Modeling Transport in Nanostrucutred Optoelectronics

Ian Rousseau[1], Vladimir Bulovic[2], Jeffrey Grossman[3]

Massachusetts Institute of Technology

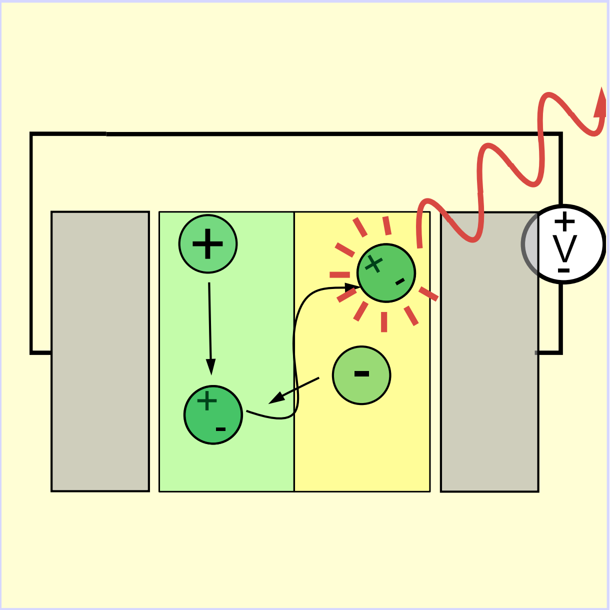
[1]Dept. of Physics

[2]Dept. of Electrical Engineering and Computer Science

[3]Dept. of Materials Science and Engineering

MOTIVATION:

- Nanostructured optoelectroncs: future potential for low-cost, large area, tunable LEDs and solar cells
- Connect macroscopic behavior to microscopic model
- Suggest methods to improve efficiency
- Device physics educational tool



PRESENTATION GOALS:

- Introduce physics in model
- Show reasonable input parameters
- Provide example results

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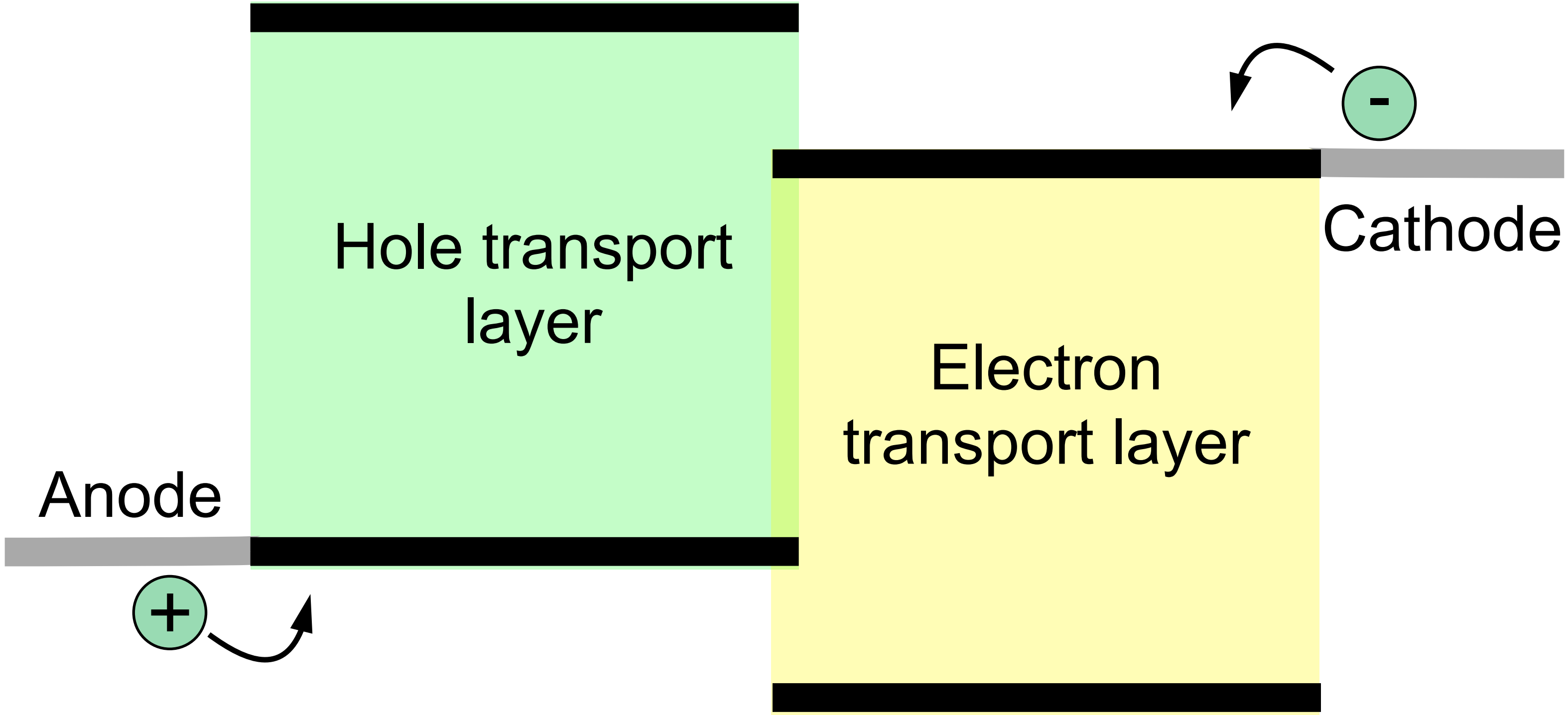
METHOD - MICROSCOPIC PROCESSES:

- Relevant processes
  - *Charge injection*: thermionic injection into broadened density of states
  - *Charge transport*: thermally-activated hopping (Miller-Abrahams or Marcus)
  - *Exciton formation*: photon absorption and charge recombination (Langevin)
  - *Exciton transport*: thermally-activated hopping or Förster energy transfer
  - *Exciton decay*: radiative, non-radiative, and dissociation into charge pairs



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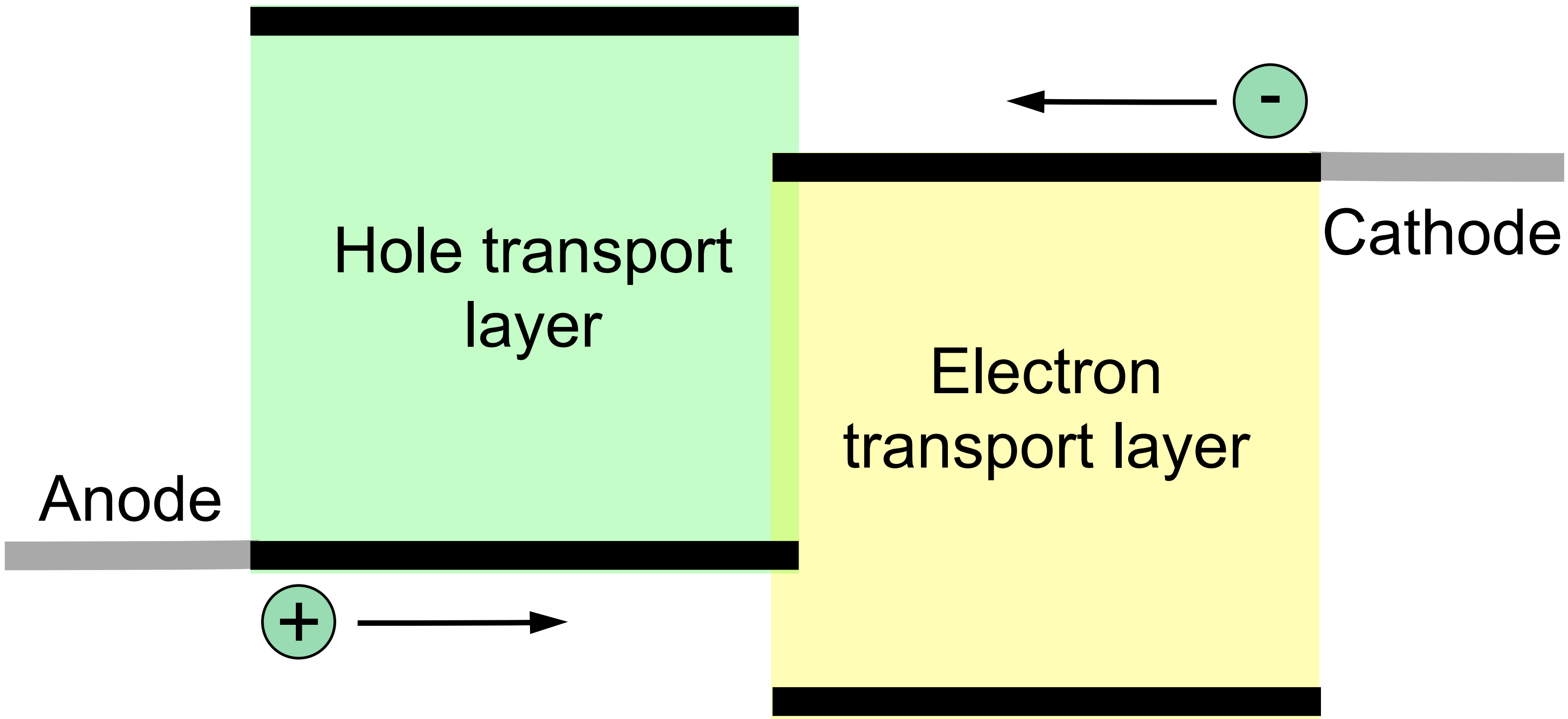

