Quantum noise for Faraday light–matter interfaces

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Abstract

In light–matter interfaces based on the Faraday effect, quite a number of quantum information protocols have been successfully demonstrated. In order to further increase the performance and fidelities achieved in these protocols, a deeper understanding of the relevant noise and decoherence processes needs to be gained. In this paper, we provide for the first time a complete description of the decoherence from spontaneous emission. We derive from first principles the effects of photons being spontaneously emitted into unobserved modes. Our results relate the resulting decay and noise terms in effective equations of motion for collective atomic spins and the forward-propagating light modes to the full atomic level structure. We illustrate and apply our results to the case of a quantum memory protocol. Our results can be applied to any alkali atoms, and the general approach taken in this paper can be applied to light–matter interfaces and quantum memories based on different mechanisms.

1. Introduction

The strong and coherent interaction of light with matter is a prerequisite for many approaches towards quantum information technologies. In particular, long-distance quantum communication relies on efficient light–matter interfaces which allow for a coherent transfer of quantum information from light to stationary carriers and back [1]. Also architectures for quantum computations based on light will depend on efficient light–matter interfaces for buffering and storing quantum information carried by light.

Optically dense atomic ensembles have proven to be a particularly promising technique for achieving strong coherent light–matter interaction. Thereby, quantum states of propagating pulses of light are mapped onto states of collective atomic (pseudo) spins. This essentially requires a mechanism to fiducially and reversibly convert photons into ground state spin excitations with long coherence times. Several mechanisms have been explored for realizing such an atomic ensemble-based light–matter interface, e.g. Raman transitions, electromagnetically induced transparency [2–4], spin echoes [5] and Faraday rotation; see [6–8] for comprehensive reviews. The Faraday effect—which will be the topic of this paper—consists in the rotation of light polarization depending on

atomic spin polarization and vice versa. In the context of the light–matter interface, it has been successfully used to create squeezed states for spin [9, 10] and light [11, 12], entangled states of collective atomic ensembles [13, 14], quantum teleportation from states of light to atoms [15], and quantum memory for light [16, 17]. Apart from the light–matter interface, the Faraday interaction has important applications also in continuous nondemolition measurement of atomic spin ensembles [18, 19], quantum-state control/tomography [20] and magnetometry [21–23].

These protocols can all be understood in terms of a rather simple model of the Faraday interaction. In this model, atoms are assumed to have a spin $\frac{1}{2}$ ground state and a spin $\frac{1}{2}$ excited state [24]. Far off resonant light probing this dipole-allowed transition will then experience a polarization rotation due to the dipole selection rules for such a $\frac{1}{2} \rightarrow \frac{1}{2}$ transition. By the same effect, the atomic polarization will be rotated by light. For far off resonant light, atoms will be only very weakly excited such that this birefringence of the atomic medium can be understood as being due to the polarizability of the atomic ground states alone [25, 26]. The *coherent* interaction of light with atoms arises from the *real* part of the atomic polarizability. By the Kramers–Kronig relation, it is clear that the corresponding imaginary part will necessarily be non-zero
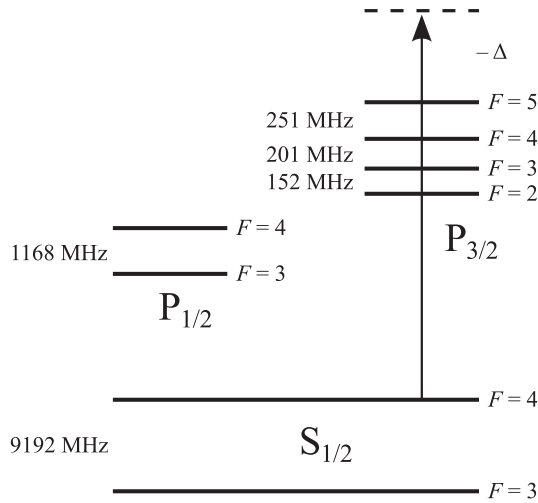
Figure 1. Level structure of caesium. We assume that our laser light is blue detuned by an amount $-\Delta$ from the transition $F = 4 \to F' = 5$.

and add some *incoherent* effects to the dynamics. Sure enough, these effects can be understood as resulting from spontaneous emission events. They will cause both decoherence of light (absorption) and decoherence of atoms (spin decay). While both effects can be kept small as compared to the coherent dynamics, they are ultimately unavoidable on a fundamental level.

Both decoherence effects—light losses and spin decay—can be included on the level of the simple spin $\frac{1}{2}$ model giving a qualitative understanding of the tradeoff between coherent and incoherent contributions to the light–matter interaction [27–29]. However, in view of the experimental achievements, it will become increasingly important to further gain a detailed and more quantitative understanding of these effects and the resulting tradeoff. A theoretical description based on a realistic atomic level scheme, such as the one shown in figure 1, and starting from first principles has not been given so far. In this paper we will provide such a description.

From the standard dipole interaction of a single multi-level atom with the three-dimensional electromagnetic field, we consistently derive effective equations of motion for the collective ground state spin and the forward-propagating light modes relevant to the description of the light–matter interface. In the derivation, we will keep track of the effects of events where photons are emitted to any other than the forward direction. This eventually adds decay and noise terms to the equations of motions whose origin and dependence on the details of the atomic level structure are fully understood and explained for the first time in this paper. Important steps towards a deeper understanding have been taken before in [[30]. The insight gained by the detailed knowledge of losses and decoherence will enable an optimized operation of the light–matter interface based on the Faraday interaction, and therefore contribute to boost its performance and fidelity.

The paper is organized as follows. In section 2, we will briefly summarize the Faraday rotation effect and connect it to the atomic polarizability. In section 3, we give a description of the coherent part of the dynamics, deriving effective equations

of motion for collective atomic spins and forward-propagating modes. We will write down and solve these equations of motion also for canonical operators for light and atoms, and illustrate the working principle of a quantum memory protocol. In section 4, we arrive at the main results of this paper and include in our derivation spontaneous emission and decoherence starting from first principles. The resulting equations of motion can again be expressed and integrated in terms of canonical operators. As an illustration we apply the result to the quantum memory protocol. In section 5, we provide a self-contained executive summary of our results. Readers just interested in applying the correct model for decoherence in the Faraday based light–matter interface can directly consult this section.

2. Effective interaction

We consider alkali atoms, which have a single electron outside a closed shell. This electron will be in an $S_{1/2}$-state, giving two stable ground levels: a higher and a lower hyperfine manifold, describing respectively a state where the electronic spin $J = \frac{1}{2}$[3] is parallel to the nuclear spin I giving a total of $F = I + \frac{1}{2}$ and a state where the spins are pointing opposite giving $F = I - \frac{1}{2}$; see figure 1 where we show the level structure of the specific case of Cs where $I = \frac{7}{2}$ so that $F = 3, 4$. There will be two dipole-allowed transitions to the excited states $P_{1/2}$ and $P_{3/2}$, each of which consists of several hyperfine states with spin F' (primed variables always refer to electronically excited states). In this paper, we will mainly consider the case where atoms are initially prepared in the $S_{1/2}(F = I + \frac{1}{2})$-state and are driven on the D_2-transition to $P_{3/2}$ as indicated in figure 1 for the example of ^{133}Cs.

In the standard dipole and rotating wave approximation, the coupling of light to atoms is given by

$$H_{\text{int}} = -(\mathbf{d}^{(+)}\mathbf{E}^{(-)} + \mathbf{d}^{(-)}\mathbf{E}^{(+)}),$$

where $\mathbf{E}^{(\pm)}$ and $\mathbf{d}^{(\pm)}$ are the positive/negative frequency component of, respectively, the electric field $\mathbf{E} = \mathbf{E}^+ + \mathbf{E}^-$ and the dipole moment operator $\mathbf{d} = \mathbf{d}^+ + \mathbf{d}^-$. We will be interested here in the interaction with far off-resonant light only. In this case, the excited states will be only very weakly populated. In the limit of low saturation, excited states can be adiabatically eliminated (appendix A), and the light–atom interaction is described by the effective Hamiltonian

$$H_{\text{int}}^{\text{eff}} = \mathbf{E}^{(-)}\boldsymbol{\alpha}\,\mathbf{E}^{(+)}. \tag{1}$$

We have here introduced the polarizability tensor operator

$$\boldsymbol{\alpha} = -\sum_{F'}\frac{P_g\mathbf{d}P_{F'}\mathbf{d}P_g}{\Delta_{F'}}, \tag{2}$$

where the projection operators are defined as

$$P_F = \sum_m |F, m\rangle\langle F, m|, \qquad P_g = \sum_F P_F, \tag{3}$$

$$P_{F'} = \sum_{m'} |F', m'\rangle\langle F', m'|, \qquad P_e = \sum_{F'} P_{F'}, \tag{4}$$

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[3] In this paper, we put $\hbar = c = 1$.

such that $P_g + P_e = 1$. Here $\Delta_{F'}$ is the detuning of the light from the $S_{1/2}F \rightarrow P_{3/2}F'$ transition; see figure 1. The polarizability operator $\boldsymbol{\alpha}$ in (2) has to be understood as a 3×3 matrix whose Cartesian components are given by $\boldsymbol{\alpha}_{ij} = -\sum_{F'} P_g \mathbf{d}_i P_{F'} \mathbf{d}_j P_g / \Delta_{F'}$. Each of these components is an operator acting exclusively within the subspace of ground states $S_{1/2}(F = I \pm \frac{1}{2})$.

The effective interaction in (1) describes second order processes where a photon is absorbed and re-emitted, while the atom makes a transition from a ground to an excited state and back again to a ground state. In such a process, the initial and final ground states can either have the same or different total spin $F = I \pm \frac{1}{2}$. In order to describe processes involving transitions within the F-subspace, we introduce $H_{FF} = P_F H_{\text{int}}^{\text{eff}} P_F$. Transitions between $F \leftrightarrow F - 1$ are described by $H_{FF-1} = P_{F-1} H_{\text{int}}^{\text{eff}} P_F + P_F H_{\text{int}}^{\text{eff}} P_{F-1}$. In general such second order processes could also give rise to changes of F by 2, but since we are considering $S_{1/2}$ ground levels there are only two ground levels, e.g. as shown in figure 1 for Cs $F = 3, 4$.

Consider first transitions within one F-subspace as described by the Hamiltonian H_{FF}. Due to the fact that we have conservation of angular momentum in the interaction, we can decompose the polarizability operator $\boldsymbol{\alpha}_{FF} = P_F \boldsymbol{\alpha} P_F$ into its tensor parts [26, 20], namely

$$\boldsymbol{\alpha}_{FF} = -\frac{d_0^2}{\Delta}(a_0 + \mathrm{i}a_1 \mathbf{j} \times + a_2 \mathbf{Q}). \tag{5}$$

Here the dipole matrix element is defined as $d_0^2 = (2J' + 1)|\langle J'\|d\|J\rangle|^2$ with J and J' being the electronic angular momenta of the ground and excited states, respectively (see appendix B), and Δ is the detuning from resonance as shown in figure 1. The three terms are the scalar, vector and second rank tensor part, respectively. $\mathbf{j}$ is the spin operator for the total ground state spin, that is, $\mathbf{j}^2|F, m\rangle = F(F + 1)|F, m\rangle$. In the vector component, we use here the short-hand notation

$$\mathbf{j} \times = \begin{pmatrix} 0 & j_z & j_y \\ j_z & 0 & -j_x \\ -j_y & j_x & 0 \end{pmatrix},$$

which is equivalent to taking the cross product of $\mathbf{j}$ with the vector to the right and then the scalar product with the vector to the left. One can use vector product properties in order to transform the expression as follows: $\mathrm{i}\mathbf{E}^{(-)} \cdot [\mathbf{j} \times \mathbf{E}^{(+)}] = -\mathrm{i}\mathbf{j} \cdot [\mathbf{E}^{(-)} \times \mathbf{E}^{(+)}]$. The latter form could be more convenient since it is just a scalar product of the spin and a vector for light which resembles the Stokes operator characterizing the polarization of light, as will be discussed in more detail below. The second rank tensor part $\mathbf{Q}$ is defined componentwise as

$$Q_{ij} = -(j_i j_j + j_j j_i) + \delta_{ij} \tfrac{2}{3} \mathbf{j}^2.$$

The tensor decomposition of $\boldsymbol{\alpha}$ is given in detail in appendix B where we also give the explicit expressions for the real coefficients a_0, a_1 and a_2 and their dependence on the detuning. In figure 2, we show these coefficients for the case of Cs. Note in particular that the second rank tensor polarizability vanishes for large detunings.
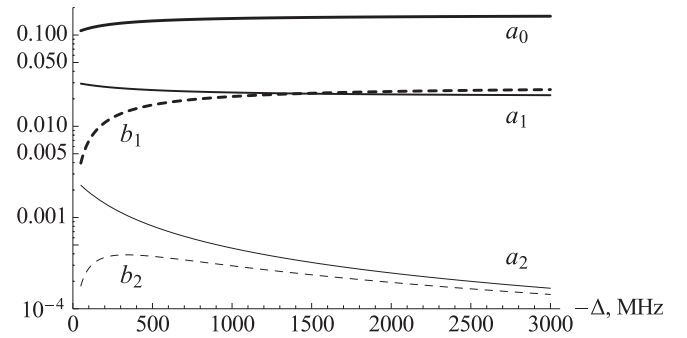

Figure 2. a and b coefficients as functions of detuning $-\Delta$ in MHz for caesium with $F = 4$; a_0 (thick), a_1 (medium), a_2 (thin), b_1 (thick dashed), b_2 (thin dashed). In the limit of high detuning, we obtain $a_0 \rightarrow \frac{1}{6}$, $a_1 \rightarrow \frac{1}{48}$, $b_1 \rightarrow \frac{1}{16\sqrt{5}}$ and $\{a_2, b_2\} \rightarrow 0$.

For the other case where the final spin state has a different total spin than the initial state, we have

$$\boldsymbol{\alpha}_{FF\pm 1} = -\frac{d_0^2}{\Delta}(b_1 \mathbf{T}^{(1)} + b_2 \mathbf{T}^{(2)}). \tag{6}$$

The coefficients b_1 and b_2 and the irreducible tensor operators $\mathbf{T}^{(1)}$ and $\mathbf{T}^{(2)}$ are given in appendix B. We do not have a scalar tensor component here because in these processes the spin state is changed such that the corresponding Hamiltonian cannot have a component proportional to the identity operator, i.e. a scalar component. In figure 2, we show b_1 and b_2 as a function of the detuning. Note that b_1 and b_2 have the same scale as a_1 and a_2, respectively.

The effective interaction Hamiltonian (1) describes elastic Rayleigh and inelastic Raman scattering of light on atoms. It is a Hermitian operator and as such gives rise to a fully coherent evolution for the overall system comprised of the atom and the quantized electromagnetic field. The quantum coherent effects resulting from this effective interaction have been widely used in the field of quantum information, both theoretical and experimental [16]. Note, however, that within the regime of its validity—that is far below saturation of excited states—the interaction Hamiltonian (1) *still* contains and correctly describes the effects of spontaneous emission on both atom and light. If light is treated as a reservoir and we trace over the random position of the atoms we will show that we can describe spontaneous emission which leads to decoherence effects within the ground state levels. Vice versa, the atom (or an ensemble of atoms) can provide an effective absorptive medium for light. In the following sections, we will first treat the *coherent interaction* and then include the *decoherence effects* resulting from the effective interaction Hamiltonian (1).

3. Coherent interaction

3.1. Interaction with a single atom

We consider now a situation as shown in figure 3. A cloud of atoms interacts with an incoming pulse of light described by the forward-propagating electric field $\mathbf{E}_F$ and we are interested in the coherent evolution of the spin of atoms and the forward-propagating field modes [31, 32]. The total electric field
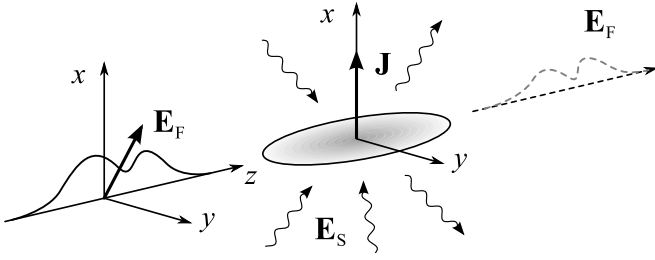
Figure 3. The spin-polarized atomic sample interacts with the off-resonant light field $\mathbf{E}_F$ and non-forward modes $\mathbf{E}_S$ in vacuum states.

