based on simulation and analysis using APSYS, we find

- **Triplet profile determined by band alignment.**
- **White color stability relies on multiple factors such as single/triplet ratio, field dependent exciton lifetime, triplet-triplet quenching and dopant to dopant energy transfer.**
- **Simulation may help gain insight into and help design better WOLED.**

Contents

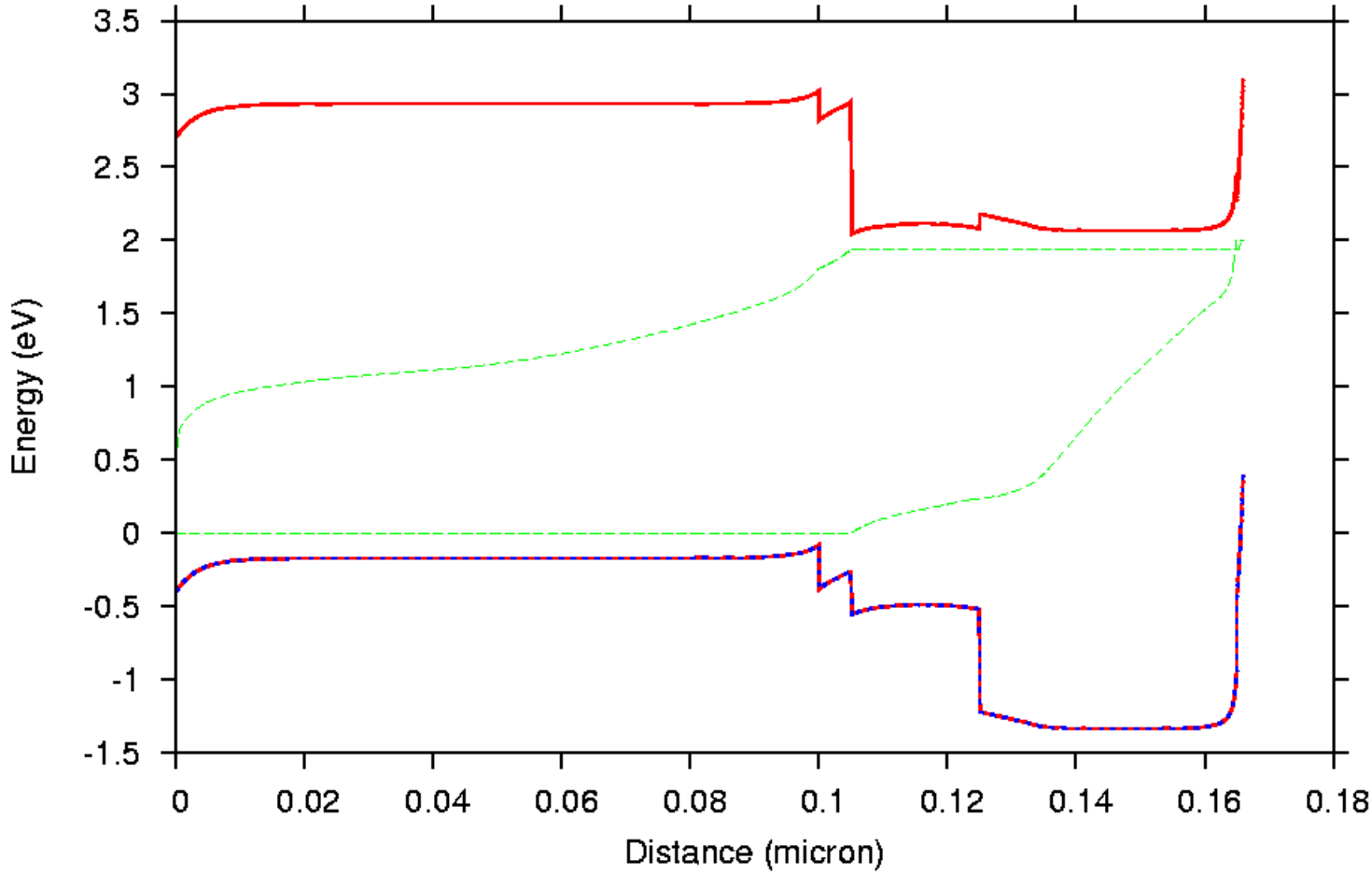
- Treating organic semiconductor with quantum drift-diffusion model.
- Light extraction using 3D ray-tracing (non-microcavity model).
- Electroluminescent spectra modeling with Frenkel exciton.
- Inclusion of microcavity effects.
- Application for AMOLED.
- Triplet diffusion and WOLED
- **Low-voltage PIN structures**
- Tandem OLED

