Radiation field quantization in a nonlinear dielectric with dispersion and absorption

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Abstract

The problem of quantizing the radiation field inside a nonlinear dielectric is studied. Based on the quantization of radiation in a linear dielectric which includes absorption and dispersion, we extend the theory in order to treat also nonlinear optical processes. We derive propagation equations in space and time for the quantized radiation field including the effects of linear absorption and dispersion as well as nonlinear optical effects. As a special case we derive the propagation equation of a narrow-frequency band light pulse in a Kerr medium.

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I. INTRODUCTION

Non-classical properties of quantum light pulses propagating through nonlinear optical media are of increasing interest. E.g. quantum solitons in fibers may in future play an important role in communication technology [1]. But the proper consideration of quantum effects as well as changes of quantum properties of light pulses propagating through nonlinear optical media requires an adequate description of the quantized radiation field in such media. In addition to the nonlinear optical properties of the dielectric under consideration, also effects of linear absorption and dispersion must be included. As it is well known the existence of solitons in fibers with a Kerr-like nonlinearity requires also the dispersion of the group velocity of the wave [4,3]. On the other hand non-classical phenomena such as squeezing are very sensitive to degradation effects as for instance absorption of radiation. Therefore it is necessary, to develop a theory of the quantized radiation field in dielectrics which considers both nonlinear optical effects and dispersion as well as absorption effects. The quantization of the radiation field in dielectrics has been performed in the past in different ways. Some of this treatments are presented in [4–21]. The quantization of the electromagnetic field in linear, lossless, and dispersionless dielectric media has been developed in [5–10]. The case of lossless and dispersionless nonlinear dielectrics has been treated by Hillery and Mlodinov in [11]. Squeezing of solitons in dispersive nonlinear dielectrics was successfully explained in [12]. A canonical quantization schema for a general dispersive nonlinear dielectric has been presented by Drummond [15]. The methods of the phase-space formulation of the equations for quantum optical pulse propagation in nonlinear and dispersive single-mode fibers are discussed in [22]. A very interesting approach to the quantization of the radiation field in a dielectric composed of two-level atoms has been recently proposed in [21].

Huttner and Barnett [16] presented a canonical quantization scheme for the radiation field in a linear dielectric, employing the Hopfield model of the dielectric [1]. It is worth noting that in this approach both the dispersion and the absorption by the medium are taken into account in a quantum-mechanically consistent way. A phenomenological generalization
of this theory was given in \[18–20,23\], and the theory has been applied to the propagation of quantum pulses in \[24\]. In this paper we develop a theory of the quantized radiation field in dielectrics which includes both nonlinear optical effects and dispersion as well as absorption effects. We employ the quantization scheme developed by Huttner and Barnett \[16\]. This quantization schema is based on a microscopic model of Hopfield type \[4\], where the dielectric medium is represented by a polarization field \(X(x)\). We extend this quantization scheme by introducing nonlinear polarization field terms into the Hamiltonian. In reality the material system is not a linear one, and these additional terms describe the intrinsic nonlinear properties of the dielectric which can give rise to nonlinear optical effects. In Sec.II we review the main results of the quantization scheme for a linear dielectric described in \[16,20\]. In Sec.III the theory is extended to nonlinear dielectrics, and the quantization schema in this case is described, including linear dispersion and absorption. A propagation equation for a narrow-frequency band quantum pulse is derived in Sec.IV and the special case of a Kerr-like nonlinearity is considered in more detail. A summary is given in Sec.V.

II. LINEAR MEDIUM

In this section we review some results of the quantization of the electromagnetic field in a dispersive and absorptive linear dielectric. We employ the quantization scheme developed by Huttner and Barnett \[16\] and extended in \[20\]. This quantization schema is based on a microscopic model of Hopfield type \[4\] where the dielectric medium is represented by a polarization field \(X(x)\), which is coupled to a reservoir composed of a continuum of harmonic oscillators \(Y(x,\omega)\) to allow for absorption \((0 < \omega < \infty\), the continuum of bath modes at a fixed space point \(x\) is labeled by the parameter \(\omega\)). For convenience we consider only a one-dimensional model of the dielectric, the propagation direction of the radiation field is the x-direction and the vector potential \(A(x)\) is linear polarized in the z-direction. The electromagnetic field is coupled to the polarization field in the minimal coupling version, and the resulting Hamiltonian can be diagonalized introducing bosonic operators \(\hat{f}(x,\omega)\)
and \( \hat{f}^{\dagger}(x, \omega) \), which describe polariton-like excitations inside the dielectric \([16,20]\). After diagonalization the Hamiltonian of the transverse electromagnetic field interacting with the linear dielectric can be written as

\[
\hat{\mathbf{H}} = \hbar \hat{\mathbf{H}}_L = \hbar \int_{-\infty}^{\infty} dx \, \hat{H}_L(x),
\]

where

\[
\hat{H}_L(x) = \int_{0}^{\infty} d\omega \, \omega \, \hat{f}^{\dagger}(x, \omega) \, \hat{f}(x, \omega).
\]

