Quantum Theory of Light and Noise Polarization in Nonlinear Optics

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We present a consistent quantum theory of the electromagnetic field in nonlinearly responding causal media, with special emphasis on $\chi^{(2)}$ media. Starting from QED in linearly responding causal media, we develop a method to construct the cubic Hamiltonian expressed in terms of the complex nonlinear susceptibility in a quantum mechanically consistent way. In particular, we show that the method yields the nonlinear noise polarization, which together with the linear one is responsible for intrinsic quantum decoherence.

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Recent advances in quantum information technologies have been the main driving forces behind the desire to build parametric down-conversion sources of entangled photon pairs (in the low-intensity limit) [1–5] or two-mode squeezed states (in the high-intensity limit) [6] with high fidelity. It is known that single-photon states of nonunit efficiency, as produced by heralded single-photon sources using parametric down-conversion, cannot be purified by using linear optical elements and photo detection to yield states with higher efficiency [7,8]. That in turn means that postprocessing of single-photon sources is impossible and the sources themselves have to be improved. In order to achieve maximal purity of heralded single-photon states or correlated (entangled) twin-beam photons it is therefore necessary to investigate the theoretical limits nature imposes on us.

An important step in this direction is to provide a quantum theory of light that takes into account nonlinear processes such as parametric down-conversion, and at the same time decoherence mechanisms due to unavoidable absorption losses of the nonlinear material the light interacts with. The theory of quantized electromagnetic fields in linearly and causally responding materials (with the linear response function satisfying the Kramers-Kronig relations) is well established [see, e.g., Refs. [9–11]]. It has been known for some time that analogous Kramers-Kronig relations do also hold for nonlinear susceptibilities [12]. Hence, it will be interesting to see how these causal relations appear in a nonlinear quantum theory.

Previous work on electromagnetic field quantization in nonlinear materials have focused on strictly lossless materials where Lagrangian methods and mode decompositions apply [13–16]. A first attempt to include in the field quantization both linear and nonlinear losses was made in Ref. [17] for Kerr media, by extending the linear harmonic-oscillator model used in the Huttner-Barnett quantization scheme [9] to a nonlinear one. A consistent approach that includes—for given nonlinear susceptibility—absorption and dispersion has not yet been formulated within the frame of (macroscopic) QED.

In this Letter we will show how to consistently quantize the electromagnetic field in the presence of nonlinearly responding causal materials. Although we will focus here on $\chi^{(2)}$ media, the theory is not restricted to this particular type of nonlinearity. This theory provides the starting point for further investigations of theoretical limits to the performance of nonlinear optical elements as sources of nonclassical light. Starting from the cubic Hamiltonian expressed in terms of the canonically conjugated variables as used in QED in linear causal media, we first express the nonlinear polarization field in terms of these variables as well. This is compared with the classical nonlinear response which enables us to identify the nonlinear noise contributions.

We begin by recalling the quantization scheme for the electromagnetic field in the presence of a linearly (and locally) responding causal dielectric medium of permittivity $\varepsilon(\mathbf{r},\omega)=\varepsilon'(\mathbf{r},\omega)+i\varepsilon''(\mathbf{r},\omega)$ [10]. In this case the Hamiltonian is bilinear,

$$H_L=\int d^3r\int_0^\infty d\omega\hbar\omega\mathbf{f}^\dagger(\mathbf{r},\omega)\cdot\mathbf{f}(\mathbf{r},\omega), \tag{1}$$

with the annihilation and creation operators $f_i(\mathbf{r},\omega)$ and $f_i^\dagger(\mathbf{r},\omega)$, respectively, playing the role of the canonically conjugate dynamical variables which are attributed to collective excitations of the electromagnetic field and the dispersing and absorbing dielectric matter and obeying the bosonic commutation rules $[f_i(\mathbf{r},\omega),f_j^\dagger(\mathbf{r}',\omega')]=\delta_{ij}\delta(\omega-\omega')\delta(\mathbf{r}-\mathbf{r}')$. The electromagnetic field (in the Schrödinger picture) can be expressed in terms of the dynamical variables as follows. The electric field reads

$$\mathbf{E}(\mathbf{r})=\int_0^\infty d\omega\underline{\mathbf{E}}(\mathbf{r},\omega)+\text{H.c.}, \tag{2}$$

$$\underline{\mathbf{E}}(\mathbf{r},\omega)=i\sqrt{\frac{\hbar}{\pi\varepsilon_0}}\frac{\omega^2}{c^2}\int d^3s\sqrt{\varepsilon''(\mathbf{s},\omega)}\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)\cdot\mathbf{f}(\mathbf{s},\omega), \tag{3}$$