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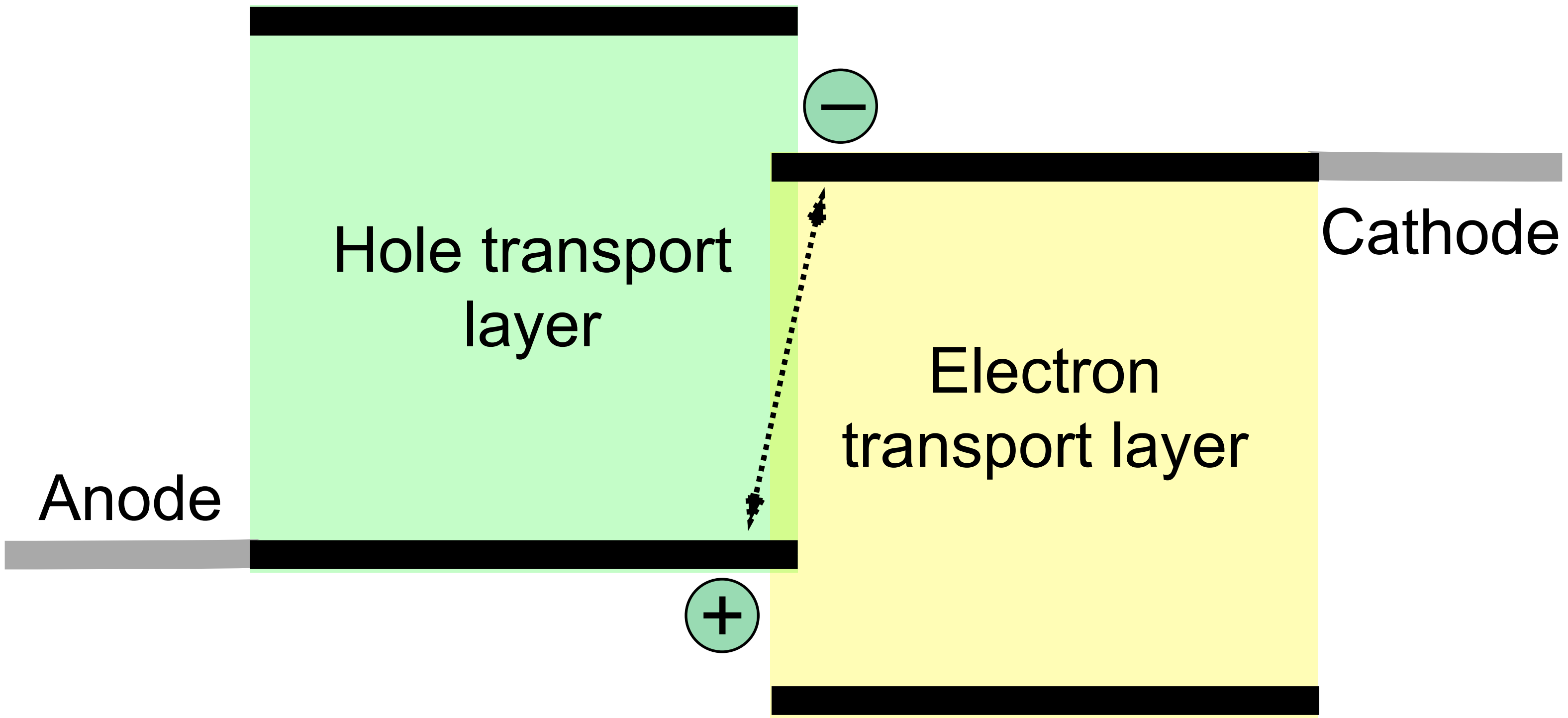

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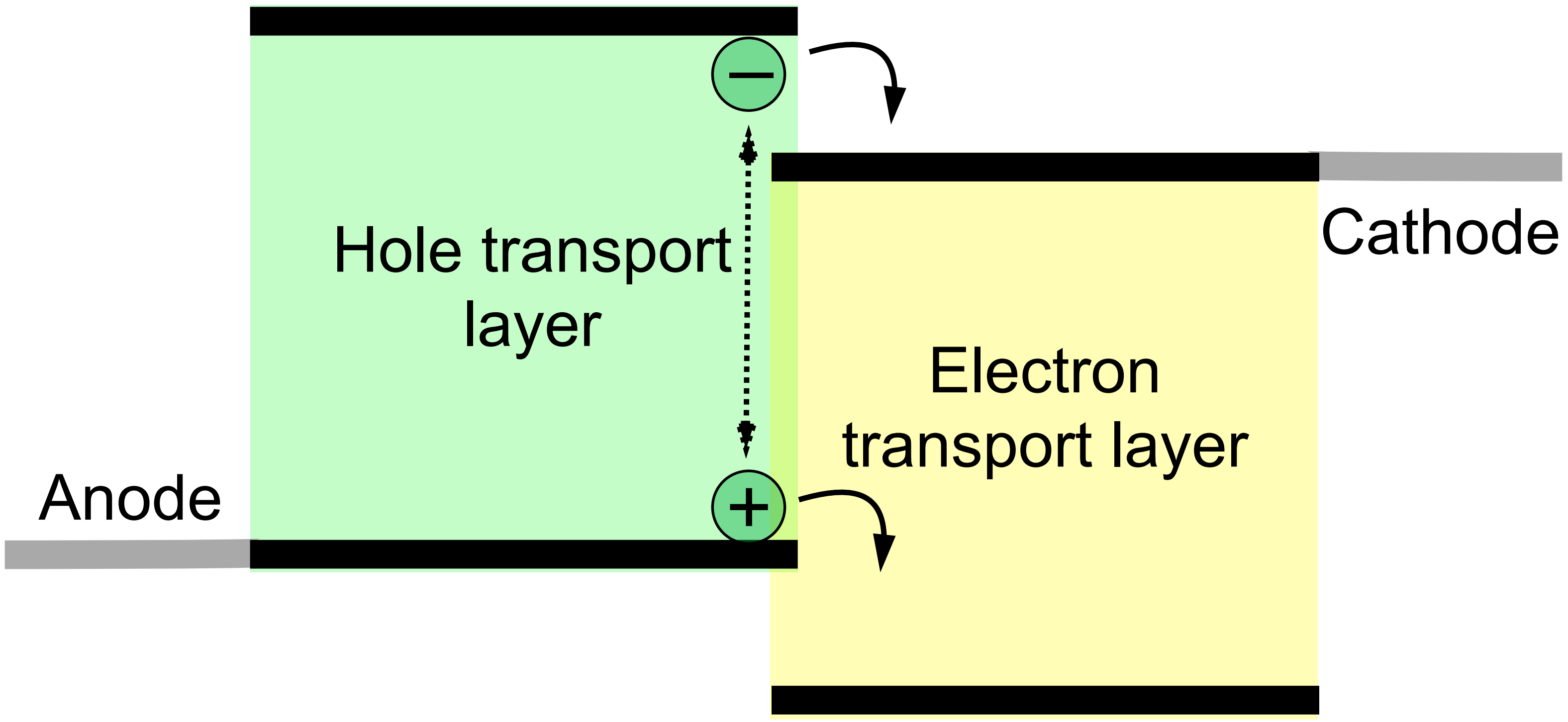