The creation and destruction operators \( \hat{f}^{\dagger}(x, \omega) \) and \( \hat{f}(x, \omega) \) satisfy the familiar bosonic equal time commutation relations

\[
\left[ \hat{f}(x, \omega), \hat{f}^{\dagger}(x', \omega') \right] = \delta(x-x') \, \delta(\omega-\omega'),
\]

\[
\left[ \hat{f}(x, \omega), \hat{f}(x', \omega') \right] = 0.
\]

The operators of the electromagnetic field, e.g. the vector potential \( \hat{\mathbf{A}}(x) \) and the electric field strength \( \hat{\mathbf{E}}(x) \) as well as the polarization field operator \( \hat{\mathbf{X}}(x) \) and the bath operators \( \hat{\mathbf{Y}}(x, \omega) \) may be written in terms of the bosonic operators \( \hat{f}^{\dagger}(x, \omega) \) and \( \hat{f}(x, \omega) \) as follows

\[
\hat{\mathbf{A}}(x) = \hat{\mathbf{A}}^{(+)}(x) + \hat{\mathbf{A}}^{(-)}(x) = \int_{0}^{\infty} d\omega \left[ \hat{\mathbf{A}}(x, \omega) + \hat{\mathbf{A}}^{\dagger}(x, \omega) \right],
\]

\[
\hat{\mathbf{E}}(x) = \hat{\mathbf{E}}^{(+)}(x) + \hat{\mathbf{E}}^{(-)}(x) = \int_{0}^{\infty} d\omega \left[ \hat{\mathbf{E}}(x, \omega) + \hat{\mathbf{E}}^{\dagger}(x, \omega) \right],
\]

\[
\hat{\mathbf{X}}(x) = \hat{\mathbf{X}}^{(+)}(x) + \hat{\mathbf{X}}^{(-)}(x) = \int_{0}^{\infty} d\omega \left[ \hat{\mathbf{X}}(x, \omega) + \hat{\mathbf{X}}^{\dagger}(x, \omega) \right],
\]

where

\[
\hat{\mathbf{A}}(x, \omega) = \int_{-\infty}^{\infty} dx' \, G_{\mathbf{A}}(x, x', \omega) \, \hat{f}(x', \omega),
\]

\[
\hat{\mathbf{E}}(x, \omega) = i\omega \hat{\mathbf{A}}(x, \omega),
\]
\[
\frac{\rho}{\epsilon_0} \hat{X}(x, \omega) = (\varepsilon(\omega) - 1) \hat{E}(x, \omega) - 2i\alpha c \sqrt{\varepsilon(\omega)} \hat{f}(x, \omega). \tag{10}
\]

The Green function \(G_A(x, x', \omega)\) is given by
\[
G_A(x, x', \omega) = -i\alpha \sqrt{\frac{\varepsilon(\omega)}{\varepsilon(\omega)}} e^{ik(\omega)|x-x'|}, \tag{11}
\]
where \(\varepsilon(\omega) = \varepsilon_r(\omega) + i\varepsilon_i(\omega)\) denotes the complex permittivity of the dielectric. The complex wave number \(k(\omega)\) is related to the complex permittivity \(\varepsilon(\omega)\) and the complex index of refraction \(n(\omega)\) by
\[
k(\omega) = \frac{\omega}{c} \sqrt{\varepsilon(\omega)} = \frac{\omega}{c} n(\omega) = \frac{\omega}{c} (n_r(\omega) + n_i(\omega)) = k_r + ik_i. \tag{12}
\]

A normalization constant \(\alpha\) has been introduced for convenience
\[
\alpha = \sqrt{\frac{\hbar}{4\pi c^2 \epsilon_0 A}}, \tag{13}
\]
where \(\epsilon_0\) is vacuum permittivity constant, and \(A\) is the normalization area perpendicular to the \(x\) direction. The coefficient \(\rho\) denotes the coupling constant between the polarization field \(\hat{X}(x)\) and the electromagnetic field. It is worth noting that the polarization field operator \(\hat{X}(x, \omega)\) may be expressed as a sum of the electric field operator \(\hat{E}(x, \omega)\) multiplied by \((\varepsilon(\omega) - 1)\) and the bosonic operator \(\hat{f}(x, \omega)\) multiplied by the square root of the imaginary part of the complex permittivity, which describes absorption in the dielectric. Explicit expression for the complex permittivity \(\varepsilon(\omega)\) can be found in [16], but these are not of relevance for our task, and any phenomenologically introduced expression consistent with the Kramers-Kronig relations can be used. Also the explicit expression of the reservoir field operators \(\hat{Y}(x, \omega)\) in terms of the bosonic operators \(\hat{f}(x, \omega)\) are not needed in the following.