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and the displacement field reads

$$\mathbf{D}_L(\mathbf{r}) = \int_0^\infty d\omega \underline{\mathbf{D}}_L(\mathbf{r}, \omega) + \text{H.c.}, \tag{4}$$

$$\underline{\mathbf{D}}_L(\mathbf{r}, \omega) = (\mu_0\omega^2)^{-1}\nabla \times \nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega) = \varepsilon_0\varepsilon(\mathbf{r}, \omega)\underline{\mathbf{E}}(\mathbf{r}, \omega) + \underline{\mathbf{P}}_L^{(N)}(\mathbf{r}, \omega) \tag{5}$$

with the linear noise polarization

$$\underline{\mathbf{P}}_L^{(N)}(\mathbf{r}, \omega) = i\sqrt{\frac{\hbar\varepsilon_0}{\pi}\varepsilon''(\mathbf{r}, \omega)}\mathbf{f}(\mathbf{r}, \omega). \tag{6}$$

The induction field is obtained by replacing in Eq. (2) $\mathbf{E}(\mathbf{r})$ and $\underline{\mathbf{E}}(\mathbf{r}, \omega)$, respectively, with $\mathbf{B}(\mathbf{r})$ and $\underline{\mathbf{B}}(\mathbf{r}, \omega) = 1/(i\omega)\nabla \times \underline{\mathbf{E}}(\mathbf{r}, \omega)$. In Eq. (3) the dyadic Green function $\boldsymbol{G}(\mathbf{r}, \mathbf{s}, \omega)$ is the unique fundamental solution of the inhomogeneous Helmholtz equation $\nabla \times \nabla \times \boldsymbol{G}(\mathbf{r}, \mathbf{s}, \omega) - \frac{\omega^2}{c^2}\varepsilon(\mathbf{r}, \omega)\boldsymbol{G}(\mathbf{r}, \mathbf{s}, \omega) = \delta(\mathbf{r} - \mathbf{s})\boldsymbol{I}$ and contains all relevant information about the material properties and the geometry of the system.

To turn over to the nonlinear media, let us first fix some notation. From now on we will abbreviate spatial and frequency variables $(\mathbf{r}_k, \omega_k)$ by their label $\boldsymbol{k}$, e.g., $\mathbf{1} \equiv (\mathbf{r}_1, \omega_1)$ and write $\int d\boldsymbol{k} \equiv \int d^3r_k \int d\omega_k$. In the latter integrals, the spatial integration extends over all space. The frequency integral, which we initially will assume to range over all positive frequencies, will be restricted later on. On recalling the physical meaning of the dynamical variables $f_i(\mathbf{r}, \omega)$ and $f_i^\dagger(\mathbf{r}, \omega)$, the most general normal-order form of the nonlinear interaction energy that corresponds to a $\chi^{(2)}$ medium reads

$$H_{NL} = \int d\mathbf{1}d\mathbf{2}d\mathbf{3}\alpha_{i(jk)}(\mathbf{1},\mathbf{2},\mathbf{3})f_i^\dagger(\mathbf{1})f_j(\mathbf{2})f_k(\mathbf{3}) + \text{H.c.} \tag{7}$$

The unknown tensor function $\alpha_{i(jk)}(\mathbf{1}, \mathbf{2}, \mathbf{3})$, which has to be symmetrized over its last two indices to avoid double counting, has to be determined from constraints imposed by generally accepted relations.

We first note that Faraday's law can be written as

$$\nabla \times \mathbf{E}(\mathbf{r}) = -\dot{\mathbf{B}}(\mathbf{r}) = -\frac{1}{i\hbar}[\mathbf{B}(\mathbf{r}), H_L + H_{NL}]. \tag{8}$$

