Quantum kinetic theory of photovoltaic devices with low dimensional absorbers

U. Aeberhard
IEF-5: Photovoltaik, Forschungszentrum Jülich, 52425 Jülich, Germany
u.aeberhard@fz-juelich.de

Many third generation photovoltaic concepts require an advanced control and manipulation of the optoelectronic properties of the active device structure, leading to a prominent role of low dimensional absorbers such as quantum wells, wires and dots in the implementation of these concepts. However, the quantum effects governing the optoelectronic characteristics of the nanostructures not only provide the desired design degrees of freedom, but also request new models for the description of the photovoltaic operation, since conventional classical macroscopic theories of generation, transport and recombination do not allow for a consistent consideration of such effects and the related device behaviour.

One of the common features of quantum photovoltaic devices is the dominant contribution from the strongly localized states of the low dimensional absorbers to generation and recombination, while transport is mediated mainly via extended states. This means that one either has to find the ideal degree of localization which provides the best compromise between efficient transport and strong absorption, as indicated e.g. in the case of the quantum dot superlattices used in tandems, where miniband transport is required, or to optimize the processes that couple maximally localized absorbing states with maximally extended current carrying states, which corresponds to the situation encountered in quantum well solar cells.

The conventional approach to the problem described above is to combine a microscopic model for the physical processes involving confined states with a macroscopic, semiclassical theory for charge transport via the use of detailed balance rates determined within the microscopic theory. The resulting hybrid approach often provides effective fitting models able to quantitatively reproduce experimental device characteristics. However, at the same time, the large number of required ad-hoc assumptions tends to obscure the underlying mechanisms of quantum photovoltaic device operation, especially concerning the crucial processes of carrier escape and capture which couple localized and extended states.

To go beyond the existing approaches in capturing the essential physics of quantum photovoltaic devices, a microscopic theory based on the non-equilibrium Green's function formalism for the electronic, optical and vibrational degrees of freedom was developed and applied to the simulation of quantum well solar cells[1,2]. The theory is capable to describe both optical and transport properties including quantum effects such as confinement and tunneling in an open non-equilibrium system under consideration of elastic and inelastic scattering effects leading to incoherence and relaxation.

Here, the approach is applied to different problems encountered in quantum photovoltaic devices, namely the generation, escape and capture of carriers in quantum wells, incoherent transport in a Si-SiO_x superlattice and the photovoltaic properties of single quantum dot devices.

Quantum well solar cells have demonstrated their ability to outperform single-junction bulk devices under concentration, where the efficiency of these devices starts to be radiatively limited. Despite the large number of experiments on MQW structures for photovoltaic and laser applications, there are still some remaining open questions concerning the operating mechanisms of these devices, such as the questions related to carrier escape and capture close to the operating point in the photovoltaic regime, and the impact of quantum well coupling and geometry on the device performance. Here, we use our microscopic theory to investigate on the one hand the scenario of phonon mediated carrier escape by analyzing the photocurrent spectrum at different photon energies (Fig. 1) and on the other hand the influence of QW coupling on the device characteristics.

In devices relying on transport in minibands formed by the coupling of quantum wells or quantum dots, several issues arise when considering realistic situations, such as disorder in the spatial correlation of the potential or strong field effects, which both affect the transport properties in the sense that they tend to break up the minibands and to localize the wavefunction of the carriers. We analyze this behaviour by the determination of the density of states and the current spectrum in regular and disordered superlattices of finite extent and with strong built-in fields (Fig. 2).

The potential applications of quantum dots in photovoltaic devices are numerous. As compared to quantum well devices, additional challenges are encountered in these devices, such as the three dimensional confinement for both electrons and and phonons, which renders the calculation of the corresponding single particle spectra much more cumbersome. In many cases, the dots are embedded in amorphous matrix materials with strongly reduced spatial correlations and where the large number of defects has significant impact on both transport and recombination. Furthermore, the dots themselves usually exhibit a certain amount of disorder in both size and spacing. In order to include the additional degrees of freedom into the model, it is useful to simplify the transport problem by reducing it to an effective coupling of the absorber to respective majority carrier contacts. By this way, a minimal quantum photovoltaic model is obtained as illustrated in Fig. 3, where injection and extraction of charge under bias and illumination can be described in a framework of far from equilibrium transport that is at the same time consistent with quantum optics. Here, the behavior of a single quantum dot device under both bias and illumination is analyzed under consideration of the degree of coupling to contacts.

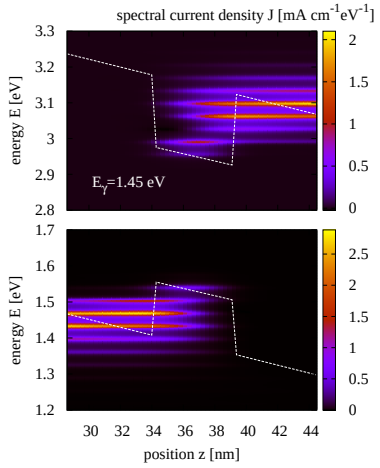

FIG. 1: Current spectrum at photon energy $E_\gamma = 1.45$ eV, bias voltage $V_{bias} = 1$ V and illumination intensity $I_\gamma = 17.7$ kW/m^2.

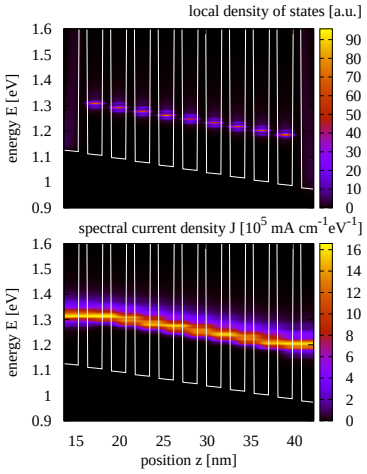

FIG. 2: Local density of states and dark current spectrum in a regular SL segment formed by 10 Si QW and SiO_x barriers with assumed height of 1.5 eV and embedded in the i-region of a nin-diode, under a bias of 0.2 V.

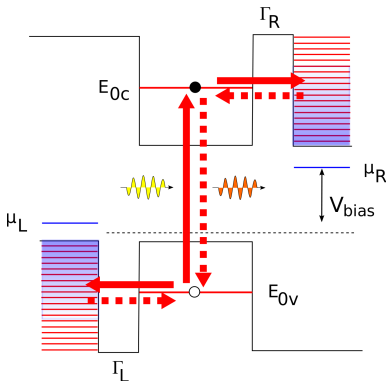

FIG. 3: Minimal model of a quantum photovoltaic device including a low dimensional absorber with discrete density of states and injection as well as extraction of charge via coupling to majority carrier contacts.

[1] U. Aeberhard and R. Morf, Phys. Rev. B **77**, 125343 (2008).

[2] U. Aeberhard, *A Microscopic Theory of Quantum Well Photovoltaics*, PhD thesis, ETH Zuerich, 2008.