Three-dimensional theory for light-matter interaction

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We present a full quantum mechanical three-dimensional theory describing an electromagnetic field interacting with an ensemble of identical atoms. The theory is constructed such that it describes recent experiments on light-matter quantum interfaces, where the quantum fluctuations of light are mapped onto the atoms and back onto light. We show that the interaction of the light with the atoms may be separated into a mean effect of the ensemble and a deviation from the mean. The mean effect of the interaction effectively gives rise to an index of refraction of the gas. We formally change to a dressed state picture, where the light modes are solutions to the diffraction problem, and develop a perturbative expansion in the fluctuations. The fluctuations are due to quantum fluctuations as well as the random positions of the atoms. In this perturbative expansion we show how the quantum fluctuations are mapped between atoms and light while the random positioning of the atoms give rise to decay due to spontaneous emission. Furthermore we identify limits, where the full three-dimensional theory reduces to the one-dimensional theory typically used to describe the interaction.

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I. INTRODUCTION

For several applications in quantum information science, such as long-distance quantum communication [1], it is essential to create an interface linking the photonic states used for transmitting quantum information to a material state suitable for storing and processing the information. The generation of the required strong coherent coupling of light to a single emitter has proven difficult to achieve in practice, although substantial progress has been made [2–6]. In recent years optically dense atomic ensembles has emerged as a promising alternative [7–22]. In this approach one can for instance use classical laser pulses to engineer a suitable interaction such that an incoming light field is reversibly stored into the coherence between, e.g., two stable ground states in the atoms [9].

Some experiments on atomic ensembles uses atoms that are enclosed inside a cavity to enhance the coupling [18]. In this situation the cavity defines a unique mode of the light field and the theoretical description consists of describing a single optical mode coupled to the atomic ensembles. Most experiments are, however, performed with atoms in free space not enclosed in a cavity, and in this situation the theoretical description is more complicated. Typically this situation is described in a one-dimensional approximation, where one only considers a single transverse mode and solves a one-dimensional propagation equation for this mode [7,12,13].

In this paper we explore the range of validity of the one-dimensional approximation by making a full three-dimensional description of the interaction between light and an atomic ensemble. Our calculations directly apply to an experimental situations similar to the ones described in Refs. [8–11], where the light is detuned far from the atomic transition, but we expect the general features of our results to be valid for a much broader class of problems.

Some justification for the one-dimensional description may be found in the literature on superfluorescence, e.g., Refs. [23–25]. In this context it was found that the one-dimensional description is valid provided that the Fresnel number is of order unity $\mathcal{F}\equiv A/\lambda L\approx 1$, where A is the transverse beam area, λ is the wavelength of the light, and L is the length of the ensemble. Based on this work it has been argued that it is also necessary to have a Fresnel number of order unity in order for the one-dimensional approximation to be applicable to the quantum interfaces between light and atomic ensembles [7,12,13]. It is, however, essential to realize that the physical situations are very different in the two cases. The work on superfluorescence typically concerns the temporal distribution of the output light measured by impinging the outgoing light on a photodetector. Because the photodetector just measures the incoming flux I, this is essentially a multimode measurement

$$I \propto \sum_m \hat{a}_m^\dagger \hat{a}_m, \tag{1.1}$$

where the sum is over all modes m hitting the detector, and each of these modes are described by the photon creation (annihilation) operators $\hat{a}_m^\dagger$ ($\hat{a}_m$). In particular the sum here includes all transverse modes. This is in contrast to the quantum interface work, where one is interested in the outgoing state of a single light mode, e.g., in Refs. [8–11] the measurement is essentially a homodyne measurement of a single mode, defined by the field of the strong classical laser. In other experiments the outgoing light is sent through a single mode optical fiber, which filters out everything except a single transverse mode. Furthermore, the superfluorescence work applies to a nonperturbative situation with a large optical gain, whereas the quantum interfaces typically operate in the few excitations regime. The previous analysis is thus not applicable to the present situation and it is therefore not to be expected that the condition $\mathcal{F}\sim 1$ is the correct condition for the validity of the one-dimensional approximation. In fact, the experiments in Refs. [8–10] are performed with $\mathcal{F}\sim 10^4$, and still give very good agreement with the one-

dimensional description. Here we make a full three-dimensional description of the experiments in Refs. [8–10], and we find that it reduces to the one-dimensional description in the paraxial approximation provided that $\mathcal{F} \gg 1$.

In a related work a three-dimensional description was also presented in Ref. [26]. Whereas our procedure assumes nonmoving atoms, i.e., cold atoms, that work considered the opposite limit, where the motion of the atoms wash out any spatial structure of the atomic spin state. Unlike the situation in Ref. [26], where the motion of the atoms always lead to certain inefficiencies, the fact that we consider stationary atoms, allows us to identify certain limits, where we exactly reproduce the simple result of the one-dimensional theory as discussed in Sec. VI B.

Our theory is developed as a perturbative expansion of the interaction between light and the atomic ensembles. It is, however, essential to be very careful about the way this perturbative expansion is performed. Below we shall present results up to second order in the interaction between the light and the atoms. We shall use an effective Hamiltonian, where the excited atomic state has been eliminated, i.e., a Hamiltonian of the form

$$H \sim \sum_{\mathbf{k},\mathbf{k}'} \sum_{i} g_{\mathbf{k},\mathbf{k}'} u_{\mathbf{k}}(\mathbf{r}_i) u^*_{\mathbf{k}'}(\mathbf{r}_i) \hat{a}^\dagger_{\mathbf{k}'} \hat{a}_{\mathbf{k}}, \tag{1.2}$$

where $g_{\mathbf{k},\mathbf{k}'}$ is a coupling constant for the two modes $\mathbf{k}$, and $\mathbf{k}'$ described by photon creation (annihilation) operators $\hat{a}^\dagger_{\mathbf{k}}$ ($\hat{a}_{\mathbf{k}}$) with mode functions $u_{\mathbf{k}}$, and $\mathbf{r}_i$ is the position of the ith atom. If we take the mode functions to be simple plane waves with an input field in a certain mode $\mathbf{k}_0$ and calculate the intensity in a certain direction described by $\mathbf{k}_1$, we find the intensity

$$I \propto \left| \sum_i e^{i\Delta\mathbf{k}\cdot\mathbf{r}_i} \right|^2 = \sum_{i,j} e^{i\Delta\mathbf{k}\cdot(\mathbf{r}_i - \mathbf{r}_j)}, \tag{1.3}$$

where $\Delta\mathbf{k} = \mathbf{k}_1 - \mathbf{k}_0$. The standard way to proceed from here is to say that the exponential varies rapidly when $i \neq j$ and therefore neglect all terms except $i=j$ so that one is left with something proportional to the number of atoms N_A, which is known as spontaneous emission. For the problem we are interested in here, we are, however, mainly concerned with the properties of the light in the forward direction, where $\Delta\mathbf{k} \approx 0$. In this case it seems unjustified to neglect the cross terms which give rise to collective scattering scaling as N^2. Since N is typically a very large number, the presence of such large N^2 contributions may limit the applicability of perturbation theory.

In order to avoid the problems associated with this collective scattering, we use a different basis for our perturbative expansion: instead of starting from the eigenmodes of the propagation equation in vacuum, we use the solutions to the classical diffraction problem in the presence of the medium, i.e., we take into account that the atoms give rise to an index of refraction of the gas, which changes the propagation of the light. Specifically, we write the Hamiltonian as

$$H = \langle \mathcal{H} \rangle_{\text{atoms}} + \delta H, \tag{1.4}$$

where $\langle \mathcal{H} \rangle_{\text{atoms}}$ is the quantum mechanical expectation value of the Hamiltonian with respect to the atomic spin state averaged over the random positions of the atoms. This averaged Hamiltonian gives rise to a continuous quadratic Hamiltonian in the light field operators similar to a Hamiltonian describing the interaction with a dielectric medium. When we formally change to the interaction picture with respect to this averaged Hamiltonian, we obtain a new set of basis modes. Doing perturbation theory on these modes, the only effect on the light comes from the quantum mechanical fluctuations and the fluctuations caused by the random position of the atoms. These fluctuations are described by the Hamiltonian $\delta H = H - \langle \mathcal{H} \rangle_{\text{atoms}}$. When we average the first-order term in the perturbative expansion with respect to the position of the atoms the resultant expression describe that the quantum fluctuations of the atoms are mapped onto the light in analogy with the results derived in a one-dimensional theory in Ref. [7].

If we go to second order in the interaction, our expression will give terms quadratic in δH. In order to take the spatial average of such terms we need to know the density-density correlation function of the atoms. Inserting the density-density correlation function for an ideal gas we no longer find the collective scattering terms described above, i.e., the collective scattering is essentially the classical diffraction of the light, which is explicitly taken into account by our average Hamiltonian, and therefore it does not appear in our perturbation theory. The spatial average of the second-order term does, however, produce a new term associated with the point particle nature of the atoms and their random positions. This term is equivalent to the results obtained by just keeping the $i=j$ terms in Eq. (1.3), and represents the effect of spontaneous emission.

Unlike most approaches to the interaction between atoms and light, which derive coupled equations for the atomic states and the electric field, our approach considers the electric displacement field $\mathbf{D}$ instead of the electric field. The reason we chose to use the displacement field is that it is convenient to work with a purely transverse field, which is the case for the displacement field due to the macroscopic Maxwell equation $\boldsymbol{\nabla} \cdot \mathbf{D} = 0$, whereas this is not necessarily the case for the electric field in a medium. Formally the two approaches are equivalent and may be related through a unitary transformation [27].

The full theory is quite involved. Readers who are mainly interested in the consequences of our theory for experimental implementations are therefore advised to skip to Sec. VI, where we discuss such consequences. The sections prior to this mainly focus on building the theoretical frame using a first-principles strategy. The paper is organized as follows: In Sec. II we give the details of the model used to describe the interaction. In Sec. III we derive a set of equations of motion describing the system of atoms and light, using Heisenberg's equation of motion. The wave equation describing the light is expressed in a form that ideally suits a perturbative treatment. In Sec. IV we express the general solution to the wave equation in terms of Green's functions and derive the pertur-

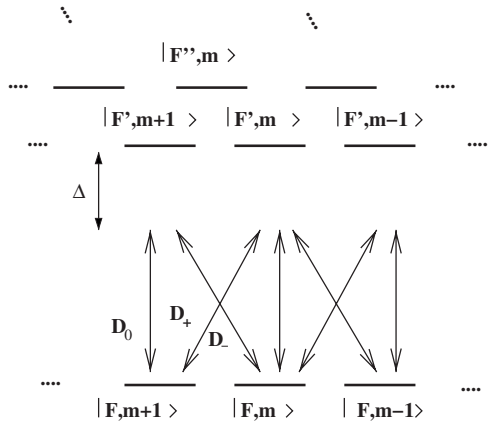


FIG. 1. Example of an atomic level structure. The atoms have a single ground level with spin F and one or more exited levels. The fields have a large detuning Δ so that the exited states may be adiabatically eliminated and we obtain an effective ground-state Hamiltonian equation (2.3).

bative expansion of the solution to the wave equations as well as the equation describing the atoms. This is represented in terms of Feynman diagrams. In addition we develop the appropriate theoretical tools to describe point particle effects such as density correlations, and derive a formal expression for the Green's function. In Sec. V we present our results where we discuss higher order effects such as spin decay and light scattering. We define operators that describe photon measurements, and demonstrate how these are calculated in the theory. In Sec. VI we discuss various limits where the general three-dimensional theory reduces to the usually employed one-dimensional model [7]. We also describe how a detailed understanding of the spatial modes can be used to achieve storage and retrieval of information in several transverse modes of light and atoms simultaneously. In Sec. VII we conclude the work, and in the appendixes we give several details omitted from the main text.

II. MODEL

The model we consider describes the interaction between an ensemble of atoms and an incoming light field. The atomic ensemble is considered to be an ideal gas of identical atoms. The atoms are described as nonmoving randomly distributed point particles and the interaction with the light field is described within the dipole approximation. Each atom is assumed to have a ground level of total spin F. In addition we assume that the atoms have no other stable ground states to which they can decay (see Fig. 1). We shall assume that the electric fields are sufficiently far detuned that we may adiabatically eliminate the exited states, and work with an effective Hamiltonian involving only the ground states. In the following we first discuss the interaction between light and a single atom, and then move on to discuss the interaction with an ensemble of atoms.

A. Interaction with single atoms

The aim of this work is to describe the interaction between an electromagnetic field and an ensemble of identical atoms. The problem is therefore both to deal with the microscopic behavior of a single atom, and also the collective effect of many atoms. We choose here to work in the so-called length gauge, where the basic interaction is given as the product of the displaced electric field and the polarization of the media [27,28]

$$\mathcal{H}_{\text{int}} = -\sum_{j}^{\text{Atoms}} \frac{1}{\epsilon_0} \mathbf{D}(\mathbf{r}_j,t) \cdot \mathbf{P}(\mathbf{r}_j,t). \tag{2.1}$$

Our gauge choice ensures $\boldsymbol{\nabla} \cdot \mathbf{D}(\mathbf{r},t)=0$. We will assume that the fields have a large detuning and do not saturate the atomic transition, so that the exited levels may be adiabatically eliminated. This procedure is described in Appendix A. The polarization of the atomic ensemble then depends linearly on the displaced electric field, that is $\mathbf{P}(\mathbf{r},t) = \bar{\bar{V}}[\hat{\mathbf{J}}]\mathbf{D}(\mathbf{r},t)$. We introduce here the argument $\hat{\mathbf{J}}$ to indicate that the interaction matrix $\bar{\bar{V}}[\hat{\mathbf{J}}]$ depends on the spin of the atoms. Next we write the displaced electric field as a sum of a positively oscillating part and a negatively oscillating part,

$$\mathbf{D}(\mathbf{r},t) = \mathbf{D}^{(+)}(\mathbf{r},t) + \mathbf{D}^{(-)}(\mathbf{r},t). \tag{2.2}$$

In Appendix A we show that the effective interaction Hamiltonian, assuming such linear dependence of the polarization on the displaced electric field, reads as

$$\mathcal{H}_{\text{int}} = -\frac{1}{2\epsilon_0} \sum_{j}^{\text{Atoms}} [(\bar{\bar{V}}[\hat{\mathbf{J}}_j]^t \mathbf{D}_j^{(-)}) \cdot \mathbf{D}_j^{(+)} + \mathbf{D}_j^{(-)} \cdot (\bar{\bar{V}}[\hat{\mathbf{J}}_j] \mathbf{D}_j^{(+)})], \tag{2.3}$$

where we have also employed the rotating-wave approximation. Here the superscript t denotes matrix transposition.

Since the Hamiltonian must be rotationally invariant it can only contain irreducible tensors of at most rank 2. In the vector representation the interaction may thus in general be written as

$$\bar{\bar{V}}[\mathbf{J}_j] = \beta(c_0 \hat{\mathbf{J}}_j^2 - i c_1 \hat{\mathbf{J}}_j \times + c_2 \times \hat{\mathbf{J}}_j) \cdot (\hat{\mathbf{J}}_j \times \ ). \tag{2.4}$$

The meaning of the notation is that when inserted into the Hamiltonian the result of, e.g., the last term on the right-hand side of Eq. (2.4) is

$$\beta c_2 \sum_{j}^{\text{Atoms}} [\mathbf{D}^{(-)}(\mathbf{r}_j,t) \times \hat{\mathbf{J}}_j] \cdot [\hat{\mathbf{J}}_j \times \mathbf{D}^{(+)}(\mathbf{r}_j,t)]. \tag{2.5}$$

Note that we have here chosen a description which has a simple analytical representation, but this means the c_2 term is not a pure rank-2 irreducible tensor, but consist of a combination of tensors of rank 0, rank 1, and rank 2. In matrix form the interaction may be written as

$$\bar{\bar{V}}[\bar{\mathbf{J}}] = \beta \begin{bmatrix} (c_0 - c_2)\hat{\mathbf{J}}^2 + c_2\hat{J}_x^2 & ic_1\hat{J}_z + c_2\hat{J}_y\hat{J}_x & -ic_1\hat{J}_y + c_2\hat{J}_z\hat{J}_x \\ -ic_1\hat{J}_z + c_2\hat{J}_x\hat{J}_y & (c_0 - c_2)\hat{\mathbf{J}}^2 + c_2\hat{J}_y^2 & ic_1\hat{J}_x + c_2\hat{J}_z\hat{J}_y \\ ic_1\hat{J}_y + c_2\hat{J}_x\hat{J}_z & -ic_1\hat{J}_x + c_2\hat{J}_y\hat{J}_z & (c_0 - c_2)\hat{\mathbf{J}}^2 + c_2\hat{J}_z^2 \end{bmatrix}. \tag{2.6}$$

In general the atoms may have several exited levels as shown in Fig. 1. The effect of several exited levels can be included in the coefficients c_0, c_1, and c_2 that will then depend on the detuning. For atoms with $F=\frac{1}{2}$ or for an alkali atom, where the fields are detuned by more than the hyperfine structure of the exited state, the c_2 term disappears [29] and the interaction matrix is given by

$$\bar{\bar{V}}[\hat{\mathbf{J}}_j] = \beta(c_0\hat{\mathbf{J}}_j^2 - ic_1\hat{\mathbf{J}}_j\times\ ). \tag{2.7}$$

Here c_0 and c_1 are constants which depend on the atomic structure as well as the detuning. The coupling constant β in Eq. (2.7) is given by

$$\beta = \frac{\pi\gamma}{2\Delta k_L^3}, \tag{2.8}$$

where γ is the linewidth of the exited level, Δ the detuning of the laser field with respect to the atomic transition, and k_L is the wave vector. With this choice of β the coefficients c_0, c_1, and c_2 will be of order unity or less. Throughout this paper we shall only consider the simple interaction in (2.7). A discussion of the effect of the c_2 term is given in Refs. [21,22] in a one-dimensional description.

We will consider a perturbative regime, where the product of the atomic density ρ and β is small $\beta\rho \ll 1$, and make a perturbative expansion in β. Note, however, that this condition does not imply that the total effect of the interaction is small. On the contrary, we are most interested in situations, where the integrated effect of the interaction significantly alters the light beam as it passes through the sample. To take into account these collective effects we explicitly include, e.g., the diffraction of the light caused by the propagation through a medium. To describe these effects we discuss in the following section how to quantize the field in a medium.

B. Mode expansion

To quantize the electromagnetic fields we could (i) impose the canonical commutation relations on the vector potential and displaced electric field or (ii) expand the electromagnetic fields on an orthonormal set of spatial mode functions $\{\mathbf{f_k}\}$ conveniently chosen to diagonalize the Hamiltonian (in vacuum this is the set of plane waves), and then quantizing the mode amplitudes. Here we will use the latter. The Hamiltonian describing the electromagnetic field in a medium is given by [27]

$$\mathcal{H} = \frac{1}{2}\int d^3r\left(\frac{\mathbf{D}^2}{\epsilon_0} + \frac{(\boldsymbol{\nabla}\times\mathbf{A})^2}{\mu_0}\right) + \mathcal{H}_{\text{int}}, \tag{2.9}$$

where $\mathcal{H}_{\text{int}}$ is given in Eq. (2.3). A careful analysis of how to quantize the electromagnetic field in a medium, is given in Ref. [30], and here we shall only go through the steps briefly.

By introducing the spin field

$$\hat{\mathbf{J}}(\mathbf{r},t) = \sum_j^{\text{Atoms}} \hat{\mathbf{J}}_j\delta(\mathbf{r}-\mathbf{r}_j), \tag{2.10}$$

the Hamiltonian may be set in an all-integral form. The main idea in our approach is to divide the full Hamiltonian into a spatially averaged part, and a point particle part, describing the fluctuations from the average caused by the atoms being point particles. For now we only consider the spatially averaged part of the theory. We will use calligraphic font to denote that we have made a spatial average. We thus write the spatially averaged interaction from Eq. (2.7) as

$$\bar{\bar{\mathcal{V}}}[\bar{\mathbf{J}}] = \beta\rho(\mathbf{r})[c_0\overline{\mathbf{J}^2} - ic_1\bar{\mathbf{J}}(\mathbf{r})\times\ \ ]. \tag{2.11}$$

Here a bar denotes a single-atom operator, that is $\bar{\mathbf{J}}(\mathbf{r})$ is the spin operator of a single atom at position $\mathbf{r}$. We use the bar to distinguish between the spatially averaged single-atom spin operator, and the general spin field in Eq. (2.10). The two may be related by $\langle\hat{\mathbf{J}}(\mathbf{r},t)\rangle_{\text{s.a.}} = \rho(\mathbf{r})\bar{\mathbf{J}}(\mathbf{r},t)$, where $\langle\cdots\rangle_{\text{s.a.}}$ denotes spatial average. The function $\rho(\mathbf{r})$ denotes the average atomic density, which in this model is a continuous scalar field.

In the following we will define a mean Hamiltonian, where we have taken into account the quantum mechanical average of the spatially averaged interaction. We then write the Hamiltonian as a sum of the average Hamiltonian and a point particle Hamiltonian

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{pp}}, \tag{2.12}$$

where

$$\mathcal{H}_0 = \frac{1}{2}\int d^3r\left(\frac{\mathbf{D}(\bar{\bar{\mathcal{M}}}^t\mathbf{D}^{(-)} + \bar{\bar{\mathcal{M}}}\mathbf{D}^{(+)})}{\epsilon_0} + \frac{(\boldsymbol{\nabla}\times\mathbf{A})^2}{\mu_0}\right), \tag{2.13}$$

$$\mathcal{H}_{\text{pp}} = -\frac{1}{2\epsilon_0}\int d^3r\mathbf{D}\cdot(\bar{\bar{m}}[\hat{\mathbf{J}}]^t\mathbf{D}^{(-)} + \bar{\bar{m}}[\hat{\mathbf{J}}]\mathbf{D}^{(+)}), \tag{2.14}$$

$$\bar{\bar{\mathcal{M}}} = \mathbb{1} - \bar{\bar{\mathcal{V}}}[\mathbf{J}], \tag{2.15}$$

and

$$\bar{\bar{m}}[\hat{\mathbf{J}}] = \bar{\bar{V}}[\hat{\mathbf{J}}] - \bar{\bar{\mathcal{V}}}[\mathbf{J}]. \tag{2.16}$$

Here we simply write $\mathbf{J}$ (without the hat) to denote that this is now a classical field describing the classical expectation of

the spin of the atoms. In analogy with Ref. [30] we introduce the mode functions $\{\mathbf{f}_\mathbf{k}\}$ defined by

$$\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\bar{\bar{\mathcal{M}}}\mathbf{f}_\mathbf{k}(\mathbf{r})=\frac{\omega_\mathbf{k}^2}{c^2}\mathbf{f}_\mathbf{k}(\mathbf{r}), \tag{2.17a}$$

$$\boldsymbol{\nabla}\cdot\mathbf{f}_\mathbf{k}(\mathbf{r})=0. \tag{2.17b}$$

We also define the appropriate inner product on the space spanned by these mode functions:

$$\langle\phi(\mathbf{r})|\psi(\mathbf{r})\rangle=\int d^3r\,\phi(\mathbf{r})^*\cdot\bar{\bar{\mathcal{M}}}\psi(\mathbf{r}). \tag{2.18}$$

We will assume that the average interaction term $\bar{\bar{\mathcal{V}}}[\mathbf{J}]$ does not evolve in time, and our appropriate mode functions are therefore time independent vector fields. One can show that the functions $\mathbf{f}_\mathbf{k}$ span a complete orthonormal basis for the space in which we work. To diagonalize the Hamiltonian we expand the vector potential and the displaced electric field in these mode functions

$$\mathbf{D}(\mathbf{r},t)=-\sum_\mathbf{k}\sqrt{\epsilon_0}p_\mathbf{k}(t)\mathbf{f}_\mathbf{k}^*(\mathbf{r}), \tag{2.19a}$$

$$\mathbf{A}(\mathbf{r},t)=\sum_\mathbf{k}c\sqrt{\mu_0}q_\mathbf{k}(t)(1-\bar{\bar{\mathcal{V}}}[\mathbf{J}])f_\mathbf{k}(\mathbf{r}). \tag{2.19b}$$

The minus sign in Eq. (2.19a) is conventional and stems from the relation between the displaced electric field and the canonical conjugate field given in terms of the vector potential.