Layer Structure & Model

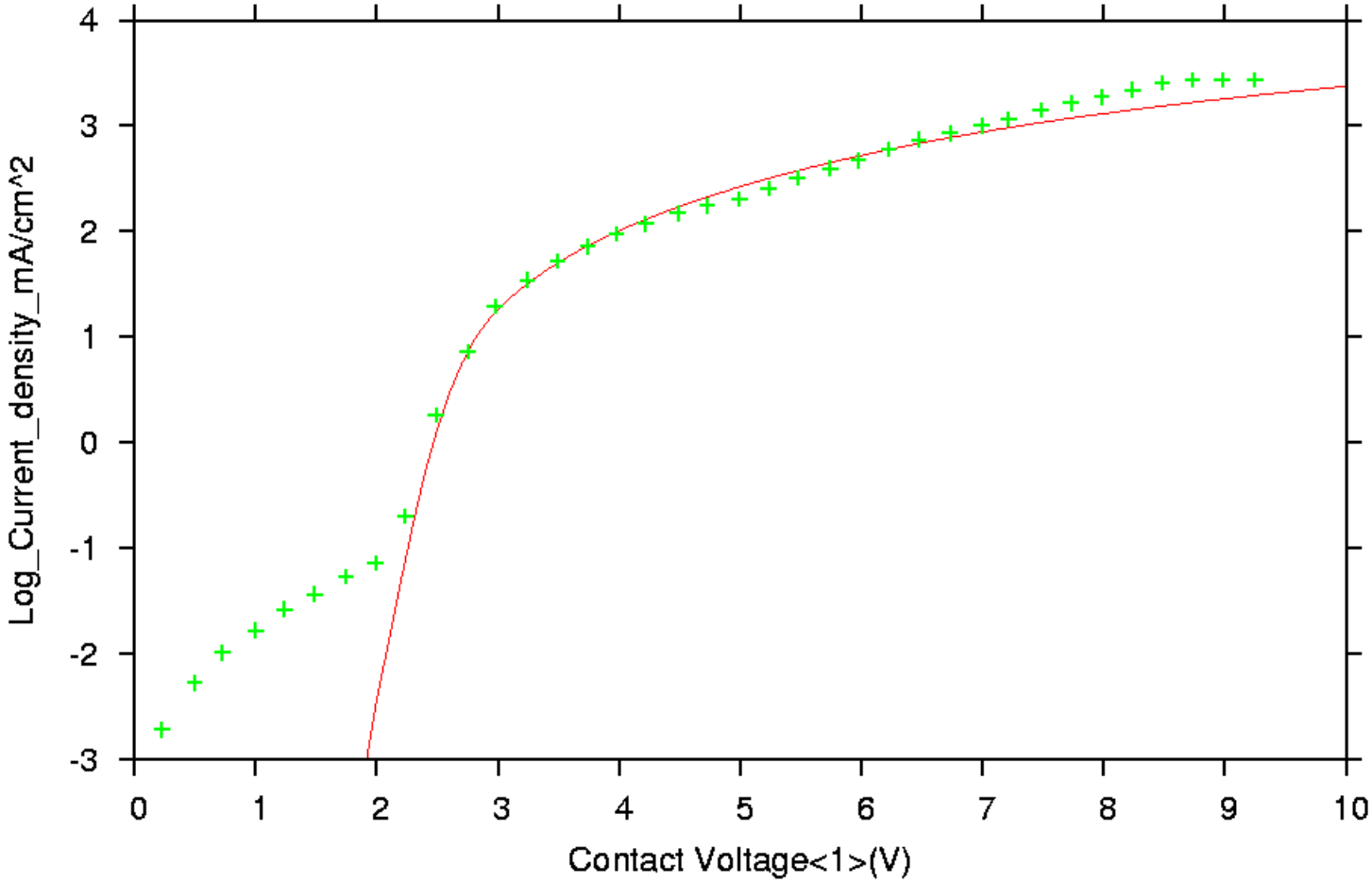
- ITO/glass substrate
- p-doped HTL (100 nm) : m-MTDATA:F_4-TCNQ (2% mol.)
- TPD (5 nm): carrier confinement
- Alq3 (20 nm) : active region @ 530 nm
- Bphen (10 nm) : carrier confinement
- n+-doped ETL (30 nm) : Bphen:Li (1:1 ratio)
- LiF (1 nm)/Al contact
- Contacts defined as Schottky
- Energy levels of electrical dopants included to account for incomplete ionization
- Quantum tunneling defined for contact and TPD regions due to band alignment
- RCLED model used

Based on J. Huang & al., “Low-voltage organic electroluminescent devices using *pin* structures”, Appl. Phys. Lett., Vol. 80 No.1, 7 Jan. 2002

