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PAPER

Optimal photon generation from spontaneous Raman processes in cold atoms

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Abstract

Spontaneous Raman processes in cold atoms have been widely used in the past decade for generating single photons. Here, we present a method to optimise their efficiencies for given atomic coherences and optical depths. We give a simple and complete recipe that can be used in present-day experiments, attaining near-optimal single photon emission.

1. Introduction

On-demand single photon sources are appealing ingredients for many quantum information tasks. Examples include the distribution of entanglement over long distances using quantum repeaters or quantum communications with security guarantees which remain valid, independent of the details of the actual implementation [1, 2]. These tasks necessitate stringent purity and efficiency requirements on the performance of the single photon sources used. Techniques based on spontaneous Raman processes in cold atoms are among the most advanced single-photon sources with such characteristics. The basic principle is to use an ensemble of three-level atoms in a Λ-configuration and two pulsed laser fields (see figure 1(a)). The first write pulse—the write control field—off-resonantly excites one transition, which can spontaneously produce a frequency-shifted photon—the write photon field—along the second transition through a Raman process. Since all the interacting atoms participate in the process, and there is no information about which atom emitted the photon, the detection of this write photon heralds the existence of a single delocalised excitation across the sample—an atomic spin wave. Once the spin wave has been prepared, the atomic sample is ready to be used as a source, and a second pulse—the read control field—along the second transition performs a conversion of the atomic spin wave into a second photon—the read photon field. If the duration of the process is short enough with respect to the atomic coherence times, and the optical depth of the sample sufficiently high, then the read photon is emitted efficiently in a well defined mode and the protocol provides a viable single photon source.

Such sources have been at the core of numerous experiments during the last decade following the seminal paper of Duan, Lukin, Cirac and Zoller [3], showing how they could be used for long-distance quantum communication based on quantum repeater architectures (for reviews, see [4–7]). Recently, they have been used as quantum memories with storage times up to 200 ms [8, 9] or as a source producing pure single photons with a temporal duration that can be varied over up to 3 orders of magnitude while maintaining constant efficiencies [10]. We stress that the efficiency of such a source is a critical parameter for the implementation of efficient quantum repeater architectures. While very high efficiencies of $\sim$90% are essential, a reduction of the source efficiency by 1% can reduce the repeater distribution rate by 10%–20%, depending on the specific architecture [4].

Several solutions can be envisioned to ensure high efficiencies. One solution relies on the use of an optical cavity to enhance the spinwave–light conversion efficiency. Experimental efforts along this direction have

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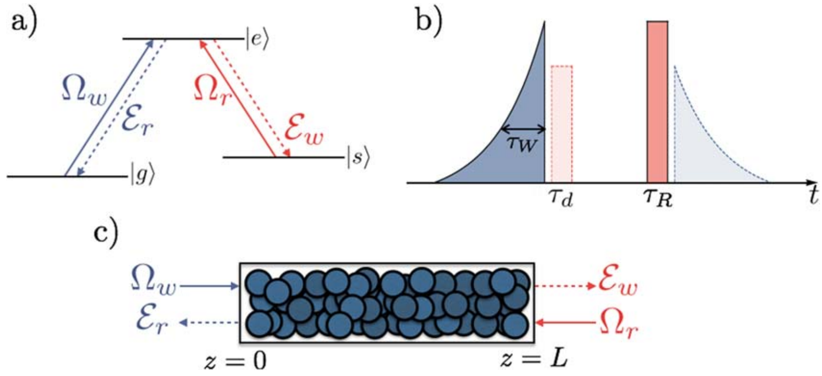

Figure 1. Λ energy level scheme and schematic of the proposed single photon source. (a) Write (read) control fields are indicated with Rabi frequencies Ω_W (Ω_R) and write (read) photon fields are indicated with quantum fields $\mathcal{E}_w$ ($\mathcal{E}_r$), each along their respective transitions. In our model the excited level $|e\rangle$ is capable of spontaneous emission to the metastable states $|g\rangle$ and $|s\rangle$. (b) A schematic of the protocol indicates the sequence of events. A fast resonant write control field of duration τ_W followed by a write photon field detection in a short time window τ_d heralds a spatially varying spin wave. A fast π-pulse of duration τ_R then enables the retrieval of the stored excitation. Laser pulses are shaded darker to indicate their stronger intensities as compared to the weaker photon emissions. (c) A backward retrieval configuration with counterpropagating control fields results in photon field emissions in opposing directions.

resulted in efficiencies of up to 84% [11, 12]. An alternative solution involves increasing the atomic density in order to obtain a larger optical depth [13]. This however makes operations like optical pumping and noise free operations more challenging. This naturally raises the following question: *What is the optimal efficiency that can be achieved with a bulk atomic ensemble having a certain optical depth?* This question has been previously addressed for memory protocols where single photons are first absorbed before subsequently retrieved in a well defined mode [14, 15].

