Leibniz Association
Weierstrass Institute for
Applied Analysis and Stochastics

ECMath
Einstein Center
for Mathematics Berlin

On p(x)-Laplace thermistor models describing electrothermal feedback in organic semiconductor devices

Matthias Liero

joint work with

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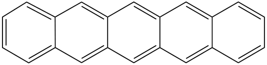
Axel Fischer, Reinhard Scholz (IAPP, TU Dresden)
Miroslav Bulìček (Charles University, Prague)

Organic semiconductors

- Carbon-based materials conducting electrical current
- Used in smartphone and TV displays, photovoltaics, and lighting applications
- OLED: whole area emits light, flexible

C_{60}

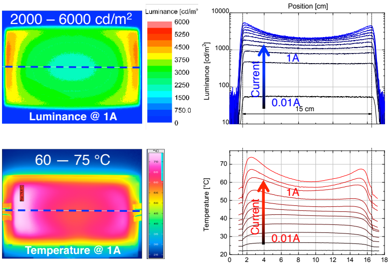


pentacene

Problem

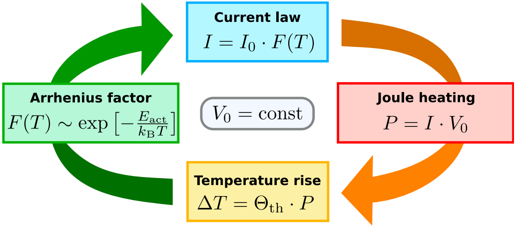
- Luminance inhomogeneities emerge when operating at high currents
- Strong self-heating effects
- Counter-intuitive nonlinear phenomena

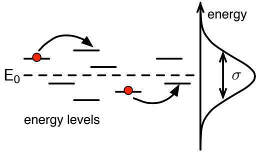
$\Rightarrow$ PDE model needed!



Measurements by A. Fischer

Organic semiconductors: Temperature-activated hopping transport of charge carriers





Organic semiconductors: Temperature-activated hopping transport of charge carriers

Current law
$I = I_0 \cdot F(T)$

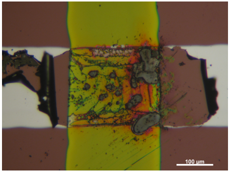
Joule heating
$P = I \cdot V_0$

Temperature rise
$\Delta T = \Theta_{\mathrm{th}} \cdot P$

Arrhenius factor
$F(T) \sim \exp\left[-\frac{E_{\mathrm{act}}}{k_{\mathrm{B}}T}\right]$

$V_0 = \mathrm{const}$

Fischer et al., Org. Elec., 2012



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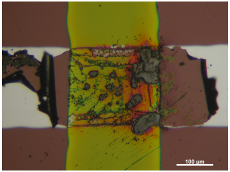
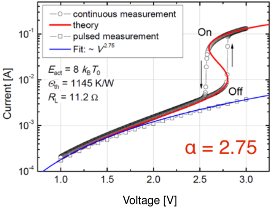


Zero-dimensional model with Arrhenius law, non-Ohmic current-voltage relation, and global heat balance

$$I(V, T) = I_{\text{ref}} F(T) \left[\frac{V}{V_{\text{ref}}}\right]^{\alpha} \qquad (\alpha \geq 1)$$

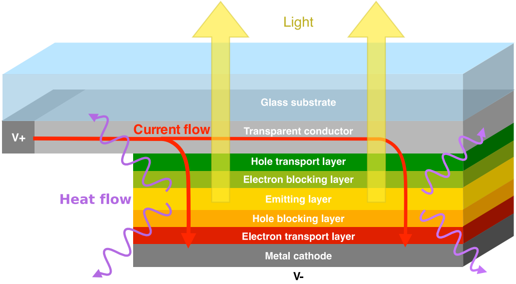
$$F(T) = \exp\left[-\frac{E_{\text{act}}}{k_{\text{B}}}\left(\frac{1}{T} - \frac{1}{T_{\text{a}}}\right)\right]$$

$$\frac{1}{\Theta_{\text{th}}}(T - T_{\text{a}}) = I(V, T) \cdot V$$



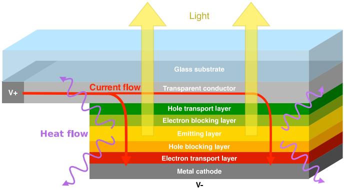
Thermistor behavior with regions of negative differential resistance

Fischer et al. PRL, 2013



- OLEDs consist of various layers and have huge aspect ratios
- Optical transparent top electrode has large sheet resistance
- Sheet resistance leads to significant lateral potential drop

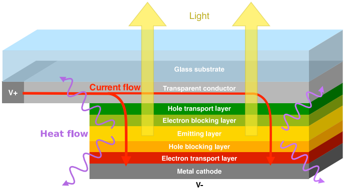
Spatially resolved models needed.