$\mathbf{E} = \mathbf{E}_F + \mathbf{E}_S$ will be the sum of forward-propagating modes and the remaining, non-forward-propagating modes $\mathbf{E}_S$, cf figure 3. These modes will be treated as a reservoir in the later sections and give rise to the decoherence effects to be discussed there.

For the moment, we will thus restrict ourselves to light propagating along the z-direction. In principle, the forward modes will also experience some spreading [33, 34], but here we take the opening angle to be roughly zero, and use a one-dimensional description of $\mathbf{E}_F(z)$. The separation into a forward-propagating mode $\mathbf{E}_F$ described by a one-dimensional equation of motion and a set of non-forward modes $\mathbf{E}_S$ is further justified in [30, 6] and is valid if the Fresnel number of the ensemble is much larger than unity. In this model the field is described in terms of position-dependent annihilation (creation) operators, defined by

$$a_\sigma(z, t) = \int \frac{\mathrm{d}k}{2\pi} a_{k\sigma}(t)\, \mathrm{e}^{\mathrm{i}kz}, \tag{7}$$

where $\sigma = x, y$ labels the transverse polarizations. These operators obey $[a_\sigma(z, t), a_{\sigma'}^\dagger(z', t)] = \delta_{\sigma\sigma'}\delta(z - z')$. In terms of $a_x(z, t)$ and $a_y(z, t)$, the Stokes operators $S_i(z, t)$ can be introduced:

$$S_x = \tfrac{1}{2}\big(a_x^\dagger a_x - a_y^\dagger a_y\big), \tag{8}$$

$$S_y = \tfrac{1}{2}\big(a_x^\dagger a_y + a_y^\dagger a_x\big), \tag{9}$$

$$S_z = \frac{1}{2i}\big(a_x^\dagger a_y - a_y^\dagger a_x\big), \tag{10}$$

$$S_0 = \tfrac{1}{2}\big(a_x^\dagger a_x + a_y^\dagger a_y\big), \tag{11}$$

which are a convenient tool to describe the polarization state of light propagating along the z-direction. They obey the commutation relations for an angular momentum density $[S_i(z), S_j(z')] = \mathrm{i}\delta(z - z')\epsilon_{ijk}S_k(z)$.

Each part in the decomposition (5) of the atomic polarizability operator will give rise to a term in the effective interaction Hamiltonian, such that $H_{FF} = H^{(0)} + H^{(1)} + H^{(2)}$ corresponding to the scalar, vector and tensor parts, respectively. These terms can be conveniently expressed and interpreted in terms of Stokes operators. The scalar Hamiltonian—the tensor-0 term—is

$$H^{(0)} = ga_0 S_0. \tag{12}$$

Here we have defined $g = -\frac{d_0^2}{\Delta}\frac{\omega_0}{\epsilon_0 A}$ where Δ is the detuning from resonance as shown in figure 1, ω_0 the atomic transition frequency and A the beam cross section. It can be interpreted as a Stark shift, which equally shifts all atomic energy levels proportional to the light intensity. Conversely it can also be interpreted as an equal shift of the frequency for all light modes (off-resonant with a detuning Δ), that is, as a new index of refraction seen by light.

The vector Hamiltonian, which for our purpose is the most interesting part, is

$$H^{(1)} = ga_1 S_z j_z. \tag{13}$$

S_z describes the circularity of light and j_z the z-component of the atomic spin. In the interaction, the atomic spin is rotated around the z-axis by an amount proportional to S_z. Likewise, the Stokes vector is rotated about the z-axis by an amount proportional to j_z. This is a circular birefringence effect and this interaction gives us the desired Faraday interaction.

Finally we also have a complicated tensor Hamiltonian

$$H^{(2)} = -ga_2\big(S_x\big(j_x^2 - j_y^2\big) + S_y\{j_x, j_y\} + 2S_0\big(3j_z^2 - \mathbf{j}^2\big)/3\big). \tag{14}$$

This amounts to a dynamical Stark shift. The effect vanishes for large detunings since the coefficient a_2 goes to zero as shown in figure 2. It can be interesting for coherent dynamics in some experiments for tomography and collective squeezing purposes [35, 21, 14, 32, 33]. We keep it for the calculation of spontaneous emission, and neglect it for simplicity in the coherent dynamics.

3.2. Interaction with an atomic ensemble

So far we have considered a single atom. We will now assume that light is interacting with a cloud of N_a atoms of length L and constant atomic density ρ as shown in figure 3. For the atomic spin, we therefore introduce the continuous spin density operators, which are a sum of the single atom (j^a) angular momentum operators evaluated at their respective positions:

$$j_k(z) = \sum_{a=1}^{N_a} \delta(z - z_a) j_k^a, \qquad k = x, y, z. \tag{15}$$

Then, the commutator is

$$[j_m(z), j_n(z')] = \sum_{a,b}^{N_a} \delta(z - z_a)\delta(z' - z_b)\big[j_m^a, j_n^b\big]$$
$$= \mathrm{i}\,\epsilon_{mnk}\, j_k(z)\delta(z - z'), \tag{16}$$

in perfect analogy to the commutation relation for the Stokes operators. For details regarding the definition of these continuous spin operators, in particular concerning transverse effects, we refer to [6, 30].

We are now in a position to describe the coherent interaction of the forward-propagating light modes with the atomic ensemble. We ignore here the contributions from $H^{(0)}$ since it merely accounts for the overall refractive index. The effective interaction can then be written as

$$H_{\mathrm{coh}}^{\mathrm{eff}} = g\int_0^L (\boldsymbol{\gamma} \cdot \mathbf{S})(z, t)\,\mathrm{d}z, \tag{17}$$

where $\boldsymbol{\gamma} = (-a_2(j_x^2 - j_y^2), -a_2\{j_x, j_y\}, a_1 j_z)$ is a vector describing atomic polarization. In appendix C, we show how the time derivative of the light operators can be transformed to a derivative in position (z), such that when we form the Heisenberg equation of motion we get from the relation $[S_i(z), S_j(z')] = i\delta(z - z')\epsilon_{ijk}S_k(z)$ that

$$\frac{\partial}{\partial z}\mathbf{S}(z, t) = g(\boldsymbol{\gamma} \times \mathbf{S})(z, t). \tag{18}$$

If we replace the operators in $\boldsymbol{\gamma}$ with their expectation values, we can see that in the interaction, the Stokes operator $\mathbf{S}$ gets rotated about the vector $\boldsymbol{\gamma}$. Written out in full detail the equation reads

$$\frac{\partial}{\partial z}\begin{pmatrix} S_x \\ S_y \\ S_z \end{pmatrix}(z, t)$$
$$= g\begin{pmatrix} 0 & -a_1 j_z & -a_2\{j_x, j_y\} \\ a_1 j_z & 0 & a_2(j_x^2 - j_y^2) \\ a_2\{j_x, j_y\} & -a_2(j_x^2 - j_y^2) & 0 \end{pmatrix}\begin{pmatrix} S_x \\ S_y \\ S_z \end{pmatrix}(z, t). \tag{19}$$

These equations have first been derived by Julsgaard [31]. The rotation of $\mathbf{S}$ can be seen to be composed of a big rotation (proportional to a_1) around the z-axis and proportional to the atomic spin along z and a small rotation (proportional to a_2) in the (x, y) plane by an angle which depends on the relative angle between the mean atomic spin and the Stokes vector. If we only consider the a_1-terms above, and assume the x component of the Stokes vector to be much larger than the other two projections (as is the case for x polarized light), and assuming the rotation angle to be small we arrive at the effective equation of motion

$$\frac{\partial}{\partial z}\begin{pmatrix} S_y \\ S_z \end{pmatrix}(z, t) = ga_1 S_x \begin{pmatrix} j_z \\ 0 \end{pmatrix}. \tag{20}$$

These simpler equations show us that S_z is conserved in the interaction, while S_y receives some contribution from the spin component j_z.

A similar analysis can be performed for the coherent dynamics of the atoms. We will write the coherent Hamiltonian as

$$H_{\text{coh}} = g[\boldsymbol{\gamma} \cdot \mathbf{S} + \gamma_0 S_0], \tag{21}$$

where $\boldsymbol{\gamma}$ is given as above and $\gamma_0 = -a_2 j_z^2$. From this we can determine the coherent evolution of the spin vector $\mathbf{j}$:

$$\frac{\partial}{\partial t}\begin{pmatrix} j_x \\ j_y \\ j_z \end{pmatrix} = g\left[\begin{pmatrix} a_2\{j_y, j_z\} & -a_2\{j_x, j_z\} & -a_1 j_y \\ a_2\{j_x, j_z\} & a_2\{j_y, j_z\} & a_1 j_x \\ -2a_2\{j_x, j_y\} & 2a_2(j_x^2 - j_y^2) & 0 \end{pmatrix}\begin{pmatrix} S_x \\ S_y \\ S_z \end{pmatrix}\right.$$
$$\left. + a_2\begin{pmatrix} -\{j_y, j_z\} \\ \{j_x, j_z\} \\ 0 \end{pmatrix}S_0\right]. \tag{22}$$

While the overall form of these equations is rather complicated, we can get valuable insight by treating the different terms separately [31]. For instance if we only consider a_1-terms as it was done with light and also assume small Faraday rotations of atomic spins with strong polarization along x then we get the equation

$$\frac{\partial}{\partial t}\begin{pmatrix} j_y \\ j_z \end{pmatrix}(z, t) = ga_1 j_x \begin{pmatrix} S_z \\ 0 \end{pmatrix}, \tag{23}$$

in perfect analogy to the effective equations of motion for light found above, cf (19) and (20). j_z is unaltered in the interaction, whilst j_y gets a contribution from S_z.

3.3. Canonical operators

In order to further emphasize the similarity in the atom and light evolution, we will consider them on a more equal footing by introducing canonical variables. From the definition of the Stokes operators for light (8)–(11), one can see that for a large classical field polarized in the x-direction and sufficiently small angles of Faraday rotations the S_y and S_z components are proportional to the X and P quadratures of the weak quantum field in the orthogonal polarization (y):

$$S_y(z, t) = \sqrt{\langle S_x \rangle}X_L(z, t), \tag{24}$$

$$S_z(z, t) = \sqrt{\langle S_x \rangle}P_L(z, t). \tag{25}$$

The quadratures introduced here obey the standard commutation relation $[X_L(z, t), P_L(z', t)] = i\delta(z - z')$.

Next we consider canonical spin densities. Experimentally two different configurations are typically employed. Either the light polarization is oriented along the same axis as the spins of the atoms (x) or the light is orthogonal to the atomic polarization (y). For simplicity, we will restrict our discussion of the general method to the case where the light is polarized along the polarization of the spins. The argument we give can however easily be generalized also to the other orientations and for completeness we give the results for both orientations below. Let us assume that the atomic spin is polarized along the x-axis. Then, the commutation relation (16) averaged over the random positions of the atoms reads

$$\overline{[j_y(z, t), j_z(z', t)]} = i\langle j_x \rangle\delta(z - z'), \tag{26}$$

where $\langle j_x \rangle = n\langle j_x^a \rangle$ with n being the average linear density of atoms. The field-like canonical variables for the spin subsystem,

$$X_A(z, t) = \frac{j_y(z, t)}{\sqrt{\langle j_x \rangle}}, \qquad P_A(z, t) = \frac{j_z(z, t)}{\sqrt{\langle j_x \rangle}}, \tag{27}$$

obey the canonical commutation relation

$$\overline{[X_A(z, t), P_A(z', t)]} = i\delta(z - z'). \tag{28}$$

Now we can write the equations of motion for light and atoms given by (20) and (23) in terms of the canonical operators. Upon introducing the coupling constant $\kappa = ga_1\sqrt{\frac{F}{2}N_a N_p}$ where N_p is a number of photons in the driving field, one obtains

$$\frac{\partial}{\partial z}\begin{pmatrix} X_L \\ P_L \end{pmatrix}(z, t) = \frac{\kappa}{\sqrt{LT}}\begin{pmatrix} P_A \\ 0 \end{pmatrix},$$
$$\frac{\partial}{\partial t}\begin{pmatrix} X_A \\ P_A \end{pmatrix}(z, t) = \frac{\kappa}{\sqrt{LT}}\begin{pmatrix} P_L \\ 0 \end{pmatrix}. \tag{29}$$

As opposed to the initial set of equations (19) and (22) these simplified equations are not coupled to each other, since we have dropped the term $H^{(2)}$ of the interaction Hamiltonian (14) due to the second rank tensor polarizability. This is justified

in the limit of large detuning. Their solution can be expressed in the form of input–output relations for collective variables which are introduced by

$$X_L^{\text{in(out)}} = \frac{1}{\sqrt{T}} \int_T \mathrm{d}t \, X_L(0(L), t), \tag{30}$$

$$X_A^{\text{in(out)}} = \frac{1}{\sqrt{L}} \int_L \mathrm{d}z \, X_A(z, 0(T)). \tag{31}$$

Here T is the duration of the light pulse and L is the length of the atomic sample. The same definitions are applied to the conjugated canonical variables of light and atoms. Finally, the input–output relations obtained from the equations of motion (29) read

$$\begin{pmatrix} X_L^{\text{out}} \\ P_L^{\text{out}} \end{pmatrix} = \begin{pmatrix} X_L^{\text{in}} \\ P_L^{\text{in}} \end{pmatrix} + \kappa \begin{pmatrix} P_A^{\text{in}} \\ 0 \end{pmatrix}, \tag{32}$$

$$\begin{pmatrix} X_A^{\text{out}} \\ P_A^{\text{out}} \end{pmatrix} = \begin{pmatrix} X_A^{\text{in}} \\ P_A^{\text{in}} \end{pmatrix} + \kappa \begin{pmatrix} P_L^{\text{in}} \\ 0 \end{pmatrix}. \tag{33}$$

These relations constitute the foundation for the light–matter interface based on the Faraday rotation; see [6].

A specific example for the application of the Faraday interaction is the possibility of creating a memory for light. The quantum state of a propagating pulse of light is thereby mapped onto the collective spin of a cloud of atoms. The mapping protocol applied in [16] works as follows. The light pulse first interacts with the atomic cloud such that the evolution is described by (32) and (33). One then measures X_L^{out} via homodyne detection and uses the result for a feedback on the atomic spin. The feedback should subtract the measurement outcome for X_L^{out} from P_A^{out} with a gain ν. One can show that the evolution of the atoms is then described by the relations [16]

$$X_A^{\text{out}} = X_A^{\text{in}} + \kappa P_L^{\text{in}}, \tag{34}$$

$$P_A'^{\text{out}} = P_A^{\text{out}} - \nu X_L^{\text{out}} = P_A^{\text{in}}(1 - \kappa\nu) - \nu X_L^{\text{in}}. \tag{35}$$

Assuming $\kappa = \nu = 1$ we have

$$X_A^{\text{out}} = X_A^{\text{in}} + P_L^{\text{in}}, \tag{36}$$

$$P_A^{\text{out}} = -X_L^{\text{in}}, \tag{37}$$

meaning that we have stored the light quadratures in atoms

$$\langle X_A^{\text{out}} \rangle = \langle P_L^{\text{in}} \rangle, \tag{38}$$

$$\langle P_A^{\text{out}} \rangle = -\langle X_L^{\text{in}} \rangle. \tag{39}$$

The quality of the mapping can be characterized by the fidelity $\mathcal{F}$. Assuming a random set of coherent states as the input state, the fidelity is found to be [38]

$$\mathcal{F} = \left(\tfrac{1}{2} + \Delta X_A^{2,out}\right)^{-\frac{1}{2}} \times \left(\tfrac{1}{2} + \Delta P_A^{2,out}\right)^{-\frac{1}{2}}. \tag{40}$$

Assuming light and atoms to be initially shot noise limited we get a fidelity of $\mathcal{F} = \sqrt{2/3} \approx 82\%$. The fidelity is limited by the initial noise of the X_A^{in} spin component which was not cancelled during the mapping process. If this quadrature would be squeezed before the pass of the light pulse, the fidelity can approach unity.

