Zurich University
of Applied Sciences

School of
Engineering

ICP Institute of
Computational Physics

FLUXiM

AEVIOM

Advanced Simulation Methods for Charge Transport in OLEDs

Evelyne Knapp, B. Ruhstaller

Overview

1. Introduction
2. Physical Models
3. Numerical Methods
4. Outlook

www.icp.zhaw.ch

ICP Team

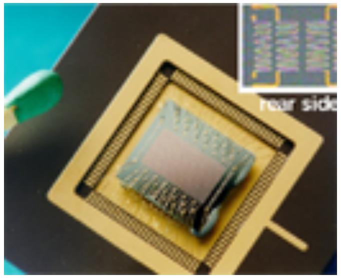
- Interdisciplinary team of 8 physicists, 4 mathematicians und 3 engineers

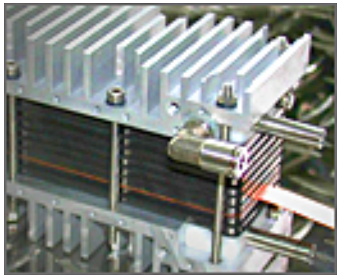
1996 Section NMSA
2002 Foundation CCP
2007 Foundation ICP

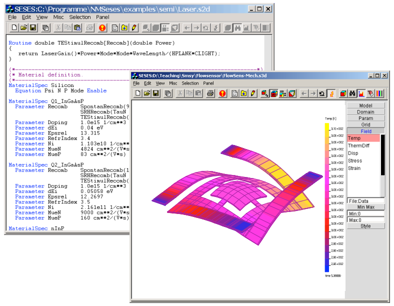
Spin-offs:
Numerical Modeling GmbH, www.nmtec.ch
Fluxim AG, www.fluxim.com

Research Activities

- The main focus is applied research and development in the following areas:
  - Micro systems, sensors, actors
  - Fuel cells
  - Organic optoelectronic and photovoltaics
  - Simulation software







AEVIOM

Advanced Experimentally Validated OLED model

Philips Research Eindhoven

Project Coordinator:
Reinder Coehoorn

Philips Research Aachen

Zürich University of Applied Sciences

Fluxim

Eindhoven University of Technology

Technical University Dresden

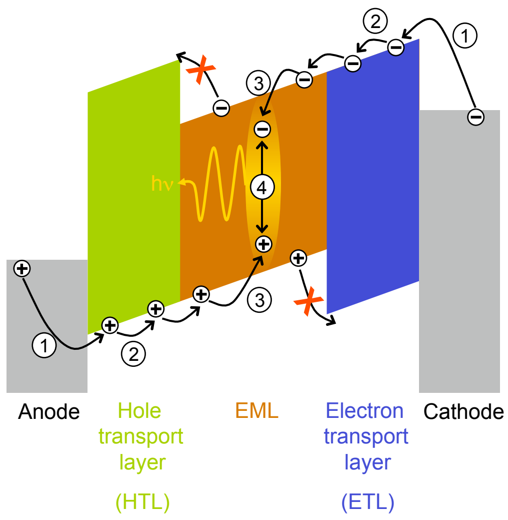
Sim4Tec

University of Groningen

University of Cambridge

Institute of Computational Physics

Principle of OLED Operation

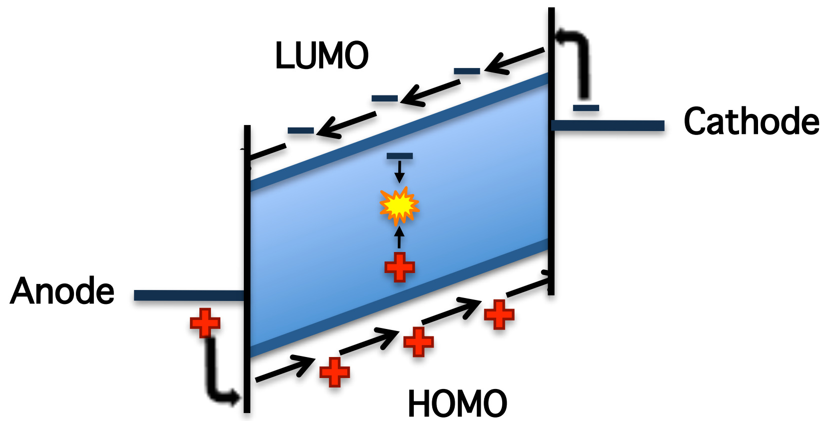


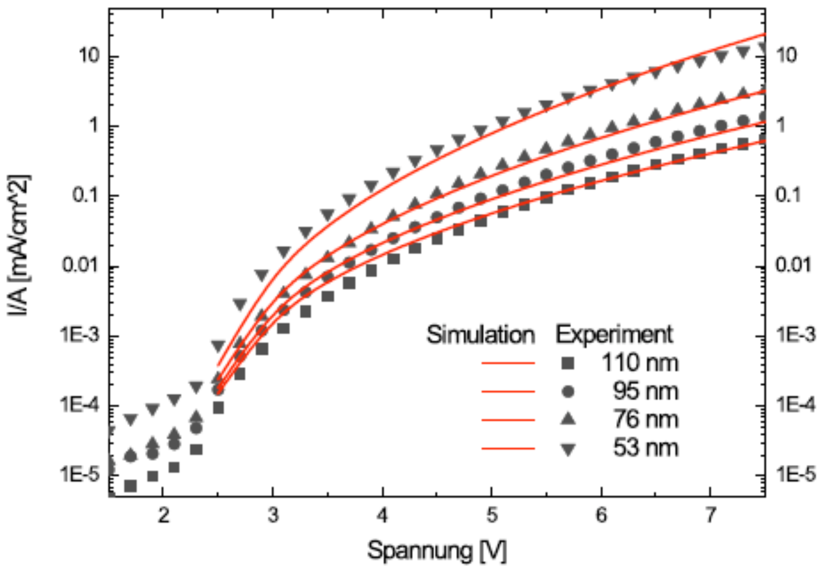
Fundamental Processes:

1. Charge Injection
2. Charge Carrier Transport
3. Exciton Formation
4. Radiative Decay
5. Light Extraction

Real stack consists of up to 12 layers!

Simulation of Organic LEDs





- Novel physical models require better numerical methods
- Transient simulations and IV curves need multiple simulations

➡ Efficient simulations are crucial

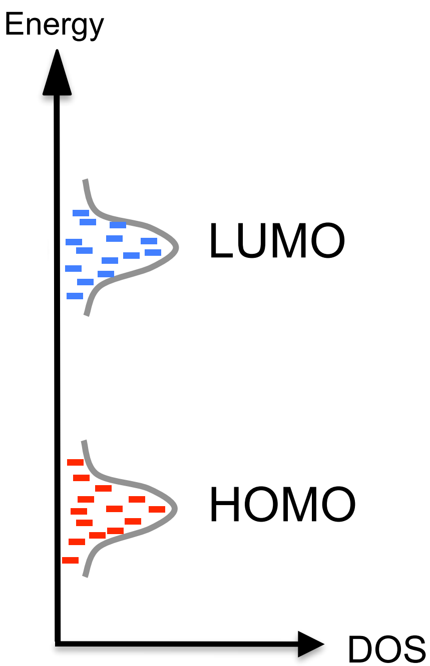
Experimental data from CSEM, simulation by ICP

Overview-Task list

- ✓ Modeling of charge carrier transport
  - › Gummel solver
  - › Newton solver
- ✓ Bipolar
- ✓ Injection
- ✓ Organic material properties
  - › Disorder (Gaussian DOS)
  - › Mobility
  - › Generalized Einstein relation
- ✓ Traps (Exponential DOS)
- ✓ Multilayer OLEDs
- Exciton dynamics
- Parameter extraction
- Coupling to optical model
- Impedance simulations

Organic Materials

Gaussian Disorder



- Small molecules and polymer LEDs/solar cells
- Charge transport by hopping between uncorrelated sites
- Width of DOS-disorder parameter σ (50-150 meV)

$$DOS(\epsilon) = \frac{N_t}{\sqrt{2\pi}\sigma} \exp\left[-\left(\frac{\epsilon - \epsilon_0}{\sqrt{2}\sigma}\right)^2\right]$$

Governing Equations in OLEDs

Poisson equation: $\epsilon \Delta \psi = q(n-p)$

Continuity equation: $\nabla \cdot J_p + q \frac{\partial p}{\partial t} = -qR(p,n)$

Drift-Diffusion: $J_p = -q\mu_p p \nabla \psi - qD_p \nabla p$

similar for electrons

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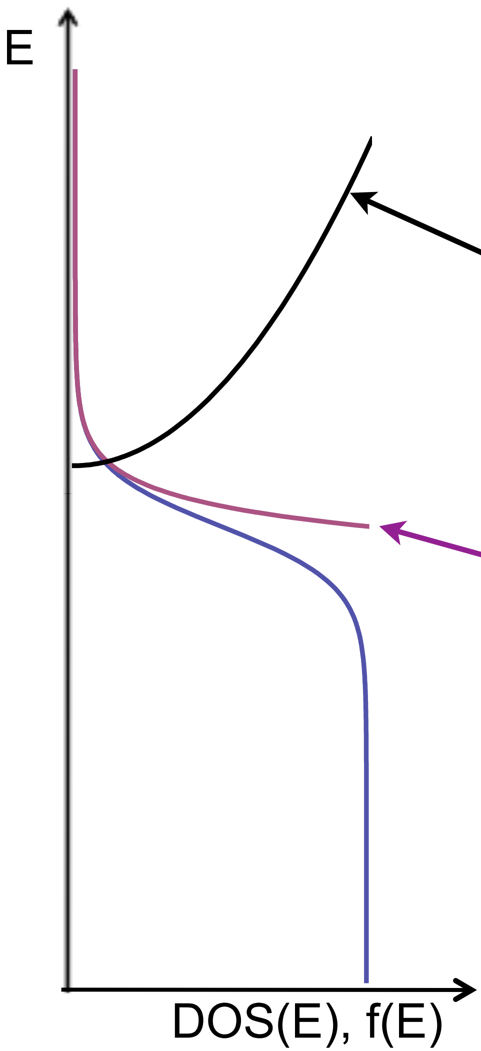
similar for electrons

mobility & diffusion coefficient

are affected by the Gaussian DOS!

