Microscopic approach to reciprocity and photon recycling in ultrathin solar cells

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***Abstract*—In contrast to their bulk counterparts, ultrathin solar cells exhibit bias-dependent optical properties. This has severe implications for the validity of conventional opto-electronic reciprocity relations. We review the predictions of this phenomenon from quantum-kinetic theory and discuss the experimental confirmation, as well as the extension of the theoretical framework for the rigorous assessment of photon recycling effects in the electrical characteristics of ultrathin-film solar cell devices.**

I. Introduction

While photovoltaic solar energy conversion has established itself as one of the main components for the energy supply of sustainable societies, the accelerated transformation of the energy sector in response to climate change demands an ever increasing utilization of the available solar resource. Conventional silicon photovoltaics is thereby approaching its theoretical efficiency limit, and alternative approaches are under investigation, such as multijunction architectures based on high-efficiency silicon bottom cells. For the top cells, high-efficiency thin films solar cells need to be considered, such as those based on III-V semiconductors [1], but also the more recent metal-halide perovskites [2]. These materials are highly absorbing, with steep absorption edges, and have been shown to operate close to the radiative limit [3], which together promotes the appearance of photon recycling (PR) effects [4], such as enhancement of open circuit voltage (V_{OC}) [5].

While for tandem applications, the subcell thickness can be kept in the sub-micron range, even thinner architectures have been proposed for light-weight, flexible and low-cost single junction applications, in conjunction with intricate light-trapping schemes [6]. In these ultra-thin solar cells, absorption and emission of light depend on the applied bias voltage via the built-in field, as was predicted theoretically [7], and recently confirmed experimentally [8]. This dependence has important implications for the proper application of the reciprocity relations [9] as frequently used for the analysis of solar cells by means of luminescence experiments. Furthermore, wave optical simulations indicate maximum V_{OC} enhancement due to PR at absorber thicknesses below 100 nm [4].

Here, we review the theoretical assessment of the optoelectronic reciprocity relations between absorption and emission in ultra-thin absorber solar cells as provided by simulations based on a non-equilibrium Green's function (NEGF) framework [7]. The simulation of electronic and optical excitations in thin semiconductor slabs is then analyzed with regard to the consideration of photon-recycling effects that arise naturally in such a coupled description. In contrast to predictions of V_{OC} enhancement from detailed balance [10] and optical modelling [11], we obtain ΔV_{OC} directly from the renormalized current-voltage characteristics.

II. Approach

The comprehensive NEGF framework for the microscopic description of optoelectronic processes in nanostructure-based solar cells is presented in recent reviews (see, e.g. , Ref. [12]). Details on the implementation of the formalism for consideration of photonic modes for the photogeneration in thin film absorbers can be found in [13], while application to the assessment of reciprocity in ultrathin absorber photovoltaics is shown in [7].

The core of the formalism is the coupled solution of the equations for Green's functions (G, $\mathcal{D}_\gamma$) and self-energies (Σ, Π_γ) for charge carriers and photons, respectively, as depicted in Fig. 1. The formalism considers electronic and optical modes in open systems under electronic and optical excitation (bias voltage and illumination) and provides electronic (scattering) rates and currents as well as optical rates (absorption, emission) and photon fluxes, which are evaluated consistently and on equal footing as derived directly from the NEGF.

The relation between absorptance and emitted photon flux required for the assessment of opto-electronic reciprocity relations are obtained from the electron-hole polarization function defined as a convolution of electron and hole Green's functions and which forms the main component of the photon self-energy Π_γ. The latter is used in the photonic Keldysh equation for the renormalized photon propagator, which provides the

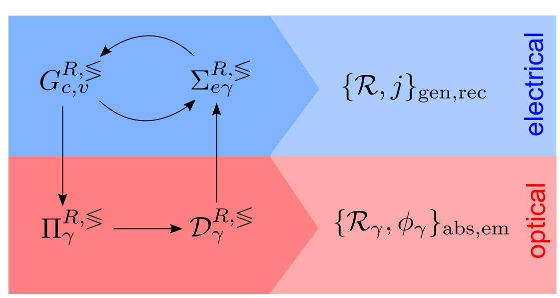

Fig. 1. Schematic representation of the self-consistent opto-electronic NEGF formalism used to assess reciprocity and photon recycling. The coupling between electronic and optical degrees of freedom proceeds via the electron-hole polarization function and the electron-photon self-energy.