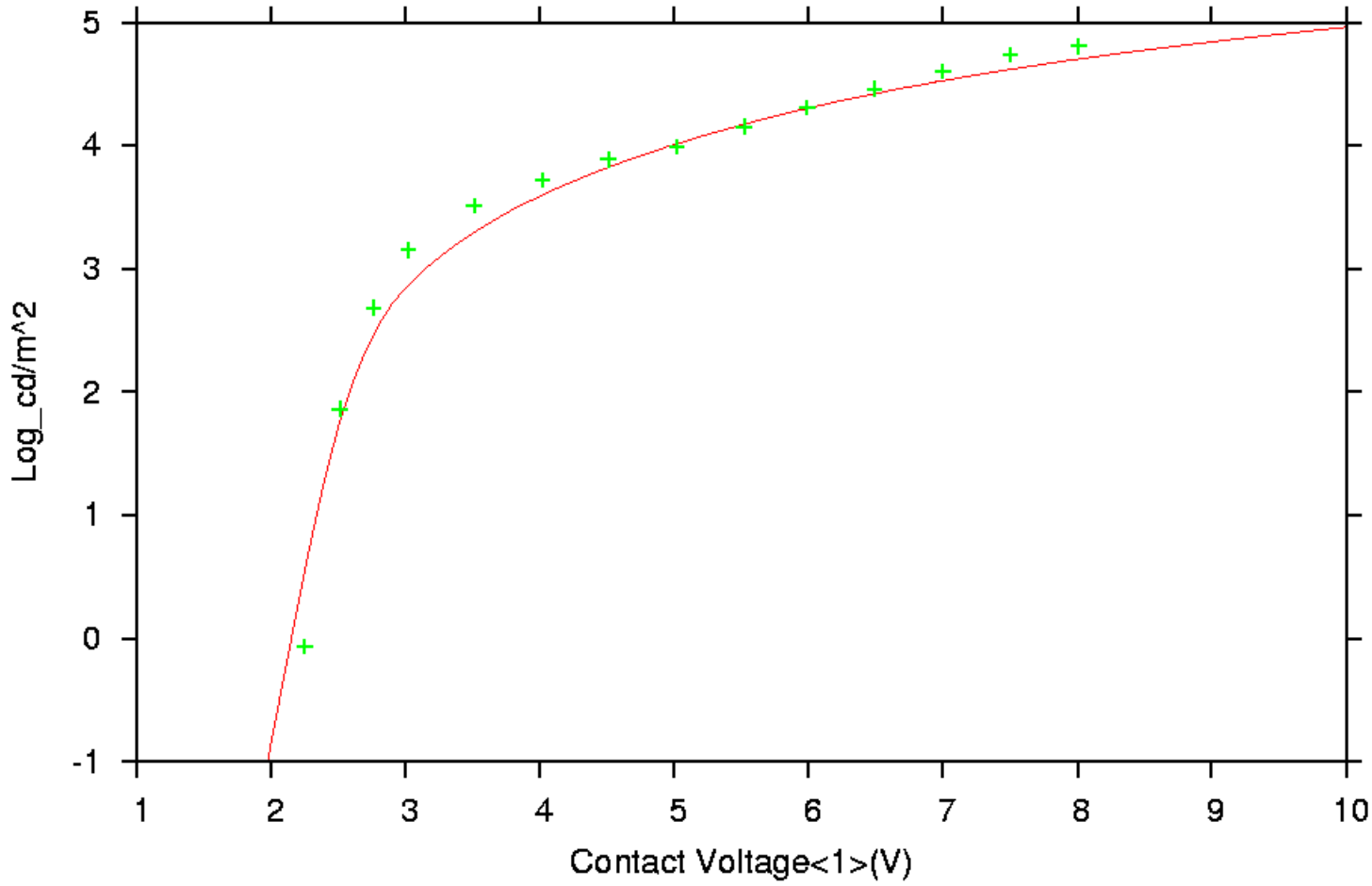
Band diagram @ 2V bias



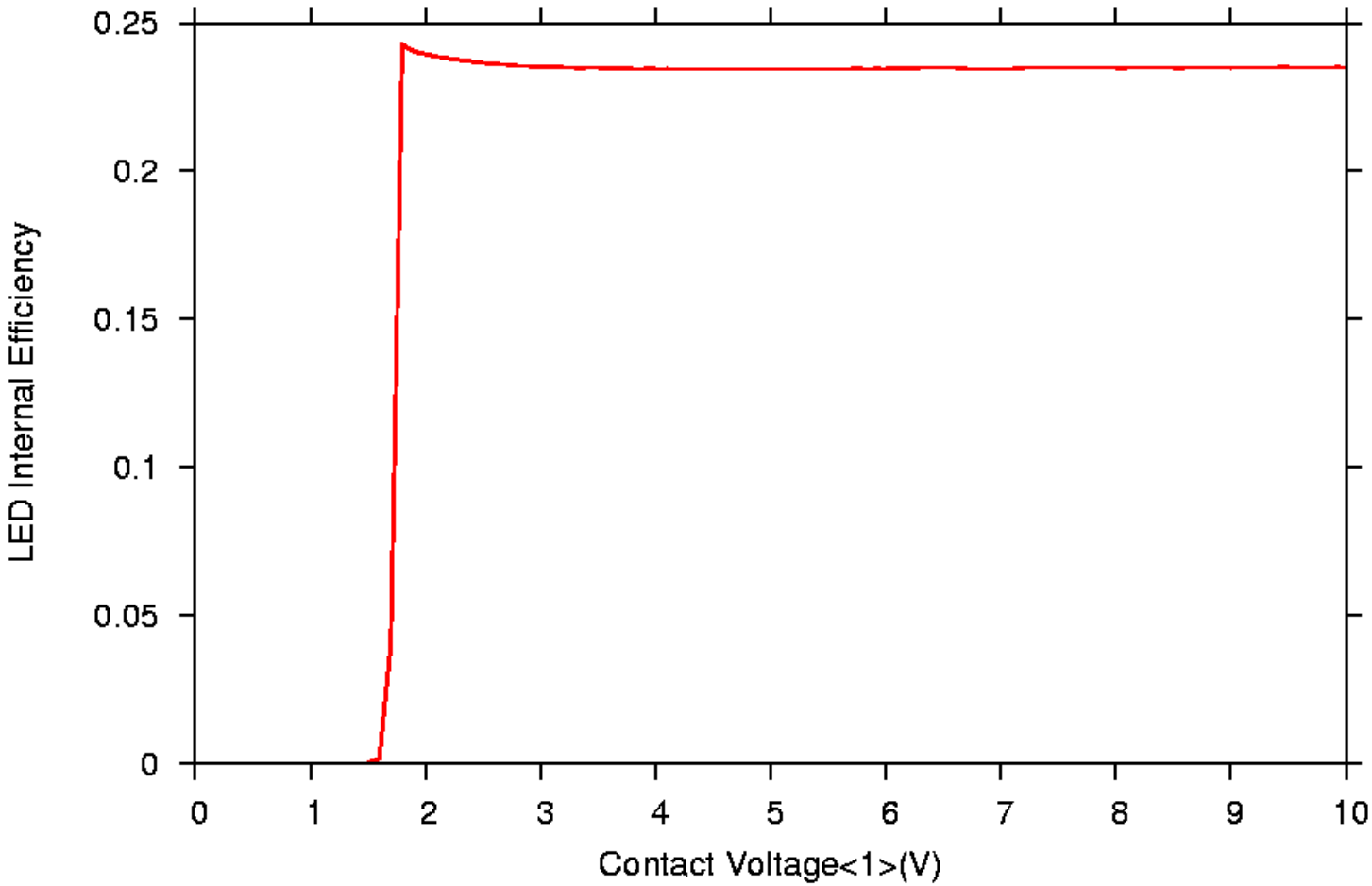
Simulated I-V versus experiment



Simulated L-V versus experiment



Internal Quantum Efficiency



Note: Efficiency limited to 25% since there is no triplet harvesting.