It is known from earlier works [6, 27] that the coupling constant is related to the optical depth by $\kappa^2 = \mathrm{d}\,\eta$, where the optical depth is $d = N_a\sigma/A$, the scattering cross section is $\sigma = 3\lambda^2/2\pi$ and the transversal area of the atomic sample is A. The coefficient $\eta \sim N_p/\Delta^2$ was called atomic depumping as it is closely related to a spin decay rate. The fact that the coupling constant depends on the decoherence rate is a very important observation. It means that for the great variety of quantum protocols based on such light–matter interaction a good performance is a compromise between the interaction strength and the decoherence. For example, in order to reach a value of $\kappa = 1$ in the memory protocol for a given optical depth d, a nonzero atomic depumping $\eta \sim 1/d$ is required. By working with an ensemble with sufficiently large optical depth $d \gg 1$, the atomic depumping can be made small and this is the basis of the protocols for quantum interfaces based on the Faraday interaction which have been implemented in practice. In reality, however, there is a limit to how large an optical depth that can be obtained in practice. Hence, there will always be some atomic depumping for any implementation of a quantum information protocol. A proper optimization of such protocols requires knowledge of how exactly the depumping η is related to the spin decay rates. In the next section, we give the answer to this question.

4. Decoherence

4.1. Example of decoherence

Before presenting the rigorous treatment of the general case of spontaneous emission for multilevel atoms, let us include decay in a phenomenological basis and examine how an arbitrary decay would affect the atom–light interaction. We expect that the spins and light would decay according to the Heisenberg–Langevin equations

$$\frac{\partial}{\partial z} a_\mu = -\frac{\gamma_\mu}{2L} a_\mu + F_{L,\mu}, \qquad \mu = \{x, y\}, \tag{41}$$

$$\frac{\partial}{\partial t} j_i = -\frac{\Gamma_i}{T} j_i + F_i, , \qquad i = \{x, y, z\}. \tag{42}$$

Since the decay process for light is due to scattering of the photons out of the mode of interest, we will see below that the added Langevin noise is simpler for light than the one for atoms. For light, the noise is just the minimal noise required to preserve the commutation relation for the field. The noise is delta correlated in time and space and has a vanishing mean value

$$\langle F_{L,\mu}(z, t) F_{L,\nu}^\dagger(z', t') \rangle = \frac{\gamma_\mu}{L} \delta_{\mu\nu} \delta(t - t') \delta(z - z'), \tag{43}$$

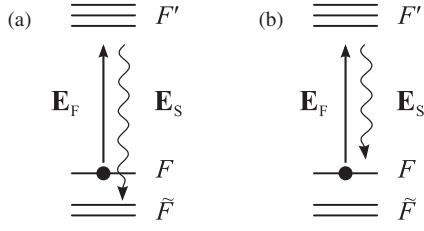$$\langle F_{L,\mu}(z, t) \rangle = 0. \tag{44}$$

Figure 4. Scheme of two possible decay processes.

The atomic spin decays in a number of ways. First, it can decay to another hyperfine level $(F - 1)$ and disappear from the interaction as it is shown in figure 4(a). This is the same process which happens to photons. Another way for an atom to decay is to decay to the same hyperfine level as shown in figure 4(b). This results in essentially a random rotation of the original spin and it creates extra noise. We will derive the correlators of the spin noise operators in the following section, as well as the corresponding decay rates.

Using the definition of canonical variables for atoms (27), we obtain the following expressions for the decay in canonical variables

$$\frac{\partial}{\partial t} X_A = \frac{1}{\sqrt{\langle j_x \rangle}} \frac{\partial}{\partial t} j_y - \frac{1}{2} \frac{j_y}{\sqrt{\langle j_x \rangle}} \cdot \frac{1}{\langle j_x \rangle} \frac{\partial}{\partial t} \langle j_x \rangle$$
$$= -\frac{\Gamma_X}{T} X_A + F_X(z, t), \qquad (45)$$

$$\frac{\partial}{\partial t} P_A = -\frac{\Gamma_P}{T} P_A + F_P(z, t). \qquad (46)$$

Here, $\Gamma_{X(P)} = \Gamma_{y(z)} - \frac{1}{2}\Gamma_x$ and $F_{X(P)} = F_{y(z)}/\sqrt{\langle j_x \rangle}$. Now we can consider equations of motion for the light–matter interaction in the presence of the decoherence in terms of the canonical variables

$$\frac{\partial}{\partial z} \begin{pmatrix} X_L \\ P_L \end{pmatrix}(z, t) = \frac{\kappa(z, t)}{\sqrt{LT}} \begin{pmatrix} P_A \\ 0 \end{pmatrix} - \frac{\gamma_y}{2L} \begin{pmatrix} X_L \\ P_L \end{pmatrix} + \begin{pmatrix} F_{L,X} \\ F_{L,P} \end{pmatrix}(z, t),$$
$$(47)$$

$$\frac{\partial}{\partial t} \begin{pmatrix} X_A \\ P_A \end{pmatrix}(z, t) = \frac{\kappa(z, t)}{\sqrt{LT}} \begin{pmatrix} P_L \\ 0 \end{pmatrix} - \frac{1}{T} \begin{pmatrix} \Gamma_X X_A \\ \Gamma_P P_A \end{pmatrix} + \begin{pmatrix} F_X \\ F_P \end{pmatrix}(z, t).$$
$$(48)$$

Here, the coupling constant $\kappa(z, t)$ is a function of position and time due to the decay of the classical spin component and of the driving wave amplitude according to (41), (42)

$$\kappa(z, t) = \frac{1}{2} g a_1 \sqrt{\langle j_x(z, t) \rangle} \langle a_x(z, t) \rangle \sqrt{LT}$$
$$= \kappa \, e^{-\frac{1}{2}(\gamma_x z/L + \Gamma_x t/T)}. \qquad (49)$$

Integration of the equations of motion over space and time gives us the input–output relations

$$\begin{pmatrix} X_L^{\text{out}} \\ P_L^{\text{out}} \end{pmatrix} = \begin{pmatrix} X_L^{\text{in}} \\ P_L^{\text{in}} \end{pmatrix} e^{-\frac{\gamma_y}{2}} + \kappa_L \begin{pmatrix} P_A^{\text{in}} \\ 0 \end{pmatrix} + \begin{pmatrix} F_{X_L} \\ F_{P_L} \end{pmatrix}, \qquad (50)$$

$$\begin{pmatrix} X_A^{\text{out}} \\ P_A^{\text{out}} \end{pmatrix} = \begin{pmatrix} X_A^{\text{in}} \, e^{-\Gamma_X} \\ P_A^{\text{in}} \, e^{-\Gamma_P} \end{pmatrix} + \kappa_A \begin{pmatrix} P_L^{\text{in}} \\ 0 \end{pmatrix} + \begin{pmatrix} F_{X_A} \\ F_{P_A} \end{pmatrix}. \qquad (51)$$

Due to the space and time dependence of the function $\kappa(z, t)$, the integration of the equations of motion can be done by expanding light and atomic variables over Legendre

polynomials in space and time domain [39, 34]. However, we consider the decay rates to be small and therefore take into account only the zeroth order Legendre modes for atoms and light which are just integrals over length of the ensemble and over time of the interaction. The obtained relations are a generalization of (32) and (33) for an arbitrary decay. The following notations were used:

$$\kappa_L = \kappa \, h\left(\frac{\Gamma_x + 2\Gamma_P}{2}\right) h\left(\frac{\gamma_x - \gamma_y}{2}\right) e^{-\frac{\gamma_y}{2}}, \qquad (52)$$

$$\kappa_A = \kappa \, h\left(\frac{\Gamma_x - 2\Gamma_X}{2}\right) h\left(\frac{\gamma_x + \gamma_y}{2}\right) e^{-\Gamma_x}, \qquad (53)$$

$$h(x) = \frac{1}{x}(1 - e^{-x}).$$

The protocol of quantum memory based on the direct mapping requires $\kappa_A = 1$, as described above. It is different from the constraint $\kappa = 1$ without decoherence. For a given detuning, an increase of the number of atoms leads to higher scattering for the signal field and an increase of the driving field strength gives rise to decay of atomic spins. Overall, it means that if the number of atoms for a given detuning is too low there is no way to meet the constraint by increasing the field power and vice versa.

The noise terms in the input–output relations (50) and (51) are a bit lengthy to express exactly. One can show that the noise terms have the following approximate values:

$$\langle F_{X_L}^2 \rangle \approx \frac{1}{2}\gamma_y + \frac{\kappa^2}{3}\langle F_P^2 \rangle, \qquad (54)$$

$$\langle F_{P_L}^2 \rangle \approx \frac{1}{2}\gamma_y, \qquad (55)$$

$$\langle F_{X_A}^2 \rangle \approx \langle F_X^2 \rangle + \frac{\kappa^2}{6}\gamma_y, \qquad (56)$$

$$\langle F_{P_A}^2 \rangle \approx \langle F_P^2 \rangle, \qquad (57)$$

where the collective noise correlators for atoms are defined as follows:

$$\langle F_{X(P)}^2 \rangle = \frac{1}{LT} \int_L dz \, dz' \int_T dt \, dt' \langle F_{X(P)}(z, t) F_{X(P)}(z', t') \rangle.$$

In order to preserve mean values of the canonical variables during the mapping of the field state to the atoms, one has to choose the feedback gain parameter to be $\nu = e^{\frac{\gamma_y}{2}}$. Assuming input states of light and atoms to be coherent, we find the fidelity according to (40) which for low decay rates reads

$$\mathcal{F} \approx \sqrt{\frac{2}{3}} \left(1 - \frac{11}{36}\gamma_y - \frac{1}{3}\left[\langle F_X^2 \rangle + 2\langle F_P^2 \rangle - \Gamma_X\right]\right). \qquad (58)$$

In order to evaluate the fidelity, we need to know all these decay rates and Langevin noises for a particular atomic ensemble. Here we come to the central part of the paper—the proper treatment of spontaneous emission in multilevel atoms.

4.2. General method

In this section, we find the full equations of motion from the single atom theory and assume that the formalism can be extended for a large collection of atoms that couple to their own reservoir and do not interact with each other. For a justification of this approach, we refer to [[30], where it is shown that the approximation is suitable for dilute elongated samples for which $\rho\lambda^3 \ll 1$ and the Fresnel number of the ensemble is much larger than unity. The interaction Hamiltonian including both forward and non-forward modes $\mathbf{E} = \mathbf{E}_F + \mathbf{E}_S$ is

$$
\begin{aligned}
H_j &= \mathbf{E}^{(-)}\boldsymbol{\alpha}_j\mathbf{E}^{(+)} \\
&\simeq \mathbf{E}_F^{(-)}\boldsymbol{\alpha}_j\mathbf{E}_F^{(+)} + \mathbf{E}_S^{(-)}\boldsymbol{\alpha}_j\mathbf{E}_F^{(+)} + \mathbf{E}_F^{(-)}\boldsymbol{\alpha}_j\mathbf{E}_S^{(+)} \\
&= H_{\mathrm{coh}}^j + V_j,
\end{aligned}
\tag{59}
$$

where j labels the jth atom. We have neglected the much weaker contribution $\mathbf{E}_S^{(-)}\boldsymbol{\alpha}_j\mathbf{E}_S^{(+)}$ which has no enhancement by the strong field in $\mathbf{E}_F$. The coupling of forward modes to forward modes is identified as the coherent interaction

$$
H_{\mathrm{coh}} = \mathbf{E}_F^{(-)}\boldsymbol{\alpha}\,\mathbf{E}_F^{(+)} \equiv \frac{d_0^2}{\Delta}\mathbf{E}_F^{(-)}\alpha\mathbf{E}_F^{(+)}.
\tag{60}
$$

Here we have introduced dimensionless α which will be more convenient to use in what follows. The interaction with the environment $\mathbf{E}_S$ will be treated in the Wigner–Weisskopf approximation. To do this, we write the perturbation V_j in the form

$$
V_j = \frac{d_0^2}{\Delta}\sum_\sigma\int\frac{\mathrm{d}^3k}{(2\pi)^3}\sqrt{\frac{\omega_k}{2\epsilon_0}}\boldsymbol{\epsilon}_{k\sigma}\left(b_{k\sigma}^\dagger\alpha_j\mathbf{E}_F^{(+)} + \mathbf{E}_F^{(-)}\alpha_j b_{k\sigma}\right).
\tag{61}
$$

The summation here is performed over all directions and polarizations of the non-forward electromagnetic modes. The resulting interaction will depend on the relative orientation of the atomic dipole moments hidden in the polarizability tensor α and the light field polarization vector. The net effect of this directional dependence appears as a difference in the decay rates and added noises for parallel and orthogonal light and atomic spin orientations. The modes in $\mathbf{E}_S$ have a Hamiltonian

$$
H_R = \sum_\sigma\int\frac{\mathrm{d}^3k}{(2\pi)^3}\omega_k b_{k\sigma}^\dagger b_{k\sigma}.
\tag{62}
$$

We assume the system to start out in vacuum and proceed in a Wigner–Weisskopf approach by first finding the time evolution of the b-operators

$$
\begin{aligned}
\frac{\partial}{\partial t}b_{k\sigma}(t) &= \mathrm{i}[H_R + V_j, b_{k\sigma}] \\
&= -\mathrm{i}\omega_k b_{k\sigma}(t) - \mathrm{i}\frac{d_0^2}{\Delta}\sqrt{\frac{\omega_k}{2\epsilon_0}}\boldsymbol{\epsilon}_{k\sigma}\alpha_j(t)\mathbf{E}_F^{(+)}(t),
\end{aligned}
\tag{63}
$$

which has the formal solution

$$
\begin{aligned}
b_{k\sigma}(t) &= b_{k\sigma}(0)\,\mathrm{e}^{-\mathrm{i}\omega_k t} \\
&\quad -\mathrm{i}\frac{d_0^2}{\Delta}\sqrt{\frac{\omega_k}{2\epsilon_0}}\boldsymbol{\epsilon}_{k\sigma}\int_0^t\mathrm{d}t'\alpha_j(t')\mathbf{E}_F^{(+)}(t')\,\mathrm{e}^{-\mathrm{i}\omega_k(t-t')}.
\end{aligned}
\tag{64}
$$

We are interested in the equation of motion of some operator A (which can belong to either atoms or light). We insert the found expression for $\mathbf{E}_S$ into V_j

$$
\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}A(t) &= [H_{\mathrm{coh}}^j, A] + \mathrm{i}\frac{d_0^2}{\Delta}\sum_\sigma\int\frac{\mathrm{d}^3k}{(2\pi)^3}\sqrt{\frac{\omega_k}{2\epsilon_0}}\boldsymbol{\epsilon}_{k\sigma} \\
&\quad \times\left(b_{k\sigma}^\dagger(0)\mathrm{e}^{\mathrm{i}\omega_k t}[\alpha_j\mathbf{E}_F^{(+)}, A](t)\right. \\
&\quad \left.+ [\mathbf{E}_F^{(-)}\alpha_j, A](t)b_{k\sigma}(0)\,\mathrm{e}^{-\mathrm{i}\omega_k t}\right) \\
&\quad -\left(\frac{d_0}{\Delta}\right)^2\sum_\sigma\int\frac{\mathrm{d}^3k}{(2\pi)^3}\frac{\omega_k}{2\epsilon_0}\epsilon_{k\sigma}^2\int_0^t\mathrm{d}t' \\
&\quad \times\left(\mathbf{E}_F^{(-)}(t')\alpha_j(t')\,\mathrm{e}^{\mathrm{i}\omega_k(t-t')}[\alpha_j\mathbf{E}_F^{(+)}, A](t)\right. \\
&\quad \left.- [\mathbf{E}_F^{(-)}\alpha_j, A](t)\alpha_j(t')\mathbf{E}_F^{(+)}(t')\,\mathrm{e}^{-\mathrm{i}\omega_k(t-t')}\right).
\end{aligned}
\tag{65}
$$

