Steady-state quantum-kinetic theory of nanostructure-based photovoltaic devices

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INTRODUCTION

Many of the concepts for novel photovoltaic devices (intermediate band, multi-exciton-generation, multi-junctions, etc.) require an advanced control and manipulation of the optoelectronic properties of the latter, resulting in the prominent role of nanostructures - such as quantum wells, wires and dots - in the implementation of these concepts (Fig.1). However, the quantum effects governing the optoelectronic characteristics of the nanostructures, e.g., confinement and tunneling, not only provide the desired design degrees of freedom, but also request new models beyond the conventional macroscopic theories of generation, transport and recombination. Therefore, a quantum kinetic theory of photovoltaic devices is devised, which treats quantum optics and dissipative quantum transport on equal footing.

THEORETICAL FRAMEWORK

The theory is based on the non-equilibrium Green's function formalism for the relevant electronic, optical and vibrational degrees of freedom. The key element in the modification of the NEGF approach to quantum optoelectronics for the simulation of novel solar cell devices based on low-dimensional semiconductor nanostructures consists in an adequate description of photogeneration involving electronic states of arbitrary degree of localization, the coupling to extended states via tunneling or scattering processes, as well as transport of charge to and extraction at carrier selective contacts (Fig.2), in terms of corresponding self-energies renormalizing the charge carrier non-equilibrium Green's functions. The intraband quantum transport aspect of the theory is identical to that encountered in unipolar devices and is thus well covered by the literature [1]. The latter also applies to the pure interband quantum kinetics in optoelectronic devices such as quantum well lasers [2]. Only recently, the formalism is being used to describe the combination of both processes for the computation of radiative generation and recombination currents in photodetectors [3], [4] or light emitting diodes [5]. The extension to the photovoltaic regime of device operation, first discussed in [6] and reviewed in [11], represents a particular challenge, since the figure of merit results from the competition of generation and recombination processes at sizable separation of electron and hole contact Fermi levels.

APPLICATIONS

Applications of the approach to study relevant processes in two different prototypical quantum photovoltaic devices are discussed: photo-carrier escape from single direct gap semiconductor quantum wells in a photodiode ([7], [8], Fig. 3), and photocurrent flow in indirect semiconductor quantum well superlattice absorbers ([9], [10], Figs. 4,5). In both cases, the spectrally resolved information provides unique insight into the mechanisms of generation and transport.

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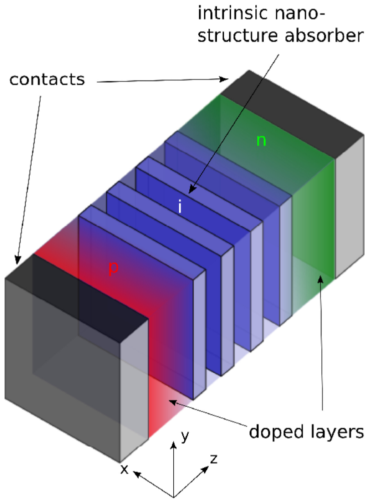

Fig. 1. Basic layout and functional elements of a quantum photovoltaic device.

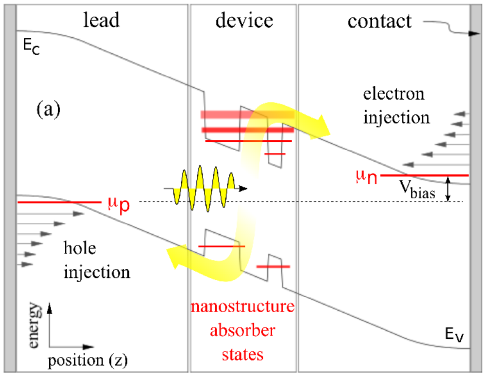

Fig. 2. Energy band diagram and optoelectronic processes in a device corresponding to a structure similar to that shown in Fig. 1.