Using the commutation relations (3,4) it can be shown [16,20] that the field operators \(\hat{A}(x)\) and \(\hat{E}(x)\) and the polarization field operators \(\hat{X}(x)\) satisfy the well known canonical commutation relations
\[
[\hat{A}(x), \hat{E}(x')] = -\frac{i\hbar}{\epsilon_0 A} \delta(x-x'), \tag{14}
\]

where \(\epsilon_0\) is vacuum permittivity constant, and \(A\) is the normalization area perpendicular to the \(x\) direction. The coefficient \(\rho\) denotes the coupling constant between the polarization field \(\hat{X}(x)\) and the electromagnetic field. It is worth noting that the polarization field operator \(\hat{X}(x, \omega)\) may be expressed as a sum of the electric field operator \(\hat{E}(x, \omega)\) multiplied by \((\varepsilon(\omega) - 1)\) and the bosonic operator \(\hat{f}(x, \omega)\) multiplied by the square root of the imaginary part of the complex permittivity, which describes absorption in the dielectric. Explicit expression for the complex permittivity \(\varepsilon(\omega)\) can be found in [16], but these are not of relevance for our task, and any phenomenologically introduced expression consistent with the Kramers-Kronig relations can be used. Also the explicit expression of the reservoir field operators \(\hat{Y}(x, \omega)\) in terms of the bosonic operators \(\hat{f}(x, \omega)\) are not needed in the following.

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\[
[\hat{A}(x), \hat{E}(x')] = -\frac{i\hbar}{\epsilon_0 A} \delta(x-x'), \tag{14}
\]
\[
\left[ \hat{X}(x), \hat{E}(x') \right] = 0, \tag{15}
\]

\[
\left[ \hat{X}(x), \hat{A}(x') \right] = 0. \tag{16}
\]

Using the Green function (11), Eq. (8) may be written as

\[
\hat{A}(x, \omega) = \hat{A}_{(-\rightarrow)}(x, \omega) + \hat{A}_{(\leftarrow)}(x, \omega) \tag{17}
\]

where

\[
\hat{A}_{(-\rightarrow)}(x, \omega) = \int_{-\infty}^{x} dx' \ G_A(x, x', \omega) \hat{f}(x', \omega) = -i\alpha \sqrt{\frac{\xi_1}{\varepsilon}} \int_{-\infty}^{x} dy \ e^{ik(x-y)} \hat{f}(y, \omega) \tag{18}
\]

and

\[
\hat{A}_{(\leftarrow)}(x, \omega) = \int_{x}^{\infty} dx' \ G_A(x, x', \omega) \hat{f}(x', \omega) = -i\alpha \sqrt{\frac{\xi_1}{\varepsilon}} \int_{x}^{\infty} dy \ e^{-ik(x-y)} \hat{f}(y, \omega) \tag{19}
\]

denote vector potential operator components describing wave propagation in the positive and negative x-direction respectively. From Eqs. (18,19) we easily find

\[
\partial_x \hat{A}_{(-\rightarrow)}(x, \omega) = ik \hat{A}_{(-\rightarrow)}(x, \omega) - i\alpha \sqrt{\frac{\xi_1}{\varepsilon}} \hat{f}(x, \omega), \tag{20}
\]

\[
\partial_x \hat{A}_{(\leftarrow)}(x, \omega) = -ik \hat{A}_{(\leftarrow)}(x, \omega) + i\alpha \sqrt{\frac{\xi_1}{\varepsilon}} \hat{f}(x, \omega). \tag{21}
\]

These relations can be considered as quantum Langevin equations governing the spatial evolution of the vector potential operator components $\hat{A}_{(-\rightarrow)}(x, \omega)$ and $\hat{A}_{(\leftarrow)}(x, \omega)$. Here the operators $\hat{f}(x, \omega)$ play the role of the Langevin force operators. Note that these noise terms are proportional to $\sqrt{\varepsilon_i}$ which is responsible for the absorption.

A similar decomposition as in Eq. (17) can be performed for the electric field operator components, where now the Green function $G_E(x, x', \omega) = i\omega G_A(x, x', \omega)$ has to be used.

Finally we consider the time evolution of the operators in the Heisenberg picture. We immediately find from Eqs. (18,19)

\[
i\partial_t \hat{f}(x, \omega) = \left[ \hat{f}(x, \omega), \hat{H}_L \right] = \omega \hat{f}(x, \omega) \tag{22}
\]
and similar equations for the other field operators, which are connected with \( \hat{f}(x, \omega) \). E.g. using Eq.\((18)\) we have

\[
i \partial_t \hat{A}(\rightarrow)(x, \omega) = \left[ \hat{A}(\rightarrow)(x, \omega), \hat{H}_L \right] = \omega \hat{A}(\rightarrow)(x, \omega).
\]

It is seen from Eqs. \((22, 23)\) that the operators show a harmonic time evolution in the case of a linear dielectric:

\[
\hat{f}(x, \omega, t_0 + t) = e^{-i\omega t} \hat{f}(x, \omega, t_0),
\]

\[
\hat{A}(\rightarrow)(x, \omega, t_0 + t) = e^{-i\omega t} \hat{A}(\rightarrow)(x, \omega, t_0),
\]

and \( \omega \) can be associated with the frequency of the harmonic time evolution of all the operators with the argument \( \omega \). Thus, in case of a linear dielectric the decomposition in Eq.\((5)\) can be considered as a Fourier decomposition of the operator of the vector potential in the Heisenberg picture.