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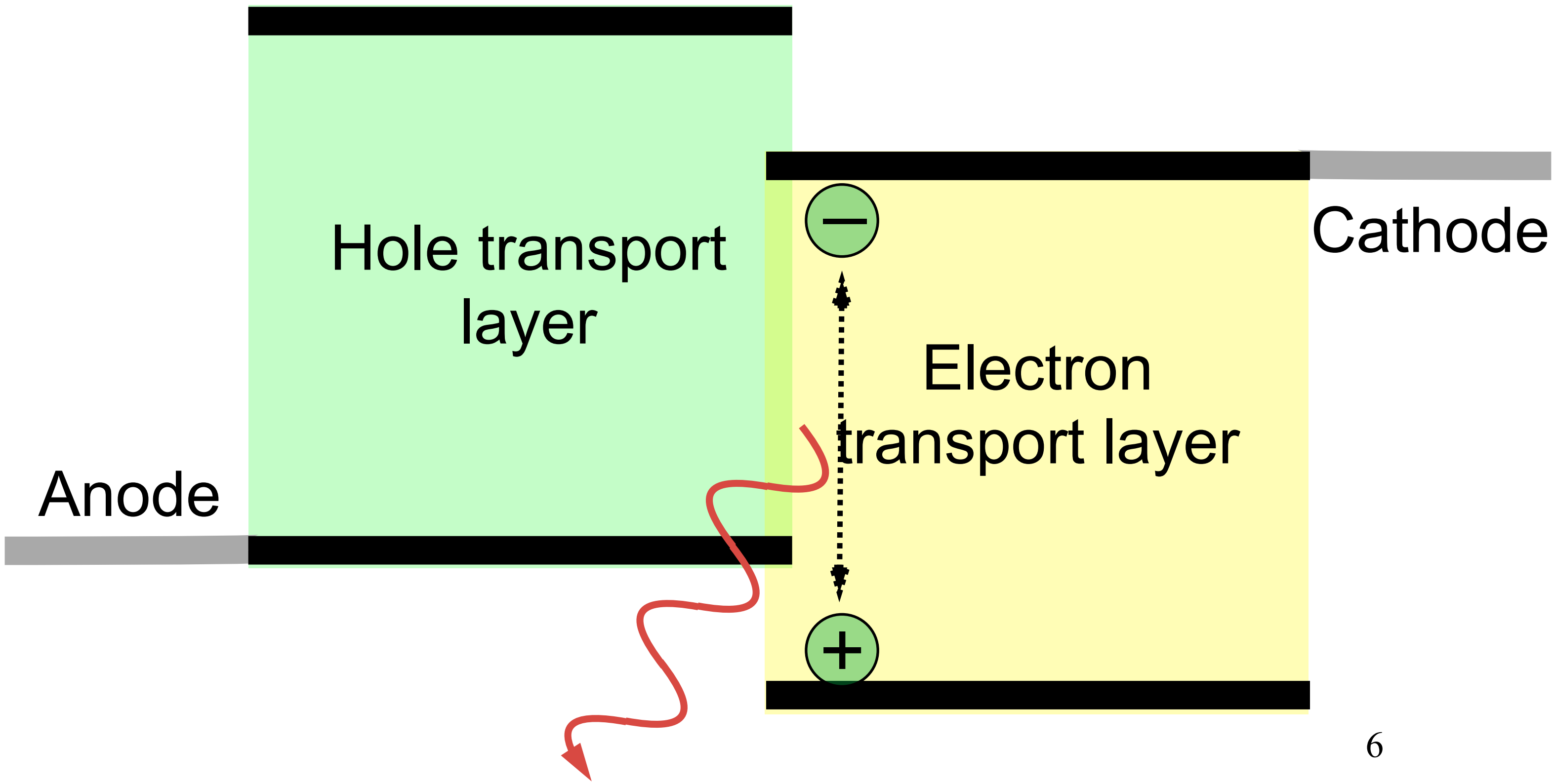
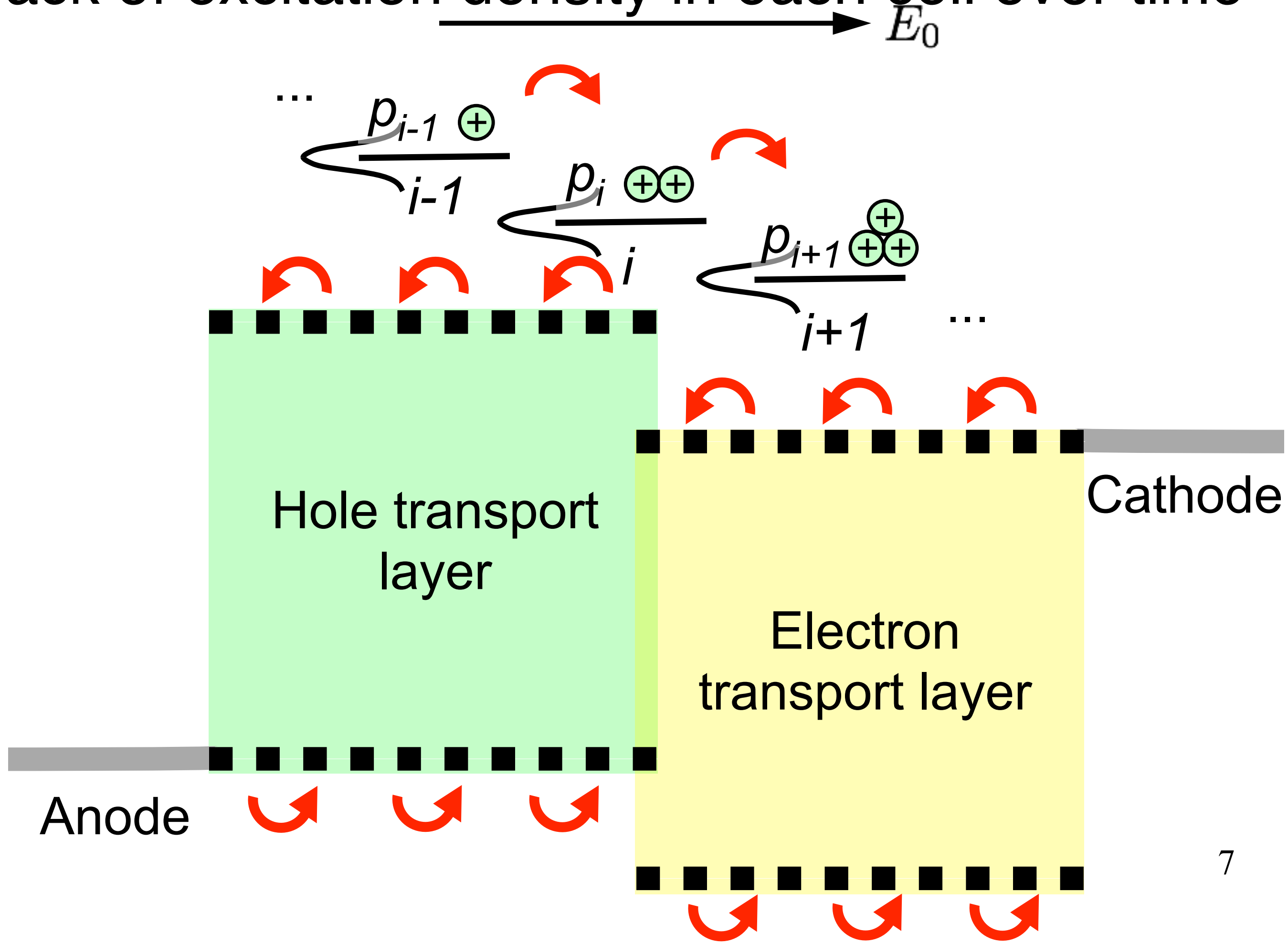


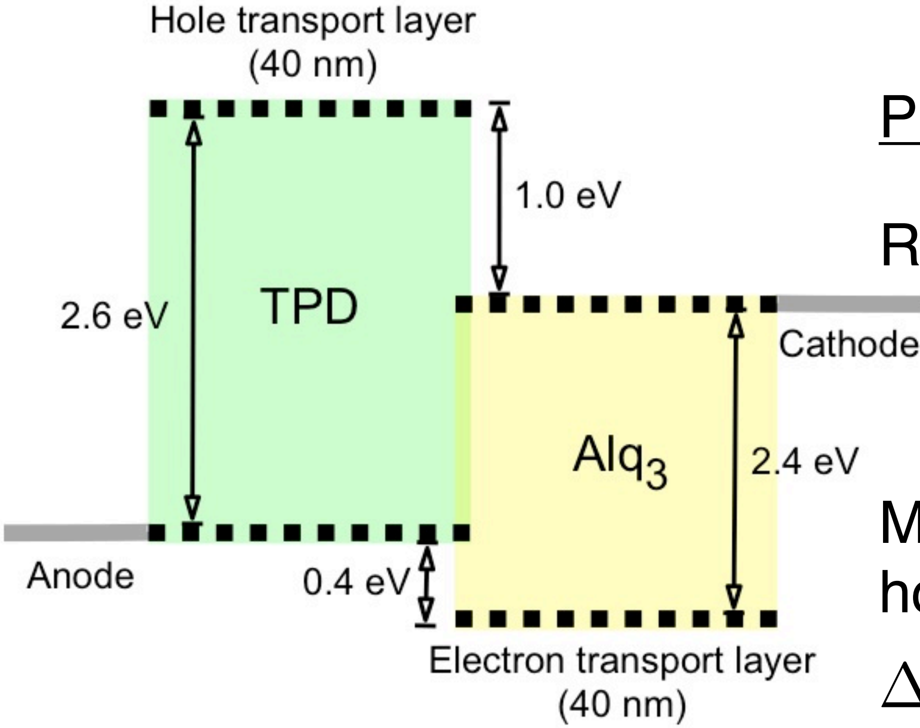
METHOD - NUMERICAL SIMULATION:

- Most transport occurs along one-dimension
- Excitations move between discrete unit cells
- Employ rate equations and add applicable rate processes
- Keep track of excitation density in each cell over time



EXAMPLE 1: BILAYER OLED

40 nm TPD/40 nm Alq_3



Processes enabled by default:

Radiative exciton decay:

$$K_x = \frac{1}{\tau_d}$$

Miller-Abrahams thermal hopping* for holes, excitons, and electrons**:

ΔE due to change in HOMO/LUMO/exciton energies, electrostatic, and polarization potentials

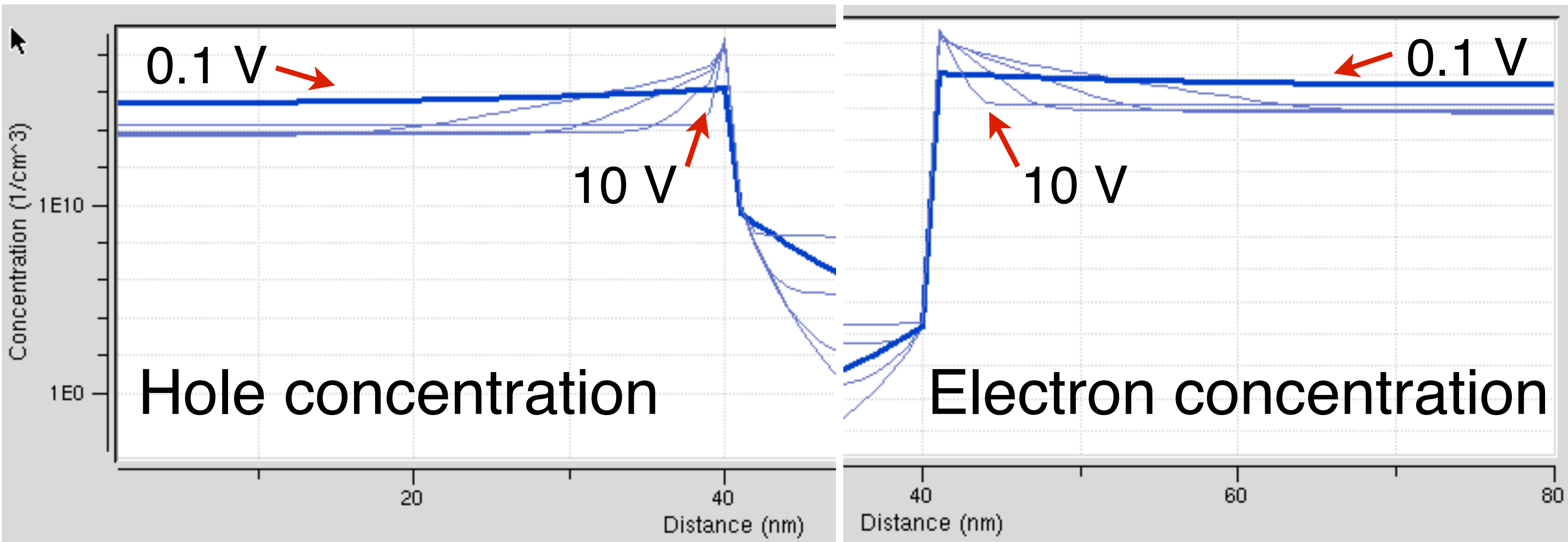
$$K = \mu kT / e d_{\mathrm{mol}}^2 \cdot \begin{cases} 1 & : \Delta E <= 0 \\ \exp(-\Delta E / kT) & : \Delta E > 0 \end{cases}$$

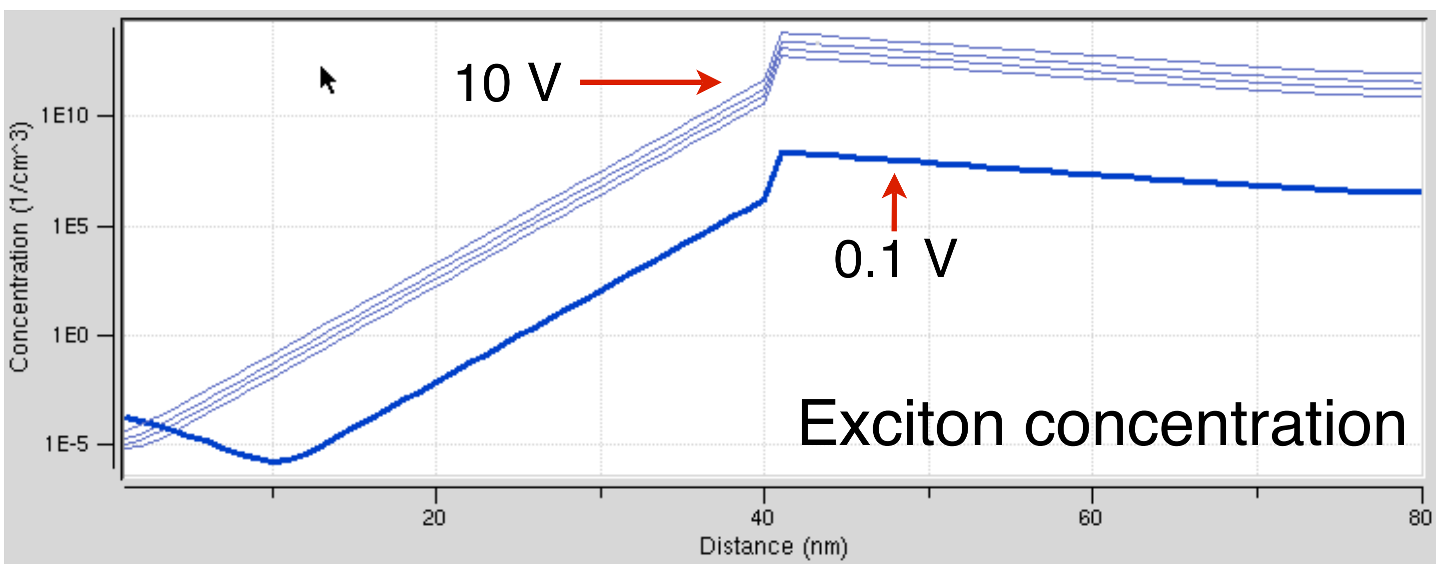
*reduces to drift-diffusion equations in limit of low field

**between neighboring molecules/nanoparticles in a delta function density of states

EFFECT OF BIAS

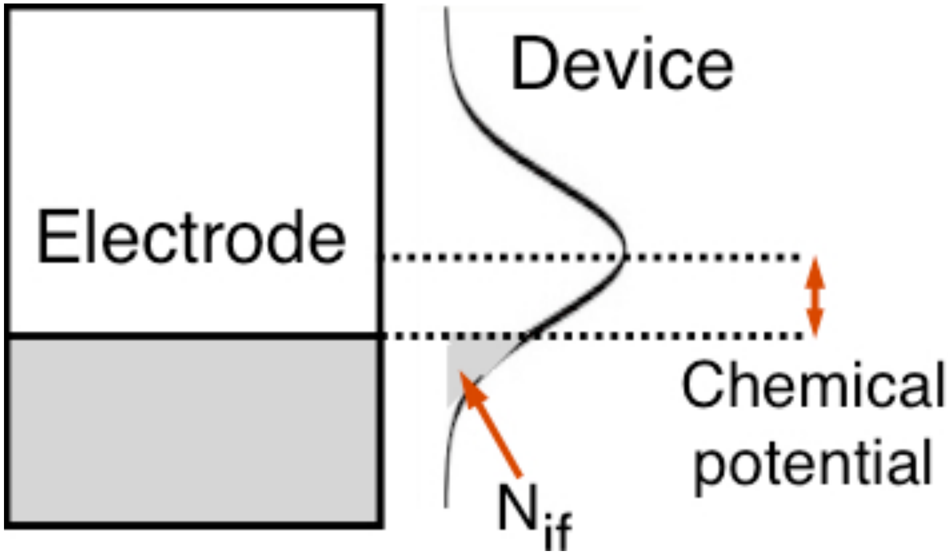
Results: carriers pile up at interface
more excitons created