Inspired by these works, we first examine the conditions on the spin wave shape for achieving optimal photon retrieval efficiencies given the optical depths and specified energy levels in the atomic species. We observe that the optimal spin waves are decreasing functions in space whose decay depends on the optical depth. The intuition is that the reemission process is a collective effect in which the fields emitted by each atom interfere with each other. The best possible way for these fields to add up constructively is that the field amplitude increases as it propagates into the medium. After finding the optimal spin wave shapes, we recognise that current approaches using off-resonant write control fields create non-ideal flat spin excitations in the sample (previously studied in works such as [16]), since such control fields do not experience significant intensity depletion during propagation. To achieve better retrieval efficiencies, we propose a concrete solution (see figure 1(b)) to spatially shape the spin wave using resonant, temporally shaped write control fields. Combined with fast read control fields during the retrieval process, we show that our recipe achieves near-optimal retrieval efficiencies.

This paper is structured as follows: in the first section we discuss the optimal retrieval efficiency from a spin excitation. For completeness, we first quickly review derivations in [15] that allow us to find the expression for the retrieval efficiency of a complete retrieval process, where we begin with only $|g\rangle - |s\rangle$ coherences and transfer all atoms to $|g\rangle$. We then find the shapes of the spin excitation that yield the optimal retrieval efficiency when complete retrieval is performed. In the second section we propose the use of a resonant write control field to create heralded spin excitations similar to those that allow for optimal retrieval. We then give explicit expressions for retrieval when using a quick read control field with a constant Rabi frequency. Finally, we include a feasibility study in the case of a gas of Rubidium-87.

2. Optimal retrieval

2.1. Efficiency of a complete retrieval process

We first review a derivation in [15] giving the dependency of the efficiency of the retrieval process on the relevant quantities in the physical setup. This yields an expression for the retrieval efficiency, that depends only on the shape of the spin wave from which the retrieval is performed, and on the optical depth of the relevant transition.

We emphasise that the work in [15] focuses on absorptive memory protocols where a field is first absorbed in an atomic medium, creating a spin wave that can be read out later to re-emit the field in a well defined spatio-temporal mode. To justify the relation to [15], in our proposal the spin wave creation is instead heralded by the detection of the write photon field, but the readout process is analogous, allowing us to make use of [15] to deduce the spin wave shapes that maximise the retrieval efficiency.

We consider a three-level atomic system in a Λ-configuration (see figure 1(a)) with spin excitations present in the form of $|g\rangle - |s\rangle$ coherences. In the situation where almost all the atoms remain in $|g\rangle$ and in a rotating frame, the backward wave propagation equation (see figure 1(a)) along with the Heisenberg–Langevin equations of motion yield

$$\partial_z \mathcal{E}_r(z, t) = -\mathrm{i}\sqrt{\frac{d\gamma_{eg}}{cL}}P(z, t),$$
$$\partial_t P(z, t) = -(\gamma_{eg} + \mathrm{i}\Delta)P(z, t) + \mathrm{i}\sqrt{\frac{d\gamma_{eg}c}{L}}\mathcal{E}_r(z, t) + \mathrm{i}\Omega_R(t)S(z, t) + F_P(z, t),$$
$$\partial_t S(z, t) = -\gamma_0 S(z, t) + \mathrm{i}\Omega_R^*(t)P(z, t) + F_S(z, t), \tag{1}$$