Concluding this section we state that the formulae given above enables one to study quantum properties of light fields propagating in a linear dielectric with absorption and dispersion \[24\] or passing through a multi-layer structure \[23\].

III. NONLINEAR CASE

In this section we extend the linear model of a dielectric in order to include non-linear optical effects. This offers the possibility for studying the propagation of quantum light fields in nonlinear optical media including dispersion and absorption. In the Hopfield model \[4\] the dielectric is described by a polarization field, the equations of motion for this field being linear. In reality the material system is not composed of harmonic oscillators and so this description is only an approximate one. In order to introduce into the Hopfield-model the really existing nonlinearities of the dielectric we add to the Lagrangian of the material system an arbitrary functional \(-\Phi[X(x)]\) of the polarization field \(X(x)\). This term does not
affect the definition of the canonical momenta conjugated to the field variables. Therefore the whole quantization scheme developed for the linear dielectric can be used also in the case of the nonlinear dielectric. The Hamiltonian describing the nonlinear dielectric interacting with the radiation field then reads as

$$\hat{H} = \hbar \left( \hat{H}_L + \hat{H}_N \right) = \hbar \int_{-\infty}^{\infty} dx \left( \hat{H}_L(x) + \hat{H}_N(x) \right),$$

(26)

where $\hat{H}_L(x)$ is the linear part of the Hamiltonian density which is defined in Eq.(2), and $\hat{H}_N(x)$ describes the intrinsic nonlinearity of the dielectric,

$$\hat{H}_N(x) = \Phi[\hat{X}(x)].$$

(27)

We emphasize that all the equations (2) – (21) between the electromagnetic field operators, the polarization field, and the bosonic creation and destruction operators $\hat{f}^\dagger(x, \omega)$ and $\hat{f}(x, \omega)$ are valid also in the case of the nonlinear dielectric interacting with the electromagnetic field. These equations have been derived in the case of the linear dielectric by the diagonalization of the linear Hamiltonian [16,20], but without using the Heisenberg equations of motion for the corresponding operators. Therefore in the case of the nonlinear dielectric described by the Hamiltonian (26,27), the same procedure can be applied. But with respect to the time development in the Heisenberg picture instead of Eqs.(22,23) now we find

$$i \partial_t \hat{f}(x, \omega) = \left[ \hat{f}(x, \omega), \hat{H}_L + \hat{H}_N \right] = \omega \hat{f}(x, \omega) + \left[ \hat{f}(x, \omega), \hat{H}_N \right],$$

(28)

$$i \partial_t \hat{A}_{(\rightarrow)}(x, \omega) = \left[ \hat{A}_{(\rightarrow)}(x, \omega), \hat{H}_L + \hat{H}_N \right] = \omega \hat{A}_{(\rightarrow)}(x, \omega) + \left[ \hat{A}_{(\rightarrow)}(x, \omega), \hat{H}_N \right].$$

(29)

These equations of motion, together with the quantum Langevin equations (20,21) governing the spatial evolution of the vector potential operator components describe the space time behavior of the radiation field in a non-linear, dispersive and absorbing dielectric. These equations have to be supplemented by the relation (10) between the polarization field operator $\hat{X}(x, \omega)$, the operator of the electric field $\hat{E}(x, \omega)$ and the bosonic operator $\hat{f}(x, \omega)$. 

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Using this equation we can express the non-linear part of the Hamiltonian completely in terms of \( \hat{E}(x, \omega) \) and \( \hat{f}(x, \omega) \) and their Hermitian conjugate operators. As a consequence Eqs. (28,29) include nonlinear optical effects in the time development of the radiation field. As can be seen from the Heisenberg equations of motion (28,29) due to the nonlinear part of the Hamiltonian the time development of the corresponding operators is much more complicated as in the linear case. Therefore the decomposition Eq. (5) now cannot be considered as a Fourier decomposition of the operator of the vector potential in the Heisenberg picture, because these components show a very complicated time behavior, and the parameter \( \omega \) cannot be associated with the frequency in the Fourier space.

Finally we would like to note that the diagonalization procedure remains the same also in case of a more general nonlinear part \( \hat{H}_N(x) \) of the Hamiltonian, which may be a functional of the operators \( \hat{A}(x), \hat{X}(x), \hat{Y}(x, \omega) \) and its spacial derivatives (see e.g. [21]), and the Eqs. (4) – (21) as well as the Heisenberg equations of motion (28,29) may be used in order to investigate the radiation field inside the nonlinear dielectric. This offers the possibility to generalize the Hopfield model if necessary.

**IV. PULSE PROPAGATION EQUATION.**

In order to study the propagation of quantum pulses in nonlinear dispersive and absorptive dielectrics we have to solve the nonlinear propagation equations in time (28,29) together with the quantum Langevin equations (20,21) governing the spatial evolution. In this section we shall consider these equations in more detail in the case of narrow-frequency band quantum pulses, and eventually for a Kerr medium we derive the quantum version of the soliton equation including also the effect of absorption.

