Quantum Memory for Light

Retrieval of stored photons from Λ-type atomic ensembles involving coupling of transverse modes

BACHELOR'S THESIS
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Abstract in Danish - Resumé på dansk

Overordnet handler denne bacheloropgave om hvorledes kvantetilstanden for en lyspuls kan gemmes i, og senere udlæses fra, en gas af atomer, hvorved sidstnævnte fungerer som en kvantehukommelse. Dette hører ind under feltet kvanteoptik, som netop beskæftiger sig med lysets kvanteegenskaber, som de manifesterer sig i vekselvirkningen med atomer. Kvantehukommelse udgør en fundamental byggesten i mange kvanteinformationsteknologiske anvendelser, herunder kvantecomputeren, hvorfor arbejdet med at forstå denne komponents virkemåde, samt hvordan dens begrænsende faktorer bedst håndteres, er et vigtigt led i bestræbelserne på at konstruere effektive kvanteinformationssystemer.

En kvantehukommelse baseret på en gas af atomer kan konstrueres på en række forskellige måder. Denne opgave beskæftiger sig med det tilfælde, hvor interaktionen mellem lys og atomer er bestemt ved en Hamiltonoperator af strålesplitter-typen. Dette tilfælde er allerede blevet analyseret i ganske stor detaljegrad i [3, 5], men til forskel fra disse studier vil udledningerne i denne opgave blive foretaget under antagelse af en cylindrisk symmetrisk fordeling af atomerne i gasskyen; en sådan antagelse medfører, at der forekommer kobling mellem forskellige transverse modes.

Formålet med denne opgave er at arbejde hen imod en forståelse af, hvor høj grad af pålidelighed der kan opnås i forbindelse med udlæsning fra en kvantehukommelse af ovenfor beskrevne type. Mere specifikt vil et formelt udtryk for effektiviteten af udlæsning fra kvantehukommelsen blive udledt. Ud fra dette udtryk er det muligt at foretage en numerisk bestemmelse af systemets egenmodes og de respektive effektiviteter, hvormed de kan udlæses.

I opgaven redegøres ydermere for den bagvedliggende kvantemekaniske formalisme og de approksimationer, der anvendes for at simplificere udledningerne.

Contents

1 Introduction

The present thesis is concerned with the theoretical aspects of a certain approach to constructing an interface between light and an atomic ensemble with the purpose of making the latter serve as a quantum memory. In the following subsections this task will be laid out in the context of both its theoretical foundation in *quantum optics* and its applicability in the field of *quantum information processing*. This will provide the necessary motivation and background for this largely theoretical treatment of the subject at hand.

1.1 Quantum Optics and Quantum Information Processing

The field of quantum optics deals with the quantum features of light and the manifestations of these in the interaction between light and matter. For this, a fully quantum mechanical treatment of the light field is needed (a procedure for quantizing the electromagnetic field is outlined in section 2.1.1). As implied by the word 'light', quantum optics is characterized by being concerned with electromagnetic radiation fields in the optical wavelength domain, that is, in the neighborhood of 400 to 700 nm.

The light-atoms interface, where the quantum state of light is transferred to atoms and vice versa, plays a fundamental role in the field of quantum information processing. One physical realization of this interface is the single-mode cavity containing only a single atom or a few atoms. However, this approach (so-called *cavity QED*) has proven to be impractical for applications due to the delicate nature of the setup.

Rather, we shall deal with the interaction between light and an optically dense ensemble of many atoms. An important feature of this setup is that we can map light mode excitations to collective superposition states of many atoms. The coupling between light field and atomic ensemble can be achieved using various different techniques, as will be touched upon in the following subsection, but a common feature is the *optical depth* d as an important parameter in determining the coupling strength; the greater the optical depth of the atomic ensemble, the stronger the coupling. [3, 5] A great deal of the current research activity in the field of quantum information processing, both theoretical and experimental, is directed towards the ensemble approach.

A basic quantum information component is the *quantum memory*, which can be realized by interfacing light with an atomic ensemble, the latter serving as the memory, and using a classical control field to facilitate read and write operations. This *ensemble based atomic memory for light* is the subject of this thesis.

We will by no means give a comprehensive introduction to the field of quantum information processing here. However, a few examples of the role of the quantum memory is in order: For instance, any kind of quantum computer will consist of at least a light interface, a memory unit, and a processor manipulating the data in that memory. Another example is the quantum repeater component that enables long-distance quantum communications; quantum memory plays an integral part in this device, too.

In this thesis, we will only consider the direct interaction between light and atoms. However, the generation and utilization of entanglement between light and an atomic ensemble, as well as entanglement between atomic ensembles, is of vast importance. The fascinating concept of *quantum teleportation*, which has been demonstrated experimentally, relies heavily on entanglement.

1.2 Interaction types and atomic level diagrams

Several techniques can be used to realize the interaction between light and an atomic ensemble. From a theoretical standpoint these can be grouped into interaction types based on the structure of the interaction Hamiltonian pertaining to each technique. In this thesis, we will focus on a single such interaction type: *Beam splitter* interaction, being the class of techniques which are formally equivalent to the beam splitter (as far as the structure of the interaction Hamiltonian goes). Examples of techniques with beam splitter type interaction include the Electromagnetically Induced Transparency and far-off-resonant Raman techniques. In this subsection we will explain how this interaction type arises between light and atoms.

We will consider physical situations where, essentially, only a few specific levels of the atom come into play (as will be commented on below), so that it is a reasonable approximation to disregard all other levels.[1] In this sense, we will consider 3-level atoms with ground states $|0\rangle, |1\rangle$ and an excited state $|e\rangle$ in the Λ-type configuration depicted in the figure below. The light field will consist of two components: A weak quantum signal (with central frequency ω_0) carrying the information to be stored into, or that has been read out from, the atomic quantum memory; and a strong classical control field (of frequency ω_0') that facilitates the storage and retrieval operations.

[1] In a detailed treatment of the impact of spontaneous emission, however, one would have to consider additional levels.

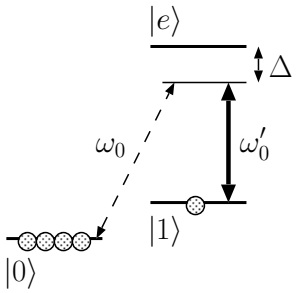

Figure 1: Λ-type model atom with a quantum field (dashed line) acting on the $|0\rangle \leftrightarrow |e\rangle$ transition and a classical control field (solid line) acting on the $|1\rangle \leftrightarrow |e\rangle$ transition. These fields have a common detuning Δ with respect to the excited level $|e\rangle$.

The quantum signal component of the light field acts on the $|0\rangle \leftrightarrow |e\rangle$ transition, whereas the classical control field acts on the $|1\rangle \leftrightarrow |e\rangle$ transition. This specific arrangement of light field components and atomic levels results in beam splitter type interaction. As indicated in the figure, we will concern ourselves with the situation where almost all atoms in the ensemble are in the $|0\rangle$ state, while relatively few are in the other ground state $|1\rangle$ (in this respect, figure 1 is not only representing a single atom in the ensemble). The collective state of all atoms being in their respective $|0\rangle$ states corresponds to an empty memory. Roughly speaking, the storing of photons works as follows: For each excitation removed from the light field, one atom is brought from $|0\rangle$ into the $|1\rangle$ state via the "temporary" state $|e\rangle$. However, this is an oversimplification: Rather than the photon being stored in a single atom state, it is stored in a collective state of the ensemble; (this is sometimes referred to as a *spin-wave* excitation since the ensemble can be described using a spin formalism as we shall see below).

We will briefly discuss what requirements the above modeling puts on real atoms and light fields for the setup to be realizable. Firstly, the spacings to the atomic levels not drawn in figure 1 need to be sufficiently large that transitions to these level are very strongly suppressed and hence negligible. Secondly, we need to ensure that the constituents of the light field, quantum signal and control field, only couple to the transitions indicated in figure 1. This can be realized for instance by making the two components have (the mutually orthogonal) left and right circular polarization, respectively, and use the hyperfine levels $S_{1/2}(f = i \pm 1/2)$ to play the role of the ground states $|0\rangle$ and $|1\rangle$ [5]. Note that electric dipole transitions between these hyperfine levels are forbidden by parity; hence, as desired, the light field cannot cause transitions to happen between the ground states $|0\rangle, |1\rangle$ (at least in the dipole approximation to be introduced in subsection 3.1.1). In experiments, alkali gases are often used as the atomic medium, a typical choice being Caesium.

1.3 Figure of merit and sources of loss

To be able to assess the performance of the quantum memory in a quantitative fashion, we need a figure of merit. Such a measure will provide us with the understanding of how various parameters affect the performance and will thereby enable us to optimize it. Moreover, it allows for comparison with the best classical device serving the same function, which is an important benchmark. The choice of figure of merit is not unique; furthermore, which quantity is the best measure of performance depends on the specific application. Similarly, the amount of error that can be tolerated depends on the application and kind of error in question.

In this thesis, we will focus on the process of retrieval from an atomic memory. We will use as our figure of merit the *retrieval efficiency* defined as follows: Given an initial excitation in one of the collective atomic modes, the retrieval efficiency is the probability of finding a photon in some output mode after the retrieval operation. Note that with this definition we make no demands as to which output mode(s) the excitation is read out into. This corresponds to the experimental situation of photon counting.

In general, the efficiency depends on how well-adapted the control field is to the quantum signal. However, a study similar[2] to this one has shown that for the case of retrieval where we only care about the number of photons (and not their modes), the efficiency is independent of the control field (and its detuning Δ) provided that it is of sufficient strength or duration to remove all excitations from the atomic ensemble [3]. The efficiency does, however, depend on the mode of the stored excitation. These aspects are discussed in great detail in said reference.

Various sources of loss are present in the realization of a quantum memory presented here, some more fundamental than others. Among those inherent to the physical system, an important example is the finite lifetime

[2]The analysis in [3] is carried out using an assumption of the atomic number density being independent of the transverse coordinates $n(\vec{r}) = n(z)$; we shall use a different assumption here.

of the excited atomic level; spontaneous emission from the excited state $|e\rangle$ (with decay rate γ) is a source of decoherence that must be taken into account (and we will do so in the present analysis). A similar, but less pronounced, source of loss is decay from the meta-stable atomic state $|1\rangle$ through "forbidden" transitions. For simplicity, we will ignore this effect here; a non-zero γ_1 will simply result in exponential decay in time of the output light field at the output end of the ensemble as discussed in subsection VI E of [3].

Other sources of loss in connection with retrieval include the undesired transitions due to atoms colliding with one another or the container wall. Furthermore, the movement of atoms will smear out the stored modes, generally resulting in less efficiently retrieved modes.

As always, deviation from ideal conditions will introduce errors. In this respect, the unification provided by the beam splitter interaction type introduced in subsection 1.2 cannot be retained; the impact of imperfections on the reliability of various techniques varies a great deal and each mechanism must be analyzed separately. However, these issues are outside the scope of this thesis.

1.4 Outline of thesis

The goal of this thesis is to derive a formal expression for the retrieval efficiency of an atomic ensemble quantum memory where the light-atoms interaction is governed by a beam splitter Hamiltonian. This analysis has already been carried out in references [3, 5], but under somewhat simplistic assumptions in some respects. The present analysis will be carried out using a slightly more realistic assumption regarding the number density distribution $n(\vec{r})$ of the atomic ensemble, which entails coupling between distinct modes of the light field and the collective atomic excitations (using the same set of modes for both) as we shall see below.

The analysis is organized as follows:

- ✿ In section 2, we will introduce the respective formalisms that will be used to treat the light field and the atomic ensemble.
- ✿ In subsection 3.1, an effective interaction Hamiltonian for the light field and a single atom will be derived using various approximations.
- ✿ In subsection 3.2, we superpose these interaction Hamiltonians and rewrite in terms of continuous collective atomic operators to obtain the light - ensemble interaction Hamiltonian. Additional assumptions regarding the physical setup will be made.
- ✿ In section 4, the equations of motion of the system are determined and then solved formally in section 5 using the specific basis of Bessel beams.
- ✿ In subsection 5.5, the formal solution is restated in terms of dimensionless variables and from this an expression for the retrieval efficiency is derived in subsection 6.
- ✿ In section 7, we will briefly touch upon how the formal expression for the retrieval efficiency can be evaluated numerically.

Details of some derivations have been deferred to appendices in an attempt to achieve a certain degree of continuity in the presentation.

2 Formalism

We will follow the approach of reference [5] with regard to the formalism being used to describe the light field and the atomic ensemble. Accordingly, we will work almost exclusively with operators rather than state kets. It is, however, still useful to consider what parts of the ket space are acted upon by the various operators. The total ket space of the system is $\mathcal{E} = \mathcal{E}_{\text{EM}} \otimes \mathcal{E}_{\text{ensemble}}$; light mode operators act exclusively on the former part, while atomic mode operators act exclusively on the latter, whereas operator products including operators from both categories will couple the two parts of the ket space.

In the process of setting up the formalism and deriving the Hamiltonian, we will work in the Schrödinger picture. But eventually we will derive the equations of motion in a Heisenberg rotating frame.

We will use CGS units and set $\hbar = 1$, whereby the units of energy and frequency coincide.

2.1 Description of the light field

2.1.1 Quantization of the electromagnetic field

In order to properly treat the interaction between matter and light, a quantum mechanical description of the electromagnetic field is needed. Such a description can be achieved through the procedure outlined in the following (see references [6, 7] for a full treatment):

Write down the classical equations of motion for the combined system of matter and the electromagnetic field, which are the Newton-Lorentz equations for the former part and Maxwell's equations for the latter. We then obtain from these the classical Hamiltonian $H(\{q_i\},\{p_j\})$ by means of a variant (involving Lagrangian and Hamiltonian densities) of the usual procedure known from analytical mechanics. The canonical position variables $\{q_i\}$ corresponding to the degrees of freedom of the electromagnetic field turn out to be the transverse components[3] $\vec{A}_\perp$ of the vector potential $\vec{A}$, whereas the scalar potential ϕ is not an independent dynamical variable and the longitudinal component $\vec{A}_\parallel$ is a non-physical gauge degree of freedom (which we fix by choosing Coulomb gauge $\nabla\cdot\vec{A}=0$).

For now we will only consider the EM field part $H_{\rm EM}$ of the total classical Hamiltonian $H = H_{\rm EM}+H_{\rm matter}+H_{\rm int}$, while deferring the treatment of the remaining parts.[4] By suitable canonical coordinate transformations to so-called normal variables $a_{\vec{k},\mu}$ (and their complex conjugates $a^*_{\vec{k},\mu}$), the electromagnetic field part $H_{\rm EM}$ of the classical Hamiltonian can be cast in the form of a sum of harmonic oscillator Hamiltonian functions,

$$H_{\rm EM} = \sum_{(\vec{k},\mu)} \omega_k |a_{\vec{k},\mu}|^2 \tag{1}$$

(having set $\hbar=1$), where the sum[5] is over EM field modes, each being characterized by a wave vector $\vec{k}$ and a polarization index μ (there are two independent polarizations for each $\vec{k}$); the corresponding polarization vector is denoted $\vec{\epsilon}_{\vec{k},\mu}$. The angular frequency $\omega_{\vec{k}}$ is related to $\vec{k}$ through the vacuum dispersion relation $\omega_{\vec{k}} = |\vec{k}|c$.

The vector potential $\vec{A}$ for the free field in Coulomb gauge is to obey the wave equation

$$\nabla^2\vec{A}(\vec{r},t) - \frac{1}{c^2}\frac{\partial^2\vec{A}(\vec{r},t)}{\partial t^2} = 0; \tag{2}$$

using separation of variables $\vec{A}(\vec{r},t) = \vec{A}(\vec{r})f(t)$ and denoting $-\frac{1}{c^2}\frac{\partial^2 f(t)}{\partial t^2} = k^2$, we obtain the spatial part of the equation - the Helmholtz equation:

$$(\nabla^2 + k^2)\vec{A}(\vec{r}) = 0. \tag{3}$$

At this point, we may pick any complete set of solutions $\{\vec{\epsilon}_{\vec{k},\mu}g_{\vec{k}}(\vec{r})\}_{\vec{k},\mu}$ to this equation; we will need two independent polarization vectors $\vec{\epsilon}_{\vec{k},\mu}$ ($\mu=1,2$) for each $\vec{k}$ as $\vec{A}(\vec{r},t)$ is a transverse vector field in Coulomb gauge. Then arbitrary $\vec{A}(\vec{r},t)$ can be expanded on this set with time-dependent coefficients $a_{\vec{k},\mu}(t)$ whose equations of motion are given by eq. (2). In the case of the interacting field, we can equally well expand $\vec{A}(\vec{r},t)$ on the set $\{\vec{\epsilon}_{\vec{k},\mu}g_{\vec{k}}(\vec{r})\}$; the equations of motion for the expansion coefficients, however, will be given by eq. (2) with the source term $-\frac{4\pi}{c}\vec{J}_\perp$ added to the right-hand side. The quality of using a complete set of solutions to eq. (3) as a basis is that these modes represent waves of definite $\vec{k}$; hence the EM Hamiltonian function $H_{\rm EM}$ in eq. (1) makes sense as a measure of the energy of the radiation field. In either case, the expansion is conventionally written as:

$$\vec{A}(\vec{r},t) = \sqrt{\frac{2\pi c^2}{V}}\sum_{(\vec{k},\mu)}\frac{1}{\sqrt{\omega_{\vec{k}}}}(\vec{\epsilon}_{\vec{k},\mu}a_{\vec{k},\mu}(t)g_{\vec{k}}(\vec{r}) + \vec{\epsilon}^{\,*}_{\vec{k},\mu}a^*_{\vec{k},\mu}(t)g^*_{\vec{k}}(\vec{r})), \tag{4}$$

where V is the quantization volume. A similar expression for the transverse part of the electric field $\vec{E}_\perp(\vec{r},t)$ can be obtained[6]

$$\vec{E}_\perp(\vec{r},t) = \sqrt{\frac{2\pi}{V}}\sum_{(\vec{k},\mu)}\sqrt{\omega_{\vec{k}}}(i\vec{\epsilon}_{\vec{k},\mu}a_{\vec{k},\mu}(t)g_{\vec{k}}(\vec{r}) - i\vec{\epsilon}^{\,*}_{\vec{k},\mu}a^*_{\vec{k},\mu}(t)g^*_{\vec{k}}(\vec{r})); \tag{5}$$

[3] See reference [6] for a discussion of transverse and longitudinal vector fields.

[4] The results obtained in the following hold true for both free and interacting EM fields; however, the equations of motion for the canonical variables involved depend on whether one considers interaction or not.

[5] We assume a discretized $\vec{k}$-space as would result from imposing periodic boundary conditions.

[6] Although $\vec{E}_\perp(\vec{r},t) = -\frac{1}{c}\frac{\partial\vec{A}(\vec{r},t)}{\partial t}$, this is not the easiest way of getting eq. (5) as the time-dependence of $a_{\vec{k},\mu}(t)$ may be complicated in the case of the interacting field. See reference [6] for details.

it is exactly $\vec{E}_\perp(\vec{r},t)$ that will appear in the final form of the interaction Hamiltonian $H_{\rm int}$, as we shall see in subsection 3.1.1.[7]

To quantize the EM field Hamiltonian $H_{\rm EM}$, we reinterpret $a_{\vec{k},\mu}$ and $a^*_{\vec{k},\mu}$ as quantum harmonic oscillator annihilation and creation operators $a_{\vec{k},\mu}$ and $a^\dagger_{\vec{k},\mu}$, respectively, satisfying the usual commutation relations $[a_{\vec{k},\mu}, a^\dagger_{\vec{k}',\mu'}] = \delta_{\vec{k},\vec{k}'}\delta_{\mu,\mu'}$ and $[a_{\vec{k},\mu}, a_{\vec{k}',\mu'}] = [a^\dagger_{\vec{k},\mu}, a^\dagger_{\vec{k}',\mu'}] = 0$. In the classical Hamiltonian, writing $|a_{\vec{k},\mu}|^2$ is unambiguous since $a_{\vec{k},\mu}a^*_{\vec{k},\mu} = a^*_{\vec{k},\mu}a_{\vec{k},\mu}$; however, the ordering does matter once we reinterpret the normal variables as quantum operators and we will choose the normally ordered version.[8] Hence, the Hamiltonian *operator* for the electromagnetic field is

$$H_{\rm EM} = \sum_{(\vec{k},\mu)} \omega_{\vec{k}} a^\dagger_{\vec{k},\mu} a_{\vec{k},\mu}. \tag{6}$$

By reinterpreting the normal mode expansions of $\vec{A}$ and $\vec{E}_\perp$, eqs. (4) and (5), in a similar fashion, we obtain the following quantum fields (operator valued functions) in the Schrödinger picture

$$\begin{aligned}
\vec{A}(\vec{r}) &= \sqrt{\frac{2\pi c^2}{V}} \sum_{(\vec{k},\mu)} \frac{1}{\sqrt{\omega_{\vec{k}}}} (\vec{\epsilon}_{\vec{k},\mu} a_{\vec{k},\mu} g_{\vec{k}}(\vec{r}) + \vec{\epsilon}^*_{\vec{k},\mu} a^\dagger_{\vec{k},\mu} g^*_{\vec{k}}(\vec{r})) \\
\vec{E}_\perp(\vec{r}) &= \sqrt{\frac{2\pi}{V}} \sum_{(\vec{k},\mu)} \sqrt{\omega_{\vec{k}}} (i\vec{\epsilon}_{\vec{k},\mu} a_{\vec{k},\mu} g_{\vec{k}}(\vec{r}) - i\vec{\epsilon}^*_{\vec{k},\mu} a^\dagger_{\vec{k},\mu} g^*_{\vec{k}}(\vec{r})) \\
&= \sqrt{\frac{2\pi}{V}} \sum_{(\vec{k},\mu)} \sqrt{\omega_{\vec{k}}} (i\vec{\epsilon}_{\vec{k},\mu} a_{\vec{k},\mu} g_{\vec{k}}(\vec{r}) + \text{H.C.}),
\end{aligned} \tag{7}$$

where "H.C." stands for Hermitian conjugate.