The reality condition on the displaced electric field $\{[\mathbf{D}(\mathbf{r},t)]^\dagger=\mathbf{D}(\mathbf{r},t)\}$ allows us to write the following:

$$\mathbf{D}(\mathbf{r},t)=-\sum_\mathbf{k}\frac{\sqrt{\epsilon_0}}{2}[p_\mathbf{k}^\dagger(t)\mathbf{f}_\mathbf{k}(\mathbf{r})+p_\mathbf{k}(t)\mathbf{f}_\mathbf{k}^*(\mathbf{r})]. \tag{2.20}$$

Using the results in Eq. (2.18) and the expansion in Eqs. (2.19), the Hamiltonian attains the desired diagonal form

$$\mathcal{H}_0=\frac{1}{2}\int d^3r\left(\frac{\mathbf{D}(1-\bar{\bar{\mathcal{V}}}[\mathbf{J}])\mathbf{D}}{\epsilon_0}+\frac{(\boldsymbol{\nabla}\times\mathbf{A})^2}{\mu_0}\right)$$
$$=\frac{1}{2}\sum_\mathbf{k}[p_\mathbf{k}^\dagger(t)p_\mathbf{k}(t)+\omega_\mathbf{k}^2q_\mathbf{k}^\dagger(t)q_\mathbf{k}(t)]. \tag{2.21}$$

The mode functions $\{\mathbf{f}_\mathbf{k}\}$ are thus the spatial basis diagonalizing the spatially averaged Hamiltonian, and as we shall see the proper basis describing the diffraction problem.

The splitting in Eq. (2.12) allows us to consider the problem as comprised of two types of properties. The effect of single atoms, and the spatially averaged Hamiltonian. The effect of the spatially averaged Hamiltonian is well understood in terms of the mode functions defined in Eq. (2.17). The point particle effect we will discuss in greater detail when considering the equations of motion for the full system. Before deriving these equations of motion we, however, briefly need to discuss the commutation relations describing the system.

C. Quantization and commutation relations

Above we expanded the fields in convenient spatial modes. The coordinates $p_\mathbf{k}(t)$ and $q_\mathbf{k}(t)$ are canonically conjugate variables, and we can thus quantize our theory by imposing the commutation relations

$$[q_\mathbf{k}(t),p_{\mathbf{k}'}(t)]=i\hbar\delta_{\mathbf{k}\mathbf{k}'}. \tag{2.22}$$

It will however be convenient to have the commutation relations for the fields which we may derive from the mode-amplitude commutation relations. It will also be convenient to separate the displaced electric field into a positively and a negatively oscillating part $\mathbf{D}=\mathbf{D}^{(+)}+\mathbf{D}^{(-)}$, where $\mathbf{D}^{(-)}$ is in accordance with convention chosen so that it only contains terms oscillating like $e^{i\omega_\mathbf{k}t}$. Our choice of gauge is reflected in the transversality of the mode functions defined in Eq. (2.17). We expect this transversality condition to be represented in the commutation relations as well. With the quantization procedure above one finds the following expression for the negative frequency part of the relevant fields:

$$\hat{\mathbf{D}}^{(-)}(\mathbf{r},t)=-i\sum_\mathbf{k}\sqrt{\frac{\hbar\omega_\mathbf{k}\epsilon_0}{2}}\hat{a}_\mathbf{k}^\dagger e^{i\omega_\mathbf{k}t}\mathbf{f}_\mathbf{k}^*(\mathbf{r}), \tag{2.23a}$$

$$\hat{\mathbf{A}}^{(-)}(\mathbf{r},t)=\sum_\mathbf{k}c\sqrt{\frac{\hbar\mu_0}{2\omega_\mathbf{k}}}\hat{a}_\mathbf{k}^\dagger e^{i\omega_\mathbf{k}t}(1-\bar{\bar{\mathcal{V}}}[\mathbf{J}])^t\mathbf{f}_\mathbf{k}^*(\mathbf{r}). \tag{2.23b}$$

The positive frequency part may be found by Hermitian conjugation. The above result is found from Eq. (2.20) along with the definitions of creation and annihilation operators given by

$$q_\mathbf{k}(t)=\sqrt{\frac{\hbar}{2\omega_\mathbf{k}}}(\hat{a}_\mathbf{k}(t)+\sum_{\mathbf{k}'}U_{\mathbf{k}\mathbf{k}'}^*\hat{a}_{\mathbf{k}'}^\dagger(t)), \tag{2.24a}$$

$$p_\mathbf{k}(t)=i\sqrt{\frac{\hbar\omega_\mathbf{k}}{2}}(\hat{a}_\mathbf{k}^\dagger(t)-\sum_{\mathbf{k}'}U_{\mathbf{k}\mathbf{k}'}\hat{a}_{\mathbf{k}'}(t)), \tag{2.24b}$$

where the matrix $U_{\mathbf{k}\mathbf{k}'}$ is defined as

$$U_{\mathbf{k}\mathbf{k}'}=\int d^3r\bar{\bar{\mathcal{M}}}\mathbf{f}_\mathbf{k}(\mathbf{r})\cdot\mathbf{f}_{\mathbf{k}'}(\mathbf{r}). \tag{2.25}$$

A detailed discussion of this procedure is found in Ref. [30].

From these definitions and the commutation relations (2.22) we obtain

$$[\hat{a}_\mathbf{k}(t),\hat{a}_\mathbf{k}^\dagger(t)]=\delta_{\mathbf{k}\mathbf{k}'}. \tag{2.26}$$

Going to the field operators we obtain

$$[\hat{\mathbf{D}}^{(+)}(\mathbf{r},t),\hat{\mathbf{A}}^{(+)}(\mathbf{r}',t)]=0, \tag{2.27}$$

$$[\hat{\mathbf{D}}^{(+)}(\mathbf{r},t),\hat{\mathbf{A}}^{(-)}(\mathbf{r}',t)]=\frac{i\hbar}{2}\bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}'), \tag{2.28}$$

where

$$\bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}') = \sum_{\mathbf{k}} \mathbf{f}_{\mathbf{k}}(\mathbf{r})[\bar{\bar{\mathcal{M}}}^t \mathbf{f}_{\mathbf{k}}^*(\mathbf{r}')]. \tag{2.29}$$

Here $\bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}')$ is a generalized transverse δ function [30]. This may be seen by considering its action on some transverse vector field $[\boldsymbol{\nabla}\cdot\boldsymbol{\psi}(\mathbf{r},t)=0]$. Since $\{\mathbf{f}_{\mathbf{k}}\}$ is a complete basis on the set of transverse fields, we may expand $\boldsymbol{\psi}(\mathbf{r},t)$ as

$$\boldsymbol{\psi}(\mathbf{r},t) = \sum_{\mathbf{k}} \hat{C}_{\mathbf{k}}(t)\mathbf{f}_{\mathbf{k}}(\mathbf{r}). \tag{2.30}$$

If we calculate the effect of the transverse δ function on a transverse field we find

$$\begin{aligned}\int d^3r' \bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}')\cdot\boldsymbol{\psi}(\mathbf{r}',t) &= \int d^3r' \sum_{\mathbf{k}\mathbf{k}'} \hat{C}_{\mathbf{k}}(t)\mathbf{f}_{\mathbf{k}'}(\mathbf{r}) \\ &\times[\bar{\bar{\mathcal{M}}}^t \mathbf{f}_{\mathbf{k}'}^*(\mathbf{r}')\cdot\mathbf{f}_{\mathbf{k}}(\mathbf{r}')]\sum_{\mathbf{k}\mathbf{k}'} \hat{C}_{\mathbf{k}}(t)\mathbf{f}_{\mathbf{k}'}(\mathbf{r})\delta_{\mathbf{k}\mathbf{k}'} = \boldsymbol{\psi}(\mathbf{r},t),\end{aligned} \tag{2.31}$$

where we have used the orthonormality condition of the basis functions.

We shall also need the equal-space commutation relations

$$[\hat{\mathbf{D}}^{(+)}(\mathbf{r},t),\hat{\mathbf{D}}^{(-)}(\mathbf{r},t')].$$

A formal expression of this commutation relation can be found from Eq. (2.23a) to be

$$[\hat{\mathbf{D}}^{(+)}(\mathbf{r},t),\hat{\mathbf{D}}^{(-)}(\mathbf{r},t')] = \frac{\hbar\epsilon_0}{2}\bar{\bar{\eta}}(\mathbf{r},t,t'), \tag{2.32}$$

where

$$\bar{\bar{\eta}}(\mathbf{r},t,t') = \sum_{\mathbf{k}} \omega_{\mathbf{k}}\mathbf{f}_{\mathbf{k}}(\mathbf{r})\mathbf{f}_{\mathbf{k}}^*(\mathbf{r})e^{-i\omega_{\mathbf{k}}(t-t')}. \tag{2.33}$$

In vacuum $\bar{\bar{\eta}}(\mathbf{r},t,t')$ is simple to evaluate, but for complex systems it is nontrivial to gain knowledge of the basis functions $\{\mathbf{f}_{\mathbf{k}}\}$. In Appendix B we calculate $\bar{\bar{\eta}}$ using the rotating-wave approximation and the local density approximation, where we assume that $\rho(\mathbf{r})$ varies slowly with respect to $\mathbf{r}$.

III. EQUATIONS OF MOTION

In this section we derive the equations of motion for the system, and consider their general properties. In the preceding section we discussed that the theory could be divided into an average part and a part representing the deviation from the average. To derive the equations of motion we will, however, work with the full Hamiltonian and later make the splitting into the average part and the deviations from it. The strategy we will use is to first derive the quantum mechanical Maxwell equations, and then to combine them into an effective wave equation for the field.

We will now, as an example, derive one of the quantum mechanical Maxwell equations from Heisenberg's equation of motion,

$$\begin{aligned}\frac{d}{dt}\hat{\mathbf{D}}(\mathbf{r}) &= \frac{i}{\hbar}[\hat{\mathcal{H}},\hat{\mathbf{D}}(\mathbf{r})] = \frac{i}{2\hbar\mu_0}\int d^3r'\{[\boldsymbol{\nabla}\times\hat{\mathbf{A}}(\mathbf{r}')]^2,\hat{\mathbf{D}}(\mathbf{r})\} \\ &= \frac{i}{2\hbar\mu_0}\int d^3r'\{[\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\hat{\mathbf{A}}(\mathbf{r}')]\cdot[\hat{\mathbf{A}}(\mathbf{r}'),\hat{\mathbf{D}}(\mathbf{r})] \\ &+ [\hat{\mathbf{A}}(\mathbf{r}'),\hat{\mathbf{D}}(\mathbf{r})]\cdot[\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\hat{\mathbf{A}}(\mathbf{r}')]\}.\end{aligned} \tag{3.1}$$

Here we have used the Hamiltonian given in Eq. (2.9), and the boundary condition that the physical fields vanish at infinity. To shorten the notation we have suppressed the explicit time dependence. The commutation relation may be found from (2.27) and (2.28) to be

$$[\hat{\mathbf{A}}(\mathbf{r}'),\hat{\mathbf{D}}(\mathbf{r})] = -i\hbar\bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}'). \tag{3.2}$$

Since the field $\boldsymbol{\nabla}\times\hat{\mathbf{A}}$ is transverse by definition, this gives us the first quantum mechanical Maxwell equation

$$\frac{d}{dt}\hat{\mathbf{D}}(\mathbf{r}) = \frac{1}{\mu_0}\boldsymbol{\nabla}\times\hat{\mathbf{B}}(\mathbf{r}), \tag{3.3}$$

where

$$\hat{\mathbf{B}}(\mathbf{r}) = \boldsymbol{\nabla}\times\hat{\mathbf{A}}(\mathbf{r}). \tag{3.4}$$

Similarly we may derive the Maxwell equation $\boldsymbol{\nabla}\times\hat{\mathbf{E}} = -\partial_t\hat{\mathbf{B}}$, where $\hat{\mathbf{E}} = -d\hat{\mathbf{A}}/dt = \hat{\mathbf{D}} - \hat{\mathbf{P}}$. The remaining Maxwell equations $\boldsymbol{\nabla}\cdot\hat{\mathbf{B}}=0$ and $\boldsymbol{\nabla}\cdot\hat{\mathbf{D}}=0$ follow immediately from the definition of $\hat{\mathbf{B}}$ in Eq. (3.4) and from the transversality of $\hat{\mathbf{D}}$.

Because of the nature of the interaction part of the Hamiltonian, it is convenient to consider the two frequency components of the displaced electric field separately. The quantum mechanical Maxwell equations may be combined into a single wave equation

$$\begin{aligned}\left(\frac{d^2}{dt^2} + c^2\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\right)\hat{\mathbf{D}}^{(-)}(\mathbf{r},t) \\ = c^2\int d^3r\,\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}')\cdot\bar{\bar{V}}[\hat{\mathbf{J}}]^t\hat{\mathbf{D}}^{(-)}(\mathbf{r}',t),\end{aligned} \tag{3.5}$$

where the positive frequency part can be found by Hermitian conjugation. Similarly we may derive equations for the spin of the atoms, and for the simple interactions given in Eq. (2.7), one finds

$$\frac{d}{dt}\hat{\mathbf{J}}(\mathbf{r},t) = \frac{i\beta c_1}{\hbar\epsilon_0}\hat{\mathbf{J}}(\mathbf{r},t)\times[\hat{\mathbf{D}}^{(-)}(\mathbf{r},t)\times\hat{\mathbf{D}}^{(+)}(\mathbf{r},t)]. \tag{3.6}$$

In the remainder of this paper we will solve these coupled partial differential equations.

The expression in Eq. (3.5) is a second-order differential equation in time. The solution of this equation will in general not only depend on the initial value $\mathbf{D}(\mathbf{r},t=t_0)$, but also the time derivative $\partial_t\mathbf{D}(\mathbf{r},t)|_{t=t_0}$. In deriving our interaction we have, however, already used the rotating-wave approximation, where we ignore the dynamics on a time scale similar to the inverse of the optical frequency. Similarly we shall here make a slowly varying envelope approximation and write the displaced electric field as

$$\hat{\mathbf{D}}(\mathbf{r},t) = \tilde{\mathbf{D}}^{(-)}(\mathbf{r},t)e^{i\omega_L t} + \tilde{\mathbf{D}}^{(+)}(\mathbf{r},t)e^{-i\omega_L t}, \tag{3.7}$$

where $\tilde{\mathbf{D}}^{(\pm)}$ are slowly varying in time. If we ignore the second derivative of the slowly varying operators $(\partial_t^2 \tilde{\mathbf{D}}^{(\pm)} \times(\mathbf{r},t) \approx 0)$, then Eq. (3.5) reduces to a first-order differential equation in time.

Since we are heading toward a perturbation theory in the point particle part of the Hamiltonian (2.12), we will add and subtract the average part of the source term in Eq. (3.5). That is we write

$$\bar{\bar{V}}[\hat{\mathbf{J}}] = \bar{\bar{V}}[\hat{\mathbf{J}}] - \bar{\bar{\mathcal{V}}}[\mathbf{J}] + \bar{\bar{\mathcal{V}}}[\mathbf{J}] \equiv \bar{\bar{m}}[\hat{\mathbf{J}}] + \bar{\bar{\mathcal{V}}}[\mathbf{J}]. \tag{3.8}$$

The idea in this separation is that now $\bar{\bar{\mathcal{V}}}[\mathbf{J}]$ represents the average effect of the ensemble, which may have a big effect, whereas $\bar{\bar{m}}[\hat{\mathbf{J}}]$ represents the fluctuations around this average. To take advantage of this we first consider the average term

$$\int d^3r\, \boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}') \cdot \bar{\bar{\mathcal{V}}}[\mathbf{J}]^t \hat{\mathbf{D}}^{(-)}(\mathbf{r}',t). \tag{3.9}$$

This term is continuous and we may use partial integration 2 times. Using the expression for the general transverse δ function one finds

$$\int d^3r\, \boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}') \cdot \bar{\bar{\mathcal{V}}}[\mathbf{J}]^t \hat{\mathbf{D}}^{(-)}(\mathbf{r}',t)$$

$$= \boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \bar{\bar{\mathcal{V}}}[\mathbf{J}]^t \hat{\mathbf{D}}^{(-)}(\mathbf{r},t). \tag{3.10}$$

We move this term to the left-hand side of Eq. (3.5), and we are left with a diffusion equation involving only the fluctuations as a source term on the right-hand side,

$$\left(2i\omega_L \frac{d}{dt} - \omega_L^2 + c^2\, \boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \bar{\bar{\mathcal{M}}}^t\right) \tilde{\mathbf{D}}^{(-)}(\mathbf{r},t)$$

$$= c^2 \int d^3r\, \boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}') \cdot \bar{\bar{m}}[\hat{\mathbf{J}}]^t \tilde{\mathbf{D}}^{(-)}(\mathbf{r}',t). \tag{3.11}$$

If we set the right-hand side of this equation to zero, i.e., ignore the fluctuations, this equation describes the propagation and diffraction of the field in a medium. For instance if we take the simplest case where the medium is isotropic so that the matrix $\bar{\bar{\mathcal{V}}}[\mathbf{J}]$ is just a scalar, this equation describes the propagation through a medium with an index of refraction given by $n = 1/\sqrt{1 - \bar{\bar{\mathcal{V}}}[\mathbf{J}]}$, see Ref. [30].

IV. GENERAL SOLUTION AND FEYNMAN DIAGRAMS

In this section we discuss the solution of Eq. (3.11) in terms of its Green's function. Let us for convenience define the differential operator

$$\mathcal{D} = 2i\omega_L \frac{d}{dt} - \omega_L^2 + c^2\, \boldsymbol{\nabla} \times \boldsymbol{\nabla} \times \bar{\bar{\mathcal{M}}}^t(\mathbf{r}). \tag{4.1}$$

We then define the Green's function by

$$\mathcal{D}\bar{\bar{G}}^{(-)}(\mathbf{r},t|\mathbf{r}_0,t_0) = \bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}_0)\delta(t-t_0). \tag{4.2}$$

The right-hand side of this equation describes an identity functional on the inner product space we are working in. We want the Green's function to describe an evolution of the system forward in time. We therefore define a cutoff on the Green's function in time

$$\bar{\bar{G}}^{(-)}(\mathbf{r},t|\mathbf{r}_0,t_0) = 0 \quad \text{for } t < t_0. \tag{4.3}$$

The general solution to Eq. (3.11) in terms of Green's functions is discussed in detail in Appendix C, and reads as

$$\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t) = 2i\omega_L \int d^3r'\, \bar{\bar{\mathcal{M}}}^t(\mathbf{r}')\bar{\bar{G}}^{(-)}(\mathbf{r},t|\mathbf{r}',t_0) \cdot \tilde{\mathbf{D}}^{(-)}(\mathbf{r}',t_0)$$

$$+ c^2 \int \int_{t_0}^{t^+} d^3r'\, dt'\, \bar{\bar{\mathcal{M}}}^t(\mathbf{r}')$$

$$\times \bar{\bar{G}}^{(-)}(\mathbf{r},t|\mathbf{r}',t') \cdot \int d^3r''\, \boldsymbol{\nabla}' \times \boldsymbol{\nabla}'$$

$$\times \bar{\bar{\delta}}^T(\mathbf{r}',\mathbf{r}'') \cdot \bar{\bar{m}}[\hat{\mathbf{J}}]^t \tilde{\mathbf{D}}^{(-)}(\mathbf{r}'',t'). \tag{4.4}$$

The upper limit is understood to be $t^+ = \lim_{\epsilon \to 0}(t+\epsilon)$. Before continuing a few comments are in order. Here we have used the boundary conditions, that all fields vanish at infinity, i.e., we imagine that at time $t=0$ we have generated an optical pulse inside the volume we are describing, which travels toward the atomic medium. Alternatively we could have described the incoming field by a boundary term. The positive frequency part may be found by Hermitian conjugation.

Let us now consider the last term of Eq. (4.4). We notice that the involved fields are all continuous and differentiable with respect to the primed spatial coordinates. Using partial integration 2 times and introducing the propagator defined by

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r}',t') = \boldsymbol{\nabla}' \times \boldsymbol{\nabla}' \times \bar{\bar{\mathcal{M}}}^t(\mathbf{r}')\bar{\bar{G}}^{(-)}(\mathbf{r},t|\mathbf{r}',t') \tag{4.5}$$

the last term of Eq. (4.4) may be written as

$$c^2 \int \int_{t_0}^{t} d^3r'\, dt' \int d^3r'' \bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r}',t')$$

$$\cdot \bar{\bar{\delta}}^T(\mathbf{r}',\mathbf{r}'') \cdot \bar{\bar{m}}[\hat{\mathbf{J}}]^t \tilde{\mathbf{D}}^{(-)}(\mathbf{r}'',t'). \tag{4.6}$$

Due to the cross product in Eq. (4.5) the propagator is transverse with respect to primed coordinates and the transverse δ function in (4.6) may be integrated out, giving

$$c^2 \int \int_{t_0}^{t} d^3r'\, dt'\, \bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r}',t') \cdot \bar{\bar{m}}[\hat{\mathbf{J}}]^t \tilde{\mathbf{D}}^{(-)}(\mathbf{r}',t'). \tag{4.7}$$

We denote the first term on the right-hand side of Eq. (4.4) as $\tilde{\mathbf{D}}_0^{(-)}(\mathbf{r},t)$,

$$\tilde{\mathbf{D}}_0^{(-)}(\mathbf{r},t) = 2i\omega_L \int d^3r' \bar{\bar{M}}^t(\mathbf{r}') \bar{\bar{G}}^{(-)}(\mathbf{r},t|\mathbf{r}',t_0) \cdot \tilde{\mathbf{D}}^{(-)}(\mathbf{r}',t_0). \tag{4.8}$$

If there were no deviation from the mean, i.e., $\bar{\bar{m}}[\hat{\mathbf{J}}]=0$, the solution would simply be $\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t)=\tilde{\mathbf{D}}_0^{(-)}(\mathbf{r},t)$. $\tilde{\mathbf{D}}_0^{(-)}(\mathbf{r},t)$ thus denotes the solution to the diffraction problem, where the atomic medium is treated as a continuous medium with a diffraction matrix $\bar{\bar{\mathcal{M}}}$.

