Modeling and simulation of electrically driven quantum light emitters

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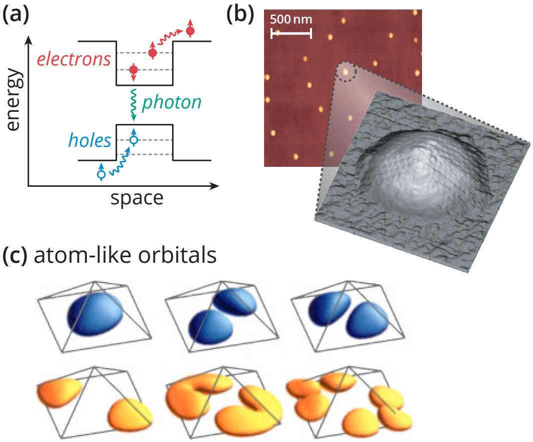
Semiconductor Nanophotonics
SFB 787

Semiconductor quantum dots

- "artificial atoms"
  - discrete energy levels
  - confinement
- nanocrystal structure
- tunable electro-optical properties
- integration in semiconductor devices and photonic resonators
- electrical control

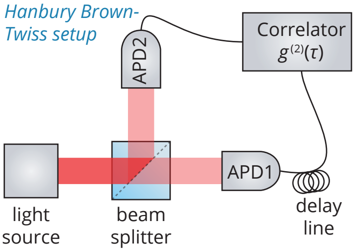
⇒ **promising candidate for novel opto-electronic devices!**

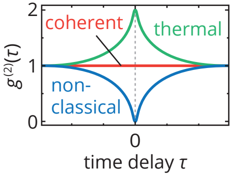


O. Stier et al., *Phys. Rev. B* **59**, 5688–5701 (1999), P. Lodahl et al., *Rev. Mod. Phys.* **87**, 347–400 (2015)

Quantum dot based light sources

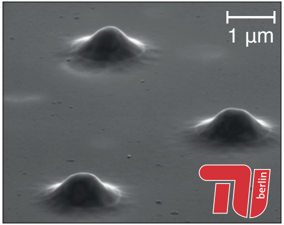
- single-/entangled photon sources
  - emission of non-classical light
  - anti-bunching
- single (or few) QD nanolasers
  - thresholdless lasing

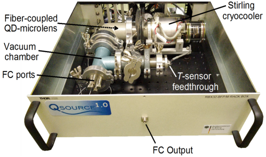




Applications

- quantum information processing
  - quantum cryptography
  - quantum computing
- quantum metrology





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A. Schlehahn et al., *Sci. Rep.* **8**, 1340 (2018), M. Gschrey et al., *Nature Communications* **6**, 7662 (2015)

Modeling

Modeling

Semiconductor transport equations

- transport of electrons and holes in self-consistent electrostatic field

van Roosbroeck system

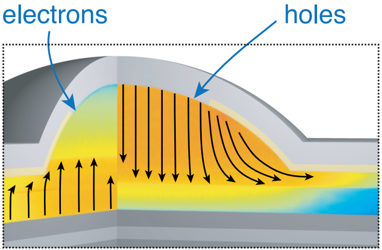
$$-\nabla \cdot \varepsilon \nabla \phi = q\,(C + p - n)$$

$$\partial_t n - \frac{1}{q} \nabla \cdot \mathbf{j}_n = -R(n,p)$$

$$\partial_t p + \frac{1}{q} \nabla \cdot \mathbf{j}_p = -R(n,p)$$

- current density: drift + diffusion

$$\mathbf{j}_n = -q M_n n \nabla \phi + q D_n(n) \nabla n$$



- Fermi Dirac statistics

$$n = N_c F_{1/2}\left(\frac{\mu_c - E_c + q\phi}{k_B T}\right)$$

- device driven by applied voltage
  →boundary conditions

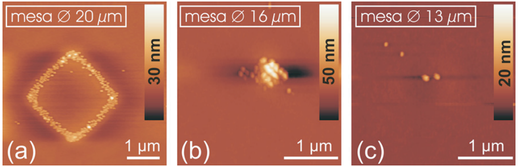
S. Selberherr, *Analysis and Simulation of Semiconductor Devices* (1984)