2.1.2 Paraxial approximation, mode functions, and slowly varying operators

In the particular model for interaction between light and atoms to be investigated, we will consider forward-propagating light beams with $\vec{k}$-vectors that we assume to be essentially pointing in the positive $\hat{z}$-direction; this is the *paraxial approximation*. The gas is taken to be confined in a "cylinder" of length L and infinite radius whose symmetry axis coincides with the aforementioned z-axis.

We will assume that each mode in the paraxial subset of $\{g_{\vec{k}}(\vec{r})\}_{\vec{k}}$ can be factorized as $g_{\vec{k}}(\vec{r}) = u_n(\vec{r}_\perp, z)e^{ikz}$, where $k = |\vec{k}|$, so that each $\vec{k}$ corresponds to a pair of indices $(n,k) \in \mathbb{Z}\times\mathbb{R}_+$, at least in an approximative sense within the paraxial regime. In subsection 5.2 we will solve the Helmholtz equation in cylindrical coordinates to obtain a specific mode set $\{g_{\vec{k}}(\vec{r})\}_{\vec{k}}$ and carry out the approximate factorization just mentioned; but for now, we will be slightly more general and work with the unspecified mode functions $g_{\vec{k}}(\vec{r}) = u_n(\vec{r}_\perp, z)e^{ikz}$. Moreover, we will assume the set $\{u_m(\vec{r}_\perp, z)\}$ to be complete and orthogonal in any plane $z = z_0$ transverse to the beam direction, that is to say

$$\int d^2\vec{r}_\perp u^*_m(\vec{r}_\perp, z) u_n(\vec{r}_\perp, z) = \delta_{mn}, \tag{8}$$

so that any operator $O(\vec{r},t)$ can be expanded on the set as $O(\vec{r},t) = \sum_m u_m(\vec{r}_\perp, z)O_m(z,t)$ with $O_m(z,t) = \int d^2\vec{r}_\perp u^*_m(\vec{r}_\perp, z)O(\vec{r},t)$.

As described in subsection 1.2, the light field will consist of two components, a weak quantum signal and a strong control field:

$$\vec{E}_\perp(\vec{r}) = \vec{E}_q(\vec{r}) + \vec{E}_c(\vec{r}) = E_q(\vec{r})\vec{\epsilon}_q + E_c(\vec{r})\vec{\epsilon}_c, \tag{9}$$

introducing the additional simplification of only allowing one polarization vector for each part; we choose these polarization vectors in a way that realizes the beam splitter coupling scheme described in subsection 1.2.

The control field (of frequency ω_0') is assumed to be of sufficient strength to be treated as a classical field so that we can replace the field operator by its expectation value with respect to a coherent state $E_c(\vec{r},t) \approx \langle E_c(\vec{r})\rangle$. Hence, for this part of the field we do not need the quantized description developed above.

[7] Meanwhile, the longitudinal part of the electric field $\vec{E}_\parallel(\vec{r},t)$, which arises from the electrostatic potential of atomic charges, will not enter the interaction Hamiltonian in its final form.

[8] Normal ordering results in a vanishing vacuum energy, for instance. That the quantization procedure cannot be entirely deductive stems from the fact that quantum mechanics contains more information than classical mechanics. In the end, experiments must decide whether the quantized theory is correct or not.

For the remainder of this section we will focus on the quantum signal part $\vec{E}_q(\vec{r},t)$ of which we will assume that all frequency components lie in a narrow band around ω_0, $\omega_k \approx \omega_0$. Using this and the paraxial approximation as well as the fact that we are only considering one polarization $\vec{\epsilon}_q$ for the quantum signal, eq. (7) can be written[9]

$$\vec{E}_q(\vec{r}) = \sqrt{\frac{2\pi\omega_0}{l}} \sum_{(m,k)} (\vec{\epsilon}_q u_m(\vec{r}_\perp, z) a_{m,k} e^{ikz} + \text{H.C.}), \tag{10}$$

where we have absorbed a factor of i into the definition of the (generally complex) polarization vector.

We now introduce the slowly varying, position-dependent annihilation operators defined as

$$a_m(z,t) \equiv \sqrt{\frac{c}{l}} \sum_k e^{i(k-k_0)z + i\omega_0 t} a_{m,k}, \tag{11}$$

where the dominant time dependence of $a_{m,k}$ (in the Heisenberg picture) has been removed. In the continuum limit $l \to \infty$, the slowly varying operators $a_m(z,t)$ can be shown to have the (equal time) commutation relations $[a_m(z), a^\dagger_{m'}(z')] = c\delta_{m,m'}\delta(z-z')$ and $[a_m(z), a_{m'}(z')] = 0$. $a^\dagger_m(z)a_m(z)$ is the flux of photons ("photons per time") at z in the mode m. Rearranging eq. (11) to get $\frac{1}{\sqrt{c}} a_m(z) e^{i(k_0 z - \omega_0 t)} = \frac{1}{\sqrt{l}} \sum_k e^{ikz} a_{m,k}$ and plugging this into eq. (10) we get

$$\vec{E}_q(\vec{r}) = \sqrt{\frac{2\pi\omega_0}{c}} \sum_m (\vec{\epsilon}_q u_m(\vec{r}_\perp, z) a_{\mathrm{L},m}(z,t) e^{i(k_0 z - \omega_0 t)} + \text{H.C.}), \tag{12}$$

where we have tagged a subscript 'L' onto the light annihilation operators to distinguish them from the collective atomic annihilation operators to be introduced below. Equation (12) is the final expression for the Schrödinger operator $\vec{E}_q(\vec{r})$ that we will use below; note that its explicit time dependence is only apparent, whereas the slowly varying $a_{\mathrm{L},m}(z,t)$ does have explicit time dependence.

2.1.3 The light field Hamiltonian

The electromagnetic field Hamiltonian was derived in subsection 2.1.1 and stated in eq. (6). In the previous subsection we introduced the paraxial approximation, allowed only a single polarization vector $\vec{\epsilon}_q$ for the quantum field and expanded on the orthogonal set of mode functions $\{g_{\vec{k}}(\vec{r}) = u_m(\vec{r}_\perp, z)e^{ikz}\}$; with these changes, the Hamiltonian of the light field can now be stated as

$$H_{\mathrm{EM}} = \sum_{(m,k)} kc a^\dagger_{\mathrm{L},mk} a_{\mathrm{L},mk}, \tag{13}$$

where we have inserted the dispersion relation $\omega_k = kc$ ($k > 0$, as we consider only forward propagating light components). The light mode Hamiltonian will enter the equation of motion of the quantum signal light field annihilation operators $a_{\mathrm{L},m}(z,t)$.

The state space $\mathcal{E}_{\mathrm{EM}}$ that the operators $a_{\mathrm{L},mk}, a^\dagger_{\mathrm{L},mk}$ act on is a so-called Fock space.[10] A natural basis consists of states designated by a list of occupation numbers, one for each field mode: $|\{n_{m,k}\}\rangle = \prod_{(m,k)} \otimes |n_{m,k}\rangle$, these are eigenstates of H_{EM}.

2.2 Description of the atomic ensemble

In section 1.2, we described how we will model the individual atoms of the ensemble as effective 3-level atoms. In this section, we will transform the atomic Hamiltonian H_{A} to a rotating frame and develop a way of treating the ensemble as a whole in terms of an angular momentum formalism. In turn, these collective angular momentum-like operators can be rephrased in terms of harmonic oscillator annihilation and creation operators in a certain approximation. Finally, we will make the shift to continuous, position-dependent operators. We will not, however, reexpress the atomic Hamiltonian in terms of the continuous operators at this point for reasons to become clear below.

We will label operators pertaining to an individual atom (or rather, operators that act specifically on the associated single atom Hilbert space) with an integer index i. Ultimately, we are interested in describing an

[9] Here we are normalizing with respect to the length l (in the $\hat{z}$ direction) of the virtual quantization volume. The physical limit, in which the k-values form a continuum, is $l \to \infty$. Note that l is not to be confused with the length of the gas container L.

[10] A Fock space has an uncountable basis, whereas the basis of a Hilbert space must be countable. This distinction will not concern us here. (A countably infinite product of countably infinite sets is uncountable.)

ensemble of atoms for which the corresponding Hilbert space $\mathcal{E}_{\text{ensemble}}$ is the tensor product of a number of single atom Hilbert spaces $\mathcal{E}_i$:

$$\mathcal{E}_{\text{ensemble}} = \mathcal{E}_1 \otimes \mathcal{E}_2 \otimes \ldots \otimes \mathcal{E}_{N_\text{A}}, \tag{14}$$

where N_A is the number of atoms in the ensemble.

2.2.1 The atomic Hamiltonian in a rotating frame

We will decompose the atomic Hamiltonian H_A of the entire ensemble into a sum of Hamiltonians for the individual atoms, $H_\text{A} = \sum_i H_\text{A}^{(i)}$, corresponding to the decomposition of the Hilbert space seen in eq. (14). In what follows, we will work in the Hilbert space of the i'th atom.

Given that the only accessible states of the atom are two ground states $\{|0\rangle^{(i)}, |1\rangle^{(i)}\}$ with energies E_0 and E_1, respectively, and an excited state $\{|e\rangle^{(i)}\}$ of energy E_e, the atomic Hamiltonian $H_\text{A}^{(i)}$ can be written in diagonal form in terms of its eigenstates as

$$H_\text{A}^{(i)} = \sum_{g=0}^{1} E_g |g\rangle^{(i)}\langle g| + E_e |e\rangle^{(i)}\langle e|.$$

In this section and below we will transform the various parts of the total Hamiltonian to a rotating frame in order to simplify our calculations and to be able to carry out certain approximations. More specifically, we will go to a rotating frame in the sense described in Appendix A; as stated there, the rotating frame Hamiltonian is given by

$$H_\text{I} = i\dot{U}^\dagger U + U^\dagger H_\text{S} U, \tag{15}$$

where U is a unitary operator. Our focus in this section is on the purely atomic part of the Hamiltonian, but of course we have to be consistent in applying the transformation by considering the effect on all parts of the Hamiltonian. The physics behind the transformation to be performed is that only frequency differences matter,[11] it is therefore desirable to get rid of the dominant time dependence due to the free evolution of the system.

Now consider the following unitary transformation which is defined in terms of Schrödinger picture operators that act on the Hilbert space of the i'th atom $\mathcal{E}_i$,

$$U^{(i)} = \exp\left[-it\left(\sum_{g=0}^{1} E_g |g\rangle^{(i)}\langle g| + (E_e - \Delta)|e\rangle^{(i)}\langle e|\right)\right], \tag{16}$$

where we have introduced the common detuning of the light fields with respect to the excited state (see figure 1) $\Delta = (E_e - E_0) - \omega_0 = (E_e - E_1) - \omega_0'$, which is the difference between resonance frequency and light frequency. Transforming $H_\text{A}^{(i)}$ by $U^{(i)}$ according to eq. (15), noting that $\dot{U}^{(i)\dagger} = U^{(i)\dagger} i\left(\sum_{g=0}^{1} E_g |g\rangle^{(i)}\langle g| + (E_e - \Delta)|e\rangle^{(i)}\langle e|\right)$ since the exponent of $U^{(i)\dagger}$ commutes with its own partial derivative w.r.t. t, we get

$$\begin{aligned} H'^{(i)}_\text{A} &= -U^{(i)\dagger}\left(\sum_{g=0}^{1} E_g |g\rangle^{(i)}\langle g| + (E_e - \Delta)|e\rangle^{(i)}\langle e|\right) U^{(i)} + U^{(i)\dagger}\left(\sum_{g=0}^{1} E_g |g\rangle^{(i)}\langle g| + E_e |e\rangle^{(i)}\langle e|\right) U^{(i)} \\ &= (E_e - (E_e - \Delta))|e\rangle^{(i)}\langle e| = \Delta |e\rangle^{(i)}\langle e|, \end{aligned} \tag{17}$$

where we have made use of the fact that $[U^{(i)}, |e\rangle^{(i)}\langle e|] = 0 = [U^{(i)}, |g\rangle^{(i)}\langle g|]$ for $g = 0, 1$, which follows from the following obvious property of the orthogonal projection operators $[|g\rangle^{(i)}\langle g|, |e\rangle^{(i)}\langle e|] = 0 = [|g\rangle^{(i)}\langle g|, |g'\rangle^{(i)}\langle g'|]$ for $g, g' \in \{0, 1\}$.

As mentioned above, we must be careful about keeping track of how the transformation affects the other parts of the total Hamiltonian. We note that the transformation $U^{(i)}$ leaves H_EM and $H_\text{A}^{(j)}$, $j \neq i$, invariant because $U^{(i)}$ does not act on these degrees of freedom of the total state space. However, we will need to pay attention to the effect of the transformation specified by $U^{(i)}$ on $H_\text{int}^{(i)}$, the interaction Hamiltonian of the i'th atom, to be considered below.

[11] For the purposes of this thesis, this is true. But if one are to derive how spontaneous emission enters the equations of motion, the absolute frequencies matter in general.

2.2.2 Angular momentum operator formulation

The ground states $|0\rangle, |1\rangle$ of the Λ-type system can be described in terms of spin-$\frac{1}{2}$ angular momentum operators; for each m we define the following operators, using the x-axis as quantization axis

$$j_{x,m} = \frac{1}{2}(|0\rangle^{(m)}\langle 0| - |1\rangle^{(m)}\langle 1|); j_{y,m} = \frac{1}{2}(|0\rangle^{(m)}\langle 1| + |1\rangle^{(m)}\langle 0|); j_{z,m} = \frac{1}{2i}(|0\rangle^{(m)}\langle 1| - |1\rangle^{(m)}\langle 0|)$$
$$j_{+,m} = j_{y,m} + ij_{z,m} = |0\rangle^{(m)}\langle 1|; j_{-,m} = j_{y,m} - ij_{z,m} = |1\rangle^{(m)}\langle 0|.$$

The operators $j_{i,m}$ $(i = x, y, z)$ obviously obey the standard angular momentum commutation relation $[j_{i,m}, j_{j,n}] = \sum_k i\epsilon_{ijk} j_{k,m}\delta_{m,n}$. From these we can define collective total angular momentum operators J_i acting on $\mathcal{E}_{\text{ensemble}}$: $J_i = \sum_m j_{i,m}$ for $i = x, y, z, \pm$. The collective operators J_i $(i = x, y, z)$, too, obey the standard commutation relation

$$[J_i, J_j] = \sum_{m,n}[j_{i,m}, j_{j,n}] = \sum_{m,n}\sum_k i\epsilon_{ijk} j_{k,m}\delta_{m,n} = \sum_k i\epsilon_{ijk} J_k.$$

2.2.3 Harmonic oscillator formalism

We will be interested in collective states where almost all of the atoms are in the $|0\rangle$ state. Using standard notation, the eigenstate of J_x (in $\mathcal{E}_{\text{ensemble}}$) in which all atoms are in the state $|0\rangle$ ("spin up") is designated $|J = \frac{N_\text{A}}{2}, M_x = \frac{N_\text{A}}{2}\rangle$, N_A being the number of atoms in the ensemble. Assuming that the system is close to this state at all times, the J_x operator can be approximated by its expectation value $J_x \approx \langle J_x\rangle$, which can be taken to be positive under the circumstances just described. This is effectively an approximation of the ensemble as a harmonic oscillator, since the following collective annihilation and creation operators

$$a_\text{A} \equiv \frac{1}{\sqrt{2\langle J_x\rangle}}\sum_m j_{+,m} = \frac{J_+}{\sqrt{2\langle J_x\rangle}},\ a_\text{A}^\dagger \equiv \frac{1}{\sqrt{2\langle J_x\rangle}}\sum_m j_{-,m} = \frac{J_-}{\sqrt{2\langle J_x\rangle}} \tag{18}$$

will now obey the harmonic oscillator commutation relation

$$[a_A, a_A^\dagger] = \frac{1}{2\langle J_x\rangle}\sum_{m,n}[j_{+,m}, j_{-,n}] = \frac{1}{2\langle J_x\rangle}\sum_{m,n} 2j_{x,m}\delta_{m,n} = \frac{1}{2\langle J_x\rangle}2J_x \approx 1,$$

where we have used $[j_{+,m}, j_{-,n}] = 2j_{x,m}\delta_{m,n}$ which follows from the angular momentum commutation relation of the j_i's.

This reformulation using harmonic oscillator formalism can be seen as arising from the Holstein-Primakoff transformation, which is briefly discussed in Appendix B.1. Note that the annihilation operator a_A consists of a sum of raising operators $j_{+,m}$ rather than lowering operators $j_{-,m}$. This stems from the fact that the Holstein-Primakoff transformation maps the fully polarized state $|J = \frac{N_\text{A}}{2}, M_x = \frac{N_\text{A}}{2}\rangle$ to the vacuum state of the harmonic oscillator; these states must vanish when acted upon by J_+ and a_A, respectively.

2.2.4 Continuous, position-dependent mode operators

Continuous, position-dependent annihilation and creation operators can now be introduced:

$$a_\text{A}(\vec{r}) = \frac{1}{\sqrt{2\langle j_x(\vec{r})\rangle}}\sum_m \delta^{(3)}(\vec{r} - \vec{r}_m) j_{+,m} = \frac{j_+(\vec{r})}{\sqrt{2\langle j_x(\vec{r})\rangle}}\ a_\text{A}^\dagger(\vec{r}) = \frac{1}{\sqrt{2\langle j_x(\vec{r})\rangle}}\sum_m \delta^{(3)}(\vec{r} - \vec{r}_m) j_{-,m} = \frac{j_-(\vec{r})}{\sqrt{2\langle j_x(\vec{r})\rangle}}, \tag{19}$$

where $\vec{r}_m$ is the position of the m'th atom and we have defined the continuous spin operators

$$j_i(\vec{r}) \equiv \sum_m \delta^{(3)}(\vec{r} - \vec{r}_m) j_{i,m} \quad (i = x, \pm).$$

We will take $\langle j_x(\vec{r})\rangle$ (assumed to be positive) to be the average over the random atomic positions $\{\vec{r}_m\}$ of the quantum mechanical expectation value of $j_x(\vec{r})$. The commutation relation for the continuous harmonic oscillator operators is seen to be

$$[a_\text{A}(\vec{r}), a_\text{A}^\dagger(\vec{r}')] = \frac{j_x(\vec{r})}{\langle j_x(\vec{r})\rangle}\delta^{(3)}(\vec{r} - \vec{r}') \approx \delta^{(3)}(\vec{r} - \vec{r}'),$$

using $[j_{+,m}, j_{-,n}] = 2j_{x,m}\delta_{m,n}$ and assuming small fluctuations in the spin density $j_x(\vec{r})$ compared to $\langle j_x(\vec{r})\rangle$.

In section 3.2, we will reexpress the total Hamiltonian in terms of the continuous, position-dependent atomic mode operators defined in this section. The equation of motion for $a_{\mathrm{A}}(\vec{r})$, as given by Heisenberg's equation, involves the commutator of $a_{\mathrm{A}}(\vec{r})$ and the Hamiltonian. Rather than working with a Hamiltonian expressed in terms of both $j_x(\vec{r})$ and $a_{\mathrm{A}}(\vec{r})$ in this connection, we will carry out the following replacement reminiscent of the (exact) Holstein-Primakoff expression $J_x = \frac{N_{\mathrm{A}}}{2} - a_{\mathrm{A}}^\dagger a_{\mathrm{A}}$ stated in eq. (82):

$$j_x(\vec{r}) \to \frac{n(\vec{r})}{2} - a_{\mathrm{A}}^\dagger(\vec{r}) a_{\mathrm{A}}(\vec{r}), \tag{20}$$

thereby following reference [5]. However, this replacement does not follow directly from the discrete operator expression $J_x = \frac{N_{\mathrm{A}}}{2} - a_{\mathrm{A}}^\dagger a_{\mathrm{A}}$; hence, eq. (20) should be considered a prescription for obtaining the correct Hamiltonian in terms of the continuous operator $a_{\mathrm{A}}(\vec{r})$. To make the shift to continuous operators $a_{\mathrm{A}}(\vec{r})$ in a more stringent way, we would have to derive the equations of motion for the discrete operators before introducing the continuous operators. One could also simply verify by inspection that $[a_{\mathrm{A}}(\vec{r}), j_x(\vec{r})] = [a_{\mathrm{A}}(\vec{r}), \frac{n(\vec{r})}{2} - a_{\mathrm{A}}^\dagger(\vec{r}) a_{\mathrm{A}}(\vec{r})]$ and note that the replacement under consideration preserves the vacuum expectation value of $j_x(\vec{r})$.