A. Perturbative expansion

Below we shall develop a perturbative expansion in the deviation from the mean due to quantum fluctuations and from the fact that the medium is not continuous but consists of a large number of point particles. The starting point for the perturbative expansion will be the *field equation*

$$\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t) = \tilde{\mathbf{D}}_0^{(-)}(\mathbf{r},t) + c^2 \int \int_{t_0}^{t} d^3r' dt' \times \bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r}',t') \cdot \bar{\bar{m}}[\hat{\mathbf{J}}]^t \tilde{\mathbf{D}}^{(-)}(\mathbf{r}',t'). \tag{4.9}$$

In addition to this we shall also need the solution to the equations of motion for the spin (3.6), which may be formally solved to give the *spin equation*

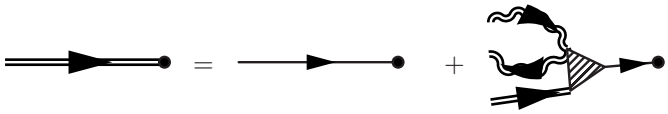
$$\hat{\mathbf{J}}(\mathbf{r},t) = \hat{\mathbf{J}}(\mathbf{r},t_0) + \frac{i\beta c_1}{\hbar \epsilon_0} \int_{t_0}^{t} dt' \hat{\mathbf{J}}(\mathbf{r},t') \times [\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t') \times \tilde{\mathbf{D}}^{(+)}(\mathbf{r},t')]. \tag{4.10}$$

These are the equations we wish to treat using the Born approximation, where we make an expansion in the interaction parameter β. [In Eq. (4.9) the interaction $\bar{\bar{m}}[\hat{\mathbf{J}}]^t$ is proportional to the expansion parameter β.]

In terms of notation this expansion gets extremely cumbersome. It is therefore convenient to introduce Feynman diagrams to represent the various terms of the expansion. We will be dealing with two types of interactions: the one given in Eq. (4.9) which we will represent with a shaded circle, and the one given in Eq. (4.10) which we will represent with a shaded triangle. We diagrammatically represent the field equation as

(4.11)

and the spin equation is represented as



(4.12)

The orientation of the diagram is such that time is going from left to right, and the evaluation at time t is marked by a dot. Spin propagation is represented by a line with an arrow pointing in the positive-time direction. A wiggly line represents propagation of the displaced electric field. The arrow denotes whether the line represents the photon-generating part of the field, $\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t)$, where the arrow points forward in time, or the photon-annihilating part of the field, where the arrow points backward in time. The full solution to the spin $\hat{\mathbf{J}}(t)$ is denoted with a double straight line, and the full solution to the displaced electric field is denoted with a double wiggly line.

The field equation and the spin equation can be represented as a perturbation series, and in the following we shall discuss the effect of the terms in this perturbation series. An important feature of our system is the random distribution of the atoms in the ensemble. The equations that we have derived so far apply to each realization of the atomic distribution $\{\mathbf{r}_1,\mathbf{r}_2,\ldots,\mathbf{r}_N\}$. However, since we have no control of the position of the atoms we must make a spatial average of our equations, that is of the terms in the perturbation series. To do this we need to know the density correlations of the gas.

B. Density correlations

We assume that we are dealing with an ideal gas, i.e., we assume that the distribution of the atoms is completely random but has a distribution given by the possible spatially varying density $\rho(\mathbf{r})$, and we assume that there are no correlations between the positions of different atoms. The correlation function for the density distribution $\rho(\mathbf{r})=\Sigma_j \delta(\mathbf{r}-\mathbf{r}_j)$ is thus

$$\langle \rho(\mathbf{r})\rho(\mathbf{r}')\rangle_{\text{s.a.}} = \left\langle \sum_{jl} \delta(\mathbf{r}-\mathbf{r}_j)\delta(\mathbf{r}'-\mathbf{r}_l)\right\rangle_{\text{s.a.}}$$
$$= \sum_{j\neq l} \langle \delta(\mathbf{r}-\mathbf{r}_j)\delta(\mathbf{r}'-\mathbf{r}_l)\rangle_{\text{s.a.}}$$
$$+ \sum_{j} \delta(\mathbf{r}-\mathbf{r}')\langle \delta(\mathbf{r}-\mathbf{r}_j)\rangle_{\text{s.a.}}$$
$$= \langle \rho(\mathbf{r})\rangle_{\text{s.a.}}\langle \rho(\mathbf{r}')\rangle_{\text{s.a.}} + \delta(\mathbf{r}-\mathbf{r}')\langle \rho(\mathbf{r})\rangle_{\text{s.a.}}. \tag{4.13}$$

Here $\langle\cdots\rangle_{\text{s.a.}}$ denotes spatial averaging. In the last step we used that the distribution is independent for different atoms, and we ignored the small difference between N_A^2 and $N_A(N_A-1)$, where N_A is the number of atoms. We have also neglected the effect that two different atoms cannot be found at the same point in space. While this may seem insignificant for a low density gas, we show in Appendix D that including this effect to all orders in the perturbation series gives the Lorentz-Lorenz correction to the index of refraction.

Below we shall also use the correlation functions for the spin. Similar to the calculation above we find

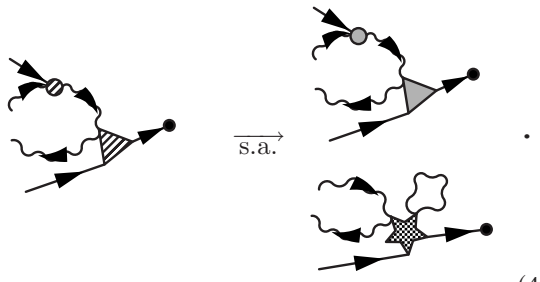
$$\langle \hat{J}_n(\mathbf{r})\hat{J}_m(\mathbf{r}')\rangle_{\text{s.a.}} = \rho(\mathbf{r})\rho(\mathbf{r}')\bar{J}_n(\mathbf{r})\bar{J}_m(\mathbf{r}')$$
$$+ \rho(\mathbf{r})\delta(\mathbf{r}-\mathbf{r}')\bar{J}_n(\mathbf{r})\bar{J}_m(\mathbf{r}), \tag{4.14}$$

where the index n,m refer to the spatial components of the operators. To shorten notation we have written $\rho(\mathbf{r})$ instead of $\langle\rho(\mathbf{r})\rangle_{\text{s.a.}}$. As discussed previously, the bar denotes a single atom operator. We will preserve the quantum mechanical behavior of the operators by not taking the quantum mechanical mean. The first term on the right-hand side of Eq. (4.14) arises from the contribution from different atoms (signified by the prime on the second spin operator). In the second term, on the other hand, the two operators refer to the same atom, and the operator product should be evaluated for a single atom. For example, for a spin-$\frac{1}{2}$ system, we have the following relation between products of spin operators on single atoms:

$$\bar{J}_n(\mathbf{r})\bar{J}_m(\mathbf{r}) = \frac{i}{2}\epsilon_{nml}\bar{J}_l(\mathbf{r}). \tag{4.15}$$

The generalization to even higher order density correlations is straightforward.

These considerations become important when we calculate the spatial average of the second-order terms of the perturbation series. Let us, as an example, consider the second-order term of the spin equation representing a photon first interacting with one atom and then later with the atom in consideration,



(4.16)

When taking spatial average, this term generates two terms in the perturbative expansion as indicated with the arrow in Eq. (4.16). The first term involving the spin of two different atoms is referred to as a coherent interaction, which we will discuss later. The second term involving the δ function corresponds to the incoherent interaction (for reasons which will become clear below). We include this situation in the diagrammatic notation by introducing a hatched star and a loop signifying the infinitely short propagation stemming from the δ-function term of the correlation function equation (4.14), i.e.,

$$\int d^3r \bar{\bar{P}}^{\pm}(\mathbf{r},t|\mathbf{r}',t')\cdot \boldsymbol{\psi}(\mathbf{r}',t')\delta(\mathbf{r}-\mathbf{r}')$$
$$= \bar{\bar{P}}^{\pm}(\mathbf{r},t|\mathbf{r},t')\cdot \boldsymbol{\psi}(\mathbf{r},t'). \tag{4.17}$$

The loop is placed on the top of the star when it comes from the positively oscillating propagator $\bar{\bar{P}}^{(-)}$, and in the bottom of the star when we refer to the negatively oscillating propagator $\bar{\bar{P}}^{(+)}$. A star scales with the expansion coefficient β squared since it involves two interactions. In the next section we will calculate the infinitely short propagator appearing in these expressions in the local density approximation.

C. Green's function and propagator

In this section we first derive a formal expression for the Green's function. Within our inner product space the Green's function is defined by (4.1) and (4.2). Expanding our Green's function in the basis $\mathbf{f}_{\mathbf{k}}^*(\mathbf{r})$ we find the representation

$$\bar{\bar{G}}^{(-)}(\mathbf{r},t|\mathbf{r}',t') = \sum_{\mathbf{k}} \mathbf{f}_{\mathbf{k}}^*(\mathbf{r})\mathbf{f}_{\mathbf{k}}(\mathbf{r}')g_{\mathbf{k}}^{(-)}(t,t'). \tag{4.18}$$

We have here expanded on the complex conjugated set $\mathbf{f}_{\mathbf{k}}^*(\mathbf{r})$ to match the expansion of the displaced electric field in Eq. (2.23a). The transverse δ function has the representation

$$\bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}') = \sum_{\mathbf{k}} \mathbf{f}_{\mathbf{k}}^*(\mathbf{r})\mathbf{f}_{\mathbf{k}}(\mathbf{r}'), \tag{4.19}$$

where we are now working in the inner-product space with inner product defined in Eq. (2.18). The scalar function $g_{\mathbf{k}}^{(-)}(t,t')$ is defined by

$$\left(2i\omega_L\frac{d}{dt} - \omega_L^2 + \omega_{\mathbf{k}}^2\right)g_{\mathbf{k}}^{(-)}(t,t') = \delta(t-t'), \tag{4.20}$$

along with the condition that the function $g_{\mathbf{k}}(t,t')$ vanish for

$t<t'$. We will consider the following form of the scalar function, where we explicitly write this cutoff in terms of a step function

$$g_{\mathbf{k}}^{(-)}(t,t') = Ce^{i\gamma_{\mathbf{k}}(t-t')}\Theta(t-t'). \tag{4.21}$$

The coefficients $\gamma_{\mathbf{k}}$ and C is found by inserting this result into Eq. (4.20).

$$\gamma_{\mathbf{k}} = \frac{\omega_{\mathbf{k}}^2 - \omega_L^2}{2\omega_L} \approx \omega_{\mathbf{k}} - \omega_L, \tag{4.22a}$$

$$C = \frac{-i}{2\omega_L}. \tag{4.22b}$$

The Green's function is thus given by

$$\bar{\bar{G}}^{(-)}(\mathbf{r},t|\mathbf{r}',t') = -i\sum_{\mathbf{k}} \mathbf{f}_{\mathbf{k}}^*(\mathbf{r})\mathbf{f}_{\mathbf{k}}(\mathbf{r}')\frac{e^{i(\omega_{\mathbf{k}}-\omega_L)(t-t')}}{2\omega_L}\Theta(t-t'). \tag{4.23}$$

Next we will look at the infinitely short propagator in Eq. (4.17). Using the Green's function given in Eq. (4.23) along with definition (2.17a) and (4.5) the propagator may be written as

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r},t') = \frac{-i}{2\omega_L c^2}\sum_{\mathbf{k}} \omega_{\mathbf{k}}^2\mathbf{f}_{\mathbf{k}}^*(\mathbf{r})\mathbf{f}_{\mathbf{k}}(\mathbf{r})e^{i(\omega_{\mathbf{k}}-\omega_L)(t-t')}, \tag{4.24}$$

where we have omitted the step function since it automatically gives unity for the integration limits we are using here. We will now relate this infinitely short propagator to some already known parameter. If we go back and consider the general result for the equal-space commutator, this may in terms of the basis functions $\{\mathbf{f}_{\mathbf{k}}\}$ be written as

$$[\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t);\tilde{\mathbf{D}}^{(+)}(\mathbf{r},t')] = -\frac{\hbar\epsilon_0}{2}\sum_{\mathbf{k}} \omega_{\mathbf{k}}\mathbf{f}_{\mathbf{k}}^*(\mathbf{r})\mathbf{f}_{\mathbf{k}}(\mathbf{r})e^{i(\omega_{\mathbf{k}}-\omega_L)(t-t')}. \tag{4.25}$$

Comparing with (4.24) we immediately get a formal relationship between this commutator and the infinitely short propagator

$$\left(\frac{d}{dt'} - i\omega_L\right)[\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t);\tilde{\mathbf{D}}^{(+)}(\mathbf{r},t')] = -\hbar\epsilon_0\omega_L c^2\bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r},t'). \tag{4.26}$$

Using Eq. (2.32) this relation can also be written as

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r},t') = \frac{1}{2c^2}\left(\frac{d}{dt'} - i\omega_L\right)\bar{\bar{\eta}}^{*t}(\mathbf{r},t,t'). \tag{4.27}$$

To illustrate how the indefinitely short propagator enters into the equations we will again consider the second-order term in the spin equation represented in Eq. (4.16). The term prior to spatial average is given as

$$\frac{i\beta c_1 c^2}{\hbar\epsilon_0}\int_{t_0}^{t} dt'\hat{\mathbf{J}} \times \left(\int\int_{t_0}^{t'} dt''d^3r'[\bar{\bar{P}}^{(-)}(\mathbf{r},t'|\mathbf{r}',t'') \cdot \bar{\bar{m}}[\hat{\mathbf{J}}]^t\tilde{\mathbf{D}}_0^{(-)}(\mathbf{r}',t'')] \times \tilde{\mathbf{D}}_0^{(+)}(\mathbf{r},t')\right). \tag{4.28}$$

After spatial average we get two terms, representing the coherent and the incoherent interaction. The incoherent interaction may then be written as

$$\frac{i\beta c_1}{2\hbar\epsilon_0}\int_{t_0}^{t} dt'\int_{t_0}^{t'} dt''\bar{\mathbf{J}}(\mathbf{r}) \times \left[\left(\frac{\partial}{\partial t''} - i\omega_L\right)[\bar{\bar{\eta}}^{*t}(\mathbf{r},t',t'')\bar{\bar{\mathcal{V}}}^t[\bar{\mathbf{J}}] \times \tilde{\mathbf{D}}_0^{(-)}(\mathbf{r},t'')] \times \tilde{\mathbf{D}}_0^{(+)}(\mathbf{r},t')\right]. \tag{4.29}$$

To simplify notation, we have signified spatial averaging with calligraphic letters, e.g., $\langle \mathbf{D}(\mathbf{r},t)\rangle_{\text{s.a.}} \equiv \mathcal{D}(\mathbf{r},t)$. This convention will be used in the remainder of this paper.

We have now developed all the necessary theoretical tools to describe the system. In the next section we shall use these tools to discuss a perturbative expansion of the evolution of the system.

V. TIME EVOLUTION

This section is divided into three parts. In the first part we examine the general behavior of the atomic spin in the presence of a light field. The aim is to understand the effect of the loops introduced in the Feynman diagrams. In the second part we consider the light field and we show how the theory introduce a decay of the field strength of the light as it interacts with the atoms. Again this is connected to the loops introduced in the Feynman diagrams. Finally we will introduce and discuss Stokes operators, which are the appropriate operators for describing the experiments in Refs. [8–10].

A. Evolution of the spin

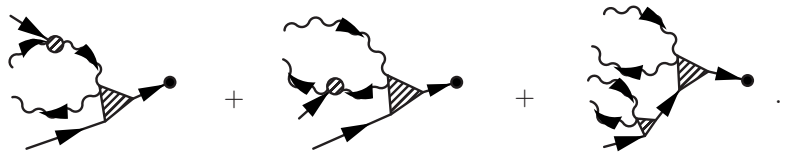
In this section we will consider the spin equation in detail for the simple interaction (4.10). We will begin our analysis by considering the first-order term in the perturbative expansion of the solution to the spin equation, formally given by the diagram

. (5.1)

This term gives no extra contributions when doing the spatial averaging, and we readily write the expression describing this term,

$$\frac{i\beta c_1\rho(\mathbf{r})}{\hbar\epsilon_0}\int_{t_0}^{t} dt'\bar{\mathbf{J}}(\mathbf{r},t_0) \times [\tilde{\mathcal{D}}_0^{(-)}(\mathbf{r},t') \times \tilde{\mathcal{D}}_0^{(+)}(\mathbf{r},t')]. \tag{5.2}$$

We now continue with the second-order terms represented by the following Feynman diagrams:

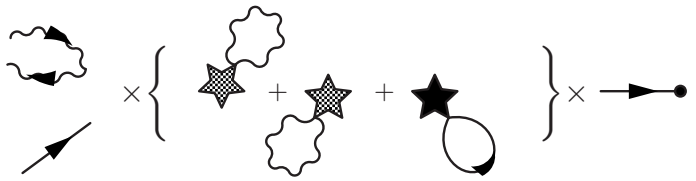


(5.3)

When taking spatial average of these terms, we have argued that the first two diagrams will give an additional set of Feynman diagrams containing loops and stars. It still remains to consider the last diagram of (5.3), representing two photons interacting with the same atom at time t and t'. In this diagram it is necessary to pay special attention to the case where the two interactions happen at the same time $t=t'$. The contribution of this term is proportional to $\mathcal{D}^{(-)}(t'')\mathcal{D}^{(+)}(t'')\mathcal{D}^{(-)}(t')\mathcal{D}^{(+)}(t')$ which is not normal ordered, and it will be convenient to separate it into normal-ordered terms. When commuting $\mathcal{D}^{(-)}(\mathbf{r},t'')$ and $\mathcal{D}^{(+)}(\mathbf{r},t')$ we once again get an infinitely short propagator, cf. (2.32). This extra term will be denoted by a filled star with a loop. This commutator term will produce an interaction which is linear in the field intensity (involves $\mathcal{D}^{(-)}\mathcal{D}^{(+)}$), whereas the normally ordered term ($\mathcal{D}^{(-)}\mathcal{D}^{(-)}\mathcal{D}^{(+)}\mathcal{D}^{(+)}$) will be quadratic in the intensity. Ignoring for now this quadratic term as well as the coherent interactions, the second-order diagrams for the spin equation after spatial average reads as

(5.4)

which can also be written as



(5.5)

The interpretation of the diagrams is given below.

To simplify the expression we will make the slowly varying envelope approximation which simplifies Eq. (4.27) to

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r},t') \approx \frac{-i\omega_L}{2c^2}\bar{\bar{\eta}}^{*t}(\mathbf{r},t,t'). \tag{5.6}$$

Second, we shall evaluate η in a local density approximation, where we assume that $\eta(\mathbf{r},t,t')$ is the same as if we were in an infinite medium with a constant density $\rho(\mathbf{r})$ and spin density $\mathbf{J}(\mathbf{r})$. By doing this we ignore the reflection of the field on the surface of the ensemble or other inhomogeneities. The infinitely short propagator which expresses the amplitude for the field to be found at the same position at some later time, therefore becomes a δ function in time. This approximation is valid provided that the diffraction matrix $\bar{\bar{\mathcal{M}}}(\mathbf{r})$ varies slowly on the scale of the wavelength of the light. Furthermore, $\bar{\bar{\eta}}(\mathbf{r},t,t')$ also contain the Lamb shift which we ignore for simplicity. A detailed calculation of $\bar{\bar{\eta}}$ is presented in Appendix B, where we find

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r},t') = \frac{-i\delta(t-t')}{c^2}\begin{bmatrix} \varrho_{\parallel}(\mathbf{r}) & 0 & 0 \\ 0 & \varrho_{\perp}(\mathbf{r}) & -i\varrho_{\Gamma}(\mathbf{r}) \\ 0 & i\varrho_{\Gamma}(\mathbf{r}) & \varrho_{\perp}(\mathbf{r}) \end{bmatrix} \equiv \frac{-i\delta(t-t')}{c^2}\bar{\bar{A}}^{(-)}(\mathbf{r}), \tag{5.7}$$

where the coefficients $\varrho_{\parallel}$, $\varrho_{\perp}$, and ϱ_{Γ} may be found in Eq. (B13). Here the result is given in a Euclidean basis, where $\mathbf{J}$ is assumed to be along the x axis. The result may also be expressed in a coordinate-independent form as

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r},t') = \frac{-i\delta(t-t')}{c^2}\{[\varrho_{\perp}(\mathbf{r}) - i\gamma(\mathbf{r})\hat{\mathbf{j}}\times\ ] + [\varrho_{\parallel}(\mathbf{r}) - \varrho_{\perp}(\mathbf{r})]\hat{\mathbf{j}}(\hat{\mathbf{j}}\cdot\ )\}, \tag{5.8}$$

where $\hat{\mathbf{j}}$ is a unit vector parallel to $\mathbf{J}$. This infinitely short propagator is inserted into the second-order terms in the spin equation. The second-order incoherent interaction given in Eq. (5.5) then reads

$$\frac{\beta^2}{\hbar\epsilon_0}\int_{t_0}^{t} dt' \Big(c_1 c_0 \bar{\mathbf{J}}^2 [\bar{\bar{A}}^{(-)}\mathcal{D}_0^{(-)}(\bar{\mathbf{J}}\cdot\mathcal{D}_0^{(+)}) - \mathcal{D}_0^{(-)}(\bar{\mathbf{J}}\cdot\bar{\bar{A}}^{(+)}\mathcal{D}_0^{(+)}) + \text{H.c.}] + \frac{c_1^2}{2}[\bar{\bar{A}}^{(+)}\mathcal{D}_0^{(-)}(\bar{\mathbf{J}}\cdot\mathcal{D}_0^{(+)}) - (\mathcal{D}_0^{(-)}\cdot\mathcal{D}_0^{(+)})\bar{\bar{A}}^{(-)}\bar{\mathbf{J}} - \mathrm{Tr}[\bar{\bar{A}}^{(-)}]\mathcal{D}_0^{(-)}(\bar{\mathbf{J}}\cdot\mathcal{D}_0^{(+)}) + \mathcal{D}_0^{(-)}(\bar{\mathbf{J}}\cdot\bar{\bar{A}}^{(-)}\mathcal{D}_0^{(+)}) + \text{H.c.}] \Big), \tag{5.9}$$

where we have suppressed the space and time dependencies.

