

# Effective Microscopic Theory of Quantum Dot Superlattice Solar Cells

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**Abstract**—We introduce a quantum dot orbital tight-binding non-equilibrium Green’s function approach for the simulation of novel solar cell devices where both absorption and conduction is mediated by quantum dot states. By the use of basis states localized on the quantum dots, the computational real space mesh of the Green’s function is coarse-grained from atomic resolution to the quantum dot spacing, which enables the simulation of extended devices consisting of many quantum dot layers.

## I. INTRODUCTION

Extended quantum dot superlattices (QDSL) as found in third generation solar cell devices [1], [2] consist of a large number of weakly coupled quantum dots, with a number of involved atoms that exceeds the limit that may still be handled by even today’s largest supercomputers. For that reason, atomistic simulations of QDSL make use of the symmetry and periodicity properties present in the idealized structure. However, large built-in fields and any kind of disorder make the electronic structure deviate from the minibands found for the latter. Furthermore, even though the electronic minibands be narrow and there may exist gaps in the phonon spectrum, there will be a large amount of scattering, leading to a further localization of the wavefunction and preventing coherent transport in the device. It is thus desirable to derive a computational model for the extraction of the device characteristics that is able to benefit from the localized nature of the QD wavefunctions and at the same time allows for the use of advanced quantum-kinetic theories for the computation of photogeneration and transport over the full device, such as the non-equilibrium Green’s function (NEGF) approach developed for quantum well solar cells in [3], [4].

## II. APPROACH

The hybridization approach uses the perturbative expansion of the QDSL wave function in terms of the eigenstates of the isolated dots. This approximation is reasonable in the regime of high confining barriers and corresponding small overlap of the wavefunction of neighboring dots. The resulting molecular orbital approach can be interpreted as a tight-binding theory with quantum dot orbitals replacing the atomic orbitals, and is in spirit similar to the first NEGF models of quantum well superlattices for quantum cascade laser simulations [5], [6].

Since the wave functions and energies of the quantum dot states are determined separately for each individual dot, the corresponding computational domain is small enough to allow, in principle, for the use of accurate ab-initio methods. In this paper, a simple effective mass approximation is used to compute the QD orbitals. To agree with full real space calculation, the local variation of the electrostatic mean-field potential needs to be included in the solution of the Schrödinger equation.

Each quantum dot provides a set of eigenfunctions  $\psi_{im}(\mathbf{r})$  and eigenenergies  $\varepsilon_{im}$ ,  $i = 1, 2, \dots, N_D$ ,  $m = 1, 2, \dots, M$ , where  $N_D$  is the number of QD,  $M$  the number of states considered and  $b$  is the particle species index, i.e. electron or hole, respectively. In order to use the single particle QD eigenstates as a tight-binding basis in analogy to the atomic case, a Löwdin-orthogonalization procedure [7] needs to be applied. Fig. 1 shows the first three QD orbitals for a cubic Si QD of 2 nm edge length embedded laterally ( $x$ - $y$ ) in  $\text{SiO}_2$  and vertically ( $z$ ) in  $\text{SiC}$  as determined within simple effective mass theory (EMA).

The non-interacting nearest-neighbor tight-binding Hamiltonian in this QD orbital basis is

$$\begin{aligned} \mathcal{H}_0(t) &= - \sum_{\langle i,j \rangle} \sum_{m,n=1}^M t_{ij}^{nm} \hat{d}_{in}^\dagger(t) \hat{d}_{jm}(t) + \sum_{i=1}^{N_D} \sum_{m=1}^M \tilde{\varepsilon}_{im} \hat{n}_{im}(t) \\ &\equiv \sum_{i,j=1}^{N_d} \sum_{m,n=1}^M h_{ij}^{mn} \hat{d}_{in}^\dagger(t) \hat{d}_{jm}(t), \end{aligned} \quad (1)$$

where  $\langle i, j \rangle$  are nearest-neighbor sites,  $\mathbf{t}$  is the hopping matrix,  $\hat{n}$  the density operator and  $\tilde{\varepsilon}_{im} = \varepsilon_{im} + \bar{U}_i$ , with  $\bar{U}_i$  the average value for the Hartree potential of Coulomb interaction at the dot position. The heterostructure potential does not longer appear explicitly, since it has been considered in the determination of the TB-parameters  $\varepsilon$  and  $\mathbf{t}$ .

The above Hamiltonian is used in the equations for the steady state non-equilibrium Green’s functions, including interaction self-energies for the coupling to photons and phonons, as well as the contact self-energy. The expressions for the interaction self-energies are determined as the Fock term within many-body perturbation theory on the level of a selfconsistent Born approximation.