Both the (transverse) electric and the induction fields are pure electromagnetic fields without being related to the material degrees of freedom and hence their equal-time commutation relations are as in vacuum QED. Therefore, their functional form in terms of the dynamical variables $f_i(\mathbf{r}, \omega)$ and $f_i^\dagger(\mathbf{r}, \omega)$ are the same as in the linear (noninteracting) theory. From here it immediately follows that

$$[\mathbf{B}(\mathbf{r}), H_{NL}] = 0. \tag{9}$$

Using Faraday's law, we rewrite Ampere's law as

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) = -\mu_0\ddot{\mathbf{D}}_L(\mathbf{r}) - \mu_0\ddot{\mathbf{P}}_{NL}(\mathbf{r}), \tag{10}$$

where we have split up the dielectric displacement field $\mathbf{D}(\mathbf{r})$ into the linear part $\mathbf{D}_L(\mathbf{r})$ and the nonlinear polarization $\mathbf{P}_{NL}(\mathbf{r})$. Employing Heisenberg's equation of motion, we may rewrite Eq. (10) as

$$\nabla \times \nabla \times \mathbf{E}(\mathbf{r}) - \frac{\mu_0}{\hbar^2}[[\mathbf{D}_L(\mathbf{r}), H_L], H_L] = \frac{\mu_0}{\hbar^2}\{[[\mathbf{D}_L(\mathbf{r}), H_L], H_{NL}] + [[\mathbf{D}_L(\mathbf{r}), H_{NL}], H_L] + [[\mathbf{P}_{NL}(\mathbf{r}), H_L], H_L]\}, \tag{11}$$

where we have retained only terms that are in zeroth or first order in the nonlinear coupling coefficient $\alpha_{i(jk)}(\mathbf{1}, \mathbf{2}, \mathbf{3})$. The left-hand side of Eq. (11) is zero by the definition of the linear displacement field. Note that the time dependence is carried by the time-dependent dynamical variables $f_i(\mathbf{r}, \omega, t)$ and $f_i^\dagger(\mathbf{r}, \omega, t)$. The first term on the right-hand side of Eq. (11) vanishes by virtue of the constraint (9). Hence, we are left with a relation between double commutators of the linear displacement and nonlinear polarization fields with the linear and nonlinear parts of the Hamiltonian, $[[\mathbf{D}_L(\mathbf{r}), H_{NL}], H_L] = -[[\mathbf{P}_{NL}(\mathbf{r}), H_L], H_L]$. A particular solution is certainly

$$[\mathbf{D}_L(\mathbf{r}), H_{NL}] = -[\mathbf{P}_{NL}(\mathbf{r}), H_L]. \tag{12}$$

The general solution would additionally include commutants with the Hamiltonian H_L. Terms commuting with H_L are functionals of the number (density) operator $\mathbf{f}^\dagger(\mathbf{r}, \omega) \cdot \mathbf{f}(\mathbf{r}, \omega)$. However, these terms have to be discarded as they would lead to diverging contributions to $\mathbf{P}_{NL}(\mathbf{r})$. Hence, the solution to Eq. (12) is already the physically relevant general solution.

The expression on the right-hand side of Eq. (12) is nothing but the Liouvillian $\mathcal{L}_L$ generated by the Hamiltonian H_L acting on the nonlinear polarization field. Therefore, Eq. (12) can be solved for $\mathbf{P}_{NL}(\mathbf{r})$ to yield

$$\mathbf{P}_{NL}(\mathbf{r}) = -\frac{1}{i\hbar}\mathcal{L}_L^{-1}[\mathbf{D}_L(\mathbf{r}), H_{NL}], \tag{13}$$

the linear displacement field, Eqs. (4) and (5), consists of a reactive part related to the electric field and a noise part $\mathbf{P}_L^{(N)}(\mathbf{r})$. Inserting Eq. (4) together with Eq. (5) into Eq. (13), we see that the nonlinear polarization also decomposes into a reactive part, which can be related to the nonlinear response, and a noise part, which determines the nonlinear noise polarization

$$\mathbf{P}_{NL}^{(N)}(\mathbf{r}) = -\frac{1}{i\hbar}\mathcal{L}_L^{-1}[\mathbf{P}_L^{(N)}(\mathbf{r}), H_{NL}]. \tag{14}$$

Because of the relation (6), $\mathbf{P}_{NL}^{(N)}(\mathbf{r})$ vanishes if the imaginary part of the linear permittivity, $\varepsilon''(\mathbf{r}, \omega)$, and hence the noise associated with it tends to zero [18].

