1.1 Hybrid Quantum-classical Modeling of Electrically Driven Quantum Light Sources

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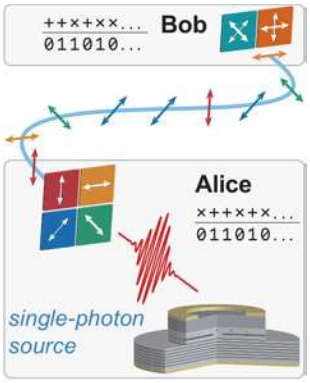

Fig. 1: *Quantum key distribution with single photons: A secret message is transferred from Alice to Bob using the BB84 protocol*

The quantum theory of light started more than a century ago when Max Planck calculated the black body radiation spectrum by assuming that light is emitted in discrete, fundamental units of energy that we denote today as *photons*. Based on the same hypothesis, which involves a particle-like conception of the electromagnetic field that was formerly understood as an entirely wave-like phenomenon, Albert Einstein gave an explanation of the photoelectric effect for which he was awarded with the Nobel prize in 1921. Subsequently, also wave-like properties of electrons in the form of matter waves were discovered, which finally lead to the advent of quantum mechanics – a scientific revolution continuing until the present day. The classical theory of electromagnetism was superseded in the following by quantum electrodynamics by the mid of the 20th century, which forms the basis of our modern understanding of light, matter, and their interaction on a fundamental level. Soon it was discovered that light can exist in different states, e.g., coherent states (lasers), thermal states (blackbody radiation) and more exotic states such as squeezed states. However, it was not until 1977 when H.J. Kimble et al. first demonstrated the emission of a single photon from a single atom at one time, which gave further evidence that light consists of photons. Such a single-photon state of the electromagnetic field is a truly non-classical state of light. The radiation generated by a single emitter shows phenomena like *photon anti-bunching* [1] (i.e., the photons emitted by the source tend to keep a distance due to non-classical intensity fluctuations) that can only be understood in terms of a quantized electromagnetic field theory.

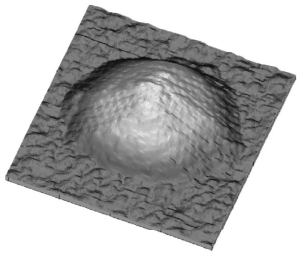

Fig. 2: *STM-image of an InAs semiconductor QD. Picture taken from Márquez et al., Appl. Phys. Lett.,* ***78*** *(2001), 2309.*

The insights obtained in quantum optics with single photons and entangled photon pairs stimulated progress in quantum information theory, which aims at, e.g., using single photons as *qubits* – units of quantum information – for optical quantum computing and information processing tasks. Some of the most promising applications in that field are the various cryptographic methods for secure data transmission based on *quantum key distribution* (e.g., BB84 protocol, E91 protocol); see Figure 1. The security of quantum key distribution relies on well-approved quantum mechanical effects (no-cloning theorem, collapse of the wave function etc.) rather than on assumptions on the available computational power or the efficiency of algorithms as in classical encryption methods.

The experimental preparation of single-photon states using single atoms requires a huge technical effort making the technology extremely expensive and inappropriate for real-world applications. However, with the advent of semiconductor quantum dots (QDs), which are nano-crystalline structures (see Figure 2) that provide an atom-like three-dimensional confinement of electrons within solid-state structures, the fundamental research in quantum optics merged with the well-developed semiconductor technology. Semiconductor QDs are frequently denoted as *artificial atoms* as they represent a solid-state analogue of a single atom with tailorable electro-optical properties. Moreover, QDs can be directly integrated into semiconductor devices and micro-resonators by standard manufacturing techniques, which has lead to many novel concepts for opto-electronic and photonic devices including single-photon sources and ultimately downsized QD nanolasers.