3 Light - atoms interaction

3.1 Interaction between light and a single atom

The Hamiltonian of the entire system consisting of the light fields and the atomic ensemble has the following general form $H = H_{\mathrm{A}} + H_{\mathrm{EM}} + H_{\mathrm{int}}$, where H_{int} accounts for the interaction between the light field and the atomic ensemble.

Like we did with the atomic Hamiltonian, we will decompose the interaction Hamiltonian H_{int} into a sum of Hamiltonians for the individual atoms, $H_{\mathrm{int}} = \sum_i H_{\mathrm{int}}^{(i)}$. For the remainder of this subsection we will restrict our attention to working out an effective expression for $H_A^{(i)} + H_{\mathrm{int}}^{(i)}$ (which acts on $\mathcal{E}_{\mathrm{EM}} \otimes \mathcal{E}_i$), that is, for the single atom case.

3.1.1 Electric dipole interaction

We outlined a procedure for quantizing the electromagnetic field in section 2.1.1 including the case of the field interacting with charged matter; but our focus thus far has solely been on the $H_{\mathrm{EM}}, H_{\mathrm{A}}$ parts of the Hamiltonian. In this section, we will be concerned with the interaction of the quantized EM field with the charged constituents of an atom (whose nucleus is assumed to be essentially motionless as compared to the dynamics of the electrons). The total Hamiltonian of the combined system will be subjected to a unitary transformation and expanded in terms of the various electric and magnetic multipole moments of the atom of which only the dominant term, the electric dipole contribution, will be retained. This is the *electric dipole approximation* which is commonly used in the domain of quantum optics. This subsection is based on [8] (chapter 4) and [9], see these for a detailed treatment.

The dipole approximation is basically to take the transverse electric field operator $\vec{E}_\perp$ to be spatially uniform over the volume occupied by the atom $\vec{E}_\perp(\vec{r}) \approx \vec{E}_\perp(\vec{r}_{\mathrm{A}})$, where $\vec{r}_{\mathrm{A}}$ is the position of the atom under consideration. That this is reasonable under the circumstances we are dealing with can be argued as follows: Considering typical atomic dimensions on the order of Ångströms (10^{-10}m) over which $|\vec{r}|$ varies and optical wavelengths λ in the neighborhood of 400 to 700 nm, the quantity $\vec{k} \cdot \vec{r} \leq \frac{2\pi}{\lambda}|\vec{r}|$, as would appear in a plane wave expansion of $\vec{E}_\perp(\vec{r}) \sim \sum_k e^{i\vec{k}\cdot\vec{r}}$, is certainly rather small.

Higher order terms account for the so-called electric quadrupole interaction and the magnetic dipole interaction, which are both smaller than the electric dipole contribution by about a factor of the fine structure constant $\alpha \approx \frac{1}{137}$ under the circumstances we are concerned with. These higher order terms, along with others, will be neglected.

The total Hamiltonian operator of the EM field/atom system in the Schrödinger picture is

$$H' = \frac{1}{2m}\sum_\alpha \left[\vec{p}_\alpha + \frac{e}{c}\vec{A}(\vec{r}_\alpha)\right]^2 + \frac{1}{2}\int d^3\vec{r}\sigma(\vec{r})\phi(\vec{r}) + \frac{1}{8\pi}\int d^3\vec{r}\left[\vec{E}_\perp(\vec{r})^2 + \vec{B}(\vec{r})^2\right], \tag{21}$$

where $\vec{r}_\alpha, \vec{p}_\alpha$ are position and momentum operators, respectively, for the α'th electron, $\sigma(\vec{r})$ is the charge density (including proton charges), and $\phi(\vec{r})$ is the non-retarded[12] Coulomb potential of the charges; the last term in

[12]The non-retarded part of $\vec{E}_\perp$ cancels the non-retarded part of $\vec{E}_\parallel = -\nabla\phi$, thereby preserving causality.

eq. (21) corresponds to $H_{\rm EM}$ in eq. (6), the transverse field Hamiltonian. In this form of the total Hamiltonian, the interaction terms are seen to be $\sum_\alpha \left(\frac{e}{mc}\vec{p}_\alpha \cdot \vec{A}(\vec{r}_\alpha) + \frac{e^2}{2mc^2}\vec{A}(\vec{r}_\alpha)^2\right)$.

We will now subject the total Hamiltonian in eq. (21) to the unitary transformation $U = \exp\left(-\frac{i}{\hbar}\int d^3\vec{r}\vec{P}(\vec{r})\cdot\vec{A}(\vec{r})\right)$, where $\vec{P}(\vec{r})$ is the atomic polarization, yielding transformed Hamiltonian and kets given by $H = UH'U^\dagger$ and $|\psi\rangle = U|\psi'\rangle$, respectively. The H so obtained is called the Power-Zienau-Woolley Hamiltonian in which the EM field is solely represented by the physical fields $\vec{E}$ and $\vec{B}$.

Transforming the first two terms of H' results in the atomic Hamiltonian $H_{\rm A} = \frac{1}{2m}\sum_\alpha \vec{p}_\alpha^2 + \frac{1}{2}\int d^3\vec{r}\sigma(\vec{r})\phi(\vec{r})$ as well as magnetic contributions, that we will neglect, from the $\vec{p}_\alpha\cdot\vec{A}(\vec{r}_\alpha), \vec{A}(\vec{r}_\alpha)^2$ terms. The electric dipole term appears when transforming $\vec{E}_\perp^2$ in the final term of H' as the lowest order contribution of a multipole expansion. New terms generated by $U\vec{B}U^\dagger$ are neglected. This leaves us with the free-field Hamiltonian $H_{\rm EM} = \frac{1}{8\pi}\int d^3\vec{r}\left[\vec{E}_\perp(\vec{r})^2 + \vec{B}(\vec{r})^2\right]$ (equivalent to eq. (13)) and the electric dipole interaction Hamiltonian

$$H_{\rm int} = -\vec{D}\cdot\vec{E}_\perp(\vec{r}_{\rm A}),$$

where $\vec{D} = -e\sum_\alpha \vec{x}_\alpha$ is the electric dipole moment operator of the atom and $\vec{r}_{\rm A}$ is its position.

The total transformed Hamiltonian for the single atom system in the dipole approximation is then $H = H_{\rm A} + H_{\rm EM} + H_{\rm int}$. Note that the forms of $H_{\rm A}$ and $H_{\rm EM}$ did not change, which tells us that we can still use the non-interacting atomic Hamiltonian $H_{\rm A}^{(i)}$ of section 2.2.1 and the light field Hamiltonian $H_{\rm EM}$ of section 2.1.3. Since we will ultimately be dealing with an ensemble of atoms, we will write the result of this section for the i'th atom as $H_{\rm int}^{(i)}(\vec{r}_i) = -\vec{D}^{(i)}\cdot\vec{E}_\perp(\vec{r}_i)$. Below we will simplify this single atom interaction Hamiltonian by making further approximations.

3.1.2 The rotating wave approximation

We will now express $H_{\rm int}^{(i)} = -\vec{D}^{(i)}\cdot\vec{E}_\perp(\vec{r}_i)$ in the basis of atomic states $\{|x\rangle^{(i)}\langle y|\}$ where $x, y \in \{0, 1, e\}$ (in the following we will suppress the $'(i)'$ labels on $\vec{D}^{(i)}$ and the $\vec{r}_i$ argument of the electric field). We assume that the states we use as our basis in the single atom Hilbert space are eigenstates of parity and, moreover, that the ground states $\{|0\rangle, |1\rangle\}$ have the same parity while the excited state $|e\rangle$ have the opposite parity. According to the Laporte selection rule then, the diagonal elements of the dipole operator vanish $\langle g|\vec{D}|g'\rangle = 0 = \langle e|\vec{D}|e\rangle$ for $g, g' \in \{0, 1\}$; this rule is proved in appendix B.2. Then defining so-called positive and negative frequency components $\vec{D}_{g,e}^{(+)} \equiv \langle g|\vec{D}|e\rangle$ and $\vec{D}_{e,g}^{(-)} \equiv (\vec{D}_{g,e}^{(+)})^* = \langle e|\vec{D}|g\rangle$, $g = 0, 1$, the dipole operator $\vec{D}$, being Hermitian, can be represented in the single atom Hilbert space as $\vec{D} = \sum_{g=0}^{1}(\vec{D}_{g,e}^{(+)}|g\rangle\langle e| + \vec{D}_{e,g}^{(-)}|e\rangle\langle g|)$.

The transverse electric field can be decomposed into positive and negative frequency components too, $\vec{E}_\perp = \vec{E}^{(+)} + \vec{E}^{(-)}$. In the case of the quantum signal part of the electric field, $\vec{E}_q^{(+)}$ is the sum of all terms containing annihilation operators and $\vec{E}_q^{(-)}$ is the sum of the terms containing creation operators.[13] Recall that in the steps that lead to our final expression for the quantum part of the light field, eq. (12), we split out the dominant part of the (Heisenberg picture) time dependence by introducing slowly varying annihilation and creation operators. For the purpose of the present argument, we will work only with this dominant part of the time dependence and therefore split out these dominant terms from the electric field operators $\vec{E}_q^{(+)} = \vec{E}_{0,q}^{(+)}\cdot e^{-i\omega_0 t}$ and $\vec{E}_q^{(-)} = \vec{E}_{0,q}^{(-)}\cdot e^{+i\omega_0 t}$, and similarly for the classical control field $\vec{E}_c^{(\pm)}(t) = \vec{E}_{0,c}^{(\pm)}\cdot e^{\mp i\omega_0' t}$; the slowly varying[14] operators $\vec{E}_{0,q}^{(\pm)}$ are defined by these equations.

With these definitions in place, the interaction Hamiltonian of a single atom at $\vec{r}_i$ can be stated as

$$H_{\rm int}^{(i)} = -\vec{D}\cdot(\vec{E}^{(+)} + \vec{E}^{(-)}) = -(\vec{D}_{0,e}^{(+)}|0\rangle\langle e| + \vec{D}_{e,0}^{(-)}|e\rangle\langle 0|)\cdot\left[\vec{E}_{0,q}^{(+)}\cdot e^{-i\omega_0 t} + \vec{E}_{0,q}^{(-)}\cdot e^{i\omega_0 t}\right] - (\vec{D}_{1,e}^{(+)}|1\rangle\langle e| + \vec{D}_{e,1}^{(-)}|e\rangle\langle 1|)\cdot\left[\vec{E}_{0,c}^{(+)}\cdot e^{-i\omega_0' t} + \vec{E}_{0,c}^{(-)}\cdot e^{i\omega_0' t}\right],$$

by making use of the property that the quantum field couples exclusively to the $|0\rangle \leftrightarrow |e\rangle$ transition, while the classical control field couples only to the $|1\rangle \leftrightarrow |e\rangle$ transition.

In section 2.2.1 we transformed the atomic Hamiltonian $H_{\rm A}$ to a rotating frame by means of a unitary operator $U = \prod_i \otimes U^{(i)}$, where $U^{(i)}$, given by eq. (16), acts exclusively on the Hilbert space of the i'th atom. As pointed

[13] Note that whereas $\vec{E}^{(\pm)}$ are vectors of operators, $\vec{D}_{i,j}^{(\pm)}$ are vectors of $\mathbb{C}$-numbers.

[14] In the sense that they are slowly varying when viewed in the Heisenberg picture.

out there, this transformation has to be applied consistently to all parts of the total Hamiltonian. Note however that the $i\hbar\dot{U}^\dagger U$ term of the transformation equation (15) was absorbed into the definition of the transformed atomic Hamiltonian $H'_{\rm A}$; hence, this term is accounted for, once and for all. The transformed single atom interaction Hamiltonian $H'^{(i)}_{\rm int}$ is then simply $H'^{(i)}_{\rm int} = U^{(i)\dagger} H^{(i)}_{\rm int} U^{(i)}$. To carry out this transformation we need to do the following operations (suppressing (i) most places)

$$U^{(i)\dagger}|g\rangle\langle e| = \exp\left(itE_g|g\rangle\langle g|\right)|g\rangle\langle e| = \exp\left(itE_g\right)|g\rangle\langle e| \quad (g=0,1)$$

where we used that $(|g\rangle\langle g|)^n = |g\rangle\langle g|$ for any integer $n \geq 1$, as is true of any projection operator. Similarly, we find that $U^{(i)\dagger}|e\rangle\langle g| = \exp(it(E_e - \Delta))|e\rangle\langle g|$ $(g = 0, 1)$ and related results for $U^{(i)}$ through Hermitian conjugation. Putting these together we get

$$\begin{aligned}
&U^{(i)\dagger} H^{(i)}_{\rm int} U^{(i)} = \\
&-\left(\vec{D}^{(+)}_{0,e} \exp\left(it[E_0 - (\omega_0 + E_0)]\right)|0\rangle\langle e| + \vec{D}^{(-)}_{e,0} \exp\left(it[(\omega_0 + E_0) - E_0]\right)|e\rangle\langle 0|\right) \cdot \left[\vec{E}^{(+)}_{0,q} \cdot e^{-i\omega_0 t} + \vec{E}^{(-)}_{0,q} \cdot e^{i\omega_0 t}\right] \\
&-\left(\vec{D}^{(+)}_{1,e} \exp\left(it[E_1 - (\omega'_0 + E_1)]\right)|1\rangle\langle e| + \vec{D}^{(-)}_{e,1} \exp\left(it[(\omega'_0 + E_1) - E_1]\right)|e\rangle\langle 1|\right) \cdot \left[\vec{E}^{(+)}_{0,c} \cdot e^{-i\omega'_0 t} + \vec{E}^{(-)}_{0,c} \cdot e^{i\omega'_0 t}\right]
\end{aligned}$$

$$\begin{aligned}
= &-\left[\vec{D}^{(+)}_{0,e} \cdot \vec{E}^{(-)}_{0,q} + \vec{D}^{(+)}_{0,e} \cdot \vec{E}^{(+)}_{0,q} \exp(-2i\omega_0 t)\right]|0\rangle\langle e| - \left[\vec{D}^{(-)}_{e,0} \cdot \vec{E}^{(+)}_{0,q} + \vec{D}^{(-)}_{e,0} \cdot \vec{E}^{(-)}_{0,q} \exp(2i\omega_0 t)\right]|e\rangle\langle 0| \\
&-\left[\vec{D}^{(+)}_{1,e} \cdot \vec{E}^{(-)}_{0,c} + \vec{D}^{(+)}_{1,e} \cdot \vec{E}^{(+)}_{0,c} \exp(-2i\omega'_0 t)\right]|1\rangle\langle e| - \left[\vec{D}^{(-)}_{e,1} \cdot \vec{E}^{(+)}_{0,c} + \vec{D}^{(-)}_{e,1} \cdot \vec{E}^{(-)}_{0,c} \exp(2i\omega'_0 t)\right]|e\rangle\langle 1|
\end{aligned}$$

Since ω_0, ω'_0 are optical frequencies, the $\exp(\pm 2i\omega_0 t), \exp(\pm 2i\omega'_0 t)$ terms vary rapidly compared to the time we allow the system to evolve (i.e. the duration of an experiment) and effectively average to zero; hence, we will neglect these terms. In this *Rotating Wave Approximation* the single atom interaction Hamiltonian reads

$$\begin{aligned}
H'^{(i)}_{\rm int} &= -\vec{D}^{(+)}_{0,e} \cdot \vec{E}^{(-)}_{0,q}|0\rangle\langle e| - \vec{D}^{(-)}_{e,0} \cdot \vec{E}^{(+)}_{0,q}|e\rangle\langle 0| - \vec{D}^{(+)}_{1,e} \cdot \vec{E}^{(-)}_{0,c}|1\rangle\langle e| - \vec{D}^{(-)}_{e,1} \cdot \vec{E}^{(+)}_{0,c}|e\rangle\langle 1| \\
&= -\sum_{g=0}^{1}\left(\vec{D}^{(+)}_{g,e} \cdot \vec{E}^{(-)}_{0,g}|g\rangle\langle e| + \vec{D}^{(-)}_{e,g} \cdot \vec{E}^{(+)}_{0,g}|e\rangle\langle g|\right), \qquad (22)
\end{aligned}$$

where $\vec{E}^{(\pm)}_{0,g=0} \equiv \vec{E}^{(\pm)}_{0,q}$ and $\vec{E}^{(\pm)}_{0,g=1} \equiv \vec{E}^{(\pm)}_{0,c}$.

3.1.3 Adiabatic elimination of the excited state

The result of the previous section, eq. (22), can be further simplified in a way that removes any explicit appearance of the excited state $|e\rangle^{(i)}$ in $H'^{(i)}_{\rm int}$ through a process called *adiabatic elimination*. But we will go further and use this approximation procedure to obtain an effective expression for the combination $H'^{(i)}_{\rm A} + H'^{(i)}_{\rm int}$, so that all occurrences of $|e\rangle^{(i)}$ are eliminated from our equations. In this thesis, we will settle for a somewhat crude adiabatic elimination procedure; but in reference [2] it is shown how the result that we obtain arises rigorously from a series expansion of the exact solution (in the case of classical, external light fields).

We consider the Heisenberg equation of motion in the rotating frame (see appendix A) for the operator $|g\rangle^{(i)}\langle e|$ (for $g = 0, 1$): $i\frac{d}{dt}[|g\rangle^{(i)}\langle e|_{\rm I,H}] = [|g\rangle^{(i)}\langle e|, H']_{\rm I,H}$, where $H' = H'_{\rm A} + H_{\rm EM} + H'_{\rm int}$, but obviously $[|g\rangle^{(i)}\langle e|, H_{\rm EM}] = 0$ and $[|g\rangle^{(i)}\langle e|, H'^{(j)}_{\rm int}] = 0 = [|g\rangle^{(i)}\langle e|, H'^{(j)}_{\rm A}]$ for $i \neq j$ because these operators do not act on the Hilbert space $\mathcal{E}_i$ of the i'th atom. Hence, $[|g\rangle^{(i)}\langle e|, H'] = [|g\rangle^{(i)}\langle e|, H'^{(i)}_{\rm A} + H'^{(i)}_{\rm int}]$, where $H'^{(i)}_{\rm A} + H'^{(i)}_{\rm int}$ is given by eqs. (17) and (22). We find that

$$i\frac{d}{dt}[|g\rangle^{(i)}\langle e|_{\rm I,H}] = \left[\Delta|g\rangle^{(i)}\langle e| - \sum_{g'=0}^{1}\left(\vec{D}^{(-)}_{e,g'} \cdot \vec{E}^{(+)}_{0,g'}|g\rangle^{(i)}\langle g'|\right) + \vec{D}^{(-)}_{e,g} \cdot \vec{E}^{(+)}_{0,g}|e\rangle^{(i)}\langle e|\right]_{\rm I,H}.$$

Now assuming $\vec{D}^{(-)}_{e,g} \cdot \vec{E}^{(+)}_{0,g} \ll \Delta$, weak excitation $\vec{D}^{(-)}_{e,g} \cdot \vec{E}^{(+)}_{0,g}|e\rangle^{(i)}\langle e| \approx 0$ and slow dynamics $\frac{d}{dt}[|g\rangle^{(i)}\langle e|_{\rm I,H}] \approx 0$ as compared to the detuning Δ, we obtain for the Schrödinger picture operators:

$$0 \approx \Delta|g\rangle^{(i)}\langle e| - \sum_{g'=0}^{1}\left(\vec{D}^{(-)}_{e,g'} \cdot \vec{E}^{(+)}_{0,g'}|g\rangle^{(i)}\langle g'|\right) \Leftrightarrow |g\rangle^{(i)}\langle e| \approx \frac{1}{\Delta}\sum_{g'=0}^{1}\left(\vec{D}^{(-)}_{e,g'} \cdot \vec{E}^{(+)}_{0,g'}|g\rangle^{(i)}\langle g'|\right).$$

It is this approximative expression for $|g\rangle^{(i)}\langle e|$ $(g = 0, 1)$ that allows us to eliminate the excited state from $H'^{(i)}_{\mathrm{A}} + H'^{(i)}_{\mathrm{int}}$; either by directly replacing instances of $|g\rangle^{(i)}\langle e|$ by the approximate expression, or indirectly through the identity $|e\rangle^{(i)}\langle e| = (|e\rangle^{(i)}\langle g|) \cdot (|g\rangle^{(i)}\langle e|)$. In this way we find that

$$H'^{(i)}_{\mathrm{A}} = \Delta|e\rangle^{(i)}\langle e| \approx \frac{1}{\Delta}\sum_{g,g'} \vec{D}^{(+)}_{g,e} \cdot \vec{E}^{(-)}_{0,g}|g\rangle^{(i)}\langle g'|\vec{D}^{(-)}_{e,g'} \cdot \vec{E}^{(+)}_{0,g'}$$

and from eq. (22)

$$H'^{(i)}_{\mathrm{int}} \approx -\frac{1}{\Delta}\sum_{g,g'} \vec{E}^{(-)}_{0,g} \cdot \vec{D}^{(+)}_{g,e}|g\rangle^{(i)}\langle g'|\vec{E}^{(+)}_{0,g'} \cdot \vec{D}^{(-)}_{e,g'} + \mathrm{H.C.} = -\frac{2}{\Delta}\sum_{g,g'} \vec{E}^{(-)}_{0,g} \cdot \vec{D}^{(+)}_{g,e}|g\rangle^{(i)}\langle g'|\vec{E}^{(+)}_{0,g'} \cdot \vec{D}^{(-)}_{e,g'}, \tag{23}$$

using normal ordering of the operators $\vec{E}^{(\pm)}_{0,q}$, positioning $\vec{E}^{(-)}_{0,q}$ (which contains creation operators) to the left of $\vec{E}^{(+)}_{0,q}$ (which contains annihilation operators). Adding these equations yields an effective expression for $H'^{(i)}_{\mathrm{A}} + H'^{(i)}_{\mathrm{int}}$ which can be written as

$$H^{(i)}_{\mathrm{Ai}}(\vec{r}_i) \equiv \sum_{g,g'} V_{g,g'}(\vec{r}_i)|g\rangle^{(i)}\langle g'|, \text{ with } V_{g,g'}(\vec{r}) = -\frac{1}{\Delta}(\vec{D}^{(+)}_{g,e} \cdot \vec{E}^{(-)}_{0,g}(\vec{r}))(\vec{D}^{(-)}_{e,g'} \cdot \vec{E}^{(+)}_{0,g'}(\vec{r})), \tag{24}$$

where the $\vec{D}^{(\pm)}_{m,n}$ correspond to the dipole operator of the i'th atom. Note that the $V_{g,g'}(\vec{r})$ are operators acting on the light field Fock space $\mathcal{E}_{\mathrm{EM}}$ and that $H^{(i)}_{\mathrm{Ai}}$ is in the rotating frame even though we have dropped the prime.