Modeling

Electrically driven single-photon emitter

- site-controlled QD nucleation above aperture

  A. Strittmatter et al., *Appl. Phys. Lett.* **100**, 093111 (2012)

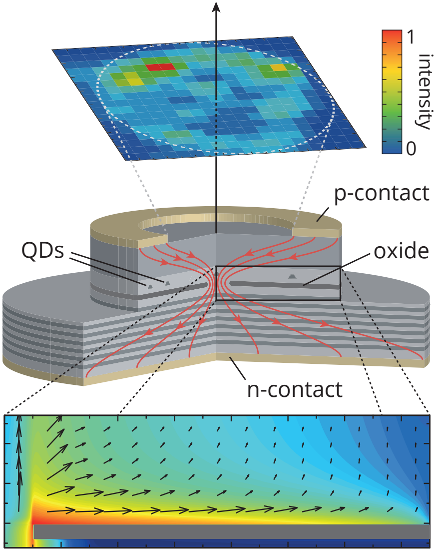


- *goal:* single-photon emission from central QD

  W. Unrau et al., *Appl. Phys. Lett.* **101**, 211119 (2012)
- lateral current spreading above oxide
- analysis and improvement of the design

  M. Kantner et al., *IEEE T. Electron Dev.* **63**, 2036–2042 (2016)

... how about quantum optical properties?



Modeling

Quantum theory of quantum dot light emitters

- electronic transitions between discrete states
- interaction with quantized optical field

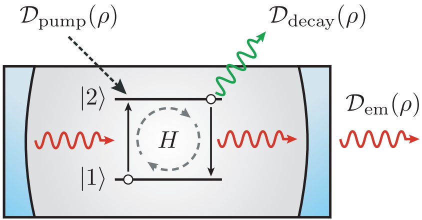
Lindblad master equation

$$\partial_t \rho = -\frac{i}{\hbar}[H, \rho] + \mathcal{D}(\rho)$$

$$H = H^0_{\text{e,h}} + H^0_{\text{ph}} + H_{\text{Coulomb}} + H_{\text{light-matter}}$$

$$\mathcal{D}(\rho) = \sum_\alpha \gamma_\alpha \left(A_\alpha \rho A^\dagger_\alpha - \frac{1}{2}\{A^\dagger_\alpha A_\alpha, \rho\} \right)$$

- computation of quantum optical properties

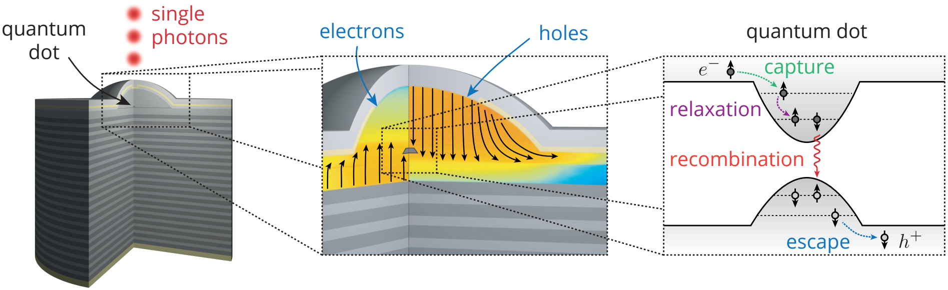


- *Hamiltonian system*: unitary time-evolution
- *open quantum system*: energy dissipation + decoherence

W. Chow, F. Jahnke, *Prog. Quant. Electron.* **37**, 109–194 (2013)

H.-P. Breuer, F. Petruccione, *Theory of open quantum systems* (2002)

Modeling



Can we combine both levels of description?