To begin with we divide the \( \omega \)-interval \([0.. \infty]\) into subintervals and decompose the field operators as follows:

\[
\hat{A}^{(+)}(x) = \sum_{i=0}^{\infty} \hat{A}_i^{(+)}(x),
\]  

(30)
\begin{align}
\hat{A}^{(+)}(x) &= \frac{\Delta \omega}{2} \int_{-\Delta \omega/2}^{\Delta \omega/2} d\Omega \, \hat{A}(x, \omega^i + \Omega), \\
\text{where} \quad \omega^i &= (i + 1/2) \Delta \omega.
\end{align}

The analogous relations can be written for all the other operators which can be expressed by an integral over \( \omega \). Now we assume that the light pulse under consideration has a small spectral bandwidth, so that only components of the field operator in one of the intervals, characterized by an index \( i_0 \), must be considered. It means that the bandwidth should not exceed the length of the corresponding subinterval \( \Delta \omega \). This also gives a criterion for choosing the value of \( \Delta \omega \). In the following we restrict the parameter \( \omega \) to this interval, and we write

\[ \omega = \omega_0 + \Omega \]

where \( \omega_0 = \omega^{i_0} \). Next we introduce slowly varying operators \( \hat{A}(\rightarrow)(x, \Omega) \) (operators corresponding to the other direction can be introduced in a similar way) and \( \hat{f}(\rightarrow)(x, \Omega) \) according to

\begin{align}
\hat{A}(\rightarrow)(x, \omega) &= e^{i\varphi(\rightarrow)} \hat{A}(\rightarrow)(x, \Omega), \\
\hat{f}(x, \omega) &= e^{i\varphi(\rightarrow)} \hat{f}(\rightarrow)(x, \Omega)
\end{align}

(the index \( i_0 \) will be suppressed here and in the following), where

\[ \varphi(\rightarrow) = k_{\varphi} x - \omega_0 t, \]

and \( k_{\varphi} \) is a real number to be determined later.

Using the Heisenberg equations of motion \((28, 29)\) we find

\begin{align}
\Omega \, \hat{A}(\rightarrow)(x, \Omega) &= (i \partial_t + \mathcal{H}_N^\times) \hat{A}(\rightarrow)(x, \Omega), \\
\Omega \, \hat{f}(\rightarrow)(x, \Omega) &= (i \partial_t + \mathcal{H}_N^\times) \hat{f}(\rightarrow)(x, \Omega),
\end{align}
where the notation
\[
\hat{H}_N^x \hat{O} \equiv \left[ \hat{H}_N, \hat{O} \right]
\]
has been introduced. We rewrite also the quantum Langevin equation (20) in terms of the new slowly varying operators (Eqs. (34)):
\[
\partial_x \hat{A}^{(\rightarrow)}(x, \Omega) = i(k - k_\varphi) \hat{A}^{(\rightarrow)}(x, \Omega) - i\alpha \sqrt{\frac{\varepsilon}{\varepsilon_i}} \hat{f}^{(\rightarrow)}(x, \Omega) + R.S. \quad (39)
\]
To be able to treat these equations (36,37,39) in more detail we expand the wave number \( k \) as a function of \( \omega \) near \( \omega_0 \) into a Taylor series
\[
k = \sum_{m=0}^{\infty} k_m \Omega^m m!,
\]
and in a similar way we have
\[
\sqrt{\frac{\varepsilon_i}{\varepsilon}} = \sum_{m=0}^{\infty} p_m \Omega^m m!,
\]
where the coefficients \( k_m \equiv k_{mr} + ik_{mi} \) and \( p_m \equiv p_{mr} + ip_{mi} \) are in general complex. Substituting these expansions into Eq.(39) and employing Eqs.(36,37), we arrive at
\[
\partial_x \hat{A}^{(\rightarrow)}(x, \Omega) = i \left( \sum_{m=0}^{\infty} \frac{k_m}{m!} \left( i\partial_t + \hat{H}_N^x \right)^m - k_\varphi \right) \hat{A}^{(\rightarrow)}(x, \Omega) - i\alpha \sum_{m=0}^{\infty} \frac{p_m}{m!} \left( i\partial_t + \hat{H}_N^x \right)^m \hat{f}^{(\rightarrow)}(x, \Omega) + R.S. \quad (42)
\]
Integration over \( \Omega \) gives
\[
\partial_x \hat{A}^{(+)}(x) = i \left( \sum_{m=0}^{\infty} \frac{k_m}{m!} \left( i\partial_t + \hat{H}_N^x \right)^m - k_\varphi \right) \hat{A}^{(\rightarrow)}(x) + R.S. \quad (43)
\]
where \( \hat{A}^{(\rightarrow)}(x) \) is given by the relations
\[
\hat{A}^{(+)}(x) = \hat{A}^{(\rightarrow)}(x) + \hat{A}^{(\leftarrow)}(x), \\
\hat{A}^{(\rightarrow)}(x) = e^{i\varphi(x)} \hat{A}^{(\leftarrow)}(x) \quad (44)
\]
and

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\[
\text{R.S.} = -i\alpha \sum_{m=0}^{\infty} \frac{p_m}{m!} (i\partial_t + \hat{H}_N^\times)^m \int_{-\Delta\omega/2}^{\Delta\omega/2} d\Omega \hat{f}_{\rightarrow}(x,\Omega).
\] (45)

Note that the various operators \( \hat{A} \) and \( \hat{f} \) should be labeled by the index \( i_0 \), describing the parameter range to which these operators belong (see e.g. Eq.(31)). The equations (43 – 45) describe the propagation of a quantum pulse with a narrow bandwidth spectrum in the dielectric with any order of dispersion, absorption and nonlinearity. In general we need similar equations for the fields running in the other direction because they are included in \( \hat{H}_N \). It should be mentioned that the restriction to a pulse with narrow spectral bandwidth is not necessary, but in the general case we must consider for each \( \omega \)-interval such an equation as Eq.(43).