Going into the rotating frame of the forward field, which has the carrier frequency ω_0 and performing the Markov approximation, we may take the polarizability outside the integral and find that the general equation of motion for an observable A is given by the quantum Langevin equation

$$
\begin{aligned}
\frac{\mathrm{d}}{\mathrm{d}t}A &= \mathrm{i}[H_{\mathrm{coh}}^j, A] + \left(\frac{d_0}{\Delta}\right)^2\mathcal{L}_j(A) \\
&\quad + \mathrm{i}\sqrt{\gamma}\frac{d_0}{\Delta}\left([\mathbf{E}_F^{(-)}\alpha_j, A]\mathbf{f}_j + \mathbf{f}_j^\dagger[\alpha_j\mathbf{E}_F^{(+)}, A]\right).
\end{aligned}
\tag{66}
$$

Here, γ is the decay rate $\gamma = d_0^2\omega_0^3/3\pi\epsilon_0$ which is related to the radiative decay rate of an atom with excited state electronic angular momentum J' as $\gamma_{\mathrm{rad}} = \gamma/(2J' + 1)$. We have neglected the Lambshift, which is assumed to be incorporated in the transition frequency ω_0; also we ignore collective effects in the limit of a dilute ensemble (see [[30] for details) as discussed in the beginning of section 4.2. The decay is described by the Lindblad form

$$
\begin{aligned}
\mathcal{L}_j(A) &= \frac{\gamma}{2}\left(2\mathbf{E}_F^{(-)}\alpha_j A\alpha_j\mathbf{E}_F^{(+)} - \mathbf{E}_F^{(-)}\alpha_j^2\mathbf{E}_F^{(+)}A\right. \\
&\quad \left.- A\mathbf{E}_F^{(-)}\alpha_j^2\mathbf{E}_F^{(+)}\right).
\end{aligned}
\tag{67}
$$

The decay gives two different contributions, one where the atom decays to the same state and will involve the a-coefficients belonging to α_{FF} and one to different states through $\alpha_{FF\pm1}$ and be described by b-coefficients. But we are not going to have any cross terms of the form ab since they will oscillate at much higher frequencies corresponding to the hyperfine splitting $\sim$ GHz and average out to zero in (65). Above we have introduced the noise operators

$$
\mathbf{f}_j(t) = \frac{d_0}{\sqrt{\gamma}}\sum_\sigma\int\frac{\mathrm{d}^3k}{(2\pi)^3}\sqrt{\frac{\omega_k}{2\epsilon_0}}\boldsymbol{\epsilon}_{k\sigma}b_{k\sigma}^j(0)\mathrm{e}^{-\mathrm{i}(\omega_k-\omega_0)t},
\tag{68}
$$

which in the Markov approximation are delta-correlated in time

$$
\langle[f_{i,\mu}(t), f_{j,\nu}^\dagger(t')]\rangle = \delta_{ij}\delta_{\mu\nu}\delta(t - t').
\tag{69}
$$

Below we will apply these equations of motion to observables for both light and atoms. There is an essential difference between the decoherence of light and of atoms. The reason is that each time the atoms undergo spontaneous emission they remain in the ensemble. For light on the other hand, a lost photon simply attenuates the beam but there is no change in its coherence. It means that we can model the resulting noise for light as vacuum operators when considering the field a, $a^\dagger$.

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4.3. Light

In the last section, we showed how to treat the spontaneous emission and how to calculate the decay for the operators of interest. We will now apply this to the light field operators $a_x(z, t)$ and $a_y(z, t)$. First, one has to generalize the dynamics of a single atom studied above to a spatially extended ensemble. The continuous equations of motion are obtained from the single atom version (66) by replacing operators with the space-dependent ones defined in (15) and then integrating the right-hand side over the ensemble volume.

In order to obtain the required equations for the field propagation, one has to consider the field evolution including the interaction Hamiltonian and the free field Hamiltonian. The resulting equation is obtained from the Heisenberg–Langevin equation (66) by replacing $\frac{\partial}{\partial t} \rightarrow \frac{\partial}{\partial z} + \frac{\partial}{\partial t}$ as showed in appendix C and [40]. Since the light pulse is usually much longer than the size of the atomic ensemble, we can omit the time derivative as it only describes the retardation effect. Using the commutation relation for light field operators, one obtains the following equations of motion:

$$
\frac{\partial}{\partial z} a_\mu(z, t) = i \int_L dz [H_{\text{coh}}, a_\mu(z, t)]
$$
$$
- \frac{|g|\gamma}{4\Delta} \frac{N_a}{L} (\langle \alpha^2 \rangle_{\mu\mu} a_\mu(z, t) + \langle \alpha^2 \rangle_{\mu\nu} a_\nu(z, t)) + F_{L,\mu}(z, t),
$$
$$(70)$$

$$
F_{L,\mu}(z, t) = i \sqrt{\frac{|g|\gamma}{2\Delta} \frac{N_a}{L}} (\alpha_{\mu\mu} f_\mu(z, t) + \alpha_{\mu\nu} f_\nu(z, t)), \quad (71)
$$

where $\mu = \{x, y\}$ and $\mu \neq \nu$. Here we have assumed that the atoms are evenly distributed so that $\langle \alpha^2 \rangle_{\mu\mu}(z) = \frac{N_a}{L} \langle \alpha^2 \rangle_{\mu\mu}$ and the element $\langle \alpha^2 \rangle_{ij}$ is to be understood as the ij'th element of the matrix α^2. Using (69), we obtain the following properties of the averaged commutators of the noise operators $F_{L,\mu}$:

$$
\langle [F_{L,\mu}(z, t), F_{L,\nu}^\dagger(z', t')] \rangle
$$
$$
= \frac{|g|\gamma}{2\Delta} \frac{N_a}{L} \langle \alpha^2 \rangle_{\mu\nu} \delta(t - t') \delta(z - z'). \quad (72)
$$

Since the matrix α^2 depends on the atomic operators, the expressions derived here incorporate that the decoherence of the light field depends on the state of the atoms. To evaluate the formulae, we therefore need to specify the atomic state. Here we assume that the atoms are initially polarized along the x-axis and evaluate the expressions in a coherent spin state. In principle, there is also a possibility for cross-scattering between the x and y polarization, which is contained in the matrix element $\langle \alpha^2 \rangle_{xy}$. In appendix D, we show, however, that this matrix element vanishes for this particular spin configuration. As noted above, experiments sometimes use a different configuration with the atoms polarized perpendicular to the light polarization. Also in this case, the cross polarization scattering vanishes, but this is not the case for a general orientation. Experimentally it is advantageous to avoid the cross polarization scattering since this avoids a rotation of the mean polarization of the light. Such rotation may be difficult to control precisely and this classical noise could therefore overwhelm the quantum noise around the mean which we are trying to control. Therefore, the geometry with spin polarized parallel or orthogonal to the direction of light polarization
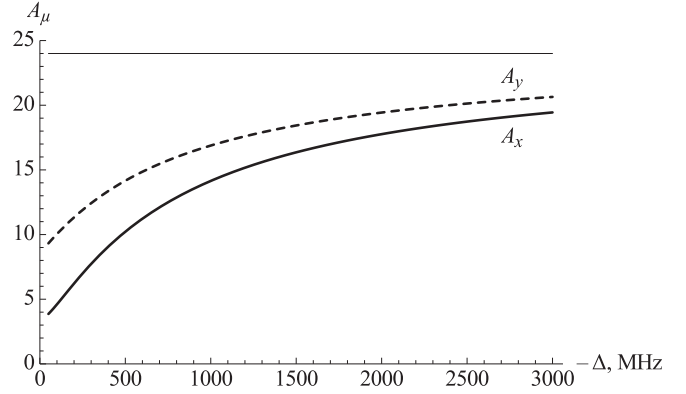

Figure 5. Light attenuation and noise are described by the matrix A_μ. Solid line shows the matrix element A_x as a function of detuning $-\Delta$ in MHz and the dashed line corresponds to A_y. We assume atoms to be polarized along x. Cs in $F = 4$.

is preferable. In both these cases, we obtain the equation of motion for light in the form (41) with the decay rates

$$
\gamma_\mu = \frac{|g|\gamma}{2\Delta} N_a \langle \alpha^2 \rangle_{\mu\mu} = 2 \frac{N_a}{N_p} \frac{\kappa^2}{d} A_\mu, \quad (73)
$$
$$
A_\mu = \frac{\langle \alpha^2 \rangle_{\mu\mu}}{F a_1^2}.
$$

Here the difference in the orientation of the atoms is again contained in the matrices $\langle \alpha^2 \rangle_{\mu\mu}$. For caesium with $F = 4$, the A_μ coefficients are shown in figure 5. In the limit of large detuning, the coefficients read

$$
A_{x(y)} \rightarrow 24. \quad (74)
$$

Taking into account the coherent part of the interaction Hamiltonian, we arrive at the full equations for the light field

$$
\frac{\partial}{\partial z} a_y(z, t) = \frac{g}{2\sqrt{T}} a_x(z, t)[a_1 j_z(z, t) + i a_2 \{j_x, j_y\}(z, t)]
$$
$$
- \left[\frac{\gamma_y}{2} - 2 i a_2 j_y^2(z, t) \right] a_y(z, t) + \sqrt{\gamma_y} f_y(z, t), \quad (75)
$$

$$
\frac{\partial}{\partial z} a_x(z, t) = \frac{-g}{2\sqrt{T}} a_y(z, t)[a_1 j_z(z, t) - i a_2 \{j_x, j_y\}(z, t)]
$$
$$
- \left[\frac{\gamma_x}{2} - 2 i a_2 j_x^2(z, t) \right] a_x(z, t). \quad (76)
$$

Since the x-polarized field is supposed to be strong and classical, we neglect its quantum noise.

4.4. Atoms

In this section, we consider atomic evolution in a similar way as was done for light. To describe the decay of the single atomic spin, we use expressions (66) and (67) from above to obtain

$$
\frac{\partial}{\partial t} j_i(z, t) = \left(\frac{d_0}{\Delta} \right)^2 \mathcal{L}(j_i)(z, t), \quad (77)
$$

$$
\mathcal{L}(j_i)(z, t) = -\frac{\gamma}{2} \mathbf{E}_F^{(-)}(z)[\alpha^2 j_i + j_i \alpha^2 - 2\alpha j_i \alpha](z) \mathbf{E}_F^{(+)}(z). \quad (78)
$$

We expect this decay to be proportional to the total flux of photons and we can as a good approximation ignore the position dependence of the light field, allowing us to write for the collective spin

$$\mathcal{L}(j_i)(t) = -\frac{\gamma}{2}\mathbf{E}_\mathrm{F}^{(-)}\int \mathrm{d}z[\alpha^2 j_i + j_i\alpha^2 - 2\alpha j_i\alpha](z)\mathbf{E}_\mathrm{F}^{(+)}.$$

The most notable contribution comes from the *x* polarized part of light. Since we already expect the effect of spontaneous emission to be small in the regime where the quantum interface is operating (remember that it scales as $1/\Delta^2$) we can safely only consider the *xx* element of the matrix

$$\xi_i = \alpha^2 j_i + j_i\alpha^2 - 2\alpha j_i\alpha.$$

With this assumption, we find that we can make the linearization

$$\mathcal{L}(j_i)(t) = -\frac{\gamma}{2}|E|^2\frac{N_p}{T}\Xi_i\, j_i(t), \tag{79}$$

where the field amplitude is given by $|E|^2 = \omega_0/2\epsilon_0 A$ and the decay rate coefficient Ξ_i is defined through the expression

$$\Xi_i\, j_i(t) = \int_0^L \mathrm{d}z\,\xi_i(z, t). \tag{80}$$

This element gives the magnitude of the decay of the respective component of the spin. For the decay to a different hyperfine level *F* (the *b* terms), we will neglect the term $2\alpha j_i\alpha$, which describes the increase in the population of the final state. The reason is that once an atom decays to another hyperfine level *F* it will no longer be interesting for us, since we restrict our analysis to the collective behaviour of many atoms in the same *F* state. This is reasonable since the energy spacing between the two ground $F = I \pm \frac{1}{2}$ hyperfine levels is big. For the same reasons, we obviously need to keep $2\alpha j_i\alpha$ for the *a*-terms. Written in short form, the decay reads

$$\frac{\partial}{\partial t}j_i(t) = -\frac{|g|\gamma}{4\Delta}\frac{N_p}{T}\Xi_i\, j_i(t) + \text{Noise}. \tag{81}$$

This expression gives us the decay rates of the spin components defined in (42). The noise terms will be treated in detail below. The decay rates depend on the relative orientation of spin and light polarizations as it is the case with the light decay. The expression for the decay rates now reads

$$\Gamma_{i_{\|(\perp)}} = \frac{|g|\gamma}{4\Delta}N_p\,\Xi_{i_{\|(\perp)}} = \frac{\kappa^2}{d}B_{i_{\|(\perp)}}, \tag{82}$$

$$B_{i_{\|(\perp)}} = \frac{\Xi_{i_{\|(\perp)}}}{Fa_1^2}.$$

Similar to the discussion of spontaneous emission in section 3.3, we have expressed here the atomic scattering in terms of the coupling constant κ and the optical depth *d*. The details of the interaction and the difference between different atoms, which is our main interest here, is thus contained in the coefficient *B*. The required coefficients Ξ are calculated in appendix D.3 for an arbitrary atom. The B_i coefficients for caesium atoms are shown in figure 6, and explicit expressions

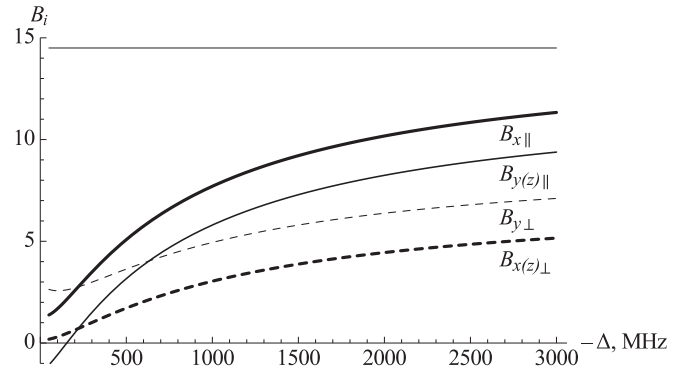**Figure 6.** Spin decay coefficients B_i as functions of detuning $-\Delta$ in MHz for parallel configuration (spin and light are *x* polarized) are shown by a thick solid line for the *x* spin component and by a thin solid curve for *y* and *z* spin projections. The orthogonal configuration (spin is *x* polarized and the light is *y* polarized) is presented by the dashed curves, thick line is *x* and *z*, thin is the *y* spin component. The plot is calculated for caesium atoms with ground state $F = 4$.

can be found in appendix E. In the limit of high detuning, the coefficients for caesium atoms with $F = 4$ read

$$B_{x_\|} \to \frac{29}{2}, \qquad B_{y_\|},\, B_{z_\|} \to \frac{25}{2},$$
$$B_{y_\perp} \to \frac{37}{4}, \qquad B_{x_\perp},\, B_{z_\perp} \to \frac{1}{2}B_{x_\|}.$$

The remaining term in the Heisenberg–Langevin equation (66) gives us the spin noise components

$$\frac{\partial}{\partial t}j_i(z, t) = \mathrm{i}\sqrt{\gamma}\frac{d_0}{\Delta}\left(\mathbf{E}_F^{(-)}[\alpha, j_i]\mathbf{f} + \mathbf{f}^\dagger[\alpha, j_i]\mathbf{E}_F^{(+)}\right).$$

From here, the noise operators for canonical spin density variables defined above as $F_X(z, t)$ and $F_P(z, t)$ are given by

$$F_{X(P)}(z, t) = \frac{\mathrm{i}\sqrt{\gamma}d_0}{\Delta\sqrt{n\langle j_x^a\rangle}}\left(\mathbf{E}_F^{(-)}[\alpha, j_{y(z)}]\mathbf{f} + \mathbf{f}^\dagger[\alpha, j_{y(z)}]\mathbf{E}_F^{(+)}\right). \tag{83}$$

These are the noise operators introduced in (48). The collective noise operators are obtained by averaging it over the light pulse duration and the length of the atomic ensemble

$$F_{X(P)} = \frac{1}{\sqrt{LT}}\int_L \mathrm{d}z\int_T \mathrm{d}t\, F_{X(P)}(z, t). \tag{84}$$

