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Causal Nonlinear Quantum Optics

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Abstract. We present a quantization scheme for the electromagnetic field in nonlinearly responding dielectric materials with dispersion and absorption. Starting from QED in linear dielectrics, we construct an effective nonlinear interaction Hamiltonian and show the emergence of a nonlinear noise polarization whose magnitude is related to the magnitude of the pump fields.

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1. Introduction

Quantum electrodynamics is rightly regarded as one of the most successful theories in physics. Its predictions have been tested to an astonishing accuracy in, e.g., measurement of the anomalous magnetic moment of the electron. In recent years we have seen remarkable progress in combining the quantum theory of the electromagnetic field with (linear) response theories associated with absorbing dielectric matter.

The usual route to obtain a quantum theory of light in (linearly) responding dielectric materials is as follows. The starting point is vacuum quantum electrodynamics, i.e. the quantized Maxwell equations without matter. Elementary charged particles are then coupled in a relativistically invariant form to the Maxwell field. In quantum optics, an approximation is usually made at this point. Often it is not necessary to keep the theory fully relativistically invariant, but a non-relativistic approach to the matter suffices. The resulting interaction Hamiltonian describes, in minimal coupling, the atom-field interaction in leading order of the particles' velocities.

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In this minimal coupling, the particles are described by their microscopic quantities such as position and momentum. For systems containing many particles, such a description can be very inconvenient. Power and Zienau [1, 2] have shown that there is an alternative description in terms of collective variables such as polarization and magnetization which makes the transition to macroscopically large atomic systems rather straightforward.

The determination of polarization and magnetization itself requires knowledge about the interaction between the material system under consideration with the electromagnetic field. Within the framework of (linear) response theory, the polarization and magnetization are expanded in (linear) powers of the electric and magnetic fields, respectively. The response functions connecting polarization and magnetization on one hand and electric and magnetic fields on the other are known as the dielectric and magnetic susceptibilities. They have the benefit of being experimentally accessible so that detailed information of the microscopic properties of the material system are not needed. Quantization of the macroscopic electromagnetic field along these lines has been successfully performed by several authors [3–8].

The next obvious step is to extend the formalism to include nonlinearly responding materials. However, standard nonlinear optics typically neglects absorption associated with nonlinear couplings which is known to be present even in the nonlinear case as Kramers–Kronig relations also exist for nonlinear susceptibilities [9]. This observation immediately leads one to conclude that, on a macroscopic level, standard mode decompositions cannot be performed for all frequencies, and hence a consistent field theory cannot be built upon them.

In Sec. 2 we will briefly review the main concept of field quantization in linear dielectrics which serves as the starting point of the following considerations. In particular, we will show that we can find a bilinear Hamiltonian that generates the time-dependent Maxwell equations. We then present an effective Hamiltonian incorporating nonlinear couplings in Sec. 3 and give arguments in favour of the generality of its analytical form. In Sec. 4 we derive the nonlinear polarization. We end with some concluding remarks in Sec. 5.

2. Linear Response Theory

Here we briefly describe the scheme for quantizing the electromagnetic field in the presence of linearly and locally responding dielectric material. For simplicity, we restrict our discussion to non-magnetic materials. The constitutive relation that provides the relation between the polarization and the electric field has the general form

$$\mathbf{P}_L(\mathbf{r},t) = \varepsilon_0 \int_0^\infty d\tau\, \chi(\mathbf{r},\tau)\mathbf{E}(\mathbf{r},t-\tau) + \mathbf{P}_L^{(N)}(\mathbf{r},t)\,, \tag{1}$$

where a noise polarization $\mathbf{P}_L^{(N)}(\mathbf{r},t)$ has been added to the usual causal response. This contribution describes a Langevin noise term with zero mean and is there to preserve Poisson brackets and, in the quantized theory, commutation rules. The response function $\chi(\mathbf{r},\tau)$ is the Fourier transformed dielectric susceptibility, the latter of which fulfils the well-known Kramers–Kronig relations which states that its real and imaginary parts form Hilbert transform pairs.