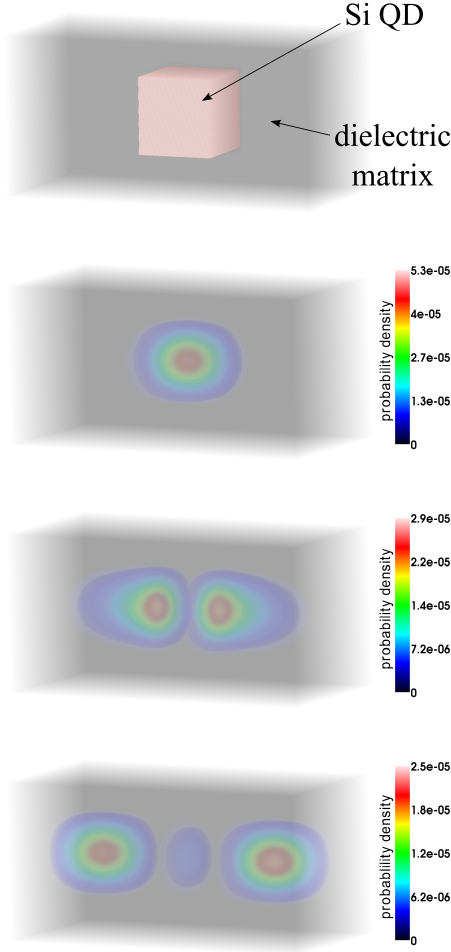


Fig. 1. Orbitals of the first three electronic states of a cubic silicon quantum dot embedded laterally in SiO<sub>2</sub> and vertically in SiC as computed within EMA.

The density of states and the charge carrier density are obtained from the solution of the above equations and the QD eigenfunctions via

$$\begin{aligned} \mathcal{D}(\mathbf{r}, E) &= \sum_{i,j} \sum_{m,n} A_{im,jn}(E) \psi_{im}^*(\mathbf{r}) \psi_{jn}(\mathbf{r}), \\ n(\mathbf{r}) &= \int \frac{dE}{2\pi} \sum_{i,j} \sum_{m,n} [-iG_{im,jn}^<(E)] \psi_{im}^*(\mathbf{r}) \psi_{jn}(\mathbf{r}), \end{aligned} \quad (2)$$

$$(3)$$

where  $\mathbf{A} \equiv i(\mathbf{G}^R - \mathbf{G}^A)$  is the charge carrier spectral function. In the same way, the local current density is obtained as [8]

$$\begin{aligned} \mathbf{j}(\mathbf{r}) &= -\frac{\hbar}{2m_0} \sum_{i,j} \sum_{m,n} \int \frac{dE}{2\pi} [-iG_{im,jn}^<(E)] \\ &\times \lim_{\mathbf{r}' \rightarrow \mathbf{r}} [\psi_{im}(\mathbf{r}) \nabla \psi_{jn}^*(\mathbf{r}') - \psi_{jn}^*(\mathbf{r}') \nabla \psi_{im}(\mathbf{r})]. \end{aligned} \quad (4)$$

### III. RESULTS

Fig. 2 shows the spatial evolution pattern of electron and hole photocurrent contributions of a 20 quantum dot superlat-

tice, resolved at the quantum dot positions. The overall current is perfectly conserved. In the present case, the superlattice is intrinsic, and charge separation is enabled by introduction of carrier selective contacts, i.e. the closure of minority carrier contacts via adjustment of the respective contact self-energy terms.

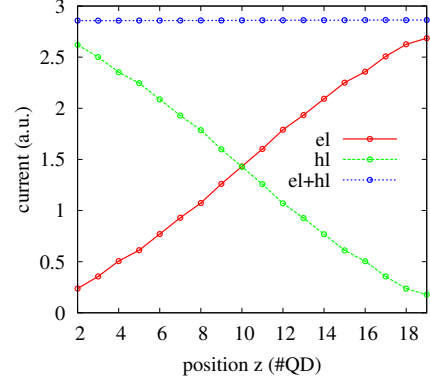


Fig. 2. Spatial evolution of electron and hole photocurrent contributions resolved at the quantum dot locations. The overall current is perfectly conserved.

### IV. CONCLUSION

The QDTB-NEGF approach extends the applicability of powerful quantum-kinetic methods to extended optoelectronic devices based on quantum dots in the low to intermediate coupling regime, and is thus able to provide insight into the microscopic processes underlying their operating mechanisms.

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