hybrid model = van Roosbroeck system + Lindblad master equation

= semiconductor transport + open quantum systems

= partial differential equations + operator evolution equation

Modeling

electrostatic interation
Poisson's equation

control
bias, device design

continuum carriers (bulk)
- drift-diffusion + recombination
- quasi-equilibrium distributions

confined carriers (QD)
- quantum many-body problem
- non-equilibrium distribution

carrier capture/ escape
dissipation superoperators

quantum optics
- photon emission
- correlation functions

Hybrid quantum-classical model

$$-\nabla\cdot\varepsilon\nabla\phi = q\left(C + p - n + Q(\rho)\right)$$

$$\partial_t n - \frac{1}{q}\nabla\cdot\mathbf{j}_n = -R - S_n(\rho; n, p, \phi)$$

$$\partial_t p + \frac{1}{q}\nabla\cdot\mathbf{j}_p = -R - S_p(\rho; n, p, \phi)$$

$$\partial_t \rho = -\frac{i}{\hbar}[H, \rho] + \mathcal{D}(\rho; n, p, \phi)$$

Kantner, Mittnenzweig, Koprucki
Phys. Rev. B **96**, 205301 (2017)

Modeling

Quantum master equation: $\partial_t \rho = -\frac{i}{\hbar}[H, \rho] + \mathcal{D}(\rho)$

Hamiltonian

- single-particle energies + interactions

$$H = H_0 + H_I$$

H_0 electrons, holes + photons
H_I Coulomb interaction, light-matter interaction

- Hamiltonian conserves (net-)charge

$$\boxed{[H, N] = 0} \qquad N = n_e - n_h$$

Modeling

Quantum master equation: $\partial_t \rho = -\frac{i}{\hbar}[H,\rho] + \mathcal{D}(\rho; n, p, \phi)$

Dissipation superoperator

- capture + escape, spontaneous emission, dephasing, relaxation, ...

$$\mathcal{D}(\rho; n, p, \phi) = \mathcal{D}_e(\rho; n, p, \phi) + \mathcal{D}_h(\rho; n, p, \phi) + \mathcal{D}_0(\rho; n, p, \phi)$$

with

$$\underbrace{\mathrm{tr}\left(N\mathcal{D}_{e/h}(\rho; n, p, \phi)\right) \neq 0,}_{\text{not charge conserving}} \qquad \underbrace{\mathrm{tr}\left(N\mathcal{D}_0(\rho; n, p, \phi)\right) = 0}_{\text{charge conserving}}$$

$$\begin{tikzcd} |\psi_i\rangle \arrow[r, bend left, "A_\alpha(\omega)"] & |\psi_f\rangle \arrow[l, bend left, "A_\alpha^\dagger(\omega)"] \end{tikzcd}$$

- general form with *Lindbladian* $L_A(\rho) = A\rho A^\dagger - \frac{1}{2}\{A^\dagger A, \rho\}$

$$\mathcal{D}(\rho; n, p, \phi) = \sum_{\omega > 0} \sum_{\alpha} \Big(\gamma_\alpha(\omega; n, p, \phi) L_{A_\alpha(\omega)}(\rho) + \gamma_\alpha(-\omega; n, p, \phi) L_{A_\alpha^\dagger(\omega)}(\rho) \Big)$$

A_α: quantum jump operators, γ_α: transition rates, $\hbar\omega$: transition energy

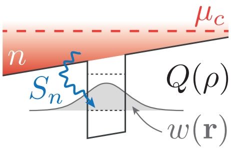
Modeling

Coupling terms

- charge density of (single) quantum dot

$$-\nabla \cdot \varepsilon \nabla \phi = q\,(C + p - n + Q(\rho)) \quad \longrightarrow \quad Q(\rho) = -w(\mathbf{r})\,\mathrm{tr}\,(N\rho)$$

with spatial profile $\int_\Omega \mathrm{d}^3 r\, w(\mathbf{r}) = 1$



- carrier transport equations

$$\partial_t n - \frac{1}{q}\nabla \cdot \mathbf{j}_n = -R - S_n(\rho; n, p, \phi) \quad \longrightarrow \quad S_n(\rho; n, p, \phi) = +w(\mathbf{r})\,\mathrm{tr}\,(N\mathcal{D}_e(\rho; n, p, \phi))$$

$$\partial_t p + \frac{1}{q}\nabla \cdot \mathbf{j}_p = -R - S_p(\rho; n, p, \phi) \quad \longrightarrow \quad S_p(\rho; n, p, \phi) = -w(\mathbf{r})\,\mathrm{tr}\,(N\mathcal{D}_h(\rho; n, p, \phi))$$