We consider elliptic system consisting of current-flow equation for electrostatic potential φ and heat equation for temperature T

$$-\nabla \cdot \big(\sigma(x, T, \nabla\varphi)\nabla\varphi\big) = 0$$

$$-\nabla \cdot \big(\lambda(x)\nabla T\big) = \eta(x)\sigma(x, T, \nabla\varphi)|\nabla\varphi|^2$$



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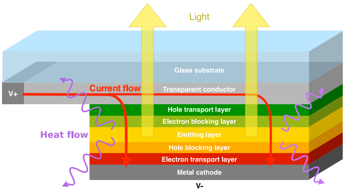
with electrical conductivity $\sigma : \Omega \times \mathbb{R} \times \mathbb{R}^d \to [0, \infty)$ given by

$$\sigma(x, T, \nabla\varphi) = \sigma_0(x)F(x, T)\left[\frac{|\nabla\varphi|}{V_{\text{ref}}/\ell_{\text{ref}}}\right]^{p(x)-2}$$

$$F(x, T) = \exp\left[-\frac{E_{\text{act}}(x)}{k_{\text{B}}}\left(\frac{1}{T} - \frac{1}{T_{\text{a}}}\right)\right]$$



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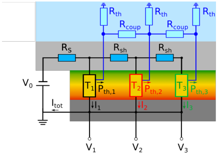
PDE model can be motivated from equivalent circuit model s.t. finite-volume discretization of PDE system correspond to Kirchhoff's circuit rules

(see Fischer et al. 2014, $\rightsquigarrow$ talk by A. Fischer, Wed)



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Mixed boundary conditions

$$\varphi = \varphi^D \quad \text{on } \Gamma_D \qquad \sigma(x,T,\nabla\varphi)\nabla\varphi\cdot\nu = 0 \quad \text{on } \Gamma_N$$

$$-\lambda(x)\nabla T\cdot\nu = \kappa(x)(T - T_{\mathsf{a}}) \quad \text{on } \Gamma := \partial\Omega$$

$$-\nabla \cdot \left(\sigma_0(x) F(x, T) |\nabla \varphi|^{p(x)-2} \nabla \varphi\right) = 0$$
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Properties

- Current-flow equation is of $p(x)$-Laplacian type
- Abrupt change of $p(x)$ between materials:

  Exponent $p(x)$ describes non-Ohmic behavior of materials, $p(x) = 2$ in electrodes (Ohmic) and $p(x) > 2$ in organic materials (e.g. $p(x) = 9.7$)
- For $\nabla\varphi \in L^{p(x)}(\Omega)^d$, Joule heat term in general only in L^1

$$-\nabla \cdot \left(\sigma_0(x) F(x, T) |\nabla \varphi|^{p(x)-2} \nabla \varphi\right) = 0$$
$$-\nabla \cdot \left(\lambda(x) \nabla T\right) = \eta(x) \sigma_0(x) F(x, T) |\nabla \varphi|^{p(x)}$$

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- **(i)** Effective elec. conductivity $\sigma_0 \in L^\infty(\Omega)$ s.t. $0 < \underline{\sigma_0} \le \sigma_0 \le \overline{\sigma_0}$ a.e. in Ω
- **(ii)** Thermal conductivity $\lambda \in L^\infty_+(\Omega)$ s.t. $0 < \underline{\lambda_*} \le \lambda \le \overline{\lambda_*} < \infty$ a.e. in Ω
- **(iii)** Activation energy $E_{\mathsf{act}} \in L^\infty_+(\Omega)$
- **(iv)** Heat transfer coefficient $\kappa \in L^\infty_+(\Omega)$, $\int_\Gamma \kappa(x)\,\mathrm{d}\Gamma > 0$
- **(v)** Light-outcoupling factor $\eta \in L^\infty(\Omega)$, $\eta \in [0, 1]$ a.e. in Ω
- **(vi)** Ambient temperature $T_{\mathsf{a}} > 0$ is constant
- **(vii)** Dirichlet data $\varphi_D \in W^{1,\infty}(\Omega)$
- **(viii)** Power-law exponent $x \mapsto p(x)$ is measurable and

$$1 < p_- := \operatorname*{ess\,inf}_{x\in\Omega} p(x) \le \operatorname*{ess\,sup}_{x\in\Omega} p(x) =: p_+ < \infty$$