where $P(z, t) = \sqrt{N}\sigma_{ge}(z, t)\mathrm{e}^{-\mathrm{i}\omega_1\frac{L-z}{c}}$ and $S(z, t) = \sqrt{N}\sigma_{gs}(z, t)\mathrm{e}^{-\mathrm{i}(\omega_1-\omega_2)\frac{L-z}{c}}$ are rescaled and slowly varying atomic operators (see appendix for details), with ω_1 (ω_2) referring to the energy transition of the $|e\rangle - |g\rangle$ ($|e\rangle - |s\rangle$) transition. γ_{eg} (γ_0) refers to the decay rate of the $|e\rangle - |g\rangle$ ($|g\rangle - |s\rangle$) transition. L denotes the length of the atomic sample and N the number of atoms within this sample. F_S and F_P indicate the noise operators associated to S and P, respectively. Ω_{R} (Δ) refers to the Rabi frequency (detuning) of the classical write control field on the $|e\rangle - |g\rangle$ transition, and $\mathcal{E}_{\mathrm{r}}$ denotes the quantum field of the retrieval emission. The optical depth d characterises the absorption of resonant light in the sample, such that the outgoing light intensity is $I_0(z = L) = e^{-2d}I(z = 0)$, valid when the spectrum of the incoming light is well contained within the atomic bandwidth.

Here, we consider the situation where retrieval is completed well within the spin wave decoherence time, and thus ignore γ_0. In computing the spin and photon numbers, we also ignore the Langevin noise terms F_S and F_P as they appear in normal ordered form, and in the situation where almost all the atoms are in the ground state these do not contribute.

Defining first the reversed functions $\bar{P}(L - z, t) = P(z, t)$, $\bar{S}(L - z, t) = S(z, t)$ and $\bar{\mathcal{E}}_{\mathrm{r}}(L - z, t) = \mathcal{E}_{\mathrm{r}}(z, t)$, then taking the Laplace transforms of equations (1) from $L - z = z' \to u$, we begin with the following set of transformed equations

$$\bar{\mathcal{E}}_{\mathrm{r}}(u, t) = \mathrm{i}\sqrt{\frac{\gamma_{eg}d}{cL}}\frac{1}{u}\bar{P}(u, t), \tag{2}$$

$$\partial_t \bar{P}(u, t) = -\left[\gamma_{eg}\left(1 + \frac{d}{Lu}\right) + \mathrm{i}\Delta\right]\bar{P}(u, t) + \mathrm{i}\Omega_{\mathrm{R}}(t)\bar{S}(u, t) \tag{3}$$

$$\partial_t \bar{S}(u, t) = \mathrm{i}\Omega_{\mathrm{R}}^*(t)\bar{P}(u, t). \tag{4}$$

From equations (3) and (4) we first obtain the following result

$$\frac{\mathrm{d}}{\mathrm{d}t}(\langle\bar{P}^\dagger(u_1, t)\bar{P}(u_2, t) + \bar{S}^\dagger(u_1, t)\bar{S}(u_2, t)\rangle) = \gamma_{eg}\left(-2 - \frac{d}{Lu_1} - \frac{d}{Lu_2}\right)\langle\bar{P}^\dagger(u_1, t)\bar{P}(u_2, t)\rangle. \tag{5}$$

With equation (2) we can then rewrite the number of emitted photons η in terms of $P(u, t)$

$$\eta = \frac{c}{L}\int_0^\infty \mathrm{d}t\ \langle\mathcal{E}_r^\dagger(z = 0, t)\mathcal{E}_r(z = 0, t)\rangle = \frac{c}{L}\mathcal{L}_2^{-1}\int_0^\infty \mathrm{d}t\ \frac{\gamma_{eg}d}{cL}\frac{1}{u_1u_2}\langle\bar{P}^\dagger(u_1, t)\bar{P}(u_2, t)\rangle\Bigg|_{\substack{z_1'\to L\\ z_2'\to L}},$$

where $\mathcal{L}_2^{-1}$ indicates the instruction to take the Laplace inverses of both u_1 and u_2 separately. With the use of equation (5) we can next rewrite $\langle\bar{P}^\dagger(u_1, t)\bar{P}(u_2, t)\rangle$ as a full derivative and perform the integral to get