In the simple case, where the matrix $\bar{\bar{A}}^{(\pm)}$ is proportional to the identity matrix ($\varrho_\Gamma \approx 0$, $\varrho_\parallel \approx \varrho_\perp = \varrho$), which is the case to lowest order, the terms proportional to $c_1 c_0$ cancels and the expression reduces to

$$-\frac{\beta^2 c_1^2 \varrho}{2\hbar\epsilon_0}\int_{t_0}^{t} dt' [(\mathcal{D}_0^{(-)}\cdot\mathcal{D}_0^{(+)})\bar{\mathbf{J}} + \mathcal{D}_0^{(-)}(\bar{\mathbf{J}}\cdot\mathcal{D}_0^{(+)}) + \text{H.c.}]. \tag{5.10}$$

This term scale with the power of the incident light, and linearly polarized light, will affect the spin component parallel to the field with 2 times the rate than the perpendicular spin components. To see this we may introduce a decay rate $\Gamma_{\mathcal{D}}$, and write expression (5.10) in a differential form, we thus see that the term indeed describes a decay of the spin components

$$\partial_t \bar{J}_x = -2\Gamma_{\mathcal{D}}\bar{J}_x, \tag{5.11a}$$

$$\partial_t \bar{J}_y = -\Gamma_{\mathcal{D}}\bar{J}_y, \tag{5.11b}$$

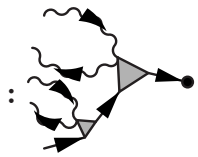
$$\partial_t \bar{J}_z = -\Gamma_{\mathcal{D}}\bar{J}_z, \tag{5.11c}$$

where

$$\Gamma_{\mathcal{D}} = \frac{\beta^2 c_1^2 \varrho}{\hbar\epsilon_0}\langle \mathcal{D}_{0,x}^{(-)}\mathcal{D}_{0,x}^{(+)}\rangle,$$

and where we have assumed that the light is linearly polarized in the x direction.

Let us now turn to the coherent part of the interaction represented by the Feynman diagrams in Eq. (5.3). The first two terms containing a dot are by construction very small, and will vanish when taking the quantum mechanical average, as discussed in Sec. IV B. The only important second-order coherent interaction is therefore the following Feynman diagram for normal-ordered fields:



(5.12)

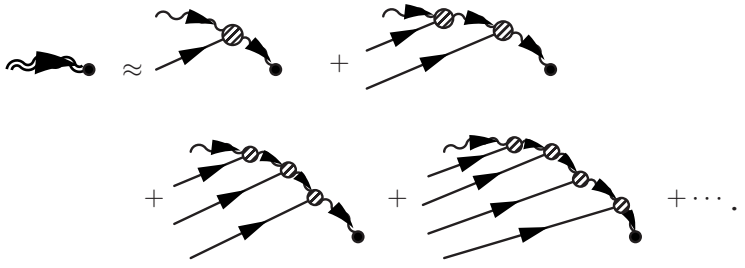
Suppressing the spatial dependence of the displaced electric field, this normal-ordered coherent interaction is given in vector representation by

$$-\frac{\beta^2 c_1^2}{\hbar^2\epsilon_0^2}\int_{t_0}^{t} dt' \{\tilde{\mathcal{D}}_0^{(-)}(t')[\tilde{\mathcal{D}}_0^{(-)}(t)\cdot\tilde{\mathcal{D}}_0^{(+)}(t')][\bar{\mathbf{J}}_0\cdot\tilde{\mathcal{D}}_0^{(+)}(t)] - [\tilde{\mathcal{D}}_0^{(-)}(t)\cdot\tilde{\mathcal{D}}_0^{(-)}(t')][\bar{\mathbf{J}}_0\cdot\tilde{\mathcal{D}}_0^{(+)}(t)]\tilde{\mathcal{D}}_0^{(+)}(t') + \text{H.c.}\}. \tag{5.13}$$

In the case of linearly polarized light, say $\tilde{\mathcal{D}}_0^{(-)} \| \mathbf{e}_x$ this term vanishes, but this is in general not the case. In Sec. VI we examine the term in some simplified system.

B. Evolution of the light

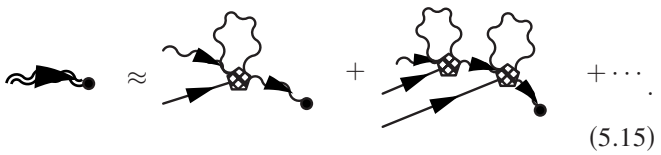
The treatment of the displaced electric field is similar to the spin, but there are a few important differences. Let us consider the negative-frequency part of the field, and write the expansion of the displaced electric field ignoring for now the evolution of the spin



(5.14)

When we take spatial average of diagrams such as these, we introduce δ-function correlations between vertex points. So far we have treated the atoms in the ideal gas approximation, where we ignore any correlation in the position of the atoms but in reality we should include a short-range correlation function describing that two different atoms cannot be at the same position. In Appendix D we show that including this leads to the Lorentz-Lorenz or Clausius-Mossotti relation. In the following we will only discuss loops, where two consecutive vertex points are evaluated for the same atom. Since we have subtracted the quantum mechanical average from the vertex, no first-order vertex will give a contribution to the

evolution of the light, and therefore these second-order loop diagrams are the most important effects apart from the diffraction effects included in the mode functions $\{\mathbf{f}_\mathbf{q}\}$. Later in Sec. V D we shall discuss the operator nature of the light field and then we keep the first-order vertex in the calculations. In the current approximation, Eq. (5.14) reduces to



(5.15)

We have here introduced an interaction denoted by a hatched pentagon which scales with $\beta^2\rho k_L^3$, and describes two ⬟ connected by the infinitely short propagator. Using the results for the infinitely short propagator, and taking quantum mechanical average this interaction reads in matrix form as follows:

$$= i\beta^2\rho(\mathbf{r}) \begin{bmatrix} \Gamma_{||}(\mathbf{r}) & 0 & 0 \\ 0 & \Gamma_{\perp,1}(\mathbf{r}) & i\Gamma_\Gamma(\mathbf{r}) \\ 0 & -i\Gamma_\Gamma(\mathbf{r}) & \Gamma_{\perp,2}(\mathbf{r}) \end{bmatrix} \equiv i\bar{\bar{\mathcal{M}}}'^t(\mathbf{r}), \tag{5.16}$$

where the coefficients entering the matrix are given by

$$\Gamma_\parallel(\mathbf{r}) = c_0^2\mathbf{J}^4\varrho_\parallel + c_1^2\varrho_\perp(J_z^2 + J_y^2), \tag{5.17a}$$

$$\Gamma_{\perp,1}(\mathbf{r}) = c_0^2\mathbf{J}^4\varrho_\perp + 2c_0c_1\varrho_\Gamma\mathbf{J}^2J_x + c_1^2(\varrho_\parallel J_z^2 + \varrho_\perp J_x^2), \tag{5.17b}$$

$$\Gamma_{\perp,2}(\mathbf{r}) = c_0^2\mathbf{J}^4\varrho_\perp + 2c_0c_1\varrho_\Gamma\mathbf{J}^2J_x + c_1^2(\varrho_\parallel J_y^2 + \varrho_\perp J_x^2), \tag{5.17c}$$

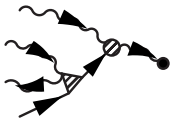
$$\Gamma_\Gamma(\mathbf{r}) = \varrho_\perp 2c_1c_0\mathbf{J}^2J_x - \varrho_\parallel\frac{c_1^2}{2}J_x + \varrho_\Gamma(c_0^2\mathbf{J}^2 + c_1^2J_x^2). \tag{5.17d}$$

We have here suppressed the spatial dependence to shorten notation. The series in Eq. (5.15) can be included in the differential equation describing the displaced electric field,

$$\left(2i\omega_L\frac{d}{dt} - \omega_L^2 + c^2\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times[\bar{\bar{\mathcal{M}}}^t(\mathbf{r}) + i\bar{\bar{\mathcal{M}}}'^t(\mathbf{r})]\right)\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t)$$
$$= c^2\int d^3r\,\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\bar{\bar{\delta}}^T(\mathbf{r},\mathbf{r}')\cdot\bar{\bar{m}}[\hat{\mathbf{J}}]^t_{\mathrm{mod}}\tilde{\mathbf{D}}^{(-)}(\mathbf{r}',t), \tag{5.18}$$

where the perturbation is modified accordingly. Because of the anti-Hermitian matrix, we see that these types of loop diagrams correspond to a decay of the field, i.e., the differential operator on the left-hand side describes the propagation through a lossy medium. On the basis of this analysis and the analysis in Sec. V A we thus link the loops in the Feynman diagrams with the decay associated with spontaneous emission.

It remains to discuss the effect of light interacting with an atom that was previously subject to an interaction such that the atomic spin state has been changed. In terms of Feynman diagrams this is described as



(5.19)

We shall postpone the analysis of this term and discuss it in connection with relating the fields to photon counting operators below.

C. Photon counting and Stokes operators

So far we have mainly been concerned with calculating the field $\tilde{\mathbf{D}}(\mathbf{r},t)$. For experiments which eventually involve counting photons we are more interested in quantities such as photon flux, and in particular the flux in some particular polarization state. We shall now discuss how to describe such photon counting experiments within our theory.

The general idea in this section is that we shall assume that we are able to measure the light flux in a certain spatial mode by projecting the light field onto the mode and then integrating the flux of the light field at some detector plane, that we assume to be far away from the atomic ensemble. We will formulate such a measuring process in terms of an inner product,

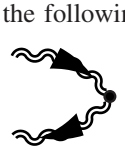
$$\langle\langle\boldsymbol{\varphi}(\mathbf{r},t)|\boldsymbol{\psi}(\mathbf{r},t)\rangle\rangle \equiv \int_{-\infty}^{\infty}dt\int_{\mathbb{R}^2}d^2r_\perp\boldsymbol{\varphi}^\dagger(\mathbf{r},t)\cdot\boldsymbol{\psi}(\mathbf{r},t). \tag{5.20}$$

We assume that the fields in general have some axis of propagation say $\mathbf{r}_\parallel$. The spatial integral is then performed in some plane perpendicular to this axis at some point $r_\parallel$ on this axis. This measuring process could be realized by, e.g., sending the light field through a single mode optical fiber prior to detection.

We are interested in the polarization of the field which is conveniently described by the so-called Stokes operators defined below. These operators can be derived from a Stokes generator defined in a bra-ket notation by

$$\bar{\bar{S}} \equiv |\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t)\rangle\rangle\langle\langle\tilde{\mathbf{D}}^{(-)}(\mathbf{r},t)|, \tag{5.21}$$

which we represent as the following diagram:



(5.22)

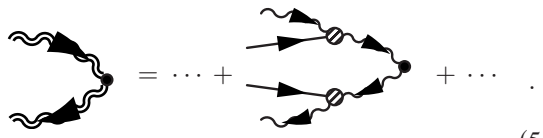
Measuring certain light modes according to the inner product in Eq. (5.20), correspond to picking out a certain matrix element of the Stokes generator. As an example we assume that in some experiment we are able to measure the photon flux of some linear polarization in some mode say $\tilde{\mathbf{f}}_{\mathbf{q},x}(\mathbf{r},t)$ after the interaction with the atoms. The time dependence is here $\tilde{\mathbf{f}}_{\mathbf{q},x}(\mathbf{r},t) = \mathbf{f}_{\mathbf{q},x}(\mathbf{r})e^{-i(\omega_{\mathbf{q},x}-\omega_L)t}$. The integrated photon flux

measured at the detector plane, is then given by

$$\frac{2c^2}{\hbar\epsilon_0\omega_L}\langle\langle\tilde{\mathbf{f}}^*_{\mathbf{q},x}|\ \bar{\bar{\mathcal{S}}}\ |\tilde{\mathbf{f}}^*_{\mathbf{q},x}\rangle\rangle, \tag{5.23}$$

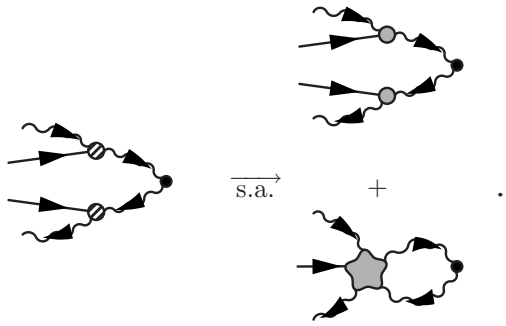
where we normalize the outcome to count the number of photons. We have here taken a spatial average of the Stokes generator as indicated by the calligraphic font.

Expanding this operator to second order, gives an additional term not covered by the analysis above. This extra term describes a process where both the negative frequency part and the positive frequency part of the displaced electric field interacts with the same atom. This extra term comes from the following contribution to the Stokes generator:



(5.24)

When taking the spatial average of this term we again generate a term representing that the interaction happens at the same point. This particular term would not have been there if we only considered the spatial average of the displaced electric field. The generated term we will illustrate as



(5.25)

We constructed the interaction represented in the Feynman diagram as a gray circle, such that when taking the quantum mechanical average the term vanishes. The new term generated when taking the spatial average, given as the lower right-hand diagram of Eq. (5.25), describes the square of the fluctuations which is not vanishing. This was also the case for the terms containing the infinitely short propagator. The new term however differs from the second-order terms containing the infinitely short propagators because here we need to use the full macroscopic propagator. To calculate the effect of this term in detail, we therefore need to have an expression for the spatial modes describing the system. We will consider this term for a simplified system in Sec. V D.

To describe the experiments in Ref. [10] it is convenient to define a set of polarization-dependent photon counting operators denoted as Stokes operators. These are defined in accordance with Eq. (5.23) as

$$\hat{s}_1^{\mathbf{q},\mathbf{q}'} = \frac{K}{2}(\langle\langle\tilde{\mathbf{f}}^*_{\mathbf{q}}|\ \bar{\bar{\mathcal{S}}}\ |\tilde{\mathbf{f}}^*_{\mathbf{q}}\rangle\rangle - \langle\langle\tilde{\mathbf{f}}^*_{\mathbf{q}'}|\ \bar{\bar{\mathcal{S}}}\ |\tilde{\mathbf{f}}^*_{\mathbf{q}'}\rangle\rangle), \tag{5.26a}$$

$$\hat{s}_2^{\mathbf{q},\mathbf{q}'} = \frac{K}{2}(\langle\langle\tilde{\mathbf{f}}^*_{\mathbf{q}}|\ \bar{\bar{\mathcal{S}}}\ |\tilde{\mathbf{f}}^*_{\mathbf{q}'}\rangle\rangle + \langle\langle\tilde{\mathbf{f}}^*_{\mathbf{q}'}|\ \bar{\bar{\mathcal{S}}}\ |\tilde{\mathbf{f}}^*_{\mathbf{q}}\rangle\rangle), \tag{5.26b}$$

$$\hat{s}_3^{\mathbf{q},\mathbf{q}'} = \frac{K}{2i}(\langle\langle\tilde{\mathbf{f}}^*_{\mathbf{q}}|\ \bar{\bar{\mathcal{S}}}\ |\tilde{\mathbf{f}}^*_{\mathbf{q}'}\rangle\rangle - \langle\langle\tilde{\mathbf{f}}^*_{\mathbf{q}'}|\ \bar{\bar{\mathcal{S}}}\ |\tilde{\mathbf{f}}^*_{\mathbf{q}}\rangle\rangle), \tag{5.26c}$$

where $K=\frac{2c^2}{\hbar\epsilon_0\omega_L}$. Using commutation relations for the creation and annihilation operators these Stokes operators are seen to have the commutation relations for angular momentum operators,

$$[\hat{s}_n^{\mathbf{q},\mathbf{q}'};\hat{s}_m^{\mathbf{q},\mathbf{q}'}] = i\epsilon_{nml}\hat{s}_l^{\mathbf{q},\mathbf{q}'}. \tag{5.27}$$

We will calculate and discuss these Stokes operators to second order in the coupling coefficient β in the following.

D. Calculation of Stokes operators

In this section we shall calculate the Stokes operators to second order. In the experiments in Refs. [8–10] the Stokes operators are measured by sending the light onto polarizing beam splitters followed by a measurement of the difference in the intensity of the two outputs. For instance, if we take the indices $\mathbf{q}$ and $\mathbf{q}'$ to refer to the x and y polarizations of the light, the operator $\hat{s}_1^{x,y}$ in Eq. (5.26) can be measured by measuring the difference in the intensity of the x and y polarizations. The remaining operators $\hat{s}_2^{x,y}$ and $\hat{s}_3^{x,y}$ can, respectively, be related to the difference intensity with the polarizing beam splitter rotated by 45° and the difference intensity between the two circular polarizations. For a general light beam, however, diffraction will cause the polarization of the light to depend on the spatial position and there is no well-defined polarization. The simple measurement scheme is thus only applicable in the paraxial approximation, where we can separate out a position-independent polarization vector. Far away from the ensemble we will therefore assume a paraxial approximation. That is, the mode functions $\tilde{\mathbf{f}}_{\mathbf{q}}(\mathbf{r},t)$ and $\tilde{\mathbf{f}}_{\mathbf{q}'}(\mathbf{r},t)$ describing the Stokes operators far away from the atomic ensemble resemble plane waves with transverse profiles that change slowly compared to the wavelength. The detector plane is placed far away from the atomic ensemble, and at this plane we will assume that the general set of basis functions $\{\mathbf{f}_{\mathbf{q}}\}$ can be approximated as

$$\mathbf{f}_q(\mathbf{r}) = \frac{1}{\sqrt{2\pi}}U_n(\mathbf{r}_\perp)\mathbf{e}_j e^{ikz}. \tag{5.28}$$

We have here set the direction of propagation to be along the z axis. The index $\mathbf{q}$ is now given as the set $\mathbf{q}=(k,n,j)$, where k is some wave number, n is an index referring to the transverse shape of the mode described by the scalar field $U_n(\mathbf{r})$, and j describes the polarization of this mode, that can be either x or y polarized. The completeness relation equation (2.29), and orthonormality condition in this approximation thus gives

$$\sum_n U_n^*(\mathbf{r}_\perp)U_n(\mathbf{r}'_\perp) = \delta(\mathbf{r}_\perp - \mathbf{r}'_\perp), \tag{5.29a}$$

$$\int d^2r_\perp U_n^*(\mathbf{r}_\perp)U_{n'}(\mathbf{r}_\perp) = \delta_{nn'}, \tag{5.29b}$$

and the dispersion relation equation (2.17a) at the detector plane is $\omega_{\mathbf{q}}^2 = c^2k^2$.

The paraxial approximation above is convenient for expressing the measured observable in terms of the polarization of the field, but may not be sufficient to accurately describe experiments, where tightly focused beams are used. We shall therefore only assume this approximation to be applicable far away from the sample, and not necessarily inside the ensemble. Physically this could correspond to a situation, where an initially paraxial beam is focused onto the ensemble with a lens and converted back into a paraxial beam after the interaction by another lens, as shown in Fig. 2. A similar treatment was used in Ref. [31].

Inside the ensemble we make the much weaker approximation that the set of spatial mode functions $U_{nq}(\mathbf{r})$ is independent of the polarization of the field, so that the set $\mathbf{f}_{\mathbf{q}}(\mathbf{r})$ is given by

$$\mathbf{f}_{\mathbf{q}}(\mathbf{r}) = \frac{1}{\sqrt{2\pi}} U_{nq}(\mathbf{r})\mathbf{e}_j(\mathbf{r}). \tag{5.30}$$

The mode $U_{nq}(\mathbf{r})$ now takes into account that the spatial shape of the beam may change through the ensemble, and likewise the polarization vector $\mathbf{e}_j(\mathbf{r})$, which we shall assume to be real valued. The index j will still be either x or y, corresponding to the polarization of the mode far away from the sample, but the vector $\mathbf{e}_j(\mathbf{r})$ will not necessarily be parallel to the x or the y axis. A more general description of the mode functions would include a dependence of the polarization vector $\mathbf{e}_j$ on the polarization state $U_{mq}(\mathbf{r})$, i.e., $\mathbf{e}_{mj}(\mathbf{r})$. The correction this generalization gives to the Stokes operators, is presented in Appendix H, in relation to Sec. VI C. When we make the relevant calculations to describe the Stokes operators defined in Eq. (5.26), we will choose to consider modes corresponding to the index $\mathbf{q}=(k,m,x)$ and $\mathbf{q}'=(k,m',y)$. We note that the set $\{\mathbf{f}_{\mathbf{q}}\}$ defined in this way is in general not complete, since, e.g., the assumption that the polarization vector is independent of the transverse mode number applies in the paraxial approximation but does not apply in general. When calculating the effect on the forward scattered field to first order we only get contributions from the near paraxial modes in the forward direction. When we go to second order there will, however, be effects of all the transverse modes, and in this case a correct treatment requires a more accurate treatment of the complete set of modes. Above we have already employed such a more general set of modes, when we discussed the effect of spontaneous emission, which involve all the transverse modes. In addition to this, a more accurate set of modes is also required for describing the effect of dipole-dipole interactions, which also involves all of the transverse modes.

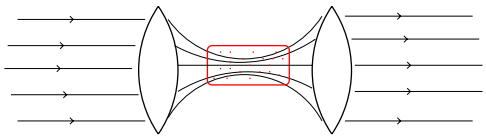
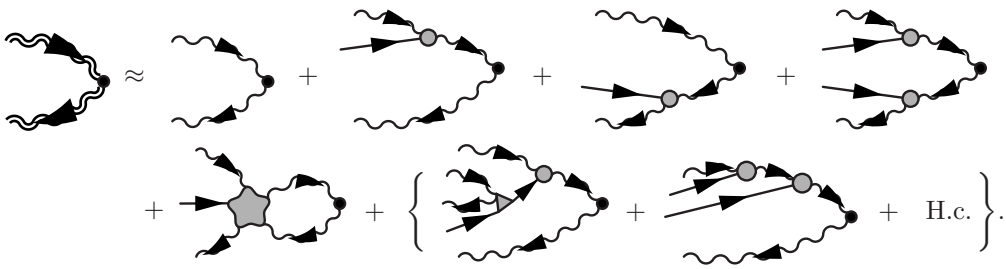


FIG. 2. (Color online) Schematic setup. We assume that away from the ensemble, the light-mode resembles a plane wave with some transverse profile. A set of lenses focus the beam down into the ensemble.