Comments

- Very reasonable fit to experiment over 4 decades of current values and 6 decades of luminance
- Low turn-on voltage, close to thermodynamic limit of Alq3
- High efficiency limited by singlet-only design. Can be improved by triplet harvesting as in previous WOLED design
- Efficiency fit can be improved by adding quenching terms which are important at higher bias values
- PIN design lends itself well to solar cell applications

Contents

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- Low-voltage PIN structures
- **Tandem OLED**

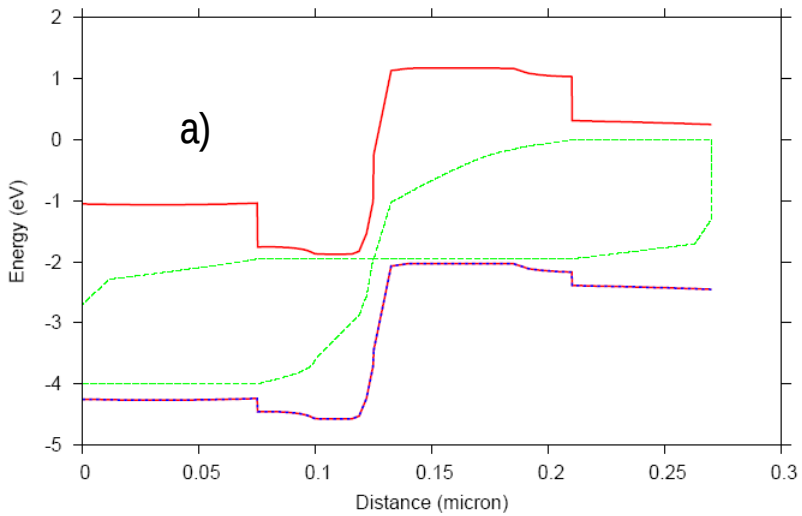
Layer structure of tandem OLED

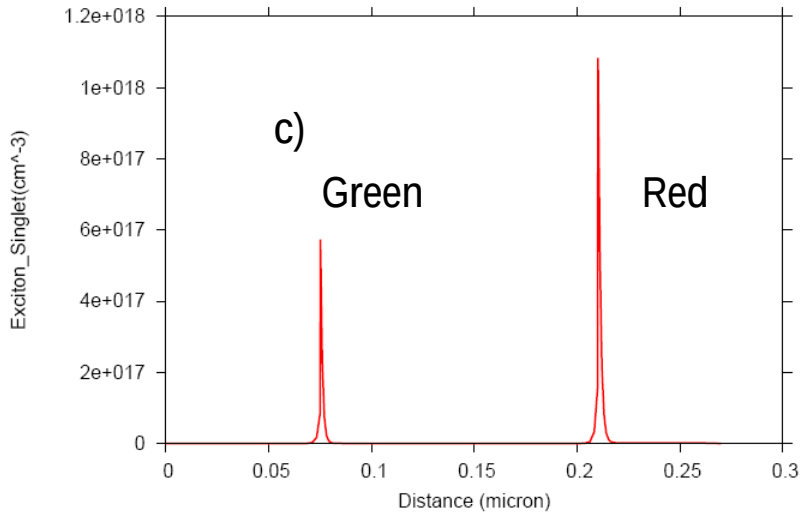
Device or unit	Layer structure
Device A	ITO/SA1/**EL-G**/OC1/**EL-R**/SC1/Mg:Ag
Device B	ITO/SA1/**EL-G**/**EL-R**/SC1/Mg:Ag
Device C	ITO/SA1/**EL-G**/SC1/Mg:Ag
Device D	ITO/SA1/**EL-R**/SC1/Mg:Ag
SA1	NPB (50 nm)
EL-G	NPB (25 nm)/Alq (20 nm)/Alq (5 nm)
OC1	Alq:Li (25 nm)/NPB:$FeCl_3$ (60 nm)
EL-R	NPB (25 nm)/Alq:DCJTB (20 nm)/Alq (5 nm)
SC1	Alq (35 nm)

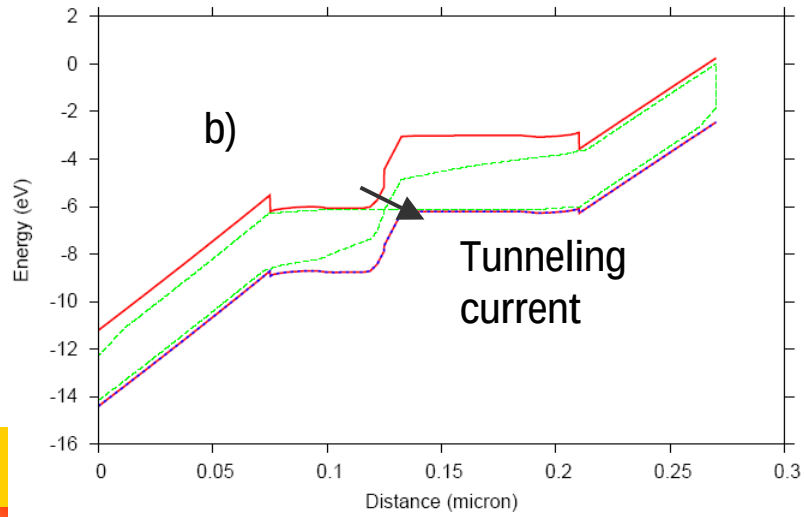
Based on Liao, Klubek, and Tang, "High-efficiency tandem organic light-emitting diodes," Appl. Phys. Lett., Vol. 84, No. 2, 12 January 2004, p.167

Model requirements: treatment of n-type and p-type dopings, tunnel junction, and microcavity effects due to multiple colors with dipole source distributed in multiple locations.