Theorem (Bulíček-Glitzky-L. 2016 a)

There exists a weak solution to the coupled system

$$\text{(Current flow)} \qquad -\nabla\cdot\big(\sigma(x,T,|\nabla\varphi|)\nabla\varphi\big) = 0$$

$$\text{(Heat flow)} \qquad -\nabla\cdot\big(\lambda(x)\nabla T\big) = \eta(x)\sigma(x,T,|\nabla\varphi|)|\nabla\varphi|^2$$

with

$$\varphi = \varphi^D \quad \text{on } \Gamma_D \qquad \sigma(x,T,\nabla\varphi)\nabla\varphi\cdot\nu = 0 \quad \text{on } \Gamma_N$$

$$-\lambda(x)\nabla T\cdot\nu = \kappa(x)(T - T_a) \quad \text{on } \Gamma := \partial\Omega$$

where $\varphi \in W^{1,p(x)}(\Omega)$ *and* $T \in W^{1,q}(\Omega)$ *with* $1 \le q < d/(d-1)$, *and*

$$\operatorname{ess\,inf}\varphi^D \le \varphi(x) \le \operatorname{ess\,sup}\varphi^D, \qquad T \ge T_{\mathrm{a}} \quad \text{a.e. in } \Omega.$$

Note: More general constitutive equations for the electrical current can be considered.

1. For $\varepsilon > 0$, introduce regularization

$$f_\varepsilon(x,T,\nabla\varphi) := \eta(x)\frac{\sigma(x,T,\nabla\varphi)|\nabla\varphi|^2}{1+\varepsilon\sigma(x,T,\nabla\varphi)|\nabla\varphi|^2} \le \frac{1}{\varepsilon}$$

2. Solve regularized problem via Galerkin approximation (use strict monotonicity of current law)

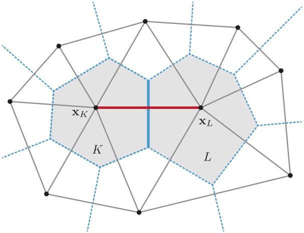
3. For solutions $(\varphi_\varepsilon, T_\varepsilon)$ of regularized problem derive uniform estimates by testing weak formulation with suitable functions, e.g. $\tilde{T} = T_\varepsilon^{-\delta}$ ($\delta \in (0,1)$)

$$\|\nabla\varphi_\varepsilon\|_{p(\cdot)} \le C, \qquad \|T_\varepsilon\|_{W^{1,q}} \le C(q), \quad \text{where } q \in \left[1, \frac{d}{d-1}\right)$$

4. Pass to the limit $\varepsilon \to 0$ in the weak formulation and identify limits exploiting monotonicity again

Our numerical scheme is based on a hybrid finite element/volume approach, see also Bradji–Herbin 2008

$$\int_K \nabla \cdot \mathbf{J}\, \mathrm{d}x = \sum_{L|K} \int_{s_{KL}} \mathbf{J} \cdot \mathbf{n}_{KL}\, \mathrm{da}$$
$$\approx \sum_{L|K} |s_{KL}| J_{KL}$$



Finite volume scheme

Construct approximation J_{KL} of normal flux $\mathbf{J} \cdot \mathbf{n}_{KL}$

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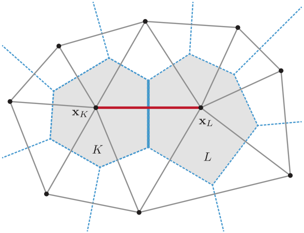


Finite volume scheme

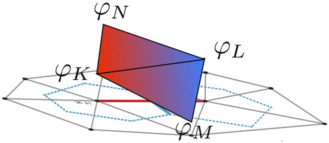
Construct approximation J_{KL} of normal flux $\mathbf{J} \cdot \mathbf{n}_{KL}$

- Heat equation

$$\int_K \nabla \cdot (\lambda(x)\nabla T)\,\mathrm{d}x \approx \sum_{L|K} |s_{KL}| \lambda_{KL} \frac{T_L - T_K}{|x_L - x_K|}$$

Problem: In current-flow equation the conductivity σ depends not only on normal component $\nabla\varphi \cdot \mathbf{n}$ on Voronoi surface but also on tangential components.