When forming the combinations such as $\langle F_X F_X\rangle$, since we have assumed that there are no photons in the reservoir with frequency at ω_0, the only combination which survives is $\langle \mathbf{f}\,\mathbf{f}^\dagger\rangle$. Moreover, we have made the assumption that the *x* (or *y* in the case of orthogonal configuration of light and spin polarizations) component of the light field is dominating, so we will only have to consider the *xx*(*yy*) element of the matrix $\zeta_i^2 = (i[\alpha, j_i])^2$. So ultimately what we will have is

$$\langle F_{X(P)}^2\rangle_{\|(\perp)} = \frac{|g|\gamma}{2\Delta}\frac{N_p}{F}\langle\zeta_{y(z)}^2\rangle_{\|(\perp)} = \frac{2}{F}\frac{\kappa^2}{d}C_{y(z)_{\|(\perp)}}, \tag{85}$$

$$C_{i_{\|(\perp)}} = \frac{\langle\zeta_i^2\rangle_{\|(\perp)}}{(Fa_1)^2}.$$

The different $\langle\zeta^2\rangle$ are calculated in appendix D.2 and are generally quite complicated. Since any orientation besides the parallel and orthogonal will give too much noise on the
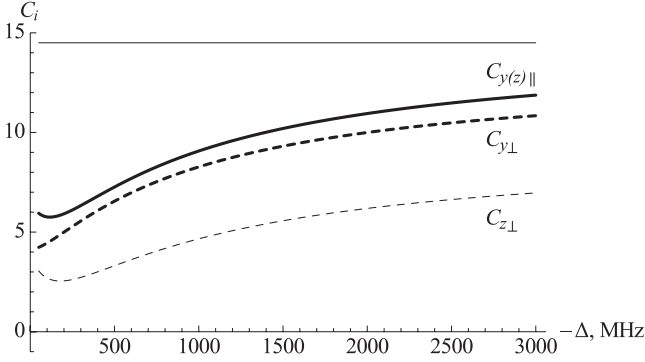
Figure 7. Spin noise coefficients C_i as functions of detuning $-\Delta$ in MHz for caesium with the ground state $F = 4$. The solid line represents y and z noise coefficients for x polarized atoms and light. The dashed curves correspond to the orthogonal configuration of atomic spin and light. The thick dashed curve shows $C_{y_\perp}$ and the thin dashed line is $C_{z_\perp}$.

light as discussed above, we only consider these two settings. The spin noise coefficients C_i required for the calculation of canonical spin variable noises are shown in figure 7, and the explicit expressions are given in appendix E. In the limit of high detuning, the coefficients for caesium atoms with $F = 4$ read

$$C_{y_\parallel}, C_{z_\parallel} \rightarrow \frac{29}{2}, \qquad C_{y_\perp} \rightarrow \frac{53}{4}, \qquad C_{z_\perp} \rightarrow \frac{37}{4}.$$

The cross correlations of the Langevin noise are closely related to the spin decay rates as shown in appendix D.4. The commutator for noise reads

$$\langle [F_X, F_P] \rangle_{\parallel(\perp)} = \frac{\mathrm{i}}{F} (\Gamma_y + \Gamma_z - \Gamma_x)_{\parallel(\perp)}. \tag{86}$$

Moreover, one can show that the anticommutator of the noise terms is equal to zero.

5. Summary

In this section, we summarize the obtained results and as an example we apply the results to determine the decay rates and Langevin noises for the specific case of caesium atoms. First of all, the decay of the spins and light is described by the Heisenberg–Langevin equations

$$\frac{\partial}{\partial z} a_\mu = -\frac{\gamma_\mu}{2L} a_\mu + F_{L,\mu}, \qquad \mu = \{x, y\},$$

$$\frac{\partial}{\partial t} j_i = -\frac{\Gamma_i}{T} j_i + F_i, \qquad i = \{x, y, z\}.$$

As was mentioned earlier, there are two preferable configurations available. Either the light polarization is oriented along the same axis as the spins of the atoms (x) or the light is orthogonal to the atomic polarization (y). In these cases, the decay rates for light are given by (73) and the atomic decay rates can be found in (82). They read

$$\gamma_\mu = 2\frac{N_a}{N_p}\frac{\kappa^2}{d} A_\mu, \qquad \Gamma_{i_{\parallel(\perp)}} = \frac{\kappa^2}{d} B_{i_{\parallel(\perp)}},$$

$$A_\mu = \frac{\langle \alpha^2 \rangle_{\mu\mu}}{F a_1^2}, \qquad B_{i_{\parallel(\perp)}} = \frac{\Xi_{i_{\parallel(\perp)}}}{F a_1^2}.$$

From these expressions we see that one can obtain a strong interaction $\kappa \sim 1$ with negligible decoherence provided that the atomic ensemble has a sufficiently high optical depth $d \gg 1$. Furthermore, the decoherence of the light modes can be suppressed relative to the atomic decay provided a larger number of photons than atoms is used $N_P \gg N_A$. Most of these expressions are well known from the previous work in the field [6]. The main new result in this work are the exact expressions for the coefficients A, B and C, which are numbers of order unity, which contain the information specific to the particular atom one is considering. We will evaluate the required coefficients for ^{133}Cs optically pumped to the ground state $F = 4$. In this case, we have $I = 7/2$, $J = 1/2$, $J' = 3/2$, $F = 4$, $\tilde{F} = 3$. The coefficient a_1 is given by (B.9), which is evaluated for $F = \tilde{F} = 4$ and $k = 1$:

$$a_1 = \frac{7}{5760} \left(\frac{176}{7} - \frac{3}{1 - \frac{\Delta_{45}}{\Delta}} - \frac{5}{1 - \frac{\Delta_{35}}{\Delta}} \right). \tag{87}$$

The matrix $\langle \alpha^2 \rangle$ whose diagonal components enter in A_μ can be found in appendix D. The Cartesian components of the matrix for the spin oriented along the x-axis are expressed via its spherical components by (D.4) and (D.5). The required spherical components of the matrix are given by (D.3). One needs to know the coefficients $a_k^{F\tilde{F}}$ mentioned above and the coefficients c_k given by (B.11). The result for Cs atoms is

$$\langle \alpha^2 \rangle_{xx} = \frac{1}{80} \left(1 + \frac{7}{3\left(1 - \frac{\Delta_{45}}{\Delta}\right)^2} \right) \rightarrow \frac{1}{24},$$

$$\langle \alpha^2 \rangle_{yy} = \frac{1}{720} \left(23 + \frac{21}{8\left(1 - \frac{\Delta_{45}}{\Delta}\right)^2} + \frac{35}{8\left(1 - \frac{\Delta_{35}}{\Delta}\right)^2} \right) \rightarrow \frac{1}{24}.$$

Since the spin is considered to be oriented along x, the xx-element of the scattering matrix represents decay of the field polarized along the spin, and the yy-component is correspondingly the decay rate coefficient for the orthogonally polarized field configuration.

The calculation of the spin decay coefficients Ξ entering the B coefficients is similar but a bit more lengthy since it involves three spin projections and two possible configurations. The basic expressions for parallel and orthogonal configurations of spin and light are given by (D.25)–(D.30). The required spherical components of $\langle \xi_\mu j_\nu \rangle$ are defined in (D.22)–(D.24). The definitions involve the same coefficients $a_k^{F\tilde{F}}$ and c_k used above for the evaluation of the light decay rates. For reference, we provide the result of the calculation for the expression $\langle \Xi_y \rangle_\parallel$ for caesium atoms $F = 4$.

$$\langle \Xi_y \rangle_\parallel = \frac{7}{1152} \left(-\frac{152}{175} - \frac{7}{12\left(1 - \frac{\Delta_{35}}{\Delta}\right)\left(1 - \frac{\Delta_{45}}{\Delta}\right)} \right.$$

$$\left. + \frac{1}{2\left(1 - \frac{\Delta_{35}}{\Delta}\right)^2} + \frac{76}{75\left(1 - \frac{\Delta_{45}}{\Delta}\right)} + \frac{351}{100\left(1 - \frac{\Delta_{45}}{\Delta}\right)^2} \right).$$

The Langevin noise operators for the light modes (72) are simply the minimal noise required to preserve the commutation relation for the free field:

$$\langle F_{L,\mu}(z, t) F_{L,\nu}^\dagger(z', t') \rangle = \frac{\gamma_\mu}{L} \delta_{\mu\nu} \delta(t - t') \delta(z - z').$$

In the context of quantum information protocols, it is more convenient to study the evolution of the canonical variables for the spin. In this case, the decay process is described according to (45) by

$$\frac{\partial}{\partial t} X_A = -\frac{\Gamma_X}{T} X_A + F_X(z, t),$$

$$\frac{\partial}{\partial t} P_A = -\frac{\Gamma_P}{T} P_A + F_P(z, t).$$

Here, $\Gamma_{X(P)} = \Gamma_{y(z)} - \frac{1}{2}\Gamma_x$ and $F_{X(P)} = F_{y(z)}/\sqrt{\langle j_x \rangle}$. The Langevin noise correlator for the collective canonical variables is found in (84) and (85) and it reads

$$\langle F^2_{X(P)} \rangle_{\parallel(\perp)} = \frac{2}{F}\frac{\kappa^2}{d} C_{y(z)\parallel(\perp)}, \qquad C_{i\parallel(\perp)} = \frac{\langle \zeta_i^2 \rangle_{\parallel(\perp)}}{Fa_1^2}.$$

The averaged commutator for the noise is given by (86):

$$\langle [F_X, F_P] \rangle_{\parallel(\perp)} = \frac{i}{F}(\Gamma_X + \Gamma_P)_{\parallel(\perp)}.$$

To evaluate this expression one needs to calculate the matrix $\langle \zeta_i^2 \rangle$. The calculation of this is essentially the same as for the spin decay coefficient. The expressions for the Cartesian components via the spherical one are given by (D.13)–(D.18). Spherical components for different F and $\tilde{F}$ are found in (D.10)–(D.12). For reference we provide the result of the calculation for the expression $\langle \zeta_y^2 \rangle_{\parallel}$ for caesium atoms with $F = 4$.

$$\langle \zeta_y^2 \rangle_{\parallel} = \frac{7}{2400}\left(\frac{176}{21} - \frac{175}{72\left(1 - \frac{\Delta_{35}}{\Delta}\right)\left(1 - \frac{\Delta_{45}}{\Delta}\right)} \right.$$
$$\left. + \frac{25}{12\left(1 - \frac{\Delta_{35}}{\Delta}\right)^2} - \frac{88}{9\left(1 - \frac{\Delta_{45}}{\Delta}\right)} + \frac{83}{8\left(1 - \frac{\Delta_{45}}{\Delta}\right)^2} \right).$$

Expressions for all of the coefficients A, B, C for ^{133}Cs are explicitly given in appendix E.

5.1. Example of application

Now we can use the derived expressions for the light and and atomic decay rates to evaluate the fidelity obtained in section 4.1. First, let us consider the coupling constant κ_A given by (53) for the case of ^{133}Cs. In figure 8 we show the solution of the equation $\kappa_A = 1$ which gives us the required parameter regime for the quantum memory based on direct mapping protocol. One can see that within a wide range of values for the optical depth $d = N_a\sigma/A$ the requirement $\kappa_A = 1$ coincides with the condition $\kappa = 1$. However, when it reaches extreme values the decay processes significantly affect the system evolution and one is required to provide $\kappa > 1$ in order to fulfill the quantum memory condition for transfer of the mean amplitude.

For a given optical depth d one can express the fidelity (58) by the approximation of small decay probability. Consequently for the parameter regime where $\kappa_A = 1$ coincides with the constriction $\kappa = 1$ we have

$$\mathcal{F} \approx \sqrt{\frac{2}{3}}\left(1 - c_L\left(\frac{\gamma}{\Delta}\right)^2 d - c_A \frac{1}{d}\right) \qquad (88)$$
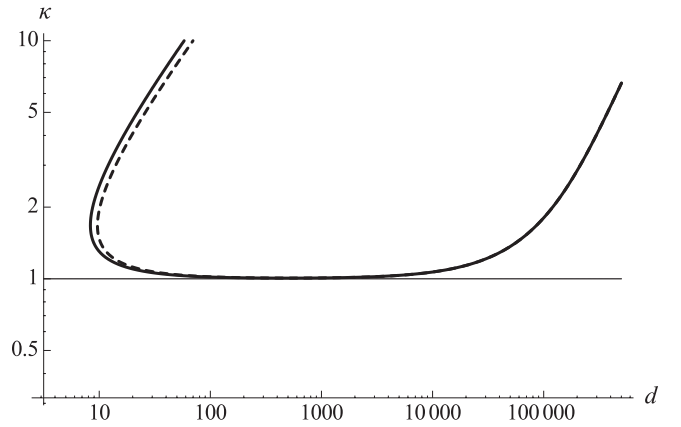

Figure 8. Solution of the equation $\kappa_A = 1$ as a function of the optical depth $d = N_a\sigma/A$. The thin solid line corresponds to the constraint without decoherence $\kappa = 1$. Thick solid and dashed curves represent respectively the solutions for the parallel and orthogonal atomic spin and light polarization configurations. The plot is calculated for caesium atoms with the ground state $F = 4$ and detuning $\Delta = -500$ MHz.

$$c_L = \frac{11}{12}\frac{1}{Fa_1^2}\langle \alpha^2 \rangle \qquad (89)$$

$$c_A = \frac{2}{3(Fa_1)^2}\left[\langle \zeta_y^2 \rangle + 2\langle \zeta_z^2 \rangle - \frac{F}{2}\left(\Xi_y - \frac{1}{2}\Xi_x\right) \right]. \qquad (90)$$

In the limit of $\Delta \to -\infty$, we can neglect the noise on the light the coefficients and the fidelity read

$$\mathcal{F} \to \sqrt{\frac{2}{3}}\left(1 - c_A\frac{1}{d}\right), \qquad (91)$$

$$c_{A_{\parallel(\perp)}} \to \frac{11}{2}\left(\frac{41}{12}\right). \qquad (92)$$

In agreement with [6], the deviation of the fidelity from the ideal one is inversely proportional to the optical depth for large detunings. The results of the paper allow us to identify the coefficient of this proportionality. It shows an advantage of the orthogonal configuration of light and spin polarizations over the parallel one in the particular quantum memory protocol discussed here for illustration.

For a finite detuning, the fidelity can be optimized by varying the ratio of photon number to the number of atoms and by changing the detuning from the resonance. The resulting optimal fidelity as a function of optical depth is shown in figure 9. The corresponding optimal detuning and optimal ratio of photons to atoms as functions of the optical depth are shown in figures 10 and 11, respectively. One has to take into account that these are very flat maxima, and hence the precise value of the detuning is not that important. This is illustrated in figure 12 where fidelities for several optical depths are shown as a function of detuning. Furthermore, one should also bear in mind that experimentally other considerations such as Doppler broadening may be important. Furthermore, to illustrate our method we have for simplicity neglected the tensor part of the coherent interaction, and this may alter the conclusion reached here.
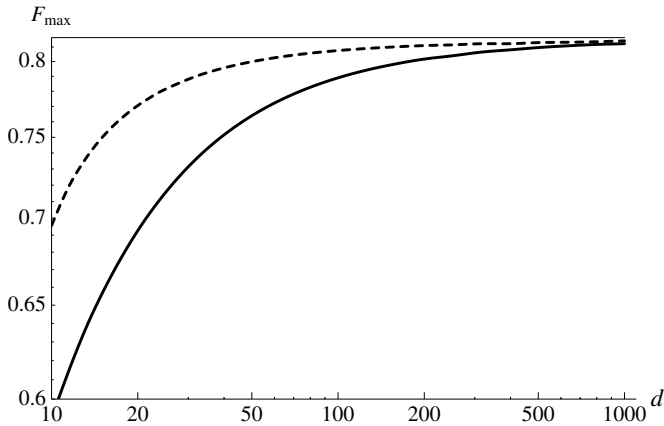
Figure 9. Optimal fidelity as a function of optical depth for caesium with $F = 4$. Solid and dashed lines correspond respectively to the parallel and orthogonal configurations of atomic spin and light polarization. Thin solid line on top shows the limiting value $\sqrt{2/3}$.
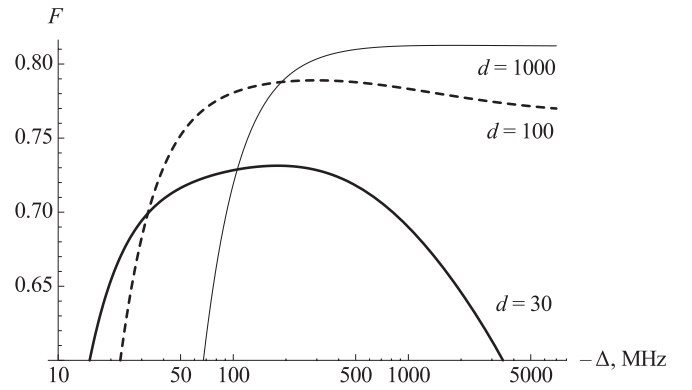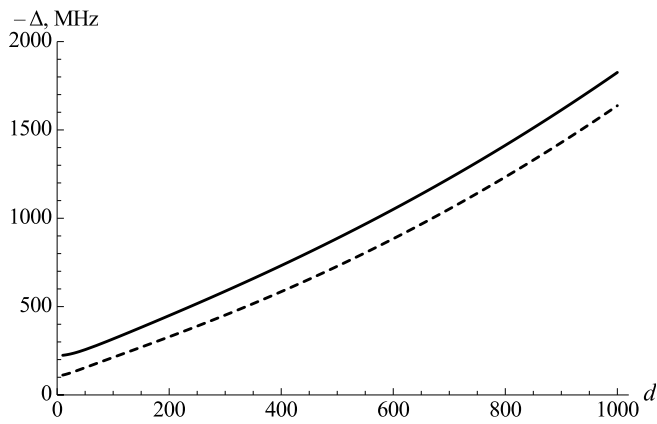

Figure 10. Optimal detuning as a function of optical depth for caesium with $F = 4$. Solid and dashed lines correspond respectively to the parallel and orthogonal configurations of atomic spin and light polarization.