Let us consider a special case with small dispersion, where the absorption in the frequency interval around \( \omega_{i_0} \) can be neglected \( (\varepsilon_i(\omega) = \text{Im}(\varepsilon(\omega)) = 0, k_{mi} = 0, k_{1r} = 1/v_{gr}) \), where \( v_{gr} \) is the group velocity, and \( k_{mr} = 0 \) for \( m \geq 2 \). Choosing \( k_\varphi = k_{0r} \) we arrive at the simplest case of a propagation equation for a quantum light pulse in a nonlinear dielectric,

\[
\left[ \partial_x + \frac{1}{v_{gr}} \partial_t \right] \hat{A}_{\rightarrow}^{(+)}(x) = \frac{i}{c} \left[ \hat{H}_N, \hat{A}_{\rightarrow}^{(+)}(x) \right].
\] (46)

Note that the assumption of zero absorption resulted in the vanishing of the term proportional to the operator \( \hat{f}_{\rightarrow}(x,\Omega) \), which may be considered as a Langevin fluctuation operator. Such operators have to be included in the Heisenberg equation of motion in order to describe correctly also losses.

### A. Second order dispersion approximation.

Now we consider the propagation of a quantum pulse with narrow spectral bandwidth including to some extend dispersion and absorption. We neglect all terms proportional to \( \Omega^m \) with \( m > 2 \). That means we take into account the dispersion only up to the second order. Moreover we neglect all terms in which the nonlinear Hamiltonian \( \hat{H}_N \) and the Langevin fluctuation operators \( \hat{f}_{\rightarrow}(x,\Omega) \) are coupled with higher order dispersion and absorption
effects (this means terms proportional to $k_m \hat{H}_N^x$, $m \geq 2$ and $p_m \hat{f}_{(\rightarrow)} (x, \Omega)$, $m \geq 1$). Choosing $k_{\varphi} = k_{0r}$, from Eq.(13) we arrive at the following propagation equation for a quantum light pulse

$$
\left( i \partial_x + i k_{0i} + i k_1 \partial_t - 1/2 k_2 \partial_{tt} + k_1 \hat{H}_N^x \right) \hat{\Delta}^{(+)} (x) \Delta \frac{\Delta \omega}{2} = \alpha p_0 \int d\Omega \hat{f}_{(\rightarrow)} (x, \Omega), \quad (47)
$$

which will be discussed in connection with a Kerr medium in the next subsection.

**B. Approximation for the propagation in a Kerr medium.**

In this section as an application of Eq.(17) we discuss the pulse propagation in a Kerr medium. At first we specify the nonlinear part of the Hamiltonian $\hat{H}_N$. The most simple extension of the linear dielectric to include nonlinearities is in case of a nonlinear dielectric with inversion symmetry the Ansatz

$$
\Phi[\hat{X} (x)] = \frac{\lambda}{4!} \hat{X}^4 (x), \quad (48)
$$

from which we find (see Eq.(27))

$$
\hat{H}_N = \frac{\lambda}{4!} \int_{-\infty}^{\infty} dx' \hat{X}^4 (x'), \quad (49)
$$

where $\lambda$ is a constant characterizing the nonlinearity in a homogeneous dielectric. In considering the propagation of a quantum pulse with narrow spectral bandwidth into the positive x-direction (see Eq. (17)) we may rewrite $\hat{H}_N$ approximately as

$$
\hat{H}_N = \frac{\lambda}{4} \int_{-\infty}^{\infty} dx' \hat{X}^{(-)} (x') \hat{X}^{(-)} (x') \hat{X}^{(+)} (x') \hat{X}^{(+)} (x'), \quad (50)
$$

where (remember Eq.(33))

$$
\hat{X}^{(+)} (x') = \int \frac{\Delta \omega}{2} d\Omega \left[ i \omega \{ \varepsilon (\omega) - 1 \} \hat{A}_{(\rightarrow)} (x, \Omega) \right.
-2i\alpha c \sqrt{\varepsilon \Omega} \hat{f}_{(\rightarrow)} (x, \Omega), \quad (51)
$$
and the operators $\hat{A}_{(\rightarrow)}(x, \Omega)$ and $\hat{f}_{(\rightarrow)}(x, \Omega)$ are defined in Eq.(34). Here we have also introduced a normal ordering of the operators and quadratic terms in $\hat{X}$ have been neglected. Straightforward calculations give for the commutator term $[\hat{H}_N, \hat{A}_{(\rightarrow)}^{(+)}(x)]$ of Eq. (47)

