Advanced Simulation Methods for Charge Transport in OLEDs

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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:
Fluxim AG, www.fluxim.com

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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
Advanced Experimentally Validated OLED model

Philips Research Eindhoven
Project Coordinator: Reinder Coehoorn

Eindhoven University of Technology

University of Groningen

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Philips Research Aachen

Zürich University of Applied Sciences
Fluxim

Technical University Dresden

Sim4Tec

University of Cambridge
Principle of OLED Operation

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Real stack consists of up to 12 layers!

Fundamental Processes:

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

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Gaussian Disorder

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

\[
DOS(\epsilon) = \frac{N_t}{\sqrt{2\pi\sigma}} \exp \left[ - \left( \frac{\epsilon - \epsilon_0}{\sqrt{2\sigma}} \right)^2 \right]
\]
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: \[ J_p = -q \mu_p p \nabla \psi - q D_p \nabla p \]

similar for electrons
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

mobility & diffusion coefficient are affected by the Gaussian DOS!
Generalized Einstein Relation

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

\[ \frac{D}{\mu} = \frac{kT}{q} \]

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Generalized Einstein Relation

\[ p = \int_{-\infty}^{\infty} \text{DOS}(E) f(E) dE \]

- **Einstein relation**
  \[ \frac{D}{\mu} = \frac{kT}{q} \]
  \[ \frac{D}{\mu} = \frac{p}{q \frac{\partial p}{\partial E_F}} \]

- **DOS**
  \[ \propto \sqrt{E} \]

- **Statistics**
  - Boltzmann
  - Fermi-Dirac

- **Ordered material**
- **Disordered material**

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## Generalized Einstein Relation

The Generalized Einstein Relation is given by:

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

### DOS and Statistics

<table>
<thead>
<tr>
<th>ordered material</th>
<th>disordered material</th>
</tr>
</thead>
<tbody>
<tr>
<td>DOS</td>
<td>$\propto \sqrt{E}$</td>
</tr>
<tr>
<td>Statistics</td>
<td>Boltzmann</td>
</tr>
<tr>
<td>Einstein relation</td>
<td>Fermi-Dirac</td>
</tr>
</tbody>
</table>

### Einstein Relation

For ordered material:

$$ \frac{D}{\mu} = \frac{kT}{q} $$

For disordered material:

$$ \frac{D}{\mu} = \frac{p}{q} \frac{\partial p}{\partial E_F} $$
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

Assumption of ohmic contact: Dirichlet boundary conditions

\[ n_1 = 0.5N_t \]
\[ n_2 = 0.5N_t \]
\[ 16V \]
Assumption of ohmic contact: Dirichlet boundary conditions

\[ n_1 = 0.5N_t \]
\[ n_2 = 0.5N_t \]

16V

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Assumption of ohmic contact: Dirichlet boundary conditions

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16V
Assumption of ohmic contact:
Dirichlet boundary conditions

\[ n_1 = 0.5N_t \]
\[ n_2 = 0.5N_t \]

16V
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)

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Recombination Profiles

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

Contact Region

metal

organic

LUMO

$\Phi_e$

Fermi energy
workfunction

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Thermionic Injection

\[ \Phi_{image} = \frac{e^2}{16\pi\varepsilon\varepsilon_0} \frac{1}{x} \]

Contact Region

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Thermionic Injection

$\Phi_{image} = \frac{e^2}{16\pi \varepsilon \varepsilon_0} \frac{1}{x}$

Contact Region

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Thermionic Injection

\[ \Phi_e - eE x - \frac{e^2}{16\pi\varepsilon\varepsilon_0 x} \]

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Density at contact depends on position of Gaussian DOS

Dependent boundary conditions

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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:
Trap Effects in OLEDs

- localized sites with higher electron affinity
  - impurities, chemical defects
- Model
  - trap distribution: Exponential, 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:
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

![Diagram](Anode to Cathode)

- Reformulation of problem

\[
F_1(\psi, p, n) = \epsilon \Delta \psi - q(n - p) \stackrel{!}{=} 0
\]

\[
F_2(\psi, p, n) = \nabla \cdot (-q\mu_p p \nabla \psi - qD_p \nabla p) + q \frac{\partial p}{\partial t} + qR \stackrel{!}{=} 0
\]

\[
F_3(\psi, p, n) = \nabla \cdot (-q\mu_n n \nabla \psi + qD_n \nabla n) - q \frac{\partial n}{\partial t} - qR \stackrel{!}{=} 0
\]

- Integration over each box
Neglecting recombination and assuming a constant current density through the device:

\[ q\mu_n(U_t \frac{\partial n}{\partial x} - n \frac{\partial \psi}{\partial x}) = c \]

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

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

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

\[
\vec{F}(\vec{x}) = \begin{pmatrix}
\vec{F}_1(\vec{x}) \\
\vec{F}_2(\vec{x}) \\
\vec{F}_3(\vec{x})
\end{pmatrix}
\]

\[
\vec{x} = \begin{pmatrix}
\psi_1 \\
\vdots \\
\psi_n \\
n_1 \\
\vdots \\
n_n \\
p_1 \\
\vdots \\
p_n
\end{pmatrix}
\]

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

• Variables sets
  › carrier concentrations \((\psi, p, n)\)
  › quasi-Fermi level \((\psi, \phi_p, \phi_n)\)
    • 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 \((\psi, \Phi_p, \Phi_n)\)

\[
\Phi_p = \exp \left( \frac{q\phi_p}{kT} \right) \\
p = p_i \Phi_p \exp \left( -\frac{q\psi}{kT} \right)
\]
\[
\Phi_n = \exp \left( -\frac{q\phi_n}{kT} \right) \\
n = n_i \Phi_n \exp \left( \frac{q\psi}{kT} \right)
\]
Discretized Equations

- **De-coupled solving**
  - Gummel algorithm

- **Coupled solving**
  - Newton algorithm

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

$$F(x) = F(x^*) + J(x^*)(x - x^*)$$

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

$$\Rightarrow x^{k+1} = x^k - J(x^k)^{-1} F(x^k)$$
Algorithms

- Gummel
  - steady-state
  - transient
- Newton
  - steady-state
  - transient
- Initial guess
  - no bias applied, Boltzmann approximation
- Gummel steady-state
  - Damping
- Newton
  - Damping
  - Homotopy
L2-Norm: \[ |F| = \sqrt{\sum_{k=1}^{n} |F_k|^2} \]
Convergence - Steady State

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

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

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Transient Simulations

- Implicit Euler time step
Outlook

✓ Modeling of charge carrier transport
  › Gummel solver
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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
  \[ \frac{\partial n(x)}{\partial t} = \frac{1}{e} \frac{\partial J_n(x)}{\partial x} - r(x) \cdot p(x) \cdot n(x) + G_{opt} n(x) \]

- Exciton Current
  \[ J_s(x) = D_s \cdot \frac{\partial S(x)}{\partial x} \]

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

**Electro-optical Coupling Terms**

**Opto-electronic Coupling Terms**


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Outlook

✓ Modeling of charge carrier transport (1st generation)
   › Gummel
   › Newton
✓ Bipolar (1st generation)
✓ Injection (2nd generation)
✓ Organic material properties
   › Disorder (2nd generation)
   › Mobility (2nd 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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Acknowledgement

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• Thanks for your attention!