3.2 Interaction between light and the atomic ensemble

To obtain the effective interaction and atomic Hamiltonian of the ensemble, we simply sum the single atom Hamiltonians $H^{(i)}_{\mathrm{Ai}}(\vec{r}_i)$: $H_{\mathrm{Ai}}(\{\vec{r}_i\}_{i\in\{1,\ldots,N_{\mathrm{A}}\}}) \equiv \sum_i H^{(i)}_{\mathrm{Ai}}(\vec{r}_i)$. Hence, we now have an effective total Hamiltonian in the rotating frame $H' = H_{\mathrm{Ai}} + H_{\mathrm{EM}}$. The next steps are: 1) to reexpress the Hamiltonian in terms of light mode annihilation and creation operators and the continuous atomic mode operators introduced in subsection 2.2.4; 2) to reduce the Hamiltonian from 3-D, where $\vec{r}$ enters, to 1-D, where only the z-coordinate enters, while the transverse coordinate integrals over $\vec{r}_\perp$ have been carried out (at least formally).

3.2.1 Effective Hamiltonian in terms of continuous mode operators

In this section, we will be concerned with expressing the effective atomic ensemble and interaction Hamiltonian H_{Ai} in terms of the continuous mode operators introduced in section 2.2.4. The key to doing this is to introduce the density operator $n(\vec{r}) = \sum_i \delta^{(3)}(\vec{r} - \vec{r}_i)$ as follows

$$H_{\mathrm{Ai}}(\{\vec{r}_i\}_{i\in\{1,\ldots,N_{\mathrm{A}}\}}) = \sum_i H^{(i)}_{\mathrm{Ai}}(\vec{r}_i) = \int d^3\vec{r}\sum_i \delta^{(3)}(\vec{r} - \vec{r}_i)H^{(i)}_{\mathrm{Ai}}(\vec{r}), \tag{25}$$

where we are integrating over the volume of the cylinder. Plugging eq. (24) into eq. (25), a number of terms appear, one of which is (the V_{ij}'s are understood to depend on the integration variable $\vec{r}$)

$$\int d^3\vec{r}\sum_i \delta^{(3)}(\vec{r} - \vec{r}_i)V_{01}|0\rangle^{(i)}\langle 1| = \int d^3\vec{r}\sum_i \delta^{(3)}(\vec{r} - \vec{r}_i)V_{01}j_{+,i} = \int d^3\vec{r}V_{01}j_+(\vec{r}), \tag{26}$$

now using the relation between $j_+(\vec{r})$ and $a_{\mathrm{A}}(\vec{r})$ eq. (19) we get

$$= \int d^3\vec{r}V_{01}\sqrt{2\langle j_x(\vec{r})\rangle}a_{\mathrm{A}}(\vec{r}) \approx \int d^3\vec{r}V_{01}\sqrt{n(\vec{r})}a_{\mathrm{A}}(\vec{r}), \tag{27}$$

where we have used the approximation that the atomic ensemble is close to the fully polarized state $\langle j_x(\vec{r})\rangle \approx \frac{n(\vec{r})}{2}$. Similarly, the Hermitian conjugate of eq. (26) appears in eq. (25); this term can be written as $\int d^3\vec{r}V_{10}\sqrt{n(\vec{r})}a^\dagger_{\mathrm{A}}(\vec{r})$.

The remaining terms are

$$\int d^3\vec{r}\sum_i \delta^{(3)}(\vec{r} - \vec{r}_i)(V_{00}|0\rangle^{(i)}\langle 0| + V_{11}|1\rangle^{(i)}\langle 1|) = \int d^3\vec{r}((V_{00} - V_{11})j_x(\vec{r}) + \frac{n(\vec{r})}{2}(V_{00} + V_{11}), \tag{28}$$

where we used the definition of $j_x(\vec{r})$ and the fact that $\sum_i \left(|0\rangle^{(i)}\langle 0| + |1\rangle^{(i)}\langle 1|\right) = 1$ is a resolution of the identity on the effective atomic Hilbert space $\mathcal{E}_{\text{ensemble}}$ (having adiabatically eliminated the excited states $|e\rangle^{(i)}$). Now using the prescription eq. (20) for replacing $j_x(\vec{r})$, eq. (28) becomes

$$= \int d^3\vec{r}\left(n(\vec{r})V_{00} + (V_{11} - V_{00})a_{\rm A}^\dagger(\vec{r})a_{\rm A}(\vec{r})\right). \tag{29}$$

The effective interaction plus atomic ensemble Hamiltonian expressed in terms of continuous collective mode operators is then the sum of eq. (27), its Hermitian conjugate and eq. (29):

$$H_{\rm Ai} = \int d^3\vec{r}\left[(\sqrt{n(\vec{r})}a_A(\vec{r})V_{01} + \text{H.C.}) + n(\vec{r})V_{00} + a_A^\dagger(\vec{r})a_A(\vec{r})(V_{11} - V_{00})\right], \tag{30}$$

where the V_{ij}'s contain the light field operators. We will neglect the $a_A^\dagger(\vec{r})a_A(\vec{r})V_{00}$ term which represents the contribution to the AC Stark shift from the quantum field, which is small compared to that of the classical control field.

Next, we evaluate the V_{ij}'s in accordance with the beam splitter light field setup described in section 2.1.2. Starting with V_{00}, we have from eq. (24) that

$$V_{00}(\vec{r}) = -\frac{1}{\Delta}(\vec{D}_{0,e}^{(+)} \cdot \vec{E}_{0,q}^{(-)}(\vec{r}))(\vec{D}_{e,0}^{(-)} \cdot \vec{E}_{0,q}^{(+)}(\vec{r})); \tag{31}$$

here, the quantum signal part of the light field alone enters and we use expression (12) to plug in for $\vec{E}_{0,q}^{(\pm)}$ in eq. (31), thereby achieving

$$V_{00}(\vec{r}) = -\frac{1}{\Delta}\frac{2\pi\omega_0}{c}|D_0|^2 \sum_{m,n} \zeta_{m,n}(\vec{r}_\perp, z)a_{{\rm L},m}^\dagger(z,t)a_{{\rm L},n}(z,t),$$

where we have defined $D_0 \equiv \vec{D}_{e,0}^{(-)} \cdot \vec{\epsilon}_q$ and $\zeta_{m,n}(\vec{r}_\perp, z) \equiv u_m^*(\vec{r}_\perp, z)u_n(\vec{r}_\perp, z)$. Then, by further defining the coupling function $g(\vec{r}) = \sqrt{\frac{2\pi\omega_0 n(\vec{r})}{c}}D_0$, the term $n(\vec{r})V_{00}$ in eq. (30) can be written

$$n(\vec{r})V_{00} = -\frac{|g(\vec{r})|^2}{\Delta}\sum_{m,n}\zeta_{m,n}(\vec{r}_\perp, z)a_{{\rm L},m}^\dagger(z)a_{{\rm L},n}(z).$$

Next, we turn to V_{11}; in parallel to eq. (31), we have

$$V_{11}(\vec{r}) = -\frac{1}{\Delta}(\vec{D}_{1,e}^{(+)} \cdot \vec{E}_{0,c}^{(-)}(\vec{r}))(\vec{D}_{e,1}^{(-)} \cdot \vec{E}_{0,c}^{(+)}(\vec{r})), \tag{32}$$

but this time it is the $|1\rangle \leftrightarrow |e\rangle$ transition that enters, which the classical control field couples to exclusively in the beam splitter setup. Defining the slowly varying *Rabi frequency* $\Omega(\vec{r}, t) = 2\vec{D}_{e,1}^{(-)} \cdot \langle\vec{E}_{0,c}^{(+)}(\vec{r})\rangle e^{-ik_0' z}$ (where $\langle\vec{E}_{0,c}^{(+)}(\vec{r})\rangle$ is the expectation value w.r.t. a coherent state) and noting that $\left(\vec{D}_{e,1}^{(-)} \cdot \vec{E}_{0,c}^{(+)}(\vec{r})\right)^\dagger = \vec{D}_{1,e}^{(+)} \cdot \vec{E}_{0,c}^{(-)}(\vec{r})$, we see that eq. (32) becomes

$$V_{11}(\vec{r}) = -\frac{|\Omega(\vec{r}, t)|^2}{4\Delta}.$$

Finally, for V_{01} (note that $V_{10} = V_{01}^\dagger$) we have from eq. (24)

$$V_{01}(\vec{r}) = -\frac{1}{\Delta}(\vec{D}_{0,e}^{(+)} \cdot \vec{E}_{0,q}^{(-)}(\vec{r}))(\vec{D}_{e,1}^{(-)} \cdot \vec{E}_{0,c}^{(+)}(\vec{r}));$$

here, both transitions enter the equation and each part of the field is treated as above. Hence,

$$\begin{aligned} V_{01}(\vec{r}) &= -\frac{1}{\Delta}(\sqrt{\frac{2\pi\omega_0}{c}}D_0^* \sum_m u_m^*(\vec{r}_\perp, z)a_{{\rm L},m}^\dagger(z,t)e^{-ik_0 z})(\vec{D}_{e,1}^{(-)} \cdot \langle\vec{E}_{0,c}^{(+)}(\vec{r})\rangle) \\ &= -\frac{1}{2\Delta}(\sqrt{\frac{2\pi\omega_0}{c}}D_0^* e^{i(k_0' - k_0)z} \sum_m u_m^*(\vec{r}_\perp, z)a_{{\rm L},m}^\dagger(z,t))(2\vec{D}_{e,1}^{(-)} \cdot \langle\vec{E}_{0,c}^{(+)}(\vec{r})\rangle e^{-ik_0' z}), \end{aligned}$$

so that the term $\sqrt{n(\vec{r})}a_A(\vec{r})V_{01}$ in eq. (30) becomes

$$\sqrt{n(\vec{r})}a_{\rm A}(\vec{r})V_{01} = -\frac{g^*(\vec{r})\Omega(\vec{r}, t)}{2\Delta}e^{i(k_0' - k_0)z}(\sum_m u_m^*(\vec{r}_\perp, z)a_{{\rm L},m}^\dagger(z))a_{\rm A}(\vec{r}).$$

The mode independent phase factor $e^{i(k_0-k_0')z}$ is now absorbed into the mode set $\{u_m(\vec{r}_\perp,z)\}$ by defining $u'_m(\vec{r}_\perp,z)\equiv u_m(\vec{r}_\perp,z)e^{i(k_0-k_0')z}$; note that $\zeta'_{m,n}(\vec{r}_\perp,z)\equiv u'^*_m(\vec{r}_\perp,z)u'_n(\vec{r}_\perp,z)=\zeta_{m,n}(\vec{r}_\perp,z)$. We drop the prime on $u'_m(\vec{r}_\perp,z)$ henceforth.

One final manipulation of eq. (30) remains: We expand all occurrences of $a_{\rm A}(\vec{r},t)$ in terms of the set $\{u_m(\vec{r}_\perp,z)\}$ as $a_{\rm A}(\vec{r},t)=\sum_m u_m(\vec{r}_\perp,z)a_{{\rm A},m}(z,t)$ as described in section 2.1.2; the operators $a_{{\rm A},m}(z,t)$ are easily shown to have the equal time commutation relations $[a_{{\rm A},m}(z),a^\dagger_{{\rm A},m'}(z')]=\delta_{m,m'}\delta(z-z')$ and $[a_{{\rm A},m}(z),a_{{\rm A},m'}(z')]=0$. We then arrive at the following restatement of eq. (30)

$$H_{\rm Ai}=\int d^3\vec{r}\left[-\frac{|\Omega(\vec{r},t)|^2}{4\Delta}\sum_{m,n}\zeta_{m,n}(\vec{r}_\perp,z)a^\dagger_{{\rm A},m}(z,t)a_{{\rm A},n}(z,t)-\frac{|g(\vec{r})|^2}{\Delta}\sum_{m,n}\zeta_{m,n}(\vec{r}_\perp,z)a^\dagger_{{\rm L},m}(z,t)a_{{\rm L},n}(z,t)\right.$$
$$\left.-\left(\frac{g^*(\vec{r})\Omega(\vec{r},t)}{2\Delta}\sum_{m,n}\zeta_{m,n}(\vec{r}_\perp,z)a^\dagger_{{\rm L},m}(z,t)a_{{\rm A},n}(z,t)+\text{H.C.}\right)\right], \tag{33}$$

where, as above, occurrences of the mode functions $u_n(\vec{r}_\perp,z)$ have been absorbed into $\zeta_{m,n}(\vec{r}_\perp,z)\equiv u^*_m(\vec{r}_\perp,z)u_n(\vec{r}_\perp,z)$.

3.2.2 Simplifying assumptions and reduction of the Hamiltonian to one dimension

In the treatments mentioned in the introduction [3, 5], the next step is to make assumptions about the atomic number density $n(\vec{r})$ ($=n(z)$ in the former, $=n$ in the latter) in order to be able to carry out the integral over the perpendicular plane in eq. (33) by exploiting the orthonormality property of the set $\{u_n(\vec{r}_\perp,z)\}$.

In the present treatment, we will use a somewhat more realistic assumption about the number density $n(\vec{r})$, namely, a cylindrically symmetric Gaussian distribution $n(\vec{r})=n(\rho)=n_0e^{-\frac{\rho^2}{2\sigma_\perp^2}}$, the symmetry axis being the z-axis so that $\rho=|\vec{r}_\perp|$. The quantity $\sigma_\perp$ is the standard deviation of the atomic distribution in the perpendicular plane. $n_0=n(\rho=0)$ is determined through the following normalization, where $N_{\rm A}$ is the total number of atoms and the integrals are over the entire cylindrical volume (of length L and infinite radius):

$$N_{\rm A}=\int n(\vec{r})dV=\int n(\rho)\rho d\theta d\rho dz=2\pi L\int_0^\infty n(\rho)\rho d\rho$$
$$=2\pi L\int_0^\infty n_0e^{-\frac{\rho^2}{2\sigma_\perp^2}}\rho d\rho=-2\pi Ln_0\sigma_\perp^2\left[e^{-\rho^2/2\sigma_\perp^2}\right]_0^\infty=2\pi Ln_0\sigma_\perp^2,$$

through integration by substitution; thus, $n_0=\frac{N_{\rm A}}{2\pi L\sigma_\perp^2}$. Then $g(\vec{r})$, defined above, simplifies to $g(\rho)=\sqrt{\frac{2\pi\omega_0}{c}n(\rho)}D_0=g_0e^{-\frac{\rho^2}{4\sigma_\perp^2}}$, where we have defined $g_0\equiv\sqrt{\frac{2\pi\omega_0}{c}n_0}D_0=\sqrt{\frac{\omega_0N_{\rm A}}{cL\sigma_\perp^2}}D_0$.

Furthermore, we will assume the "slowly varying" Rabi frequency to be a constant $\Omega(\vec{r},t)=\Omega$, corresponding to an essentially monochromatic control field. The effective interaction plus atomic ensemble Hamiltonian can now be written as

$$H_{\rm Ai}=\int dz\left[-\frac{|\Omega|^2}{4\Delta}\sum_{m,n}\left[\int d^2\vec{r}_\perp\zeta_{m,n}(\vec{r}_\perp,z)\right]a^\dagger_{{\rm A},m}(z,t)a_{{\rm A},n}(z,t)\right.$$
$$-\frac{|g_0|^2}{\Delta}\sum_{m,n}\left[\int d^2\vec{r}_\perp\zeta_{m,n}(\vec{r}_\perp,z)e^{-\frac{\rho^2}{2\sigma_\perp^2}}\right]a^\dagger_{{\rm L},m}(z,t)a_{{\rm L},n}(z,t) \tag{34}$$
$$\left.-\left(\frac{g_0^*\Omega}{2\Delta}\sum_{m,n}\left[\int d^2\vec{r}_\perp\zeta_{m,n}(\vec{r}_\perp,z)e^{-\frac{\rho^2}{4\sigma_\perp^2}}\right]a^\dagger_{{\rm L},m}(z,t)a_{{\rm A},n}(z,t)+\text{H.C.}\right)\right],$$

where we have factored out the part of the integral which is not over the perpendicular coordinates. The first of the transverse integrals $\int d^2\vec{r}_\perp\zeta_{m,n}(\vec{r}_\perp,z)$ can be carried out immediately due to the orthogonality property of $\{u_m(\vec{r}_\perp,z)\}$, eq.(8). Then, stated in different notation, eq. (34) becomes

$$H_{\rm Ai}=\int dz\left[-\frac{|\Omega|^2}{4\Delta}\sum_n a^\dagger_{{\rm A},n}(z,t)a_{{\rm A},n}(z,t)-\frac{|g_0|^2}{\Delta}\sum_{m,n}\beta_{m,n}(z)a^\dagger_{{\rm L},m}(z,t)a_{{\rm L},n}(z,t)\right.$$
$$\left.-\left(\frac{g_0^*\Omega}{2\Delta}\sum_{m,n}\gamma_{m,n}(z)a^\dagger_{{\rm L},m}(z,t)a_{{\rm A},n}(z,t)+\text{H.C.}\right)\right] \tag{35}$$

where we have introduced the coupling functions

$$\beta_{m,n}(z) = \int d^2\vec{r}_\perp \zeta_{m,n}(\vec{r}_\perp, z) e^{-\frac{\rho^2}{2\sigma_\perp^2}}, \quad \gamma_{m,n}(z) = \int d^2\vec{r}_\perp \zeta_{m,n}(\vec{r}_\perp, z) e^{-\frac{\rho^2}{4\sigma_\perp^2}},$$

that govern the coupling between modes; these functions cannot be evaluated before a set of mode functions $\{u_n(\vec{r}_\perp, z)\}$ has been chosen. We see that with eq. (35) we have in fact obtained a one dimensional effective Hamiltonian describing the atomic ensemble and its interaction with the light field. Note that the Hermitian conjugate of $\sum_{m,n} \gamma_{m,n}(z) a^\dagger_{\mathrm{L},m}(z,t) a_{\mathrm{A},n}(z,t)$, which enters eq. (35), is (suppressing z and t dependencies)

$$\left[\sum_{m,n} \gamma_{m,n} a^\dagger_{\mathrm{L},m} a_{\mathrm{A},n}\right]^\dagger = \sum_{m,n} \gamma^*_{m,n} a^\dagger_{\mathrm{A},n} a_{\mathrm{L},m} = \sum_{m,n} \gamma_{n,m} a^\dagger_{\mathrm{A},n} a_{\mathrm{L},m} = \sum_{m,n} \gamma_{m,n} a^\dagger_{\mathrm{A},m} a_{\mathrm{L},n},$$

since the Hermiticity of $\underline{\underline{\gamma}}$ is equivalent to $\underline{\underline{\gamma}}^* = \underline{\underline{\gamma}}^t$ or, in component form, $\gamma^*_{m,n} = \gamma_{n,m}$.