$$\left[\hat{H}_N, \hat{A}_{(\rightarrow)}^{(+)}(x)\right] = \frac{\lambda}{2} \int_{-\infty}^{\infty} dx' \, G_H(x, x') \hat{X}_{(\rightarrow)}^{(-)}(x') \hat{X}_{(\rightarrow)}^{(+)}(x'),$$

where

$$G_H(x, x') = \alpha^2 \epsilon_0 \rho \int_{-\Delta \Omega/2}^{\Delta \Omega/2} d\Omega \exp\left(i \omega (\epsilon^* - 1) \frac{\epsilon_i}{2k_i |\epsilon|} - 2c \frac{\epsilon_i}{\sqrt{\epsilon}} U(\Delta x)\right) \times \exp \left[-k_i |\Delta x| + i (k_r - k_{0r}) \Delta x\right],$$

$U(\Delta x)$ denotes the unity step function, and $\Delta x = x - x'$. We note that the relation (52) in general describes a nonlocal, nonlinear response of the matter, and Eq. (47) is an integro-differential equation with respect to the space coordinate $x$.

Now we consider the limiting case of weak absorption in the interval around $\omega_0$. Employing the approximation

$$\epsilon_1 \ll \epsilon_r,$$

the term $\frac{\epsilon_i}{2k_i |\epsilon|}$ may be approximated by

$$\frac{\epsilon_i}{2k_i |\epsilon|} \sim 2 \frac{\epsilon_i}{\epsilon_r \sqrt{\epsilon_r} \Im(1 + i \frac{\epsilon_i}{2 \epsilon_r}) \epsilon_r} \sim \frac{1}{k_r},$$

and the term containing the step function can be neglected, because it is proportional to $\epsilon_1$. As a further approximation we expand the integrand of the integral kernel in powers of $\Omega$ and retain only the lowest order terms arriving at

$$G_H(x, x') = \alpha^2 \epsilon_0 \rho i \omega_0 (\epsilon^* - 1) \frac{1}{k_r} \int_{-\Delta \Omega/2}^{\Delta \Omega/2} d\Omega \exp \left[i \Omega \Delta x k_{1r}\right],$$

$$= \alpha^2 \epsilon_0 \rho i \omega_0 (\epsilon^* - 1) \frac{2}{k_r \Delta x k_{1r}} \sin \left[\Delta \omega k_{1r} \Delta x/2\right].$$

Assuming that the operators $\hat{X}_{(\rightarrow)}^{(+)}$ in Eq.(52) are slowly varying on a scale defined by the characteristic length $(\Delta \omega k_{1r})^{-1}$ of $G_H(x, x')$, we can replace the function $G_H(x, x')$ by a delta-function, and the commutator term in Eq.(52) reads as

$$[\hat{H}_N, \hat{A}_{(\rightarrow)}^{(+)}(x)] = \frac{\lambda}{2} \int_{-\infty}^{\infty} dx' \, \delta(\Delta x) \hat{X}_{(\rightarrow)}^{(+)}(x'),$$

and the operators $\hat{A}_{(\rightarrow)}(x, \Omega)$ and $\hat{f}_{(\rightarrow)}(x, \Omega)$ are defined in Eq.(34). Here we have also introduced a normal ordering of the operators and quadratic terms in $\hat{X}$ have been neglected. Straightforward calculations give for the commutator term $[\hat{H}_N, \hat{A}_{(\rightarrow)}^{(+)}(x)]$ of Eq. (47)

$$[\hat{H}_N, \hat{A}_{(\rightarrow)}^{(+)}(x)] = \frac{\lambda}{2} \int_{-\infty}^{\infty} dx' \, G_H(x, x') \hat{X}_{(\rightarrow)}^{(-)}(x') \hat{X}_{(\rightarrow)}^{(+)}(x'),$$

where

$$G_H(x, x') = \alpha^2 \epsilon_0 \rho \int_{-\Delta \Omega/2}^{\Delta \Omega/2} d\Omega \exp\left(i \omega (\epsilon^* - 1) \frac{\epsilon_i}{2k_i |\epsilon|} - 2c \frac{\epsilon_i}{\sqrt{\epsilon}} U(\Delta x)\right) \times \exp \left[-k_i |\Delta x| + i (k_r - k_{0r}) \Delta x\right],$$

$U(\Delta x)$ denotes the unity step function, and $\Delta x = x - x'$. We note that the relation (52) in general describes a nonlocal, nonlinear response of the matter, and Eq. (47) is an integro-differential equation with respect to the space coordinate $x$.