The inverse Liouvillian can be calculated using standard techniques, and we obtain from Eq. (13)

$$\mathbf{P}_{NL}(\mathbf{r}) = \frac{i}{\hbar}\lim_{s\to 0}\int_0^\infty d\tau e^{-s\tau}e^{-i/\hbar H_L\tau}[\mathbf{D}_L(\mathbf{r}), H_{NL}]e^{i/\hbar H_L\tau}, \tag{15}$$

where the real positive number s ensures convergence of the integral. In the next step we compute the commutator $[\mathbf{D}_L(\mathbf{r}), H_{NL}]$ and evaluate the integral in Eq. (15). First, we evaluate the commutator between the dynamical variables and the cubic Hamiltonian H_{NL}, leading to [here, $\mathbf{0} \equiv (\mathbf{s}, \omega)$]

$$[f_m(\mathbf{0}), H_{NL}] = \int d\mathbf{2}d\mathbf{3}\alpha_{m(jk)}(\mathbf{0}, \mathbf{2}, \mathbf{3})f_j(\mathbf{2})f_k(\mathbf{3}) + \int d\mathbf{1}d\mathbf{2}\alpha^*_{i(jm)}(\mathbf{1}, \mathbf{2}, \mathbf{0})f_j^\dagger(\mathbf{2})f_i(\mathbf{1}). \quad (16)$$

In what follows, we will concentrate on the contribution to the nonlinear displacement and polarization that comes from terms containing two annihilation operators such as $f_j(\mathbf{2})f_k(\mathbf{3})$. We will label these contributions with the superscript $^{(++)}$ in analogy with the standard notation for positive-frequency parts. The inverse Liouvillian of the bilinear combination of annihilation operators is readily found to be $\mathcal{L}^{-1}f_j(\mathbf{2})f_k(\mathbf{3}) = i/(\omega_2 + \omega_3)f_j(\mathbf{2})f_k(\mathbf{3})$. Combined with Eq. (15) we finally obtain for the nonlinear polarization field

$$P^{(++)}_{NL,l}(\mathbf{r}) = \frac{1}{i\hbar}\sqrt{\frac{\hbar\varepsilon_0}{\pi}}\int d\mathbf{0}d\mathbf{2}d\mathbf{3}\frac{\sqrt{\varepsilon''(\mathbf{0})}}{\omega_2 + \omega_3}\alpha_{m(jk)}(\mathbf{0}, \mathbf{2}, \mathbf{3}) \times \frac{\omega^2}{c^2}\varepsilon(\mathbf{r}, \omega)G_{lm}(\mathbf{r}, \mathbf{0})f_j(\mathbf{2})f_k(\mathbf{3}) + P^{(N,++)}_{NL,l}(\mathbf{r}), \quad (17)$$

where the noise polarization reads

$$P^{(N,++)}_{NL,l}(\mathbf{r}) = \frac{1}{i\hbar}\sqrt{\frac{\hbar\varepsilon_0}{\pi}}\int d\mathbf{0}d\mathbf{2}d\mathbf{3}\frac{\sqrt{\varepsilon''(\mathbf{0})}}{\omega_2 + \omega_3}\alpha_{l(jk)}(\mathbf{0}, \mathbf{2}, \mathbf{3}) \times \delta(\mathbf{r} - \mathbf{s})f_j(\mathbf{2})f_k(\mathbf{3}). \quad (18)$$

In order to make contact with standard notation, let us recall the definition of the nonlinear polarization within the framework of response theory:

$$P_{NL,l}(\mathbf{r}, t) = \varepsilon_0\int_{-\infty}^{t} d\tau_1 d\tau_2 \check{\chi}^{(2)}_{lmn}(\mathbf{r}, t - \tau_1, t - \tau_2) \times E_m(\mathbf{r}, \tau_1)E_n(\mathbf{r}, \tau_2) + P^{(N)}_{NL,l}(\mathbf{r}, t). \quad (19)$$

The first term on the right-hand side of Eq. (19) is the causal response well known from nonlinear optics [19], with $\check{\chi}^{(2)}_{lmn}(\mathbf{r}, t_1, t_2)$ being the response function of the $\chi^{(2)}$ medium. The term $P^{(N)}_{NL,l}(\mathbf{r}, t)$ is a (yet unknown) nonlinear noise polarization commonly disregarded in classical nonlinear optics. In most cases of interest it is sufficient to evaluate Eq. (19) in the slowly varying amplitude approximation in the sense that

$$\mathbf{E}(\mathbf{r}, t) = \sum_{\nu=1}^{3}\tilde{\mathbf{E}}(\mathbf{r}, \Omega_\nu, t)e^{-i\Omega_\nu t} + \text{H.c.}, \quad (20)$$