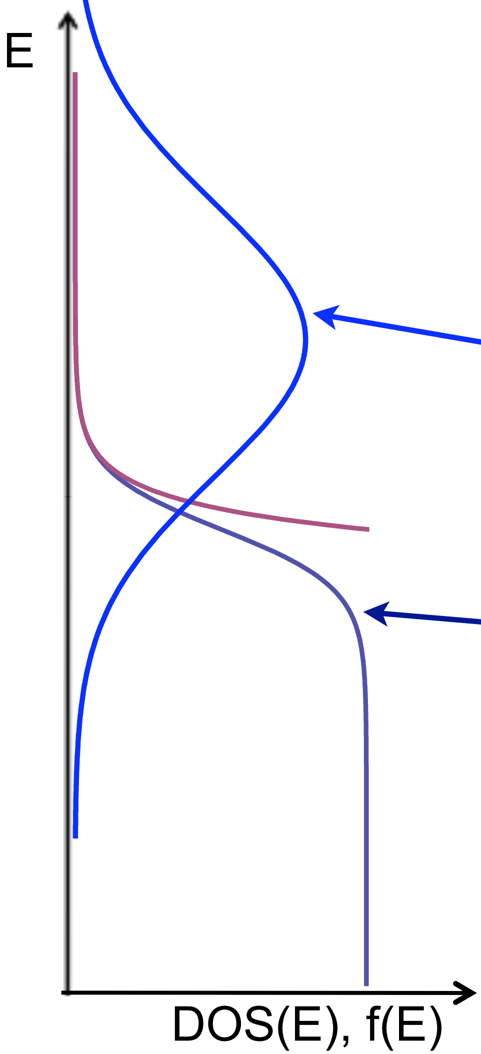
Generalized Einstein Relation

$$p = \int_{-\infty}^{\infty} DOS(E) f(E) dE$$



	ordered material
DOS	$\propto \sqrt{E}$
Statistics	Boltzmann
Einstein relation	$\frac{D}{\mu} = \frac{kT}{q}$

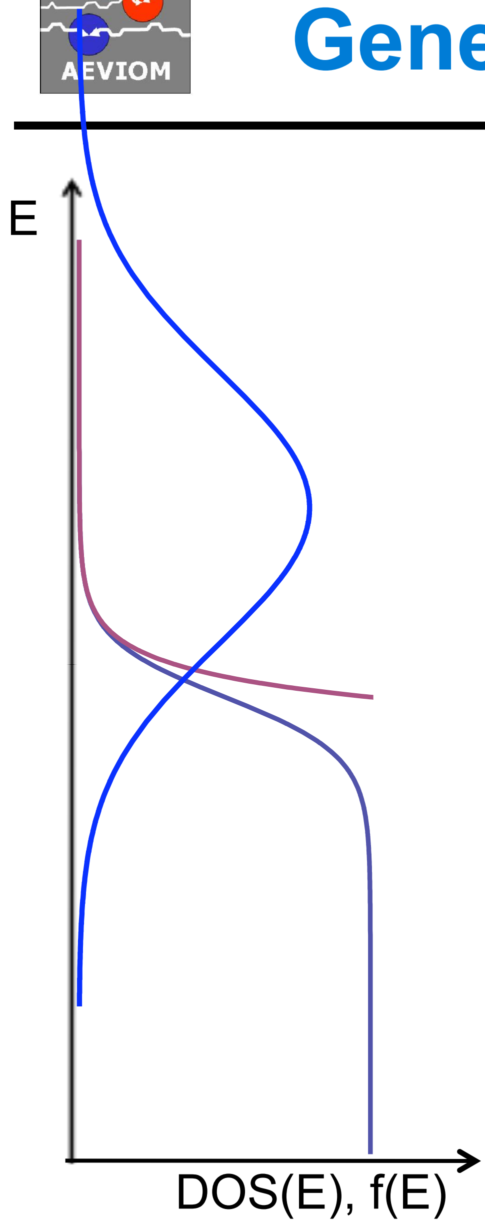
Generalized Einstein Relation



$$p = \int_{-\infty}^{\infty} DOS(E) f(E) dE$$

	ordered material	disordered material
DOS	$\propto \sqrt{E}$	Gaussian
Statistics	Boltzmann	Fermi-Dirac
Einstein relation	$\frac{D}{\mu} = \frac{kT}{q}$	$\frac{D}{\mu} = \frac{p}{q \frac{\partial p}{\partial E_F}}$

Generalized Einstein Relation



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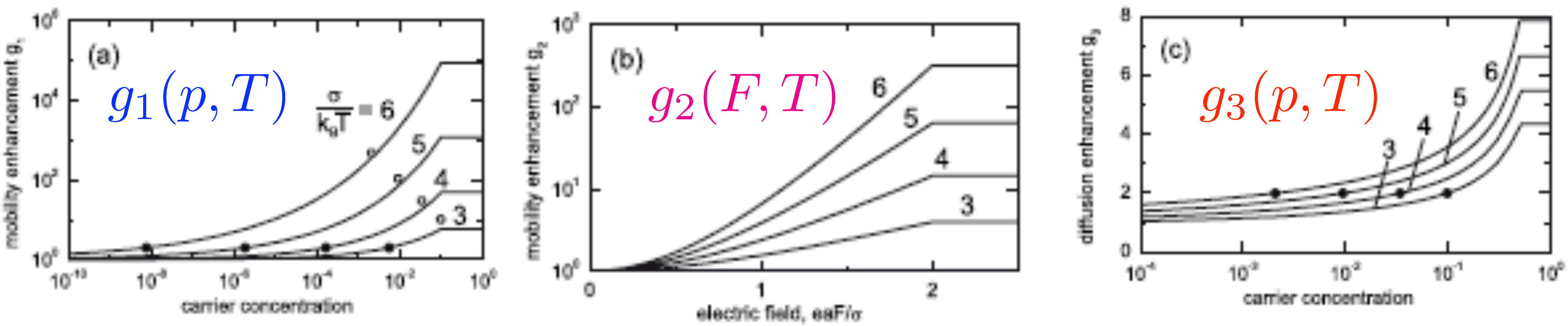
Extended Gaussian Disorder Model (EGDM)

$$D_p = \frac{k_B T}{q}\mu_0(T,p,F)g_3(p,T)$$

$$\mu_p(T,p,F) = \mu_0(T)g_1(p,T)g_2(F,T)$$

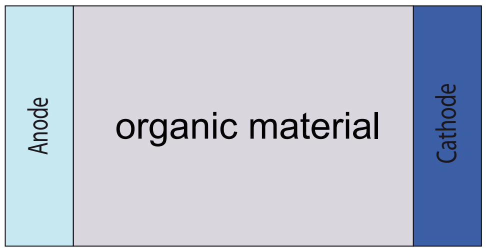


Nonlinear equations for mobility and diffusion coefficient

Mobility depends on temperature, field and density

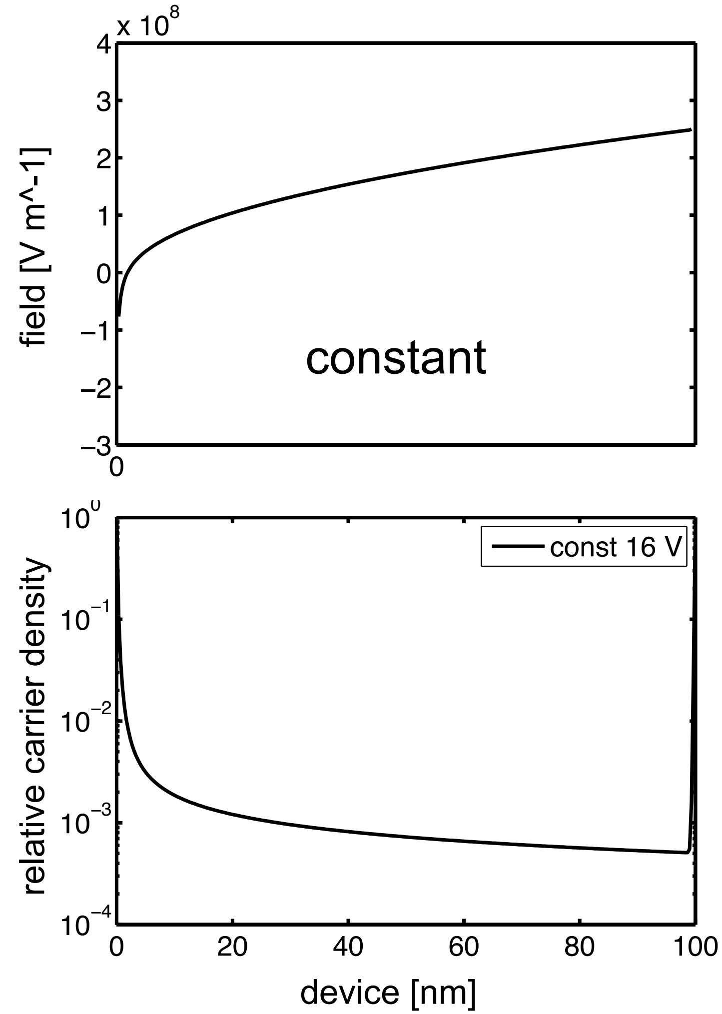
S. L. M. van Mensfoort, R. Coehoorn, Phys. Rev. B 78, 085207 (2008)

Effects of EGDM Transport



Assumption of ohmic contact:
Dirichlet boundary conditions

$$n_1 = 0.5N_t$$
$$n_2 = 0.5N_t$$
$$16V$$



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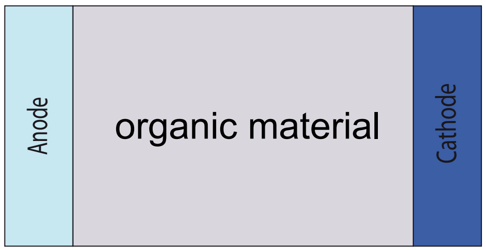

