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# QUANTUM THEORY OF LIGHT IN LOSSY NONLINEAR MEDIA[1]

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We present a canonical quantum theory of radiation in nonlinear media taking into account the effects of linear dispersion and absorption in a consistent way. We apply recently developed concepts of the quantization of radiation in dispersive and absorbing linear media and extend the theory in order to include nonlinear optical processes. In particular, the method enables us to systematically derive the noise sources that arise naturally in the nonlinear terms in the space-time propagation equations of the radiation fields.

## 1. Introduction

The study of the propagation of nonclassical light in nonlinear media, such as quantum solitons in Kerr media, has been of increasing interest because of potential applications in optical communication systems [1]. The quantization of light in dielectric media can be performed in different ways. One approach to the problem is to describe the medium phenomenologically by a permittivity (see, e.g., [2]). Another way is to treat the medium microscopically on the basis of appropriately chosen dynamical degrees of freedom (see, e.g., [3, 4]). In what follows we present a microscopic theory and show that the nonlinearities can be incorporated within a fully canonical model including both dispersion and losses. For this purpose we start from the model presented in [3] and extend it to nonlinear media, representing the matter by an *anharmonic* oscillator field. We express the Hamiltonian of the nonlinear system in terms of the eigenoperators of the linear system and derive quantum-mechanically consistent nonlinear evolution equations for the radiation field in which both dispersion and absorption are included. Throughout the paper we freely use results and formulae from references [3, 5] for a

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linear medium. For the sake of brevity we do not fully reproduce the derivations of all the results from this paper but refer the reader there for a complete treatment.

The paper is organized as follows. In Section 2 the Lagrangian and Hamiltonian for the nonlinear dielectric are introduced. Evolution equations for the field operators are derived in Section 3 and it is shown that additional noise sources appear in the nonlinear terms. A summary and concluding remarks are given in Section 4.

## 2. Hamiltonian of the nonlinear system

Our starting point is a nonlinear extension to Hopfield's microscopic model of a dielectric [6, 7]. In the linear model the matter is represented by a harmonic-oscillator polarization field coupled to a continuum of harmonic-oscillator reservoir fields. We extend this by the addition of a nonlinear term, but otherwise follow the procedure given in [3]. There are, however, some important differences when nonlinear media are considered, and these will be emphasized at the appropriate places.

It is usual to begin by writing down a Lagrangian for the system under consideration,

$$L = \int d^3r\, \mathcal{L}(\mathbf{r}). \tag{1}$$

In our case the Lagrangian density $\mathcal{L}(\mathbf{r})$ has the form

$$\mathcal{L}(\mathbf{r}) = \mathcal{L}_{\mathrm{l}}(\mathbf{r}) + \mathcal{L}_{\mathrm{nl}}(\mathbf{r}), \tag{2}$$

$$\mathcal{L}_{\mathrm{l}}(\mathbf{r}) = \mathcal{L}_{em}(\mathbf{r}) + \mathcal{L}_{mat}(\mathbf{r}) + \mathcal{L}_{res}(\mathbf{r}) + \mathcal{L}_{int}(\mathbf{r}), \tag{3}$$

where $\mathcal{L}_{\mathrm{l}}(\mathbf{r})$ governs the linear dynamics of the system and $\mathcal{L}_{\mathrm{nl}}(\mathbf{r})$ introduces the nonlinear behaviour. The Lagrangian density $\mathcal{L}_{\mathrm{l}}(\mathbf{r})$ is the same as in [3]. We have the radiation-field part (we work in the Coulomb gauge with $\nabla \cdot \mathbf{A}(\mathbf{r}) = 0$)

$$\mathcal{L}_{em}(\mathbf{r}) = \frac{\epsilon_0}{2}\left[\dot{\mathbf{A}}(\mathbf{r}) \cdot \dot{\mathbf{A}}(\mathbf{r}) - c^2\left(\nabla \times \mathbf{A}(\mathbf{r})\right) \cdot \left(\nabla \times \mathbf{A}(\mathbf{r})\right)\right], \tag{4}$$