where ν numbers the three relevant field amplitudes of midfrequencies Ω_ν $[\Omega_1 \equiv \Omega_{23} = \Omega_2 + \Omega_3]$ corresponding to the $\chi^{(2)}$ interaction. In Eq. (20), the time scale on which the amplitude function $\tilde{E}_i(\mathbf{r}, \Omega_\nu, t)$ noticeably changes is long compared with Ω_ν^{-1} and the characteristic time of variation of $\check{\chi}^{(2)}_{lmn}(\mathbf{r}, t_1, t_2)$ with respect to both t_1 and t_2 [see, e.g., the treatment in Ref. [20]]. Hence the slowly varying field amplitudes can be taken out of the integral at the upper integration limit t, and we are left with the Fourier transform of $\check{\chi}^{(2)}_{lmn}(\mathbf{r}, t_1, t_2)$, $\chi^{(2)}_{lmn}(\mathbf{r}, \omega_1, \omega_2)$, which slowly varies with ω_1 and ω_2. In this way we derive

$$\tilde{P}^{(++)}_{NL,l}(\mathbf{r}, \Omega_{23}) = \varepsilon_0\chi^{(2)}_{lmn}(\mathbf{r}, \Omega_2, \Omega_3)\tilde{E}_m(\mathbf{r}, \Omega_2)\tilde{E}_n(\mathbf{r}, \Omega_3) + \tilde{P}^{(N,++)}_{NL,l}(\mathbf{r}, \Omega_{23}), \quad (21)$$

where the time argument t of the $\sim$ quantities has been omitted for notational convenience.

The validity of the approximation leading from Eq. (19) to Eq. (21) may be regarded as being a prerequisite for substantiating the effective interaction Hamiltonian (8). At the same time, it suggests further specification of the Hamiltonian as therein the introduction of slowly varying variables is desirable. In view of Eqs. (2) and (3), we define, on assuming the Green tensor and the linear susceptibility are slowly varying with ω, the slowly varying bosonic variables $\tilde{\mathbf{f}}(\mathbf{r}, \Omega_\nu) = (\Delta\Omega_\nu)^{-1/2} \times \int_{\Delta\Omega_\nu} d\omega\mathbf{f}(\mathbf{r}, \omega, t)e^{i\Omega_\nu t}$ ($\Delta\Omega_\nu$, relevant frequency interval around Ω_ν), and Eq. (8) reduces to

$$H_{NL} = \int d^3s_1 d^3s_2 d^3s_3 \alpha_{i(jk)}(\mathbf{s}_1, \Omega_{23}, \mathbf{s}_2, \Omega_2, \mathbf{s}_3, \Omega_3) \times \sqrt{\Delta\Omega_1\Delta\Omega_2\Delta\Omega_3}\tilde{f}_i^\dagger(\mathbf{s}_1, \Omega_{23})\tilde{f}_j(\mathbf{s}_2, \Omega_2)\tilde{f}_k(\mathbf{s}_3, \Omega_3) + \text{H.c.} \quad (22)$$

Introducing in Eqs. (17) and (21) the slowly varying variables $\tilde{\mathbf{f}}(\mathbf{r}, \Omega_\nu)$, from a comparison of the reactive parts of the nonlinear polarization as given by the two equations we derive the following integral equation for determining the nonlinear coupling coefficient $\alpha_{i(jk)}(\mathbf{s}_1, \Omega_{23}, \mathbf{s}_2, \Omega_2, \mathbf{s}_3, \Omega_3)$ in terms of the nonlinear susceptibility $\chi^{(2)}_{lmn}(\mathbf{r}, \Omega_2, \Omega_3)$:

$$\int d^3s\sqrt{\varepsilon''(\mathbf{s}, \Omega_{23})}\alpha_{m(jk)}(\mathbf{s}, \Omega_{23}, \mathbf{s}_2, \Omega_2, \mathbf{s}_3, \Omega_3)G_{lm}(\mathbf{r}, \mathbf{s}, \Omega_{23}) = \frac{\hbar^2}{i\pi c^2}\sqrt{\frac{\pi}{\hbar\varepsilon_0}}\frac{\Omega_2^2\Omega_3^2}{\Omega_{23}\varepsilon(\mathbf{r}, \Omega_{23})}\sqrt{\varepsilon''(\mathbf{s}_2, \Omega_2)\varepsilon''(\mathbf{s}_3, \Omega_3)}\chi^{(2)}_{lmn}(\mathbf{r}, \Omega_2, \Omega_3)G_{mj}(\mathbf{r}, \mathbf{s}_2, \Omega_2)G_{nk}(\mathbf{r}, \mathbf{s}_3, \Omega_3). \quad (23)$$