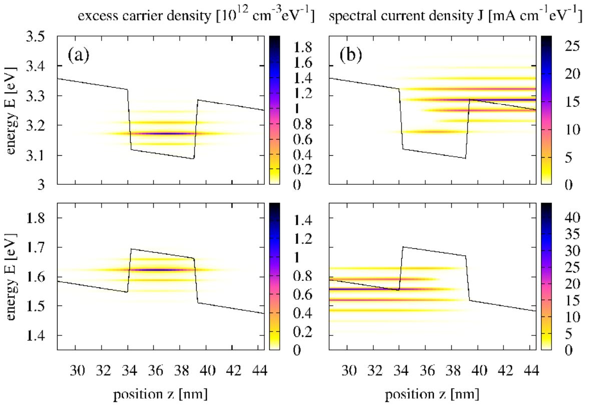

Fig. 3. Spectral properties of charge carriers in a single QW AlGaAs-photodiode under bias (1.3 V) and illumination (1.55 eV, 17.7 kW/m^2): (a) spectral excess carrier density with phonon satellites and (b) resulting (net) current spectrum.

$$\begin{tikzcd}
\Sigma^{ep,npo}_{\Gamma_v} \arrow[r, bend right] & G_{\Gamma_v} \arrow[l, bend right] \arrow[d, bend right] & \Sigma^{e\gamma}_{\Gamma_v} \arrow[l] & & \\
& \Sigma^{e\gamma}_{\Gamma_c} \arrow[r] & G_{\Gamma_c} \arrow[u, bend right] \arrow[d, bend right] & \Sigma^{ep,\Gamma-X}_{\Gamma_c} \arrow[l] & \\
& & \Sigma^{ep,\Gamma-X}_{X_c} \arrow[r] & G_{X_c} \arrow[u, bend right] \arrow[r, bend right] & \Sigma^{ep,X-X}_{X_c} \arrow[l, bend right]
\end{tikzcd}$$

Fig. 4. Self-consistent computation of Green's functions and scattering self-energies in silicon enabling the description of phonon-assisted indirect optical transitions between Γ_v (valence band) and X_c (conduction band) states via a virtual intermediate Γ_c state. [10]. A similar procedure can be used to compute non-radiative recombination via midgap defect states.

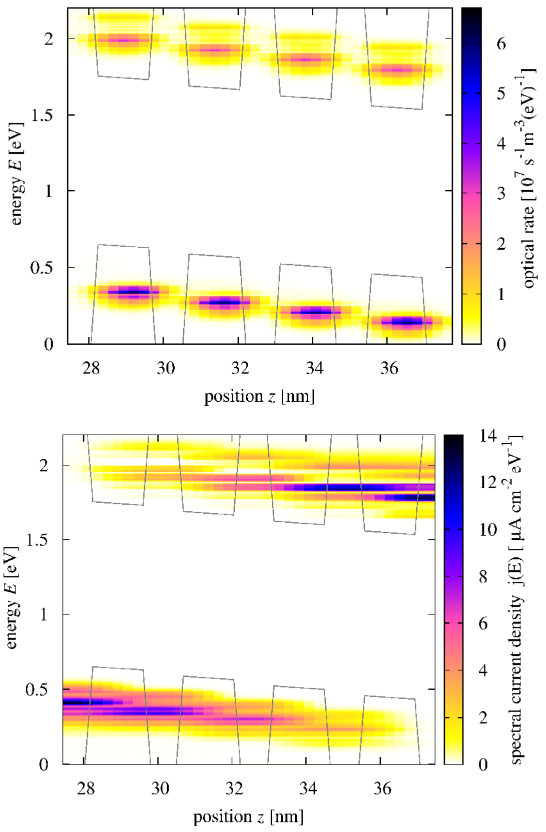

Fig. 5. Spatially and energy resolved charge carrier photogeneration rate and short-circuit photocurrent density in a Si-SiO$_x$ superlattice absorber, where transport is restricted to superlattice states, under monochromatic illumination with energy $E_\gamma = 1.65$ eV and intensity $I_\gamma = 10$ kW/m^2 [9].