the matter or polarization part

$$\mathcal{L}_{mat}(\mathbf{r}) = \frac{\rho}{2}\left[\dot{\mathbf{X}}(\mathbf{r}) \cdot \dot{\mathbf{X}}(\mathbf{r}) - \omega_0^2 \mathbf{X}(\mathbf{r}) \cdot \mathbf{X}(\mathbf{r})\right], \tag{5}$$

and the reservoir part

$$\mathcal{L}_{res}(\mathbf{r}) = \frac{\rho}{2}\int_0^\infty d\omega \left[\dot{\mathbf{Y}}(\mathbf{r},\omega) \cdot \dot{\mathbf{Y}}(\mathbf{r},\omega) - \omega^2 \mathbf{Y}(\mathbf{r},\omega) \cdot \mathbf{Y}(\mathbf{r},\omega)\right]. \tag{6}$$

The interaction part is

$$\mathcal{L}_{int}(\mathbf{r}) = -\alpha \mathbf{A}(\mathbf{r}) \cdot \dot{\mathbf{X}}(\mathbf{r}) - \int_0^\infty d\omega\, v(\omega)\, \mathbf{X}(\mathbf{r}) \dot{\mathbf{Y}}(\mathbf{r},\omega), \tag{7}$$

which governs the coupling between the radiation, matter and reservoir fields. The coupling between the radiation field and polarization field is given by the constant $\alpha$, and the function $v(\omega)$ describes the coupling between the polarization field and the reservoir fields. The same assumptions apply to $v(\omega)$ as were described in [3].



With regard to the nonlinear interaction, we assume that $\mathcal{L}_{\mathrm{nl}}(\mathbf{r})$ is a function of the fields and/or their spatial derivatives. Since $\mathcal{L}_{\mathrm{nl}}(\mathbf{r})$ does not depend on the temporal derivatives of the fields, the conjugate field momenta are related to the fields in the same way as in the linear case, so that the relations between the fields and the conjugate field momenta are the same as in [3]. In what follows we will restrict attention to the case when $\mathcal{L}_{\mathrm{nl}}(\mathbf{r})$ is a function of the polarization field $\mathbf{X}(\mathbf{r})$,

$$\mathcal{L}_{\mathrm{nl}}(\mathbf{r}) = f[\mathbf{X}(\mathbf{r})], \tag{8}$$

such as $\mathcal{L}_{\mathrm{nl}}(\mathbf{r}) \sim (\mathbf{X} \cdot \mathbf{X})^2$ for a Kerr medium. This means that the nonlinear dynamics of the system is assumed to arise from the *nonlinear* motion of the *polarization* field.

Since we use the Coulomb gauge the field $\mathbf{A}(\mathbf{r})$ is transverse. However, the fields $\mathbf{X}(\mathbf{r})$ and $\mathbf{Y}(\mathbf{r},\omega)$ can have longitudinal components [3]. It is clear that $\mathcal{L}_{\mathrm{nl}}(\mathbf{r})$ provides, in general, a coupling between transverse and longitudinal components of these fields. Therefore, in contrast with the linear model, the Lagrangian does not separate into two independent parts for the transverse and longitudinal fields. The longitudinal-field effects, however, will be negligible in the limit of intense transverse light beams.