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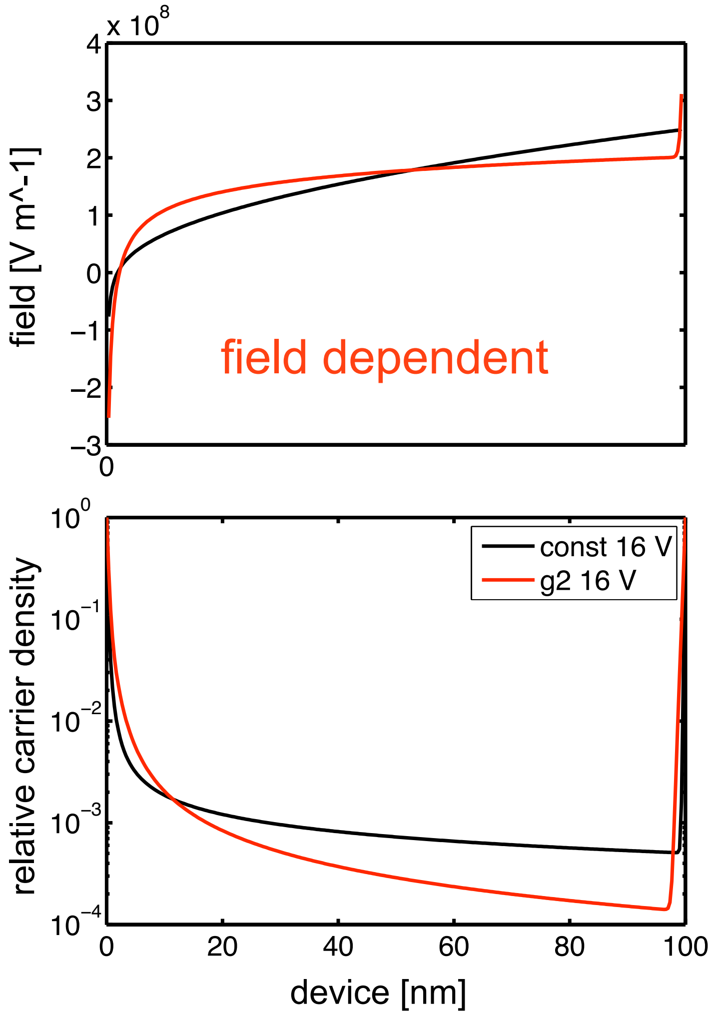

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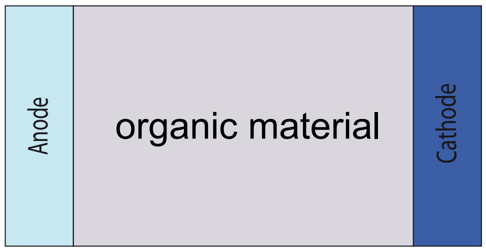

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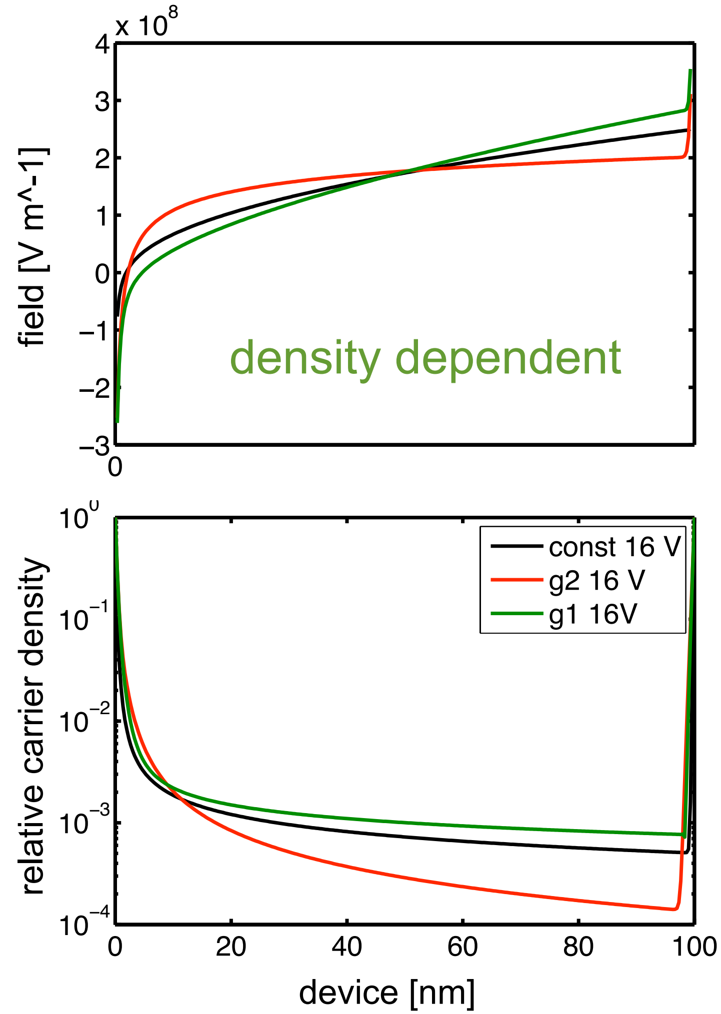

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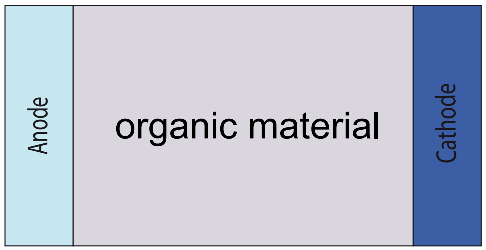

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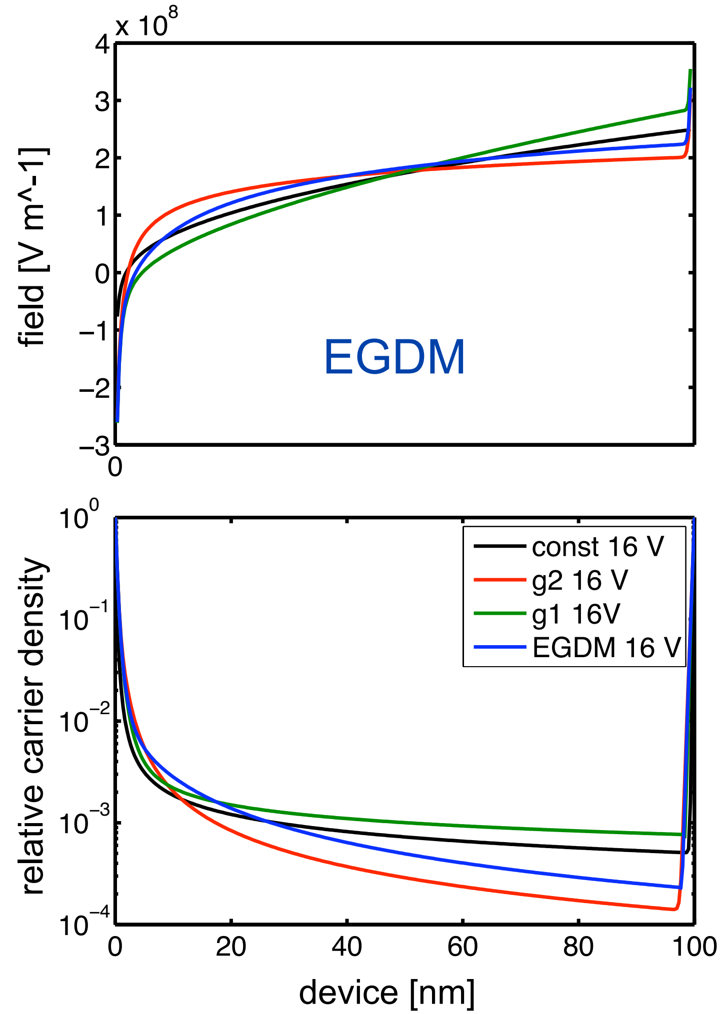
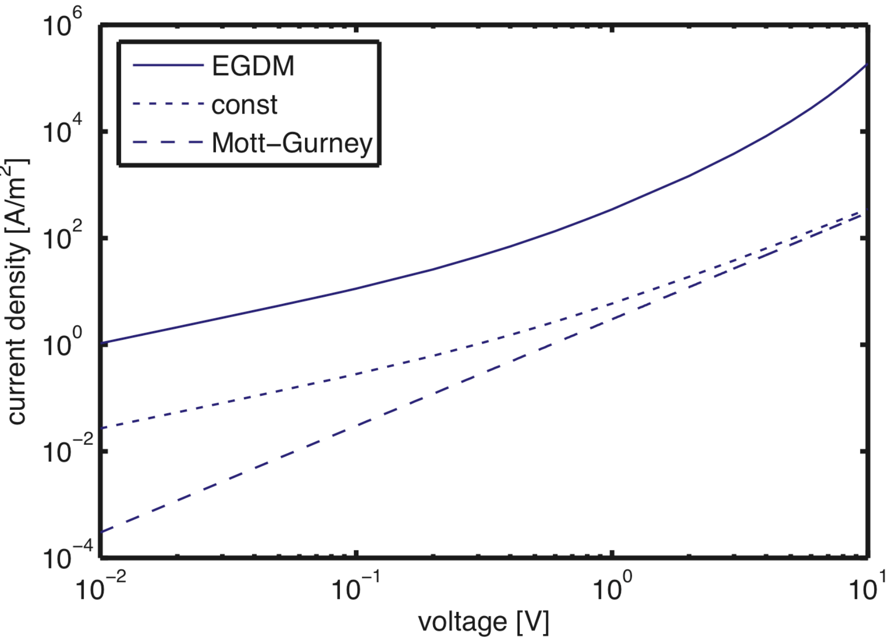