Given nodal values $\{\varphi_K\}$ construct
$P1$ finite element interpolant $\widehat{\varphi}_{\mathcal{T}}$



Numerics for thermistor scheme

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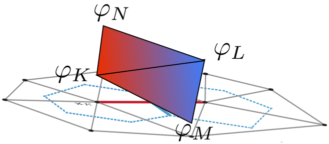

Current-flux approximation (for same material region)

$$\int_K \nabla \cdot \big(\sigma(x, T, |\nabla\varphi|)\nabla\varphi\big)\,\mathrm{d}x = \sum_{L|K} \int_{s_{KL}} \sigma_0(x) F(x,T) |\nabla\varphi|^{p(x)-2} \nabla\varphi \cdot \mathbf{n}\,\mathrm{d}a$$

$$\approx \sum_{L|K} |s_{KL}| F_{s_{KL}}(T_{KL}) \left|\overline{\nabla\varphi_{\mathcal{T}}}\right|^{p_{s_{KL}}-2} \frac{\varphi_L - \varphi_K}{|x_K - x_L|}$$

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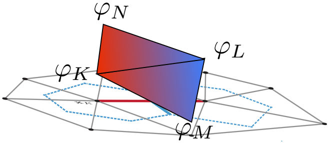

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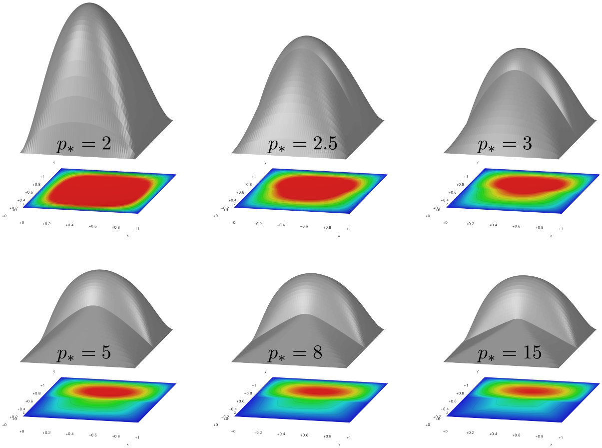
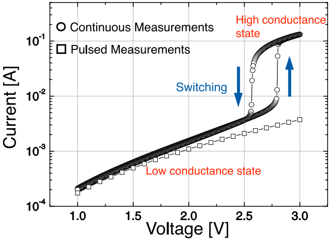
- Averaging of $|\nabla\varphi|$ is necessary for stable schemes on general grids
- At material interface no averaging is performed to avoid artificial lateral diffusion
- Needs boundary conforming Delaunay grids

Simulation study for $p(x)$-Laplace equation $-\nabla\cdot\left(|\nabla u|^{p(x)-2}\nabla u\right)=1$ with Dirichlet boundary conditions



Problem: In voltage-controlled simulations, solutions have to jump into high conductance state, current-controlled simulations numerically unstable



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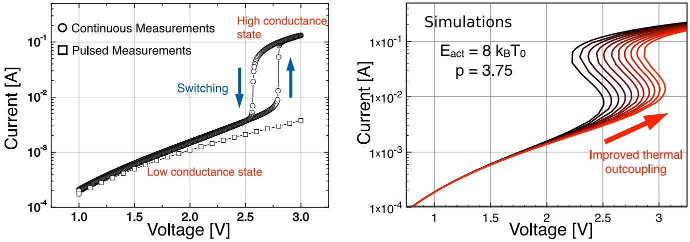

- Simulation of IV curves via numerical path following methods
- Solve extended system for (φ, T, V) with V applied voltage
- Predictor-corrector scheme with step-size control

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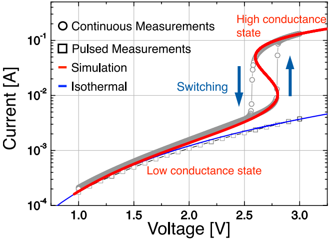

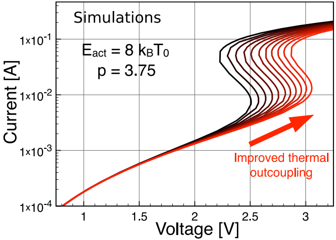

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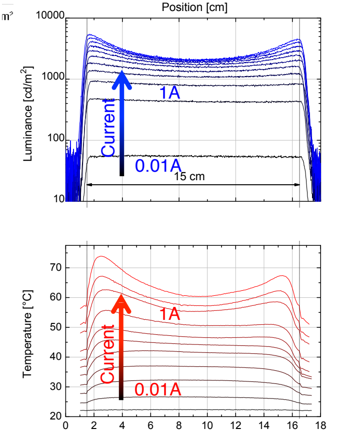
Recall: Counter-intuitive phenomena in large-area OLEDs