4 Equations of motion

4.1 Determining the equations of motion

In this section we wish to find differential equations governing the evolution of the collective atomic mode operators $a_{\mathrm{A},m}(z,t)$ and the light mode operators $a_{\mathrm{L},m}(z,t)$ in the Heisenberg rotating frame.

Heisenberg's equation of motion for an operator in the rotating frame is derived in Appendix A. It reads

$$\frac{dA_{\mathrm{I,H}}}{dt} = \left(i\left[H_\mathrm{I}, A_\mathrm{S}\right] + \frac{\partial A_\mathrm{S}}{\partial t}\right)_{\mathrm{I,H}},$$

where subscript 'I,H' denotes conversion from the Schrödinger picture to the Heisenberg rotating frame, and $H_\mathrm{I} = i\dot{U}^\dagger U + U^\dagger H_\mathrm{S} U$ is the rotating frame Hamiltonian obtained in the course of the previous sections. We found $H_\mathrm{I} = H_\mathrm{Ai} + H_\mathrm{EM}$, where the constituents are given by eqs. (35) and (13), respectively.

In the following we are dealing with Schrödinger picture operators unless otherwise stated. We begin with setting up the equation of motion for $a_{\mathrm{L},\tilde{m}}(z,t)_{\mathrm{I,H}}$. Since $\frac{\partial a_{\mathrm{L},\tilde{m}}(z,t)}{\partial t} = i\omega_0 a_{\mathrm{L},\tilde{m}}(z,t)$, as is seen from eq. (11), the equation of motion for $a_{\mathrm{L},\tilde{m}}(z,t)_{\mathrm{I,H}}$ is given by

$$\frac{d}{dt} a_{\mathrm{L},\tilde{m}}(z,t)_{\mathrm{I,H}} = \left(-i[a_{\mathrm{L},\tilde{m}}(z,t), H_\mathrm{Ai} + H_\mathrm{EM}] + i\omega_0 a_{\mathrm{L},\tilde{m}}(z,t)\right)_{\mathrm{I,H}}. \tag{36}$$

To start out with, we determine $[a_{\mathrm{L},\tilde{m}}(z,t), H_\mathrm{EM}]$ using the relation between the Schrödinger picture operators $a_{\mathrm{L},\tilde{m}}$ and $a_{\mathrm{L},\tilde{m}k}$ in eq. (11), the commutator identity $[A, BC] = [A,B]C + B[A,C]$, $[a_{\mathrm{L},\tilde{m}k}, a^\dagger_{\mathrm{L},mk'}] = \delta_{\tilde{m},m}\delta_{k,k'}$, and $[a_{\mathrm{L},\tilde{m}k}, a_{\mathrm{L},mk'}] = 0$:

$$[a_{\mathrm{L},\tilde{m}}(z,t), H_\mathrm{EM}] = \sum_k \sqrt{\frac{c}{l}} e^{i(k-k_0)z + i\omega_0 t} kc a_{\mathrm{L},\tilde{m}k} = c\left(\frac{1}{i}\frac{\partial a_{\mathrm{L},\tilde{m}}(z,t)}{\partial z} + k_0 a_{\mathrm{L},\tilde{m}}(z,t)\right),$$

plugging this into the expression for the equation of motion eq. (36) and using $k_0 c = \omega_0$ we get

$$\frac{d}{dt} a_{\mathrm{L},\tilde{m}}(z,t)_{\mathrm{I,H}} = \left(-c\frac{\partial}{\partial z} a_{\mathrm{L},\tilde{m}}(z,t) - i[a_{\mathrm{L},\tilde{m}}(z,t), H_\mathrm{Ai}]\right)_{\mathrm{I,H}}. \tag{37}$$

We now introduce a rescaled time $\tau = t - \frac{z}{c}$ and define new rotating frame annihilation operators that depend on this variable $a'_{\mathrm{X},\tilde{m}}(z,\tau) \equiv a_{\mathrm{X},\tilde{m}}(z, t = \tau + \frac{z}{c})$, where X=L,A. It is shown in Appendix B.3 that the rescaled time equivalent of eq. (37) is

$$\frac{\partial}{\partial z} a'_{\mathrm{L},\tilde{m}}(z,\tau)_{\mathrm{I,H}} \quad = \quad \left(-\frac{i}{c}[a'_{\mathrm{L},\tilde{m}}(z,\tau), H_\mathrm{Ai}]\right)_{\mathrm{I,H}}, \tag{38}$$

which is now a differential equation in z; whereas for $a'_{\mathrm{A},\tilde{m}}(z,\tau)_{\mathrm{I,H}}$, we have

$$\frac{\partial}{\partial \tau} a'_{\mathrm{A},\tilde{m}}(z,\tau)_{\mathrm{I,H}} = \left.\frac{\partial a_{\mathrm{A},\tilde{m}}(z,t)_{\mathrm{I,H}}}{\partial t}\right|_{t=\tau+\frac{z}{c}}. \tag{39}$$

Taking note of eqs. (38) and (39), we will drop the prime on $a'_{\mathrm{X},\tilde{m}}$ and simply denote the rescaled time by t in all that follows.

Evaluating the commutator in eq. (38) by using eq. (35) and the various (equal time) commutation relations involving $a_{\mathrm{L},m}(z,t)$, we achieve the final expression for the equations of motion for the light modes

$$\begin{aligned}
&\frac{\partial}{\partial z}a_{\mathrm{L},\tilde{m}}(z,t)_{\mathrm{I,H}} \\
&= \frac{i}{c}\int dz' \left(\frac{|g_0|^2}{\Delta}\sum_{m,n}\beta_{m,n}(z')c\delta_{\tilde{m},m}\delta(z-z')a_{\mathrm{L},n}(z',t) + \frac{g_0^*\Omega}{2\Delta}\sum_{m,n}\gamma_{m,n}(z')c\delta_{\tilde{m},m}\delta^{(3)}(z-z')a_{\mathrm{A},n}(z',t)\right)_{\mathrm{I,H}} \\
&= i\left(\frac{|g_0|^2}{\Delta}\sum_{n}\beta_{\tilde{m},n}(z)a_{\mathrm{L},n}(z,t) + \frac{g_0^*\Omega}{2\Delta}\sum_{n}\gamma_{\tilde{m},n}(z)a_{\mathrm{A},n}(z,t)\right)_{\mathrm{I,H}}.
\end{aligned}$$

Next, we wish to find the equation of motion for the atomic modes $a_{\mathrm{A},\tilde{m}}(z,t)$; in this case we have no explicit time dependence contributing to the Heisenberg equation of motion (also, recall that H_{A} has been absorbed into H_{Ai}):

$$\begin{aligned}
&\frac{\partial}{\partial t}a_{\mathrm{A},\tilde{m}}(z,t)_{\mathrm{I,H}} = (-i[a_{\mathrm{A},\tilde{m}}(z,t),H_{\mathrm{Ai}}])_{\mathrm{I,H}} \\
&= -i\int dz' \left(-\frac{|\Omega|^2}{4\Delta}\sum_{n}\delta_{\tilde{m},n}\delta(z-z')a_{\mathrm{A},n}(z') - \frac{g_0\Omega^*}{2\Delta}\sum_{m,n}\gamma_{m,n}(z')\delta_{\tilde{m},m}\delta(z-z')a_{\mathrm{L},n}(z')\right)_{\mathrm{I,H}} \\
&= i\left(\frac{|\Omega|^2}{4\Delta}a_{\mathrm{A},\tilde{m}}(z) + \frac{g_0\Omega^*}{2\Delta}\sum_{n}\gamma_{\tilde{m},n}(z)a_{\mathrm{L},n}(z)\right)_{\mathrm{I,H}}.
\end{aligned}$$

At this point we can drop the 'I,H' subscript as the coupled differential equations for $a_{\mathrm{X},\tilde{m}}(z,t)_{\mathrm{I,H}}$ (X=L,A) arrived at above are expressed entirely in terms of Heisenberg rotating frame operators. In summary, then,

$$\frac{\partial}{\partial z}a_{\mathrm{L},\tilde{m}}(z,t) = \frac{i}{\Delta}|g_0|^2\sum_{n}\beta_{\tilde{m},n}(z)a_{\mathrm{L},n}(z,t) + \frac{i}{2\Delta}g_0^*\Omega\sum_{n}\gamma_{\tilde{m},n}(z)a_{\mathrm{A},n}(z,t) \tag{40}$$

$$\frac{\partial}{\partial t}a_{\mathrm{A},\tilde{m}}(z,t) = \frac{i}{4\Delta}|\Omega|^2 a_{\mathrm{A},\tilde{m}}(z,t) + \frac{i}{2\Delta}g_0\Omega^*\sum_{n}\gamma_{\tilde{m},n}(z)a_{\mathrm{L},n}(z,t). \tag{41}$$

4.2 Spontaneous emission from the excited state

In the case of beam splitter interaction, which is what we are concerned with, spontaneous emission from the excited state $|e\rangle$ with decay rate γ can be included in the equations of motion by making the replacement $\Delta \to \Delta - i\frac{\gamma}{2}$. The details behind this are rather involved and we will refer to Appendix D of reference [5] for a treatment of these aspects. Having made this replacement, the equations of motion (40) and (41) read

$$\frac{\partial}{\partial z}a_{\mathrm{L},\tilde{m}}(z,t) = \frac{i}{\Delta - i\frac{\gamma}{2}}|g_0|^2\sum_{n}\beta_{\tilde{m},n}(z)a_{\mathrm{L},n}(z,t) + \frac{i}{2(\Delta - i\frac{\gamma}{2})}g_0^*\Omega\sum_{n}\gamma_{\tilde{m},n}(z)a_{\mathrm{A},n}(z,t) \tag{42}$$

$$\frac{\partial}{\partial t}a_{\mathrm{A},\tilde{m}}(z,t) = \frac{i}{4(\Delta - i\frac{\gamma}{2})}|\Omega|^2 a_{\mathrm{A},\tilde{m}}(z,t) + \frac{i}{2(\Delta - i\frac{\gamma}{2})}g_0\Omega^*\sum_{n}\gamma_{\tilde{m},n}(z)a_{\mathrm{L},n}(z,t). \tag{43}$$

5 Solving the equations of motion

The equations of motion, eqs. (42) and (43), are operator differential equations. But in fact, the operators $a_{\mathrm{X},\tilde{m}}(z,t)$ (X = L,A) can be reinterpreted as $\mathbb{C}$-valued functions, which are mathematically simpler, without thereby throwing away essential information. This point is argued in two different ways in section II C of reference [5], one of which is based on the facts that the set of coherent states spans the ket space and that taking expectation values w.r.t. coherent states preserves the form of the equations.

Since we are concerned with the process of reading out from the atomic ensemble (starting at $t = 0$), we need to solve for $a_{\mathrm{L}}(z,t)$ with initial conditions $a_{\mathrm{L}}(z,t=0) = 0$ and some $a_{\mathrm{A}}(z,t=0)$ assumed to be given.

5.1 Elimination of $a_{\mathbf{A}}(z,t)$

We start out by eliminating $a_{\mathrm{A}}(z,t)$ from the set of coupled differential equations (42) and (43) by means of the Laplace transform, which is defined as

$$F(\omega) = \mathcal{L}\{f(t)\} = \int_0^\infty e^{-\omega t} f(t) dt,$$

where ω is a complex frequency variable. The Laplace transform is a kind of one-sided Fourier transform yielding a frequency-domain representation of a time-domain function.

Laplace transforming eq. (43) and exploiting that $\mathcal{L}\{\frac{\partial}{\partial t} a_{A,\tilde{m}}(z,t)\} = \omega \cdot \mathcal{L}\{a_{A,\tilde{m}}(z,t)\} - a_{A,\tilde{m}}(z,t=0)$ we get

$$\omega a_{\mathrm{A},\tilde{m}}(z,\omega) - a_{\mathrm{A},\tilde{m}}(z,t=0) = \frac{i}{4(\Delta - i\frac{\gamma}{2})}|\Omega|^2 a_{\mathrm{A},\tilde{m}}(z,\omega) + \frac{i}{2(\Delta - i\frac{\gamma}{2})} g_0 \Omega^* \sum_n \gamma_{\tilde{m},n}(z) a_{\mathrm{L},n}(z,\omega), \tag{44}$$

where we have defined $a_{\mathrm{A},\tilde{m}}(z,\omega) \equiv \mathcal{L}\{a_{\mathrm{A},\tilde{m}}(z,t)\}$ with a similar definition for $a_{\mathrm{L},n}(z,\omega)$. Switching to matrix notation, where for instance $(\underline{\underline{\beta}}(z))_{m,n} = \beta_{m,n}(z)$ and the m'th component of $\vec{a}_{\mathrm{A}}(z,\omega)$ is $a_{\mathrm{A},m}(z,\omega)$, eq. (44) can be rearranged to read

$$\left(\omega - \frac{i}{4(\Delta - i\frac{\gamma}{2})}|\Omega|^2\right)\vec{a}_{\mathrm{A}}(z,\omega) = \vec{a}_{\mathrm{A}}(z,t=0) + \frac{i}{2(\Delta - i\frac{\gamma}{2})} g_0 \Omega^* \underline{\underline{\gamma}}(z)\vec{a}_{\mathrm{L}}(z,\omega).$$

Assuming $\omega - \frac{i}{4(\Delta - i\frac{\gamma}{2})}|\Omega|^2 \neq 0$, we can solve for $\vec{a}_{\mathrm{A}}(z,\omega)$:

$$\vec{a}_{\mathrm{A}}(z,\omega) = \left(\omega - \frac{i}{4(\Delta - i\frac{\gamma}{2})}|\Omega|^2\right)^{-1}\left(\vec{a}_{\mathrm{A}}(z,t=0) + \frac{i}{2(\Delta - i\frac{\gamma}{2})} g_0 \Omega^* \underline{\underline{\gamma}}(z)\vec{a}_{\mathrm{L}}(z,\omega)\right); \tag{45}$$

then Laplace transforming eq. (42) and plugging in eq. (45) for $\vec{a}_{\mathrm{A}}(z,\omega)$ we obtain in matrix notation

$$\begin{aligned}\frac{\partial}{\partial z}\vec{a}_{\mathrm{L}}(z,\omega) &= \frac{i}{\Delta - i\frac{\gamma}{2}}|g_0|^2 \underline{\underline{\beta}}(z)\vec{a}_{\mathrm{L}}(z,\omega) + \frac{i}{2(\Delta - i\frac{\gamma}{2})} g_0^* \Omega \underline{\underline{\gamma}}(z)\vec{a}_{\mathrm{A}}(z,\omega) \\ &= |g_0|^2 \frac{i}{\Delta - i\frac{\gamma}{2}}\underline{\underline{\underline{M}}}(z,\omega)\vec{a}_{\mathrm{L}}(z,\omega) - 2\left(|\Omega|^2 + 4i(\Delta - i\frac{\gamma}{2})\omega\right)^{-1} g_0^* \Omega \underline{\underline{\gamma}}(z)\vec{a}_{\mathrm{A}}(z,t=0),\end{aligned} \tag{46}$$

with $\underline{\underline{\underline{M}}}(z,\omega) = \underline{\underline{\beta}}(z) - \left(1 + 4i(\Delta - i\frac{\gamma}{2})\frac{\omega}{|\Omega|^2}\right)^{-1}\underline{\underline{\gamma}}^2(z)$.

5.2 Bessel beams as basis modes

To proceed, we will need to choose a specific basis $\{g_{\vec{k}}(\vec{r})\}$ of solutions to the Helmholtz equation, which, in turn, gives rise to the set of modes $\{u_n(\vec{r}_\perp, z)\}$ as described in section 2.1.2. We will make our choice so as to make subsequent calculations as simple as possible; to this end, we would specifically like to get rid of the z dependence of the matrix $\underline{\underline{\underline{M}}}$ for reasons to be explained below. In section 3.2.2 we introduced the assumption that the number density $n(\vec{r})$ be cylindrically symmetric around the propagation axis, that is, $n(\vec{r}) = n(\rho)$, where $\rho = |\vec{r}_\perp|$ (in fact, we assumed a Gaussian distribution in the radial coordinate). To find a complete set of mode functions $\{g_{\vec{k}}(\vec{r})\}$ adapted to this symmetry, we study the solutions of the Helmholtz equation in cylindrical coordinates. It is the Sturm-Liouville theory of differential equations that guarantees the desired properties of the set of solutions: Orthogonality and completeness.

The Helmholtz equation in cylindrical coordinates reads:

$$(\nabla^2 + k^2)u(\vec{r}) = 0 \Leftrightarrow \left(\frac{\partial^2}{\partial z^2} + \frac{1}{\rho}\frac{\partial}{\partial \rho} + \frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho^2}\frac{\partial^2}{\partial \phi^2} + k^2\right)u(\rho,\phi,z) = 0.$$

Assuming separability $u(\rho,\phi,z) = \Upsilon(\rho)\Phi(\phi)Z(z)$ and dividing through by u, wee see that $\frac{1}{Z}\frac{\partial^2 Z}{\partial z^2} = \text{const}$. Choosing this constant to be $-k_\parallel^2$, the forward propagating (non-decaying) solution is (up to a multiplicative constant) $Z(z) = e^{ik_\parallel z}$; here $k_\parallel$ plays the role of a longitudinal wave number. Inserting this result back into the Helmholtz equation and multiplying by ρ^2 we now have

$$\frac{1}{\Upsilon(\rho)}\left(\rho\frac{\partial}{\partial \rho} + \rho^2\frac{\partial^2}{\partial \rho^2}\right)\Upsilon(\rho) + [k^2 - k_\parallel^2]\rho^2 + \frac{1}{\Phi(\phi)}\frac{\partial^2\Phi(\phi)}{\partial \phi^2} = 0,$$

then similarly we conclude that $\Phi(\phi) = e^{im\phi}$ with $m \in \mathbb{Z}$ from the periodic boundary condition on Φ, i.e. $\Phi(\phi) = \Phi(\phi + 2\pi)$. Plugging this back into the differential equation we get

$$(\rho\frac{\partial}{\partial\rho} + \rho^2\frac{\partial^2}{\partial\rho^2})\Upsilon(\rho) + ([k^2 - k_\parallel^2]\rho^2 - m^2)\Upsilon(\rho) = 0$$

or, by introducing the rescaled coordinate $\tilde{\rho} = \sqrt{k^2 - k_\parallel^2}\rho$ (since we interpret the constants $k_\parallel, k$ in relation to a wave vector $\vec{k}$, $k \geq |k_\parallel|$), this is exactly Bessel's differential equation:

$$(\tilde{\rho}\frac{\partial}{\partial\tilde{\rho}} + \tilde{\rho}^2\frac{\partial^2}{\partial\tilde{\rho}^2})\tilde{\Upsilon}(\tilde{\rho}) + (\tilde{\rho}^2 - m^2)\tilde{\Upsilon}(\tilde{\rho}) = 0,$$

where $\tilde{\Upsilon}(\tilde{\rho}) = \Upsilon(\tilde{\rho}/\sqrt{k^2 - k_\parallel^2})$ or $\Upsilon(\rho) = \tilde{\Upsilon}(\sqrt{k^2 - k_\parallel^2}\rho)$.

The solutions relevant to our problem are the Bessel functions of the first kind $\tilde{\Upsilon}(\tilde{\rho}) = J_m(\tilde{\rho})$, hence $\Upsilon(\rho) = J_m(\sqrt{k^2 - k_\parallel^2}\rho)$. In order to achieve a discrete set of solutions, a boundary condition must be imposed on the set of solutions. We will require $\Upsilon(\rho)$ to vanish at some virtual cut-off radius $\rho = R$, from which we get the following condition $\sqrt{k^2 - k_\parallel^2} = \lambda_{m,n}/R$ for some $n \in \mathbb{N}$, where $\lambda_{m,n}$ is the n'th zero of $J_m(\tilde{\rho})$. Rearranging the relation between $k_\parallel, k$ and $\lambda_{m,n}$ in order to get an expression for $k_\parallel$ we find that $k_{\parallel m,n} = \pm\sqrt{k^2 - \frac{\lambda_{m,n}^2}{R^2}}$, now indexing the "allowed" $k_\parallel$. Since we will consider forward propagating light only, $k_\parallel > 0$, we choose the plus sign; moreover, because we are working in the paraxial regime and $k_{\perp m,n} \equiv \frac{\lambda_{m,n}}{R}$ can be seen as the perpendicular wave number of the mode function we are determining, we get the condition $k \gg \frac{\lambda_{m,n}}{R}$.