$$\eta = \mathcal{L}_2^{-1}\frac{d}{L}\frac{-1}{(u_1 + u_2)d + 2Lu_1u_2} \times (\langle\bar{P}^\dagger(u_1, t)\bar{P}(u_2, t)\rangle + \langle\bar{S}^\dagger(u_1, t)\bar{S}(u_2, t)\rangle)\,|_{t=0}^{\infty}\,|_{\substack{z_1'\to L\\ z_2'\to L}} = \frac{1}{L^2}\mathcal{L}_2^{-1}\frac{dL}{d(u_1 + u_2) + 2Lu_1u_2}\langle\bar{S}^\dagger(u_1, 0)\bar{S}(u_2, 0)\rangle\Bigg|_{\substack{z_1'\to L\\ z_2'\to L}},$$

where the last equality comes from the conditions we assume in a complete retrieval process, i.e. that we begin with only $|g\rangle - |s\rangle$ coherences and at the end of the process all atoms are in $|g\rangle$. By performing the inverse

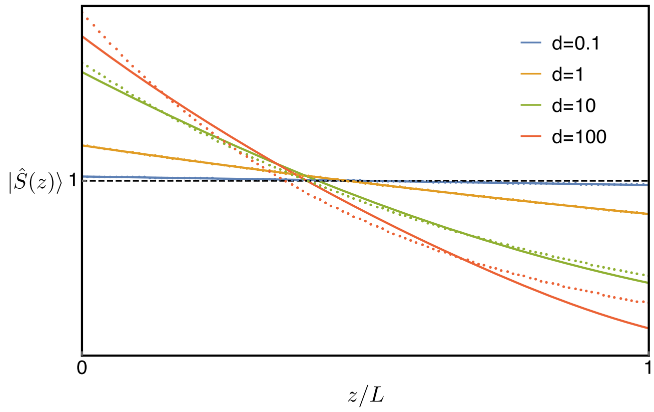

Figure 2. Optimised spin wave shapes for retrieval in the backward direction (solid lines) when compared to the best fitting exponential shapes created by our resonant write protocol (dotted lines).

Laplace transforms one sees that for complete retrieval in the backward direction[6],

$$\eta = \frac{1}{L}\int_0^L \mathrm{d}z_1 \frac{1}{L}\int_0^L \mathrm{d}z_2\ k_r(L - z_1, L - z_2) \times \langle S^\dagger(L - z_1, 0)S(L - z_2, 0)\rangle, \tag{6}$$

where $k_r(z_1, z_2) = \frac{d}{2}\mathrm{e}^{-d\frac{z_1+z_2}{2L}}I_0\left(\frac{d}{L}\sqrt{z_1 z_2}\right)$ and $I_n(x)$ indicates the modified nth Bessel function of the first kind. We proceed by considering the situation where there is originally a single spin wave in the sample (such that $\frac{1}{L}\int_0^L S^\dagger(z, 0)S(z, 0)\ \mathrm{d}z = 1$), and thus interpret η as the efficiency of the retrieval process. The retrieval efficiency η is independent of the details of the read control field used, and is a result of the ratio between desired and undesired modes that are retrieved from the spin wave.

2.2. Optimal spin shapes for complete retrieval

Having shown the dependence of the retrieval efficiency on the spin wave shape and optical depth, we now look to gain some intuition on how one might obtain the optimal retrieval efficiencies, by plotting the spin shapes that maximise the retrieval efficiency for given depths.

To do this, we recognise equation (6) as the continuous form of a product of discretised versions of k_r (in the form of a matrix) and $|S\rangle$ (in the form of a vector). Cast in this light, this integral can be computed by performing a matrix multiplication between the discretised versions of k_r and $|S\rangle$, and in this discrete approximation the optimal spin shape is the eigenvector of k_r with the largest eigenvalue. One can then interpolate the resulting vector to obtain optimised spin shapes, which are shown as solid coloured lines in figure 2.

The best spin shapes for optimal retrieval show a clear spatial dependence with a bias (depending on the optical depth d) towards placing larger excitation probabilities towards the retrieval direction (backwards in this case). Intuitively, we see these shapes representing the best way to obtain constructive interference throughout the retrieval process. As the optical field is converted from the spin wave towards the retrieval direction, it benefits from encountering a higher excitation from the atoms it next encounters. We will denote the retrieval efficiencies from these optimal spin shapes as η^*.