We will in the following calculate the Stokes operators in the limit described above. Diagrams containing a loop, we will not discuss, since these only lead to a decay of the light which we have discussed earlier. After taking spatial average, the diagrams in consideration are



(5.31)

Let us begin our discussion of this perturbation series by considering the first term on the right-hand side of Eq. (5.31). This term is the zeroth-order term of the Stokes generator $\bar{\bar{\mathcal{S}}}^{(0)}$. In the far-field limit $z \to \infty$ the matrix element we need to calculate is

$$\langle\langle \tilde{\mathbf{f}}^{*}_{kmj}(\mathbf{r},t)|\tilde{\mathcal{D}}^{(-)}_{0}(\mathbf{r},t)\rangle\rangle$$
$$= \int\int_{-\infty}^{\infty} dtd^2r_\perp \frac{1}{\sqrt{2\pi}} U_m(\mathbf{r}_\perp) e_j e^{ikz-i(\omega_k-\omega_L)t}$$
$$\times \sum_{qnl} \sqrt{\frac{\hbar\epsilon_0\omega_L}{4\pi}} U^{*}_n(\mathbf{r}_\perp) e_l e^{-iqz-i(\omega_q-\omega_L)t} \hat{a}^{\dagger}_{qnl}$$
$$= \sqrt{\frac{\hbar\epsilon_0\omega_L}{2c^2}} \hat{a}^{\dagger}_{kmj}, \tag{5.32}$$

and $\bar{\bar{\mathcal{S}}}^{(0)}$ thus gives us

$$K\langle\langle \tilde{\mathbf{f}}^{*}_{kmj}(\mathbf{r},t)|\ \bar{\bar{\mathcal{S}}}^{(0)}\ |\tilde{\mathbf{f}}^{*}_{km'j'}(\mathbf{r},t)\rangle\rangle = \hat{a}^{\dagger}_{kmj}\hat{a}_{km'j'}. \tag{5.33}$$

The zeroth-order Stokes operator $\hat{s}_1^{\mathbf{qq}'}$ for $\mathbf{q}=(k,m,x)$ and $\mathbf{q}'=(k,m',y)$ gives

$$\hat{s}_1^{\mathbf{qq}'} \equiv \hat{s}_1^{mm'} = \frac{1}{2}(\hat{a}^{\dagger}_{kmx}\hat{a}_{kmx} - \hat{a}^{\dagger}_{km'y}\hat{a}_{km'y}). \tag{5.34a}$$

The two remaining zeroth-order Stokes operators are found accordingly,

$$\hat{s}_2^{mm'} = \frac{1}{2}(\hat{a}^{\dagger}_{kmx}\hat{a}_{km'y} + \hat{a}^{\dagger}_{km'y}\hat{a}_{kmx}), \tag{5.34b}$$

$$\hat{s}_3^{mm'} = \frac{1}{2i}(\hat{a}^{\dagger}_{kmx}\hat{a}_{km'y} - \hat{a}^{\dagger}_{km'y}\hat{a}_{kmx}). \tag{5.34c}$$

In the following we will calculate the first-order components of the Stokes operators. We assume the quantum mechanical average of the atomic spin $\mathbf{J}$ to be parallel to the x axis. The relevant interaction matrix can in this case be written

$$\bar{\bar{m}}[\hat{\mathbf{J}}] = ic_1\beta \begin{bmatrix} 0 & \hat{J}_z(\mathbf{r}) & -\hat{J}_y(\mathbf{r}) \\ -\hat{J}_z(\mathbf{r}) & 0 & 0 \\ \hat{J}_y(\mathbf{r}) & 0 & 0 \end{bmatrix}, \tag{5.35}$$

and after spatial averaging we simply write

$$\langle \bar{\bar{m}}[\hat{\mathbf{J}}]\rangle_{\text{s.a.}} \equiv \bar{\bar{\mathcal{M}}}[\bar{\mathbf{J}}] = -ic_1\beta\rho(\mathbf{r}) \begin{pmatrix} 0 \\ \bar{J}_y(\mathbf{r}) \\ \bar{J}_z(\mathbf{r}) \end{pmatrix} \times \quad . \tag{5.36}$$

The second and the third term on the right-hand side of Eq. (5.31) are the first-order terms of the Stokes generator, $\bar{\bar{\mathcal{S}}}^{(1)}$. To calculate the contribution to the Stokes operators from these terms we must evaluate the expression

$$\langle\langle \tilde{\mathbf{f}}^{*}_{kmj}(\mathbf{r},t)|c^2 \int\int_{t_0}^{t} dtd^3r' \bar{\bar{P}}^{(-)}(\mathbf{r},t|\mathbf{r}',t') \cdot\ \bar{\bar{\mathcal{M}}}^{t}[\bar{\mathbf{J}}]\tilde{\mathcal{D}}^{(-)}_{0}(\mathbf{r}',t')\rangle\rangle. \tag{5.37}$$

The initial time t_0 we will set to $-\infty$, and because we assume our detector plane to be infinitely far away from the atomic ensemble, we can take $t\to\infty$. Using the expression for the set $\{\mathbf{f_q}\}$ given by Eq. (5.28) for the detector plane and Eq. (5.30) inside the ensemble, Eq. (5.37) reduces to

$$\left(-i\sqrt{\frac{\hbar\omega_L\epsilon_0}{2c^2}}\right)\frac{k_Lc_1\beta}{2}\int d^3r' \sum_{nl} \rho(\mathbf{r}')\Theta^{mn}_{jl}(\mathbf{r}')^{*}\hat{a}^{\dagger}_{knl}, \tag{5.38}$$

where

$$\Theta^{mn}_{jl}(\mathbf{r}') \equiv U_{km}(\mathbf{r}')^{*}U_{kn}(\mathbf{r}')\mathbf{e}_j(\mathbf{r}) \cdot \left[\begin{pmatrix} 0 \\ \bar{J}_y(\mathbf{r}') \\ \bar{J}_z(\mathbf{r}') \end{pmatrix} \times \mathbf{e}_l(\mathbf{r}')\right]$$
$$= \Psi^{mn}_{k}(\mathbf{r}')[\delta_{lx}\delta_{jy} - \delta_{jx}\delta_{ly}]\left[\begin{pmatrix} 0 \\ \bar{J}_y(\mathbf{r}') \\ \bar{J}_z(\mathbf{r}') \end{pmatrix} \cdot \mathbf{e}_z(\mathbf{r}')\right], \tag{5.39}$$

with

$$\Psi^{mn}_{k}(\mathbf{r}') = U_{km}(\mathbf{r}')^{*}U_{kn}(\mathbf{r}'). \tag{5.40}$$

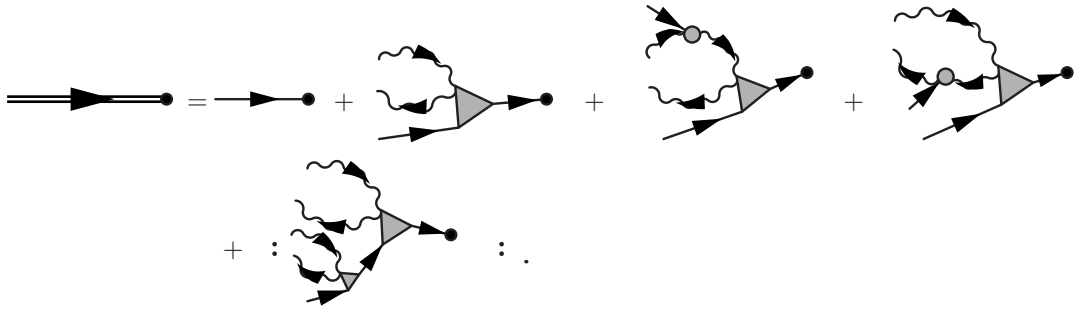
In the final equality we have introduced the local basis vector $\mathbf{e}_z(\mathbf{r})=\mathbf{e}_x(\mathbf{r})\times\mathbf{e}_y(\mathbf{r})$. The effect of the first-order term of the Stokes generator $\bar{\bar{\mathcal{S}}}^{(1)}$ to the Stokes operators thus reads

$$K\langle\langle \tilde{\mathbf{f}}^{*}_{kmj}(\mathbf{r},t)|\ \bar{\bar{\mathcal{S}}}^{(1)}\ |\tilde{\mathbf{f}}^{*}_{km'j'}(\mathbf{r},t)\rangle\rangle$$
$$= k_Lc_1\beta\int d^3r' \sum_{nl} \rho(\mathbf{r}')\frac{1}{2}[\Theta^{mn}_{jl}(\mathbf{r}')^{*}\hat{a}^{\dagger}_{knl}\hat{a}_{km'j'}$$
$$+ \Theta^{m'n}_{j'l}(\mathbf{r}')\hat{a}^{\dagger}_{kmj}\hat{a}_{knl}]. \tag{5.41}$$

The remaining terms on the right-hand side of Eq. (5.31), that is the second-order terms, can be calculated in a similar way. The results may be found in Appendix E. The calculations given in Eqs. (5.41), (E1), (E2), and (E5) is the starting point for a discussion of the dynamics of the system subject to a general light field of many modes.

The description that we have used here, where we define the Stokes operators in terms of expectation value between different orthogonal modes, is very convenient for a theoretical description of the process. It does, however, not directly correspond to the experimentally measured observables unless one, e.g., separates out particular modes with single mode optical fibers. We shall therefore defer the discussion of the consequences of these results to the next section, where we use these results to calculate the evolution of observables more relevant to experiments.

We will now give the equation for the atomic spin. The incoherent terms describing decay due to spontaneous emission have already been discussed. Here we will consider the coherent interaction up to second order in the perturbation series. Below we show the diagrammatic representation of the coherent perturbation series for the atomic spin up to second order,



(5.42)

We will denote the first-order term in the expansion, Eq. (5.42), as $\mathcal{J}^{(1)}$. Employing again the approximations done in the previous calculations, that is, using the set of light modes $\{\mathbf{f_q}\}$ given in Eq. (5.30) and setting the initial time to $-\infty$ and the final time to ∞, the term can be written as

$$\begin{aligned}\mathcal{J}^{(1)} &= -\beta c_1 k_L \sum_{kmm'} \Psi_k^{mm'}(\mathbf{r})[\bar{\mathbf{J}}(\mathbf{r}) \times \mathbf{e}_z(\mathbf{r})]\frac{1}{2i} \\ &\times (\hat{a}_{kmx}^\dagger \hat{a}_{km'y} - \hat{a}_{kmy}^\dagger \hat{a}_{km'x}) \\ &= -\beta c_1 k_L \sum_{kmm'} [\bar{\mathbf{J}}(\mathbf{r}) \times \mathbf{e}_z(\mathbf{r})] \\ &\times \{\mathrm{Re}[\Psi_k^{mm'}(\mathbf{r})]\hat{s}_3^{mm'} + \mathrm{Im}[\Psi_k^{mm'}(\mathbf{r})]\hat{s}_2^{mm'}\}. \end{aligned} \tag{5.43}$$

We notice that compared to the simple theory in Ref. [29] there is an additional term proportional to the imaginary part of the function $\Psi^{mm'}(\mathbf{r})$. A similar correction can also be found for the Stokes operators for the light. Also notice that the dynamics of the spin to first order happens in a plane orthogonal to the vector $\mathbf{e}_z(\mathbf{r})$. This is the reason why the term in Eq. (E3) vanishes, since there we are considering the effect of the dynamics of the atomic spin on an axis parallel to the $\mathbf{e}_z(\mathbf{r})$ vector. The calculation of the second-order terms is presented in Appendix F. In the following section we will examine the effect of these calculations under conditions attainable in experiments.

VI. EXPERIMENTAL APPLICATION AND VALIDITY

In this section we shall consider different limits where we can reduce our general theory to a theory resembling the simple description obtained in one-dimensional theories [7,29]. Furthermore, we discuss the validity of the approximations made to arrive at these simple limits as well as the validity of our perturbative treatment of the interaction.

A. Measurement procedure

In the preceding section we discussed how our theory could be used to calculate Stokes operators corresponding to specific transverse modes of the field. While such a treatment is appealing from a theoretical perspective, it is less desirable experimentally, since the isolation of single transverse modes is complicated (although it could be done by passing the light through single mode optical fibers). Here we shall therefore express our result in terms of a simpler experimental procedure. Suppose that the detection is performed by sending the light onto a polarizing beam splitter and recording the intensity of the two output port with two cameras. The difference between the intensities can now be used to define position-dependent Stokes operators $\hat{s}_i(\mathbf{r}_\perp)$, i.e., $\hat{s}_1(\mathbf{r}_\perp)$ corresponds to the difference in intensity between x and y polarization at position $\mathbf{r}_\perp$ in the detector plane. Similarly $\hat{s}_2(\mathbf{r}_\perp)$ and $\hat{s}_3(\mathbf{r}_\perp)$ can, respectively, be related to the difference intensity with the polarizer rotated by 45° and the difference intensity between the two circular polarizations. These operators may in general be determined by

$$\begin{aligned}\hat{s}_1(\mathbf{r}_\perp) = \sum_{kmm'} \frac{1}{2}[&U_m^*(\mathbf{r}_\perp)\hat{a}_{kmx}^\dagger \hat{a}_{km'x} U_{m'}(\mathbf{r}_\perp) \\ &- U_m^*(\mathbf{r}_\perp)\hat{a}_{kmy}^\dagger \hat{a}_{km'y} U_{m'}(\mathbf{r}_\perp)], \end{aligned} \tag{6.1a}$$

$$\begin{aligned}\hat{s}_2(\mathbf{r}_\perp) = \sum_{kmm'} \frac{1}{2}[&U_m^*(\mathbf{r}_\perp)\hat{a}_{kmx}^\dagger \hat{a}_{km'y} U_{m'}(\mathbf{r}_\perp) \\ &+ U_m^*(\mathbf{r}_\perp)\hat{a}_{kmy}^\dagger \hat{a}_{km'x} U_{m'}(\mathbf{r}_\perp)], \end{aligned} \tag{6.1b}$$

$$\begin{aligned}\hat{s}_3(\mathbf{r}_\perp) = \sum_{kmm'} \frac{1}{2i}[&U_m^*(\mathbf{r}_\perp)\hat{a}_{kmx}^\dagger \hat{a}_{km'y} U_{m'}(\mathbf{r}_\perp) \\ &- U_m^*(\mathbf{r}_\perp)\hat{a}_{kmy}^\dagger \hat{a}_{km'x} U_{m'}(\mathbf{r}_\perp)]. \end{aligned} \tag{6.1c}$$

Below we shall derive expressions for the operators (6.1) and discuss how to implement a light-matter quantum interface based on these operators. In Sec. VI B we for simplicity first consider an extreme paraxial limit, where we assume that essentially no diffraction occurs during the propagation. In this limit the dynamics becomes extremely simple. In Sec. VI C we consider a more interesting limit, where we may have multiple modes which may experience diffraction. Here we show that measurement of the operators $\hat{s}_i(\mathbf{r}_\perp)$ still allows us to simplify the dynamics of the system. In a suitable limit we find a simple two-mode transformation between transverse modes of the light field and single modes of the atomic ensembles.

B. Extreme paraxial approximation

In the extreme paraxial approximation, we completely ignore any dynamics transverse to the propagation direction of the light modes and approximate the set of modes $\{\mathbf{f_q}\}$ with Eq. (5.28) throughout the ensemble. Since the typical distance for diffraction is given by $l_d \sim A/\lambda$, the condition for the validity of this approximation is $L \ll l_d$, or expressed in terms of the Fresnel number $\mathcal{F} \gg 1$.

The full expressions for the Stokes operators are quite involved, and we therefore leave the incoherent part of the evolution to Appendix G. Keeping only the coherent part of the interaction, we find the Stokes operators to second order in the interaction to be

$$\hat{s}_{1,\mathrm{out}}(\mathbf{r}_\perp) = \hat{s}_{1,\mathrm{in}}(\mathbf{r}_\perp) - k_L c_1 \beta \int dz' \rho(z',\mathbf{r}_\perp) \bar{J}_z(z',\mathbf{r}_\perp) \hat{s}_{2,\mathrm{in}}(\mathbf{r}_\perp) - \frac{1}{2}(k_L \beta c_1)^2 \iint dz'\,dz'' \rho(z',\mathbf{r}_\perp)\rho(z'',\mathbf{r}_\perp) \times \bar{J}_z(z',\mathbf{r}_\perp)\bar{J}_z(z'',\mathbf{r}_\perp)\hat{s}_{1,\mathrm{in}}(\mathbf{r}_\perp), \tag{6.2a}$$

$$\hat{s}_{2,\mathrm{out}}(\mathbf{r}_\perp) = \hat{s}_{2,\mathrm{in}}(\mathbf{r}_\perp) + k_L c_1 \beta \int dz' \rho(z',\mathbf{r}_\perp) \bar{J}_z(z',\mathbf{r}_\perp) \hat{s}_{1,\mathrm{in}}(\mathbf{r}_\perp) - \frac{1}{2}(k_L \beta c_1)^2 \iint dz'\,dz'' \rho(z',\mathbf{r}_\perp)\rho(z'',\mathbf{r}_\perp) \times \bar{J}_z(z',\mathbf{r}_\perp)\bar{J}_z(z'',\mathbf{r}_\perp)\hat{s}_{2,\mathrm{in}}(\mathbf{r}_\perp), \tag{6.2b}$$

$$\hat{s}_{3,\mathrm{out}}(\mathbf{r}_\perp) = \hat{s}_{3,\mathrm{in}}(\mathbf{r}_\perp). \tag{6.2c}$$

In this limit we see that the Stokes operator $\hat{s}_3$ is decoupled from the coherent dynamics of the system, and only evolves due to spontaneous emission [derived in Eq. (G2)].

Similarly we may find the coherent dynamics of the atomic spin. Leaving again the incoherent part to Appendix G, we find

$$\bar{J}_{x,\mathrm{out}}(\mathbf{r}) = \bar{J}_{x,\mathrm{in}}(\mathbf{r}) - \beta c_1 k_L \sum_k \bar{J}_{y,\mathrm{in}}(\mathbf{r}) \hat{s}^k_{3,\mathrm{in}}(\mathbf{r}_\perp) - \frac{1}{2}(\beta c_1 k_L)^2 \sum_{kk'} \bar{J}_{x,\mathrm{in}}(\mathbf{r}) \hat{s}^k_{3,\mathrm{in}}(\mathbf{r}_\perp) \hat{s}^{k'}_{3,\mathrm{in}}(\mathbf{r}_\perp), \tag{6.3a}$$

$$\bar{J}_{y,\mathrm{out}}(\mathbf{r}) = \bar{J}_{y,\mathrm{in}}(\mathbf{r}) + \beta c_1 k_L \sum_k \bar{J}_{x,\mathrm{in}}(\mathbf{r}) \hat{s}^k_{3,\mathrm{in}}(\mathbf{r}_\perp) - \frac{1}{2}(\beta c_1 k_L)^2 \sum_{kk'} \bar{J}_{y,\mathrm{in}}(\mathbf{r}) \hat{s}^k_{3,\mathrm{in}}(\mathbf{r}_\perp) \hat{s}^{k'}_{3,\mathrm{in}}(\mathbf{r}_\perp), \tag{6.3b}$$

$$\bar{J}_{z,\mathrm{out}}(\mathbf{r}) = \bar{J}_{z,\mathrm{in}}(\mathbf{r}). \tag{6.3c}$$

Analogous to what we found for $\hat{s}_3$, we see that the operator $\bar{J}_z$ is decoupled from the coherent dynamics of the system. This result can directly be associated to the conservation of angular momentum along the z axis. In the extreme paraxial approximation this is true to all orders in the coherent interaction.

The results in Eqs. (6.2) and (6.3) are essentially equivalent to the simplified one-dimensional description of the system given in Refs. [7,29]. The only difference is that the expressions derived here now apply for each value of $\mathbf{r}_\perp$, whereas the previous treatments assumed the system was transversely homogeneous and only considered the variables integrated over $\mathbf{r}_\perp$.

A further simplification of Eq. (6.3) can be obtained if we introduce the rotation vector

$$\mathbf{\Omega} = \beta c_1 k_L \sum_k \hat{s}^k_{3,\mathrm{in}}(\mathbf{r}_\perp)\mathbf{e}_z. \tag{6.4}$$

With this definition we find that Eq. (6.3) describes nothing but a rotation of the spin around the $\mathbf{e}_z$ axis,

$$\bar{\mathbf{J}}_{\mathrm{out}} = \bar{\mathbf{J}}_{\mathrm{in}} + \bar{\mathbf{J}}_{\mathrm{in}} \times \mathbf{\Omega} + \frac{1}{2}(\bar{\mathbf{J}}_{\mathrm{in}} \times \mathbf{\Omega}) \times \mathbf{\Omega}. \tag{6.5}$$

C. Multimode coupling

In the preceding section we basically ignored all of the dynamics transverse to the propagation direction. Now we turn to a more interesting situation, where we may describe effects associated with diffraction of the light beams. Our goal in this section is to find a set of conditions under which we can have a simple dynamics, where the individual transverse modes of the light field talks to a single mode of the atomic ensemble. Such an interaction would enable the storage of information from several light modes into spatial modes of the ensemble, e.g., using the protocol in [9]. The realization of this interaction would thus expand the information storage capacity of the atomic ensembles. A similar problem is considered in Ref. [32]. In related work such storage of multimode memory has recently been achieved in atomic ensembles using electromagnetically induced transparency [33].

To achieve simple results in the end, we will here consider a situation, where we have a strong classical beam polarized in the x direction in a single transverse mode $U_{ok}(\mathbf{r})$ (denoted by the index o). For the y polarization we, however, include a complete set of modes, which may or may not include a term identical to the mode of the x polarization. For the strong mode we will approximate $\hat{a}^\dagger_{kox} = \hat{a}_{kox} = \sqrt{N^o_x} \gg 1$, where N^o_x is the number of photons in this particular mode. Since the Stokes operators are dominated by the terms involving the classical component, the only important contributions in the Stokes operator (6.1) are the terms containing the strong classical mode. Equations (6.1) are thus approximated by

$$\hat{s}^{(\mathrm{in})}_1(\mathbf{r}_\perp) \approx \frac{1}{2}|U_{ok_L}(\mathbf{r}_\perp)|^2 N^o_x, \tag{6.6a}$$

$$\hat{s}^{(\mathrm{in})}_2(\mathbf{r}_\perp) \approx \frac{\sqrt{N^o_x}}{2} \sum_{km} \{\mathrm{Re}[U^*_{ok}(\mathbf{r}_\perp)U_{mk}(\mathbf{r}_\perp)]\hat{X}^m_P - \mathrm{Im}[U^*_{ok}(\mathbf{r}_\perp)U_{mk}(\mathbf{r}_\perp)]\hat{P}^m_P\}, \tag{6.6b}$$

$$\hat{s}_3^{(\mathrm{in})}(\mathbf{r}_\perp) \approx \frac{\sqrt{N_x^o}}{2}\sum_{km}\{\mathrm{Re}[U_{ok}^*(\mathbf{r}_\perp)U_{mk}(\mathbf{r}_\perp)]\hat{P}_P^m + \mathrm{Im}[U_{ok}^*(\mathbf{r}_\perp)U_{mk}(\mathbf{r}_\perp)]\hat{X}_P^m\}, \tag{6.6c}$$

where

$$\hat{X}_P^m = \frac{1}{\sqrt{2}}(\hat{a}_{kmy}^\dagger + \hat{a}_{kmy}), \tag{6.7a}$$

$$\hat{P}_P^m = \frac{1}{i\sqrt{2}}(\hat{a}_{kmy}^\dagger - \hat{a}_{kmy}). \tag{6.7b}$$

In order to obtain a simple result in the measurement process, let us assume that we can choose the mode functions $U_{mk}(\mathbf{r})$ to be real in the detection plane. This could, e.g., be achieved by sending the light through a lens which converts the incoming modes into extreme paraxial beams as shown in Fig. 2 (note that since we only make this assumption in the detection plane, this assumption does not restrict the shape inside the ensemble). Experimentally the operators $\hat{X}^m$ and $\hat{P}^m$ defined here can then be measured by simply integrating the measured $\hat{s}_i(\mathbf{r}_\perp)$ with a suitable weight function, e.g.,

$$\sqrt{\frac{2}{N_x^0}}\int d\mathbf{r}_\perp \frac{U_m(\mathbf{r}_\perp)}{U_o(\mathbf{r}_\perp)}\hat{s}_2(\mathbf{r}_\perp) = \hat{X}_P^m, \tag{6.8}$$

where we have used the expansion in (6.1) as well as the orthogonality relation of the transverse mode functions (5.29b).