Tunnel Junction for OLED





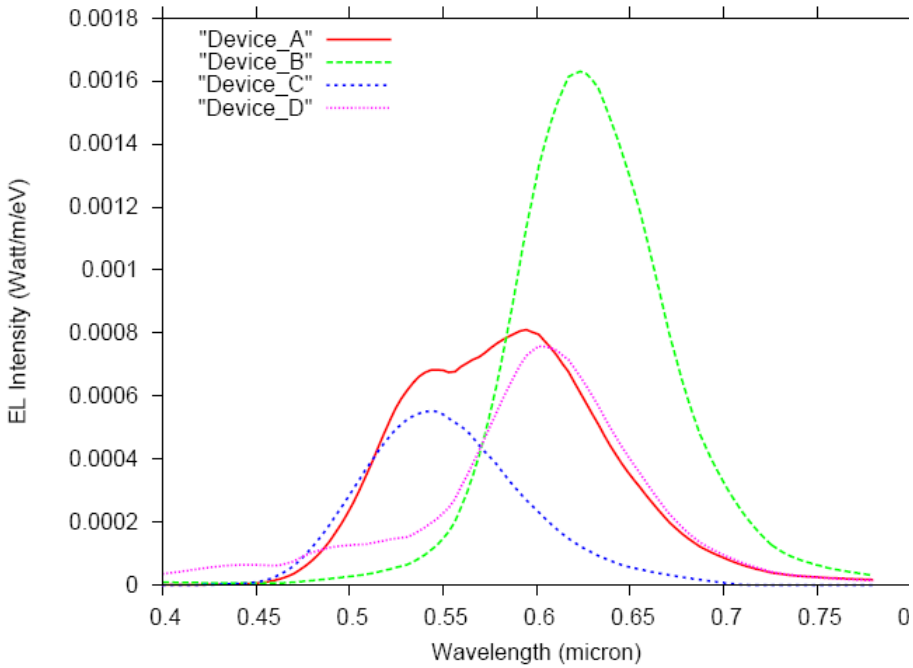


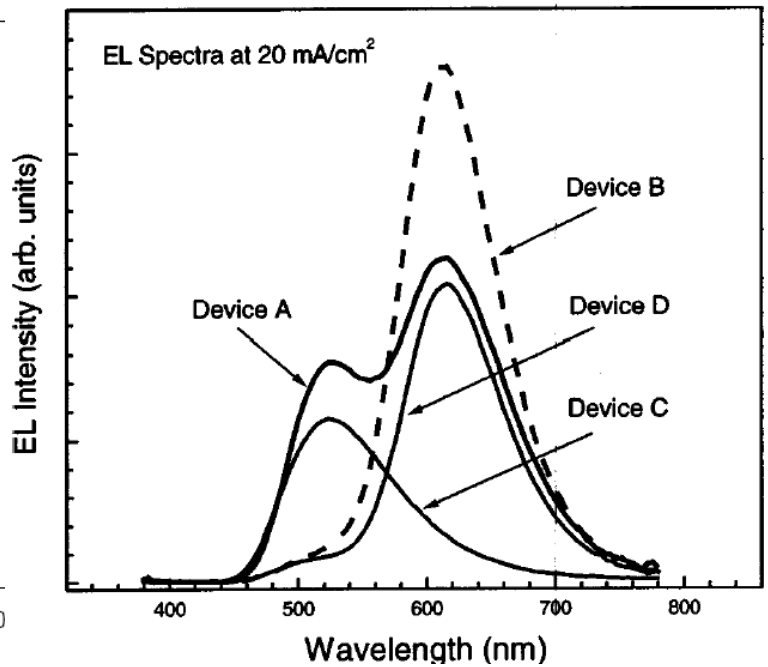
a) Band diagram at 4 volt;

b) at 14.2 volt;

c) Exciton distribution at 14.2 volt.

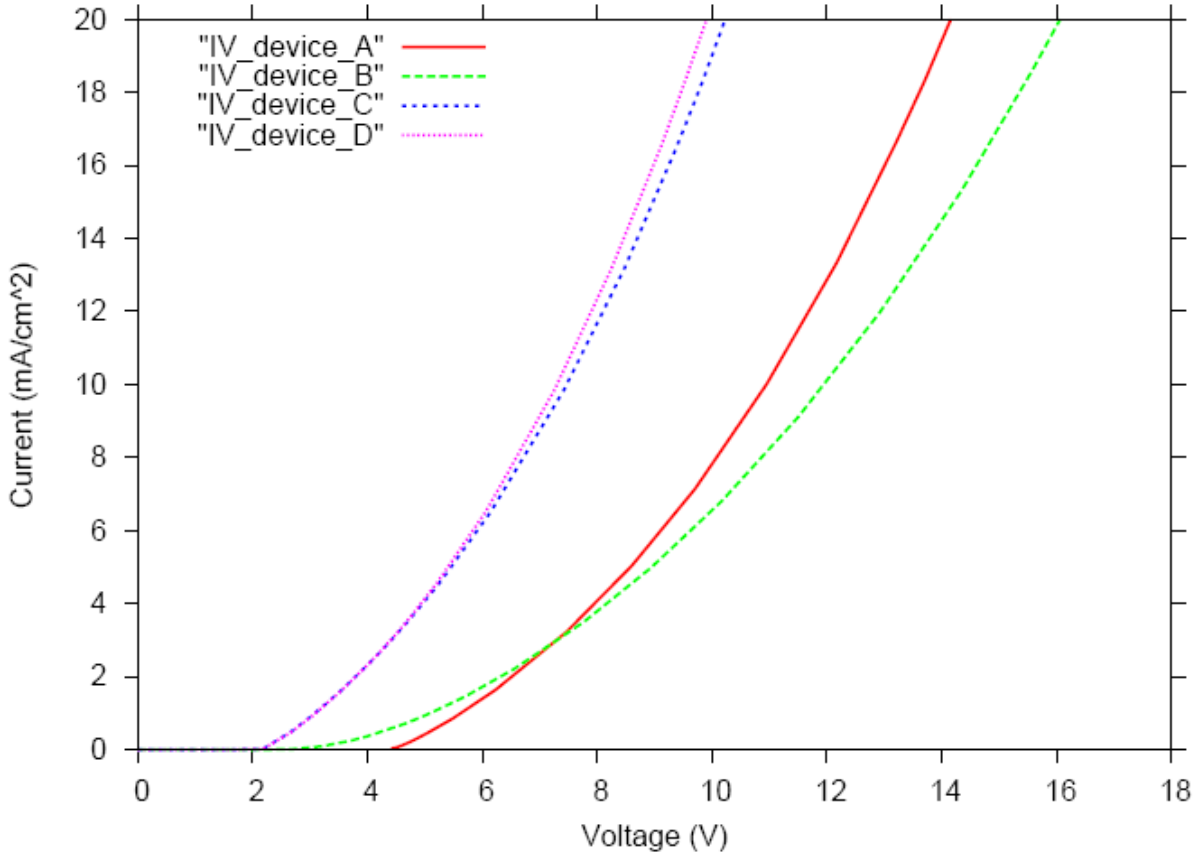
Simulated EL spectrum





Optical phase in microcavity model, band alignment and mobility were found to affect the color and intensity of the EL spectra. Reason results were obtained after careful choice of parameters.