We use the fact that the conjugate momenta have the same form as in the linear case to find the Hamiltonian of our system and perform the quantization following [3]. The Hamiltonian can be written as

$$\hat{H} = \hat{H}_{\mathrm{l}} + \hat{H}_{\mathrm{nl}} = \hat{H}_{\mathrm{l}}^{\parallel} + \hat{H}_{\mathrm{l}}^{\perp} + \hat{H}_{\mathrm{nl}}, \tag{9}$$

where the Hamiltonian $\hat{H}_{\mathrm{l}}$ that governs the linear dynamics can be taken from [3], and the nonlinear interaction term $\hat{H}_{\mathrm{nl}}$ is given by

$$\hat{H}_{\mathrm{nl}} = -\int d^3r\, f[\hat{\mathbf{X}}(\mathbf{r})]. \tag{10}$$

## 3. Nonlinear field dynamics

### A. Field representation

We now represent the transverse part of the linear Hamiltonian in diagonal form [3, 5]

$$\hat{H}_{\mathrm{l}}^{\perp} = \int d^3r \int_0^\infty d\omega\, \hbar\omega\, \hat{\mathbf{f}}^\dagger(\mathbf{r},\omega) \cdot \hat{\mathbf{f}}(\mathbf{r},\omega), \tag{11}$$

where the basic field $\hat{\mathbf{f}}(\mathbf{r},\omega)$ satisfies the commutation relations [5]

$$\left[\hat{f}_j(\mathbf{r},\omega), \hat{f}_{j'}^\dagger(\mathbf{r}',\omega')\right] = \delta(\omega - \omega')\, \delta_{jj'}^{\perp}(\mathbf{r} - \mathbf{r}'),$$
$$\left[\hat{f}_j(\mathbf{r},\omega), \hat{f}_{j'}(\mathbf{r}',\omega')\right] = 0, \tag{12}$$

$\delta^{\perp}_{jj'}(\mathbf{r}-\mathbf{r}')$ being the transverse $\delta$-function. Writing

$$\hat{\mathbf{A}}(\mathbf{r}) = \int_0^\infty d\omega\, \hat{\mathbf{A}}(\mathbf{r},\omega) + \text{H.c.}, \tag{13}$$

$$\hat{\mathbf{X}}^{\perp}(\mathbf{r}) = \int_0^\infty d\omega\, \hat{\mathbf{X}}^{\perp}(\mathbf{r},\omega) + \text{H.c.}, \tag{14}$$

we can relate $\hat{\mathbf{A}}(\mathbf{r})$ and $\hat{\mathbf{X}}^{\perp}(\mathbf{r})$ to $\hat{\mathbf{f}}(\mathbf{r},\omega)$ as [5]

$$\hat{\mathbf{A}}(\mathbf{r},\omega) = \sqrt{\frac{\hbar}{\pi\epsilon_0 c^2}\frac{\omega}{c}}\sqrt{\varepsilon_{\rm i}(\omega)} \int d^3r'\, G(\mathbf{r},\mathbf{r}',\omega)\,\hat{\mathbf{f}}(\mathbf{r}',\omega), \tag{15}$$

$$\hat{\mathbf{X}}^{\perp}(\mathbf{r},\omega) = \frac{\epsilon_0}{\alpha}\left[-i\omega\,[\varepsilon(\omega)-1]\,\hat{\mathbf{A}}(\mathbf{r},\omega) + i\sqrt{\frac{\hbar}{\pi\epsilon_0}}\sqrt{\varepsilon_{\rm i}(\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega)\right]. \tag{16}$$

Here

$$G(\mathbf{r},\mathbf{r}',\omega) = -\frac{1}{4\pi|\mathbf{r}-\mathbf{r}'|}\exp\!\left[i\frac{\omega}{c}\sqrt{\varepsilon(\omega)}\,|\mathbf{r}-\mathbf{r}'|\right] \tag{17}$$

is the Green function satisfying the equation

$$\Delta G(\mathbf{r},\mathbf{r}',\omega) + \frac{\omega^2}{c^2}\varepsilon(\omega)\, G(\mathbf{r},\mathbf{r}',\omega) = \delta(\mathbf{r}-\mathbf{r}'), \tag{18}$$

