Quantum theory of light scattering for arbitrary finite-size dielectric and conducting structures

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Abstract
We present a quantum theory of light scattering for the analysis of the quantum statistical and fluctuation properties of light scattered or emitted by micrometric and nanometric three-dimensional structures of arbitrary shape. This theory provides a general and unified basis for analysing a large class of optical processes where quantum and/or thermal fluctuations play a role.

Keywords: Quantum optics, thermal emission, quantum fluctuations

In this paper we present a quantum generalization of the classical theory of light scattering based on Green's dyadic technique. The theory presented here provides a general and unified basis for analysing a large class of optical processes where quantum and/or thermal fluctuations play a role. It is expected to be adequate to analyse a wide range of optical phenomena and experiments such as precision measurements of Casimir forces [1], light emission from sources embedded in photonic systems [2], light fluctuations in finite inverted-population media with inclusion of spatial effects [3], the spatial behaviour of scattered and/or confined nonclassical light [4, 5] and nano-scale radiative transfer [6, 7].

By using the method of Langevin forces, light has been quantized in media of increasing generality [8–15]. Here we generalize these results to media that can be anisotropic and/or with a nonlocal susceptibility [16]. Furthermore we consider explicitly media that can have finite size. This allows the analysis of quantized light scattering and allows us to derive general quantum optical input–output relations relating the output photon operators to the input photon operators and to the noise currents of the scattering system. These relations also hold for evanescent fields and thus allow us to define naturally output photon operators associated with evanescent waves.

Let us consider the most general nonmagnetic linear scattering system. It can be described by a causal and eventually nonlocal susceptibility tensor[1] $\chi_{i,j}(\boldsymbol{r}, \boldsymbol{r}', \omega)$ [17]. Thus we are considering a large class of material systems of arbitrary shape including anisotropic media and/or media driven by the electric field via a nonlocal susceptibility. We introduce the operators $\boldsymbol{L}$ for $-\nabla \times \nabla\times$ and $\boldsymbol{e}_0$ for $k^2\mathbf{1}$ and the integral operator $\boldsymbol{e}_s$, describing the effect of the scattering system. This operator applied to the electric field gives

$$(\boldsymbol{e}_s \boldsymbol{E})_i = k^2 \int \mathrm{d}^3 r' \, \chi_{ij}(\boldsymbol{r}, \boldsymbol{r}') E_j(\boldsymbol{r}').$$

Hence the wave equation relative to the material systems considered here, for the positive frequency components of the electric-field operator, can be written in compact notation as

$$(\boldsymbol{L} + \boldsymbol{e}_0 + \boldsymbol{e}_s)\hat{\boldsymbol{E}}^+ = \mathrm{i}\omega\mu_0 \hat{\boldsymbol{j}} \tag{1}$$

where the hat indicates quantum operators. The zero-mean noise currents $\hat{\boldsymbol{j}}$ can be derived from the Heisenberg–Langevin equations for the material system [17] and appear only if the susceptibility tensor is not real; they are a direct consequence of the fluctuation–dissipation theorem and obey the following commutation rules:

$$[\hat{j}_i(\boldsymbol{r}, \omega), \hat{j}_j(\boldsymbol{r}', \omega)] = 0 \tag{2}$$

$$[\hat{j}_i(\boldsymbol{r}, \omega), \hat{j}_j^\dagger(\boldsymbol{r}', \omega)] = \frac{\hbar}{\pi \mu_0} \frac{\omega^2}{c^2} |\chi^I_{ij}(\boldsymbol{r}, \boldsymbol{r}', \omega)| \delta(\omega - \omega') \tag{3}$$

χ^I being the imaginary part of the susceptibility tensor. These equations show that a nonlocal susceptibility produces noise currents that are spatially correlated. These spontaneous currents act as quantum Langevin forces. Their expectation values determine the amounts of noise that are added to optical signals that propagate through the attenuating or amplifying media. Moreover $\langle \hat{\boldsymbol{j}}^\dagger \hat{\boldsymbol{j}} \rangle$ is the source term producing light emission.