Figure 11. Optimal ratio of photons to atoms as a function of optical depth for caesium with $F = 4$. Solid and dashed lines correspond respectively to the parallel and orthogonal configurations of atomic spin and light polarization.
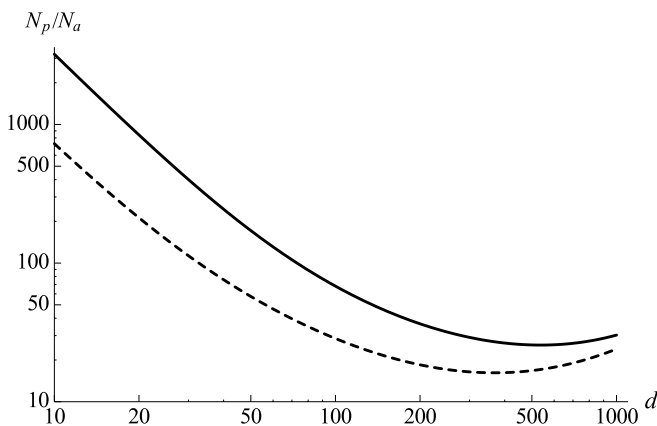

Figure 12. Fidelity for different optical depths as a function of detuning for caesium with $F = 4$. Solid, dashed and thin curves correspond to optical depths $d = 30$, 100, 1000, respectively. The calculation is done for the parallel configuration of atomic spin and light polarization.

6. Conclusion

In this paper, we have found in accordance with earlier work that spontaneous emission occurs faster for parallel polarizations than perpendicular—a consequence of selection rules and Clebsch–Gordan coefficients. We have also found the precise decay rates and noise correlations that take into account the full level structure of the atom and our procedure can easily be applied to similar systems. In this paper, we have given a comprehensive discussion of quantum noise from spontaneous emission for light matter quantum interfaces based on the Faraday effect. Taking into account the full level structure of the atoms, we derive for the first time the full expressions for the decay and quantum noise arising from spontaneous emission. In agreement with previous treatments based on simplified models, our theory shows that the decay and noise can be made to vanish for a sufficiently large optical depth of the ensemble. Given that experiments will always work with a finite optical depth, it is, however, important to have a detailed understanding of the noise. In particular, in view of the rapid experimental advances, a thorough understanding of fundamental noise sources will be crucial for further increasing the efficiency of quantum memories and for assessing the feasibility of new advances for light matter quantum interfaces. The tradeoff between the coherent dynamics and unavoidable fundamental losses, investigated in full detail here, will set the ultimate limits to the performance of future quantum networks.

Appendix A. Adiabatic elimination

The projections defined in section 2 will be used for our treatment of the operators and if we specifically apply it to

the dipole operator $\mathbf{d}$ we get (note that we do not get any contributions from the terms $P_g \mathbf{d} P_g$, $P_e \mathbf{d} P_e$ since $\mathbf{d}$ is a parity odd operator)

$$\mathbf{d} = (P_g + P_e)\mathbf{d}(P_g + P_e) = P_e \mathbf{d} P_g + P_g \mathbf{d} P_e$$
$$= \mathbf{d}^{(+)} + \mathbf{d}^{(-)}. \quad (\text{A.1})$$

Here $\mathbf{d}^{(+)}$ is the raising operator raising the atoms from the ground state $|F, m\rangle$ to the excited state $|F', m'\rangle$. In the following, we will go into the rotating frame with respect to the laser frequency ω_0, so the energy of the atom is described by the Hamiltonian

$$H_A = \sum_{F'} \Delta_{F'} P_{F'}, \quad (\text{A.2})$$

where $\Delta_{F'}$ is the detuning of the excited state from the laser frequency. To ease notation, we treat by both using a symbol g or e to represent respectively ground states $|F, m\rangle$ and excited states $|F', m'\rangle$:

$$H_{\text{int}} = -\sum_{ge} \mathbf{E}^{(-)} \mathbf{d}_{ge}^{(-)} |g\rangle\langle e| + \text{h.c.} \quad (\text{A.3})$$

Taking the commutator with $H_A + H_{\text{int}}$, we get the following equation:

$$\frac{\mathrm{d}}{\mathrm{d}t} |g\rangle\langle e| = -\mathrm{i}\Delta_e |g\rangle\langle e|$$
$$+ \mathrm{i}\mathbf{E}^{(+)} \left(\sum_{g'} \mathbf{d}_{eg'}^{(+)} |g\rangle\langle g'| - \sum_{e'} \mathbf{d}_{ge'}^{(-)} |e'\rangle\langle e| \right). \quad (\text{A.4})$$

So in the adiabatic regime, we neglect the time derivative and obtain

$$|g\rangle\langle e| = \sum_{g'} \frac{\mathbf{E}^{(+)} \cdot \mathbf{d}_{eg'}^{(+)}}{\Delta_e} |g\rangle\langle g'|. \quad (\text{A.5})$$

Inserting this into the Hamiltonian, we get the expression

$$H_{\text{int}}^{\text{eff}} = H_{\text{int}} + H_A = \mathbf{E}^{(-)} \cdot \boldsymbol{\alpha} \cdot \mathbf{E}^{(+)}$$
$$\equiv H_{\text{int}}, \quad (\text{A.6})$$

$$\boldsymbol{\alpha} = -\sum_{g'ge} \frac{\mathbf{d}_{g'e}^{(-)} \mathbf{d}_{eg}^{(+)}}{\Delta_e} |g'\rangle\langle g|. \quad (\text{A.7})$$

Here $\mathbf{d}_{g'e}^{(+)} \mathbf{d}_{eg}^{(-)} \sim \mathbf{d}_{g'e}^{(+)} \wedge \mathbf{d}_{eg}^{(-)}$ is the dyadic vector product of the dipole operator with itself.

Appendix B. Construction of the Hamiltonian

The dipole moment $\mathbf{d}$ can be expanded in spherical components by

$$\mathbf{d} = \sum_q d_q \mathbf{e}_q^* = \sum_q (-1)^q d_{-q} \mathbf{e}_q. \quad (\text{B.1})$$

The component of $\mathbf{d}$ for the $F \rightarrow F'$ transition can thus be expressed as

$$\mathbf{d}_{F'F}^{(+)} = \sum_{q,m,m'} \langle F'm'|d_q^{(+)}|Fm\rangle |F'm'\rangle\langle Fm| \mathbf{e}_q^*. \quad (\text{B.2})$$

The Wigner–Eckart theorem states that the matrix elements[4] can be expressed as

$$\langle F'm'|d_q^{(+)}|Fm\rangle = C_{Fm1q}^{F'm'} \langle F'\|d\|F\rangle$$
$$= (-1)^{F-1+m'} \sqrt{2F'+1} \begin{pmatrix} F & 1 & F' \\ m & q & -m' \end{pmatrix} \langle F'\|d\|F\rangle. \quad (\text{B.3})$$

We have here used the following notation for Clebsch–Gordan coefficients $C_{jm\,j'm'}^{JM} = \langle jm\,j'm'|JM\rangle$. It is convenient to define new operators $\sigma_{F'F}^+ = \mathbf{d}_{F'F}^{(+)}/\langle F'\|d\|F\rangle$. The adjoint is defined as $\sigma_{FF'}^- = (\sigma_{F'F}^+)^\dagger$. Then, the polarizability tensor operator can be expressed as $\boldsymbol{\alpha} = -\sum_{F'\tilde{F}F} (\langle \tilde{F}\|d\|F'\rangle\langle F'\|d\|F\rangle/\Delta_{F'F}) \sigma_{\tilde{F}F'}^- \otimes \sigma_{F'F}^+$. Using the above definition, one obtains

$$\sigma_{\tilde{F}F'}^- \otimes \sigma_{F'F}^+ = (2F'+1) \sum_{p,q} \sum_{n,m,m'} (-1)^{F+\tilde{F}-2+2m'}$$
$$\times \begin{pmatrix} \tilde{F} & 1 & F' \\ n & p & -m' \end{pmatrix} \begin{pmatrix} F & 1 & F' \\ m & q & -m' \end{pmatrix} |\tilde{F}n\rangle\langle Fm| \mathbf{e}_p \otimes \mathbf{e}_q^*. \quad (\text{B.4})$$

This expression can now be split into its operator and tensor part by inserting the identity [32]

$$\sum_{k=0}^{2} \sum_{l=-k}^{k} (2k+1) \begin{pmatrix} 1 & 1 & k \\ -q & p & l \end{pmatrix} \begin{pmatrix} 1 & 1 & k \\ -\tilde{q} & \tilde{p} & l \end{pmatrix} = \delta_{p\tilde{p}}\delta_{q\tilde{q}}, \quad (\text{B.5})$$

such that

$$\sigma_{\tilde{F}F'}^- \otimes \sigma_{F'F}^+ = (2F'+1)(-1)^{F+F'} \sum_{k,l} (2k+1)$$
$$\times \left[\sum_{\tilde{p},\tilde{q}} (-1)^{\tilde{q}} \begin{pmatrix} 1 & 1 & k \\ -\tilde{q} & \tilde{p} & l \end{pmatrix} \mathbf{e}_{\tilde{p}} \otimes \mathbf{e}_{\tilde{q}}^* \right]$$
$$\times \left[\sum_{n,m} (-1)^{2\tilde{F}+F-n} \sum_{p,q,m'} (-1)^{F'+2-p-m'-q} \begin{pmatrix} F' & 1 & F \\ -m' & q & m \end{pmatrix} \right.$$
$$\left. \times \begin{pmatrix} 1 & 1 & k \\ -q & p & l \end{pmatrix} \begin{pmatrix} 1 & F' & \tilde{F} \\ -p & m' & -n \end{pmatrix} |\tilde{F}n\rangle\langle Fm| \right]. \quad (\text{B.6})$$

The expression for the operator part can be further simplified by evaluating the sum over p, q and m' using the identity

$$\sum_{\mu_1,\mu_2,\mu_3} (-1)^{l_1+l_2+l_3-\mu_1-\mu_2-\mu_3} \begin{pmatrix} l_2 & l_3 & j_1 \\ -\mu_2 & \mu_3 & m_1 \end{pmatrix}$$
$$\times \begin{pmatrix} l_3 & l_1 & j_2 \\ -\mu_3 & \mu_1 & m_2 \end{pmatrix} \begin{pmatrix} l_1 & l_2 & j_3 \\ -\mu_1 & \mu_2 & m_3 \end{pmatrix}$$
$$= \begin{pmatrix} j_1 & j_2 & j_3 \\ m_1 & m_2 & m_3 \end{pmatrix} \begin{Bmatrix} j_1 & j_2 & j_3 \\ l_1 & l_2 & l_3 \end{Bmatrix}. \quad (\text{B.7})$$

Changing 3j-symbols to Clebsh–Gordan coefficients, we get

$$\sigma_{\tilde{F}F'}^- \otimes \sigma_{F'F}^+ = (2F'+1) \frac{(-1)^{F+F'+1}}{\sqrt{3(2\tilde{F}+1)}} \sum_{k=1}^{2} (2k+1)$$
$$\times \begin{Bmatrix} F & k & \tilde{F} \\ 1 & F' & 1 \end{Bmatrix} \times \sum_{l=-k}^{k} \left[\sum_{n,m} C_{Fm\,kl}^{\tilde{F}n} |\tilde{F}n\rangle\langle Fm| \right]$$
$$\times \left[\sum_{p,q} C_{1p\,kl}^{1q} \mathbf{e}_p \otimes \mathbf{e}_q^* \right]. \quad (\text{B.8})$$

---

[4] Some people use a different convention for the reduced matrix element which is related to the convention used here by $(F\|d\|F') = \sqrt{2F+1}\langle F\|d\|F'\rangle$.

Finally, the overall polarizability can be written as $\boldsymbol{\alpha} = -(2J'+1)|\langle J'\|d\|J\rangle|^2 \frac{1}{\Delta} \sum_{k,\tilde{F}} a_k^{F\tilde{F}}(\Delta)\, \mathbf{T}_{F\tilde{F}}^k$, where

$$
a_k^{F\tilde{F}}(\Delta) = -(-1)^F (2k+1) c_k \sqrt{\frac{2F+1}{3}} \sum_{F'} \frac{(-1)^{F'}(2F'+1)}{1 - \frac{\delta_{F'}}{\Delta}}
$$

$$
\times \begin{Bmatrix} J' & F' & I \\ F & J & 1 \end{Bmatrix} \begin{Bmatrix} J' & F' & I \\ \tilde{F} & J & 1 \end{Bmatrix} \begin{Bmatrix} F & k & \tilde{F} \\ 1 & F' & 1 \end{Bmatrix}. \tag{B.9}
$$

The hyperfine splitting is $\delta_{F'} = \Delta - \Delta_{F'}$. Here we have used that

$$
\langle \tilde{F}\|d^{(+)}\|F'\rangle \langle F'\|d^{(-)}\|F\rangle
$$
$$
= (-1)^{2(J'+I+F)+\tilde{F}-F'} \begin{Bmatrix} J' & F' & I \\ F & J & 1 \end{Bmatrix} \begin{Bmatrix} J' & F' & I \\ \tilde{F} & J & 1 \end{Bmatrix}
$$
$$
\times \sqrt{(2F+1)(2\tilde{F}+1)}\,(2J'+1)|\langle J'\|d\|J\rangle|^2. \tag{B.10}
$$

The coefficients c_k read

$$
c_0 = 1, \qquad c_1 = \frac{1}{\sqrt{2F(F+1)}},
$$
$$
c_2 = \frac{3}{\sqrt{10F(F+1)(2F-1)(2F+3)}}. \tag{B.11}
$$

These coefficients are chosen in such a way that the resulting Hamiltonian contains spin component as irreducible tensors. The irreducible tensors are defined as follows:

$$
\mathbf{T}_{F\tilde{F}}^k = \frac{1}{c_k} \sum_{l=-k}^{k} M_{l;F\tilde{F}}^k \sum_{p,q} C_{1pkl}^{1q} \mathbf{e}_p \otimes \mathbf{e}_q^*. \tag{B.12}
$$

For $\tilde{F} = F$, we have

$$
M_{l;FF}^k = \sum_{n,m} C_{Fmkl}^{Fn} |Fn\rangle\langle Fm|
$$

if $\tilde{F} \neq F$

$$
M_{l;F\tilde{F}}^k = \sum_{n,m} \left(C_{Fmkl}^{\tilde{F}n} |\tilde{F}n\rangle\langle Fm| + (-1)^l C_{Fm;k,-l}^{\tilde{F}n} |Fm\rangle\langle \tilde{F}n| \right). \tag{B.13}
$$