Simulated IV curves



Experimental findings: Device A with tunnel junction (TJ), V=14.2 V at 20 mA/cm^2; Device B, V=16 V at 20 mA/cm^2 .
➔ Electrical doping and TJ reduces operating voltage by about 2 volts.

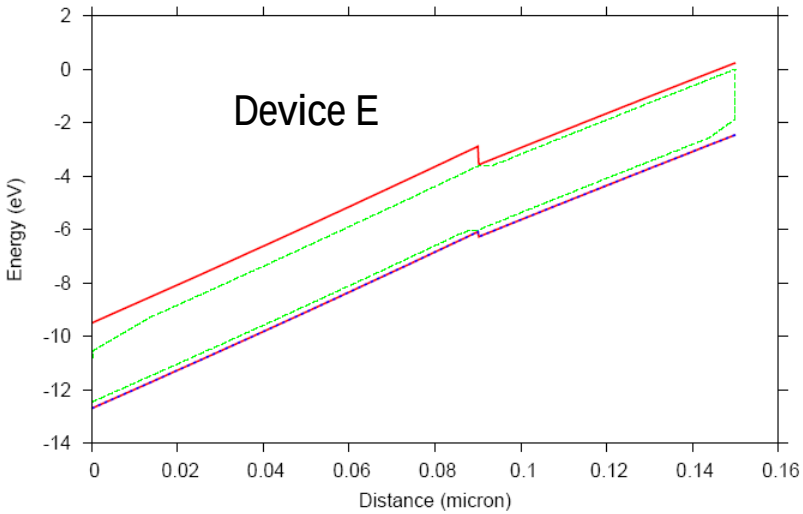
More tandem structures

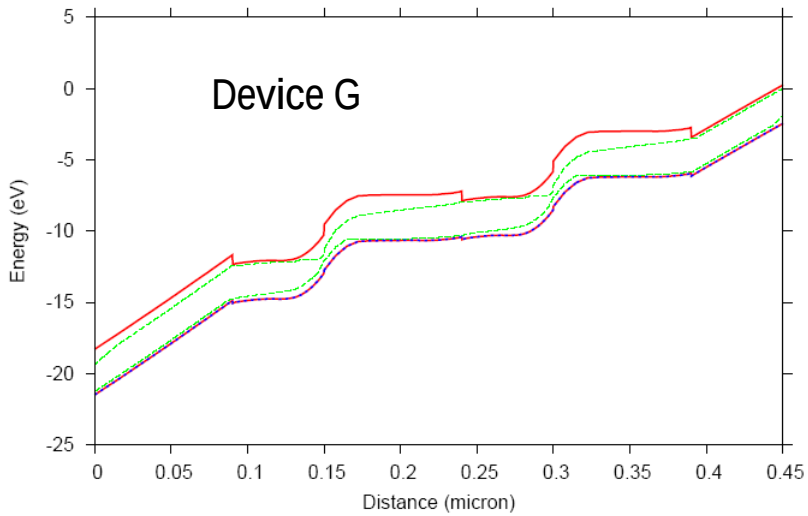
Device or unit	Layer structure
Device E	ITO/SA2/**EL2**/SC2/Mg:Ag
Device F	ITO/SA2/**EL2**/OC2/**EL2**/SC2/Mg:Ag
Device G	ITO/SA2/**EL2**/OC2/**EL2**/OC2/**EL2**/SC2/Mg:Ag
Device H	ITO/SA2/**EL2**/**EL2**/**EL2**/SC2/Mg:Ag
SA2	NPB (60 nm)
EL2	NPB (30 nm)/Alq:C545T (20 nm)/Alq (10 nm)
OC2	Alq:Li (30 nm)/NPB:$FeCl_3$ (60 nm)
SC2	Alq (30 nm)

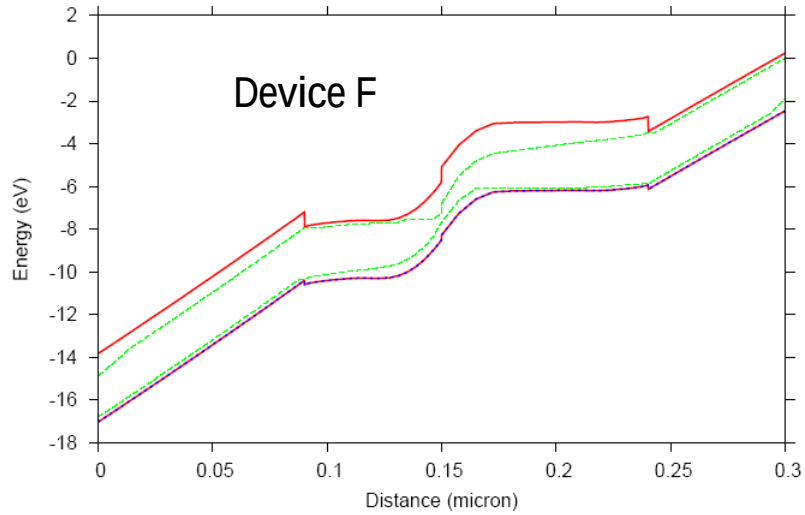
Based on Liao, Klubek, and Tang, "High-efficiency tandem organic light-emitting diodes," Appl. Phys. Lett., Vol. 84, No. 2, 12 January 2004, p.167