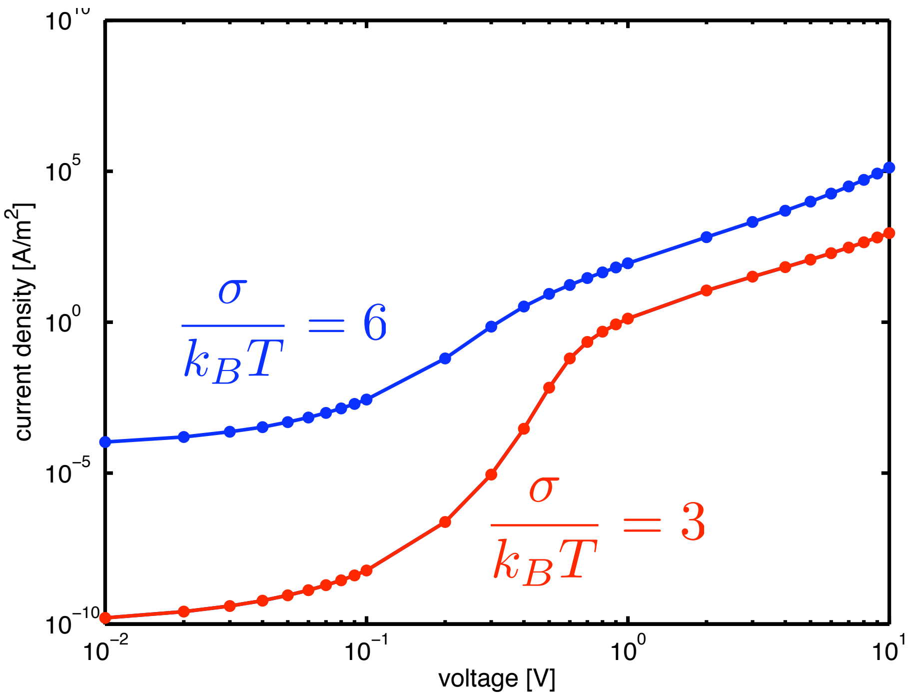
EGDM on single layer OLED

IV Curve
(hole-only device)



- Diffusion effects
- Field- and density-dependent

IV Curve
(hole-only device with 1eV built-in potential)

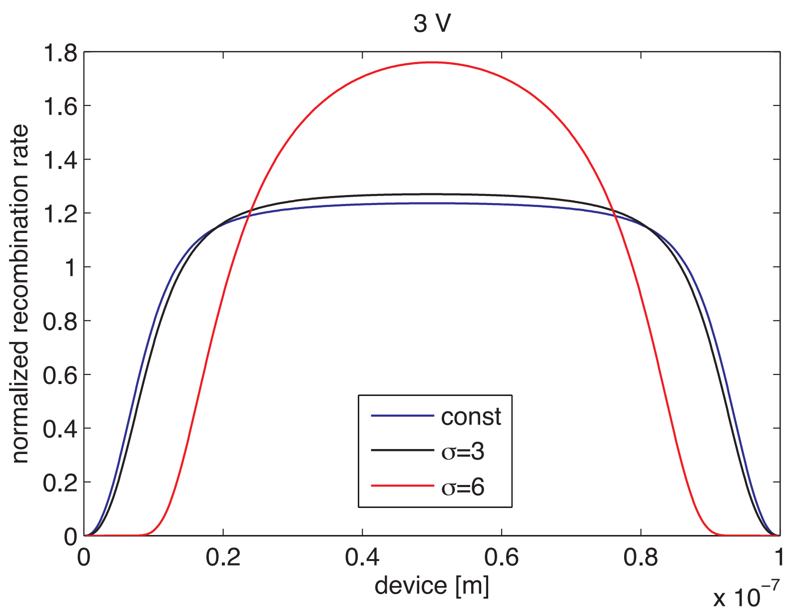


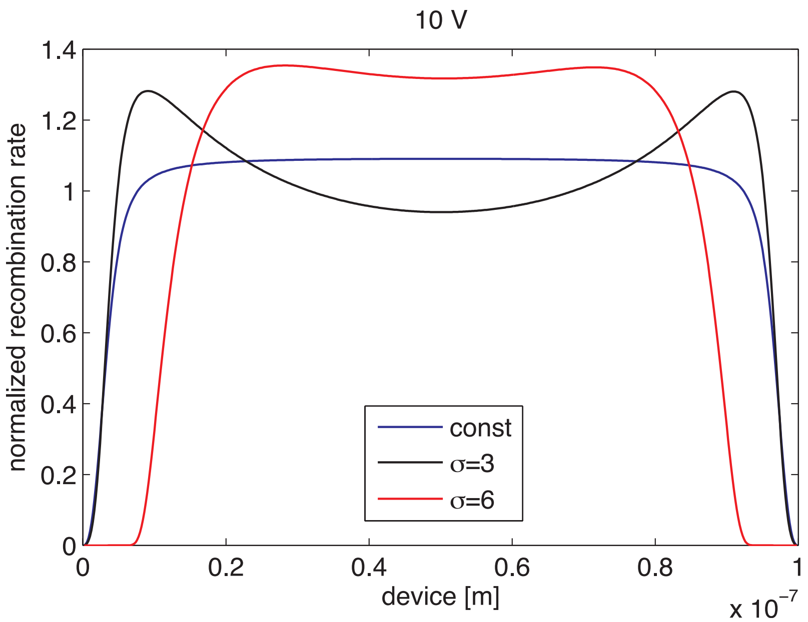
- Effects of different disorder parameters

In good agreement with:
S. L. M. van Mensfoort, R. Coehoorn, Phys. Rev. B 78, 085207 (2008, Fig 9)