In our equations of motion we for simplicity only keep terms to first order in β and $\sqrt{N_x^o}$, and neglect all other terms. The equations of motion for the Stokes operators give in this limit

$$\hat{X}_{\mathrm{out}}^m = \hat{X}_{\mathrm{in}}^m + k_L\beta c_1\sqrt{\frac{N_x^o}{2}}\int d^3r'\rho(\mathbf{r}')\begin{pmatrix}0\\ \bar{J}_y(\mathbf{r})\\ \bar{J}_z(\mathbf{r})\end{pmatrix}\cdot\mathbf{e}_z(\mathbf{r})\mathrm{Re}[\Psi_k^{mo}(\mathbf{r})], \tag{6.9a}$$

$$\hat{P}_{\mathrm{out}}^m = \hat{P}_{\mathrm{in}}^m + k_L\beta c_1\sqrt{\frac{N_x^o}{2}}\int d^3r'\rho(\mathbf{r}')\begin{pmatrix}0\\ \bar{J}_y(\mathbf{r})\\ \bar{J}_z(\mathbf{r})\end{pmatrix}\cdot\mathbf{e}_z(\mathbf{r})\mathrm{Im}[\Psi_k^{mo}(\mathbf{r})], \tag{6.9b}$$

where Ψ^{mo} is defined in terms of the mode functions U_m in Eq. (5.40). Employing the same set of approximations for the spin equation we find

$$\bar{\mathbf{J}}_{\mathrm{out}}(\mathbf{r}) \approx \bar{\mathbf{J}}_{\mathrm{in}}(\mathbf{r}) + k_L\beta c_1\sqrt{\frac{N_x^o}{2}}\sum_n\{\mathrm{Re}[\Psi_k^{no}(\mathbf{r})]\hat{P}_{\mathrm{in}}^n - \mathrm{Im}[\Psi_k^{mo}(\mathbf{r})]\hat{X}_{\mathrm{in}}^n\}[\bar{\mathbf{J}}_{\mathrm{in}}(\mathbf{r})\times\mathbf{e}_z(\mathbf{r})]. \tag{6.10}$$

Note, that the expressions we have derived here, allow for a general set of transverse modes which may experience diffraction, and thus go beyond the extreme paraxial approximation made in the preceding section. In the expressions above we do, however, still use the paraxial approximation in Eq. (5.30), where we ignore the dependence of the polarization vector on the mode number. In Appendix H we relax this approximation.

The expressions in Eq. (6.10) differ from the simple results of the preceding section because of the extra terms proportional to $\mathrm{Im}[\Psi_k^{mo}(\mathbf{r})]$. These terms complicate the dynamics and, e.g., means that one cannot use the protocol in Ref. [9] to store information in the ensemble. There are, however, certain limits where the extra terms in Eq. (6.10) disappear. One situation is when the mode we are considering in the y polarization is identical to the classical mode in the x polarization (except from the different orientation of the polarization). This situation corresponds to the experimental situation, where the weight factor U_m/U_o in Eq. (6.8) is unity, such that the final result is obtained by integrating the intensity over the transverse plane. This case therefore corresponds the experimental situation where the light is detected by photodetectors instead of cameras. In this case $\mathrm{Im}[\Psi_k^{mo}(\mathbf{r})]$ vanish identically and the evolution of the light operators again resemble the result of the preceding section, where, e.g., the $\hat{s}_2$ component was conserved, which translates into $\hat{P}_{\mathrm{out}}^m = \hat{P}_{\mathrm{in}}^m$. Note, however, that unlike the situation considered below, the atomic operators in this situation gets an admixture of several different input light modes, and will not in general reduce to the dynamics considered in Ref. [9].

Let us now consider a different limit ideally suited for a multimode memory. We assume that we are in the paraxial approximation, where we can ignore the spatial dependence of the polarization vectors. For simplicity we also assume that the classical mode $U_o(\mathbf{r})$ has a uniform intensity and that the density is constant over the region, where U_m is nonzero in the atomic ensemble. We furthermore assume that the macroscopic polarization is constant and along the x axis, $\bar{J}_x$, and finally we assume that Ψ^{mo} is real (for a discussion of the validity of this approximation we refer to the next section). In the spin equation (6.10) we will only keep terms proportional to the macroscopic spin component $\bar{J}_x$. In this situation the relevant equations reads as

$$\hat{X}_{P,\mathrm{out}}^m = \hat{X}_{P,\mathrm{in}}^m + k_L\beta c_1 U_o\sqrt{\frac{N_x^o}{2}}\rho\int d^3r'\bar{J}_z(\mathbf{r})U_m(\mathbf{r})e^{-ikz}, \tag{6.11a}$$

$$\hat{P}_{P,\mathrm{out}}^m = \hat{P}_{P,\mathrm{in}}^m, \tag{6.11b}$$

$$\bar{J}_{y,\mathrm{out}}(\mathbf{r}) = \bar{J}_{y,\mathrm{in}}(\mathbf{r}) + k_L \beta c_1 U_o \sqrt{\frac{N_x^o}{2}} \sum_n U_n(\mathbf{r}) e^{-ikz} \hat{P}_{\mathrm{in}}^n \bar{J}_{x,\mathrm{in}}, \tag{6.11c}$$

$$\bar{J}_{z,\mathrm{out}}(\mathbf{r}) = \bar{J}_{z,\mathrm{in}}(\mathbf{r}). \tag{6.11d}$$

Here the factor $\exp(-ikz)$ comes from the classical field and cancels the $\exp(ikz)$ dependence of the mode function U_m, since $U_m \exp(-ikz)$ should be real according to the assumption of Ψ being real. This set of equations can be symmetrized and simplified by introducing a set of collective operators

$$\tilde{X}_A^m = \sqrt{\frac{\rho}{J_x L}} \int d^3 r \bar{J}_y(\mathbf{r}) U_m(\mathbf{r}) e^{-ikz}, \tag{6.12a}$$

$$\tilde{P}_A^m = \sqrt{\frac{\rho}{J_x L}} \int d^3 r \bar{J}_z(\mathbf{r}) U_m(\mathbf{r}) e^{-ikz}, \tag{6.12b}$$

where L is the length of the ensemble. The coefficients here are chosen such that the operators $\hat{X}_A^m$ and $\hat{P}_A^m$ fulfill the standard commutation relation for position and momentum

$$[\tilde{X}_A^m, \tilde{P}_A^{m'}] = i \delta_{mm'}. \tag{6.13}$$

With these definitions, Eqs. (6.11) reduce to

$$\tilde{X}_{P,\mathrm{out}}^m = \tilde{X}_{P,\mathrm{in}}^m + \kappa \tilde{P}_{A,\mathrm{in}}^m, \tag{6.14a}$$

$$\tilde{P}_{P,\mathrm{out}}^m = \tilde{P}_{P,\mathrm{in}}^m, \tag{6.14b}$$

$$\tilde{X}_{A,\mathrm{out}}^m = \tilde{X}_{A,\mathrm{in}}^m + \kappa \tilde{P}_{P,\mathrm{in}}^m, \tag{6.14c}$$

$$\tilde{P}_{A,\mathrm{out}}^m = \tilde{P}_{A,\mathrm{in}}^m, \tag{6.14d}$$

where

$$\kappa = k_L \beta c_1 U_o \sqrt{\frac{N_x^o \rho \bar{J}_x L}{2}}. \tag{6.14e}$$

These equations describe a system where one transverse light-mode couples to a single mode of the atomic ensemble, which in turn couples back to the same light mode. This two-mode mode dynamics is exactly identical to the dynamics derived in Ref. [7] for a single transverse mode. The dynamics can thus, e.g., be used to realize a multimode version of the memory protocol implemented in Ref. [9]. In this protocol $\hat{P}_{P,\mathrm{in}}^m$ is stored in the atomic mode $\hat{X}_{a,\mathrm{out}}^m$, while at the same time the atomic mode $\hat{P}_{A,\mathrm{in}}^m$ is transferred to the light mode $\hat{X}_{P,\mathrm{out}}^m$, as described by Eq. (6.14). After detection of the light operator $\hat{X}_{P,\mathrm{out}}^m$ one can then realize a quantum memory by feeding back the measurement result to the atoms as it was shown in Ref. [9].

D. Validity

1. Validity of the simple multimode dynamics

In the preceding section we derived a simple multimode dynamics useful for making a multimode light-matter quantum interface. For experimental implementation of this idea an important question is the validity of the approximations leading to Eq. (6.14). First, we need that the imaginary part of $\Psi^{om}(\mathbf{r})$ in Eq. (6.9) should vanish. Furthermore, in order to define orthogonal spin-modes that do not couple different transverse modes, we need $|U_o(\mathbf{r})|$ to be uniform. Taking the classical mode to be given by $U_o(\mathbf{r}) = U_o e^{ikz}$, where U_o is real, we also need the quantum mode $U_m(\mathbf{r})$ to be real valued apart from the e^{ikz} dependence. Let us now take the modes $U_m(\mathbf{r})$ to be Hermite-Gaussian beams [34]. Such modes can be represented by

$$U_{mn}(\mathbf{r}) = \frac{B w_0}{w(z)} H_m\left(\sqrt{2}\frac{x}{w(z)}\right) H_n\left(\sqrt{2}\frac{y}{w(z)}\right) e^{i[kz-(m+n+1)\tanh z/z_0]} \times e^{ik(x^2+y^2)/2R(z)} e^{-(x^2+y^2)/w^2(z)}, \tag{6.15a}$$

where

$$w(z) = w_0 \sqrt{1 + z^2/z_0^2}, \tag{6.15b}$$

$$R(z) = z + \frac{z_0^2}{z}, \tag{6.15c}$$

$$z_0 = \frac{\pi w_0^2}{\lambda}. \tag{6.15d}$$

Here w_0 is the minimum waist of the beam, k is the wave number, λ is the wavelength, $B \in \mathbb{R}$ is a normalization coefficient, and H_n is the set of Hermite polynomials. The condition that $U_{mn}(\mathbf{r})$ must be real valued gives the conditions

$$\lambda R(z) \gg w^2(z), \quad \left|(1+m+n)\frac{z}{z_0}\right| \ll 1. \tag{6.16}$$

These are in fact equivalent conditions, and introducing the Fresnel number $\mathcal{F} \equiv w^2(z)/\lambda L$ we find the condition

$$\mathcal{F} \gg 1 + m + n. \tag{6.17}$$

2. Validity of perturbation theory

The theory we have developed in this paper is based on perturbation theory in the interaction between light and atoms. In this section we discuss the limits of validity of this perturbative treatment. We will be considering worst case scenarios to find the limit, where our perturbation series equations (5.31) and (5.42) converge. An important parameter for these estimates will be the effective coupling constant for the collective operators κ defined in Eq. (6.14). For applications to light-matter quantum interfaces this parameter should be of order unity. As we shall see below, this is still possible without violating the applicability of perturbation theory. Another important parameter is the optical depth, α, defined by $\alpha \sim \rho \lambda^2 L$. The optical depth plays an important

factor when describing the effect of the incoherent interaction, e.g., the spontaneous emission.

Throughout this work, we have assumed that the atomic ensemble is polarized along the x axis, so that the atomic spin components $\rho\bar{J}_y$, $\rho\bar{J}_z$ only carries quantum noise. Also we have assumed that the classical component of the light is linearly polarized so that, e.g., circular components are governed by quantum noise. These assumptions will be important for estimating the terms below.

We first consider the expansion of the light field (5.31), and in particular the coherent part of the interaction. The effective perturbation coefficient for the first-order term is found to scale at most as $(\beta k_L\sqrt{N_A})/A \sim \kappa/\sqrt{N_P}$ [may be found by estimating Eq. (5.41)]. Here A is the transverse area of the atomic ensemble, and N_P is the total number of photons in a pulse. Going to second order an important term is described in Eq. (E3). Since we are not including the time evolution of the macroscopic polarization in the average interaction, this term has a potential scaling as large as κ^2. We showed, however, that in the paraxial approximation, the term vanishes. Going beyond the paraxial approximation as done in Appendix H, we find that for linearly polarized light the scaling is $\kappa^2/\sqrt{N_P}$. The last contribution to Eq. (5.31) is the incoherent interaction considered in Appendix G. The scaling of this effect $\kappa^2\cdot(N_A/N_P)/\alpha$.

Now we consider the spin series (5.42) for a single atom. The incoherent part of the evolution of the spin is described in Eq. (5.9), and scales as κ^2/α, it can be ignored for sufficiently large α. The first-order term scales as $\kappa/\sqrt{N_A}$ for linearly polarized light. To increase this coefficient we need circularly polarized light, which makes it interesting to examine the second-order term describing the change of the polarization due to the interactions with atoms. This process is described in Eq. (F1), which represents the optically induced dipole-dipole interaction. This particular term vanishes when we take quantum mechanical averages, because we have subtracted the only nonvanishing component, but we can still calculate the root-mean-square contribution. The effect can then be separated into a short-range part and a long-range contribution. The long-range contribution can be estimated to give a contribution of order $\kappa^2\sqrt{d/(L\alpha)}$, where d is the smallest dimension of the setup, i.e., the smaller of the length and the transverse sizes of the beam and the ensemble. The short-range part actually diverges within our present approximations. If, however, we regularize the integral by excluding the volume, where the dipole-dipole interaction of an excited and a ground-state atom $V\sim\gamma\lambda^3/r^3$ is of the same order as the detuning Δ, we find a contribution $\kappa^2\sqrt{\Delta/\gamma}\sqrt{\lambda/(L\alpha)}$. The justification for this regularization is that when we made the adiabatic elimination we assumed a constant detuning Δ. This approximation breaks down when two atoms are sufficiently close that the dipole-dipole interaction is the strongest effect in the problem, in which case it is more appropriate to describe the atoms in terms of molecular states. Both the short- and long-range part of the interaction are thus small for sufficiently large optical depth α and for sufficiently long ensembles (large L). It should, however, be noted that here we have only performed a very rough treatment of the dipole-dipole interaction, and it would be desirable to make a more accurate treatment of the effects of these terms. Also it should be noted that the estimates we have performed here apply to nonmoving atoms, i.e., cold atoms. If we include the motion of the atoms, i.e., warm atoms as in Refs. [8–10], there will be a reduction of these terms because the sign of the interaction will change in time.

In summary, sufficient requirements for the convergence of the series for the light fields are

$$\frac{\kappa}{\sqrt{N_P}} \ll 1, \quad \frac{\kappa^2}{\sqrt{N_P}} \ll 1, \quad \frac{\kappa^2}{\alpha}\frac{N_A}{N_P} \ll 1, \tag{6.18}$$

and for the spin equation sufficient requirements are

$$\frac{\kappa}{\sqrt{N_A}} \ll 1, \quad \frac{\kappa^2}{\alpha} \ll 1, \quad \kappa^2\sqrt{\frac{d}{L\alpha}} \ll 1,$$

$$\kappa^2\sqrt{\frac{\Delta}{\gamma}}\sqrt{\frac{\lambda}{L\alpha}} \ll 1. \tag{6.19}$$

By having many atoms and photons as well as a large optical depth, it is thus possible to achieve $\kappa\sim 1$ without violating the applicability of perturbation theory.

The main idea in this work is to develop a perturbation series, where we explicitly take into account the reshaping of the light modes caused by the mean effect of the interaction. Let us for comparison compare with the series, if the mean effect of the interaction had not been subtracted. For the Stokes operators the perturbative series is given in Eq. (5.31). If we do not subtract the average effect of the interaction, the scalar part of the interaction [the c_0 component in Eq. (2.4)] will give first-order corrections to the field of order $\kappa\sqrt{N_A}/N_P$ times the incoming field. With $N_A\sim N_P$ as it is suggested in Ref. [7], this term will give a factor of order unity for $\kappa\sim 1$, and this therefore cannot be considered a small term. For the calculation of the Stokes operators, however, the two large components in the first-order terms in Eq. (5.31) cancel out. The calculation may thus yield a reasonable result even without performing the more involved procedures described in this paper, but the validity of the procedure would be questionable. (Some experiments actually use $N_p \gg N_A$ [8], where this problem may be of minor concern.) Furthermore, one of the major limiting factors identified above, is the dipole-dipole interactions. The effect of this term is much more complicated to evaluate if we had not subtracted the average interaction, but the term certainly will be larger, because the interactions in Eq. (F1) would include a nonvanishing term, and not just the quantum fluctuations. Again this term would thus seriously question the applicability of perturbation theory. In contrast the present approach allows us to rigorously apply perturbation theory in experimentally relevant regimes.

VII. CONCLUSION

In quantum optics the propagation of light through an atomic medium is often described in a one-dimensional approximation, where one completely ignores the transverse structure of the beam and only considers the longitudinal

propagation. In this paper we have investigated the validity of this approximation by developing a full three-dimensional theory describing the interaction. The challenge in this work has been to develop a theory capable of describing the microscopic interaction with single atoms as well as macroscopic effects such as the diffraction of the laser beam caused by the refractive index of the gas. In essence the theory we have developed here includes both the microscopic and macroscopic effect by separating the interaction into an average part and the fluctuation from the average. In this formulation macroscopic effects such as diffraction are naturally associated with the average part whereas the microscopic fluctuations describe processes such as the mapping of quantum fluctuations between light and atoms. Furthermore, we have shown that spontaneous emission from the atoms naturally appear as an effect caused by the fluctuations associated with the point particle nature and the random positions of the atoms.

Based on our separation into the average and the fluctuations we have developed a perturbative expansion in the fluctuations. The advantage of this procedure is that it has a wider region of applicability than a direct perturbative treatment. For instance, in an experimental setup, an index of refraction of the gas just changes the beam profile which often only has a minor effect on the experiment. On the other hand, such trivial effects may have a large influence on the theoretical calculation. If one considers perturbation theory based on the vacuum solutions to the wave equation, the perturbative expansion will include all of the terms responsible for the reshaping of the beam, and this may break the validity of perturbation theory. On the other hand, our theory performs perturbation theory on modes which are solutions to the wave equation including the index of refraction of the gas. Our theory is thus applicable even for situations where the beam is considerably distorted by the refractive index of the gas.

A major motivation for this work has been to investigate the validity of the one-dimensional approximation in the description of the experiments in Refs. [8–11]. In Sec. VI we explicitly considered some situations where we could reduce our general theory to a theory resembling the one used to describe these experiments in the one-dimensional approximation [7,29]. To achieve a simple description resembling the previous theories, an essential requirement is that we are in the paraxial approximation. If we are not in this limit, the polarization of the light changes as its propagates through the ensemble, which complicates the interaction with the atoms. Furthermore, for the particular interaction considered here, we also find it to be desirable to be in a regime where the Fresnel number is much larger than unity $\mathcal{F}\gg 1$. In these limits our theory essentially reproduces the results of the simple theory. The only difference is that instead of the vacuum mode functions, the mode functions appearing in the theory should represent the modes, which are solutions to the diffraction problem including the index of refraction of the gas.

In the present paper we have mainly focused on developing the theory and deriving how the usual approximations arise from our more complicated approach. The theory is, however, fully consistent and thus capable of including any higher order corrections not previously included in the theoretical description. In particular it could be interesting to study the effect of light-induced dipole-dipole interactions. While such processes may not be relevant for understanding the current experiments, they may play an important role in future experiments, e.g., with Bose-Einstein condensates, where the density may be fairly high. Another interesting extension of our theory could be to study different types of interactions such as, for instance, electromagnetically induced transparency [19].

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APPENDIX A: ADIABATIC ELIMINATION

In this appendix we derive an effective Hamiltonian involving only the atomic ground state. The Hamiltonian (2.1) can be expanded on the complete set of states describing the atom. Let such a set be comprised of a set of exited states $\{|e_j\rangle\}$ and a set of ground states $\{|g_i\rangle\}$ so that the Hamiltonian reads as

$$\mathcal{H}=\sum_j(\omega_j+\omega_0)|e_j\rangle\langle e_j|+\sum_i\omega_0|g_i\rangle\langle g_i|+\mathcal{H}_{\rm int}. \quad \text{(A1)}$$

For convenience we have here set $\hbar=1$ and only consider a single atom. The set of ground states are assumed to have the same energy, ω_0, and ω_j is the transition frequency from the ground state to the exited state $|e_j\rangle$. The interaction Hamiltonian is given in Eq. (2.3), and when expanded on the set of internal atomic states reads as

$$\mathcal{H}_{\rm int}=-\frac{1}{\epsilon_0}\sum_{ij}\hat{\mathbf{D}}^{(-)}(t)\cdot\langle g_i|\hat{\mathbf{P}}|e_j\rangle|g_i\rangle\langle e_j| + \langle e_j|\hat{\mathbf{P}}|g_i\rangle|e_j\rangle\langle g_i|\cdot\hat{\mathbf{D}}^{(+)}(t), \quad \text{(A2)}$$

where we have used the rotating-wave approximation as well as the fact that the matrix elements $\langle e_j|\hat{\mathbf{P}}|e_{j'}\rangle$ and $\langle g_i|\hat{\mathbf{P}}|g_{i'}\rangle$ vanish. To shorten the notation we suppress the spatial dependence. We will use that the displaced electric field primarily oscillates at the laser frequency, and changes to the interaction picture

$$\hat{\mathbf{D}}^{(-)}(t)\propto e^{i\omega_L t}. \quad \text{(A3)}$$

Using Heisenberg's equations of motion we may derive an equation of motion for $|g_i\rangle\langle e_j|$,

$$\frac{d}{dt}|g_i\rangle\langle e_j|=-i\Delta_j|g_i\rangle\langle e_j|-\frac{i}{\epsilon_0}\sum_{j'}(\langle e_j|\hat{\mathbf{P}}|g_i\rangle|e_{j'}\rangle\langle e_j| - \langle e_j|\hat{\mathbf{P}}|g_{i'}\rangle|g_i\rangle\langle g_{i'}|)\cdot\tilde{\mathbf{D}}^{(+)}(t), \quad \text{(A4)}$$

where $\tilde{\mathbf{D}}^{(\pm)}$ is slowly varying. In the limit of weak driving

we may set $\frac{d}{dt}|g_i\rangle\langle e_j|=0$, and obtain an approximate solution

$$|g_i\rangle\langle e_j| \approx \frac{1}{\epsilon_0\Delta_j}\sum_{i'}\langle e_j|\hat{\mathbf{P}}|g_{i'}\rangle|g_i\rangle\langle g_{i'}|\cdot\hat{\mathbf{D}}^{(+)}(t), \tag{A5}$$

where we have neglected the exited state population. The atomic part of the Hamiltonian can be written as

$$\mathcal{H}_0 = \sum_j \Delta_j|e_j\rangle\langle g_0|g_0\rangle\langle e_j| + \sum_i \omega_L|g_i\rangle\langle g_i| + \sum_{ij}(\omega_0-\omega_L)(|e_j\rangle\langle e_j|+|g_i\rangle\langle g_i|), \tag{A6}$$

where $|g_0\rangle$ is any ground state. By inserting expression (A5) and the Hermitian conjugate into Eqs. (A2) and (A6) we find the simple result

$$\mathcal{H} = -\frac{1}{\epsilon_0}\left(\hat{\mathbf{D}}^{(-)}(t)\cdot\sum_{jii'}\frac{1}{\epsilon_0\Delta_j}\langle g_i|\hat{\mathbf{P}}|e_j\rangle\right)|g_i\rangle \times\langle g_{i'}|[\langle e_j|\hat{\mathbf{P}}|g_{i'}\rangle\cdot\hat{\mathbf{D}}^{(+)}(t)] \tag{A7}$$

(neglecting a zero-point energy term in the Hamiltonian). We may now identify the matrix operator $\bar{\bar{V}}[\hat{J}]$,

$$\bar{\bar{V}}[\hat{J}] = \sum_{jii'}\frac{1}{\epsilon_0\Delta_j}\langle g_i|\hat{\mathbf{P}}|e_j\rangle\langle e_j|\hat{\mathbf{P}}|g_{i'}\rangle|g_i\rangle\langle g_{i'}|\cdot\quad, \tag{A8}$$

and we immediately get the result stated in Eq. (2.3). The notation "·" in this expression means usual vector product with the vector to the right-hand side. Furthermore we may also find the relation between the polarization and the displaced electric field

$$\hat{\mathbf{P}}^{(-)}(t) = \sum_{ij}|e_j\rangle\langle e_j|\hat{\mathbf{P}}|g_i\rangle\langle g_i| = \sum_{jii'}\frac{1}{\epsilon_0\Delta_j}\langle e_j|\hat{\mathbf{P}}|g_i\rangle\langle g_{i'}|\hat{\mathbf{P}}|e_j\rangle|g_{i'}\rangle\langle g_i|\cdot\hat{\mathbf{D}}^{(-)}(t) = \bar{\bar{V}}[\hat{\mathbf{J}}]\hat{\mathbf{D}}^{(-)}(t). \tag{A9}$$

We have here only written the positively oscillating component, the negatively oscillating component is found by Hermitian conjugation, which from Eq. (A8) is the same as transposition of the matrix.