Today, semiconductor quantum optics is on the leap from the lab to commercial applications [1]. To support the development of novel devices, efficient mathematical models and simulation tools are needed to optimize particular device concepts, provide insights into internal physics, and reduce the development costs. In particular, for electrically driven devices, which are desirable for practical applications, the understanding of the current flow is an essential basis for the improvement of certain device designs. For example, Figure 3 shows a single-photon-emitting diode featuring an oxide-confined aperture that is intended to efficiently funnel the current (red lines) into the central QD above the aperture. The experimentally recorded electroluminescence map, however, revealed optical activity of parasitic QDs far away from the aperture. This counterintuitive phenomenon was eventually understood on the basis of carrier transport simulations using the van Roosbroeck system, which showed a rapid lateral current spreading right above the oxide explaining the observations [2]. On the other hand, the van Roosbroeck system makes no predictions on the quantum optical properties of the radiation emitted by the device. Hence, many important figures of merit, like, e.g., the second-order intensity correlation function related to the above-mentioned photon anti-bunching effect, are not accessible by the semi-classical transport model. In order to enable a quantum optical analysis of the device and to simulate electrically driven quantum light sources on a comprehensive level, one has to combine classical device physics with cavity quantum electrodynamics. In particular, it is required to connect semi-classical semiconductor transport theory (e.g., using the van Roosbroeck system) with quantum optical models from the theory of open quantum systems, as illustrated in Figure 4. This was recently achieved in [3] by coupling the van Roosbroeck system to a quantum master equation in Lindblad form.

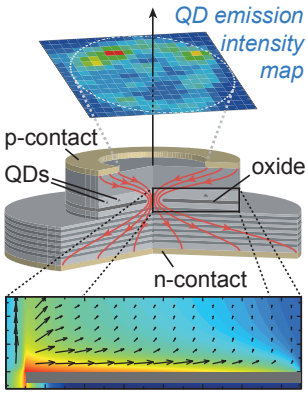

Fig. 3: *Lateral current spreading in an oxide-confined single-photon source leading to unwanted optical activity of parasitic QDs [2]*

The research on quantum-classical hybrid models at WIAS is embedded in the long-term collaborations with experimental groups on single-photon sources (Technische Universität Berlin, funded by DFG CRC 787 *Semiconductor Nanophotonics*). It was stimulated by the ERC-Advanced Grant *AnaMultiScale* on the analysis of multiscale systems driven by functionals that had a focus on the derivation of consistent multi-physics models obeying the fundamental laws of non-equilibrium thermodynamics.

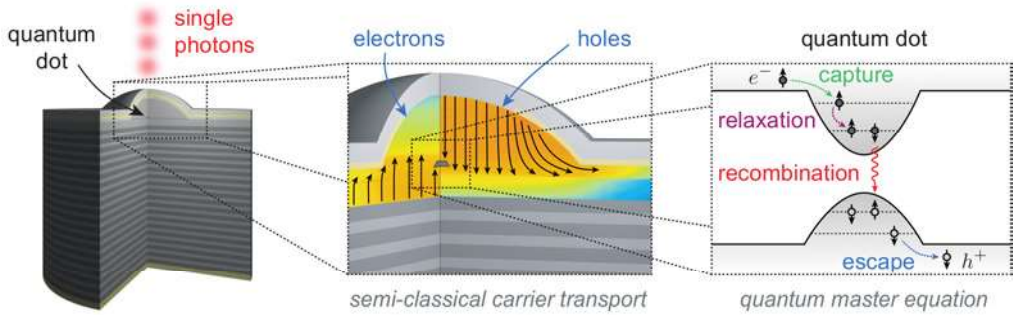

Fig. 4: *The hybrid quantum-classical modeling approach for quantum light sources combines semi-classical carrier transport theory with microscopic models for the QD-photon system*

Combining classical device physics with quantum mechanics

The van Roosbroeck system describes the transport of electrons and holes in macroscopic semiconductor structures in a semi-classical approximation. The charge transport is modeled by a system of reaction-drift-diffusion equations for the electron and hole densities n and p that are coupled to Poisson's equation describing their self-consistently generated electrostatic potential ϕ.

The gradient of the electrostatic potential in turn generates the drift part of the currents. Moreover, electron-hole pairs can be generated or recombine in the semiconductor material modeled by the net-recombination rate R. In [3], we introduced a hybrid quantum-classical model that self-consistently couples the van Roosbroeck system with a quantum master equation in Lindblad form, which is an operator equation describing the evolution of the quantum mechanical density matrix ρ:

$$-\nabla \cdot \varepsilon \nabla \phi = q\,(p - n + C + Q\,(\rho))\,, \tag{1}$$

$$\partial_t n - \frac{1}{q}\nabla \cdot \mathbf{j}_n = -R - S_n\,(\rho; n, p, \phi)\,, \tag{2}$$

$$\partial_t p + \frac{1}{q}\nabla \cdot \mathbf{j}_p = -R - S_p\,(\rho; n, p, \phi)\,, \tag{3}$$

$$\partial_t \rho = -\frac{i}{\hbar}[H, \rho] + \mathcal{D}\,(\rho; n, p, \phi)\,. \tag{4}$$