Appendix C. Transformation of the light equation of motion

We will work in the paraxial approximation assuming a flat transverse profile and write the forward electric field as

$$
\mathbf{E}_F(z,t) = \sqrt{\frac{\omega_0}{2\epsilon_0 A}} \sum_\sigma \int \frac{d^3k\, d^2\rho}{(2\pi)^3} \boldsymbol{\epsilon}_\sigma (a_{k\sigma}(t)\, e^{i(k_z z + k_\perp \rho)} + h.c.),
$$
$$
= |E| \sum_\sigma \boldsymbol{\epsilon}_\sigma (a_\sigma(z,t) + a_\sigma^\dagger(z,t)). \tag{C.1}
$$

Here, $|E| = \sqrt{\frac{\omega_0}{2\epsilon_0 A}}$ and we have defined the space-dependent operators $a_\sigma(z,t) = \int \frac{dk_z}{2\pi} a_{k_z\sigma}(t)\, e^{ik_z z}$, where it is assumed that the different k are close to k_0, so that the operators oscillate at the common frequency $\omega_0 = |k_0|$. For the radiation field, we have that

$$
H_L = \sum_\sigma \int \frac{dk}{2\pi} \omega_k a_{k\sigma}^\dagger a_{k\sigma}. \tag{C.2}
$$

Now we are to ready to form the EOM

$$
\frac{\partial}{\partial t} a_\sigma(z,t) = i[H_{int} + H_L, a_\sigma(z,t)], \tag{C.3}
$$

$$
[H_L, a_\sigma(z,t)] = \int \frac{dk}{2\pi} [H_L, a_{k\sigma}(t)]\, e^{ikz}
$$
$$
= -\int \frac{dk}{2\pi} \omega_k a_{k\sigma}(t)\, e^{ikz}. \tag{C.4}
$$

But we also have from the explicit z-dependence that

$$
\frac{\partial}{\partial z} a_\sigma(z,t) = \int \frac{dk}{2\pi} ik a_{k\sigma}(t)\, e^{ikz}, \tag{C.5}
$$

which we recognize as $-i[H_L, a_\sigma(z,t)]$. So we replace the time evolution from the radiation field by minus the derivative with respect to z and end up with

$$
\left(\frac{\partial}{\partial t} + \frac{\partial}{\partial z} \right) a_\sigma(z,t) = i[H_{int}, a_\sigma(z,t)]. \tag{C.6}
$$

This procedure is directly applicable for the Stokes operators too. In the end, we throw away the time derivative, which is the same as ignoring retardation effects, but this approximation can even be made exact by introducing a suitable rescaled time. By doing this, we have thus an equation in position and not time for light observables.

Appendix D. Calculations of atomic spin noise and decay rate coefficients for light and atoms

D.1. Light matrix α^2

The dimensionless tensor polarizability can be written as

$$
\alpha = \sum_{k,\tilde{F}} a_k^{F\tilde{F}}(\Delta)\, \mathbf{T}_{F\tilde{F}}^k. \tag{D.1}
$$

We assume that the atomic spin is polarized along the x-axis and during interaction the collective spin experiences merely small rotations. Having the quantization axis to be the x-axis, we have to linearize the spin-dependent operators around the initial state $|FF\rangle$. Using the expression for the irreducible tensors (B.12) and averaging over the initial spin state, we obtain

$$
\langle \alpha^2 \rangle = \sum_{k,k',\tilde{F}} \frac{a_k^{F\tilde{F}} a_{k'}^{F\tilde{F}}}{c_k c_{k'}} \sum_{p,q} \sum_{r,s,l,l'} C_{\tilde{F}rkl}^{FF} C_{FFk'l'}^{\tilde{F}r}
$$
$$
\times C_{1pkl}^{1s} C_{1sk'l'}^{1q} \mathbf{e}_p \otimes \mathbf{e}_q^*. \tag{D.2}
$$

We have changed the notation for Clebsch–Gordan coefficients to the shorter form $\langle jm\, j'm'|JM\rangle = C_{jm\, j'm'}^{JM}$. Taking into account restrictions on momentum projections given by Clebsch–Gordan coefficients, the sum over four indices is reduced to a sum over a single index. We have $p = q$, $l = s-q$, $l' = -l$, $r = F+q-s$, and hence

$$
\langle \alpha^2 \rangle = \sum_{k,k',\tilde{F}} \frac{a_k^{F\tilde{F}} a_{k'}^{F\tilde{F}}}{c_k c_{k'}} \sum_{q,s} C_{\tilde{F},F+q-s;k,s-q}^{FF} C_{FF;k',q-s}^{\tilde{F},F+q-s}
$$
$$
\times C_{1q;k,s-q}^{1s} C_{1s;k',q-s}^{1q} \mathbf{e}_q \otimes \mathbf{e}_q^*. \tag{D.3}
$$

The required elements of the matrix α^2 for the atomic spin aligned in the x-direction in Cartesian coordinates read

$$
\langle \alpha^2 \rangle_{xx} = \langle \alpha^2 \rangle_{00} \tag{D.4}
$$

$$
\langle \alpha^2 \rangle_{yy} = \frac{1}{2}(\langle \alpha^2 \rangle_{--} + \langle \alpha^2 \rangle_{++}) \tag{D.5}
$$

$$
\langle \alpha^2 \rangle_{xy} = 0. \tag{D.6}
$$

D.2. Atomic Langevin noise $(i[\alpha, j_i])^2$

Let us consider the matrix $\zeta_i = i[\alpha, j_i]$. The commutator of an irreducible tensor and spherical spin components reads

$$\left[M^k_{l;FF}, j_\mu\right] = -\sqrt{k(k+1)}C^{k,l+\mu}_{kl;1\mu} M^k_{l+\mu;FF}. \qquad (D.7)$$

This is true for an irreducible tensor defined for a single spin state; therefore, we consider the case of $\tilde{F} = F$:

$$\zeta^{FF}_\mu = -i\sum_k \frac{a^{FF}_k}{c_k}\sqrt{k(k+1)}\sum_{l,p,q}$$
$$\times C^{k,l+\mu}_{kl;1\mu} M^k_{l+\mu;FF}C^{1q}_{1p;kl}\mathbf{e}_p\otimes\mathbf{e}^*_q. \qquad (D.8)$$

Using the same method as in the previous subsection, one obtains

$$\langle\zeta_\mu\zeta_\nu\rangle^{FF} = -\sum_{k,k'}\frac{a^{FF}_k a^{FF}_{k'}}{c_k c_{k'}}\sqrt{k(k+1)k'(k'+1)}\sum_{p,q}\sum_{r,s,l,l'}$$
$$\times C^{k,l+\mu}_{kl;1\mu}C^{k',l'+\nu}_{k'l';1\nu}C^{FF}_{Fr;k,l+\mu}C^{Fr}_{FF;k'l'+\nu}$$
$$\times C^{1s}_{1p;kl}C^{1q}_{1s;k'l'}\mathbf{e}_p\otimes\mathbf{e}^*_q. \qquad (D.9)$$

After some transformations, one comes to the following expression:

$$\langle\zeta_\mu\zeta_\nu\rangle^{FF}_{pq} = -\sum_{k,k'}\frac{a^{FF}_k a^{FF}_{k'}}{c_k c_{k'}}\sqrt{k(k+1)k'(k'+1)}\sum_s$$
$$\times \delta_{p,q+\mu+\nu}C^{k,s-q-\nu}_{k,s-p;1\mu}C^{k',q-s+\nu}_{k',q-s;1\nu}$$
$$\times C^{FF}_{F,F+q-s+\nu;k,s-q-\nu}C^{F,F+q-s+\nu}_{FF;k',q-s+\nu}$$
$$\times C^{1s}_{1,p;k,s-p}C^{1q}_{1s;k',q-s}. \qquad (D.10)$$

The case of $\tilde{F} \neq F$ has to be considered specifically. The commutator with spin components no longer have a nice form:

$$\left[M^k_{l;F\tilde{F}}, j_\mu\right] = \sqrt{F(F+1)}\sum_{m,n,r}\left[C^{\tilde{F}n}_{Fr;kl}C^{Fr}_{Fm;1\mu}|\tilde{F}n\rangle\langle Fm|\right.$$
$$\left. -(-1)^l C^{\tilde{F}n}_{Fr;k-l}C^{Fm}_{Fr;1\mu}|Fm\rangle\langle\tilde{F}n|\right].$$

Using this expression for the commutator and doing some algebra, we obtain

$$\langle\zeta_\mu\zeta_\nu\rangle^{F\tilde{F}}_{pq} = (-1)^{s-p}\delta_{p,q+\mu+\nu}F(F+1)$$
$$\times C^{FF}_{F,F-\mu;1\mu}C^{F,F-\nu}_{FF;1\nu}\sum_{k,k'}\frac{a^{F\tilde{F}}_k a^{F\tilde{F}}_{k'}}{c_k c_{k'}}$$
$$\times \sum_s C^{\tilde{F},F+q-s+\nu}_{F,F-\mu;k,p-s}C^{\tilde{F},F+q-s+\nu}_{F,F+\nu;k',q-s}$$
$$\times C^{1s}_{1,p;k,s-p}C^{1q}_{1s;k',q-s}. \qquad (D.11)$$

The full matrix $\langle\zeta^2\rangle$ is just a sum of all of these contributions for different $\tilde{F}$:

$$\langle\zeta_\mu\zeta_\nu\rangle_{pq} = \sum_{\tilde{F}}\langle\zeta_\mu\zeta_\nu\rangle^{F\tilde{F}}_{pq}. \qquad (D.12)$$

Since the spin is prepared in a polarized state along the x direction, the spin noise coefficient for the ith spin component in the presence of the driving field polarized parallel to the spin is given by the xx components of the matrix $\langle\zeta^2_i\rangle$. The case of

the orthogonal configuration of the spin and light is described by the yy element of the matrix

$$\langle\zeta^2_x\rangle_\| = \langle\zeta_0\zeta_0\rangle_{00}, \qquad (D.13)$$
$$\langle\zeta^2_y\rangle_\| = -\frac{1}{2}\{\langle\zeta_-\zeta_+\rangle_{00} + \langle\zeta_+\zeta_-\rangle_{00}\}, \qquad (D.14)$$
$$\langle\zeta^2_z\rangle_\| = \langle\zeta^2_y\rangle_\|, \qquad (D.15)$$
$$\langle\zeta^2_x\rangle_\perp = \frac{1}{2}\{\langle\zeta_0\zeta_0\rangle_{--} + \langle\zeta_0\zeta_0\rangle_{++}\}, \qquad (D.16)$$
$$\langle\zeta^2_y\rangle_\perp = -\frac{1}{4}\{\langle\zeta_-\zeta_-\rangle_{-+} + \langle\zeta_+\zeta_+\rangle_{+-} + \langle\zeta_-\zeta_+\rangle_{--}$$
$$+ \langle\zeta_-\zeta_+\rangle_{++} + \langle\zeta_+\zeta_-\rangle_{--} + \langle\zeta_+\zeta_-\rangle_{++}\}, \qquad (D.17)$$
$$\langle\zeta^2_z\rangle_\perp = \frac{1}{4}\{\langle\zeta_-\zeta_-\rangle_{-+} + \langle\zeta_+\zeta_+\rangle_{+-} - \langle\zeta_-\zeta_+\rangle_{--}$$
$$- \langle\zeta_-\zeta_+\rangle_{++} - \langle\zeta_+\zeta_-\rangle_{--} - \langle\zeta_+\zeta_-\rangle_{++}\}. \qquad (D.18)$$

D.3. Spin decay rates and ξ_i

In this subsection, we derive expressions for the spin decay rates. The coefficients for the spin component decays are defined via linearization of the operator $\xi_i = \alpha^2 j_i + j_i\alpha^2 - 2\alpha j_i\alpha = -i[\alpha, \zeta_\mu]$ for the given polarized spin state. We want to have $\xi_i \sim \Xi_i j_i$ which implies

$$\langle\xi_i\rangle = \Xi_i\langle j_i\rangle, \qquad \langle\xi_i j_i\rangle = \Xi_i\langle j^2_i\rangle. \qquad (D.19)$$

The last equality is always nontrivial, so we can use it for defining $\Xi_i = \langle\xi_i j_i\rangle/\langle j^2_i\rangle$. Using expressions (D.1) and (D.8), we proceed

$$\xi^{FF}_\mu = -\sum_{k,k'}\frac{a^{FF}_k a^{FF}_{k'}}{c_k c_{k'}}\sqrt{k(k+1)}\sum_{p,q}\sum_{r,s,l,l'}C^{k,l+\mu}_{kl;1\mu}$$
$$\times \left[C^{Fn}_{Fr;k'l'}C^{Fr}_{Fm;k,l+\mu}C^{1s}_{1p;k'l'}C^{1q}_{1s;kl}\right.$$
$$\left. -C^{Fn}_{Fr;k,l+\mu}C^{Fr}_{Fm;k'l'}C^{1s}_{1p;kl}C^{1q}_{1s;k'l'}\right]$$
$$\times |Fm\rangle\langle Fn|\,\mathbf{e}_p\otimes\mathbf{e}^*_q. \qquad (D.20)$$

After averaging over the initial spin polarized state $|FF\rangle$, we obtain the required matrix elements in the spherical basis

$$\langle\xi_\mu j_\nu\rangle^{FF}_{pq} = -\sum_{k,k'}\frac{a^{FF}_k a^{FF}_{k'}}{c_k c_{k'}}\sqrt{k(k+1)F(F+1)}$$
$$\times \sum_s C^{k,l+\mu}_{kl;1\mu}C^{F,F+\nu}_{FF;1\nu}$$
$$\times \left[C^{FF}_{Fr;k'l'}C^{Fr}_{F,F+\nu;k,l+\mu}C^{1s}_{1p;k'l'}C^{1q}_{1s;kl}\right.$$
$$\left. -C^{FF}_{Fr;k,l+\mu}C^{Fr}_{F,F+\nu;k'l'}C^{1s}_{1p;kl}C^{1q}_{1s;k'l'}\right]. \qquad (D.21)$$

The Clebsch–Gordan coefficients require the following constraints on the indices to be fulfilled for the first term $r = F + p - s$, $l = q - s$, $l' = s - p$ and for the second term $r = F + q - s + \nu$, $l = s - p$, $l' = q - s$ plus the usual condition $p = q + \mu + \nu$. Finally, we obtain

$$\langle\xi_\mu j_\nu\rangle^{FF}_{pq} = -\delta_{p,q+\mu+\nu}\sum_{k,k'}\frac{a^{FF}_k a^{FF}_{k'}}{c_k c_{k'}}C^{F,F+\nu}_{FF;1\nu}$$
$$\times \sqrt{k(k+1)F(F+1)}\sum_s$$
$$\times \left[C^{FF}_{F,F+p-s;k',s-p}C^{F,F+p-s}_{F,F+\nu;k,q-s+\mu}\right.$$
$$\times C^{k,q-s+\mu}_{k,q-s;1\mu}C^{1s}_{1p;k',s-p}C^{1q}_{1s;k,q-s}$$
$$-C^{FF}_{F,F+q-s+\nu;k,s-p+\mu}C^{F,F+q-s+\nu}_{F,F+\nu;k',q-s}$$
$$\left. \times C^{k,s-p+\mu}_{k,s-p;1\mu}C^{1s}_{1p;k,s-p}C^{1q}_{1s;k',q-s}\right]. \qquad (D.22)$$

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In the case of $\tilde{F} \neq F$, essentially the same calculations provide us with the necessary expression

$$\langle \xi_\mu j_\nu \rangle_{pq}^{F\tilde{F}} = \delta_{p,q+\mu+\nu} \sum_{k,k'} \frac{a_k^{F\tilde{F}} a_{k'}^{F\tilde{F}}}{c_k c_{k'}} F(F+1) C_{FF;1\nu}^{F,F+\nu}$$

$$\times \sum_s (-1)^{s-p} C_{1p;k,s-p}^{1s} C_{1s;k',q-s}^{1q}$$

$$\times \left[C_{FF;k,p-s}^{\tilde{F},F-s+p} C_{F,F+\mu+\nu;k',q-s}^{\tilde{F},F-s+p} C_{F,F+\nu;1\mu}^{F,F+\mu+\nu} \right.$$

$$\left. + C_{F,F-\mu;1\mu}^{FF} C_{F,F-\mu;k,p-s}^{\tilde{F},F+q-s+\nu} C_{F,F+\nu;k',q-s}^{\tilde{F},F+q-s+\nu} \right]. \quad \text{(D.23)}$$