[1] Of course for systems or part of a system that can be described by a local susceptibility, $\chi_{ij}(\boldsymbol{r}, \boldsymbol{r}')$ reduces to $\chi_{ij}(\boldsymbol{r})\delta(\boldsymbol{r} - \boldsymbol{r}')$.

By applying the Green dyadic approach, the electric-field operator satisfying equation (1) can be expressed as

$$\hat{\boldsymbol{E}}^{+} = \hat{\boldsymbol{E}}_h^{+} + \hat{\boldsymbol{E}}_p^{+} \qquad (4)$$

with the particular solution $\hat{\boldsymbol{E}}_p^{+}$ given by

$$\hat{\boldsymbol{E}}_p^{+} = \mathrm{i}\omega\mu_0 \boldsymbol{G}\hat{\boldsymbol{j}} \qquad (5)$$

and the homogeneous solution

$$\hat{\boldsymbol{E}}_h^{+} = (\boldsymbol{1} - \boldsymbol{G}\boldsymbol{e}_s)\hat{\boldsymbol{E}}^{0+}. \qquad (6)$$

In equation (6) $\hat{\boldsymbol{E}}^{0+}$ is the electric-field operator in the absence of the scattering system; it describes the input light field and can be expanded in terms of photon operators according to the usual schemes describing light quantization in free space. The particular solution $\hat{\boldsymbol{E}}_p^{+}$ describes the light field emitted by the scattering system. The Green tensor $\boldsymbol{G}$ is the solution of the following equation:

$$(\boldsymbol{L} + \boldsymbol{e}_0 + \boldsymbol{e}_s)\boldsymbol{G} = \boldsymbol{1}$$

where $\boldsymbol{1} \equiv \delta_{ij}\delta(\boldsymbol{r} - \boldsymbol{r}')$. Equation (4) gives the electric-field operator in terms of the input photon operators and the noise current operators. Having fixed the quantum state of the input light beams and of the scattering system, by using equation (4) in principle it is possible to compute the electric-field operator in the presence of the scattering system in the whole space if the Green tensor $G_{ij}(\boldsymbol{r}, \boldsymbol{r}')$ is known.

Inside absorbing materials, owing to the presence of noise currents, it is not possible to define space-independent photon operators as in free space [9, 17]; however, we may attempt to find input and output photon operators outside the scattering system [16]. This would furnish useful input–output quantum optical relations and it would imply that, just outside the scattering system, the light field, although carrying information on the scattering process, can be quantized as in free space. We proceed by bounding the scattering system with two planes at $z = \pm L$, thus separating space into three regions: the left-hand region (I) ($z < -L$), the scattering region (II) ($-L < z < L$) and the right-hand region (III) ($z > L$). By fully exploiting the Green-tensor formalism including the Dyson and the Lippman–Schwinger equations we have derived input–output quantum optical relations for the general scattering system described above. In particular the electric-field operator in the regions of space outside the scattering system can be expanded in terms of input and output operators. By using the angular spectrum of plane waves, we obtain

$$\hat{\boldsymbol{E}}^{+}(\boldsymbol{r}, \omega) = \mathrm{i}\sqrt{\frac{\hbar\omega}{2\varepsilon_0}} \sum_K \times [\boldsymbol{\phi}_K^{>}(\boldsymbol{r}, \omega)\hat{a}_K^{>}(\omega) + \boldsymbol{\phi}_K^{<}(\boldsymbol{r}, \omega)\hat{b}_K^{<}(\omega)] \qquad z < -L \quad (7)$$

$$\hat{\boldsymbol{E}}^{+}(\boldsymbol{r}, \omega) = \mathrm{i}\sqrt{\frac{\hbar\omega}{2\varepsilon_0}} \sum_K \times [\boldsymbol{\phi}_K^{>}(\boldsymbol{r}, \omega)\hat{b}_K^{>}(\omega) + \boldsymbol{\phi}_K^{<}(\boldsymbol{r}, \omega)\hat{a}_K^{<}(\omega)] \qquad z > L \quad (8)$$

where $\hat{a}_K^{>/<}(\omega)$ are rightward (>) and leftward (<) input destruction photon operators, while $\hat{b}_K^{>/<}(\omega)$ are the output photon operators that depend on the input photon operators and on the noise currents and $K \equiv (\boldsymbol{K}, \sigma)$ is a shortcut for the wavevector projection along the xy plane and the polarization direction σ. $\boldsymbol{\phi}_K^{>/<}(\boldsymbol{r}, \omega)$ are plane waves describing light propagation in free space. They are given by

