Photon Green's functions for a consistent theory of absorption and emission in nanostructure-based solar cell devices

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***Abstract*—The light-matter interaction in planar nanostructures with applications in photovoltaic devices is investigated by means of a microscopic quantum-kinetic theory based on the non-equilibrium Green's function formalism. The Dyson and Keldysh equations for the Green's functions of photons are solved numerically. The result is used to couple the optical and electronic degrees of freedom via respective self-energies. The numerical approach for the solution of the optical problem is verified against a standard transfer-matrix formalism and applied to the fluorescent emission of colloidal quantum dots in microresonator cavities and the generation of dark- and photocurrent in ultra-thin-absorber solar cells.**

I. Introduction

Many novel solar cell architectures aim at an increase of the light absorption via a specific tayloring of the optical modes of the absorber [1]. At the same time, microresonator cavities can be used to enhance the optical rates of luminescent and/or up-converter materials for spectral photon management in third generation photovoltaic devices [2]. The treatment of the optical problem of slab systems within the non-equilibrium Green's function formalism (NEGF) has been discussed in the literature for several different applications, such as emission enhancement in microcavity lasers [3], absorption-reflection characteristics of a general non-equilibrium system [4] or the photovoltaic response of a spatially homogeneous (bulk) absorber in absence of recombination losses [5]. Here, the approach is reformulated in a representation suitable for its combination with the NEGF treatment of charge carrier photogeneration, transport and recombination in semiconductor nanostructures and bipolar thin-film devices as previously developed for nanostructure-based solar cells [6].

II. Approach

In a layer structure with homogeneous transverse ($\|$) dimensions , the equations for the Green's functions (GF) can be simplified by using the Fourier transform of the latter with respect to transverse coordinates, i.e.,

$$\mathcal{D}_{\mu\nu}(\mathbf{r},\mathbf{r}',E)=\frac{\mathcal{A}}{(2\pi)^2}\int d^2q_\parallel \mathcal{D}_{\mu\nu}(\mathbf{q}_\parallel,z,z',E)e^{i\mathbf{q}_\parallel\cdot(\mathbf{r}_\parallel-\mathbf{r}'_\parallel)}. \tag{1}$$

For each energy and transverse momentum vector, a separate set of equations for the GF needs to be solved. In the case of the retarded GF, the integral form of the Dyson equation for the dyadic form reads (omitting the energy and momentum arguments and assuming Einstein's convention for summation over repeated indices)

$$\begin{aligned}\mathcal{D}^R_{\mu\nu}(z,z') =&\mathcal{D}^R_{0\mu\nu}(z,z')+\int dz_1\int dz_2\mathcal{D}^R_{0\mu\alpha}(z,z_1)\\ &\times\Pi^R_{\alpha\beta}(z_1,z_2)\mathcal{D}^R_{\beta\nu}(z_2,z').\end{aligned} \tag{2}$$

Similarly, the kinetic equation for the correlation functions becomes

$$\begin{aligned}\mathcal{D}^{\lessgtr}_{\mu\nu}(z,z')=\int dz_1\int dz_2\mathcal{D}^R_{\mu\alpha}(z,z_1)\Big[\Pi^{\lessgtr}_{0\alpha\beta}(z_1,z_2)\\ +\Pi^{\lessgtr}_{\alpha\beta}(z_1,z_2)\Big]\mathcal{D}^A_{\beta\nu}(z_2,z'),\end{aligned} \tag{3}$$

where the self-energy component $\Pi^{\lessgtr}_0$ related to the solution of the homogeneous problem is given in terms of the GF $\mathcal{D}_0$ of the unperturbed system.

In the case of a one-dimensional dielectric perturbation potential (i.e., a 1D photonic crystal), the unperturbed GF $\mathcal{D}_0$ can be defined on the basis of solutions for homogeneous free space, which for unpolarized light are given by

$$\mathcal{D}_{0\mu\nu}(\mathbf{q},E)=\frac{\hbar^2c_0^2}{2V\hbar\omega_\mathbf{q}}\delta^\parallel_{\mu\nu}(\mathbf{q})D_0(\mathbf{q},E), \tag{4}$$

where D_0 is the scalar GF of non-interacting bosons and $\delta_\parallel(\mathbf{q})$ is the transverse delta function in reciprocal space. The corresponding GF in slab representation are then obtained from the explicit forms for the GF components of non-interacting bosons in homogeneous systems (bulk) via inverse Fourier transform with respect to q_z. Due to the $\mathbf{q}$-dependence of $\delta^\parallel$, the free GF can be separated in isotropic and anisotropic contributions,

$$\mathcal{D}_{0\mu\nu}=\mathcal{D}^{(1)}_0\delta_{\mu\nu}+\mathcal{D}^{(2)}_{0\mu\nu}. \tag{5}$$

Further evaluation of the anisotropic term requires consideration of the polarization components and the directional

dependence of the mode occupation. For normal incidence ($\mathbf{q}_\parallel = 0$), the anisotropic part vanishes in $\mathcal{D}_0$. For a simple 1D dielectric potential, the retarded photon self-energy reduces to the diagonal term $\Pi^R_{\mu\nu}(\mathbf{q}_\parallel = 0, z, z', E) = V(z)\delta_{\mu\nu}\delta(z - z')$. The corresponding integral equation for the scalar component of the retarded photon is solved via numerical quadrature [7].