Thus, for each $k = |\vec{k}|$, we have obtained the following (countably infinite) basis set of *Bessel beam* solutions to the Helmholtz equation:

$$\{g_{k,m,n}(\rho, \phi, z) = N_{m,n}e^{ik_{\parallel m,n}z}e^{im\phi}J_m(k_{\perp m,n}\rho)\}_{m\in\mathbb{Z},n\in\mathbb{N}}, \tag{47}$$

where the normalization constant is $N_{m,n} = (\pi R^2 J_{m+1}^2(\lambda_{m,n}))^{-\frac{1}{2}}$ and the components of $\vec{k}$ are given by $k_{\perp m,n} = \frac{\lambda_{m,n}}{R}, k_{\parallel m,n} = \sqrt{k^2 - \frac{\lambda_{m,n}^2}{R^2}}$.

In section 2.1.2 we assumed that we could factor out the k dependence of the solutions as $g_{k,m,n}(\rho, \phi, z) = u_{m,n}(\vec{r}_\perp, z)e^{ikz}$, thereby obtaining a mode set $\{u_{m,n}(\vec{r}_\perp, z)\}$. For the set of Bessel beams in eq. (47), this can be achieved by writing (suppressing mode indices) $e^{ik_\parallel z} = e^{i(k_\parallel - k)z}e^{ikz}$ and performing the following two approximations: Firstly, $k_\parallel - k = \sqrt{k^2 - k_\perp^2} - k = k\left(\sqrt{1 - \frac{k_\perp^2}{k^2}} - 1\right) \approx -\frac{1}{2}\frac{k_\perp^2}{k}$, valid in the paraxial region where $k_\perp \ll k$; secondly, we further approximate $-\frac{1}{2}\frac{k_\perp^2}{k} \approx -\frac{1}{2}\frac{k_\perp^2}{k_0}$ which is OK since we consider a narrow band of k-values around k_0, $\frac{\Delta k}{k_0} \ll 1$. Hence, the k-independent set of modes $\{u_{m,n}(\vec{r}_\perp, z)\}$ corresponding to the paraxial Bessel beams is[15]

$$\{u_{m,n}(\rho, \phi, z) = N_{m,n}e^{i(k_0 - k_0')z}e^{-\frac{i}{2}\frac{k_{\perp m,n}^2}{k_0}z}e^{im\phi}J_m(\lambda_{m,n}\rho/R)\}_{m\in\mathbb{Z},n\in\mathbb{N}},$$

where we have included the phase factor $e^{i(k_0 - k_0')z}$ absorbed into the $u_{m,n}$'s in subsection 3.2.1; however, at this point the $u_{m,n}$'s only enter the equations in the combination $\zeta_{(m,n);(\tilde{m},\tilde{n})} = u_{m,n}^*(\vec{r}_\perp, z)u_{\tilde{m},\tilde{n}}(\vec{r}_\perp, z)$, accordingly the factor $e^{i(k_0 - k_0')z}$ in $u_{m,n}(\vec{r}_\perp, z)$ does not make a difference. The normalization constant $N_{m,n}$ is the same as above.

These mode functions are indeed orthogonal in any transverse plane:

$$\int d^2\vec{r}_\perp u_{m,n}^*(\vec{r}_\perp, z)u_{\tilde{m},\tilde{n}}(\vec{r}_\perp, z) = \delta_{m,\tilde{m}}\delta_{n,\tilde{n}} \tag{48}$$

as can be checked by exploiting the orthogonality properties

$$\int_0^{2\pi} d\phi e^{i(\tilde{m}-m)\phi} = 2\pi\delta_{m,\tilde{m}}, \quad \int_0^1 ds J_m(\lambda_{m,n}s)J_m(\lambda_{m,\tilde{n}}s)s = \delta_{n,\tilde{n}}\frac{1}{2}J_{m+1}^2(\lambda_{m,n});$$

[15]Note that although we have written u_m in the above (with one integer index variable) rather than $u_{m,n}$ for the mode functions, the two sets $\mathbb{Z}$ and $\mathbb{Z} \times \mathbb{N}$ are in bijection. Hence the two are equivalent as index sets, although obviously the latter is the more natural one for this particular set of modes.

(notice the weight function $w(s) = s$ in the latter).

Now that we have achieved a suitable set of mode functions, we move on to work on the coupling functions introduced in eq. (35) of section 3.2.2; carrying out the angular integral using $\int_0^{2\pi} d\phi e^{i(\tilde{m}-m)\phi} = 2\pi\delta_{m,\tilde{m}}$ we get:

$$\gamma_{(m,n);(\tilde{m},\tilde{n})}(z) = \int d^2\vec{r}_\perp \zeta_{(m,n);(\tilde{m},\tilde{n})}(\vec{r}_\perp, z) e^{-\frac{\rho^2}{4\sigma_\perp^2}} = \delta_{m,\tilde{m}} e^{\frac{i}{2k_0}(k_{\perp m,n}^2 - k_{\perp m,\tilde{n}}^2)z} G_{n,\tilde{n}}^{(m)} \tag{49}$$

where we have defined

$$G_{n,\tilde{n}}^{(m)} \equiv 2\pi N_{m,n} N_{m,\tilde{n}} \int_0^R d\rho J_m(\lambda_{m,n}\rho/R) J_m(\lambda_{m,\tilde{n}}\rho/R) \rho e^{-\frac{\rho^2}{4\sigma_\perp^2}}. \tag{50}$$

By a similar calculation,

$$\beta_{(m,n);(\tilde{m},\tilde{n})}(z) = \int d^2\vec{r}_\perp \zeta_{(m,n);(\tilde{m},\tilde{n})}(\vec{r}_\perp, z) e^{-\frac{\rho^2}{2\sigma_\perp^2}} = \delta_{m,\tilde{m}} e^{\frac{i}{2k_0}(k_{\perp m,n}^2 - k_{\perp m,\tilde{n}}^2)z} B_{n,\tilde{n}}^{(m)} \tag{51}$$

where we define

$$B_{n,\tilde{n}}^{(m)} \equiv 2\pi N_{m,n} N_{m,\tilde{n}} \int_0^R d\rho J_m(\lambda_{m,n}\rho/R) J_m(\lambda_{m,\tilde{n}}\rho/R) \rho e^{-\frac{\rho^2}{2\sigma_\perp^2}}. \tag{52}$$

Thus, we see that $\beta_{(m,n);(\tilde{m},\tilde{n})}(z)$ and $\gamma_{(m,n);(\tilde{m},\tilde{n})}(z)$ are both diagonal in the m-$\tilde{m}$ indices, which is why we have introduced the more natural $B_{n,\tilde{n}}^{(m)}$, $G_{n,\tilde{n}}^{(m)}$ notation which singles out the various m-subspaces. Moreover, the z-dependence of $\beta_{(m,n);(\tilde{m},\tilde{n})}(z), \gamma_{(m,n);(\tilde{m},\tilde{n})}(z)$ is seen to be simply a phase factor $e^{\frac{i}{2k_0}(k_{\perp m,n}^2 - k_{\perp m,\tilde{n}}^2)z}$.

5.3 Discussion of the $R \to \infty$ limit and evaluation of $G_{n,\tilde{n}}^{(m)}$, $B_{n,\tilde{n}}^{(m)}$

The cut-off radius R, at which all mode functions are required to vanish, is entirely unphysical. It only serves the technical purpose of yielding a discrete set of mode functions, which is what we need in order to make connection with the theory laid out above (in which we have dealt only with expansions in terms of discrete sets of modes). Once we have obtained a solution using a finite cut-off R, we could make the transition to the more realistic case of a continuum of modes by taking the limit $R \to \infty$. This approach, however, will turn out to be troublesome as we would be taking the limit of an infinite series of infinite dimensional matrices as it turns out. It is more manageable to go back to the equations of motion, (42) and (43), and pass from a discrete mode set to a continuous one at that stage. We will demonstrate how this transition is carried out later in this section.

That we did not choose to follow that path back in section 4, was in anticipation of the fact that ultimately we will have to resort to numerical methods which necessarily entails discretization. The rationale being that the error introduced by using a finite cut-off radius R is equivalent to the error introduced by discretizing in connection with solving numerically. Thus, we might as well work with a finite cut-off radius R all along which is large compared to the characteristic radius $\sigma_\perp$ of the cloud of gas.

5.3.1 Evaluation of $G_{n,\tilde{n}}^{(m)}$, $B_{n,\tilde{n}}^{(m)}$

We will now evaluate the quantities $G_{n,\tilde{n}}^{(m)}$, $B_{n,\tilde{n}}^{(m)}$ defined in eqs. (50) and (52). We will make use of the following integral identity [4]:

$$\int_0^\infty d\rho J_m(a\rho) J_m(b\rho) \rho e^{-\mu^2\rho^2} = \frac{1}{2\mu^2} e^{-\frac{a^2+b^2}{4\mu^2}} I_m\left(\frac{ab}{2\mu^2}\right) \quad \text{for } |\arg(\mu)| < \frac{\pi}{4}; \mathrm{Re}(m) > -1; a, b > 0, \tag{53}$$

where $I_m(x) = i^{-m} J_m(ix)$ is the modified Bessel function of the first kind of order m. But since $J_{-m}(x) = (-1)^m J_m(x)$ for $m \in \mathbb{Z}$, we have the following special case of eq. (53) which is better suited to our needs

$$\int_0^\infty d\rho J_m(a\rho) J_m(b\rho) \rho e^{-\mu^2\rho^2} = \frac{1}{2\mu^2} e^{-\frac{a^2+b^2}{4\mu^2}} I_{|m|}\left(\frac{ab}{2\mu^2}\right) \quad \text{for } |\arg(\mu)| < \frac{\pi}{4}; m \in \mathbb{Z}; a, b > 0.$$

To be able to use this result, we will introduce the approximation of replacing the upper integration limit of value R in eqs. (50) and (52) by ∞;[16] this is a good approximation as long as $R \gg \sigma_\perp$. In this way we obtain

$$\begin{aligned} G_{n,\tilde{n}}^{(m)} &\approx 2\pi N_{m,n} N_{m,\tilde{n}} \int_0^\infty d\rho J_m(\lambda_{m,n}\rho/R) J_m(\lambda_{m,\tilde{n}}\rho/R) \rho e^{-\frac{\rho^2}{4\sigma_\perp^2}} \\ &= 4\left(|J_{m+1}(\lambda_{m,n}) J_{m+1}(\lambda_{m,\tilde{n}})|\right)^{-1} \frac{\sigma_\perp^2}{R^2} e^{-\frac{\sigma_\perp^2}{R^2}(\lambda_{m,n}^2 + \lambda_{m,\tilde{n}}^2)} I_{|m|}\left(2\frac{\sigma_\perp^2}{R^2}\lambda_{m,n}\lambda_{m,\tilde{n}}\right), \end{aligned} \tag{54}$$

[16] Note that we are not taking the limit $R \to \infty$ which would entail a transition to a continuum of mode functions as mentioned above; we are simply approximating the value of an integral.

$$B^{(m)}_{n,\tilde{n}} \approx 2\pi N_{m,n}N_{m,\tilde{n}}\int_0^\infty d\rho J_m(\lambda_{m,n}\rho/R)J_m(\lambda_{m,\tilde{n}}\rho/R)\rho e^{-\frac{\rho^2}{2\sigma_\perp^2}}$$
$$= 2\left(|J_{m+1}(\lambda_{m,n})J_{m+1}(\lambda_{m,\tilde{n}})|\right)^{-1}\frac{\sigma_\perp^2}{R^2}e^{-\frac{\sigma_\perp^2}{2R^2}(\lambda^2_{m,n}+\lambda^2_{m,\tilde{n}})}I_{|m|}\left(\frac{\sigma_\perp^2}{R^2}\lambda_{m,n}\lambda_{m,\tilde{n}}\right). \quad (55)$$

5.3.2 Equations of motion in the $R\to\infty$ limit

In order to demonstrate how to take the $R\to\infty$ limit, we will now restate the equations of motion (42), (43) for a_{L}, a_{A} in terms of the basis of Bessel beams. To begin with, note that the set of Bessel beam modes is indexed by a pair of integers $(m,n)\in\mathbb{Z}\times\mathbb{N}$ rather than a single integer (see the footnote on page 20), therefore the aforementioned equations must be modified accordingly $n\to(m,n)$. We then plug in the expressions for γ and β in terms of $G^{(m)}_{n,\tilde{n}}$ and $B^{(m)}_{n,\tilde{n}}$, eqs. (49) and (51), and carry out those sums made easy by Kronecker deltas to get:

$$\frac{\partial}{\partial z}a_{\mathrm{L},(m,\tilde{n})}(z,t) = \frac{i}{\Delta - i\frac{\gamma}{2}}\sum_n e^{\frac{i}{2k_0}(k^2_{\perp m,\tilde{n}}-k^2_{\perp m,n})z}\left(|g_0|^2B^{(m)}_{\tilde{n},n}a_{\mathrm{L},(m,n)}(z,t)+\frac{1}{2}g_0^*\Omega G^{(m)}_{\tilde{n},n}a_{\mathrm{A},(m,n)}(z,t)\right), \quad (56)$$

$$\frac{\partial}{\partial t}a_{\mathrm{A},(m,\tilde{n})}(z,t) = \frac{i}{2(\Delta - i\frac{\gamma}{2})}\left(\frac{1}{2}|\Omega|^2a_{\mathrm{A},(m,\tilde{n})}(z,t)+g_0\Omega^*\sum_n e^{\frac{i}{2k_0}(k^2_{\perp m,\tilde{n}}-k^2_{\perp m,n})z}G^{(m)}_{\tilde{n},n}a_{\mathrm{L},(m,n)}(z,t)\right). \quad (57)$$

When we pass into a continuum of modes by taking the limit $R\to\infty$, the sums over n will turn into integrals over $k_\perp$ since in this limit, the discrete variable n is superseded by the continuous $k_\perp$ as the variable indexing the set of modes within a given m-subspace. From here on we will suppress the m and n indices at times, however, note the following correspondences for fixed m: $k_{\perp m,n}\equiv\frac{\lambda_{m,n}}{R}\leftrightarrow k_\perp, k_{\perp m,\tilde{n}}\equiv\frac{\lambda_{m,\tilde{n}}}{R}\leftrightarrow k_{\tilde{\perp}}$. To facilitate the change of summing variable, we write $1=\Delta n=\left(\frac{\Delta k_\perp}{\Delta n}\right)^{-1}\Delta k_\perp$, where $\Delta k_\perp$ is the change in $k_\perp$ when n is increased by one.[17] As we let R go to infinity, the difference quotient becomes the derivative[18] $\frac{\Delta k_\perp}{\Delta n}\to\frac{dk_\perp}{dn}$ while $\Delta k_\perp\to dk_\perp$; we obtain the former by treating n as a continuous variable:

$$\frac{dk_\perp}{dn}=\frac{1}{R}\frac{d\lambda_{m,n}}{dn};$$

to get $\frac{d\lambda_{m,n}}{dn}$, we note that, since we are working in the $R\to\infty$ limit, it is the asymptotic behaviour ($n\to\infty$) of $\lambda_{m,n}$ that matters. Hence, we consider the asymptotic form of $J_m(x)$ as $x\to\infty$ [1]

$$J_m(x)=\sqrt{\frac{2}{\pi x}}\cos(x-\frac{1}{2}m\pi-\frac{1}{4}\pi), \quad (58)$$

from which it is easily seen that, asymptotically, the zeros obey $\lambda_{m,n}=\pi n+\mathrm{const}$; thus $\frac{d\lambda_{m,n}}{dn}=\pi$ and

$$\frac{dk_\perp}{dn}=\frac{\pi}{R}. \quad (59)$$

As for the matter at hand, the important parts of eqs. (56) and (57) are

$$\sum_n e^{\frac{i}{2k_0}(k^2_{\perp m,\tilde{n}}-k^2_{\perp m,n})z}B^{(m)}_{\tilde{n},n}a_{\mathrm{L},(m,n)}(z,t)\ \text{and}\ \sum_n e^{\frac{i}{2k_0}(k^2_{\perp m,\tilde{n}}-k^2_{\perp m,n})z}G^{(m)}_{\tilde{n},n}a_{\mathrm{X},(m,n)}(z,t),$$

where X = L,A and $G^{(m)}_{\tilde{n},n}$ and $B^{(m)}_{\tilde{n},n}$ are given by eqs. (54) and (55), respectively. The treatment of the two expressions are very similar, so we will only work out the second one in detail; plugging in for $G^{(m)}_{\tilde{n},n}$, multiplying by $1=\Delta n=\left(\frac{\Delta k_\perp}{\Delta n}\right)^{-1}\Delta k_\perp$ and reexpressing everything in terms of $k_\perp, k_{\tilde{\perp}}$,

$$\sum_n e^{\frac{i}{2k_0}(k^2_{\tilde{\perp}}-k^2_\perp)z}\left(\left|J_{m+1}(k_\perp R)J_{m+1}(k_{\tilde{\perp}}R)\right|\right)^{-1}4\frac{\sigma_\perp^2}{R^2}e^{-\sigma_\perp^2(k^2_\perp+k^2_{\tilde{\perp}})}$$
$$\times I_{|m|}\left(2\sigma_\perp^2k_\perp k_{\tilde{\perp}}\right)a_{\mathrm{X},(m,n)}(z,t)\left(\frac{\Delta k_\perp}{\Delta n}\right)^{-1}\Delta k_\perp.$$

[17] The quantity $\Delta k_\perp$ is of course generally a function of n.

[18] Although, strictly, it doesn't make sense mathematically to differentiate with respect to an integer variable since it takes on discrete values.

We are now about to take the limit $R \to \infty$ in which we can replace the occurrences of J_{m+1} by its large argument asymptotic form eq. (58) and plug in eq. (59) for $\frac{\Delta k_\perp}{\Delta n}$; we then get,

$$\int_0^\infty e^{\frac{i}{2k_0}(k_{\tilde{\perp}}^2 - k_\perp^2)z} \left(\left| \cos(k_\perp R - \frac{1}{2}(m+1)\pi - \frac{1}{4}\pi) \cos(k_{\tilde{\perp}} R - \frac{1}{2}(m+1)\pi - \frac{1}{4}\pi) \right| \right)^{-1} \times 4\frac{\pi R}{2}\sqrt{k_\perp k_{\tilde{\perp}}}\frac{\sigma_\perp^2}{R^2} e^{-\sigma_\perp^2(k_\perp^2+k_{\tilde{\perp}}^2)} I_{|m|}\left(2\sigma_\perp^2 k_\perp k_{\tilde{\perp}}\right) a_{\mathrm{X},m}(k_\perp, z, t) \left(\frac{R}{\pi}\right) dk_\perp$$

$$= \int_0^\infty e^{\frac{i}{2k_0}(k_{\tilde{\perp}}^2 - k_\perp^2)z} 2\sqrt{k_\perp k_{\tilde{\perp}}}\sigma_\perp^2 e^{-\sigma_\perp^2(k_\perp^2+k_{\tilde{\perp}}^2)} I_{|m|}\left(2\sigma_\perp^2 k_\perp k_{\tilde{\perp}}\right) a_{\mathrm{X},m}(k_\perp, z, t) dk_\perp \tag{60}$$

where we have made use of the fact that in the $R \to \infty$ limit, $k_\perp R$ is a zero of $\cos(x - \frac{1}{2}m\pi - \frac{1}{4}\pi)$, hence $|\cos(k_\perp R - \frac{1}{2}(m+1)\pi - \frac{1}{4}\pi)| = 1$ in this limit. Note that we have set ∞ as the upper integration limit despite the facts that physically $k_\perp$ cannot exceed the length of $\vec{k}$ and that, moreover, we are working in the paraxial regime. We assume, however, that the contribution from modes with $k_\perp$ values outside the paraxial region is negligible; if this were not the case, working in the paraxial regime would not make sense. By introducing rescaled, dimensionless variables $\bar{k}_\perp = k_\perp\sqrt{\frac{L}{k_0}}, \bar{k}_{\tilde{\perp}} = k_{\tilde{\perp}}\sqrt{\frac{L}{k_0}}, y = \frac{z}{L}, \bar{a}_{\mathrm{X},m}(\bar{k}_\perp, z, t) = a_{\mathrm{X},m}(k_\perp = \bar{k}_\perp\sqrt{\frac{k_0}{L}}, z, t)$, eq. (60) reads

$$\int_0^\infty e^{\frac{i}{2}(\bar{k}_{\tilde{\perp}}^2 - \bar{k}_\perp^2)y} 4\pi\sqrt{\bar{k}_\perp \bar{k}_{\tilde{\perp}}} F e^{-2\pi F(\bar{k}_\perp^2 + \bar{k}_{\tilde{\perp}}^2)} I_{|m|}\left(4\pi F \bar{k}_\perp \bar{k}_{\tilde{\perp}}\right) \bar{a}_{\mathrm{X},m}(\bar{k}_\perp, z, t) d\bar{k}_\perp,$$

where we have defined the *Fresnel number* $F = \frac{\sigma_\perp^2}{L\lambda_0}$ where $\lambda_0 = \frac{2\pi}{k_0}$; note that F is the only constant appearing in this equation. We will refer to this result later.