APPENDIX B: CALCULATION OF INFINITELY SHORT PROPAGATOR

In this appendix we calculate the infinitely short propagator in the local density approximation. We will for simplicity only consider the simple interaction given by

$$\bar{\bar{\mathcal{V}}}[\mathbf{J}] = \beta\rho(\mathbf{r})[c_0\mathbf{J}(\mathbf{r})^2 - ic_1\mathbf{J}(\mathbf{r})\times\quad]. \tag{B1}$$

We further shorten the notation by introducing the coefficients $a_0=1-\beta\rho(\mathbf{r})c_0\mathbf{J}(\mathbf{r})^2$ and $a_1=\beta\rho(\mathbf{r})c_1|\mathbf{J}(\mathbf{r})|$.

If we Fourier transform Eq. (2.17), the equation we wish to solve is

$$\hat{\mathbf{k}}\times\hat{\mathbf{k}}\times(a_0+ia_1\hat{\mathbf{j}}\times\quad)\boldsymbol{\epsilon}^{\mathbf{k}} = -\frac{\omega_{\mathbf{k}}^2}{c^2k^2}, \tag{B2a}$$

$$\hat{\mathbf{k}}\cdot\boldsymbol{\epsilon}^{\mathbf{k}} = 0, \tag{B2b}$$

where the vectors $\hat{\mathbf{k}}$ and $\hat{\mathbf{j}}$ are unit vectors representing, respectively, the direction of the plane-wave solution and the orientation of the atomic spin. The solution to the above equations is the following set of polarization vectors,

$$\boldsymbol{\epsilon}_{\pm}^{\mathbf{k}} = N_{\pm}^{\mathbf{k}}\left(\frac{\hat{\mathbf{j}}\times\hat{\mathbf{k}}}{|\hat{\mathbf{j}}\times\hat{\mathbf{k}}|}\pm i\frac{\hat{\mathbf{k}}\times(\hat{\mathbf{j}}\times\hat{\mathbf{k}})}{|\hat{\mathbf{k}}\times(\hat{\mathbf{j}}\times\hat{\mathbf{k}})|}\right) \equiv N_{\pm}^{\mathbf{k}}(\hat{\mathbf{v}}_1\pm i\hat{\mathbf{v}}_2), \tag{B3}$$

where $\hat{\mathbf{v}}_1$ and $\hat{\mathbf{v}}_2$ are unit vectors given by the first and second fraction, respectively. The normalization constant $N_{\pm}^{\mathbf{k}}$ is determined by using the inner product in Eq. (2.18). In this way we find the real space representation of the basis functions $\mathbf{f}_{\mathbf{k}}(\mathbf{r})$,

$$\mathbf{f}_{\pm}^{\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{2(2\pi)^3[a_0\pm a_1(\hat{\mathbf{j}}\cdot\hat{\mathbf{k}})]}}(\hat{\mathbf{v}}_1\pm i\hat{\mathbf{v}}_2)e^{i\mathbf{k}\cdot\mathbf{r}}. \tag{B4}$$

The dispersion relation is then derived from (B2a),

$$\omega_{\mathbf{k}\pm}^2 = c^2k^2[a_0\pm a_1(\hat{\mathbf{j}}\cdot\hat{\mathbf{k}})]. \tag{B5}$$

The infinitely short propagator can then be calculated to be the following:

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t-t') = \frac{-i}{2\omega_Lc^2}\sum_{s\in\{+,-\}}\int d^3k\omega_{\mathbf{k}s}^2[\mathbf{f}_s^{\mathbf{k}}(\mathbf{r})]^*\mathbf{f}_s^{\mathbf{k}}(\mathbf{r})e^{i(t-t')/2\omega_L(\omega_{\mathbf{k}s}^2-\omega_L^2)}. \tag{B6}$$

We introduce the matrix given by the following juxtaposition:

$$\bar{\bar{M}}(\hat{\mathbf{k}},\hat{\mathbf{j}},s) = (\hat{\mathbf{v}}_1-is\hat{\mathbf{v}}_2)(\hat{\mathbf{v}}_1+is\hat{\mathbf{v}}_2). \tag{B7}$$

Changing to spherical coordinates and making the substitutions $x=\cos\theta$ and $k'=k\sqrt{1-a_0+sa_1x}$, as well as using the dispersion relations given in Eq. (B5), the integral reduces to

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t-t') = \frac{-i}{2\omega_Lc^2}\sum_{s\in\{+,-\}}\int_0^\infty dk'\int_{-1}^1 dx\int_0^{2\pi}d\phi\frac{c^2k'^4}{2(2\pi)^3(a_0+sa_1x)^{5/2}} \times\bar{\bar{M}}(x,\phi,s)e^{ic(t-t')(k'^2-k_L^2)/(2k_L)}. \tag{B8}$$

Neglecting the dependence of k' outside the exponential and using that the difference $k'^2-k_L^2$ for large k_L runs from $-\infty$ to ∞, the k' integral gives a δ function in time. Including the ϕ integration in a matrix $\bar{\bar{M}}$ we finally obtain

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t-t') = \frac{-ik_L^3\delta(t-t')}{16\pi^2c^2}\sum_{s\in\{+,-\}}\int_{-1}^1 dx\frac{\bar{\bar{M}}(x,s)}{(a_0+sa_1x)^{5/2}}, \tag{B9}$$

with the matrix $\bar{\bar{M}}$ given by

$$\bar{\bar{M}}(x,s) = \pi \begin{bmatrix} 2(1-x^2) & 0 & 0 \\ 0 & 1+x^2 & 2isx \\ 0 & -2isx & 1+x^2 \end{bmatrix}. \quad \text{(B10)}$$

The s sum is evaluated by substitution in the integral and the final expression for the infinitely short propagator is

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t-t') = \frac{-ik_L^3\delta(t-t')}{8\pi c^2}\int_{-1}^{1} dx \frac{\bar{\bar{M}}(x,+)}{\pi(a_0+a_1x)^{5/2}}. \quad \text{(B11)}$$

Thise integral may be evaluated, and we will express the infinitely short propagator as

$$\bar{\bar{P}}^{(-)}(\mathbf{r},t-t') = \frac{-i\delta(t-t')}{c^2} \begin{bmatrix} \varrho_{\parallel} & 0 & 0 \\ 0 & \varrho_{\perp} & -i\varrho_{\Gamma} \\ 0 & i\varrho_{\Gamma} & \varrho_{\perp} \end{bmatrix}. \quad \text{(B12)}$$

The coefficients for $a_0 - a_1 > 0$, are given by

$$\varrho_{\parallel} = \frac{-k_L^3}{3\pi a_1^3}\left(\frac{-4a_0+2a_1}{\sqrt{a_0-a_1}} + \frac{4a_0+2a_1}{\sqrt{a_0+a_1}}\right), \quad \text{(B13a)}$$

$$\varrho_{\perp} = \frac{-k_L^3}{3\pi a_1^3}\left(\frac{2a_0^2-3a_0a_1+\frac{1}{2}a_1^2}{(a_0-a_1)^{3/2}} - \frac{2a_0^2+3a_0a_1+\frac{1}{2}a_1^2}{(a_0+a_1)^{3/2}}\right), \quad \text{(B13b)}$$

$$\varrho_{\Gamma} = \frac{k_L^3}{6\pi a_1^2}\left(\frac{2a_0-3a_1}{(a_0-a_1)^{3/2}} - \frac{2a_0+3a_1}{(a_0+a_1)^{3/2}}\right). \quad \text{(B13c)}$$

APPENDIX C: RECIPROCAL EQUATION FOR GREEN'S FUNCTION

In this appendix we derive the reciprocal equation for the Green's function. Before doing so we will need some results concerning the representation of the Green's function. Let us define the following inner product:

$$\langle \boldsymbol{\varphi} | \boldsymbol{\psi} \rangle = \int d^3r dt \bar{\bar{\mathcal{M}}}(\mathbf{r}) \boldsymbol{\varphi}(\mathbf{r},t) \cdot \boldsymbol{\psi}^{\dagger}(\mathbf{r},t). \quad \text{(C1)}$$

We will generally work in the L^2 space equipped with this inner product. Using that the matrix operator $\bar{\bar{\mathcal{M}}}$ is Hermitian, one finds the differential operator $\mathcal{D}$ given in Eq. (4.1) to be Hermitian in our inner product space

$$\langle \boldsymbol{\varphi} | \mathcal{D}\boldsymbol{\psi} \rangle = \langle \mathcal{D}\boldsymbol{\varphi} | \boldsymbol{\psi} \rangle. \quad \text{(C2)}$$

That $\mathcal{D}$ is Hermitian means that the eigenfunctions F_k to $\mathcal{D}$,

$$\mathcal{D}\mathbf{F}_{\mathbf{k}}(\mathbf{r},t) = \lambda_{\mathbf{k}}\mathbf{F}_{\mathbf{k}}(\mathbf{r},t), \quad \text{(C3)}$$

define a complete basis of our inner product space $\{\mathbf{F}_{\mathbf{k}}\}$. A representation of the identity functional given in Eq. (4.2) may therefore be

$$\sum_{\mathbf{k}} \mathbf{F}_{\mathbf{k}}^{\dagger}(\mathbf{r},t)\mathbf{F}_{\mathbf{k}}(\mathbf{r}_0,t_0). \quad \text{(C4)}$$

It can be checked that this is exactly a functional identity representation in our inner product space by expanding any function on the basis $\{\mathbf{F}_{\mathbf{k}}\}$.

To get a formal expression of the Green's function defined in Eq. (4.2) we expand the Green's function in this basis, and using Eqs. (C3) and (C4) we find

$$\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}_0,t_0) = \sum_{\mathbf{k}} \frac{1}{\lambda_{\mathbf{k}}} \mathbf{F}_{\mathbf{k}}^{\dagger}(\mathbf{r},t)\mathbf{F}_{\mathbf{k}}(\mathbf{r}_0,t_0). \quad \text{(C5)}$$

Starting from Eq. (4.2) we make the substitution $t \to -t$, $t_0 \to -t_1$, and $\mathbf{r}_0 \to \mathbf{r}_1$ and we write the following:

$$\mathcal{D}^* \bar{\bar{G}}(\mathbf{r},-t|r_1,-t_1) = \bar{\bar{I}}\delta(\mathbf{r},\mathbf{r}_1)\delta(t,t_1). \quad \text{(C6)}$$

In the next step we take the inner product with Eq. (4.2) and $\bar{\bar{G}}(\mathbf{r},-t|r_1,-t_1)$ from the left-hand side with respect to unprimed coordinates, and Eq. (C6) and $\bar{\bar{G}}(\mathbf{r},t|r_0,t_0)$ also from the left-hand side with respect to unprimed coordinates. The resulting two equations are then subtracted. The term containing ω_L^2 vanish trivially, and using rules for differentiating a product, the resulting equation may be written as

$$\begin{aligned} 2i\omega_L \iint d^3r dt \bar{\bar{\mathcal{M}}}(\mathbf{r}) \frac{\partial}{\partial t}[\bar{\bar{G}}(\mathbf{r},-t|\mathbf{r}_1,-t_1) \cdot \bar{\bar{G}}(\mathbf{r},t|\mathbf{r}_0,t_0)] \\ + c^2 \iint d^3dt[\bar{\bar{\mathcal{M}}}(\mathbf{r})\bar{\bar{G}}(\mathbf{r},-t|\mathbf{r}_1,-t_1) \cdot \boldsymbol{\nabla} \times \boldsymbol{\nabla} \\ \times \bar{\bar{\mathcal{M}}}(\mathbf{r})\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}_0,t_0) - \bar{\bar{\mathcal{M}}}(\mathbf{r})\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}_0,t_0) \cdot \boldsymbol{\nabla} \times \boldsymbol{\nabla} \\ \times \bar{\bar{\mathcal{M}}}(\mathbf{r})\bar{\bar{G}}(\mathbf{r},-t|\mathbf{r}_1,-t_1)] \\ = \bar{\bar{G}}(\mathbf{r}_1,t_1|\mathbf{r}_0,t_0) - \bar{\bar{G}}(\mathbf{r}_0,-t_0|\mathbf{r}_1,-t_1). \end{aligned} \quad \text{(C7)}$$

Using the cutoff property of the Green's function, the first term on the left-hand side is seen to vanish. Using the explicit expression for the Green's function (C5) along with the Gauss theorem, one may show that the second term also vanishes. The final result is therefore

$$\bar{\bar{G}}(\mathbf{r}_1,t_1|\mathbf{r}_0,t_0) = \bar{\bar{G}}(\mathbf{r}_0,-t_0|\mathbf{r}_1,-t_1). \quad \text{(C8)}$$

From Eqs. (4.2) and (C8), and using the substitutions $t \to -t'$, $t_0 \to t$, $\mathbf{r} \to \mathbf{r}'$, and $\mathbf{r}_0 \to \mathbf{r}$ we end up with the *reciprocal equation*

$$\begin{aligned} \left(-2i\omega_L\frac{\partial}{\partial t'} - \omega_L^2 + c^2\boldsymbol{\nabla}' \times \boldsymbol{\nabla}' \times \bar{\bar{\mathcal{M}}}(\mathbf{r}')\right)\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t') \\ = \bar{\bar{I}}\delta(\mathbf{r},\mathbf{r}')\delta(t,t'). \end{aligned} \quad \text{(C9)}$$

In the following we derive the general solution to the equation

$$\left(2i\omega_L\frac{\partial}{\partial t}-\omega_L^2+c^2\boldsymbol{\nabla}\times\boldsymbol{\nabla}\times\bar{\bar{\mathcal{M}}}(\mathbf{r})\right)\boldsymbol{\psi}(\mathbf{r},t)=\boldsymbol{\rho}(\mathbf{r},t), \tag{C10}$$

where $\boldsymbol{\psi}(\mathbf{r},t)$ is an unknown field, $\boldsymbol{\rho}(\mathbf{r},t)$ is a source term effecting the solution, and $\bar{\bar{\mathcal{M}}}$ is some Hermitian matrix operator, which may depend on position. We make an inner product of Eq. (C10) with $\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t')$ from the left-hand side and an inner product of Eq. (C9) with $\boldsymbol{\psi}(\mathbf{r},t)$ from the right-hand side and subtract these two equations. In this calculation we are integrating over the time interval $t'\in]t_0,t^+[$, where we understand $t^+=\lim_{\epsilon\to 0}(t+\epsilon)$. Again we find that terms containing ω_L^2 vanish. Similar to the above, we will use rules for differentiation as a product, and we eventually end up with

$$\begin{aligned}\boldsymbol{\psi}(\mathbf{r},t)-\int\int_{t_0}^{t^+}d^3r'dt'\bar{\bar{\mathcal{M}}}(\mathbf{r}')\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t')\cdot\boldsymbol{\rho}(\mathbf{r}',t')\\ =-2i\omega_L\int\int_{t_0}^{t^+}d^3r'dt'\bar{\bar{\mathcal{M}}}(\mathbf{r}')\frac{\partial}{\partial t}[\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t')\cdot\boldsymbol{\psi}(\mathbf{r}',t')]\\ +c^2\int\int_{t_0}^{t^+}d^3r'dt'\bar{\bar{\mathcal{M}}}(\mathbf{r}')[\boldsymbol{\psi}(\mathbf{r}',t')\cdot\boldsymbol{\nabla}'\times\boldsymbol{\nabla}'\\ \times\bar{\bar{\mathcal{M}}}(\mathbf{r}')\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t')-\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t')\\ \cdot\boldsymbol{\nabla}'\times\boldsymbol{\nabla}'\times\boldsymbol{\psi}(\mathbf{r}',t')].\end{aligned} \tag{C11}$$

Using the same boundary conditions as was done in the calculation leading to the reciprocal equation we conclude that the last term in Eq. (C11) vanishes. The right-hand side of the equation thus reduces to

$$\begin{aligned}-2i\omega_L\int d^3r'\bar{\bar{\mathcal{M}}}(\mathbf{r}')[\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t')\cdot\boldsymbol{\psi}(\mathbf{r}',t')]_{t_0}^{t^+}\\ =2i\omega_L\int d^3r'\bar{\bar{\mathcal{M}}}(\mathbf{r}')\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t_0)\cdot\boldsymbol{\psi}(\mathbf{r}',t_0).\end{aligned} \tag{C12}$$

Here we have used that the upper time limit vanishes due to the cutoff in the Green's function. Rearranging terms we finally arrive at the general solution to the diffusion equation

$$\begin{aligned}\boldsymbol{\psi}(\mathbf{r},t)=2i\omega_L\int d^3r'\bar{\bar{\mathcal{M}}}(\mathbf{r}')\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t_0)\cdot\boldsymbol{\psi}(\mathbf{r}',t_0)\\ +\int\int_{t_0}^{t}d^3r'dt'\bar{\bar{\mathcal{M}}}(\mathbf{r}')\bar{\bar{G}}(\mathbf{r},t|\mathbf{r}',t')\cdot\boldsymbol{\rho}(\mathbf{r}',t').\end{aligned} \tag{C13}$$

APPENDIX D: LORENTZ-LORENZ RELATION

In the main text we consider lowest order corrections to the index of refraction. To verify that our theory can also correctly reproduce higher order corrections, we shall in this appendix show how to derive the so-called Lorentz-Lorenz or Clausius-Mossotti relation for the electric permittivity within our theoretical framework [37]. To lowest order the permittivity is given by Eq. (3.11),

$$\bar{\bar{\epsilon}}(\mathbf{r})^{-1}=1-\bar{\bar{\mathcal{V}}}[\mathbf{J}]. \tag{D1}$$

To calculate the higher order correction it is convenient to first Fourier transform the Dyson equation (4.9) describing the light field with respect to time,

$$\begin{aligned}\tilde{\mathbf{D}}^{(-)}(\mathbf{r},\omega)=\tilde{\mathbf{D}}_0^{(-)}(\mathbf{r},\omega)+c^2\int d^3r'\bar{\bar{P}}^{(-)}(\mathbf{r},\mathbf{r}',\omega)\cdot\bar{\bar{m}}[\hat{\mathbf{J}}]^t\\ \times\tilde{\mathbf{D}}^{(-)}(\mathbf{r}',\omega).\end{aligned} \tag{D2}$$

This equation is the starting point for the analysis. The Fourier transformation is here defined as

$$f(\omega)=\int_0^\infty dte^{(i\omega-\eta)t}f(t), \tag{D3}$$

where η is an infinitely small convergence factor.

From Eq. (4.24) we find the Fourier transformed propagator $\bar{\bar{P}}^{(-)}$ to read as

$$\bar{\bar{P}}^{(-)}(\mathbf{r},\mathbf{r}',\omega)=\frac{1}{c^2}\sum_{\mathbf{k}}\frac{\omega_{\mathbf{k}}^2\mathbf{f}_{\mathbf{k}}^*(\mathbf{r})\mathbf{f}_{\mathbf{k}}(\mathbf{r}')}{\omega_{\mathbf{k}}^2-\omega_L^2+2\omega_L(\omega+i\eta)}. \tag{D4}$$

The real space representation of this propagator is in general difficult to calculate, however, for a scalar interaction the calculations simplify considerably. For $\omega\approx 0$ which is reasonable in our case, since we are dealing with slowly varying operators, the propagator reads as

$$\begin{aligned}\bar{\bar{P}}^{(-)}(\mathbf{n})=\int\frac{d^3k}{c^2(2\pi)^3}\sum_{\boldsymbol{\epsilon}\perp\mathbf{k}}\boldsymbol{\epsilon}\boldsymbol{\epsilon}\frac{\mathbf{k}^2e^{i\mathbf{k}\cdot\mathbf{n}}}{\mathbf{k}^2-k_L^2}\\ =-\frac{k_L^3}{c^2 4\pi}\frac{e^{ik_Ln}}{k_Ln}\left[\left(1+\frac{3i}{k_Ln}-\frac{3}{(k_Ln)^2}\right)\frac{\mathbf{nn}}{\mathbf{n}^2}\right.\\ \left.-\left(1+\frac{i}{k_Ln}-\frac{1}{(k_Ln)^2}\right)\bar{\bar{I}}\right]+\frac{2}{3}\bar{\bar{I}}\delta(\mathbf{n}),\end{aligned} \tag{D5}$$

where $\mathbf{n}=\mathbf{r}-\mathbf{r}'$, $n=|\mathbf{n}|$, and $\bar{\bar{I}}$ is the identity matrix. We notice that the propagator gives us the well-known result for the radiated field of an oscillating dipole. In addition we have a term describing a self-interaction. This propagator is also discussed in Ref. [35]. In the following we shall only be considering the self-interaction part of the propagator.