and $\varepsilon(\omega) = \varepsilon_{\rm r} + i\varepsilon_{\rm i}$ is the complex permittivity (for the connection between $\varepsilon(\omega)$ and the oscillator-medium parameters, see [3]). Note that from Eqs. (15), (17), and (18) $\hat{\mathbf{A}}(\mathbf{r},\omega)$ can be seen to satisfy the spatial propagation equation

$$\Delta\hat{\mathbf{A}}(\mathbf{r},\omega) + \frac{\omega^2}{c^2}\varepsilon(\omega)\hat{\mathbf{A}}(\mathbf{r},\omega) = \sqrt{\frac{\hbar}{\pi\epsilon_0 c^2}\frac{\omega}{c}}\sqrt{\varepsilon_{\rm i}(\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega). \tag{19}$$

Next we introduce the transverse electric field strength operator

$$\hat{\mathbf{E}}(\mathbf{r}) = i\int_0^\infty d\omega\,\omega\hat{\mathbf{A}}(\mathbf{r},\omega) + \text{H.c.}, \tag{20}$$

it can be proven [3, 5] that the components of the vector potential and the electric field strength satisfy the well-known canonical commutation relations

$$\left[\hat{A}_j(\mathbf{r}), \hat{E}_{j'}(\mathbf{r}')\right] = -\frac{i\hbar}{\epsilon_0}\delta^{\perp}_{jj'}(\mathbf{r}-\mathbf{r}'). \tag{21}$$

Relations for the longitudinal fields can be found analogously.

### B. Field evolution equations

The equation of motion for $\hat{\mathbf{f}}(\mathbf{r},\omega)$ in the Heisenberg picture reads as

$$i\hbar\partial_t\hat{\mathbf{f}}(\mathbf{r},\omega) = \left[\hat{\mathbf{f}}(\mathbf{r},\omega), \hat{H}\right] = \hbar\omega\,\hat{\mathbf{f}}(\mathbf{r},\omega) + \left[\hat{\mathbf{f}}(\mathbf{r},\omega), \hat{H}_{\rm nl}\right]. \tag{22}$$



Note that (in the nonlinear case) $\omega$ is not the frequency, but plays the role of a continuous index of the reservoir fields. It should be clear that due to the linear (equal-time) connection between $\hat{\mathbf{A}}(\mathbf{r},\omega)$, $\hat{\mathbf{X}}^{\perp}(\mathbf{r},\omega)$, and $\hat{\mathbf{f}}(\mathbf{r},\omega)$ the equations of motion for $\hat{\mathbf{A}}(\mathbf{r},\omega)$ and $\hat{\mathbf{X}}^{\perp}(\mathbf{r},\omega)$ have the same form as Eq. (22). Furthermore, from Eq. (22) we see that $\omega$ corresponds to the action of an operator

$$\hat{\omega} = \left(i\partial_t + \hbar^{-1}\hat{H}^{\times}_{\rm nl}\right), \tag{23}$$

where the action of $\hat{H}^{\times}_{\rm nl}$ on an operator $\hat{O}$ is defined by

$$\hat{H}^{\times}_{\rm nl}\,\hat{O} \equiv \left[\hat{H}_{\rm nl}, \hat{O}\right]. \tag{24}$$

In order to obtain a space-time evolution equation for the vector potential, we define [by power-series expansion of $K(\omega) = c^{-2}\omega^2\varepsilon(\omega)$] the operator function

$$\hat{K}\left(i\partial_t + \hbar^{-1}\hat{H}^{\times}_{\rm nl}\right) = c^{-2}\left(i\partial_t + \hbar^{-1}\hat{H}^{\times}_{\rm nl}\right)^2 \varepsilon\left(i\partial_t + \hbar^{-1}\hat{H}^{\times}_{\rm nl}\right). \tag{25}$$