5.4 Formal solution

Since the matrices $\underline{\underline{\beta}}(z), \underline{\underline{\gamma}}(z)$, that govern how modes couple in the equations of motion, are diagonal in $m, \tilde{m}$, there is no coupling between modes with distinct m values. Hence, we can consider each invariant m-subspace separately when solving the equations of motion.

The restriction of the matrix $\underline{\underline{M}}(z, \omega)$ to a particular m-subspace is then (in component form)

$$\begin{aligned} (\underline{\underline{M}}^{(m)})_{\alpha,\beta}(z,\omega) &= e^{\frac{i}{2k_0}(k_{\perp m,\alpha}^2 - k_{\perp m,\beta}^2)z} \left[B_{\alpha,\beta}^{(m)} - \left(1 + 4i(\Delta - i\frac{\gamma}{2})\frac{\omega}{|\Omega|^2}\right)^{-1} \sum_\sigma G_{\alpha,\sigma}^{(m)} G_{\sigma,\beta}^{(m)} \right] \\ &= e^{\frac{i}{2k_0}(k_{\perp m,\alpha}^2 - k_{\perp m,\beta}^2)z} D_{\alpha,\beta}^{(m)}(\omega) \end{aligned} \tag{61}$$

which defines the matrix $D_{\alpha,\beta}^{(m)}(\omega)$ (≡bracketed term in 1st line). The expression for $D_{\alpha,\beta}^{(m)}(\omega)$ can be simplified using the following resolution of the identity (restricted to a particular m-subspace):

$$\sum_n 2\pi\rho \left[N_{m,n} J_m(\lambda_{m,n}\rho/R)\right] \left[N_{m,n} J_m(\lambda_{m,n}\rho'/R)\right]^* = \delta(\rho - \rho'), \tag{62}$$

which can be verified by checking that for any function $f(\rho) = \sum_{n'} c_{n'} N_{m,n'} J_m(\lambda_{m,n'}\rho/R)$ it holds true that

$$\int_0^R d\rho \sum_n 2\pi\rho \left[N_{m,n} J_m(\lambda_{m,n}\rho/R)\right] \left[N_{m,n} J_m(\lambda_{m,n}\rho'/R)\right]^* f(\rho) = f(\rho');$$

using this resolution of the identity, eq. (62), it is easily shown that (here using the definition of $G_{\alpha,\beta}^{(m)}$, eq. (50), rather than the approximate expression, eq. (54))

$$\sum_\sigma G_{\alpha,\sigma}^{(m)} G_{\sigma,\beta}^{(m)} = B_{\alpha,\beta}^{(m)}. \tag{63}$$

Using this result, we can now simplify $D_{\alpha,\beta}^{(m)}(\omega)$

$$D_{\alpha,\beta}^{(m)}(\omega) = \left[1 - \left(1 + 4i(\Delta - i\frac{\gamma}{2})\frac{\omega}{|\Omega|^2}\right)^{-1}\right] B_{\alpha,\beta}^{(m)} = \left(1 + \frac{|\Omega|^2}{4i(\Delta - i\frac{\gamma}{2})\omega}\right)^{-1} B_{\alpha,\beta}^{(m)}$$

By plugging in eq. (61) for $\underline{\underline{M}}(z,\omega)$, the differential equation (46) for $a^{(m)}_{\mathrm{L},\alpha}(z,\omega)$ now reads

$$\frac{\partial}{\partial z}a^{(m)}_{\mathrm{L},\alpha}(z,\omega) = -\sum_{\beta} e^{\frac{i}{2k_0}(k^2_{\perp m,\alpha}-k^2_{\perp m,\beta})z}\left(|g_0|^2\left(i(\Delta-i\frac{\gamma}{2})+\frac{|\Omega|^2}{4\omega}\right)^{-1}B^{(m)}_{\alpha,\beta}a^{(m)}_{\mathrm{L},\beta}(z,\omega)\right.$$
$$\left.+2\left(|\Omega|^2+4i(\Delta-i\frac{\gamma}{2})\omega\right)^{-1}g_0^*\Omega G^{(m)}_{\alpha,\beta}a^{(m)}_{\mathrm{A},\beta}(z,t=0)\right) \quad (64)$$

Defining $\tilde{a}^{(m)}_{\mathrm{L},\alpha}(z,\omega) \equiv a^{(m)}_{\mathrm{L},\alpha}(z,\omega)e^{-\frac{i}{2k_0}k^2_{\perp m,\alpha}z}$, $\tilde{a}^{(m)}_{\mathrm{A},\alpha}(z,t=0) \equiv a^{(m)}_{\mathrm{A},\alpha}(z,t=0)e^{-\frac{i}{2k_0}k^2_{\perp m,\alpha}z}$ and noting that $\frac{\partial}{\partial z}\left[\tilde{a}^{(m)}_{\mathrm{L},\alpha}(z,\omega)\right]+i\frac{k^2_{\perp m,\alpha}}{2k_0}\tilde{a}^{(m)}_{\mathrm{L},\alpha}(z,\omega) = e^{-\frac{i}{2k_0}k^2_{\perp m,\alpha}z}\frac{\partial}{\partial z}\left[a^{(m)}_{\mathrm{L},\alpha}(z,\omega)\right]$ equation (64) becomes

$$\frac{\partial}{\partial z}\left[\tilde{a}^{(m)}_{\mathrm{L},\alpha}(z,\omega)\right]+i\frac{k^2_{\perp m,\alpha}}{2k_0}\tilde{a}^{(m)}_{\mathrm{L},\alpha}(z,\omega) = -\sum_{\beta}\left(|g_0|^2\left(i(\Delta-i\frac{\gamma}{2})+\frac{|\Omega|^2}{4\omega}\right)^{-1}B^{(m)}_{\alpha,\beta}\tilde{a}^{(m)}_{\mathrm{L},\beta}(z,\omega)\right.$$
$$\left.+2\left(|\Omega|^2+4i(\Delta-i\frac{\gamma}{2})\omega\right)^{-1}g_0^*\Omega G^{(m)}_{\alpha,\beta}\tilde{a}^{(m)}_{\mathrm{A},\beta}(z,t=0)\right). \quad (65)$$

Furthermore, by moving the term $i\frac{k^2_{\perp m,\alpha}}{2k_0}\tilde{a}^{(m)}_{\mathrm{L},\alpha}(z,\omega)$ to the right-hand side and defining

$$E^{(m)}_{\alpha,\beta}(\omega) \equiv -i\frac{k^2_{\perp m,\alpha}}{2k_0}\delta_{\alpha,\beta}-|g_0|^2\left(i(\Delta-i\frac{\gamma}{2})+\frac{|\Omega|^2}{4\omega}\right)^{-1}B^{(m)}_{\alpha,\beta}, \quad (66)$$

we can write eq. (65) as

$$\frac{\partial}{\partial z}\left[\tilde{a}^{(m)}_{\mathrm{L},\alpha}(z,\omega)\right] = \sum_{\beta}\left(E^{(m)}_{\alpha,\beta}(\omega)\tilde{a}^{(m)}_{\mathrm{L},\beta}(z,\omega)-2\left(|\Omega|^2+4i(\Delta-i\frac{\gamma}{2})\omega\right)^{-1}g_0^*\Omega G^{(m)}_{\alpha,\beta}\tilde{a}^{(m)}_{\mathrm{A},\beta}(z,t=0)\right). \quad (67)$$

At this point we do not have to write the matrix indices explicitly anymore and we can restate the previous equation as

$$\frac{\partial}{\partial z}\left[\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z,\omega)\right] = \underline{\underline{E}}^{(m)}(\omega)\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z,\omega)+\nu(\omega)\underline{\underline{G}}^{(m)}\tilde{\vec{a}}^{(m)}_{\mathrm{A}}(z,t=0), \quad (68)$$

where we have defined the function $\nu(\omega) = -2\left(|\Omega|^2+4i(\Delta-i\frac{\gamma}{2})\omega\right)^{-1}g_0^*\Omega$. Since the matrix $\underline{\underline{E}}^{(m)}(\omega)$ multiplying $\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z,\omega)$ is independent of z,[19] the formal solution to this equation is

$$\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z,\omega) = \nu(\omega)\int_0^z dz' \exp\left[\underline{\underline{E}}^{(m)}(\omega)(z-z')\right]\underline{\underline{G}}^{(m)}\tilde{\vec{a}}^{(m)}_{\mathrm{A}}(z',t=0), \quad (69)$$

as can be confirmed by inspection:

$$\begin{aligned}\frac{\partial}{\partial z}\left[\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z,\omega)\right] &= \nu(\omega)\exp\left[\underline{\underline{E}}^{(m)}(\omega)(z-z')\right]\underline{\underline{G}}^{(m)}\tilde{\vec{a}}^{(m)}_{\mathrm{A}}(z',t=0)\Big|_{z'=z}\\ &+\nu(\omega)\underline{\underline{E}}^{(m)}(\omega)\int_0^z dz'\exp\left[\underline{\underline{E}}^{(m)}(\omega)(z-z')\right]\underline{\underline{G}}^{(m)}\tilde{\vec{a}}^{(m)}_{\mathrm{A}}(z',t=0)\\ &= \nu(\omega)\underline{\underline{G}}^{(m)}\tilde{\vec{a}}^{(m)}_{\mathrm{A}}(z,t=0)+\underline{\underline{E}}^{(m)}(\omega)\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z,\omega).\end{aligned}$$

Since we are interested in examining the retrieval efficiency, we set $z=L$ in eq. (69), thereby obtaining

$$\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z=L,\omega) = \nu(\omega)\int_0^L dz'\exp\left[\underline{\underline{E}}^{(m)}(\omega)(L-z')\right]\underline{\underline{G}}^{(m)}\tilde{\vec{a}}^{(m)}_{\mathrm{A}}(z',t=0). \quad (70)$$

5.5 Restatement of formal solution in terms of dimensionless quantities

The formal solution for $\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z=L,\omega)$ in terms of the initial condition $\tilde{\vec{a}}^{(m)}_{\mathrm{A}}(z',t=0)$ is given in eq. (70); this expression is, however, not amenable to analytical solution. Instead, we will work towards a numerical solution. In order to bring the solution into a form suited as input to a computer, we will rewrite it solely in terms of

[19] Otherwise, in a scenario where $[\underline{\underline{E}}^{(m)}(z),\underline{\underline{E}}^{(m)}(z')] \neq 0$ for $z \neq z'$ we would have to concern ourselves with length-ordering which would enter in much the same way that time-ordering appears in the Dyson series for an interaction Hamiltonian that does not commute with itself at unequal times.

dimensionless quantities. But just as importantly, this will, in addition, provide us with physical insight into which quantities (or combinations thereof) are actually determining the dynamics of the system.

Eq. (70) involves the following dimensionful quantities: The mode frequency ω, detuning Δ, spontaneous decay rate γ, and Rabi frequency $\Omega = 2\vec{D}_{e,1}^{(-)} \cdot \langle \vec{E}_{0,c}^{(+)}(\vec{r})\rangle e^{-ik_0' z}$ all have units of $\frac{1}{\text{time}}$; the quantum field coupling constant $g_0 = \sqrt{\frac{\omega_0 N_\text{A}}{cL\sigma_\perp^2}} D_0$ has unit of $\frac{1}{\sqrt{\text{length}\cdot\text{time}}}$. The following quantities have dimensions of length: Cutoff radius R, radial spread of atomic density $\sigma_\perp$, and the length of the container L. Meanwhile, wave vector quantities $k_0, k_\perp$ have unit of $\frac{1}{\text{length}}$.

We define dimensionless length and frequency variables $y' \equiv z'/L, u \equiv \frac{\omega\gamma}{|\Omega|^2}$, dimensionless wave vector components $\bar{k}_{\perp m,n} \equiv k_{\perp m,n}\sqrt{\frac{L}{k_0}}$, and the following dimensionless function of u: $X(u) \equiv 2\left(4(\Delta/\gamma - i/2)u - i\right)^{-1}$. The dimensions of $\tilde{\vec{a}}_\text{L}^{(m)}(z = L, \omega)$ and $\tilde{\vec{a}}_\text{A}^{(m)}(z', t = 0)$, respectively, can be obtained by noting that the following integrals should yield (dimensionless) probabilities $\int |\tilde{\vec{a}}_\text{L}^{(m)}(z = L, \omega)|^2 d\omega, \int_0^L |\tilde{\vec{a}}_\text{A}^{(m)}(z', t = 0)|^2 dz'$ from which we see that the former has dimension of $\sqrt{\text{time}}$ and the latter $\frac{1}{\sqrt{\text{length}}}$. We define a new vector of rescaled, dimensionless light mode annihilation operators[20] $\hat{\vec{a}}_\text{L}^{(m)}(u) \equiv \kappa\tilde{\vec{a}}_\text{L}^{(m)}(z = L, \omega = u\frac{|\Omega|^2}{\gamma})$ which is a function of the dimensionless frequency $u = \frac{\omega\gamma}{|\Omega|^2}$; the probability integral is $\int |\hat{\vec{a}}_\text{L}^{(m)}(u)|^2 du = |\kappa|^2 \frac{\gamma}{|\Omega|^2} \int |\tilde{\vec{a}}_\text{L}^{(m)}(z = L, \omega)|^2 d\omega$, hence we can choose $\kappa = \frac{\Omega^*}{\sqrt{\gamma}}$ in the definition of $\hat{\vec{a}}_\text{L}^{(m)}(u)$. Similarly, a vector of dimensionless atomic annihilation operators is obtained as $\hat{\vec{a}}_\text{A}^{(m)}(y') \equiv \sqrt{L}\tilde{\vec{a}}_\text{A}^{(m)}(z' = y'L, t = 0)$, the normalization of which (w.r.t. the dimensionless y') follows from that of $\tilde{\vec{a}}_A^{(m)}(z', t = 0)$ (w.r.t. z'): $1 = \int_0^L |\tilde{\vec{a}}_A^{(m)}(z', t = 0)|^2 dz' = \int_0^1 |\hat{\vec{a}}_\text{A}^{(m)}(y')|^2 dy'$.

We will now restate the formal solution eq. (70) in terms of these newly introduced dimensionless quantities including the transformations of variables $\omega \to u = \frac{\omega\gamma}{|\Omega|^2}, z' \to y' = \frac{z}{L}$. To begin with, we consider the term $L\underline{\underline{E}}^{(m)}(u)$, eq.(66), that will appear below; in component form it can be written as

$$[LE_{\alpha,\beta}^{(m)}(u)] \quad = \quad -\frac{i}{2}\bar{k}_{\perp m,\alpha}^2\delta_{\alpha,\beta} + 2i\left[\frac{\omega_0 N_\text{A}}{c\gamma\sigma_\perp^2}|D_0|^2\right] uX(u)B_{\alpha,\beta}^{(m)},$$

where the dimensionless matrix $\underline{\underline{B}}^{(m)}$ has components as given by eq. (55) and the bracketed term denotes a dimensionless combination of constants.

We now transform the integration variable $z' \to y'$ in eq. (70) and then factor and move around various dimensionful quantities:

$$\begin{aligned}
[\tilde{\vec{a}}_\text{L}^{(m)}(z = L, \omega)\frac{\Omega^*}{\sqrt{\gamma}}] \quad &= \quad [\nu(\omega)\sqrt{\frac{L}{\gamma}}\Omega^*] \int_0^1 dy' \exp\left[L\underline{\underline{E}}^{(m)}(u)(1 - y')\right] \underline{\underline{G}}^{(m)}[\tilde{\vec{a}}_A^{(m)}(z' = y'L, t = 0)\sqrt{L}] \Leftrightarrow \\
\hat{\vec{a}}_\text{L}^{(m)}(u) \quad &= \quad i\left[\sqrt{\frac{\omega_0 N_\text{A}}{c\gamma\sigma_\perp^2}} D_0^*\right] X(u) \int_0^1 dy' \exp\left[L\underline{\underline{E}}^{(m)}(u)(1 - y')\right] \underline{\underline{G}}^{(m)} \hat{\vec{a}}_\text{A}^{(m)}(y'). \qquad (71)
\end{aligned}$$

The formal solution has now been put on dimensionless form in eq. (71); summarizing the constituents of this expression (all of the following are dimensionless quantities):

- ✿ Atomic input[21] state: $\hat{\vec{a}}_\text{A}^{(m)}(y')$ specifies the initial ($t = 0$) atomic state as function of the dimensionless axial position $y \in [0; 1]$.
- ✿ Integral transform kernel: $\underline{\underline{K}}^{(m)}(u, y') \equiv i[\sqrt{\frac{\omega_0 N_\text{A}}{c\gamma\sigma_\perp^2}} D_0^*] X(u) \exp\left[L\underline{\underline{E}}^{(m)}(u)(1 - y')\right] \underline{\underline{G}}^{(m)}$ governs the coupling of the atomic input modes at y' within some m-subspace to a specific frequency component u of the light field output modes (in the same m-subspace). The constituents of the integral kernel are:
  - ♥ $X(u) = 2\left(4(\Delta/\gamma - i/2)u - i\right)^{-1}$,
  - ♥ $LE_{\alpha,\beta}^{(m)}(u) = -\frac{i}{2}\bar{k}_{\perp m,\alpha}^2\delta_{\alpha,\beta} + 2i\left[\frac{\omega_0 N_\text{A}}{c\gamma\sigma_\perp^2}|D_0|^2\right] uX(u)B_{\alpha,\beta}^{(m)}$,
  - ♥ $B_{n,\tilde{n}}^{(m)} = 2\left(|J_{m+1}(\lambda_{m,n})J_{m+1}(\lambda_{m,\tilde{n}})|\right)^{-1} \frac{\sigma_\perp^2}{R^2} e^{-\pi F(\bar{k}_{\perp m,n}^2 + \bar{k}_{\perp m,\tilde{n}}^2)} I_{|m|}\left(2\pi F\bar{k}_{\perp m,n}\bar{k}_{\perp m,\tilde{n}}\right)$,
  - ♥ $G_{n,\tilde{n}}^{(m)} = 4\left(|J_{m+1}(\lambda_{m,n})J_{m+1}(\lambda_{m,\tilde{n}})|\right)^{-1} \frac{\sigma_\perp^2}{R^2} e^{-2\pi F(\bar{k}_{\perp m,n}^2 + \bar{k}_{\perp m,\tilde{n}}^2)} I_{|m|}\left(4\pi F\bar{k}_{\perp m,n}\bar{k}_{\perp m,\tilde{n}}\right)$.

[20] We are of course still working with their $\mathbb{C}$-number representations.

[21] "Input state" in the sense that it is taken as a given.

✿ Light field output state: $\hat{\vec{a}}_{\mathrm{L}}^{(m)}(u)$ represents the light field at the output end ($z = L$) of the gas volume as a function of the dimensionless frequency variable $u \equiv \frac{\omega\gamma}{|\Omega|^2}$.

The constant $\frac{\sigma_\perp}{R}$ is an artifact of the artificial discretization we have introduced (as discussed in section 5.3) and not a characteristic quantity of the physical system; accordingly, all occurrences of $\frac{\sigma_\perp}{R}$ disappear in the $R \to \infty$ limit as we demonstrated in subsection 5.3.2. The set of constants (with physical reality) appearing in the solution has thus been boiled down to the following three quantities $\sqrt{\frac{\omega_0 N_{\mathrm{A}}}{c\gamma\sigma_\perp^2}}D_0, \frac{\Delta}{\gamma}, F = \frac{\sigma_\perp^2}{L\lambda_0}$. We see that the Ω dependence has disappeared; that we were able to absorb it into the rescaled frequency variable u means that Ω only sets the frequency scale, but doesn't influence the efficiencies.