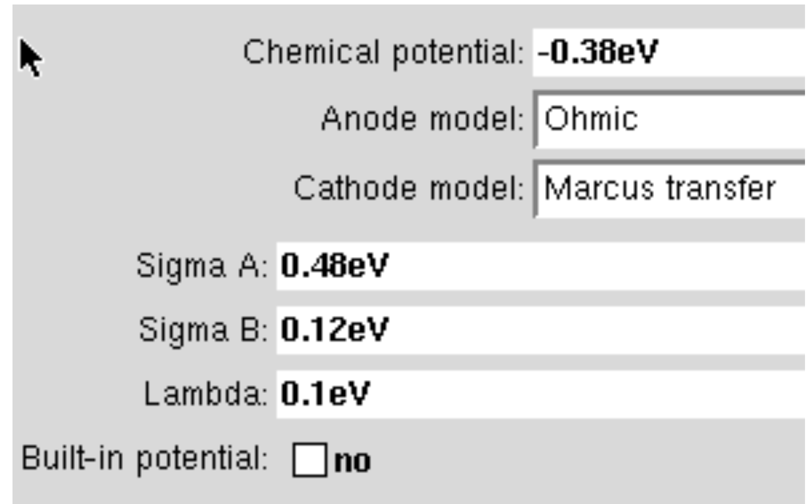




INJECTION

How charges enter and exit the device





Assume Gaussian density of states of width 0.1 eV in first device layer:

$$N_{if} = N_{\mathrm{dos}} \mathrm{erf}(\mu/\sigma)$$

Model #1: Ohmic injection

$$J = F \cdot \mu kT/ed_{mol} \cdot \begin{cases} N_{if} & : F > 0 \\ N_0 & : F \leq 0 \end{cases}$$

Where F is the electric field and N_0 is the concentration in the first device layer

INJECTION

How charges enter and exit the device

Model #2: Thermionic injection

Injection over a thermal barrier of height $\Delta E = \frac{4\pi e^2}{\epsilon} N_{if} d^2_{mol}$ with field-dependent recombination current.

See Scott and Malliaras, CPL 299 (1999) pp 115-119

Model #3: Marcus theory

- Move from first to second device layer is rate-limiting step
- Assume DOS broader at interface (A) than in first layer (B)
- Assumes degenerate carrier distribution and approximates integral for *net* transfer rate*
- Includes recombination current
- Current depends on field and chemical potential
- Broadening introduces a built-in potential

*Limketai and Baldo, PRB 71 (2005) 085207 is the starting point for this model

LANGEVIN RECOMBINATION

hole + electron ⇒ exciton

$$R = \gamma n p$$

Two types:

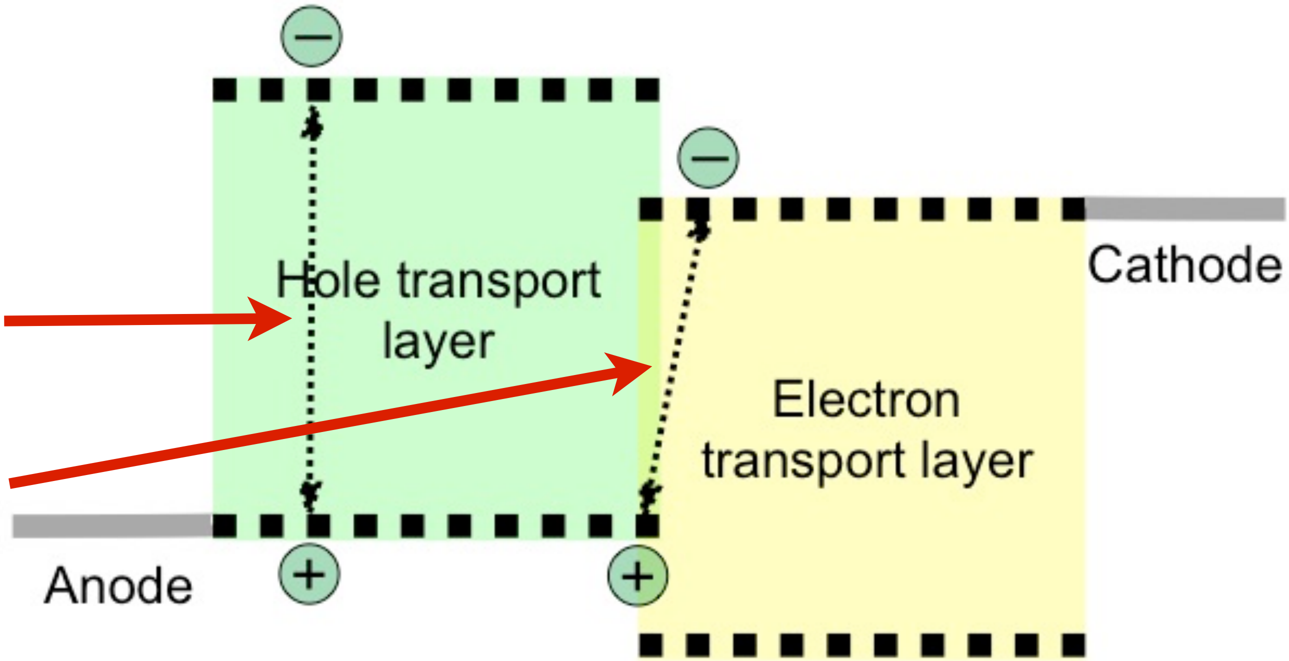
Monomolecular

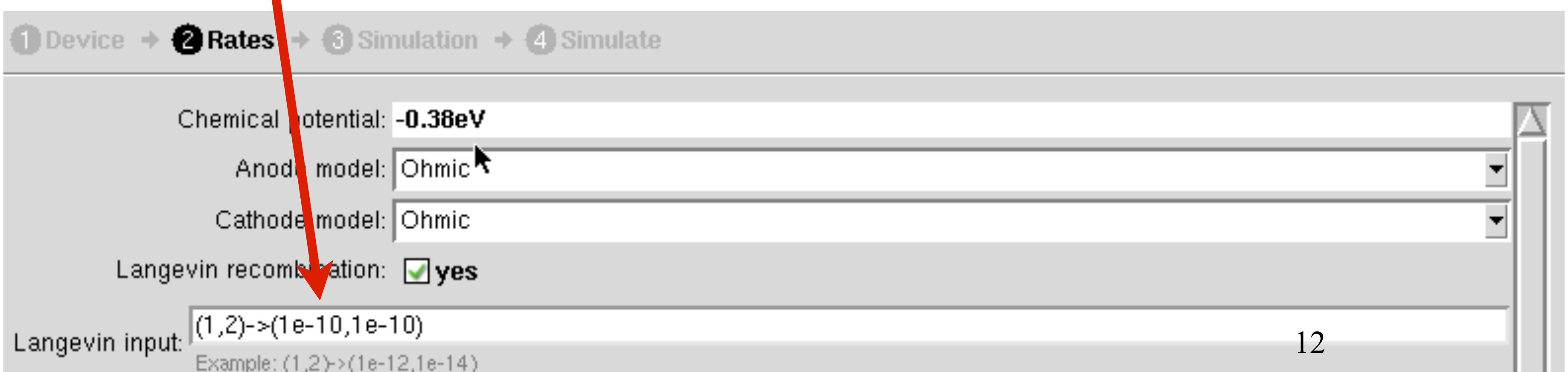
(from mobility)

$$\gamma = \frac{e}{\epsilon}(\mu_p + \mu_n)$$

Bimolecular

(user-specified [cm^3/s])