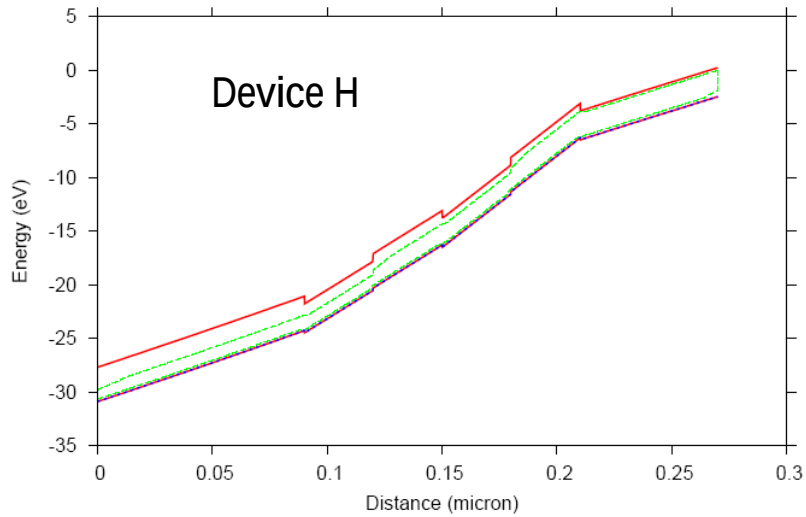
Model requirements: treatment of n-type and p-type dopings, tunnel junction. Microcavity effects neglected for simplicity here.