Upon quantization, we associate the (frequency components of the) linear noise polarization $\mathbf{P}_L^{(N)}(\mathbf{r},\omega)$, apart from a multiplicative factor, with a bosonic vector field $\hat{\mathbf{f}}(\mathbf{r},\omega)$ with the equal-time commutation rules $[\hat{\mathbf{f}}(\mathbf{r},\omega),\hat{\mathbf{f}}^\dagger(\mathbf{r}',\omega')] = \delta(\mathbf{r}-\mathbf{r}')\,\delta(\omega-\omega')\boldsymbol{I}$ such that

$$\mathbf{P}_L^{(N)}(\mathbf{r},\omega) = i\sqrt{\frac{\hbar\varepsilon_0}{\pi}\varepsilon_I(\mathbf{r},\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega)\,. \tag{2}$$

With this definition it can be shown that the (frequency components of the) electric field can be expanded as [6, 8]

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

where $\boldsymbol{G}(\mathbf{r},\mathbf{s},\omega)$ is the dyadic Green function of the classical scattering problem, i.e. the fundamental solution to the Helmholtz partial differential equation. The electric-field operator in the Schrödinger picture is obtained by integrating over all frequencies, $\hat{\mathbf{E}}(\mathbf{r}) = \int d\omega\,\hat{\mathbf{E}}(\mathbf{r},\omega) + \text{h.c.}$.

Equation (3) can be regarded as a generalization of the familiar mode expansion with the role of the creation and annihilation operator being taken on by the dynamical variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)$ which, in contrast to the vacuum case, describe collective excitations of the electromagnetic field and the absorbing matter. The time-dependent Maxwell equations are then the Heisenberg equations of motion for the magnetic induction and dielectric displacement fields generated by the bilinear Hamiltonian

$$\hat{H}_L = \int_0^\infty d\omega \int d^3r\,\hbar\omega\,\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)\cdot\hat{\mathbf{f}}(\mathbf{r},\omega)\,. \tag{4}$$

It is worth noting that a Hamiltonian equivalent to (4) has been derived by explicitly diagonalizing a (bilinear) model Hamiltonian [10–12].

Using the representation (3) and the properties of the dyadic Green function (being a response function), one can show that the equal-time commutation relation known from vacuum QED,

$$\left[\varepsilon_0\hat{\mathbf{E}}(\mathbf{r}),\hat{\mathbf{B}}(\mathbf{r}')\right] = -i\hbar\boldsymbol{\nabla}\times\delta(\mathbf{r}-\mathbf{r}')\boldsymbol{I}\,, \tag{5}$$

is still valid. Furthermore, the linear fluctuation-dissipation theorem is respected and takes the specific form

$$\langle 0|\hat{\mathbf{E}}(\mathbf{r},\omega)\hat{\mathbf{E}}^\dagger(\mathbf{r}',\omega')|0\rangle = \frac{\hbar\omega^2}{\pi\varepsilon_0 c^2}\mathrm{Im}\boldsymbol{G}(\mathbf{r},\mathbf{r}',\omega)\delta(\omega-\omega')\,. \tag{6}$$

3. Nonlinear Interaction Hamiltonian

In order to make the step to nonlinear interaction processes, we immediately face several problems. For a start, there is no clearly visible way to extend Eq. (1) to incorporate nonlinear response, in particular, it is by no means clear how to define the nonlinear extension to the noise polarization $\mathbf{P}_{NL}^{(N)}(\mathbf{r},t)$. Moreover, apart from nonlinear response theories corresponding to a $\chi^{(2)}$ nonlinear process, the extensions to the fluctuation-dissipation theorem do not contain contributions from the fully retarded Green function only [13].

Faced with these conceptual difficulties, we argue that a sensible way to proceed is to look at an effective interaction Hamiltonian, with the Hopfield model [14] of linearly absorbing dielectrics [10–12] in mind. Let the reader be reminded that in this model the free electromagnetic field is coupled in bilinear fashion to an harmonic-oscillator polarization field which in turn is coupled to a continuum of harmonic oscillators modeling a heat bath. The explicit diagonalization of this model yields an expression of the dynamical variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$ in terms of a linear combination of the original variables.

An effective nonlinear interaction Hamiltonian for absorbing matter can now be obtained by first treating the interaction between the electromagnetic field and a collection of N-level atoms without coupling to a heat bath. Considering multi-photon processes that are non-resonant with any atomic transition, the resulting effective interaction Hamiltonian will contain monomials in the creation and annihilation operators for photons only [15]. Hence, the detailed atomic level structure becomes invisible in this approximation. This observation allows us to again treat the atoms as harmonic oscillators in the spirit of the Hopfield model. Thus, the bilinear parts of the Hamiltonian can be diagonalized as before which, yet again, results in the introduction of the dynamical variables $\hat{\mathbf{f}}(\mathbf{r},\omega)$. The photon creation and annihilation operators in the effective (nonlinear) interaction Hamiltonian can then be (re-)expressed in terms of the $\hat{\mathbf{f}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}^\dagger(\mathbf{r},\omega)$. As the dynamical variables of the new theory depend linearly on the creation and annihilation operators of the original theory, the same is true in reverse. Hence, the nonlinear interaction Hamiltonian must be of the same form containing the same monomials.

