Quantum-optical radiation laws for confined semiconductors systems

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Abstract. We present a quantum-kinetically exact theoretical framework for the propagation, emission and scattering of light in bounded media in the context of semiconductor optics. The theory is based on the nonequilibrium photon Green’s functions. Its advantage is that the spatial inhomogeneity inherent to bounded media and, hence, to many semiconductor optics problems, is fully and exactly considered. The electromagnetic properties of media are treated microscopically rather than in an effective approximation, and media may be arbitrarily dispersive and absorptive. Relations for the propagation of quantized (squeezed) light as well as for the energy transport of incident and emitted light are given and discussed.

In the description of light propagation through media, the electromagnetic effects of the medium are often accounted for by the dielectric function in the form \( \varepsilon(\omega) = 1 + \chi(\omega) \), which describes frequency-dispersion. This form implies that the medium is in a steady state, and that the response of the medium is constant over its entire spatial extension.

However, the induced dipoles that give rise to the polarization field (e.g., excitons) are not point-like but have a finite spatial extension. Also, it is obvious that the polarization may change if the medium boundaries are approached (e.g., excitonic decay at surfaces, surface effects). Thus, the electromagnetic properties of media are inherently non-local. In order to account for spatial non-locality (or inhomogeneity) as well as for temporal inhomogeneity (for nonequilibrium cases), the constitutive relation for the polarization field \( \mathbf{P} \) is best established in the general form [1; 2]

\[
\mathbf{P}(\mathbf{r}, t) = \varepsilon_0 \int \! d\mathbf{r}'dt' \chi(\mathbf{r}, \mathbf{r}', t, t') \mathbf{E}(\mathbf{r}', t'),
\]

in which the linear response of the medium to the exciting field is described by the non-local susceptibility \( \chi \).

The spatial inhomogeneity gives rise to spatial dispersion effects [2], which will be shown to have a strong impact on, e.g., semiconductor transmission spectra, but it is often neglected since it is a major obstacle for the theoretical description. The approaches presented in Refs. [3–5] are able to fully consider microscopical non-local susceptibilities with the help of the nonequilibrium photon Green’s functions [6; 7]. The medium properties do not need to be specified and no further approximations are made, so the results are universal in this respect. The description of incident quantized light is possible, and useful results for the quantum optics of passive optical devices can be drawn [8].
1. The nonequilibrium photon Green’s function

The nonequilibrium photon Green’s function (PGF) $D$ [6; 7] can be defined as an expectation value expression for the electromagnetic vector potential operator $\hat{A}$ [with 1 denoting $(r_1, t_1)$],

$$D^>(1, 2) = \frac{1}{i\hbar\mu_0} \left( \langle \hat{A}(1)\hat{A}(2) \rangle - \langle \hat{A}(1) \rangle \langle \hat{A}(2) \rangle \right)$$  \hspace{1cm} (2)

which obeys Maxwell’s equation in terms of induced (bound) and external (free) currents $j_{\text{ind}}$, $j_{\text{ext}}$.

The so-called “greater” PGF $D^>$ obviously describes fluctuations of the vector potential. It is linked to the “lesser” PGF $D^<$ simply by $D^>(1, 2) = D^<(2, 1)$, and several other identities exist, such as the one defining the “retarded” PGF, which governs the (classical) wave propagation in the role of a mathematical Green function:

$$A(1) = \langle \hat{A}(1) \rangle = -\mu_0 D^{\text{ret}}(1, 2) j^{\text{ext}}(2).$$  \hspace{1cm} (3)

It shall be mentioned that these PGFs are valid for nonequilibrium cases and that they can be formally combined in a single PGF $D(1, 2)$ on the so-called Keldysh time contour.