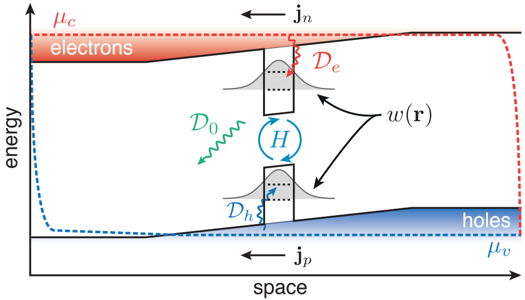
$\Rightarrow$ **local charge conservation** $\quad \nabla \cdot (\mathbf{j}_n + \mathbf{j}_p - \varepsilon \partial_t \nabla \phi) = 0$

Thermodynamics

Thermodynamics

Hybrid quantum-classical model

$$-\nabla \cdot \varepsilon \nabla \phi = q\,(C + p - n + Q(\rho))$$
$$\partial_t n - \frac{1}{q}\nabla \cdot \mathbf{j}_n = -R - S_n(\rho; n, p, \phi)$$
$$\partial_t p + \frac{1}{q}\nabla \cdot \mathbf{j}_p = -R - S_p(\rho; n, p, \phi)$$
$$\partial_t \rho = -\frac{i}{\hbar}[H, \rho] + \mathcal{D}(\rho; n, p, \phi)$$



... consistency with thermodynamics?

- thermodynamic equilibrium?
- (quantum) detailed balance?
- second law of thermodynamics?

Thermodynamic correctness crucial in device simulation!

Thermodynamic equilibrium

- minimize equilibrium grand potential

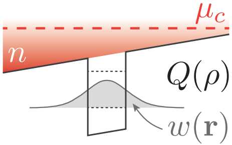
$$\Phi_{\text{eq}}[n,p,\rho] = \mathcal{F}[n,p,\rho] - \mu_{\text{eq}} \int_\Omega \mathrm{d}^3 r \, (n - p - Q(\rho))$$

with free energy functional

$$\mathcal{F}[n,p,\rho] = \mathcal{F}_{\text{classical}}[n,p] + \mathcal{F}_{\text{quantum}}[\rho] + U_{\text{electrostatic}}[n,p,\rho]$$

⇒ equilibrium density matrix

$$\rho_{\text{eq}} = \frac{1}{Z} \exp\left(-\beta\left[\left(H - \left(\mu_{\text{eq}} + q\langle\phi_{\text{eq}}\rangle_w\right)N\right)\right]\right)$$



with $\langle\phi\rangle_w = \int_\Omega \mathrm{d}^3 r \, w(\mathbf{r})\,\phi(\mathbf{r})$

Quantum detailed balance condition

- dissipation superoperator

$$\mathcal{D}(\rho; n, p, \phi) = \sum_{\omega>0} \sum_{\alpha} \Big(\gamma_\alpha(\omega; n, p, \phi) L_{A_\alpha(\omega)}(\rho) + \gamma_\alpha(-\omega; n, p, \phi) L_{A_\alpha^\dagger(\omega)}(\rho) \Big)$$

- transition rates driven by averaged macroscopic potentials

$$\gamma_\alpha(\omega; n, p, \phi) \to \gamma_\alpha(\omega; \langle\mu_c\rangle_w, \langle\mu_v\rangle_w, \langle\phi\rangle_w) \quad \longrightarrow \quad \text{fit to microscopic calculations}^{1,2}$$