This observation is sufficient to justify an ansatz for an effective interaction Hamiltonian corresponding to a $\chi^{(2)}$ nonlinear process as [16]

$$\hat{H}_{NL} = \int d\mathbf{1}\, d\mathbf{2}\, d\mathbf{3}\, \alpha_{ijk}(\mathbf{1},\mathbf{2},\mathbf{3}) \hat{f}_i^\dagger(\mathbf{1}) \hat{f}_j(\mathbf{2}) \hat{f}_k(\mathbf{3}) + \text{h.c.}\,, \tag{7}$$

where the integration over $\mathbf{k} \equiv (\mathbf{s}_k, \omega_k)$ runs over all the space and all the frequencies. The unknown tensor function $\alpha_{ijk}(\mathbf{1},\mathbf{2},\mathbf{3})$ has to be determined by consistency with requirements from standard nonlinear optics. In particular, we require it to be linear in the nonlinear susceptibility $\chi^{(2)}$.

4. Nonlinear Noise Polarization

With the help of the Hamiltonians (4) and (7) we are in the position to derive an expression for the nonlinear polarization. First note that Faraday's law $\boldsymbol{\nabla}\times\hat{\mathbf{E}}(\mathbf{r}) = -\dot{\hat{\mathbf{B}}}(\mathbf{r}) = -[\hat{\mathbf{B}}(\mathbf{r}),\hat{H}_L+\hat{H}_{NL}]/(i\hbar)$ implies that

$$\left[\hat{\mathbf{B}}(\mathbf{r}),\hat{H}_{NL}\right]=0 \tag{8}$$

per construction of $\hat{\mathbf{B}}(\mathbf{r})$.

By splitting up the total dielectric displacement into its linear part, $\hat{\mathbf{D}}_L(\mathbf{r})$, and the nonlinear polarization $\hat{\mathbf{P}}_{NL}(\mathbf{r})$, Ampère's law can be written in the form

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\hat{\mathbf{E}}(\mathbf{r})=-\mu_0\ddot{\hat{\mathbf{D}}}_L(\mathbf{r})-\mu_0\ddot{\hat{\mathbf{P}}}_{NL}(\mathbf{r})\,. \tag{9}$$

Each of the second time derivatives constitutes a double commutator with the sum of the Hamiltonians (4) and (7). The linear part of the dielectric displacement field is linear in the dynamical variables, whereas the nonlinear polarization corresponding to a $\chi^{(2)}$ nonlinear process is bilinear. Hence, the double commutators produce terms ranging from being linear to quadrilinear in the dynamical variables. This in turn means that we obtain a hierarchy of contributions to different order. In order to compute the nonlinear polarization corresponding to a given $\chi^{(n)}$ nonlinear process, in principle one has to include contributions from lower-order nonlinearities. For our current discussion this means that we neglect all double commutators that result in terms containing more than bilinear combinations of dynamical variables, all higher-order terms in principle contribute to higher-order nonlinearities. Collecting all relevant terms, we obtain

$$\begin{aligned}\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\hat{\mathbf{E}}(\mathbf{r}) &= \frac{\mu_0}{\hbar^2}\left[\left[\hat{\mathbf{D}}_L(\mathbf{r}),\hat{H}_L\right],\hat{H}_L\right]+\frac{\mu_0}{\hbar^2}\left[\left[\hat{\mathbf{D}}_L(\mathbf{r}),\hat{H}_L\right],\hat{H}_{NL}\right]\\ &\quad+\frac{\mu_0}{\hbar^2}\left[\left[\hat{\mathbf{D}}_L(\mathbf{r}),\hat{H}_{NL}\right],\hat{H}_L\right]+\frac{\mu_0}{\hbar^2}\left[\left[\hat{\mathbf{P}}_{NL}(\mathbf{r}),\hat{H}_L\right],\hat{H}_L\right].\end{aligned} \tag{10}$$

The first term on the right-hand side of Eq. (10) is just equal to the left-hand side as can be easily checked, noting that the frequency components of the linear dielectric displacement field can be written as $\hat{\mathbf{D}}_L(\mathbf{r},\omega)=\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\hat{\mathbf{E}}(\mathbf{r},\omega)/(\mu_0\omega^2)$. The second term on the right-hand side of Eq. (10) vanishes by virtue of the constraint (8). This means that we are left with a relation of the form

$$\left[\left[\hat{\mathbf{P}}_{NL}(\mathbf{r}),\hat{H}_L\right],\hat{H}_L\right]=-\left[\left[\hat{\mathbf{D}}_L(\mathbf{r}),\hat{H}_{NL}\right],\hat{H}_L\right]. \tag{11}$$

As has been shown explicitly in [17], the general solution to it is just the single-commutator relation

$$\left[\hat{\mathbf{P}}_{NL}(\mathbf{r}),\hat{H}_L\right] = -\left[\hat{\mathbf{D}}_L(\mathbf{r}),\hat{H}_{NL}\right] \tag{12}$$

as all commutants with $\hat{H}_L$ that are not included in (12) have to strictly vanish.