3. Practical recipe for achieving near-optimal retrieval efficiencies

3.1. Heralding spatially varying excitations from write photon detections

In the previous section, we have outlined how the retrieval efficiency depends on the shape of the given spin excitation, and also how the optimal spin shapes can be computed. Here we propose a method of conveniently creating spin shapes that yield near-optimal retrieval efficiencies. In contrast to creating spin excitations using spontaneous Raman processes enabled by far-detuned write control fields, we explore the use of resonant control fields instead, which create spin excitations with significantly position-dependent excitation profiles. These profiles can be controlled by tuning the duration of the write control field, which is in turn related to the

[6] In [15], equation (6) is said to describe the *optimal retrieval efficiency* from a given spin wave. For us, we see this retrieval efficiency function as a description of *complete retrieval* in the absence of spin wave decoherence, which is made optimal only when provided with the correct spin excitation.

frequency spread. A shorter (longer) write field duration implies that it has a wider (sharper) spread in frequency, and this thus affects how quickly the write pulse is depleted within the sample.

We give a detailed derivation of the write process in appendix B.1. To summarise (see figure 1(b)), beginning with all atoms in the $|g\rangle$-level, we send a short rising exponential resonant write pulse with Rabi frequency $\Omega_W(0,\, t) = \Omega_W^{\max} e^{t/\tau_W}$ that does not significantly excite the atoms to the $|e\rangle$ level ($\Omega_W^{\max} \tau_W \ll 1$). If sent with a sufficiently short duration ($\tau_W \ll 1/\gamma_{eg}, \tau_W \ll 1/\gamma_{es}$) and shut off at $t = 0$, one can consider only the dynamics along the $|g\rangle - |e\rangle$ transition, and obtain atomic coherences of the form (see appendix B.2)

$$\sigma_{ge}(z,\, 0) = e^{ik_w \cdot z}\theta_0 e^{-\frac{\alpha z}{2}}, \tag{7}$$

where $\theta_0 = i\frac{\Omega_W^{\max}\tau_W}{1+\gamma_{eg}\tau_W}$, $\alpha/2 = d\frac{\gamma_{eg}\tau_W}{1+\gamma_{eg}\tau_W}\frac{1}{L}$ and k_w indicates the wave vector for the write photon, which is described using a quantum field $\mathcal{E}_w$. Immediately after the preparation, we look for the detection of write photons within a short detection window τ_d as a herald for single spin excitations. This avoids potential dephasing effects from the decoherence of the $|e\rangle$ level. In this short detection window of duration $\tau_d \ll \min\left(\frac{1}{2\gamma_{es}},\, \frac{1}{2\gamma_{eg}}\right)$, and where $\tau_d \ll \left\{\bar{d}\ \gamma_{es}|\theta_0|^2\frac{1-e^{-\alpha L}}{\alpha L}\right\}^{-1}$, ensuring the number of emitted write photons n_w is much smaller than 1, we obtain (see appendices B.3 and B.4)

$$n_w = (\bar{d}\ \gamma_{es}\tau_d)|\theta_0|^2\frac{1-e^{-\alpha L}}{\alpha L}, \tag{8}$$

where γ_{es} ($\bar{d}$) refers to the decay rate (optical depth) of the $|e\rangle - |s\rangle$ transition. The write photon number η_w is simply the product of $\bar{d}\ \gamma_{es}\tau_d$ and the fraction of excited atoms (averaged across the sample).

In this same regime for τ_d, to leading order the corresponding heralded spin state is (see appendix B.5)

$$S^\dagger(z,\, \tau_d) = -i\sqrt{\frac{\bar{d}\gamma_{es}c}{L}}\,\theta_0 e^{-\alpha z/2}\int_0^{\tau_d} e^{-\gamma_0(\tau_d - t_a)}\mathcal{E}_w(0,\, t_a)\, dt_a, \tag{9}$$

which has an exponentially decaying spatial dependence from the $z = 0$ side of the sample. The extent of this spatial decay is characterised by α, which does depend on the given properties of the atomic sample, but can be controlled by varying the write control field duration τ_W. One can compare this class of heralded spin shapes (created from exponentially rising write pulses) to the optimal spin shapes in figure 2.

3.2. Performing fast retrieval

We now proceed with the retrieval process, and spell out the exact requirements for a certain implementation of retrieval—the fast π-pulse using a square waveform of duration τ_R. Once again, we focus on retrieval processes completed well within the spin wave decoherence time and performed under relevant experimental conditions. We thus ignore both the spin decoherence and Langevin noise terms in equation (1). Here we have implicitly assumed that the energy levels of the $|g\rangle$ and $|s\rangle$ levels are degenerate[7]. See [15, 17] for details.