. Fischer (IAPP), Adv. Func. Mat. 24 (2014) 3367–3374

- High sheet resistance of optical transparent electrode cannot explain saturation of current
- degradation of material can also be excluded due to lower temperature center of panel
- Effect is caused by interplay between S-NDR and heat conduction

Pattern formation in OLEDs induced by operation modes propagating through device

Operation modes are characterized by

local differential resistance

$$\frac{\mathrm{d}V_x}{\mathrm{d}I_x} = \left(\frac{\mathrm{d}V_x}{\mathrm{d}I_{\text{tot}}}\right)\left(\frac{\mathrm{d}I_x}{\mathrm{d}I_{\text{tot}}}\right)^{-1}$$

I_{tot} total current

I_x local current through stack

V_x local voltage drop

Modes	for	$I_{\text{tot}} \uparrow$
Normal	$V_x \uparrow$	$I_x \uparrow$
S-NDR	$V_x \downarrow$	$I_x \uparrow$
Switched-back	$V_x \downarrow$	$I_x \downarrow$
N-NDR	$V_x \uparrow$	$I_x \downarrow$

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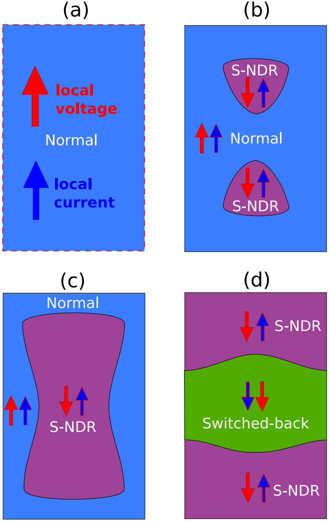
For sufficiently high applied currents center of panel is switched back.



- Introduced $p(x)$-Laplace thermistor model for electrothermal description of organic semiconductor devices
- Existence of solutions via regularization and Galerkin approximation
- Numerical approximation via hybrid finite-volume/element scheme and numerical path following techniques
- Implemented in software protoype
- Pattern formation in OLEDs can be explained by self-heating, Arrhenius law and sheet resistance

Modeling

- Fischer, Pahner, Lüssem, Leo, Scholz, Koprucki, Gärtner, and Glitzky
  Self-heating, bistability, and thermal switching in organic semiconductors, PRL 110, (2013)
- Fischer, Koprucki, Gärtner, Tietze, Brückner, Lüssem, Leo, Glitzky, and Scholz
  Feel the heat: Nonlinear electrothermal feedback in Organic LEDs, AFM 24 (2014)
- Liero, Koprucki, Fischer, Scholz, and Glitzky
  p-Laplace thermistor modeling of electrothermal feedback in organic semiconductor devices, ZAMP 66, (2015)

Analysis

- Glitzky and Liero
  Analysis of $p(x)$-Laplace thermistor models describing the electrothermal behavior of organic semiconductor devices, WIAS-Preprint 2143, (2015)
- Bulíček, Glitzky, and Liero
  Systems describing electrothermal effects with $p(x)$-Laplacian like structure for discontinuous variable exponents, WIAS preprint 2206, 2016
- Bulíček, Glitzky, and Liero
  Thermistor systems of p(x)-Laplace-type with discontinuous exponents via entropy solutions, WIAS preprint 2247, 2016

New operation mode in spatially dependent setting appears

$$\frac{\mathrm{d}V_i}{\mathrm{d}I_i} = \frac{\mathrm{d}V_i}{\mathrm{d}V_{\text{ext}}}\left(\frac{\mathrm{d}I_i}{\mathrm{d}V_{\text{ext}}}\right)$$

	$\frac{\mathrm{d}V_i}{\mathrm{d}V_{\text{ext}}} > 0$	$\frac{\mathrm{d}V_i}{\mathrm{d}V_{\text{ext}}} < 0$
$\frac{\mathrm{d}I_i}{\mathrm{d}V_{\text{ext}}} > 0$	I: normal mode	II: S-NDR
$\frac{\mathrm{d}I_i}{\mathrm{d}V_{\text{ext}}} < 0$	not present	III: switched back