Since the frequency components of $\hat{\mathbf{D}}_L(\mathbf{r},\omega)$ are composed of a reactive term, $\varepsilon_0\varepsilon(\mathbf{r},\omega)\hat{\mathbf{E}}(\mathbf{r},\omega)$, and a noise contribution, $\hat{\mathbf{P}}_L^{(N)}(\mathbf{r},\omega)$, by the structure of Eq. (12) the same is true for the nonlinear polarization. Hence, in this way we have derived a nonlinear noise polarization which is a solution to $[\hat{\mathbf{P}}_{NL}^{(N)}(\mathbf{r}),\hat{H}_L] = -[\hat{\mathbf{P}}_L^{(N)}(\mathbf{r}),\hat{H}_{NL}]$. Equation (12) can be solved by using well-known techniques for inverting Liouvillian superoperators to obtain for the positive-frequency component [16, 17]

$$\hat{P}_{NL,l}^{(++)}(\mathbf{r}) = \frac{1}{i\hbar}\sqrt{\frac{\hbar\varepsilon_0}{\pi}}\int d\mathbf{1}d\mathbf{2}d\mathbf{3}\frac{\sqrt{\varepsilon_I(\mathbf{1})}}{\omega_2+\omega_3}\alpha_{mjk}(\mathbf{1},\mathbf{2},\mathbf{3}) \\ \times\frac{\omega^2}{c^2}\varepsilon(\mathbf{r},\omega)G_{lm}(\mathbf{r},\mathbf{1})\hat{f}_j(\mathbf{2})\hat{f}_k(\mathbf{3}) + \hat{P}_{NL,l}^{(N,++)}(\mathbf{r}) \tag{13}$$

with the nonlinear noise polarization

$$\hat{P}_{NL,l}^{(N,++)}(\mathbf{r}) = \frac{1}{i\hbar}\sqrt{\frac{\hbar\varepsilon_0}{\pi}}\int d\mathbf{1}d\mathbf{2}d\mathbf{3}\frac{\sqrt{\varepsilon_I(\mathbf{1})}}{\omega_2+\omega_3}\alpha_{ljk}(\mathbf{1},\mathbf{2},\mathbf{3})\delta(\mathbf{r}-\mathbf{s})\hat{f}_j(\mathbf{2})\hat{f}_k(\mathbf{3}). \tag{14}$$

Equation (13) has to be compared with the expressions known from standard nonlinear optics. The nonlinear polarization is defined in the framework of response theory as

$$P_{NL,l}(\mathbf{r},t) = \varepsilon_0\int_{-\infty}^{t} d\tau_1 d\tau_2\,\chi_{lmn}^{(2)}(\mathbf{r},t-\tau_1,t-\tau_2) \\ \times\, E_m(\mathbf{r},\tau_1)E_n(\mathbf{r},\tau_2) + P_{NL,l}^{(N)}(\mathbf{r},t), \tag{15}$$

where $P_{NL,l}^{(N)}(\mathbf{r},t)$ is the nonlinear noise polarization that is commonly disregarded in classical nonlinear optics. However, because the validity of the approximate interaction Hamiltonian (7), as described in Sec. 3, is restricted to certain frequency regions far away from any atomic resonances, we can treat the electric field in the slowly-varying amplitude approximation and expand it in a set of non-overlapping functions centered at the mid-frequencies Ω_i. If we perform this approximation for the process of second-harmonic generation with $\Omega_1=\Omega_2+\Omega_3$, we can write Eq. (15) as

$$P_{NL,l}^{(++)}(\mathbf{r},\Omega_1) = \varepsilon_0\chi_{lmn}^{(2)}(\mathbf{r},\Omega_2,\Omega_3)E_m(\mathbf{r},\Omega_2)E_n(\mathbf{r},\Omega_3) + P_{NL,l}^{(N,++)}(\mathbf{r},\Omega_1), \tag{16}$$

where all the appearing quantities are assumed to be slowly varying with Ω_i.