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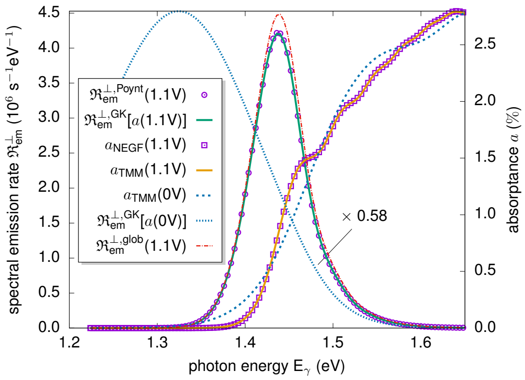

Fig. 2. Absorptance and front-side emission of a 100 nm GaAs p-i-n solar cell with gold rear reflector, reflecting the impacts of built-in field variation and reabsorption [7].

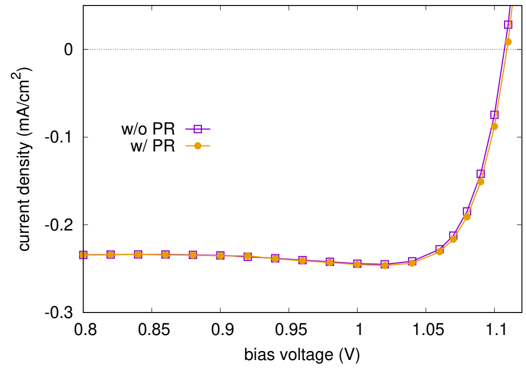

Fig. 3. Current-voltage characteristics of a 50 nm GaAs p-i-n solar cell, with PR (full symbols) and without PR (empty symbols). The additional generation from reabsorption results in a V_{OC} enhancement of about 2 mV. The peculiar shape of the curve around MPP is due to the bias dependence of the photocurrent.

photon flux in terms of the Poynting vector of the quantized fields. Consideration of the V_{OC} enhancement due to photon recycling, on the other hand, requires implementation of the full computational cycle shown in Fig. 1. The central element is an additional electron-photon self-energy component that couples the electronic NEGFs (G) to the NEGF of the internally emitted photons ($\mathcal{D}_\gamma$).

III. Results

Figure 2 shows the absorption and emission characteristics of a planar 100 nm GaAs p-i-n solar cell with gold rear reflector. The spectral emission rate is reproduced by the generalized Kirchhoff law, if the absorptance at the same bias voltage is used, while the reciprocity based on the EQE at zero bias deviates strongly from the correct result. The absorptance from NEGF (corresponding to the EQE in the case of unit collection efficiency) coincides with the same quantity as obtained from the transfer matrix method (TMM). Comparison of the spectral emission rate from the Poynting vector with that from the global emission rate - obtained via spatial integration of the local rate - reveals the effect of reabsorption.

The ability to assess the impact of photon recycling on the electrical characteristics of thin-film solar cells is demonstrated in Fig. 3, which shows the current-voltage characteristics of a 50 nm GaAs p-i-n solar cell with gold back reflector, under monochromatic illumination with $E_\gamma = 1.44$ eV and $I_\gamma = 0.1$ kW/cm^2, with PR (full symbols) and w/o PR (empty symbols). To speed up the computations, the diagonal approximation of the self-energies was used, which underestimates the optical rates by about one order of magnitude. Still, a V_{OC} enhancement of about 2 mV can be inferred from the characteristics.

IV. Conclusions

We demonstrate the rigorous assessment of optoelectronic reciprocity and photon recycling in the NEGF simulation framework for ultrathin solar cells, which enables evaluation of V_{OC} enhancement directly from the electrical characteristics, i.e., under consideration of both, optical cavity effects and the full transport problem including contact layers.

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