With a resonant square retrieve pulse in the backward direction (see figure 1(c)) one finds the following simple expression for the dynamics of the spin wave (details given in appendix A.1)

$$\ddot{\bar{S}}(u,\, t) + A\dot{\bar{S}}(u,\, t) + B\bar{S}(u,\, t) = 0, \tag{10}$$

where $A = \gamma_{eg}\left(1 + \frac{d}{Lu}\right)$ and $B = \Omega_R^2$ (for real Ω_R), and we have taken the Laplace transform $L - z = z' \to u$.

In the regime[8] where $2\Omega_R \gg \gamma_{eg}(1 + d)$, we find $4B \gg A^2$, and obtain the following solution

$$\bar{S}(u,\, t) = e^{-At/2}\cos(\Omega_R t)\bar{S}(u,\, t = \tau_d), \tag{11}$$

which yields the following expression

$$\begin{aligned}\bar{P}(u,\, t) &= \frac{1}{i\Omega_R}\partial_t\bar{S}(u,\, t)\\ &= \frac{i}{\Omega_R}e^{-\frac{A}{2}t}\left(\frac{A}{2}\cos(\Omega_R t) + \Omega_R\sin(\Omega_R t)\right)\bar{S}(u,\, t = \tau_d),\end{aligned} \tag{12}$$

where we then see that with a sufficiently fast π-pulse (such that $2\Omega_R\tau_R = \pi$) obeying $\gamma_{eg}(1 + d)\tau_R \ll 2$, one can convert S to P without loss, yielding

[7] The phase-matching condition in one dimension is fully satisfied for co-propagating pulses and emissions, even in the non-degenerate case. For counterpropagating strategies like the one we suggest, one requires the condition $|\Delta k|L \ll 1$, where $\Delta k = k_W - k_w (=k_R - k_r)$ refers to the difference in wave vector along our 1-dimensional system for the write (read) control and photon fields (see appendix C).

[8] In considering the lossless preparation of $\bar{P}(u,\, t)$ from $\bar{S}(u,\, t = \tau_d)$, requiring $2\Omega_R \gg \gamma_{eg}(1 + d)$ for the π-pulse can be demanding. However, we show in appendix A.3 that one can achieve the same retrieval efficiency even in the slow readout regime where we do not separate the P preparation process from the emission.

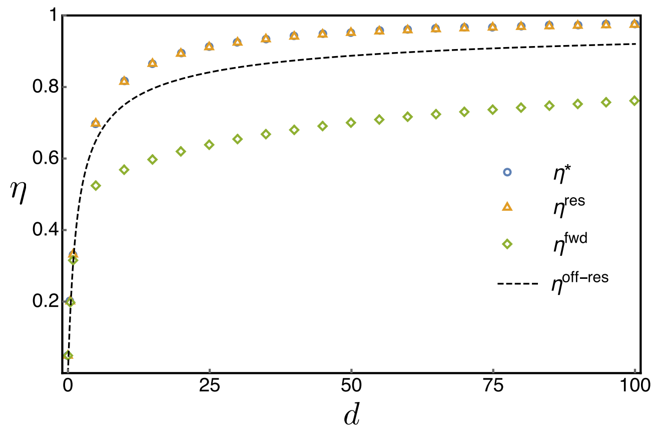

Figure 3. Retrieval efficiency as a function of the optical depth. Blue circles indicate the retrieval efficiency from the optimal spin wave. Yellow triangles (green diamonds) indicate the efficiency from backward (forward) retrieval for the proposed recipe that uses an exponentially rising write control field. The black dashed line indicates the retrieval efficiency using the standard approach with off-resonant write control fields.

$$\bar{P}(u,\, \tau_{\mathrm{R}} + \tau_{\mathrm{d}}) \approx \mathrm{i}\bar{S}(u,\, t = \tau_{\mathrm{d}}). \tag{13}$$