At the same time, for consistency, the slowly-varying amplitude approximation has to be made for the dynamical variables, details of which can be found in [16, 17]. So equipped, we can find a linear functional relation between the coupling tensor $\alpha_{mjk}(\mathbf{r},\Omega_1,\mathrm{s}_2,\Omega_2,\mathrm{s}_3,\Omega_3)$ in Eq. (13) and the nonlinear susceptibility tensor $\chi^{(2)}_{lmn}(\mathbf{r},\Omega_2,\Omega_3)$ in Eq. (16) [16,17]. Inserting this relation into Eq. (14) yields the sought expression for the nonlinear noise polarization in terms of the nonlinear susceptibility as

$$P_{NL,l}(\mathbf{r},\Omega_1) = \frac{\varepsilon_0 c^2}{\Omega_1^2} H_{li}(\mathbf{r},\Omega_1)\left[\frac{\chi^{(2)}_{imn}(\mathbf{r},\Omega_2,\Omega_3)}{\varepsilon(\mathbf{r},\Omega_1)} E_m(\mathbf{r},\Omega_2)E_n(\mathbf{r},\Omega_3)\right], \qquad (17)$$

where $H_{li}(\mathbf{r},\Omega_1) = \partial_l\partial_i - \delta_{li}\Delta - \Omega_1^2/c^2\varepsilon(\mathbf{r},\Omega_1)\delta_{li}$ is the Helmholtz differential operator, i.e. the inverse of the Green tensor.

Equation (17) puts us in the situation to estimate the strength of the nonlinear noise. Disregarding the vector character of the quantities appearing in it and neglecting the precise frequency dependence of the relevant quantities, we can estimate the order of magnitude of the nonlinear noise relative to the linear noise as $|P_{NL}/P_L| \sim |\chi^{(2)}/\varepsilon||E|$, where $|E|$ denotes the strength of the pump field. Similarly, for higher-order nonlinearities we would obtain $|P^{(n)}_{NL}/P_L| \sim |\chi^{(n)}/\varepsilon||E|^{n-1}$. These results state that for strong pumping the nonlinear noise polarization can indeed become important and is not to be neglected.

5. Conclusions

We have shown how to quantize the electromagnetic field in the presence of nonlinearly responding, absorbing dielectric materials. Based on the theory of QED in linear dielectrics which is proven to be consistent with quantum-theoretical and statistical requirements, we have constructed an effective nonlinear interaction Hamiltonian from which a nonlinear polarization could be derived. This nonlinear polarization automatically includes a contribution associated with nonlinear noise which is commonly disregarded in nonlinear optics. We could show that the strength of this nonlinear noise grows monotonically with the strength of the pump field and may ultimately limit the performance of nonlinear quantum optical processes.

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References

1. E.A. Power and S. Zienau, *Phil. Trans. R. Soc. A* **251** (1959) 427.
2. R.G. Woolley, *Phil. Trans. R. Soc. A* **321** (1971) 557.
3. T. Gruner and D.-G. Welsch, *Phys. Rev. A* **53** (1996) 1661.
4. A. Tip, *Phys. Rev. A* **57** (1998) 4818.
5. T.D. Ho, L. Knöll and D.-G. Welsch, *Phys. Rev. A* **57** (1998) 3931.

6. S. Scheel, L. Knöll and D.-G. Welsch, *Phys. Rev. A* **58** (1998) 700.
7. O. DiStefano, S. Savasta and R. Girlanda, *Phys. Rev. A* **60** (1999) 1614.
8. L. Knöll, S. Scheel and D.-G. Welsch, *Coherence and Statistics of Photons and Atoms*, ed. J. Peřina, Wiley, New York, 2001.
9. N. Bloembergen, *Nonlinear Optics*, Benjamin, New York, 1965.
10. B. Huttner and S.M. Barnett, *Phys. Rev. A* **46** (1992) 4306.
11. A. Bechler, *J. Mod. Opt.* **46** (1999) 901.
12. M. Wubs and L.G. Suttorp, *Phys. Rev. A* **70** (2004) 013816.
13. E. Wang and U. Heinz, *Phys. Rev. D* **66** (2002) 025008.
14. J.J. Hopfield, *Phys. Rev.* **112** (1958) 1555.
15. W. Vogel and D.-G. Welsch, *Quantum Optics*, Wiley-VCH, Weinheim, 2006.
16. S. Scheel and D.-G. Welsch, *Phys. Rev. Lett.* **96** (2006) 073601.
17. S. Scheel and D.-G. Welsch, *J. Phys. B: At. Mol. Opt. Phys.* **39** (2006) S711.