The model system (1)–(4) is based on a Born approximation separating continuum and confined carriers where the transport of the freely roaming continuum carriers by drift and diffusion is described by the van Roosbroeck system (1)–(3), whereas the bound QD carriers evolve according to the Lindblad master equation (4). The coupling structure is illustrated in Figure 5. The Lindblad master equation models the evolution of an open quantum many-body system, where the internal Hamiltonian dynamics of the quantum system is described by the commutator term $\sim i[H, \rho]$ and the dissipative interaction with the macroscopic environment is mediated by the dissipation superoperator $\mathcal{D}\,(\rho; n, p, \phi)$. The latter includes, e.g., capture and escape of carriers from the continuum states to the confined QD states, spontaneous decay of bound excitons, and the emission of cavity photons from the system. The dissipative interactions can change the charge of the quantum system, while the Hamiltonian evolution leaves it invariant. The backaction of the quantum system on its macroscopic environment is reflected by novel coupling terms in the van Roosbroeck system, which are the scattering rates S_n, S_p describing the loss of continuum carriers in the continuity equations (2)–(3) for electrons and holes and the (net-)charge density $Q(\rho)$ of the QD contributing to the right-hand side of Poisson's equation (1). These macroscopic coupling terms can be expressed as expectation values of certain Hermitian operators, which depend on the state of the quantum mechanical density matrix ρ [3].

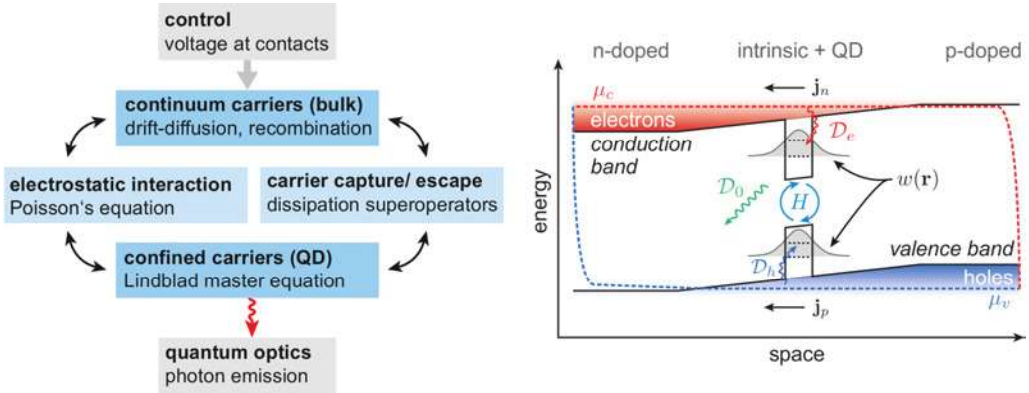

Fig. 5: *Illustration of the hybrid modeling approach and the structure of the couplings between the classical and the quantum mechanical subsystem*

Consistency with thermodynamics and GENERIC structure

The consistency with fundamental laws of non-equilibrium thermodynamics is crucial in semiconductor device modeling, in particular, when it comes to multi-physics applications where mathematical models from different fields need to be coupled in a reasonable way. In [3], we showed that the hybrid model (1)–(4) meets this requirement. The central quantity in the analysis of the system's thermodynamic properties is the free energy functional

$$\mathcal{F}(n, p, \rho) = \mathcal{F}_{\text{classical}}(n, p) + \mathcal{F}_{\text{quantum}}(\rho) + U_\phi(p - n + Q(\rho)), \tag{5}$$

which comprises the free energy contributions of the quasi-free electrons and holes that are subject to the van Roosbroeck model, the free energy of the quantum system, and the electrostatic interaction energy U_ϕ generated by the Coulomb interaction of the charges in the system. Based on (5), it can be shown that the system guarantees a non-negative entropy production rate under direct-current bias conditions, which implies consistency with the second law of thermodynamics.