$$\boldsymbol{\phi}_K^{>/<}(\boldsymbol{r}, \omega) = \alpha_{\boldsymbol{K}} \boldsymbol{e}_K^{>/<} \exp \mathrm{i}(\boldsymbol{K} \cdot \boldsymbol{R} \pm k_z z) \qquad (9)$$

where $\boldsymbol{r} = (\boldsymbol{R}, z)$, $\boldsymbol{e}_K^{\tau}$ is the polarization unit vector, $k_z = (\omega^2/c^2 - K^2)^{1/2}$ and $\alpha_{\boldsymbol{K}} = (\omega/2\pi c^2 k_z \mathcal{A})^{1/2}$, $\mathcal{A}$ being the quantization surface. The output operators $\hat{\boldsymbol{b}}_K \equiv (\hat{b}_K^{>}, \hat{b}_K^{<})$ are related to the input photon operators $\hat{\boldsymbol{a}}_K \equiv (\hat{a}_K^{>}, \hat{a}_K^{<})$ and to the noise currents $\hat{\boldsymbol{j}}(\boldsymbol{r})$ of the scattering system, according to

$$\hat{\boldsymbol{b}}_K = \sum_{K'} \boldsymbol{S}_K^{K'} \hat{\boldsymbol{a}}_{K'} + \hat{\boldsymbol{F}}_K \qquad (10)$$

where $\boldsymbol{S}_K^{K'}$ is a 2×2 scattering matrix (S matrix),

$$\boldsymbol{S}_K^{K'} = \begin{pmatrix} T_K^{K'} & R_K^{K'} \\ \mathcal{R}_K^{K'} & \mathcal{T}_K^{K'} \end{pmatrix} \qquad (11)$$

whose elements are the generalized reflection and transmission coefficients [18] and $\hat{\boldsymbol{F}}_K$ is a two-dimensional quantum noise vector,

$$\hat{\boldsymbol{F}}_K = \frac{\mathrm{i}\pi}{\varepsilon_0}\sqrt{\frac{2\varepsilon_0}{\hbar\omega}}(F_K^{>}, F_K^{<})$$

where

$$F_K^{>/<} = \int \boldsymbol{\psi}_{\bar{K}}^{</>}(\boldsymbol{r}) \cdot \hat{\boldsymbol{j}}(\boldsymbol{r})\, \mathrm{d}\boldsymbol{r} \qquad (12)$$

with

$$\boldsymbol{\psi}_K^{\tau} = (\boldsymbol{1} - \boldsymbol{G}\boldsymbol{e}_s)\boldsymbol{\psi}_K^{\tau}.$$

If the quantum state of input radiation and of the material system is known, any output photon correlation can be directly calculated by using these relations provided the classical light modes $\boldsymbol{\psi}_{\bar{K}}^{<}$ (where $\bar{K} \equiv (-\boldsymbol{K}, \sigma)$) have been computed. Light modes for specific complex structures can be calculated according to the scheme described in [19]. The obtained input–output relations (10) are based on the angular-spectrum representation. As well known, this representation also describes explicitly the evanescent waves [18] that appear for $K > k$. The relations (10) also hold for evanescent fields and naturally define output photon operators associated with evanescent waves. With the increase in techniques based on measurement and control of evanescent waves, these relations should find application for the analysis of evanescent nonclassical fields, for example arising from the scattering of nonclassical input fields by nanometric objects.