In the framework of the PGFs, the non-local electromagnetic properties of the medium appear in the photon self-energy (polarization function)

$$P^{\text{ret}}(1, 2) = -\frac{1}{\varepsilon} \frac{\partial^2 \chi(r_1, r_2, t_1, t_2)}{\partial t_1 \partial t_2},$$  \hspace{1cm} (4)

which is linked to the induced current and to the polarization field as $j_{\text{ind}} = \hat{P} = -\frac{1}{\mu_0} P^{\text{ret}} A$, and determines the behavior of the PGF via the Dyson equation

$$(D^{-1}_0(1, 2) - P^{\text{ret}}(1, 2)) D^{\text{ret}}(2, 3) = \delta(1, 3).$$  \hspace{1cm} (5)

2. Representation of incident quantized light

The vector potential fluctuations represented by $D^\Xi$ split up into the medium-induced contribution resulting from electronic processes in the medium and the vacuum-induced contribution from light incident from the free space [3]:

$$D^\Xi = D^\Xi_{\text{med}} + D^\Xi_{\text{vac}}, \quad D^\Xi_{\text{med}} = D^{\text{ret}} P^{\text{med}} D^{\text{adv}}, \quad D^\Xi_{\text{vac}} = \varepsilon_T^{-1, \text{ret}} D^\Xi_0 \varepsilon_T^{-1, \text{adv}}$$  \hspace{1cm} (6)

This is a fully universal and very useful property of the PGF. It allows to discriminate light by its source, be it internal ($D^\Xi_{\text{med}}$) or external ($D^\Xi_{\text{vac}}$, under external control via $j_{\text{ext}}$). The incident light fluctuations exhibit an interesting structure: They consist of solutions of the pure vacuum Maxwell equations, $D^\Xi_0$, which are renormalized globally by the inverse of the dielectric function tensor $\varepsilon_T^{-1}$.

As a retarded PGF, the latter ensures that the propagation of these fluctuations can always be traced back to that of classical light waves, regardless of the state of incoming light and of specific optical properties or geometrical shapes of the matter.

To analyze $D^\Xi_{\text{vac}}$ further, one may apply a normal-mode expansion for the vector potential in $D^\Xi_0$ according to Eq. (2) [3; 8]. Then, expectation values of photon operator products have to be evaluated in a given quantum state, e.g., a squeezed state: $\langle \hat{a}^+ \hat{a} \rangle = \langle \Phi_s | \hat{a}^+ \hat{a} | \Phi_s \rangle = \langle \Phi_0 | \hat{S}^+ \hat{a}^+ \hat{a} \hat{S} | \Phi_0 \rangle$ [9]. Then, photon operator commutation reveals a general structure of the pure vacuum PGF [8], namely its decomposition into the ubiquitous ground-state fluctuations of the electromagnetic field and the contribution caused by external stimulation (light source). The latter contribution is equal for both $D^\Xi_0$ and $D^\Xi_0$:

$$D^\Xi_0 = D^\Xi_{\text{sp}} + D_{0, \text{stim}} \quad D^\Xi_{\text{sp}} = \sum_{\lambda q} \frac{c}{2Vq} F(1) \otimes F^*(2),$$  \hspace{1cm} (7)
where $\mathbf{F} = e_{\lambda q} e^{i \mathbf{q} \cdot \mathbf{r} - i \omega t}$ is a free plane wave for wave vector $\mathbf{q}$ and polarization $\lambda$.

While the form of $D_{0,sp}^x$ is fixed, that of $D_{0,stim}$ depends on the preparation of the incident light. For example, a Fock (photon number) state with photon population $n_F(\omega = cq)$ results in

$$D_{0,stim}^x(1,2) = \sum_{\lambda q} \frac{c}{i V_q} n_F^{\lambda q} \text{Re} \left[ \mathbf{F}_{\lambda q}(1) \otimes \mathbf{F}^\ast_{\lambda q}(2) \right], \quad (8)$$

and a squeezed vacuum state $|sv\rangle = |\xi\rangle |0\rangle$ with squeezing strength $\xi e^{i \phi}$ yields

$$D_{0,stim}^x(1,2) = \sum_{\lambda q} \frac{c}{2i V_q} \left\{ \sinh^2 \xi F(1) \otimes F^\ast(2) - \cosh \xi \sinh \xi e^{i \phi} F(1) \otimes F(2) + \text{c.c.} \right\}. \quad (9)$$