III. Results

The general expression for the local density of photon states in terms of the photon GF reads

$$\mathcal{N}(z, E) = -\frac{\mathcal{C}}{\pi}\sum_\mu \sum_{\mathbf{q}_\parallel} \mathrm{Im}\mathcal{D}^R_{\mu\mu}(\mathbf{q}_\parallel, z, z, E), \qquad (6)$$

where the normalization constant $\mathcal{C} = \frac{2E}{(\hbar c_0)^2}$ relates the bare boson field to the quantized photon field. As a first consistency check, it is verified that in the case of free field modes, the spatially constant result $\mathcal{N}_0(z, E) = (n_0^3 E^2)/(\pi^2\hbar^3 c_0^3)$ is obtained.

To verify the computation of the GF for an inhomogeneous situation, the local DOS and spectral density of photons for a 500 nm thick dielectric slab ($n = 3$) in air ($n_0 = 1$) are compared to the corresponding quantities as computed via a standard transfer-matrix method (TMM). For the LDOS, the NEGF expression (6) is compared to the sum of the absolute value squared of the electric field for left and right incidence in TMM. For the photon density, the quantity

$$n_\gamma(\mathbf{q}_\parallel, z, E) = \frac{\mathcal{C}}{2\pi}\sum_\mu i\mathcal{D}^<_{\mu\mu}(\mathbf{q}_\parallel, z, z, E) \qquad (7)$$

with the correlation function resulting from the solution of (3) under the assumption of an asymmetric mode occupation $N_\mathbf{q} = \tilde{N}\delta(q_\parallel)\theta(q_z)$ and vanishing polarization $\Pi^<$ is compared to the absolute value of the electric field for left incidence in TMM. The results are diplayed in Fig. 1 for the full calculation domain of the GF.

In terms of the photon GF, the fully non-local electron-photon scattering self-energies required for the description of photogeneration and radiative recombination processes at the nanoscale reads

$$\Sigma^{\lessgtr}(\mathbf{k}_\parallel, z, z', E) \approx i\hbar\mu_0\big(\frac{e}{m_0}\big)^2\frac{\mathcal{A}}{(2\pi)^2}\int d^2q_\parallel \sum_{\mu\nu} p_\mu(z)p_\nu(z')$$

$$\times \int \frac{dE'}{2\pi\hbar} G^{\lessgtr}(\mathbf{k}_\parallel, z, z', E - E')\mathcal{D}^{\lessgtr}_{\nu\mu}(\mathbf{q}_\parallel, z, z', E'), \qquad (8)$$

where the $\mathbf{p}$ denote the momentum matrix elements and G the charge carrier GF in a real space band basis. Using this method in combination with the NEGF theory for fluorescent emission of quantum dots developed in [8], the total emission rate of a quantum dot located at a position z_0 in a planar resonant dielectric cavity can be obtained from

$$R_{em}(z_0) = \frac{\mathcal{A}}{(2\pi)^2}\int d^2q_\parallel \sum_{\mu\nu}\int \frac{dE}{2\pi\hbar}\Pi^<_{\mu\nu}(E)\mathcal{D}^>_{\nu\mu}(\mathbf{q}_\parallel, z_0, z_0, E) \qquad (9)$$

where the polarization $\Pi^<$ reflects the optical excitation of the quantum dot.

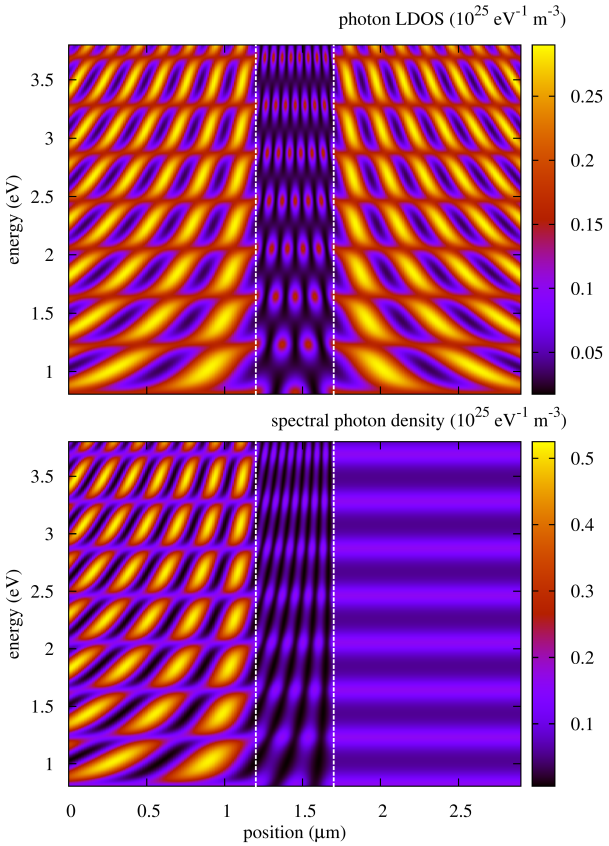

Fig. 1. LDOS and spectral photon density as computed from the photon GF for a dielectric layer ($n = 3$) in air and normal light incidence from the left.

IV. Conclusions

We presented a method for the computation of photon Green's functions suitable for integration in a comprehensive quantum-kinetic theory of absorption and emission processes in layer-based nanostructures with application in novel solar cell devices.

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