When considering the density correlation function to second order $\langle\rho(\mathbf{r}_1)\rho(\mathbf{r}_2)\rangle$ we have so far used the ideal gas approximation in Eq. (4.13), where there are no correlations between different atoms. In reality we can never have two atoms at the same position and this gives a small correction to $\langle\rho(\mathbf{r}_1)\rho(\mathbf{r}_2)\rangle$, which must vanish for $\mathbf{r}_1=\mathbf{r}_2$ (apart from the δ function, which represents the single atom contribution). This can formally be described by introducing the so-called irreducible correlation functions h_2 such that

$$\langle \rho(\mathbf{r}_1)\rho(\mathbf{r}_2)\rangle = \langle \rho(\mathbf{r}_1)\rangle\langle \rho(\mathbf{r}_2)\rangle + h_2(\mathbf{r}_1,\mathbf{r}_2), \tag{D6}$$

where h_2 now takes care of the core repulsion of the atoms (here we exclude the δ function). For $\mathbf{r}_1=\mathbf{r}_2$ we thus find that $h_2(\mathbf{r}_1,\mathbf{r}_1)=-\langle\rho(\mathbf{r}_1)\rangle^2$.

The above can be used along with the real space representation of the propagator to give the second-order correction to the permittivity. We will not consider terms that vanish when we take quantum mechanical mean. The relevant part of the second-order term thus gives in shorthand notation $-\int \bar{\bar{P}}^{(-)}(2/3)(\bar{\bar{\mathcal{V}}}[\mathbf{J}])^2\tilde{\mathbf{D}}^{(-)}$. When we introduce this interaction to the differential equation (3.11) we find the permittivity to second order,

$$\bar{\bar{\epsilon}}(\mathbf{r})^{-1} = 1 - \bar{\bar{\mathcal{V}}}[\mathbf{J}] + \frac{2}{3}(\bar{\bar{\mathcal{V}}}[\mathbf{J}])^2. \tag{D7}$$

The calculation can be continued to infinite order [36], and the result reads as

$$\bar{\bar{\epsilon}}(\mathbf{r})^{-1} = 1 - \bar{\bar{\mathcal{V}}}[\mathbf{J}] - \bar{\bar{\mathcal{V}}}[\mathbf{J}]\sum_{n=1}^{\infty}\left(\frac{-2}{3}\bar{\bar{\mathcal{V}}}[\mathbf{J}]\right)^n = \frac{1-\frac{1}{3}\bar{\bar{\mathcal{V}}}[\mathbf{J}]}{1+\frac{2}{3}\bar{\bar{\mathcal{V}}}[\mathbf{J}]}. \tag{D8}$$

This is the Lorenz-Lorenz relation, and we thus see that the effect can be included in the theory by dressing the spatial mode functions according to the result above.

APPENDIX E: CALCULATIONS OF SECOND-ORDER STOKES GENERATOR

In this appendix we present detailed calculations of the second-order terms of Eq. (5.31). We will denote the fourth term on the right-hand side of Eq. (5.31) as $\bar{\bar{\mathcal{S}}}_A^{(2)}$, and one finds

$$K\langle\langle \tilde{\mathbf{f}}^*_{kmj}(\mathbf{r},t)|\ \bar{\bar{\mathcal{S}}}_A^{(2)}\ |\tilde{\mathbf{f}}^*_{km'j'}(\mathbf{r},t)\rangle\rangle = \left(\frac{1}{2}\right)^2(k_L\beta c_1)^2\iint d^3r d^3r' \sum_{\substack{ln\\ l'n'}} \rho(\mathbf{r})\rho(\mathbf{r}') \times \Theta^{mn}_{jl}(\mathbf{r})^*\Theta^{m'n'}_{j'l'}(\mathbf{r}')\hat{a}^\dagger_{knl}\hat{a}_{kn'l'}. \tag{E1}$$

The seventh term on the right-hand side of Eq. (5.31) plus its complex conjugate is denoted as $\bar{\bar{\mathcal{S}}}_B^{(2)}$. To calculate this term we extend the limits of the time integration from minus to plus infinity. This we can do by introducing a factor of 1/2, and approximating the imaginary term $i\int_{-\infty}^0 dt\ \sin(\omega t)$ to be zero. This corresponds to the usual treatment of such terms in the Markov approximation to spontaneous emission when one ignores the Lamb shift. We then find the following contribution to the Stokes operators:

$$K\langle\langle \tilde{\mathbf{f}}^*_{kmj}(\mathbf{r},t)|\ \bar{\bar{\mathcal{S}}}_B^{(2)}\ |\tilde{\mathbf{f}}^*_{km'j'}(\mathbf{r},t)\rangle\rangle = \left(\frac{1}{2}\right)^3(k_L\beta c_1)^2\iint d^3r d^3r' \sum_{\substack{ln\\ l'n'}} \rho(\mathbf{r})\rho(\mathbf{r}') \times[\Theta^{mn}_{jl}(\mathbf{r})^*\Theta^{nn'}_{ll'}(\mathbf{r}')^*\hat{a}^\dagger_{kn'l'}\hat{a}_{km'j'} + \Theta^{m'n}_{j'l}(\mathbf{r})\Theta^{nn'}_{ll'}(\mathbf{r}')\hat{a}^\dagger_{kmj}\hat{a}_{kn'l'}]. \tag{E2}$$

One notices that the factors of 1/2 in Eqs. (E1) and (E2) exactly add up to give one-half of the square of the first-order term, as is shown in Eq. (6.2).

The sixth term on the right-hand side of Eq. (5.31), plus its Hermitian conjugate, we will denote as $\bar{\bar{\mathcal{S}}}_C^{(2)}$, and we find

$$K\langle\langle \tilde{\mathbf{f}}^*_{kmj}(\mathbf{r},t)|\ \bar{\bar{\mathcal{S}}}_C^{(2)}\ |\tilde{\mathbf{f}}^*_{km'j'}(\mathbf{r},t)\rangle\rangle = \left(\frac{1}{2}\right)^3(k_L\beta c_1)^2\int d^3r' \sum_{\substack{ln\\ ql'n'\\ n''l''}} \rho(\mathbf{r}')[\mathcal{C}^{l'l''}_{jl}(\mathbf{r}')\Psi^{mn}_k(\mathbf{r}')^* \times\Psi^{n'n''}_q(\mathbf{r}')\hat{a}^\dagger_{qn'l'}\hat{a}_{qn''l''}\hat{a}^\dagger_{knl}\hat{a}_{km'j'} + \hat{a}^\dagger_{kmj}\hat{a}^\dagger_{qn'l'}\hat{a}_{qn''l''}\hat{a}_{knl}\mathcal{C}^{l'l''}_{j'l}(\mathbf{r}')\Psi^{m'n}_k(\mathbf{r}')\Psi^{n'n''}_q(\mathbf{r}')^*], \tag{E3}$$

where we have introduced the coefficients

$$\mathcal{C}^{l'l''}_{jl}(\mathbf{r}) = \mathbf{e}_j(\mathbf{r})\cdot(\{\bar{\mathbf{J}}(\mathbf{r})\times[\mathbf{e}_{l'}(\mathbf{r})\times\mathbf{e}_{l''}(\mathbf{r})]\}\times\mathbf{e}_l(\mathbf{r})). \tag{E4}$$

This term can be shown to vanish by expanding the spin operator $\bar{\mathbf{J}}$ on the basis defined by the polarization vectors $\mathbf{e}_x(\mathbf{r})$, $\mathbf{e}_y(\mathbf{r})$, and $\mathbf{e}_z(\mathbf{r})$ and using that the indices j, l, l', and l'' only run over x and y.

Finally we will calculate the effect of the fifth term on the right-hand side of Eq. (5.31), which we will denote as $\bar{\bar{\mathcal{S}}}_D^{(2)}$. In this calculation it is important to remember that the term will scale as $\beta^2\rho$, and reads as

$$K\langle\langle \tilde{\mathbf{f}}^*_{kmj}(\mathbf{r},t)|\ \bar{\bar{\mathcal{S}}}_D^{(2)}\ |\tilde{\mathbf{f}}^*_{km'j'}(\mathbf{r},t)\rangle\rangle = \left(\frac{1}{2}\right)^2(k_L\beta)^2\int d^3r \sum_{\substack{ln\\ l'n'}} \rho(\mathbf{r})\Psi^{nm}_k(\mathbf{r})\Psi^{m'n'}_k(\mathbf{r}) \times\hat{a}^\dagger_{knl}\hat{a}_{kn'l'}\{c_1^2[\bar{\mathbf{J}}(\mathbf{r})\cdot\mathbf{e}_z(\mathbf{r})]^2(\delta_{jy}\delta_{lx}-\delta_{jx}\delta_{ly}) \times(\delta_{j'y}\delta_{l'x}-\delta_{j'x}\delta_{l'y}) + c_0^2\mathbf{J}(\mathbf{r})^4\delta_{jl}\delta_{j'l'}\}. \tag{E5}$$

APPENDIX F: CALCULATION OF SECOND-ORDER SPIN TERMS

In this section we calculate the second-order terms for the atomic spin, represented as the third and fourth terms on the right-hand side of Eq. (5.42). These terms are denoted as

$\mathcal{J}_A^{(2)}$, and using the previous notation one finds

$$\mathcal{J}_A^{(2)} = \frac{-i}{2}\left(\frac{\beta c_1 k_L}{2}\right)^2 \sum_k \int d^3r' \sum_{\substack{m'm'' \\ l}} [\bar{\mathbf{J}}(\mathbf{r}) \times \mathbf{e}_z(\mathbf{r})] \times \left[\begin{pmatrix} 0 \\ J_y(\mathbf{r}') \\ \bar{J}_z(\mathbf{r}') \end{pmatrix} \cdot \mathbf{e}_z(\mathbf{r}')\right] \rho(\mathbf{r}') \hat{a}^\dagger_{km'l} \hat{a}_{km''l} \times \sum_m [\Psi_k^{mm''}(\mathbf{r}) \Psi_k^{m'm}(\mathbf{r}') - \Psi_k^{m'm}(\mathbf{r}) \Psi_k^{mm''}(\mathbf{r}')]. \tag{F1}$$

We can examine this term by assuming that the only photon carrying modes of the light are the two modes $\mathbf{f}_{kox}$ and $\mathbf{f}_{ko'y}$ and neglect all other modes. In this case the term reduces to

$$\mathcal{J}_A^{(2)} = \left(\frac{\beta c_1 k_L}{2}\right)^2 \sum_k \int d^3r' \sum_{m \in \{(o,x),(o',y)\}} [\bar{\mathbf{J}}(\mathbf{r}) \times \mathbf{e}_z(\mathbf{r})] \times \left[\begin{pmatrix} 0 \\ J_y(\mathbf{r}') \\ \bar{J}_z(\mathbf{r}') \end{pmatrix} \cdot \mathbf{e}_z(\mathbf{r}')\right] \rho(\mathbf{r}') \hat{a}^\dagger_{knl} \hat{a}_{knl} \, \mathrm{Im}[\Psi_k^{mn}(\mathbf{r}) \Psi_k^{nm}(\mathbf{r}')]. \tag{F2}$$

This term represents an atom at position $\mathbf{r}'$ interacting with the light field and emitting a photon into mode m, which propagates to the position $\mathbf{r}$, where it is absorbed by an atom followed by stimulated emission into the classical beam. This process is also known as optically induced dipole-dipole interaction, and indeed the sum over all modes m can be used to introduce the dipole propagator in (D5). Note, however, that above we have written the term in the paraxial approximation, where we ignore the dependence of the polarization vector on the mode number. Since the sum over m involves all modes, and not just the paraxial modes, an accurate treatment requires a more complicated expression involving the polarization vectors along the lines of Appendix H (we use this more complicated expression in our estimates of the size of the effect).

The last term we will consider is the term describing an atom interacting with the light field at two different times. This term is represented as the fifth term on the right-hand side of Eq. (5.42), and is given in vector component form in Eq. (5.13). We will denote this term with $\mathcal{J}_B^{(2)}$. A short calculation gives

$$\mathcal{J}_B^{(2)} = -\frac{1}{2}\left(\frac{\beta c_1 k_L}{2}\right)^2 \sum_{kk'} \sum_{\substack{mm' \\ nn'}} \sum_{\substack{jj' \\ l}} \mathbf{e}_l(\bar{\mathbf{J}} \cdot \mathbf{e}_{j'}) \times [\Psi_k^{mn} \Psi_{k'}^{m'n'} (\hat{a}^\dagger_{kml} \hat{a}^\dagger_{k'm'j} \hat{a}_{knj} \hat{a}_{k'n'j'} - \hat{a}^\dagger_{kmj} \hat{a}^\dagger_{k'm'j} \hat{a}_{knj'} \hat{a}_{k'n'l}) + \mathrm{H.c.}], \tag{F3}$$

where we have suppressed the spatial dependence to shorten the notation. Doing the sum over j, j' and l we obtain

$$\mathcal{J}_B^{(2)} = -\frac{1}{2}\left(\frac{\beta c_1 k_L}{2}\right)^2 \sum_{kk'} \sum_{\substack{mm' \\ nn'}} [\bar{\mathbf{J}} - \mathbf{e}_z(\bar{\mathbf{J}} \cdot \mathbf{e}_z)] \times \Psi_k^{mn} \Psi_{k'}^{m'n'} (2\hat{a}^\dagger_{kmx} \hat{a}^\dagger_{k'm'y} \hat{a}_{kny} \hat{a}_{k'n'x} - \hat{a}^\dagger_{kmy} \hat{a}^\dagger_{k'm'y} \hat{a}_{knx} \hat{a}_{k'n'x} - \hat{a}^\dagger_{kmx} \hat{a}^\dagger_{k'm'x} \hat{a}_{kny} \hat{a}_{k'n'y}). \tag{F4}$$

The first-order term in Eq. (5.43) describes the first-order effect of rotation of the spin around the $e_z(\mathbf{r})$ axis. The second-order term in (F4) describes the second-order term of this rotation. From the rotation frequency in the first-order term $\propto s_3$ (assuming Ψ to be real), one would thus expect this term to scale as $\beta^2 (s_3)^2$ which is different from the term in (F4). This difference arises because we have separated the term into normal-ordered components such that the second-order term in (F4) only contributes when at least two photons are present. When we performed the normal ordering in the diagram we introduced an additional term, which we described by the third term in Eq. (5.4)

APPENDIX G: CALCULATION OF SPONTANEOUS EMISSION

In this section we calculate the corrections to Eqs. (6.2) and (6.3), due to the incoherent interaction. To do this we need a result for the infinitely short propagator. From the definition of the propagator (4.24) and the calculation of it in (5.8), we find the relation

$$\sum_n |U_n(\mathbf{r}_\perp)|^2 = \frac{2}{k_L} \varrho(\mathbf{r}_\perp), \tag{G1}$$

where $\varrho(\mathbf{r}_\perp) = k_L^3/(16\pi^2)$ is the zeroth-order term of the expansion of $\varrho_\parallel(\mathbf{r})$ in β given in Eq. (B13). This result is important when calculating $\bar{\bar{S}}_d^{(2)}$ and for relating this term with the incoherent interactions, responsible for spontaneous emission. When including this term and the decay described in Sec. V B, the incoherent interaction reduces to

$$\hat{s}_{1,\mathrm{out}}(\mathbf{r}_\perp) = \cdots - \frac{\beta^2 k_L \varrho(\mathbf{r}_\perp)}{2} \int dz' \rho(z') (c_1^2 [\bar{J}_y^2(z') - \bar{J}_z^2(z')] \times \hat{s}_{0,\mathrm{in}}(\mathbf{r}_\perp) + \{c_0^2 \mathbf{J}^4(z') + c_1^2 [4J_z^2(z') + J_y^2(z')]\} \hat{s}_{1,\mathrm{in}}(\mathbf{r}_\perp)), \tag{G2a}$$

$$\hat{s}_{2,\mathrm{out}}(\mathbf{r}_\perp) = \cdots - \frac{\beta^2 k_L \varrho(\mathbf{r}_\perp)}{2} \int dz' \rho(z') \{c_0^2 \mathbf{J}^4(z') + c_1^2 [3J_z^2(z') + J_y^2(z') + J_x^2(z')]\} \hat{s}_{2,\mathrm{in}}(\mathbf{r}_\perp), \tag{G2b}$$

$$\hat{s}_{3,\mathrm{out}}(\mathbf{r}_\perp) = \cdots - \frac{\beta^2 k_L \varrho(\mathbf{r}_\perp)}{2} \int dz' \rho(z') \{c_0^2 \mathbf{J}^4(z') + c_1^2 [J_z^2(z') + J_y^2(z') + J_x^2(z')]\} \hat{s}_{3,\mathrm{in}}(\mathbf{r}_\perp), \tag{G2c}$$

where we have only kept terms that are nonvanishing after taking quantum mechanical average of the atomic spin. The operators $\hat{s}_{0,\mathrm{in}}(\mathbf{r}_\perp)$ measures the total photon flux, and is given as

$$\hat{s}_0(\mathbf{r}_\perp) = \sum_{kmm'} \frac{1}{2} [U_m^*(\mathbf{r}_\perp) \hat{a}_{kmx}^\dagger \hat{a}_{km'x} U_{m'}(\mathbf{r}_\perp) + U_m^*(\mathbf{r}_\perp) \hat{a}_{kmy}^\dagger \hat{a}_{km'y} U_{m'}(\mathbf{r}_\perp)]. \tag{G3}$$

It is important to note that in a discussion of the various contributions to decay one should include all terms in the perturbative expansion, including the loop diagrams in Eq. (5.15). If these are not included one finds the contribution from the term in Eq. (E5) to increase the operator $\hat{s}_3$. Similarly we find the effect of spontaneous emission on the spin equation to read as

$$\bar{J}_{x,\mathrm{out}}(z) = \cdots - \beta^2 c_1^2 k_L \varrho(\mathbf{r}_\perp) \sum_k \left[\bar{J}_{x,\mathrm{in}}(z) \left(\hat{s}_{0,\mathrm{in}}^k(\mathbf{r}_\perp) + \frac{1}{2} \hat{s}_{1,\mathrm{in}}^k(\mathbf{r}_\perp) \right) + \frac{1}{2} \bar{J}_{y,\mathrm{in}}(z) \hat{s}_{2,\mathrm{in}}^k(\mathbf{r}_\perp) \right], \tag{G4a}$$

$$\bar{J}_{y,\mathrm{out}}(z) = \cdots - \beta^2 c_1^2 k_L \varrho(\mathbf{r}_\perp) \sum_k \left[\bar{J}_{y,\mathrm{in}}(z) \left(\hat{s}_{0,\mathrm{in}}^k(\mathbf{r}_\perp) + \frac{1}{2} \hat{s}_{1,\mathrm{in}}^k(\mathbf{r}_\perp) \right) + \frac{1}{2} \bar{J}_{x,\mathrm{in}}(z) \hat{s}_{2,\mathrm{in}}^k(\mathbf{r}_\perp) \right], \tag{G4b}$$

$$\bar{J}_{z,\mathrm{out}}(z) = \cdots - \beta^2 c_1^2 k_L \varrho(\mathbf{r}_\perp) \sum_k \bar{J}_{z,\mathrm{in}}(z) \hat{s}_{0,\mathrm{in}}^k(\mathbf{r}_\perp). \tag{G4c}$$

The above result is derived from Eq. (5.9) by using the paraxial approximation and only keeping terms of order β^2. A minor correction is introduced in Eq. (6.3) since we chose a representation that was in fact not normal ordered.

APPENDIX H: BEYOND PARAXIAL APPROXIMATION

In this section we will go slightly beyond the approximation made in Eq. (5.30), and consider the set

$$\mathbf{f}_\mathbf{q}(\mathbf{r}) = \frac{1}{\sqrt{2\pi}} U_{nq}(\mathbf{r}) \mathbf{e}_{nj}(\mathbf{r}). \tag{H1}$$

We will consider the correction this generalization makes to the result given in Eq. (6.9), and therefore define spin components in the local basis given by the set $\mathbf{e}_{mx}(\mathbf{r})$, $\mathbf{e}_{my}(\mathbf{r})$, and $\mathbf{e}_{mz}(\mathbf{r})$,

$$\bar{\mathcal{J}}_{\mathbf{e}_{mi}}(\mathbf{r}) = \begin{pmatrix} 0 \\ \bar{J}_y(\mathbf{r}) \\ \bar{J}_z(\mathbf{r}) \end{pmatrix} \cdot \mathbf{e}_{mi}(\mathbf{r}) \tag{H2}$$

for $i \in \{x,y,z\}$. These vectors are defined by the fact that, e.g., $\mathbf{e}_{oy}(\mathbf{r})$ should be transverse and perpendicular to the polarization vector arising from the mode function $U_{ok}(\mathbf{r})\mathbf{e}_{ox}(\mathbf{r})$. $\mathbf{e}_{oz}$ is then defined by $\mathbf{e}_{oz} = \mathbf{e}_{ox} \times \mathbf{e}_{oy}$. Similarly for the quantum modes m, the definition of $\mathbf{e}_{mx}$ follows from the fact that it should be perpendicular to the polarization vector from the mode $U_{mk}(\mathbf{r})\mathbf{e}_{my}(\mathbf{r})$.

With these definitions, Eq. (6.9) gives

$$\hat{X}_{\mathrm{out}}^m = \hat{X}_{\mathrm{in}}^m + k_L \beta c_1 \sqrt{\frac{N_x^o}{2}} \int d^3r' \rho(\mathbf{r}') \mathrm{Re}[\Psi_k^{mo}(\mathbf{r})] \{\bar{\mathcal{J}}_{\mathbf{e}_{oz}}(\mathbf{r}) \times [\mathbf{e}_{ox}(\mathbf{r}) \cdot \mathbf{e}_{mx}(\mathbf{r})] - \bar{\mathcal{J}}_{\mathbf{e}_{ox}}(\mathbf{r}) [\mathbf{e}_{ox}(\mathbf{r}) \cdot \mathbf{e}_{mz}(\mathbf{r})]\}, \tag{H3a}$$

$$\hat{P}_{\mathrm{out}}^m = \hat{P}_{\mathrm{in}}^m + k_L \beta c_1 \sqrt{\frac{N_x^o}{2}} \int d^3r' \rho(\mathbf{r}') \mathrm{Im}[\Psi_k^{mo}(\mathbf{r})] \{\bar{\mathcal{J}}_{\mathbf{e}_{oz}}(\mathbf{r}) \times [\mathbf{e}_{ox}(\mathbf{r}) \cdot \mathbf{e}_{mx}(\mathbf{r})] - \bar{\mathcal{J}}_{\mathbf{e}_{ox}}(\mathbf{r}) [\mathbf{e}_{ox}(\mathbf{r}) \cdot \mathbf{e}_{mz}(\mathbf{r})]\}. \tag{H3b}$$

Similarly we find the correction to Eq. (6.10) to give

$$\bar{\mathbf{J}}_{\mathrm{out}}(\mathbf{r}) \approx \bar{\mathbf{J}}_{\mathrm{in}}(\mathbf{r}) + k_L \beta c_1 \sqrt{\frac{N_x^o}{2}} \sum_n \{\mathrm{Re}[\Psi_k^{no}(\mathbf{r})] \hat{P}_{\mathrm{in}}^n - \mathrm{Im}[\Psi_k^{no}(\mathbf{r})] \hat{X}_{\mathrm{in}}^n\} \{\bar{\mathbf{J}}_{\mathrm{in}}(\mathbf{r}) \times [\mathbf{e}_{ox}(\mathbf{r}) \times \mathbf{e}_{ny}(\mathbf{r})]\}. \tag{H4}$$

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