This equation is of Fredholm type and can be solved by inverting the integral kernel on the left-hand side of Eq. (23). Note that the inverse of the Green tensor is just the Helmholtz operator $H_{ij}(\mathbf{r},\omega) = \partial_i^r \partial_j^r - \delta_{ij}\Delta^r - (\omega^2/c^2)\varepsilon(\mathbf{r},\omega)\delta_{ij}$: $H_{ij}(\mathbf{r},\omega)G_{jk}(\mathbf{r},\mathbf{s},\omega) = \delta_{ik}\delta(\mathbf{r}-\mathbf{s})$. Hence, from Eq. (23) it follows that

$$\alpha_{i(jk)}(\mathbf{r},\Omega_{23},\mathbf{s}_2,\Omega_2,\mathbf{s}_3,\Omega_3) = \frac{\hbar^2}{i\pi c^2}\sqrt{\frac{\pi}{\hbar\varepsilon_0}}\frac{\Omega_2^2\Omega_3^2}{\Omega_{23}}\sqrt{\frac{\varepsilon''(\mathbf{s}_2,\Omega_2)\varepsilon''(\mathbf{s}_3,\Omega_3)}{\varepsilon''(\mathbf{r},\Omega_{23})}}H_{li}(\mathbf{r},\Omega_{23}) \times[\varepsilon^{-1}(\mathbf{r},\Omega_{23})\chi^{(2)}_{imn}(\mathbf{r},\Omega_2,\Omega_3)G_{mj}(\mathbf{r},\mathbf{s}_2,\Omega_2)G_{nk}(\mathbf{r},\mathbf{s}_3,\Omega_3)]. \quad (24)$$

Reinserting Eq. (24) into Eq. (18) eventually yields, on recalling Eqs. (2), (3), and (20) and the definition of the slowly varying variables $\tilde{\mathbf{f}}(\mathbf{r},\Omega_\nu)$, the following expression for the nonlinear noise polarization:

$$\tilde{P}^{(N,++)}_{NL,l}(\mathbf{r},\Omega_{23}) = \frac{\varepsilon_0 c^2}{\Omega_{23}^2}H_{li}(\mathbf{r},\Omega_{23})[\varepsilon^{-1}(\mathbf{r},\Omega_{23}) \times \chi^{(2)}_{imn}(\mathbf{r},\Omega_2,\Omega_3)\tilde{E}_m(\mathbf{r},\Omega_2)\tilde{E}_n(\mathbf{r},\Omega_3)]. \quad (25)$$

To our knowledge, this is the first time a nonlinear noise polarization has been derived in the frame of quantum nonlinear optics. Note again that, by construction [see Eq. (14)], the nonlinear noise polarization (25) tends to zero when $\varepsilon''(\mathbf{r},\omega) \to 0$. To estimate the order of magnitude of the nonlinear noise polarization, we may disregard vector characters and the accurate frequency and space dependencies in the above equations. It then follows that $|\mathcal{P}^{(N)}_{NL}|/|\mathcal{P}^{(N)}_{L}| \sim |\chi^{(2)}/\varepsilon||\mathcal{E}|$, where $|\mathcal{P}^{(N)}_{L}|$ and $|\mathcal{P}^{(NL)}_{NL}|$ are, respectively, measures of the strengths of the linear and nonlinear noise polarizations, and $|\mathcal{E}|$ measures the strength of a pump. Hence, for strong pumping the nonlinear noise polarization can become essential.

In summary, we have presented a consistent quantum theory of the electromagnetic field in the presence of quadratically responding dielectric materials. It takes care of the causal nature of the dielectric response which implies the existence of a nonlinear noise polarization. The nonlinear (effective) interaction Hamiltonian (22) [corresponding to Eq. (7) in the slowly varying amplitude approximation], together with the nonlinear coupling coefficient from Eq. (24), allows one to study nonlinear quantum optical processes such as parametric down-conversion in the presence of realistic dielectric materials. The main advantage of our approach is that it automatically takes absorption—via the complex permittivity—and geometric boundaries—via the dyadic Green function—into account. The procedure to generalize the theory presented above is by no means restricted to quadratic responses. In fact, one can construct a hierarchy of Hamiltonians with increasing number of the dynamical variables $\mathbf{f}(\mathbf{r},\omega)$ and $\mathbf{f}^\dagger(\mathbf{r},\omega)$ corresponding to higher-order nonlinear responses. The construction ensures that the equal-time commutation relations between the relevant field operators are preserved. We believe this theory represents an important step towards further studies with the aim to understand the ultimate limits on the performance of quantum optical processes.

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