Band diagrams at 20 mA/cm^2

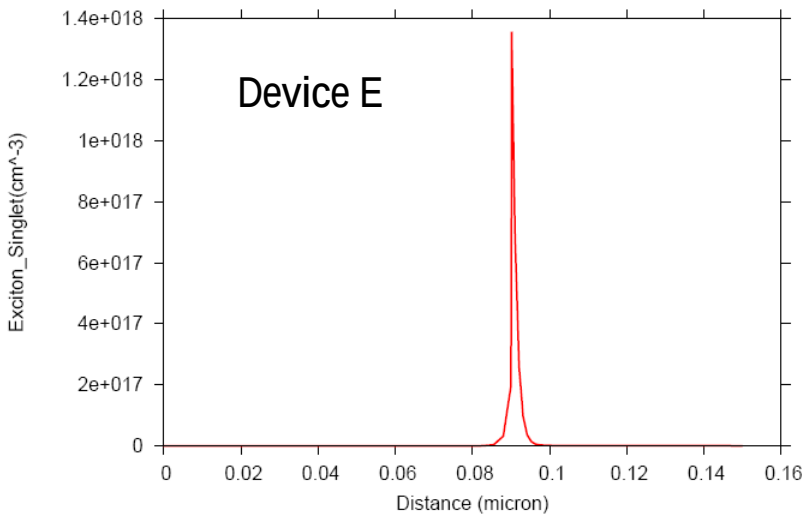


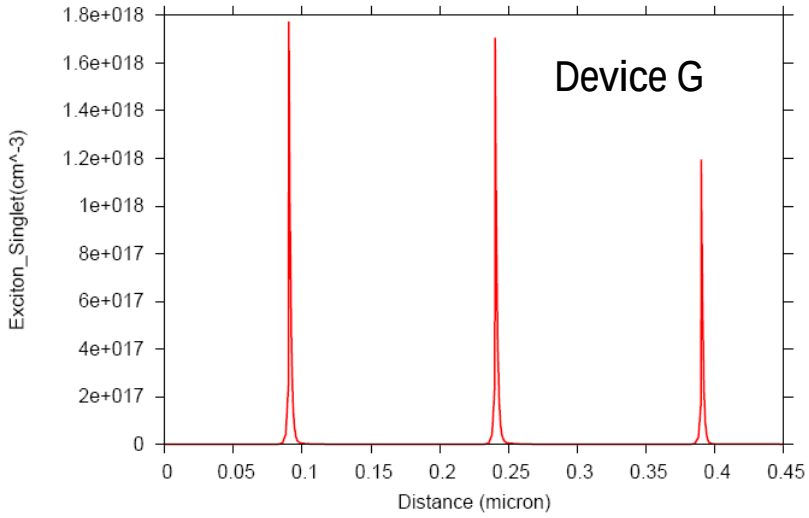


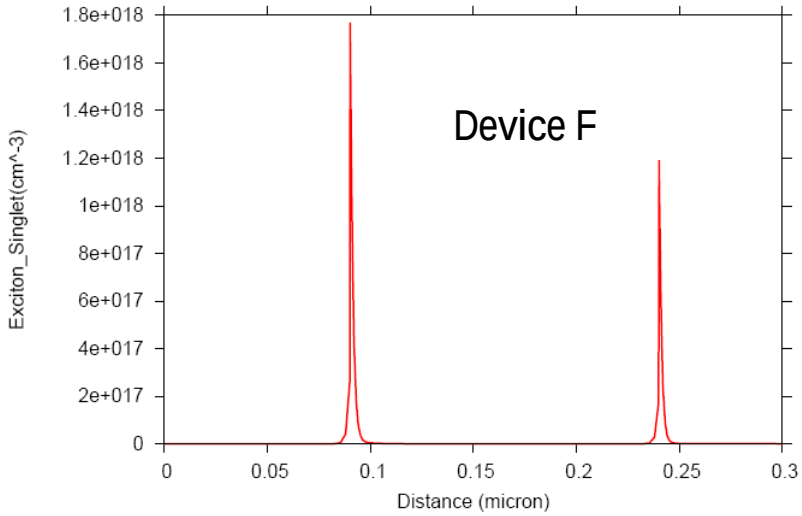


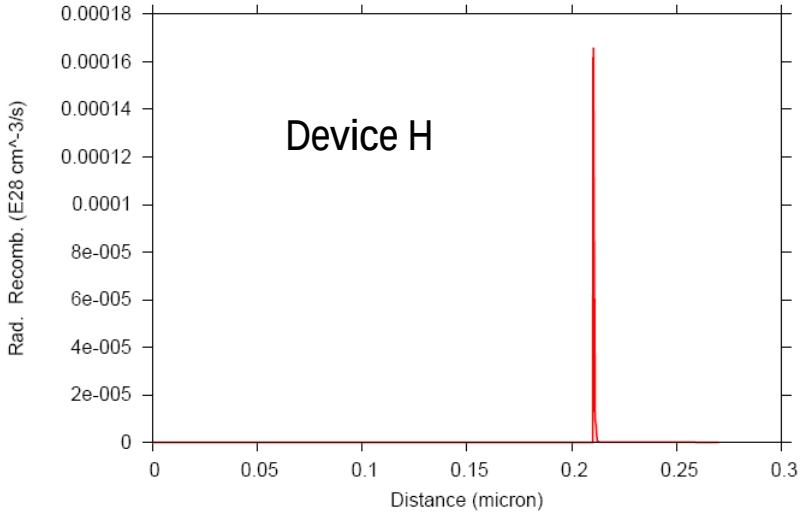


Exciton at 20 mA/cm^2

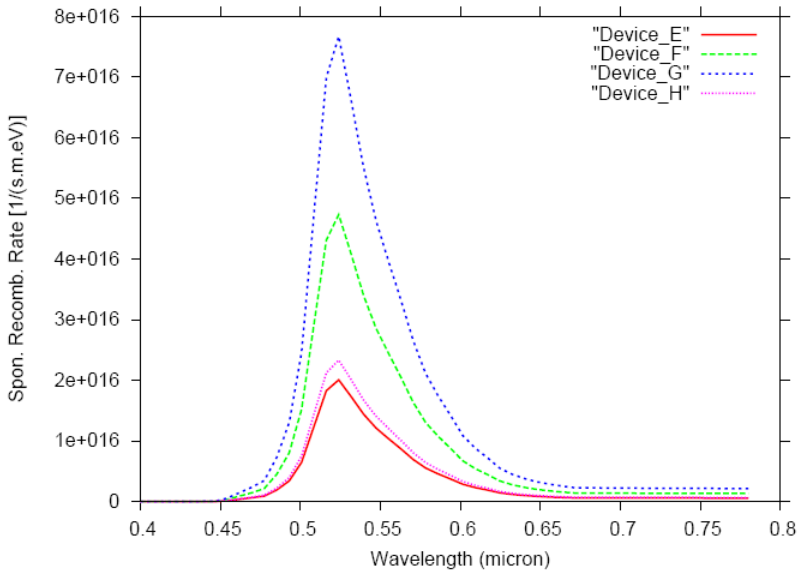


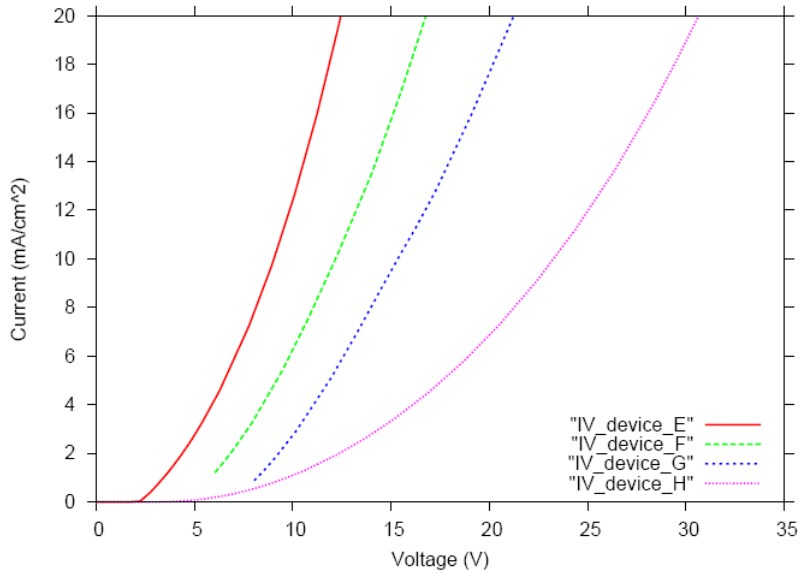


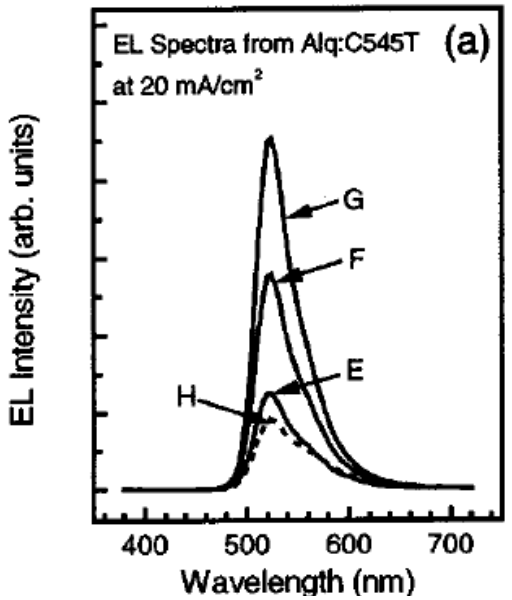


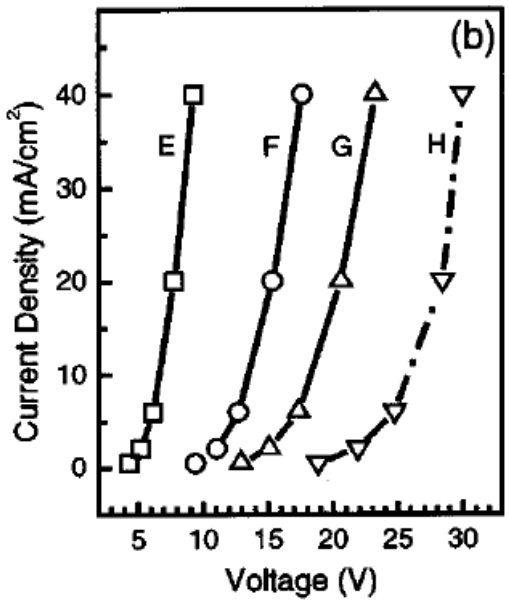


Simulated EL / IV vs experiment









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Conclusions

- A rather comprehensive set of modules suitable for OLED and WOLED simulation implemented in APSYS for both electrical and optical modeling.
- Each module may be used independently or in a fully integrated manner.
- All critical modules have been verified by experiments.