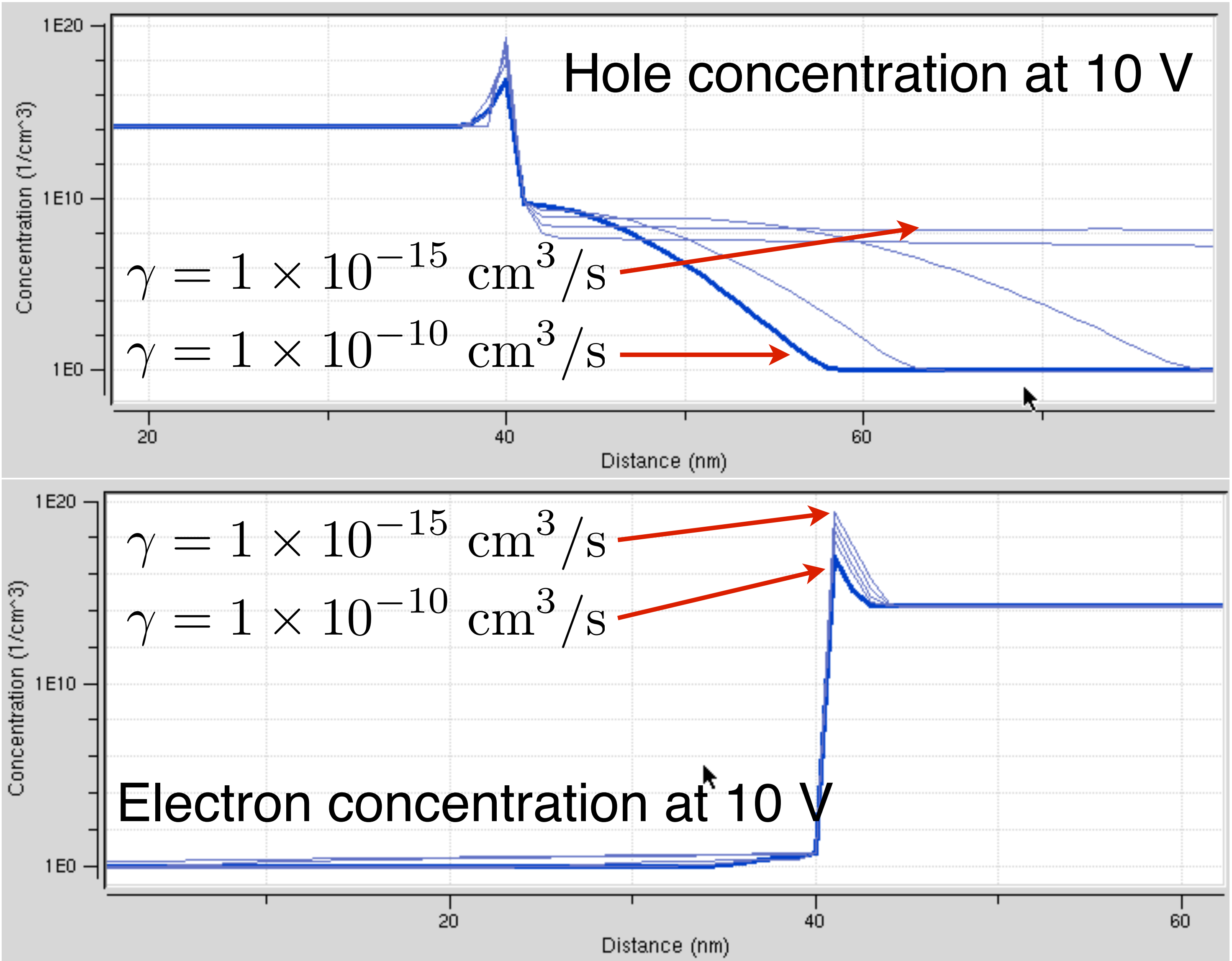




LANGEVIN RECOMBINATION

hole + electron ⇒ exciton

Result: Higher recombination ⇒ fewer charges at interface

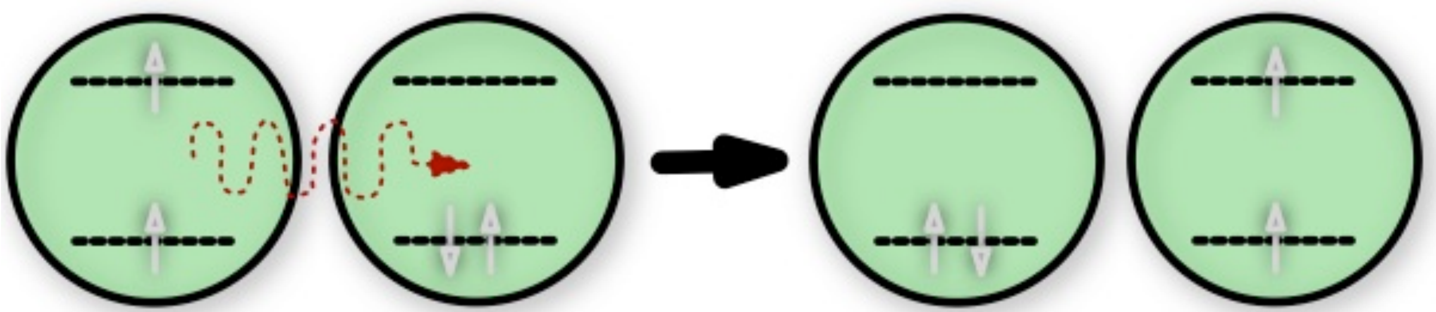


FÖRSTER RESONANT ENERGY TRANSFER

long-range exciton transport

$$K_{D\rightarrow A} = \frac{3c^4}{4\pi\epsilon_D}\frac{1}{\tau_D}\frac{1}{R^6}\int \frac{1}{\omega^4}F_D(\omega)\sigma_A(\omega)d\omega$$

Integral describes overlap between donor emission and acceptor absorbance spectra



Singlet excitons move via non-radiative dipole-dipole coupling

User specifies the effective transfer radius $R_{D\rightarrow A}$ [cm]

τ_D is the exciton natural lifetime

$$K_{D\rightarrow A} = \frac{1}{\tau_D}\left(\frac{R_{D\rightarrow A}}{R}\right)^6$$

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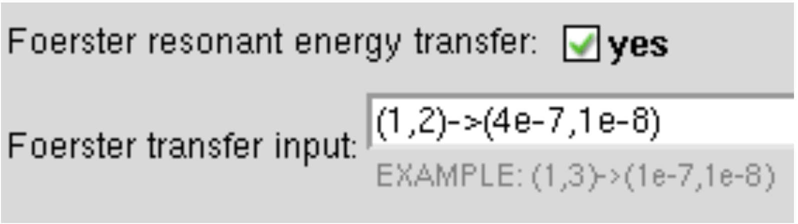
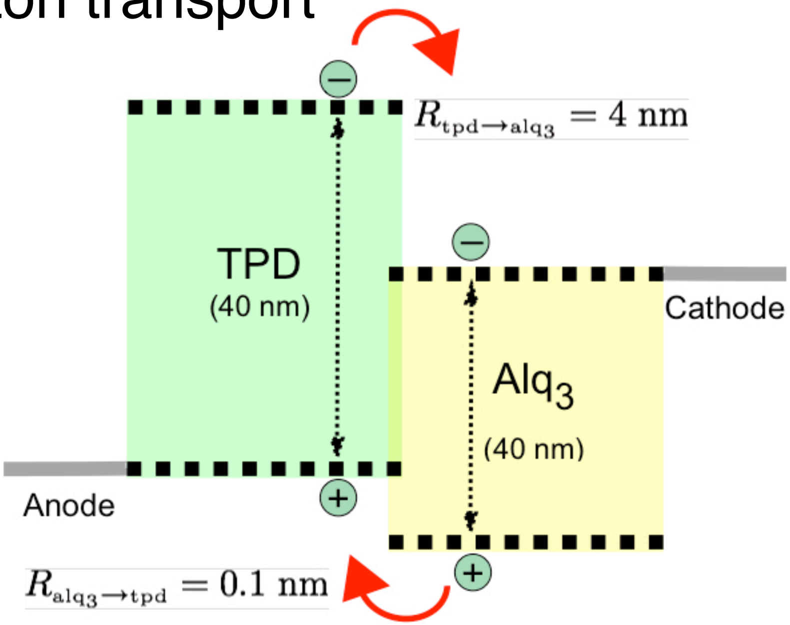
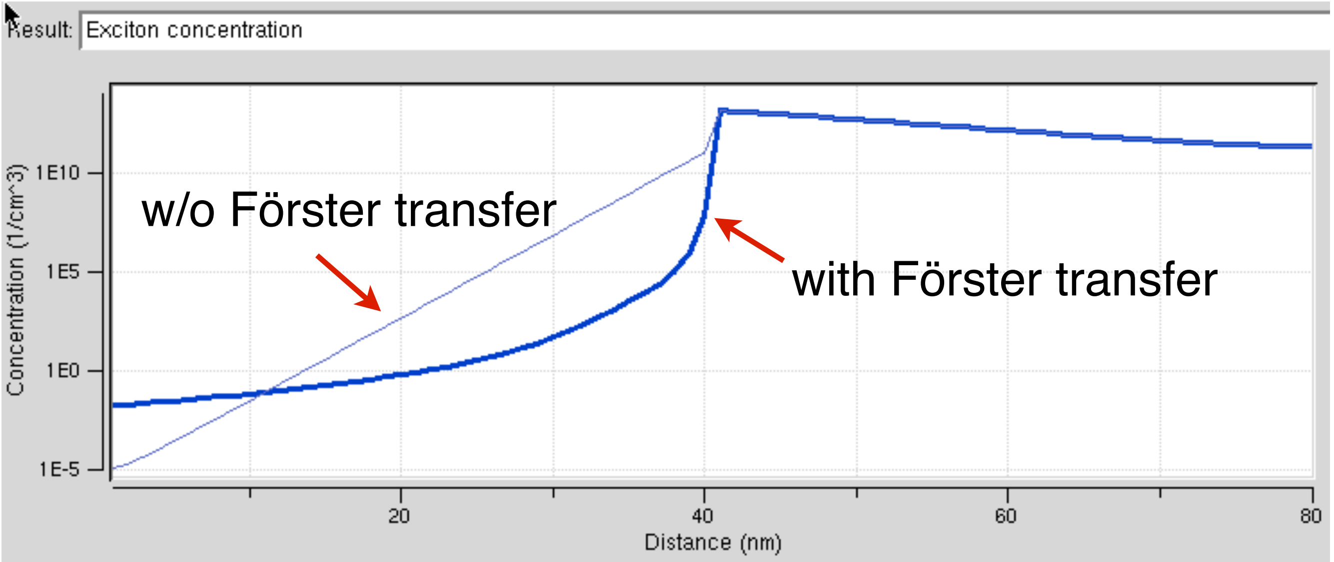