Institute of Computational Physics

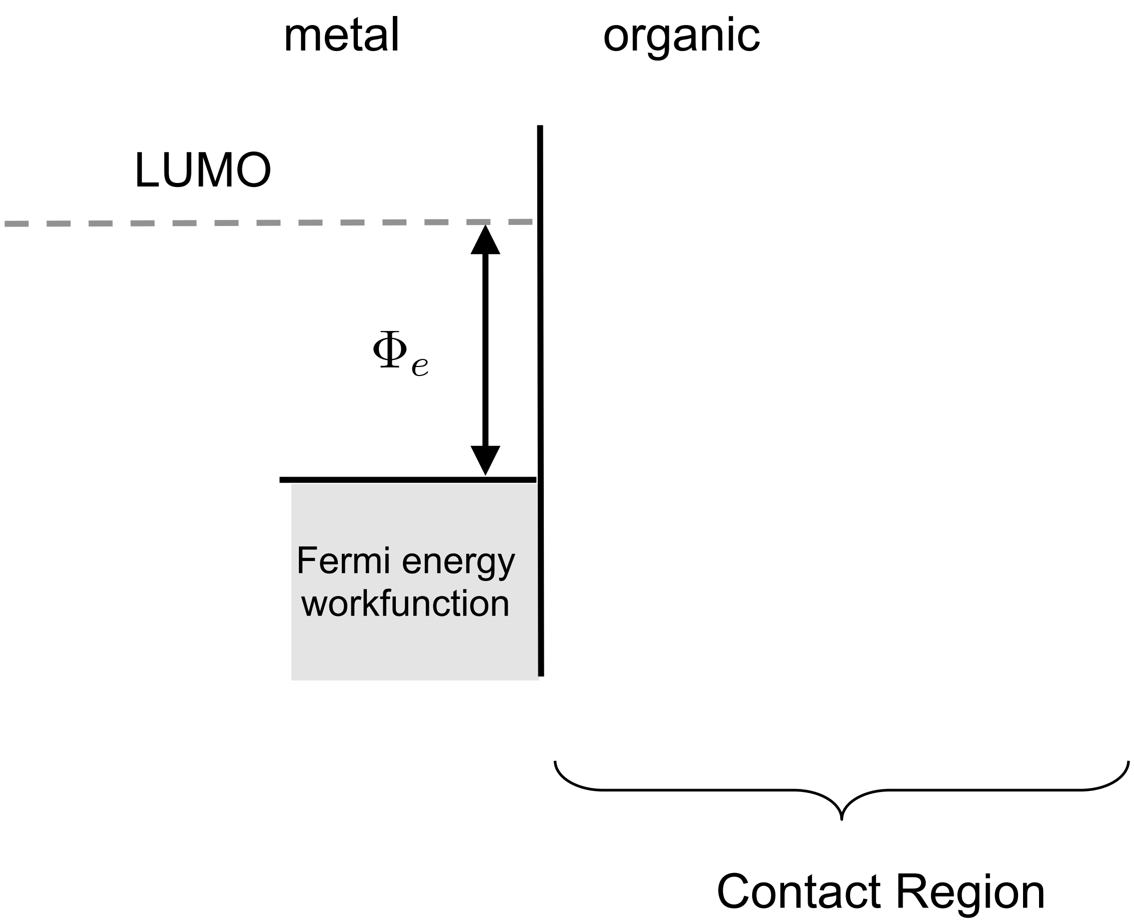
Recombination Profiles



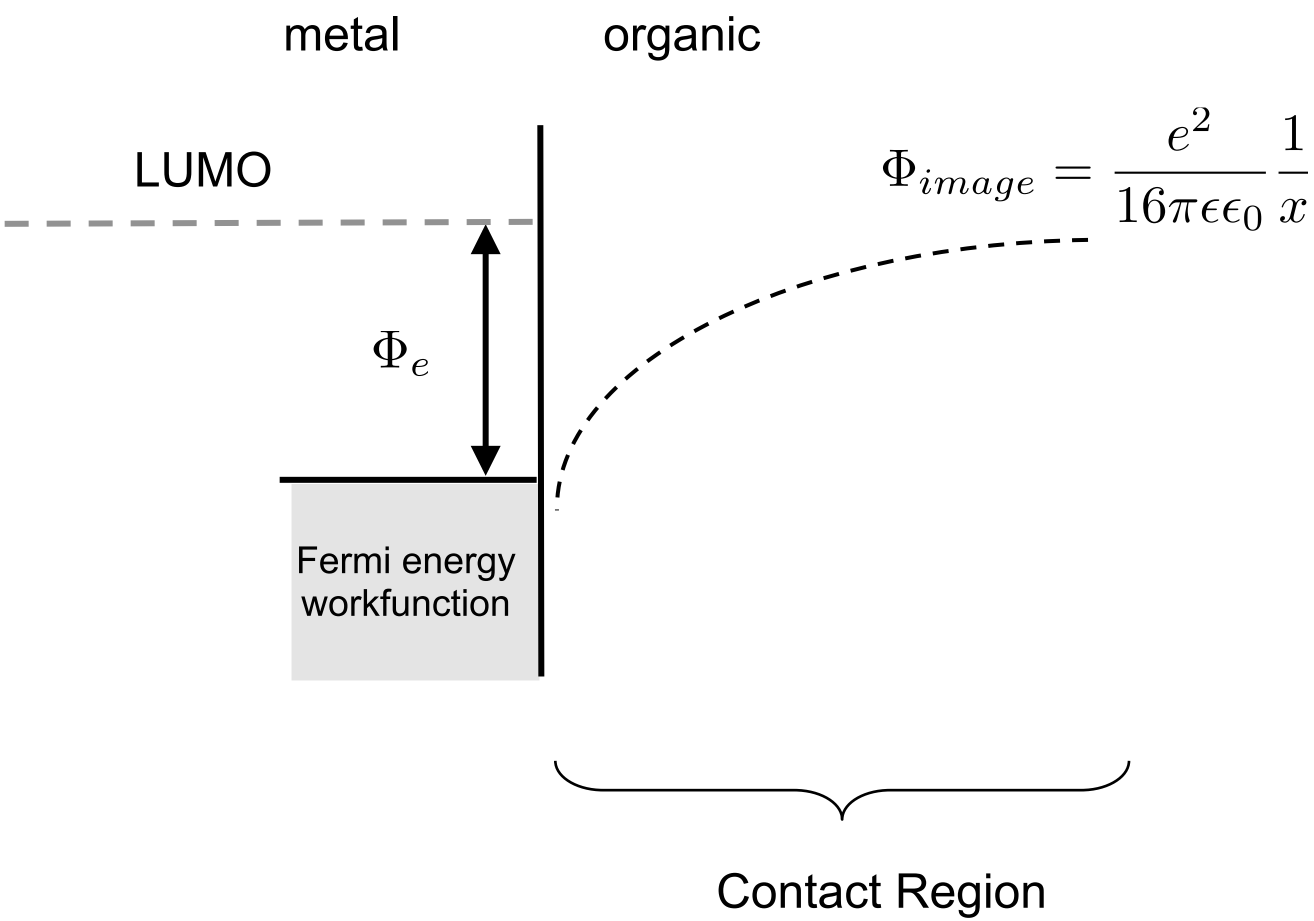


- Bipolar simulation with constant mobility and EGDM for $\hat{\sigma} = 3$ and $\hat{\sigma} = 6$
- Effects of disorder clearly visible