We then use this in Eq. (19) to replace the function $K(\omega) = c^{-2}\omega^2\varepsilon(\omega)$:

$$\Delta\hat{\mathbf{A}}(\mathbf{r},\omega) + \hat{K}\left(i\partial_t + \hbar^{-1}\hat{H}^{\times}_{\rm nl}\right)\hat{\mathbf{A}}(\mathbf{r},\omega) = \sqrt{\frac{\hbar}{\pi\epsilon_0 c^2}\frac{\omega}{c}}\sqrt{\varepsilon_{\rm i}(\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega). \tag{26}$$

Since $\hat{K}$ does not depend explicitly on $\omega$, in Eq. (26) the $\omega$ integration can be performed and the sought evolution equation can formally be given by

$$\Delta\hat{\mathbf{A}}(\mathbf{r}) + \hat{K}\left(i\partial_t + \hbar^{-1}\hat{H}^{\times}_{\rm nl}\right)\hat{\mathbf{A}}(\mathbf{r}) = \sqrt{\frac{\hbar}{\pi\epsilon_0 c^2}}\left[\int_0^\infty d\omega\,\frac{\omega}{c}\sqrt{\varepsilon_{\rm i}(\omega)}\,\hat{\mathbf{f}}(\mathbf{r},\omega) + \text{H.c.}\right]. \tag{27}$$

The properties of $\varepsilon(\omega)$ imply $[\varepsilon_{\rm r}(-\omega) = \varepsilon_{\rm r}(\omega),\ \varepsilon_{\rm i}(-\omega) = -\varepsilon_{\rm i}(\omega)]$ that $\hat{K}$ is Hermitian. It may be convenient to decompose $\hat{K}$ into two parts,

$$\hat{K} = \hat{K}_{\rm l} + \hat{K}_{\rm nl}, \tag{28}$$

where $\hat{K}_{\rm l} = \hat{K}(i\partial_t) = c^{-2}(i\partial_t)^2\varepsilon(i\partial_t)$ [together with the term on the right-hand side in Eq. (27)] describes the effects of linear dispersion and absorption. The nonlinear effects (and their mixing with the linear effects) are included in $\hat{K}_{\rm nl}$.

Equation (27) can be regarded as a basic equation for describing the propagation of quantized radiation in a nonlinear medium including both dispersion and absorption. From inspection of the equation we see that the term on the right-hand side is obviously related to the noise sources that are necessarily associated with absorption [factor $\sqrt{\varepsilon_{\rm i}(\omega)}$]. On the left-hand side we can express $\hat{\mathbf{X}}(\mathbf{r})$ in $\hat{H}_{\rm nl}$, Eq. (10), in terms of its transverse and longitudinal parts $\hat{\mathbf{X}}^{\perp}(\mathbf{r})$ and $\hat{\mathbf{X}}^{\parallel}(\mathbf{r})$, respectively, and then make use of Eqs. (14) and (16) to relate $\hat{\mathbf{X}}^{\perp}(\mathbf{r})$ to $\hat{\mathbf{A}}(\mathbf{r},\omega)$ and $\hat{\mathbf{f}}(\mathbf{r},\omega)$. In this way we find that noise sources naturally arise in the nonlinear terms in the equation of motion (27). It has previously been shown that such noise sources are a necessary component of a quantization scheme in a Kerr-type medium [8].

## 4. Summary

We have developed a quantization scheme for radiation in a nonlinear dielectric with dispersion and absorption. This has been achieved by modifying the Lagrangian of the linear model [3] by the addition of a general nonlinear term which does not change the canonically conjugated momenta of the system. This guarantees that the equal-time commutation relations of the linear theory are also valid in the extended theory. In particular, the equal-time commutation relations of the free field are preserved, and the spatial propagation equations for the field components are the same as for a linear dielectric. The nonlinearity provides a coupling between transverse and longitudinal fields, making impossible a separation of the Hamiltonian into two completely independent parts as in the linear case. As in the linear dielectric, absorption provides additional noise. Since the basic equation between the transversal polarization field and the electromagnetic field necessarily contains fluctuations, noise sources also arise naturally in the nonlinear terms.

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