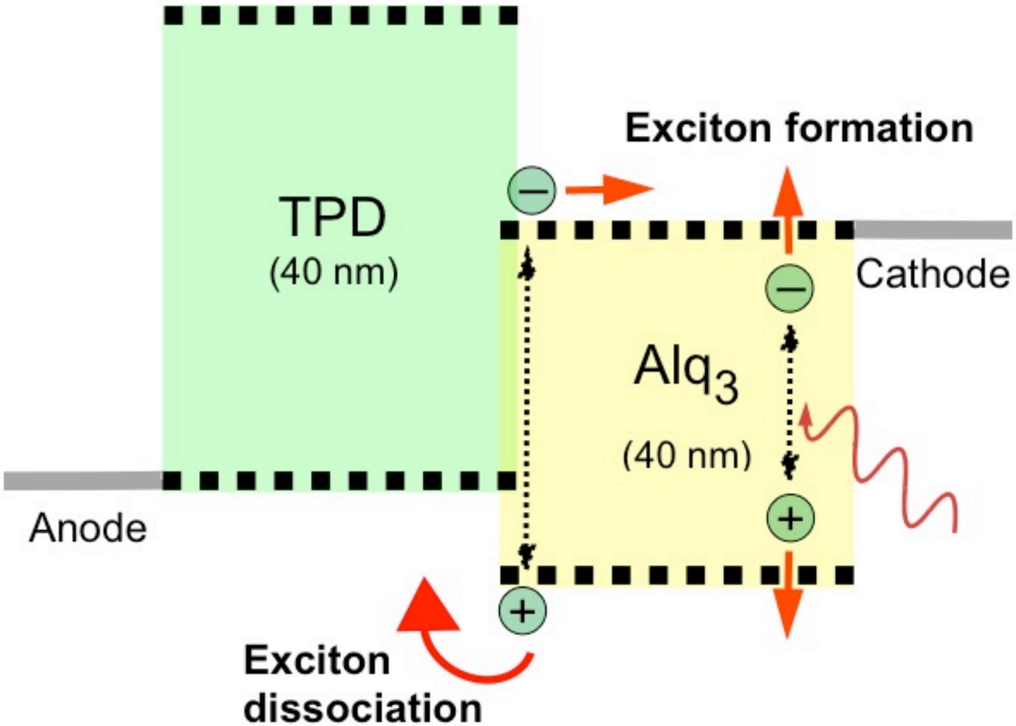


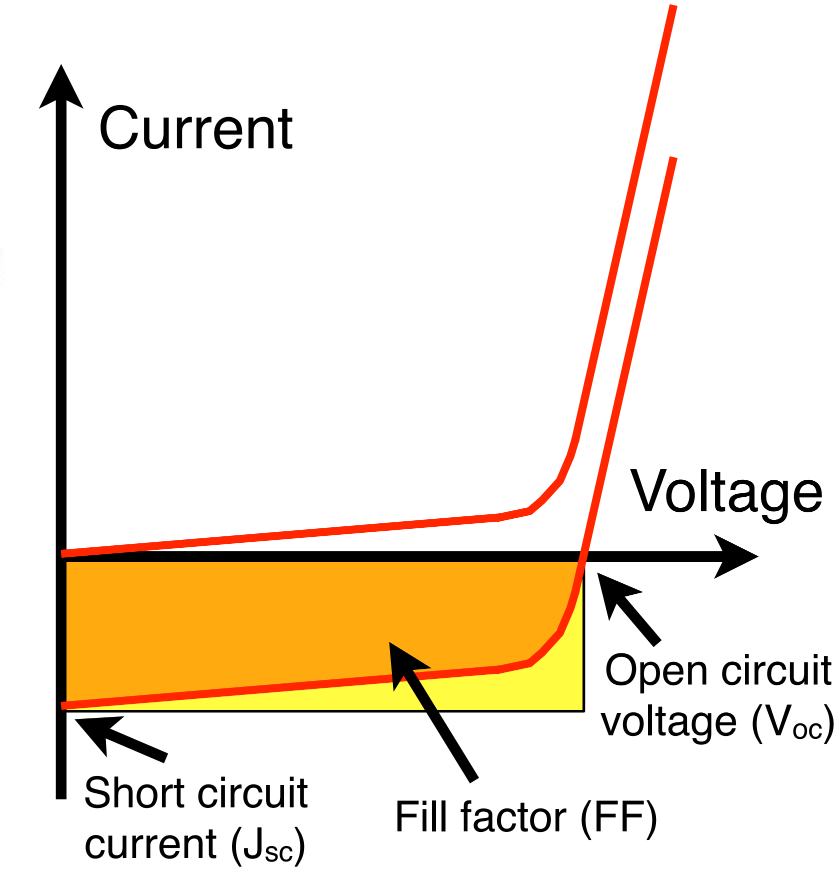
Result: fewer excitons in TPD



EXAMPLE 2: SOLAR CELL

exciton generation and dissociation





Exciton dissociation: ☑ yes

Exciton dissociation input: (1,2)->(1e0,1e10)

Example: (1,2)->(1e20,1e18); (2,3)->(1e5,0)

Photo generation: ☑ yes

Photo generation input: 2->1e23

EXAMPLE: 1->1e20; 2->1e24

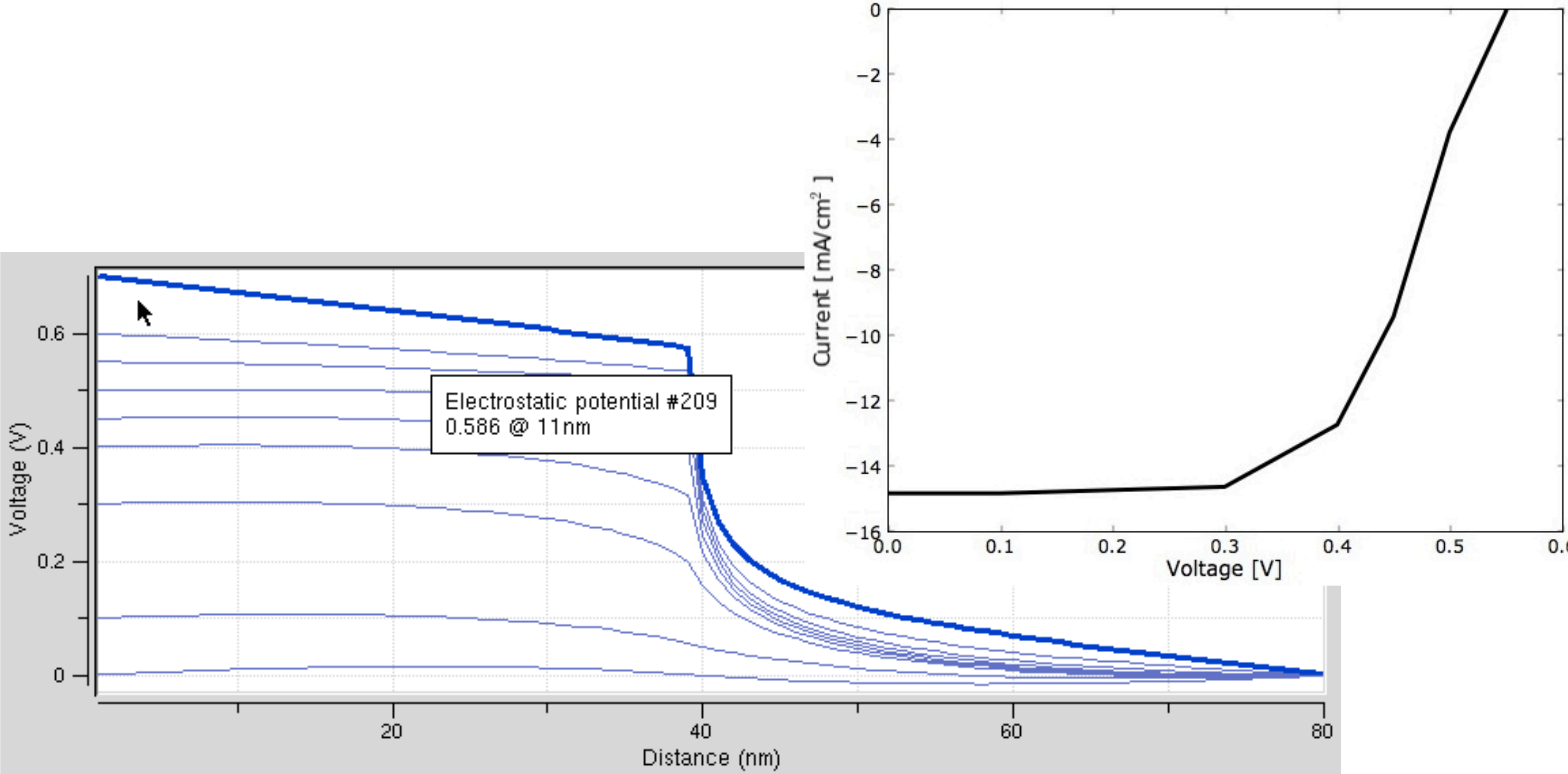
$$\text{Efficiency } [\%] = 100 \cdot \frac{J_{sc} \cdot V_{oc} \cdot FF}{I_{sun}}$$