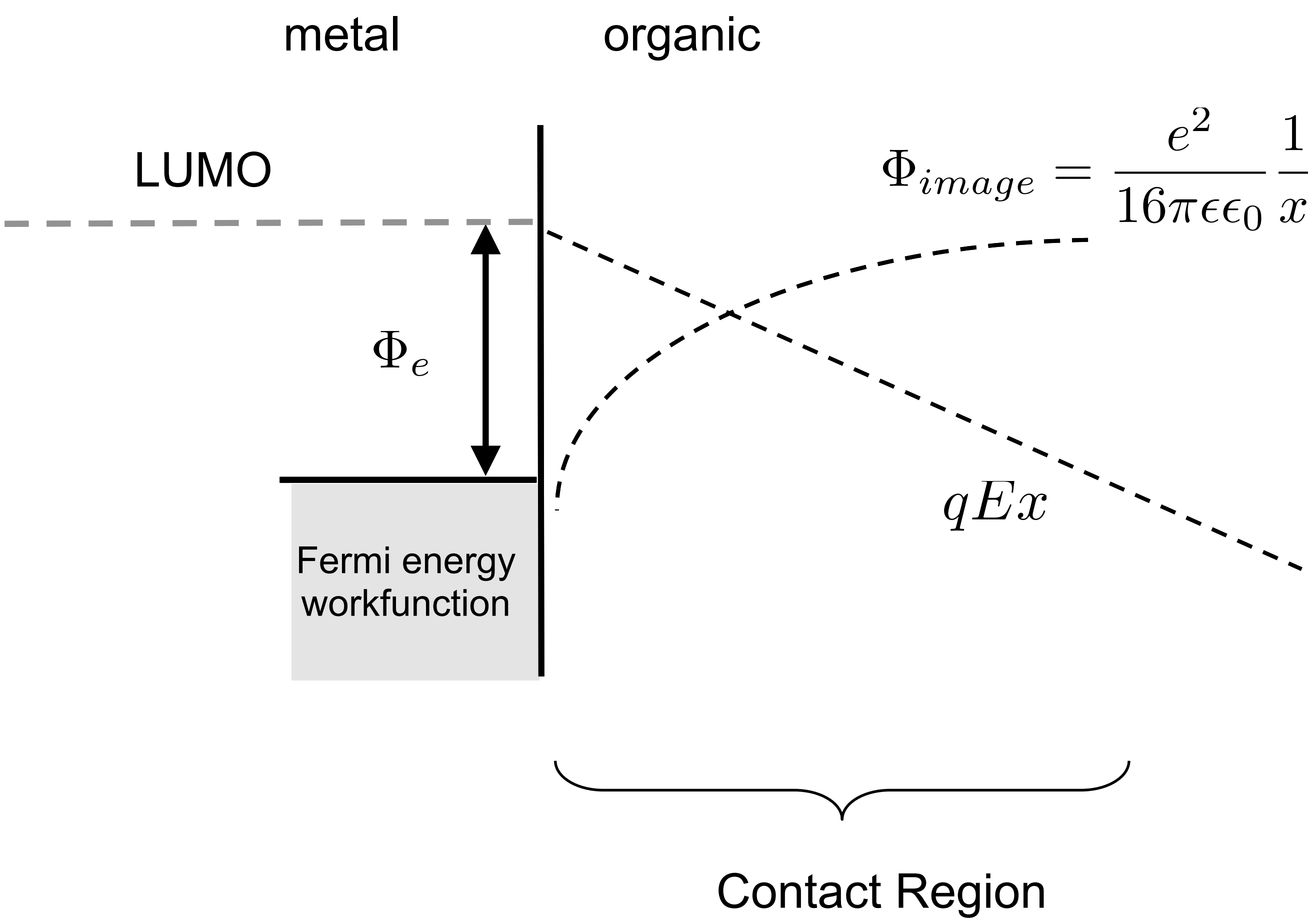
Thermionic Injection



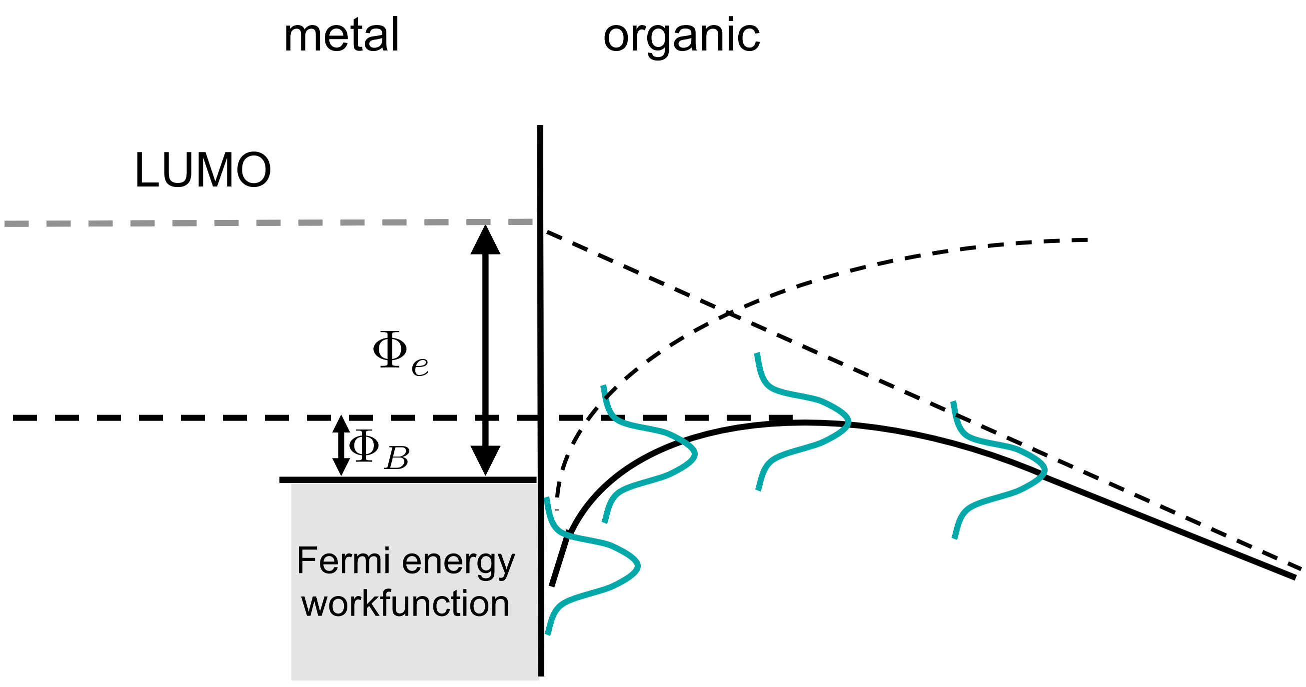
Thermionic Injection



Thermionic Injection

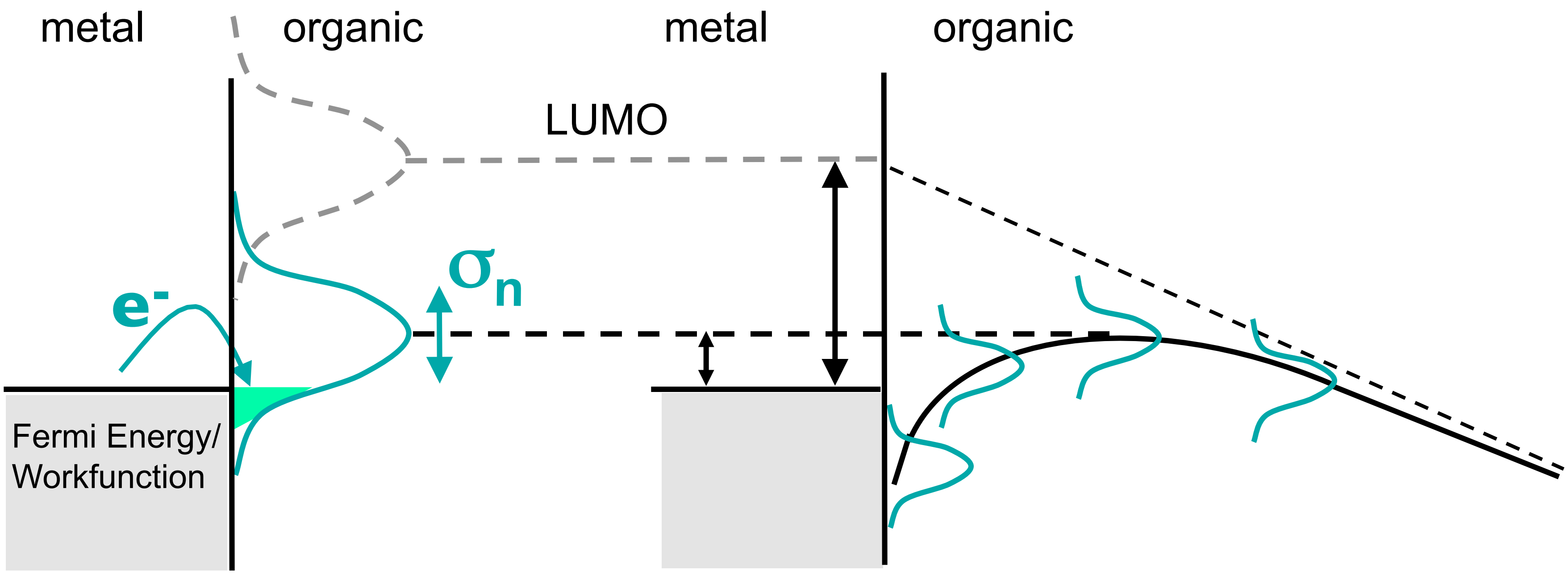


Thermionic Injection



$$\Phi_e - eEx - \frac{e^2}{16\pi\epsilon\epsilon_0 x}$$

Thermionic Injection

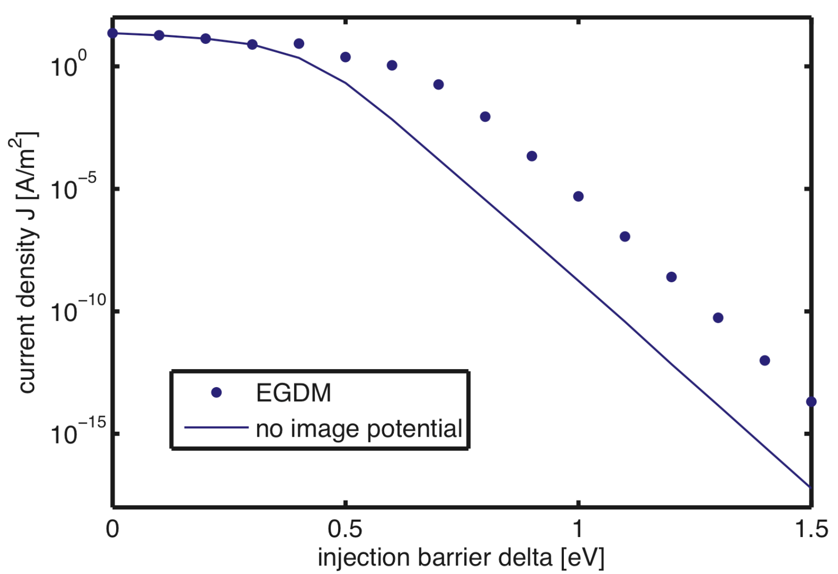


Density at contact depends on position of Gaussian DOS

Dependent boundary conditions

Effects of Injection

Dependence of the current density on the injection barrier at 2V

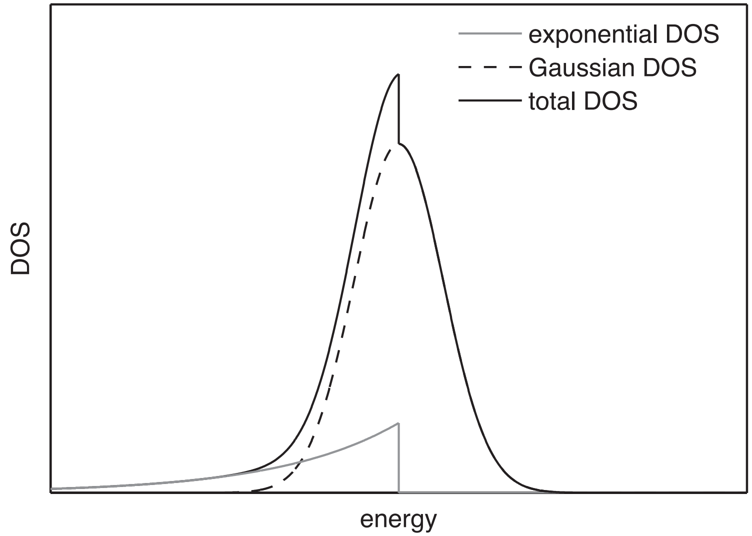


- No effect if injection barrier < 0.5 eV
- Higher currents with image potential
- Agrees with Monte Carlo results

In good agreement with:
J.J.M. van der Holst, M.A. Uijttewaal, R. Balasubramanian, R. Coehoorn, P.A. Bobbert, G.A. de Wijs and R.A. de Groot (EUT, PRE), Phys. Rev. B (2009).

Trap Effects in OLEDs

- localized sites with higher electron affinity
  - impurities, chemical defects
- Model
  - trap distribution: Expontential, Gaussian
  - discrete levels: shallow, deep

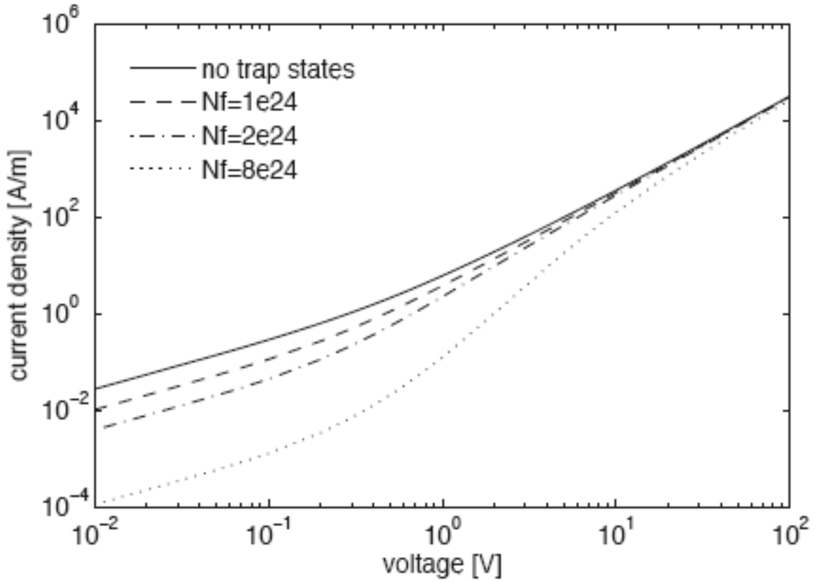


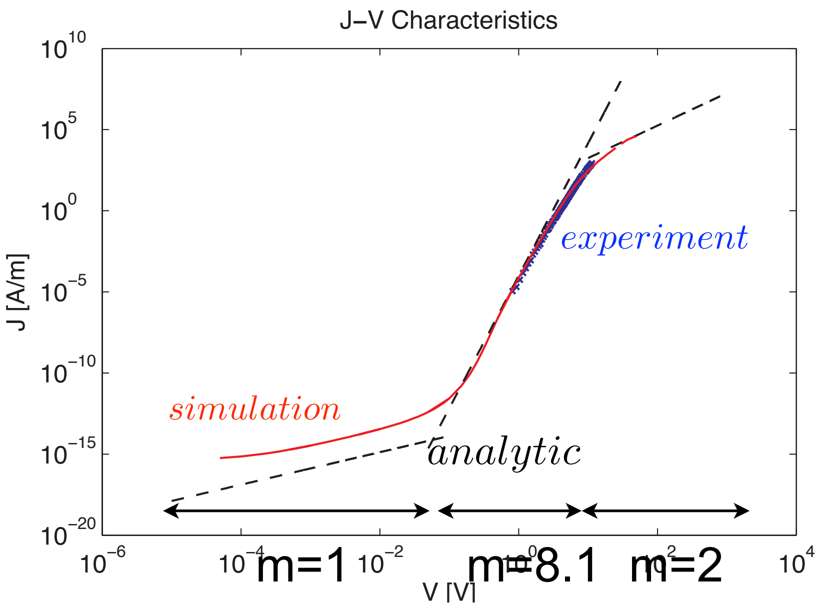
$$\epsilon \Delta \psi = q(n - p + n_t - p_t)$$