From a mathematical point of view the thermodynamic consistency of the hybrid system (1)–(4) is reflected by the fact that our model falls into the class of damped Hamiltonian systems within the GENERIC framework. GENERIC is an acronym for *General Equations for Non-Equilibrium Reversible Irreversible Coupling* and provides a thermodynamically consistent way of coupling reversible Hamiltonian dynamics with irreversible dissipative dynamics. In our case, a damped Hamiltonian GENERIC system is defined by a quadruple $(\mathbf{Z}, \mathcal{F}, \mathbb{K}, \mathbb{J})$, where $\mathbf{Z}$ is the state space, and $\mathcal{F}(z)$ is the free-energy functional on it. The state variable of the system is given by $z = (n, p, \rho)$. Moreover, the state space carries two geometric structures, namely the Poisson structure $\mathbb{J}$ that generates the Hamiltonian evolution and the Onsager operator $\mathbb{K}$ driving the dissipative dynamics. Together, the time evolution of the system is given by

$$\partial_t z = \mathbb{J}(z)\, D\mathcal{F}(z) - \mathbb{K}(z)\, D\mathcal{F}(z).$$

The Onsager operator $\mathbb{K}(z)$ is positive and symmetric, whereas $\mathbb{J}(z)$ is antisymmetric and satisfies the Jacobi identity. The second law of thermodynamics is encoded in the positivity and symmetry of $\mathbb{K}$ that follows from microscopic reversibility of the underlying microscopic dynamics. In our hybrid model, the evolution of the semi-classical part is purely dissipative such that the Poisson structure only acts on the quantum mechanical part via

$$\mathbb{J}(\rho)A = \frac{i}{\hbar}[\rho, A].$$

Inserting $A = H + k_B T \log \rho$ exactly gives the Hamiltonian part in (4). The quantum-classical coupling of (1)–(4) is generated by an Onsager operator, i.e., there exists a positive, symmetric $\mathbb{K}_{\text{coupling}}(n, p, \rho)$ such that

$$\left(S_n, S_p, \mathcal{D}\right)^T = \mathbb{K}_{\text{coupling}}(n, p, \rho) \cdot D\mathcal{F}(n, p, \rho).$$

The Onsager operator $\mathbb{K}(\rho)$ was originally introduced in [4], where it was shown that every Lindblad master equation satisfying detailed balance is a damped Hamiltonian system in the sense of GENERIC. The operator $\mathbb{K}(\rho)$ defines a transport metric on the space of density matrices and generates a non-commutative analogue of optimal transport distances for probability distributions.

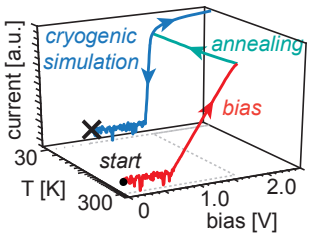

Fig. 6: *Annealing scheme for the numerical simulation of carrier transport at cryogenic temperatures [5]*

Application

The system (1)–(4) is applied to the numerical simulation of an electrically driven single-photon source shown in Figure 7(a), where a single QD is embedded in the intrinsic region of a p-i-n diode. In the case of a leaky resonator, the light-matter interaction is weak, and the quantum system can be described by a purely electronic Hamiltonian. We consider a QD that can be occupied by up to two electrons and two holes, such that the model comprises several multi-particle states including bright and dark excitons and the biexciton. The device geometry, transport parameters, capture rate models, and further details can be found in [3]. The extremely low operation temperatures of quantum light sources cause severe convergence issues for standard numerical routines that can be handled by using the annealing technique [5] illustrated in Figure 6. The simulation results for a pulsed excitation of the device show a biexciton cascade leading to a single-photon emission from the bright exciton state on the order of nanoseconds after the excitation pulse; see Figure 7(b). The numerical results are found to be in good agreement with experimental observations.

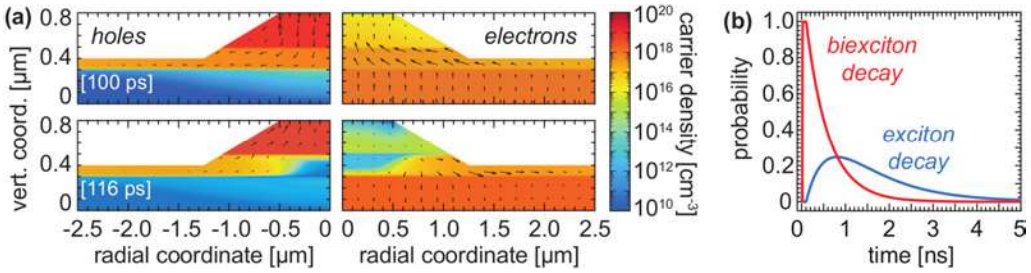

Fig. 7: *The pulsed electrical excitation of the device leads to the emission of non-classical light via the biexciton cascade*

Conclusion

By combining classical device physics with microscopic models from semiconductor quantum optics, we obtained a hybrid quantum-classical model system that can be applied to the simulation of electrically driven quantum light sources. The well-behaved thermodynamic properties of the combined model system are reflected by the underlying mathematical structure.

References

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