6 Retrieval efficiency

In subsection 5.5, eq. (71), we arrived at a formal solution for (the $\mathbb{C}$-number representation of) the dimensionless light mode annihilation operator $\hat{\vec{a}}_{\mathrm{L}}^{(m)}(u)$ which is a function of the dimensionless frequency variable u. However, the retrieval efficiency is defined in terms of the time-domain function $\vec{a}_{\mathrm{L}}^{(m)}(z = L, t)$ as

$$\eta = \sum_m \int_0^\infty |\vec{a}_{\mathrm{L}}^{(m)}(z = L, t)|^2 dt, \tag{72}$$

since the flux of photons is given by $a_{\mathrm{L}}^\dagger a_{\mathrm{L}}$. Retracing the various operator definitions we have made use of in the course of the analysis, the relation between $a_{\mathrm{L},\alpha}^{(m)}(z = L, t)$ and $\hat{a}_{\mathrm{L},\alpha}^{(m)}(u = \frac{\omega\gamma}{|\Omega|^2})$ is seen to be

$$a_{\mathrm{L},\alpha}^{(m)}(z = L, t) = \mathcal{L}^{-1}\{a_{\mathrm{L},\alpha}^{(m)}(z = L, \omega)\} = e^{\frac{i}{2k_0}k_{\perp m,\alpha}^2 z}\mathcal{L}^{-1}\{\hat{a}_{\mathrm{L},\alpha}^{(m)}(z = L, \omega)\} = e^{\frac{i}{2k_0}k_{\perp m,\alpha}^2 z}\frac{\sqrt{\gamma}}{\Omega^*}\mathcal{L}^{-1}\{\hat{a}_{\mathrm{L},\alpha}^{(m)}(u = \frac{\omega\gamma}{|\Omega|^2})\},$$

but to carry out the inverse Laplace transform of the solution eq. (71) is difficult because of the way in which u enters the exponentiated matrix herein. Instead, we will convert the time-domain integral in eq. (72) to the frequency-domain.

6.1 Plancherel-type theorem for the Laplace transform

For the case of the Fourier transform, which is equivalent to the *bilateral* Laplace transform, the well-known Plancherel theorem relates the time- and frequency-domain integrals $\int_{-\infty}^{\infty}|j(t)|^2dt = \int_{-\infty}^{\infty}|J(\omega)|^2d\omega$, where $J(\omega) = \mathcal{F}\{j(t)\}$ is the Fourier transform of $j(t)$. We need a relation along these lines, but in general there is no Plancherel theorem for the Laplace transform. However, for the special case where all of the isolated singularities $\{s_i\}$ of $F(\omega) = \mathcal{L}\{f(t)\}$ are in the left half of the complex plane ($\mathrm{Re}(s_i) < 0$ for all i), there is in fact a Plancherel-type theorem relating the time- and frequency domains.

For a frequency-domain function $F(\omega)$ with all its singularities in the left half-plane, the inverse Laplace transform is given by

$$f(t) = \mathcal{L}^{-1}\{F(\omega)\} = \frac{1}{2\pi i}\int_{-i\cdot\infty}^{i\cdot\infty} e^{\omega t}F(\omega)d\omega, \tag{73}$$

which is equivalent to the inverse Fourier transform as seen by making the change of integration variable $\omega = 2\pi i\nu$ whereby we get

$$f(t) = \int_{-\infty}^{\infty} e^{2\pi i\nu t}F(\omega = 2\pi i\nu)d\nu = \mathcal{F}^{-1}\{F(\omega = 2\pi i\nu)\}, \tag{74}$$

where we have defined the Fourier transform as $J(\nu) = \mathcal{F}\{j(t)\} = \int_{-\infty}^{\infty} e^{-2\pi i\nu t}j(t)dt$. From eq. (74) we see that the Plancherel theorem for the Fourier transform implies

$$\int_{-\infty}^{\infty}|f(t)|^2dt = \int_{-\infty}^{\infty}|F(\omega = 2\pi i\nu)|^2d\nu. \tag{75}$$

This is not exactly the result we need; furthermore, $f(t)$ is not necessarily meaningful for $t < 0$. But by plugging some $t_- < 0$ into eq. (73) we see that the factor $e^{\omega t_-}$ decays for $\mathrm{Re}(\omega) \to \infty$; then assuming that $F(\omega)$ is well-behaved for large arguments we can employ the standard technique of closing the contour with a semi-circle at infinity in the right half-plane. Now since $F(\omega)$ has no singularities in this region by assumption, Cauchy's integral theorem implies $f(t) = 0$ for $t < 0$. Hence, eq. (75) reduces to the desired relation

$$\int_0^{\infty}|f(t)|^2dt = \int_{-\infty}^{\infty}|F(\omega = 2\pi i\nu)|^2d\nu = \frac{1}{2\pi}\int_{-\infty}^{\infty}|F(\omega = i\nu')|^2d\nu'. \tag{76}$$

6.2 Formal expression for the retrieval efficiency

By inspecting eq. (71), we see that the only singularity of $\hat{a}^{(m)}_{\mathrm{L},\alpha}(u)$ is at u_0 given by $4(\Delta/\gamma - i/2)u_0 - i = 0 \Leftrightarrow u_0 = (-\frac{1}{2} + i\frac{\Delta}{\gamma})/(4\frac{\Delta^2}{\gamma^2} + 1)$, for which indeed $\mathrm{Re}(u_0) < 0$, whereby $a^{(m)}_{\mathrm{L},\alpha}(z = L, \omega)$ has its only singularity at $\omega = \frac{|\Omega|^2}{\gamma}u_0$. Furthermore, $a^{(m)}_{\mathrm{L},\alpha}(z = L, \omega)$ is seen to have the asymptotic behavior $\sim \frac{1}{|\omega|}$ for large arguments. Hence, the result of the previous subsection applies to our case and we obtain from eq. (72) the following frequency-domain expression for the retrieval efficiency

$$\begin{aligned}\eta &= \frac{1}{2\pi}\sum_m \int_{-\infty}^{\infty} |\vec{a}^{(m)}_{\mathrm{L}}(z = L, \omega = i\nu)|^2 d\nu \\ &= \frac{1}{2\pi}\sum_m \int_{-\infty}^{\infty} |\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z = L, \omega = i\nu)|^2 d\nu \\ &= \frac{1}{2\pi}\sum_m \int_{-\infty}^{\infty} |\hat{\vec{a}}^{(m)}_{\mathrm{L}}(u = iu')|^2 du' \end{aligned} \tag{77}$$

using $\tilde{a}^{(m)}_{L,\alpha}(z, \omega) \equiv a^{(m)}_{L,\alpha}(z, \omega)e^{-\frac{i}{2k_0}k^2_{\perp m,\alpha}z}$ in the second line, and $\hat{\tilde{a}}^{(m)}_{\mathrm{L}}(u) \equiv \frac{\Omega^*}{\sqrt{\gamma}}\tilde{\vec{a}}^{(m)}_{\mathrm{L}}(z = L, \omega = u\frac{|\Omega|^2}{\gamma})$ and $u \equiv \frac{\omega\gamma}{|\Omega|^2}$ in the final line. The formal solution for $\hat{\vec{a}}^{(m)}_{\mathrm{L}}(u)$ is given by eq. (71); plugging this into eq. (77), using the abbreviation $\underline{\underline{K}}^{(m)}(u, y')$ for the integral transform kernel introduced in section 5.5, we get

$$\eta = \frac{1}{2\pi}\sum_m \int_{-\infty}^{\infty} du' \int_0^1 dy' \int_0^1 dy'' \hat{\vec{a}}^{(m)\dagger}_{\mathrm{A}}(y')\underline{\underline{K}}^{(m)\dagger}(u = iu', y')\underline{\underline{K}}^{(m)}(u = iu', y'')\hat{\vec{a}}^{(m)}_{\mathrm{A}}(y''), \tag{78}$$

which is the final form of the formal expression for the retrieval efficiency η.

7 Comments on numerical evaluation

The objective of this thesis, to derive a formal expression for the retrieval efficiency, was attained with eq. (78). This provides a starting point for numerical evaluation of the retrieval efficiency, a task that will be saved for later work. We will, nevertheless, make a few remarks in this section on how to adapt the formal expression to numerical treatment.

To begin with, we will need to choose values for the quantities that we found to characterize the system in section 5.5; these are $\sqrt{\frac{\omega_0 N_{\mathrm{A}}}{c\gamma\sigma^2_\perp}}D_0, \frac{\Delta}{\gamma}, F = \frac{\sigma^2_\perp}{L\lambda_0}$. Next, we must choose a value for the artificial constant $\frac{\sigma_\perp}{R} \ll 1$ and choose reasonable finite ranges of m and n. The dimensionless perpendicular wave vector components are then given by $\bar{k}_{\perp m,n} = \lambda_{m,n}\frac{\sigma_\perp}{R}\frac{1}{\sqrt{2\pi F}}$, where $\lambda_{m,n}$ is the n'th zero of $J_m(x)$.

Further discretization of eq. (78) is needed with respect to the integration variables y', y'', u'. After having introduced such discretization, whereby $\underline{\underline{K}}^{(m)}(u = iu', y'') \to \underline{\underline{K}}^{(m)(u')}_{(y'')}$ and $\hat{\vec{a}}^{(m)}_{\mathrm{A}}(y'') \to \hat{a}^{(m)}_{\mathrm{A},n,y''}$, the contraction between these quantities appears in the equation as $\sum_{n,y''}\left(\underline{\underline{K}}^{(m)(u')}_{(y'')}\right)_{\bar{n},n}\hat{a}^{(m)}_{\mathrm{A},n,y''}$. But we can re-index so as to turn the pair of indices (n, y'') into one integer index $(n, y'') \to \bar{n}$ (see the footnote on page 20), whereby the aforementioned contraction takes the form $\sum_{\bar{n}}\left(\underline{\underline{K}}^{(m)(u')}\right)_{\bar{n},\bar{n}}\hat{a}^{(m)}_{\mathrm{A},\bar{n}}$ which we will write as $\underline{\underline{K}}^{(m)(u')}\vec{a}^{(m)}_{\mathrm{A}}$ for brevity. This transformation has a significant impact on the form of eq. (78):

$$\eta \sim \sum_m \sum_{u'} \vec{a}^{(m)\dagger}_{\mathrm{A}}\underline{\underline{K}}^{(m)(u')\dagger}\underline{\underline{K}}^{(m)(u')}\vec{a}^{(m)}_{\mathrm{A}}; \tag{79}$$

while two generally distinct matrices form the combination $\underline{\underline{K}}^{(m)\dagger}(u = iu', y')\underline{\underline{K}}^{(m)}(u = iu', y'')$ in eq. (78) (as generally $y' \neq y''$), we have in eq. (79) the product of a matrix with its own Hermitian conjugate $\underline{\underline{K}}^{(m)(u')\dagger}\underline{\underline{K}}^{(m)(u')}$. In the latter case, the resulting matrix is obviously Hermitian and hence diagonalizable according to the spectral theorem. Performing the diagonalization will yield a set of eigenmodes of the system, the associated real eigenvalues being their retrieval efficiencies; the matrix $\underline{\underline{K}}^{(m)(u')\dagger}\underline{\underline{K}}^{(m)(u')}$ is easily shown to be nonnegative-definite, so the eigenvalues are guaranteed to be nonnegative.

8 Conclusion

We have achieved a formal solution for the retrieval efficiency of an ensemble based quantum memory for the case of beam splitter type interaction in the paraxial regime assuming a cylindrically symmetric Gaussian atomic density. We saw in course of the analysis that the dependence of the atomic density on the transverse coordinates resulted in coupling between distinct modes. The theoretical derivation given here provides the foundation necessary for determining the eigenmodes of the system and their efficiencies numerically; carrying out this next step will, among other things, give insight regarding which modes are most efficiently retrieved. The present analysis also uncovered which combinations of physical quantities are determining the operation of the quantum memory.

A Heisenberg equation in a rotating frame

To obtain a slowly varying operator, we define the Schrödinger picture operator $A_{\mathrm{I,S}} \equiv U A_{\mathrm{S}} U^\dagger$ for some unitary operator U to be specified below. The corresponding Heisenberg picture operator is $A_{\mathrm{I,H}} = T^\dagger A_{\mathrm{I,S}} T$, where the time evolution operator T of the system obeys the Schrödinger equation $i\dot{T} = H_{\mathrm{S}} T$. The Heisenberg equation of motion for $A_{\mathrm{I,H}}$ is

$$\frac{dA_{\mathrm{I,H}}}{dt} = i[H_{\mathrm{H}}, A_{\mathrm{I,H}}] + \left(\frac{\partial A_{\mathrm{I,S}}}{\partial t}\right)_{\mathrm{H}}, \tag{80}$$

where $H_{\mathrm{H}} = T^\dagger H_{\mathrm{S}} T$ and subscript 'H' denotes conversion from the Schrödinger picture to the Heisenberg picture. We rewrite the commutator term as

$$[H_{\mathrm{H}}, A_{\mathrm{I,H}}] = T^\dagger [H_{\mathrm{S}}, A_{\mathrm{I,S}}] T = T^\dagger U [U^\dagger H_{\mathrm{S}} U, A_{\mathrm{S}}] U^\dagger T.$$

The second term, that accounts for the explicit time dependence of $A_{\mathrm{I,S}}$, is

$$\begin{aligned}\left(\frac{\partial A_{\mathrm{I,S}}}{\partial t}\right)_{\mathrm{H}} &= T^\dagger \left(\dot{U} A_{\mathrm{S}} U^\dagger + U A_{\mathrm{S}} \dot{U}^\dagger + U \dot{A}_{\mathrm{S}} U^\dagger\right) T \\ &= T^\dagger \left(U U^\dagger \dot{U} A_{\mathrm{S}} U^\dagger + U A_{\mathrm{S}} \dot{U}^\dagger U U^\dagger + U \dot{A}_{\mathrm{S}} U^\dagger\right) T = T^\dagger U \left(\left[-\dot{U}^\dagger U, A_{\mathrm{S}}\right] + \frac{\partial A_{\mathrm{S}}}{\partial t}\right) U^\dagger T,\end{aligned}$$

using $UU^\dagger = 1$ and $\dot{U}U^\dagger = -U\dot{U}^\dagger$. Plugging these expressions into eq. (80), we obtain a Heisenberg equation for $A_{\mathrm{I,H}}$:

$$\frac{dA_{\mathrm{I,H}}}{dt} = T^\dagger U \left(i[U^\dagger H_{\mathrm{S}} U, A_{\mathrm{S}}] - \left[\dot{U}^\dagger U, A_{\mathrm{S}}\right] + \frac{\partial A_{\mathrm{S}}}{\partial t}\right) U^\dagger T = T^\dagger U \left(i\left[H_{\mathrm{I}}, A_{\mathrm{S}}\right] + \frac{\partial A_{\mathrm{S}}}{\partial t}\right) U^\dagger T, \tag{81}$$

where we have defined the rotating frame Hamiltonian $H_{\mathrm{I}} = i\dot{U}^\dagger U + U^\dagger H_{\mathrm{S}} U$. Hence, to get $\frac{dA_{\mathrm{I,H}}}{dt}$ we take the commutator of the original Schrödinger picture operator A_{S} with H_{I}, add the contribution from the explicit time dependence, if any, and convert the result to the Heisenberg rotating frame. The latter conversion amounts to tagging a subscript 'I,H' onto the Schrödinger picture operators.

B Supplemental details of derivations

B.1 The Holstein-Primakoff transformation

The Holstein-Primakoff transformation is a mapping of angular momentum operators to bosonic harmonic oscillator operators. The *exact* Holstein-Primakoff transformation in the case of angular momentum $J = \frac{N_{\mathrm{A}}}{2}$ is

$$J_x = \frac{N_{\mathrm{A}}}{2} - a_{\mathrm{A}}^\dagger a_{\mathrm{A}}, \quad J_+ = \sqrt{N_{\mathrm{A}}}\sqrt{1 - \frac{a_{\mathrm{A}}^\dagger a_{\mathrm{A}}}{N_{\mathrm{A}}}}\, a_{\mathrm{A}}, \quad J_- = \sqrt{N_{\mathrm{A}}}\, a_{\mathrm{A}}^\dagger \sqrt{1 - \frac{a_{\mathrm{A}}^\dagger a_{\mathrm{A}}}{N_{\mathrm{A}}}}. \tag{82}$$

Given that the operators $a_{\mathrm{A}}, a_{\mathrm{A}}^\dagger$ obey the usual harmonic oscillator commutation relations, it can be shown that the operators $J_x, J_\pm$ defined in eqs. (82) will obey the angular momentum commutation relations. From the relation between J_x and the number operator $a_{\mathrm{A}}^\dagger a_{\mathrm{A}}$ found in eq. (82), we see that the fully polarized angular momentum state $|J = \frac{N_{\mathrm{A}}}{2}, M_x = \frac{N_{\mathrm{A}}}{2}\rangle$ must correspond to the vacuum state of the harmonic oscillator under

the Holstein-Primakoff transformation. Assuming that the large ensemble is very close to the fully polarized state $|J=\frac{N_\mathrm{A}}{2}, M_x=\frac{N_\mathrm{A}}{2}\rangle$, we can expand the square roots of operators in eqs. (82) in the small quantity $\frac{a_\mathrm{A}^\dagger a_\mathrm{A}}{N_\mathrm{A}}$:

$$J_x = N_\mathrm{A}(\frac{1}{2} - \frac{a_\mathrm{A}^\dagger a_\mathrm{A}}{N_\mathrm{A}}), \quad J_+ = \sqrt{N_\mathrm{A}}(1 - \frac{1}{2}\frac{a_\mathrm{A}^\dagger a_\mathrm{A}}{N_\mathrm{A}} + \ldots)a_\mathrm{A}, \quad J_- = \sqrt{N_\mathrm{A}}a_\mathrm{A}^\dagger(1 - \frac{1}{2}\frac{a_\mathrm{A}^\dagger a_\mathrm{A}}{N_\mathrm{A}} + \ldots). \tag{83}$$

B.2 The Laporte selection rule

Consider two states $|x\rangle, |y\rangle$ that are parity eigenstates $\pi|x\rangle = \varepsilon|x\rangle, \pi|y\rangle = \varepsilon|y\rangle$ with the same eigenvalue ε, where the eigenvalue is either $\varepsilon = \pm 1 \in \mathbb{R}$ from the fact that $\pi^2 = 1$; we then have, noting that the dipole operator $\vec{D}$ is odd under a parity transformation $\pi^\dagger \vec{D}\pi = -\vec{D}$,

$$\langle x|\vec{D}|y\rangle = \varepsilon^2\langle x|\vec{D}|y\rangle = \langle x|\varepsilon^*\vec{D}\varepsilon|y\rangle = \langle x|\pi^\dagger\vec{D}\pi|y\rangle = -\langle x|\vec{D}|y\rangle,$$

proving the Laporte rule that $\langle x|\vec{D}|y\rangle = 0$ for states of the same parity.

B.3 Transformation to rescaled time

All of the following are Heisenberg rotating frame operators. We introduce a rescaled time $\tau = t - \frac{z}{c}$ and define new annihilation operators that depend on this variable $a'_{\mathrm{X},\tilde{m}}(z,\tau) \equiv a_{\mathrm{X},\tilde{m}}(z, t = \tau + \frac{z}{c})$, where X=L,A. Taking the derivative and using the chain rule yields

$$\begin{aligned}\frac{\partial}{\partial z}a'_{\mathrm{L},\tilde{m}}(z,\tau) &= \left.\frac{\partial a_{\mathrm{L},\tilde{m}}(z,t)}{\partial z}\right|_{t=\tau+\frac{z}{c}} + \left.\frac{\partial a_{\mathrm{L},\tilde{m}}(z,t)}{\partial t}\right|_{t=\tau+\frac{z}{c}} \cdot \frac{\partial(\tau+\frac{z}{c})}{\partial z} \\ &= \left.\frac{\partial a_{\mathrm{L},\tilde{m}}(z,t)}{\partial z}\right|_{t=\tau+\frac{z}{c}} + \frac{1}{c}\left.\frac{\partial a_{\mathrm{L},\tilde{m}}(z,t)}{\partial t}\right|_{t=\tau+\frac{z}{c}},\end{aligned}$$

since now z, τ are considered to be the independent variables. Then, by evaluating the equation of motion for $a_\mathrm{L}(z,t)$, eq. (37),[22] at $t = \tau + \frac{z}{c}$, it is seen to be equivalent to a differential equation for $a'_\mathrm{L}(z,\tau)$ in position z

$$\frac{\partial}{\partial z}a'_{\mathrm{L},\tilde{m}}(z,\tau) = -\frac{i}{c}[a'_{\mathrm{L},\tilde{m}}(z,\tau), H_\mathrm{Ai}],$$

in light of the definition of $a'_{\mathrm{L},\tilde{m}}(z,\tau)$.

With regard to $a'_{\mathrm{A},\tilde{m}}(z,\tau)$, we simply note that the chain rule implies

$$\frac{\partial}{\partial\tau}a'_{\mathrm{A},\tilde{m}}(z,\tau) = \left.\frac{\partial a_{\mathrm{A},\tilde{m}}(z,t)}{\partial z}\right|_{t=\tau+\frac{z}{c}} \cdot \frac{\partial z}{\partial\tau} + \left.\frac{\partial a_{\mathrm{A},\tilde{m}}(z,t)}{\partial t}\right|_{t=\tau+\frac{z}{c}} \cdot \frac{\partial t}{\partial\tau} = \left.\frac{\partial a_{\mathrm{A},\tilde{m}}(z,t)}{\partial t}\right|_{t=\tau+\frac{z}{c}},$$

again considering z, τ to be the independent variables.

[22] Note that the notations $\frac{d}{dt}, \frac{\partial}{\partial t}$ are in fact equivalent in the present case.

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