As an application of the quantization scheme described above we analyse the spatial variations of vacuum and thermal fluctuations and describe their relationship. Let us start considering vacuum fluctuations in presence of a scattering system. As well known, vacuum fluctuations play a fundamental role in quantum optical processes. By using equation (4) we obtain

$$\langle \hat{\boldsymbol{E}}_i(\boldsymbol{r}_1, \omega)\hat{\boldsymbol{E}}_j(\boldsymbol{r}_2, \omega')\rangle_{0,0} = S_{ij}^0(\boldsymbol{r}_1, \boldsymbol{r}_2, \omega)\delta(\omega - \omega'). \qquad (13)$$

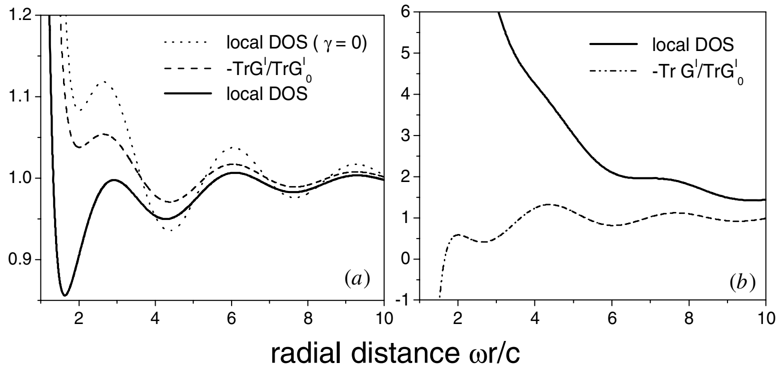

Figure 1. Radiative local DOS $\rho(r,\omega)/\rho_0(\omega)$ for light scattering from one resonant point scatterer at $r=0$ as a function of the scaled radial distance ($\rho_0(\omega)$ is the free-space local DOS) for $\gamma = 1.2\omega_0/Q$ (full curve) and $\gamma = 0$ (dotted curve). The dashed curve describes $-\,\mathrm{Tr}[\boldsymbol{G}^I]/\,\mathrm{Tr}[\boldsymbol{G}^I_0]$. The two-level scatterer is considered in the ground state (*a*) and for perfectly inverted populations (*b*). Parameters are given in the text.

In equation (13), $\langle\ \rangle_{a,b}$ indicates the expectation value, where (a,b) labels respectively the state of input light and the state of the material system. In this case $(0,0)$ indicates the vacuum state for both the Hilbert spaces. S^0_{ij} contains two contributions, one from the particular solution (6) and the other from the homogeneous solution (5). It is possible (see equation (18) below) to express these two contributions in terms of the imaginary part of the Green tensor, thus obtaining

$$S^0_{ij}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega) = -(\hbar\omega^2/\varepsilon_0\pi c^2)G^I_{ij}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega). \tag{14}$$

Equation (14) agrees with results obtained by applying the fluctuation–dissipation theorem [20].

Let us now consider a scattering system with an effective uniform temperature T embedded in a vacuum at *zero* temperature. By using equation (4) and the property [16]

$$\langle \hat{j}^\dagger_i(\boldsymbol{r},\omega)\hat{j}_j(\boldsymbol{r}',\omega)\rangle = \frac{\hbar}{\pi\mu_0}\frac{\omega^2}{c^2}\chi^I_{ij}(\boldsymbol{r},\boldsymbol{r}',\omega)N(\omega,T)\delta(\omega-\omega') \tag{15}$$

we obtain

$$\langle \hat{\boldsymbol{E}}^-_i(\boldsymbol{r}_1,\omega)\hat{\boldsymbol{E}}^+_j(\boldsymbol{r}_2,\omega')\rangle_{0,T} = W_{ij}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega)\delta(\omega-\omega') \tag{16}$$

with

$$W_{ij}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega) = N(\omega,T)(\hbar\omega^2/\varepsilon_0\pi c^2)\tilde{A}(\boldsymbol{r}_1,\boldsymbol{r}_2,\omega) \tag{17}$$

where $N(\omega,T)$ is the mode occupation described by Planck's formula,

$$\bar{N}(\omega,T) = \frac{1}{\exp(\hbar\omega/k_BT)-1}$$

and where the tensor $\tilde{A}$ can be expressed in compact notation as $\tilde{\boldsymbol{A}} = \boldsymbol{G}e^I_s\boldsymbol{G}^*$. Equation (17) is very similar to the expression used in [18] to calculate the cross-spectral density tensor of the near field thermally emitted into free space by an opaque planar source. By using the Green-tensor formalism it is possible to derive the following relationship involving $\tilde{\boldsymbol{A}}$ and the imaginary part of the Green tensor [16]:

$$\frac{\pi c^2}{2\omega}\tilde{\rho}_{i,j}(\boldsymbol{r},\boldsymbol{r}') = -\boldsymbol{G}^I_{ij}(\boldsymbol{r},\boldsymbol{r}') - \tilde{A}_{ij}(\boldsymbol{r},\boldsymbol{r}') \tag{18}$$

where

$$\tilde{\rho}_{i,j}(\boldsymbol{r},\boldsymbol{r}') = \sum_{\tau,K}\psi^\tau_{K,i}(\boldsymbol{r})\psi^{\tau*}_{K,j}(\boldsymbol{r}'). \tag{19}$$

This equation has been demonstrated for particular cases [12, 14]. Its general derivation will be presented elsewhere. Using equation (18), equation (17) can be written in the form

$$\boldsymbol{W}(\boldsymbol{r},\boldsymbol{r}',\omega) = N(\omega,T)\left[\boldsymbol{S}^0(\boldsymbol{r},\boldsymbol{r}',\omega) - \frac{\hbar\omega}{2\varepsilon_0}\tilde{\rho}(\boldsymbol{r},\boldsymbol{r}',\omega)\right]. \tag{20}$$

Equation (20) establishes a general relationship between the spatial variations of the second-order coherence tensors for vacuum fluctuations and thermal light emission. We observe that, while vacuum fluctuations originate from both the scattering system and the input light modes, light emission in a zero-temperature free space originates only from the scattering system. This explains why the spatial variation of the tensor describing light emission can be obtained by subtracting from the contribution due to the vacuum fluctuation the contribution originating from the input light modes $\tilde{\rho}(\boldsymbol{r},\boldsymbol{r}',\omega)$ and eventually reflected by the thermal source. Taking the trace of both sides of equation (18) we obtain

$$\rho(\boldsymbol{r},\omega) = -\frac{2\omega}{\pi c^2}\,\mathrm{Tr}[\boldsymbol{G}^I(\boldsymbol{r},\boldsymbol{r},\omega) + \tilde{\boldsymbol{A}}(\boldsymbol{r},\boldsymbol{r},\omega)] \tag{21}$$

where $\rho(\boldsymbol{r},\omega) \equiv \mathrm{Tr}(\tilde{\rho}(\boldsymbol{r},\boldsymbol{r},\omega))$. $\rho(\boldsymbol{r},\omega)$ describes the light intensity at $\boldsymbol{r}$ due to incoherent illumination, i.e. with input light modes arriving from all spatial directions. In systems with a real permittivity $\rho(\boldsymbol{r},\omega)$ is the local optical density of states (DOS) and can be expressed as $\rho(\boldsymbol{r},\omega) = -(2\omega/\pi c^2)\,\mathrm{Tr}\,\boldsymbol{G}^I(\boldsymbol{r},\boldsymbol{r},\omega)$, i.e. it is proportional to vacuum fluctuations. Equation (21) shows that this well known relation does not hold in the presence of systems with a complex permittivity and provides the correct extension. In the presence of absorption there are thus two possible different definitions of the DOS that coincide in absence of absorption. In the following we refer to $\rho(\boldsymbol{r},\omega)$ as the local radiative DOS and in order to inspect the differences between these two quantities we present numerical results for a pointlike scattering object which can be regarded as the building block of much more complicated scattering objects [19, 21]. The internal resonance is modelled by using the dielectric function of a homogeneously broadened two-level system with energy spacing ω_0 and with given upper- and lower-level populations (N_u and N_l respectively). We observe that, since the theory is linear, the populations are assumed not to be disturbed by the scattering and the emission process. Figure 1(*a*) displays results for the system in the ground state ($N_u = 0$, $N_l = 1$). In figure 1(*b*) results are presented for the case of perfectly inverted populations. In both cases it is possible to observe a clear influence of the correction term. The significant increase of the local DOS in figure 1(*b*) as compared with that shown in figure 1(*a*) is a direct manifestation of light amplification for inverted populations. Figure 1 has been obtained by considering a point scatterer characterized by $\omega = \omega_0$ and using a quality factor $Q = 1000$ and a homogeneous broadening $\gamma = 1.2\omega_0/Q$.

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