- **generalized Kubo-Martin-Schwinger-condition**

$$\frac{\gamma_\alpha\left(-\omega, \langle\mu_c\rangle_w, \langle\mu_v\rangle_w, \langle\phi\rangle_w\right)}{\gamma_\alpha\left(+\omega, \langle\mu_c\rangle_w, \langle\mu_v\rangle_w, \langle\phi\rangle_w\right)} = \exp\left(-\beta\left[\hbar\omega - \left(\langle\mu_\alpha\rangle_w + q\langle\phi\rangle_w\right)\ell_\alpha\right]\right)$$

- quantum detailed balance: $\boxed{\mathcal{D}_\alpha\left(\rho_{\mathrm{eq}}; n_{\mathrm{eq}}, p_{\mathrm{eq}}, \phi_{\mathrm{eq}}\right) \equiv 0}$

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[1] T. R. Nielsen et al., *Phys. Rev. B* **69**, 235314 (2004), [2] A. Wilms et al., *Phys. Rev. B* **88**, 235421 (2013)

Entropy production rate

$$0 \leq \frac{\mathrm{d}S_{\text{tot}}}{\mathrm{d}t} = \frac{1}{T}\int_\Omega \mathrm{d}^3r \, (\mu_c - \mu_v)\, R + \frac{1}{qT}\int_\Omega \mathrm{d}^3r \, (\mathbf{j}_n \cdot \nabla\mu_c + \mathbf{j}_p \cdot \nabla\mu_v) + \\ + k_B \operatorname{tr}\left(\left[\log\rho - \beta H\right] \mathcal{D}_0(\rho)\right) + \\ + k_B \operatorname{tr}\left(\left[\log\rho - \beta\left(H - \left[\langle\mu_c\rangle_w + q\langle\phi\rangle_w\right] N\right)\right] \mathcal{D}_e(\rho)\right) + \\ + k_B \operatorname{tr}\left(\left[\log\rho - \beta\left(H - \left[\langle\mu_v\rangle_w + q\langle\phi\rangle_w\right] N\right)\right] \mathcal{D}_h(\rho)\right)$$

- each term non-negative:
  - pseudo-convex form $R = r(n,p,\phi)\left(1 - e^{-\beta(\mu_c - \mu_v)}\right)$, $x(1 - e^{-x}) \geq 0 \; \forall x \in \mathbb{R}$
  - Spohn's inequality[1]: $\operatorname{tr}\left(\left(\log\rho - \log\rho_\alpha^*\right)\mathcal{D}_\alpha(\rho)\right) \geq 0$ for $\mathcal{D}_\alpha(\rho_\alpha^*) = 0$

Hybrid model obeys second law of thermodynamics!

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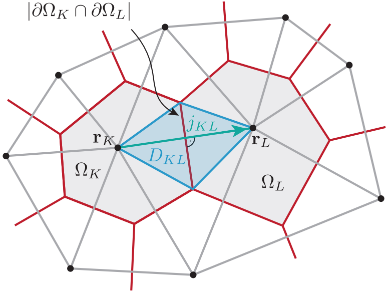
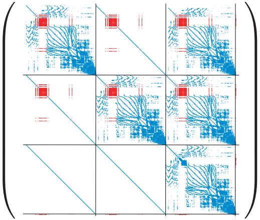
[1] H. Spohn, *J. Math. Phys.* **19**, 1227 (1978)

Numerical method

Finite volumes method

- discretization on Voronoï boxes using divergence theorem
- Scharfetter-Gummel scheme for robust discretization of drift + diffusion flux
  - smooth interpolation between upwind and central difference scheme
- implicit Euler in time
- non-local coupling with Lindblad equation
- full Newton iteration



$J =$ 

Cryogenic temperatures

- problem is ill-conditioned as $T \to 0\,\mathrm{K}$

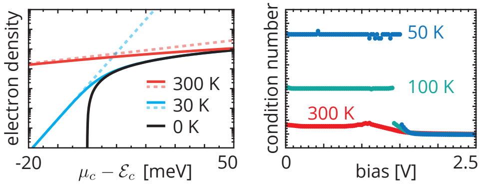
$$n \propto \exp\left(\frac{\mu_c - \mathcal{E}_c}{k_B T}\right)$$