Constant rate: exciton ⇒ free charge [1/s]

Constant rate: exciton creation [1/cm³-s]

SOLAR CELL

exciton generation and dissociation



Results: Reproduce current-voltage curve

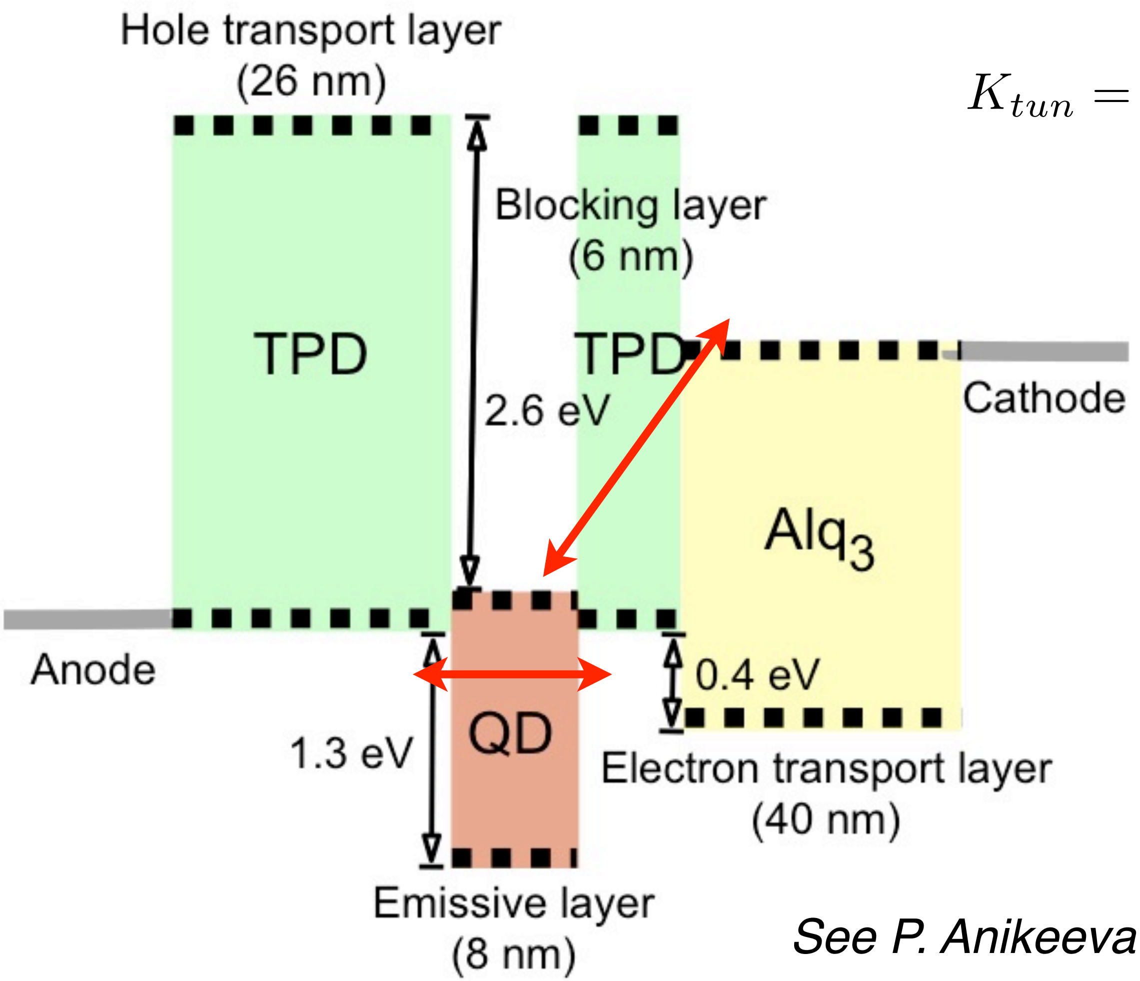
Voltage dropped near interface due to large charge buildup

EXAMPLE 3: "TUNNELING" EFFECTS:

charge movement through imperfections in blocking layers

Idea: thin "blocking" layers in devices may not be complete

Sample device: TPD/quantum dot/TPD/Alq_3 QD-LED



$$K_{tun} = K_{ma}\left(1 - \frac{\pi}{2\sqrt{3}}\right)\exp\left[-\frac{\Delta x}{x_0}\right]$$

- *Geometrical factor*
- *Miller-Abrahams rate*
- *Exponential decay of wavefunction overlap*

See P. Anikeeva's PhD thesis for more information

"TUNNELING" EFFECTS:

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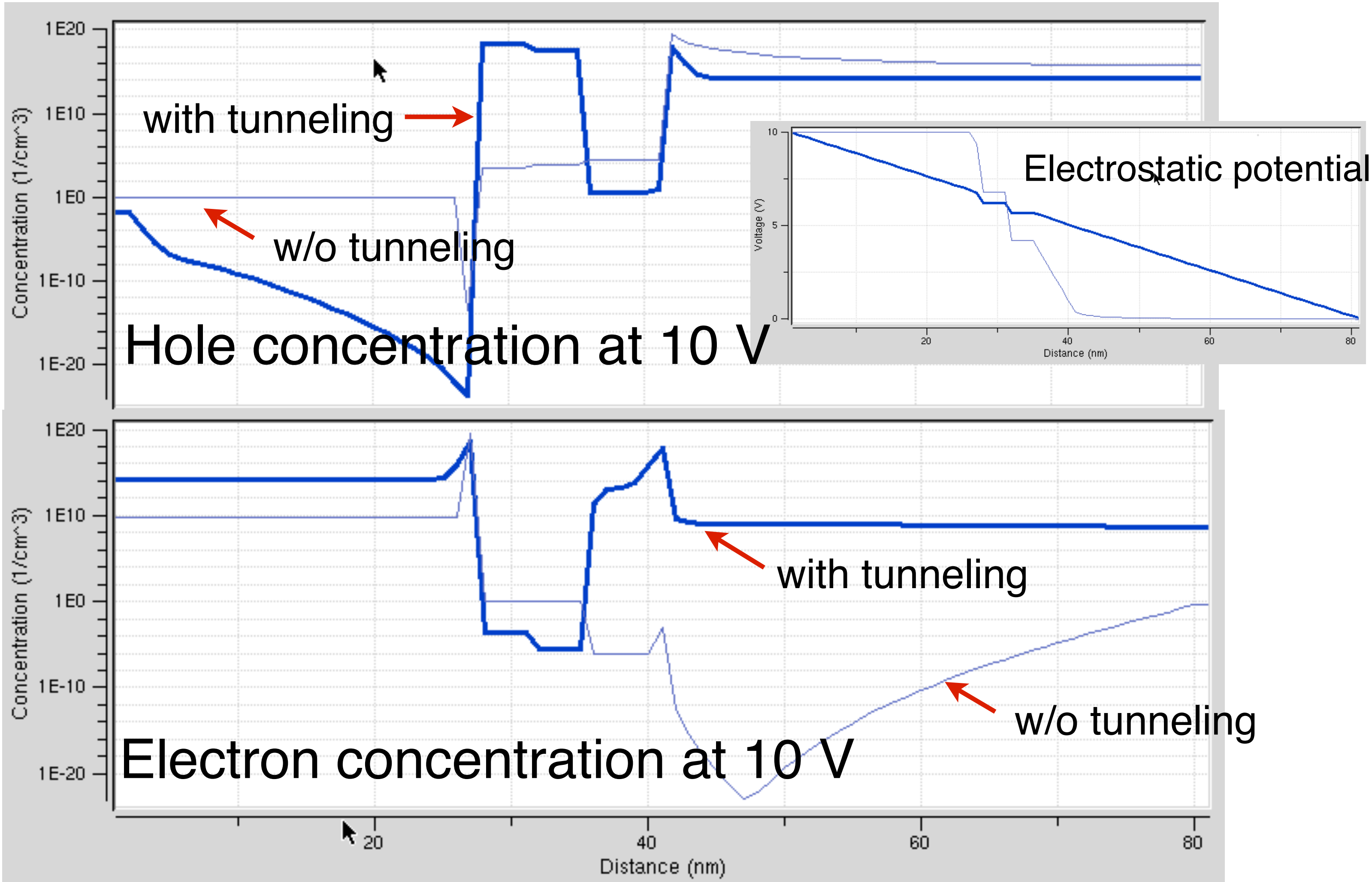

Results: Charges are able to flow ⇒ functioning device

Electric potential evenly dropped over device