Now we consider the limiting case of weak absorption in the interval around $\omega_0$. Employing the approximation

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and the term containing the step function can be neglected, because it is proportional to $\epsilon_1$. As a further approximation we expand the integrand of the integral kernel in powers of $\Omega$ and retain only the lowest order terms arriving at

$$G_H(x, x') = \alpha^2 \epsilon_0 \rho \omega_0 (\epsilon^* - 1) \frac{1}{k_r} \int_{-\Delta \Omega/2}^{\Delta \Omega/2} d\Omega \exp \left[i \Omega \Delta x k_{1r}\right],$$

$$= \alpha^2 \epsilon_0 \rho \omega_0 (\epsilon^* - 1) \frac{2}{k_r \Delta x k_{1r}} \sin \left[\Delta \omega k_{1r} \Delta x/2\right].$$

Assuming that the operators $\hat{X}_{(\rightarrow)}^{(+)}$ in Eq.(52) are slowly varying on a scale defined by the characteristic length $(\Delta \omega k_{1r})^{-1}$ of $G_H(x, x')$, we can replace the function $G_H(x, x')$ by a delta-function, and the commutator term in Eq.(52) reads as
\[
\left[ \hat{H}_N, \hat{A}^{(+)}_{(\rightarrow)}(x) \right] = \frac{\pi \alpha^2 \lambda \varepsilon_0}{k_1 k_{1r} \rho} i \omega_0 (\varepsilon^* - 1) \hat{X}^{(-)}_{(\rightarrow)}(x) \hat{X}^{(+)}_{(\rightarrow)}(x) \hat{X}^{(+)_{(\rightarrow)}}(x). \tag{57}
\]

Higher order approximations in Eq. (53) give rise to nonlocal corrections in Eq. (52), which will be neglected here. In the same way as we have arrived at the Eq. (17), we neglect in Eq. (57) the Langevin fluctuation operators \( \hat{f}_{(\rightarrow)}(x, \Omega) \), which result from the decomposition of \( \hat{X}^{(+)}_{(\rightarrow)}(x) \) (see Eq. (51)), and we substitute \( k_{1r} \) for \( k_1 \), thus finding eventually

\[
k_{1r} \left[ \hat{H}_N, \hat{A}^{(+)}_{(\rightarrow)}(x) \right] = \chi \hat{A}^{(-)}_{(\rightarrow)}(x) \hat{A}^{(+)}_{(\rightarrow)}(x) \hat{A}^{(+)}_{(\rightarrow)}(x), \tag{58}
\]

where

\[
\chi = \frac{\pi \alpha^2}{k_1} \lambda \left( \frac{\varepsilon_0}{\rho} \omega_0 |\varepsilon - 1| \right)^4. \tag{59}
\]

Substituting this in Eq. (17) we obtain finally the following equation for the propagation of a narrow-frequency band quantum pulse in a Kerr medium with dispersion and weak absorption

\[
\left( i \partial_x + i k_{0i} + i k_1 \partial_t - 1/2 k_2 \partial_{tt} + \chi \hat{A}^{(-)}_{(\rightarrow)}(x) \hat{A}^{(+)}_{(\rightarrow)}(x) \right) \hat{A}^{(+)}_{(\rightarrow)}(x)
= \alpha \sqrt{\frac{\varepsilon_1}{\varepsilon}} \int_{-\Delta \omega/2}^{\Delta \omega/2} d\Omega \hat{f}_{(\rightarrow)}(x, \Omega). \tag{60}
\]

The absorption properties of the dielectric are described by the terms proportional to \( k_{m} \) and the Langevin fluctuation operator on the right hand side of this equation. The term proportional to \( \chi \) describes the Kerr nonlinearity, and the dispersion effects are contained in the term proportional to \( k_{2r} \). Of course the solution of this nonlinear operator equation containing also a Langevin fluctuation operator will be difficult, and numerical methods must be employed.

It is clearly seen that in the case of pure real coefficients \( k_1 \) and \( k_2 \) in Eq. (60) and vanishing \( \varepsilon_1 \) we obtain the operator version of the classical (see e.g. [2,3]) Schrödinger equation similar to the one used in [13,14]. We note that such an approximation is in general not consistent with the Kramers-Kronig relations because dispersion is always accompanied by absorption. Including the absorption brings an additional fluctuation term into the Eq. (60) which is important, in order that the canonical field commutation relations Eq. (14) are preserved.
V. SUMMARY

On the basis of the quantization of the radiation field in a linear dielectric with dispersion and absorption as it has been described by Huttner and Barnett [16] and Gruner and Welsch [20], we have developed a quantization scheme which includes besides linear dispersion and absorption also nonlinear optical effects. This has been achieved by introducing into the matter part of the microscopic Huttner-Barnett model a nonlinear interaction term depending only on the polarization field operator which describes the dielectric. This guarantees, that the operator relations at equal times of the linear theory are also valid in the extended theory. Especially the equal time commutation relations of the free field case are preserved, and the space propagation equations for field components are the same as in the case of a linear dielectric. We have applied the theory to a narrow-frequency band quantum pulse propagating in a dielectric with a Kerr-like nonlinearity. Using a second order dispersion approximation and assuming weak absorption in the frequency band under consideration we have derived in the Heisenberg picture a nonlinear operator equation which governs the space-time development of the quantum pulse. It contains the Kerr nonlinearity, second order dispersion and an absorption term together with a corresponding fluctuation operator. The resulting equation can be considered as a generalization of a nonlinear Schrödinger equation describing quantum solitons.

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