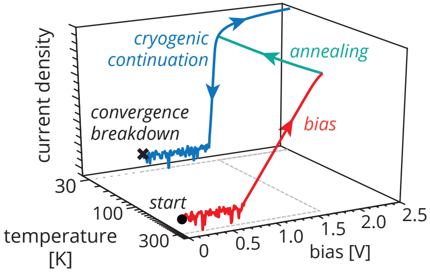
- generalized Scharfetter-Gummel scheme for degenerate carrier statistics

  T. Koprucki et al., *Opt. Quant. Elect.* **47**, 1327 (2015)

- annealing method for cryogenic temperature

  M. Kantner, T. Koprucki, *Opt. Quant. Elect.* **48**, 543 (2016)

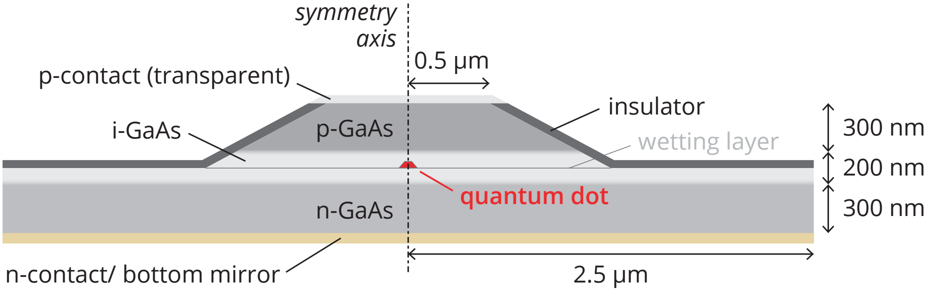




Application

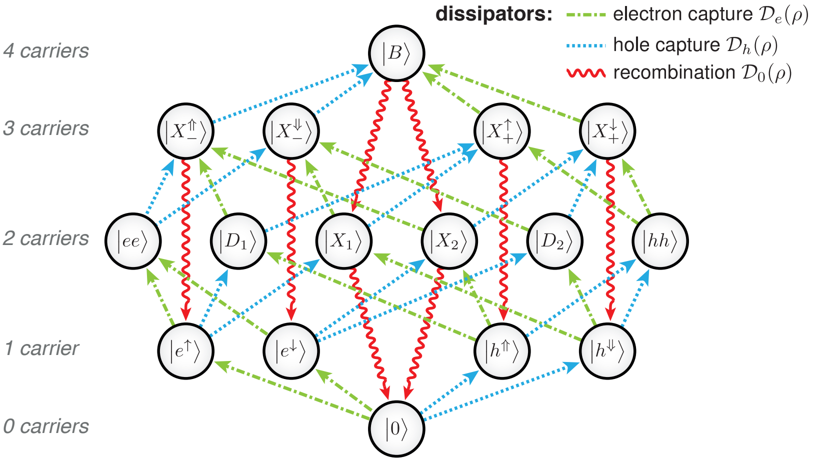
Application

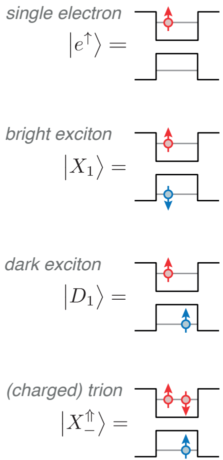
Electrically driven single-photon source



- GaAs-based pin-diode structure
- InAs-QD centered in intrinsic zone
- low *Q* resonator (no top mirror)