The full matrix $\langle \xi_\mu j_\nu \rangle$ can be found by summation over all $\tilde{F}$:

$$\langle \xi_\mu j_\nu \rangle_{pq} = \sum_{\tilde{F}} \langle \xi_\mu j_\nu \rangle_{pq}^{F\tilde{F}}. \quad \text{(D.24)}$$

We recall that the spin is prepared in a polarized state along the x direction. Then, the decay rate coefficient for j_i in the presence of the driving field polarized parallel to the spin is given by the xx-component of the matrix $\langle \xi_i j_i \rangle$. The case of the orthogonal configuration of the spin and light is described by the yy-element of the matrix:

$$\Xi_{x_\parallel} = \frac{1}{F^2} \langle \xi_0 j_0 \rangle_{00}, \quad \text{(D.25)}$$

$$\Xi_{y_\parallel} = -\frac{1}{F} \{ \langle \xi_- j_+ \rangle_{00} + \langle \xi_+ j_- \rangle_{00} \}, \quad \text{(D.26)}$$

$$\Xi_{z_\parallel} = \Xi_{y_\parallel}, \quad \text{(D.27)}$$

$$\Xi_{x_\perp} = \frac{1}{2F^2} \{ \langle \xi_0 j_0 \rangle_{--} + \langle \xi_0 j_0 \rangle_{++} \}, \quad \text{(D.28)}$$

$$\Xi_{y_\perp} = -\frac{1}{2F} \{ \langle \xi_- j_- \rangle_{-+} + \langle \xi_+ j_+ \rangle_{+-} + \langle \xi_- j_+ \rangle_{--}$$
$$+ \langle \xi_- j_+ \rangle_{++} + \langle \xi_+ j_- \rangle_{--} + \langle \xi_+ j_- \rangle_{++} \}, \quad \text{(D.29)}$$

$$\Xi_{z_\perp} = \frac{1}{2F} \{ \langle \xi_- j_- \rangle_{-+} + \langle \xi_+ j_+ \rangle_{+-} - \langle \xi_- j_+ \rangle_{--}$$
$$- \langle \xi_- j_+ \rangle_{++} - \langle \xi_+ j_- \rangle_{--} - \langle \xi_+ j_- \rangle_{++} \}. \quad \text{(D.30)}$$

D.4. Relation between the spin decay and the spin noise

Let us consider the commutator of the Langevin noises for spin-polarized atoms $\langle [F_i, F_j] \rangle$. The noise is defined by the matrix $\zeta_i = i[\alpha, j_i]$ calculated in appendix D.2. Therefore, one can consider the mean value of the commutator $\langle [\zeta_y, \zeta_z] \rangle = -\langle [[\alpha, j_y], [\alpha, j_z]] \rangle$. On the other hand, the spin decay rate is given by the operator $\xi_i = \alpha^2 j_i + j_i \alpha^2 - 2\alpha j_i \alpha$. In the previous subsection of the appendix, we have shown that for spin-polarized atoms, the operator is proportional to the spin projection $\langle \xi_i \rangle \sim \Xi_i \langle j_i \rangle$. Using this fact one can show that

$$\langle [\zeta_y, \zeta_z] \rangle = \frac{i}{2} (\Xi_y + \Xi_z - \Xi_x) \langle j_x \rangle. \quad \text{(D.31)}$$

This holds true for any cyclic permutation of x, y, z.

Appendix E. Expressions for ^{133}Cs

In this section, we evaluate the A, B, C coefficients for ^{133}Cs optically pumped to the ground state $F = 4$. In this case, we have $I = 7/2$, $J = 1/2$, $J' = 3/2$, $F = 4$, $\tilde{F} = 3$. In order to simplify the expressions, we use the following notations: a_k for a_k^{FF} and b_k for $a_k^{F\tilde{F}}$:

$$a_0 = \frac{7}{144} \left(\frac{44}{21} + \frac{1}{1 - \frac{\Delta_{45}}{\Delta}} + \frac{1}{3 \left(1 - \frac{\Delta_{35}}{\Delta} \right)} \right), \quad \text{(E.1)}$$

$$a_1 = \frac{7}{5760} \left(\frac{176}{7} - \frac{3}{1 - \frac{\Delta_{45}}{\Delta}} - \frac{5}{1 - \frac{\Delta_{35}}{\Delta}} \right), \quad \text{(E.2)}$$

$$a_2 = \frac{1}{5760} \left(16 - \frac{21}{1 - \frac{\Delta_{45}}{\Delta}} + \frac{5}{1 - \frac{\Delta_{35}}{\Delta}} \right), \quad \text{(E.3)}$$

$$b_1 = \frac{1}{128\sqrt{5}} \left(\frac{5}{1 - \frac{\Delta_{45}}{\Delta}} + \frac{3}{1 - \frac{\Delta_{35}}{\Delta}} \right), \quad \text{(E.4)}$$

$$b_2 = \frac{3}{128\sqrt{77}} \left(\frac{1}{1 - \frac{\Delta_{45}}{\Delta}} - \frac{1}{1 - \frac{\Delta_{35}}{\Delta}} \right). \quad \text{(E.5)}$$

Light coefficients A_μ:

$$A_x = \frac{1}{4a_1^2} \left[a_0^2 + 4a_1^2 + 56a_1 a_2 - \frac{112}{3} a_0 a_2 + \frac{4900}{9} a_2^2 \right.$$
$$\left. + \frac{140}{9} \left(b_1^2 + \frac{77}{5} b_2^2 + 2\sqrt{\frac{77}{5}} b_1 b_2 \right) \right], \quad \text{(E.6)}$$

$$A_y = \frac{1}{4a_1^2} \left[a_0^2 + 18a_1^2 - 28a_1 a_2 + \frac{56}{3} a_0 a_2 + \frac{2170}{9} a_2^2 \right.$$
$$\left. + \frac{70}{9} \left(b_1^2 + \frac{539}{15} b_2^2 - 2\sqrt{\frac{77}{5}} b_1 b_2 \right) \right]. \quad \text{(E.7)}$$

Spin decay coefficients B_i:

$$B_{x_\parallel} = \frac{1}{2a_1^2} \left[a_1^2 + 14a_2 a_1 + 49a_2^2 \right.$$
$$\left. + \frac{140}{9} \left(b_1^2 + \frac{77}{5} b_2^2 + 2\sqrt{\frac{77}{5}} b_1 b_2 \right) \right], \quad \text{(E.8)}$$

$$B_{y_\parallel} = \frac{1}{4a_1^2} \left[a_1^2 - 98a_2 a_1 + 273a_2^2 \right.$$
$$\left. + \frac{245}{9} \left(b_1^2 + \frac{55}{3} b_2^2 + 2\sqrt{\frac{55}{7}} b_1 b_2 \right) \right], \quad \text{(E.9)}$$

$$B_{z_\parallel} = B_{y_\parallel}, \quad \text{(E.10)}$$

$$B_{x_\perp} = \frac{1}{4a_1^2} \left[a_1^2 - 14a_2 a_1 + 105a_2^2 \right.$$
$$\left. + \frac{140}{9} \left(b_1^2 + \frac{539}{15} b_2^2 - 2\sqrt{\frac{77}{5}} b_1 b_2 \right) \right], \quad \text{(E.11)}$$

$$B_{y_\perp} = \frac{1}{2a_1^2}\left[a_1^2 + 56a_2a_1 - 35a_2^2\right.$$

$$\left. + \frac{175}{18}\left(b_1^2 + \frac{693}{25}b_2^2 - \frac{2}{5}\sqrt{\frac{77}{5}}b_1b_2\right)\right], \tag{E.12}$$

$$B_{z_\perp} = B_{x_\perp}. \tag{E.13}$$

Spin noise coefficients C_i:

$$C_{x_\parallel} = \frac{1}{a_1^2}\left[a_1^2 + 14a_2a_1 + 49a_2^2\right.$$

$$\left. + \frac{560}{9}\left(b_1^2 + \frac{77}{5}b_2^2 + 2\sqrt{\frac{77}{5}}b_1b_2\right)\right], \tag{E.14}$$

$$C_{y_\parallel} = \frac{1}{a_1^2}\left[4a_1^2 + 308a_2^2\right.$$

$$\left. + \frac{35}{6}\left(b_1^2 + \frac{1001}{45}b_2^2 + \frac{2}{3}\sqrt{\frac{77}{5}}b_1b_2\right)\right], \tag{E.15}$$

$$C_{z_\parallel} = C_{y_\parallel}, \tag{E.16}$$

$$C_{x_\perp} = \frac{1}{2a_1^2}\left[a_1^2 - 14a_2a_1 + 161a_2^2\right.$$

$$\left. + \frac{560}{9}\left(b_1^2 + \frac{539}{15}b_2^2 - 2\sqrt{\frac{77}{5}}b_1b_2\right)\right], \tag{E.17}$$

$$C_{y_\perp} = \frac{1}{2a_1^2}\left[9a_1^2 - 14a_2a_1 + 63a_2^2\right.$$

$$\left. + \frac{175}{18}\left(b_1^2 + \frac{693}{25}b_2^2 - \frac{2}{5}\sqrt{\frac{77}{5}}b_1b_2\right)\right], \tag{E.18}$$

$$C_{z_\perp} = \frac{1}{2a_1^2}[a_1^2 + 14a_2a_1 + 651a_2^2$$

$$+ \frac{175}{18}(b_1^2 + \frac{693}{25}b_2^2 - \frac{2}{5}\sqrt{\frac{77}{5}}b_1b_2)]. \tag{E.19}$$

References

[1] Kimble H J 2008 The quantum internet *Nature* **453** 1023–30
[2] Gorshkov A, André A, Lukin M and Sørensen A 2007 Photon storage in-type optically dense atomic media: i. Cavity model *Phys. Rev.* A **76** 033804
[3] Gorshkov A, André A, Lukin M and Sørensen A 2007 Photon storage in-type optically dense atomic media: ii. Free-space model *Phys. Rev.* A **76** 033805
[4] Gorshkov A, André A, Lukin M and Sørensen A 2007 Photon storage in-type optically dense atomic media: iii. Effects of inhomogeneous broadening *Phys. Rev.* A **76** 033806
[5] Tittel W, Afzelius M, Chaneliére T, Cone R, Kröll S, Moiseev S and Sellars M 2010 Photon-echo quantum memory in solid state systems *Laser and Photonics Rev.* **4** 244–67
[6] Hammerer K, Sørensen A S and Polzik E S 2010 Quantum interface between light and atomic ensembles *Rev. Mod. Phys.* **82** 1041–93
[7] Sangouard N, Simon C, Riedmatten H de and Gisin N 2011 Quantum repeaters based on atomic ensembles and linear optics *Rev. Mod. Phys.* **83** 33–80
[8] Lvovsky A I, Sanders B C and Tittel W 2009 Optical quantum memory *Nature Photon.* **3** 706–14
[9] Kuzmich A, Mandel L and Bigelow N 2000 Generation of spin squeezing via continuous quantum nondemolition measurement *Phys. Rev. Lett.* **85** 1594–7
[10] Takano T, Fuyama M, Namiki R and Takahashi Y 2009 Spin squeezing of a cold atomic ensemble with the nuclear spin of one-half *Phys. Rev. Lett.* **102** 033601
[11] Mikhailov E E and Novikova I 2008 Low-frequency vacuum squeezing via polarization self-rotation in Rb vapor *Opt. Lett.* **33** 1213–5
[12] Agha I H, Messin G and Grangier P 2010 Generation of pulsed and continuous-wave squeezed light with ^{87}Rb vapor *Optics Express* **18** 4198–205
[13] Julsgaard B, Kozhekin A and Polzik E S 2001 Experimental long-lived entanglement of two macroscopic objects *Nature* **413** 400–3
[14] Krauter H, Muschik C, Jensen K, Wasilewski W, Petersen J, Cirac J I and Polzik E S 2011 Entanglement generated by dissipation and steady state entanglement of two macroscopic objects *Phys. Rev. Lett.* **107** 080503
[15] Sherson J F, Krauter H, Olsson R K, Julsgaard B, Hammerer K, Cirac J I and Polzik E S 2006 Quantum teleportation between light and matter *Nature* **443** 557–60
[16] Julsgaard B, Sherson J, Cirac J I, Fiurasek J and Polzik E S 2004 Experimental demonstration of quantum memory for light *Nature* **432** 482–6
[17] Jensen K, Wasilewski W, Krauter H, Fernholz T, Nielsen B M, Owari M, Plenio M B, Serafini A, Wolf M M and Polzik E S 2010 Quantum memory for entangled continuous-variable states *Nature Phys.* **7** 13–6
[18] Smith G A, Chaudhury S, Silberfarb A, Deutsch I H and Jessen P S 2004 Continuous weak measurement and nonlinear dynamics in a cold spin ensemble *Phys. Rev. Lett.* **93** 163602
[19] Chaudhury S, Smith G A, Schulz K and Jessen P S 2006 Continuous nondemolition measurement of the Cs clock transition pseudospin *Phys. Rev. Lett.* **96** 043001
[20] Deutsch I H and Jessen P S 2010 Quantum control and measurement of atomic spins in polarization spectroscopy *Opt. Commun.* **283** 681–94
[21] Wasilewski W, Jensen K, Krauter H, Renema J J, Balabas M V and Polzik E S 2010 Quantum noise limited and entanglement-assisted magnetometry *Phys. Rev. Lett.* **104** 133601
[22] Napolitano M and Mitchell M W 2010 *New J. Phys.* **12** 093016
[23] Napolitano M, Koschorreck M, Dubost B, Behbood N, Sewell R J and Mitchell M W 2011 *Nature* **471** 486–9
[24] Duan L-M, Cirac J I, Zoller P and Polzik E S 2000 Quantum communication between atomic ensembles using coherent light *Phys. Rev. Lett.* **85** 5643–6
[25] Happer W and Mathur B 1967 Off-resonant light as a probe of optically pumped alkali vapors *Phys. Rev. Lett.* **18** 577–80
[26] Happer W 1972 Optical pumping *Rev. Mod. Phys.* **44** 169–249
[27] Hammerer K, Mølmer K, Polzik E S and Cirac J I 2004 Light–matter quantum interface *Phys. Rev.* A **70** 044304
[28] Madsen L B and Mølmer K 2004 *Phys. Rev.* A **70** 052324
[29] Koschorreck M and Mitchell M W 2009 *J. Phys. B: At. Mol. Opt. Phys.* **42** 195502
[30] Sørensen M W and Sørensen A S 2008 Three-dimensional theory for light–matter interaction *Phys. Rev.* A **77** 013826
[31] Julsgaard B 2003 Entanglement and quantum interactions with macroscopic gas samples *PhD Thesis* Aarhus University
[32] Hammerer K 2006 Quantum information processing with atomic quantum information processing with atomic ensembles and light *PhD Thesis* Technische Universität München, Max-Planck-Institut für Quantenoptik
[33] Vasilyev D V, Sokolov I V and Polzik E S 2008 Quantum memory for images: a quantum hologram *Phys. Rev.* A **77** 020302

[34] Vasilyev D V and Sokolov I V 2011 Double-pass quantum volume hologram *Phys. Rev.* A **83** 053851

[35] Wasilewski W, Fernholz T, Jensen K, Madsen L S, Krauter H, Muschik C and Polzik E S 2009 Generation of two-mode squeezed and entangled light in a single temporal and spatial mode *Optics Express* **17** 14444–57

[36] Mishina O, Kupriyanov D and Polzik E 2005 arXiv:quant-ph/0509220v1

[37] Mishina O S and Kupriyanov D V 2007 *Phys. Rev.* A **75** 042326

[38] Giedke G 2001 Quantum information and continuous variable systems *PhD Thesis* Institut für Theoretische Physik der Leopold-Franzens–Universität Innsbruck

[39] Hammerer K, Polzik E S and Cirac J I 2006 High-fidelity teleportation between light and atoms *Phys. Rev.* A **74** 064301

[40] Kolobov M I 1999 The spatial behavior of nonclassical light *Rev. Mod. Phys.* **71** 1539–89