$$\nabla \cdot J_p + q\frac{\partial p}{\partial t} = -qR(p,n)$$

$$J_p = -q\mu_p p \nabla \psi - qD_p \nabla p$$

Trap IV Curves



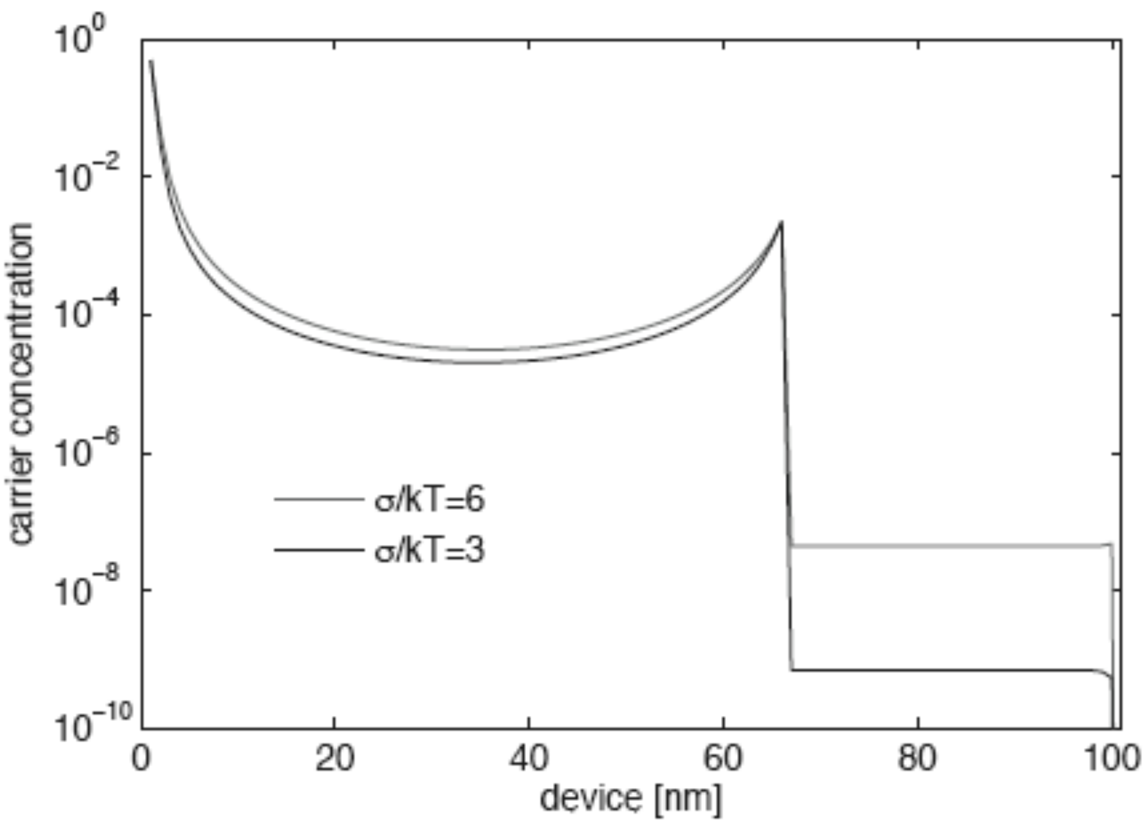


trap density influences current density

Analytical solution for Gaussian DOS:
M. M. Mandoc, B. de Boer, G. Paasch, P. W. M. Blom, Phys. Rev. B (2007).

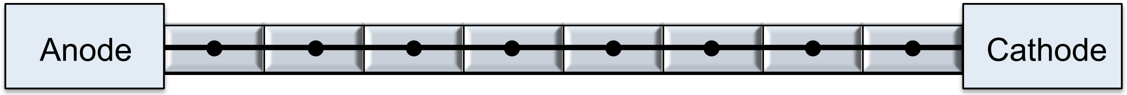
Multi-layer Devices

- Stack of organic material to optimize recombination profiles and light emission



Spatial Discretization

- 1-dimensional finite volume method
  - Domain divided into n grid points



- Reformulation of problem

$$
\begin{aligned}
F_1(\psi, p, n) &= \epsilon \Delta \psi - q(n-p) \stackrel{!}{=} 0 \\
F_2(\psi, p, n) &= \nabla \cdot (-q\mu_p p \nabla \psi - q D_p \nabla p) + q\frac{\partial p}{\partial t} + qR \stackrel{!}{=} 0 \\
F_3(\psi, p, n) &= \nabla \cdot (-q\mu_n n \nabla \psi + q D_n \nabla n) - q\frac{\partial n}{\partial t} - qR \stackrel{!}{=} 0
\end{aligned}
$$

- Integration over each box

Scharfetter-Gummel Discretization

- Neglecting recombination and assuming a constant current density through the device

$$q\mu_n \left(U_t \frac{\partial n}{\partial x} - n \frac{\partial \psi}{\partial x}\right) = c$$

- Boundary values $n(x_{i-1}) = n_{i-1}$ and $n(x_i) = n_i$
- Analytic solution

$$n(x) = n_{i-1}(1 - g(x)) + n_i g(x)$$

with

$$g(x) = \frac{1 - \exp\left(\frac{(\psi_i - \psi_{i-1})}{U_t} \frac{x - x_{i-1}}{x_i - x_{i-1}}\right)}{1 - \exp\left(\frac{\psi_i - \psi_{i-1}}{U_t}\right)}$$

- Analytic solution serves as Ansatz function
  - › Scharfetter-Gummel discretization

Spatial Discretization

- Exponential fitting for drift-diffusion (F2 and F3)
  - Scharfetter-Gummel discretization with generalized Einstein relation and density- and fielddependent mobility

- System of (3 x n) strongly coupled equations

$$\vec{F}(\vec{x}) = \begin{pmatrix} \vec{F}_1(\vec{x}) \\ \vec{F}_2(\vec{x}) \\ \vec{F}_3(\vec{x}) \end{pmatrix} \qquad \vec{x} = \begin{pmatrix} \psi_1 \\ \vdots \\ \psi_n \\ n_1 \\ \vdots \\ n_n \\ p_1 \\ \vdots \\ p_n \end{pmatrix}$$

- Dirichlet boundary conditions:
  - Values for potential and carriers given at electrodes

Problem Formulation

- Variables sets
  - › carrier concentrations (ψ, p, n)
  - › quasi-Fermi level (ψ, ϕ_p, ϕ_n)
    - Assumption: Boltzmann statistics

$$p = n_{int,eff} \exp\left(\frac{q(\phi_p - \psi)}{kT}\right)$$

$$n = n_{int,eff} \exp\left(\frac{q(\psi - \phi_n)}{kT}\right)$$

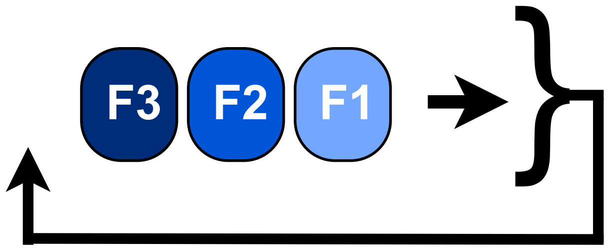
  - › Slotboom (ψ, Φ_p, Φ_n)

$$\Phi_p = \exp\left(\frac{q\phi_p}{kT}\right) \qquad p = p_i \Phi_p \exp\left(\frac{-q\psi}{kT}\right)$$

$$\Phi_n = \exp\left(\frac{-q\phi_n}{kT}\right) \qquad n = n_i \Phi_n \exp\left(\frac{q\psi}{kT}\right)$$

Discretized Equations

- De-coupled solving
  - Gummel algorithm



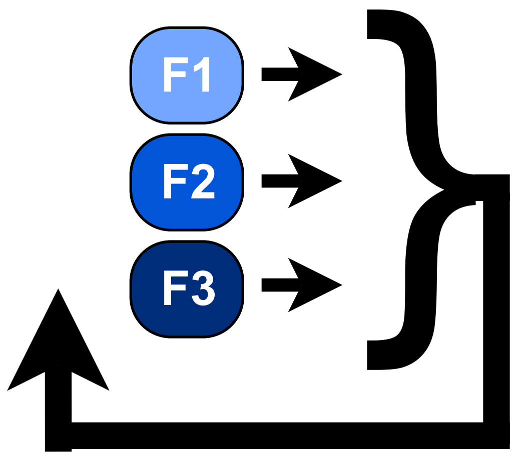
- Coupled solving
  - Newton algorithm

Find x* so that F(x*)=0.

$$F(x) = F(x^*) + J(x^*)(x - x^*) \qquad \textit{Taylor Series}$$

$$J(x) = \begin{bmatrix} \frac{\partial F_1(x)}{\partial x_1} & \cdots & \frac{\partial F_1(x)}{\partial x_N} \\ \vdots & \ddots & \vdots \\ \frac{\partial F_N(x)}{\partial x_1} & \cdots & \frac{\partial F_N(x)}{\partial x_N} \end{bmatrix} \qquad \textit{Jacobian Matrix}$$

$$\Rightarrow x^{k+1} = x^k - J(x^k)^{-1} F(x^k) \qquad \textit{Iteration function}$$

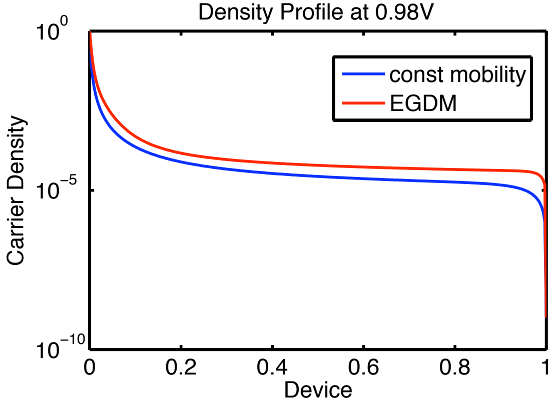


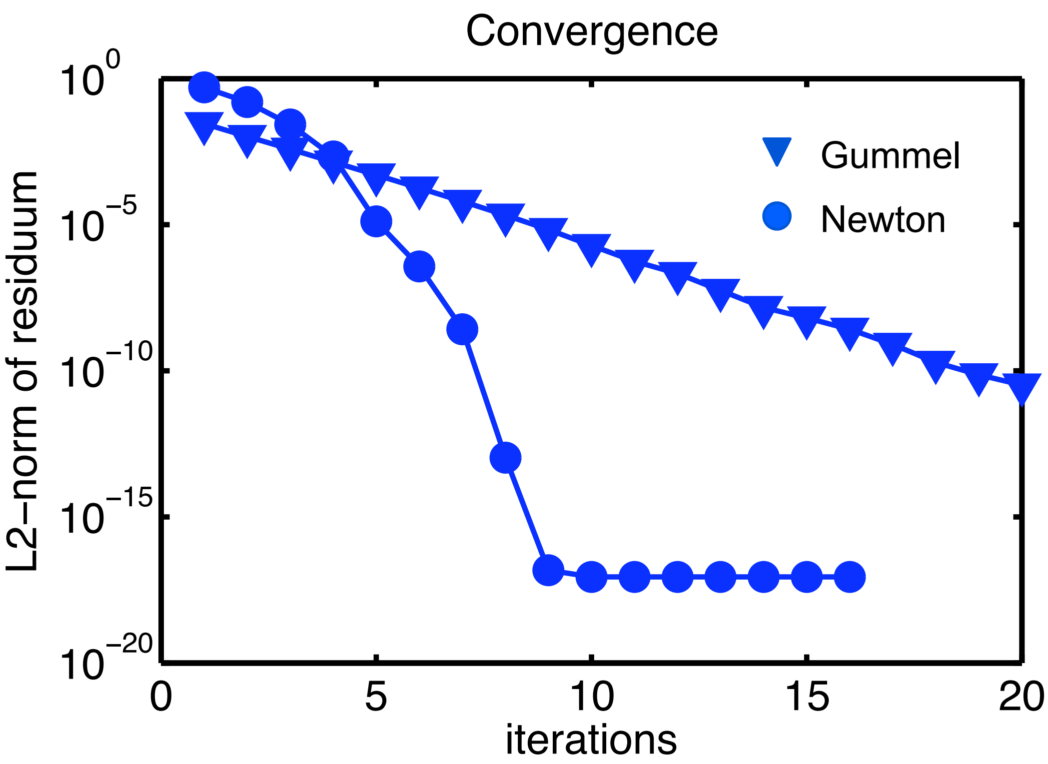
Algorithms

- Gummel
  - steady-state
  - transient
- Newton
  - steady-state
  - transient
- Initial guess
  - no bias applied, Boltzmann approximation
- Gummel steady-state
  - Damping
- Newton
  - Damping
  - Homotopy