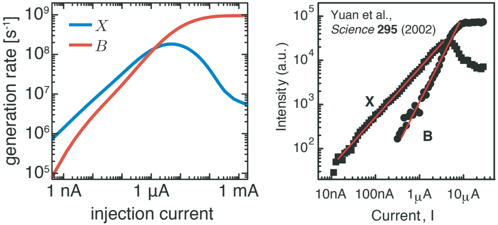
Quantum dot states



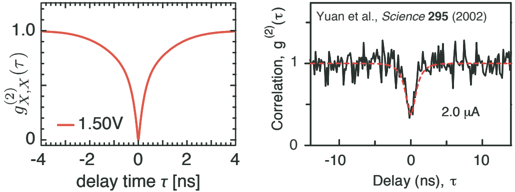


Simulation results: Stationary operation

(a) Single-photon generation rate

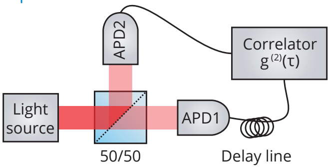


(b) 2nd order intensity correlation function



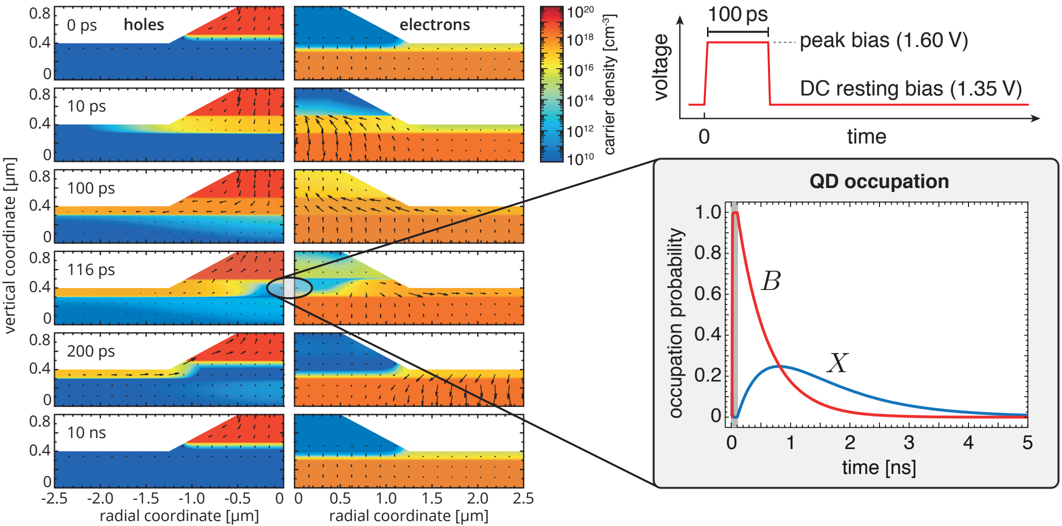
Hanbury Brown-Twiss experiment

$$g^{(2)}(\tau) = \frac{\langle a^\dagger(0)a^\dagger(\tau)a(\tau)a(0)\rangle}{\langle a^\dagger(0)a(0)\rangle^2}$$



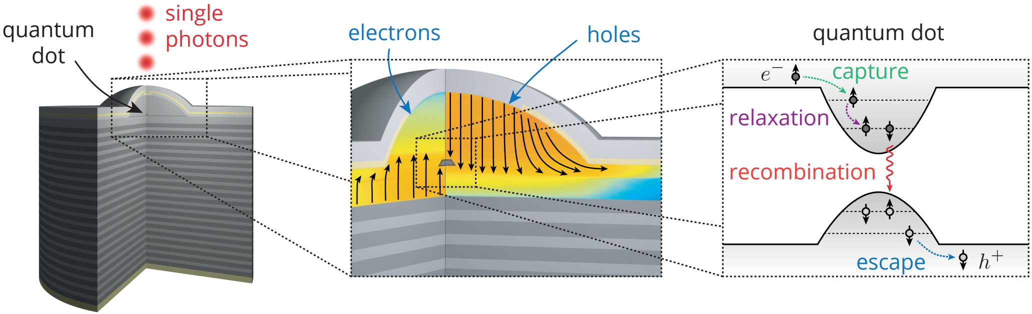
Application

Simulation results: Pulsed operation



Summary

Summary



- **hybrid quantum-classical** modeling of quantum light sources
- consistency with (non-)equilibrium **thermodynamics**
- application to electrically driven **single-photon sources**
- **carrier transport** and **quantum optics** out of one box

M. Kantner, M. Mittnenzweig and T. Koprucki, *Phys. Rev. B* **96**, 205301 (2017)

Leibniz Association

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