The renormalization of these pure vacuum fields to effective waves $A_{\lambda q}$ propagating in the bounded media system is formally trivial thanks to the simple structure of Eq. (6) [3]:

$$A_{\lambda q}(1) = \xi^{-1,ret}_{T}(1,2) F_{\lambda q}(2) \quad \Rightarrow \quad D_{vac}^x = D_{0}^x [\mathbf{F} \rightarrow \mathbf{A}] \quad (10)$$

3. Energy transport

The energy flux vector (Poynting vector) $\mathbf{S} = \frac{1}{i \mu_0} \langle \mathbf{E} \times \mathbf{B} \rangle \big|_{\text{sym}}$ [1; 7] can also be expressed by PGFs. The splitting properties (6),(7) then translate directly to $\mathbf{S}$:

$$\mathbf{S} = \mathbf{S}_{sp} + \mathbf{S}_{stim} + \mathbf{S}_{med} \quad (11)$$

The quantity $\mathbf{S}_{stim}$ is the just the energy flux caused by an external light source. It is equivalent to the normally-ordered energy flux $\langle \hat{S} \rangle$ that is measured by photodetectors, e.g., in absorption-transmission experiments [8; 9].

With the theoretical methods presented above, e.g., the energy flow caused by a squeezed vacuum transmitted through a steady-state semiconductor slab can be readily deduced [3]:

$$s_{sv,q}^t = \sinh^2 \xi |t_{\lambda q}|^2 - \cosh \xi \sinh \xi |t_{\lambda q}|^2 \cos(2cqt - 2 \text{arg} t_{\lambda q} - \phi_{\lambda q} + \Delta \phi_L) \quad (12)$$

The transmitted energy flux is governed by the classical transmittivity factor $t_{\lambda q}$, even for nonclassical light. It exhibits a stationary term and a term which oscillates with double mode frequency $2cq$. This latter term can be exploited in homodyning experiments [9] in order to overcome the quantum noise limit. $\Delta \phi_L$ denotes the usual phase delay accumulated while passing through the slab with thickness $L$. These results confirm and generalize those of former quantum-optical approaches which had to neglect spatial dispersion and resorted to effective approximations for the susceptibility.

We may now link these results to those from Ref. [4], where a nonequilibrium Kirchhoff/Planck law was established: The spectrally and directionally resolved energy flux leaving the slab surfaces is given by

$$\Delta s = (b_{\text{stim}} - n_{\text{stim}}) a. \quad (13)$$

It is the balance of (1) absorption of incident light $s_a = n_{\text{stim}} a$, where $n_{\text{stim}}$ is the photon distribution of the incident light (excluding the spontaneous fluctuations), and (2) emission of light by the slab medium $s_e = b_{\text{stim}} a$, where $b_{\text{stim}}$ is the distribution of its optical medium excitation (e.g., polaritons). Interestingly, both are governed by the classical absorptivity function $a$ [4]. In the Fock case, $n_{\text{stim}}$ is equal to $n_F$ [Eq. (8)]. The distribution $b$ can be shown to be always bosonic despite of the fermionic character of the underlying particles. It is accessible to measurement as the ratio of incoherent emission and coherent absorption [10]. The contribution of ground-state fluctuations cancels out in the net energy flow.
4. Spatial dispersion in confined excitonic systems. Squeezed vacuum transmission.  
A simple approximation for the excitonic contribution to a semiconductor susceptibility can be obtained with the oscillator model. Often, no spatial effects are considered ($\chi \rightarrow \chi(\omega)$), but if the kinetic energy of the excitons is included ($\chi \rightarrow \chi(\omega, q)$), drastic effects (polariton resonances and beats) appear, e.g., in absorptivity spectra (Fig. 1). Far more accurate, however, is a microscopic, self-consistent calculation of field and polarization as demonstrated, e.g., by [11], yielding $\chi \rightarrow \chi(x,x',\omega)$. The present theory supports all levels of susceptibility approximations. For demonstration, the squeezed vacuum transmission spectrum according to Eq. (12) is shown in Fig. 1 for the spatially dispersive case.

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