Convergence - Steady State

L2-Norm: $|F| = \sqrt{\sum_{k=1}^{n} |F_k|^2}$





Convergence - Steady State

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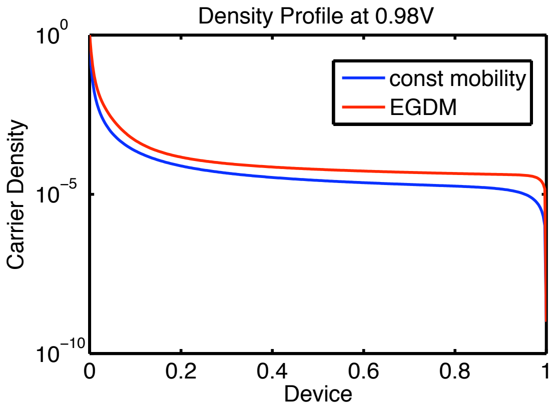
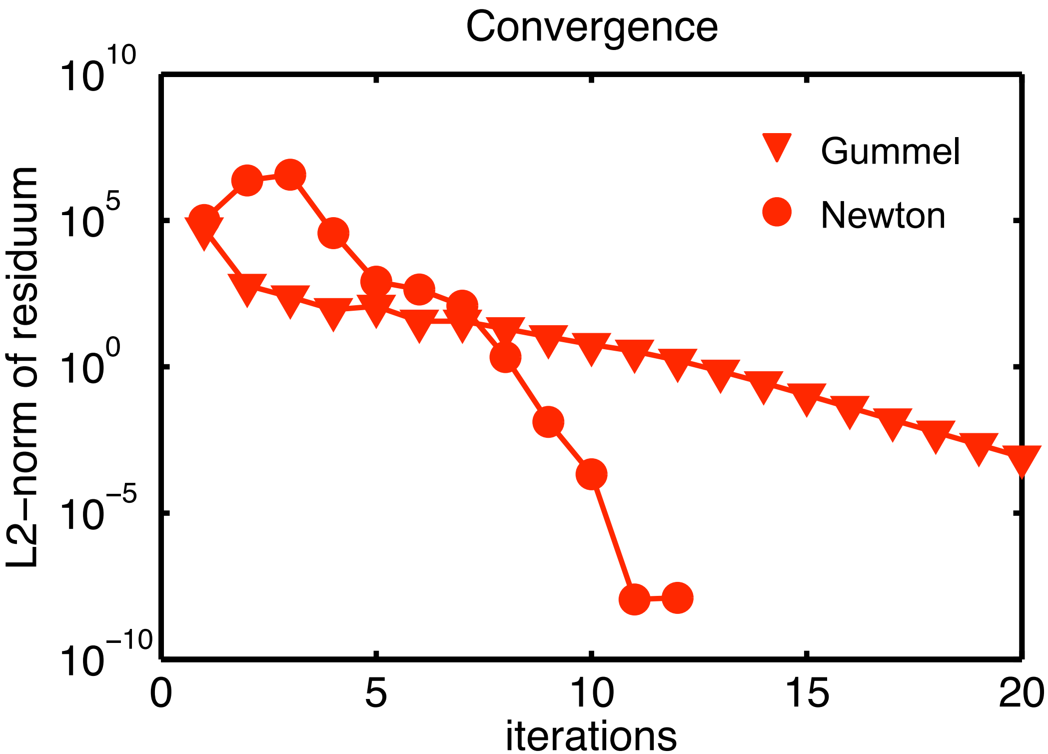
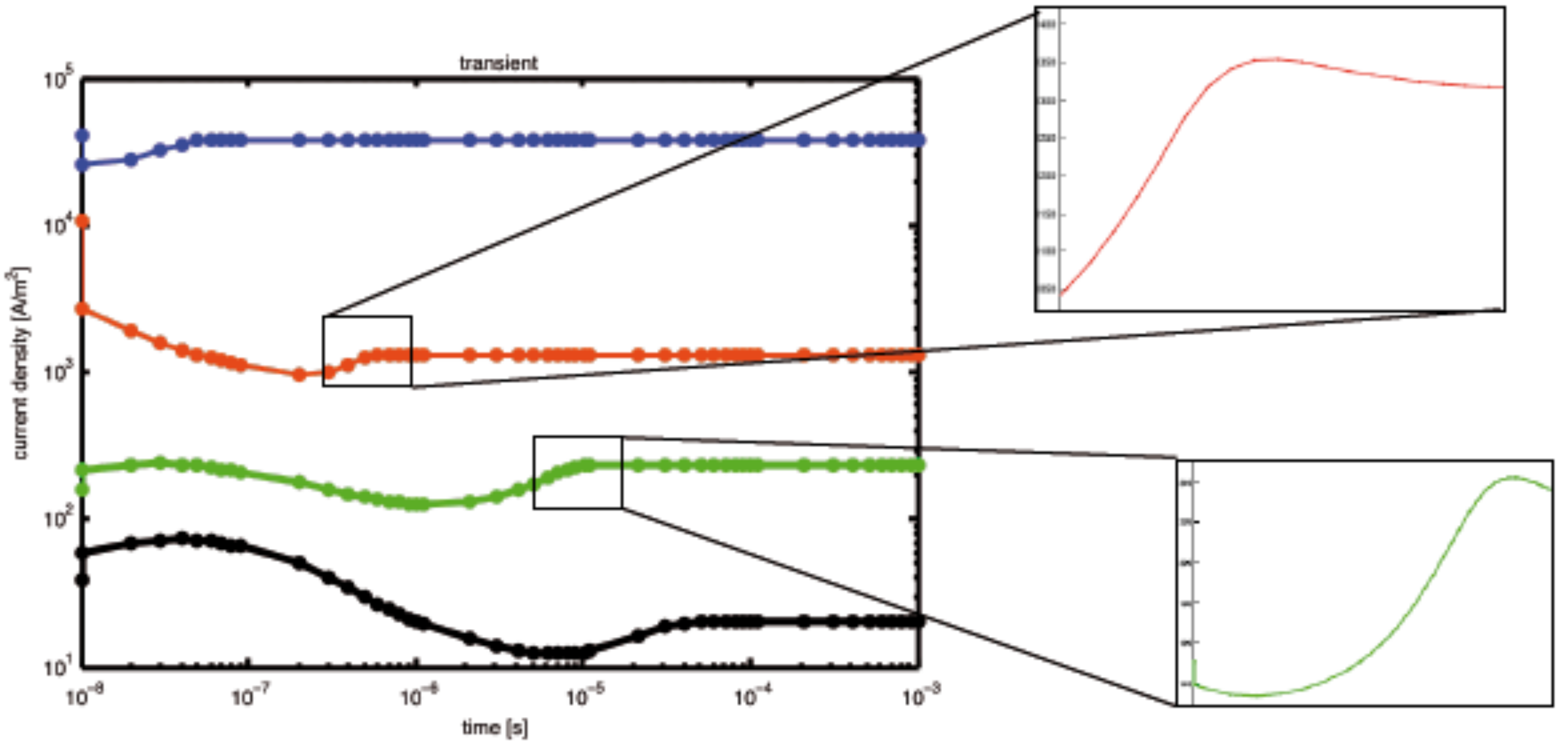




- Convergence for Gummel and Newton algorithm
- Fewer iterations needed for Newton algorithm

Transient Simulations

- Implicit Euler time step



Outlook

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  - › Newton solver
- ✓ Bipolar
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- ✓ Multilayer OLEDs
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- Impedance simulations

Exciton Dynamics

- Poisson Equation

$$\frac{\partial E(x)}{\partial x} = \frac{e}{\varepsilon\varepsilon_0}\left(p(x) - n(x)\right)$$

- Charge Current

$$J_n(x) = e\mu_n(x,E)\cdot n(x)\cdot E(x) + D(\mu)\cdot\frac{\partial n(x)}{\partial x}$$

- Charge Continuity

$$\frac{\partial n(x)}{\partial t} = \frac{1}{e}\frac{\partial J_n(x)}{\partial x} - r(x)\cdot p(x)\cdot n(x) + G_{opt_{np}}\, g_n(x)$$

- Exciton Current
- Exciton Continuity

$$J_S(x) = D_S\cdot\frac{\partial S(x)}{\partial x}$$

$$\frac{dS_i}{dt} = G_i R + \vec{\nabla}\vec{J}_{s_i} - \left(k_{rad_i} + k_{nonrad_i}\right)\cdot S_i - k_{annihilation_i}\cdot S_i^2 + \sum_{j=1}^{n_{exc}}\left(k_{ji}\cdot S_j - k_{ij}\cdot S_i\right) + G_{opt_{exc}}\, g_s$$

- Light-emission (from dipoles) & Light-incoupling

Electro-optical Coupling Terms

Opto-electronic Coupling Terms

Extended version of the models published by Ruhstaller et al., J. Appl. Phys. **89**, 4575, (2001) and Ruhstaller et al., IEEE JSTQE **9**, (3) 723, (2003)

Outlook

- ✓ Modeling of charge carrier transport (1st generation)
  - › Gummel
  - › Newton
- ✓ Bipolar (1st generation)
- ✓ Injection (2nd generation)
- ✓ Organic material properties
  - › Disorder (2nd generation)
  - › Mobility (2n generation)
  - › Generalized Einstein relation (2nd generation)
- ✓ Traps (2nd generation)
- ✓ Multilayer OLEDs (1st generation)
- Exciton dynamics (1st generation)
- Parameter extraction
- Optical simulations
- Impedance simulations

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