The emitted read photon field can then be obtained by solving the set of equations in (1) after the fast read control field has ended (see appendix A.2), giving

$$\mathcal{E}_{\mathrm{r}}(0,\, t) = \mathrm{i}\sqrt{\frac{\gamma_{eg}\, d}{cL}}\, \mathrm{e}^{-\gamma_{eg} t} \int_0^L J_0\left[2\sqrt{\frac{\gamma_{eg}\, d}{L} t (L - z_1'')}\right] P(L - z_1'',\, \tau_{\mathrm{R}} + \tau_{\mathrm{d}})\, \mathrm{d}z_1''. \tag{14}$$

Along with equation (13) and noting that $\int_0^\infty \mathrm{e}^{-\alpha x} J_\nu(2\beta\sqrt{x}) J_\nu(2\gamma\sqrt{x}) \mathrm{d}x = \frac{1}{\alpha} I_\nu\left(\frac{2\beta\gamma}{\alpha}\right) \exp\left(-\frac{\beta^2+\gamma^2}{\alpha}\right)$ [18], this emitted field then yields a retrieval efficiency given by equation (6).

3.3. Comparison

We have seen that the proposed retrieval protocol yields a dependence on the spin shape, as described by equation (6). Hence we now compare the retrieval efficiencies attainable with our protocol and compare them to the optimal ones.

We can estimate the achievable efficiency of our protocol by choosing a write pulse duration such that the heralded spin shape best fits the optimal spin shape. A good approximation to this write pulse duration is well described in [19], and given by

$$\tau_{\mathrm{W}}^{\mathrm{approx}} = \frac{1}{\gamma_{eg}} \frac{1}{1 + \frac{d}{2}}. \tag{15}$$

As we show below, this simple expression for the write pulse duration essentially produces the optimal efficiency available for a given optical depth. This is hence the write pulse duration we recommend.

We also compute the retrieval efficiencies η^{fwd} that would be obtained if the resonant write pulse of duration τ_W^{approx} were to be followed by a co-propagating retrieve pulse instead. This would result in a situation where the spin wave would be far from optimal with respect to the retrieval direction. In figure 3, we compare the optimal efficiency η^*, the efficiencies η^{res} and η^{fwd} obtained with our proposal (from a spin wave created from a resonant exponential pulse with duration τ_W^{approx}) together with the efficiency of the standard approach using far off-resonant write pulses, for which the efficiency is bounded by the complete retrieval efficiency from a flat spin wave [15]

$$\eta^{\text{off}-\text{res}} = 1 - \mathrm{e}^{-d}(I_0(d) + I_1(d)), \tag{16}$$

which we have verified numerically. This retrieval efficiency is valid for retrieval from both the forward and backward directions from a flat spin wave.

Our proposal approaches optimal efficiencies, performing within $\sim 10^{-3}$ of η^* and compares favourably with respect to the standard off-resonant case. The improvement in efficiency is dependent on the optical depth, and we present some values in table 1.

Table 1. Comparison of retrieval efficiency from different spin shapes.

d	$\eta^{\rm fwd}$	$\eta^{\rm off-res}$	$\eta^{\rm res}$	η^{*}
0.1	0.047 6	0.047 6	0.047 6	0.047 6
1	0.314 0	0.326 3	0.330 5	0.330 5
10	0.567 1	0.750 9	0.813 4	0.814 2
20	0.618 3	0.822 7	0.892 1	0.897 3
100	0.760 0	0.920 3	0.972 8	0.974 5

Finally, we have also investigated the on-resonance retrieval with an exponentially increasing write pulse using non optimised pulse widths ($\tau_W = \gamma_{eg}^{-1}$), but find a saturation of only ∼67 % of the retrieval efficiency for high optical depths.

3.4. Retrieval into a single mode

For a single photon source to be useful, one needs to not only efficiently emit a single photon, but also to ensure that the said photon is emitted in a single mode. In our model we have assumed that the write and read photons are each in a single mode.

For an actual implementation, one way to check that a single mode for the write and read photons are collected and detected, is to perform an autocorrelation measurement with two detectors after a 50-50 beamsplitter (see appendix D). Under the assumption that the write and read photons are correlated through vacuum squeezing processes, this measurement allows us to determine the number of emission modes K, as it gives $g^{(2)} \sim 1 + \frac{1}{K}$ [20] (valid in the absence of detector noise and for small emission probabilities).

4. Feasibility study of Rubidium-87

For a feasibility study we consider a Λ-system consisting of the following energy levels from the D-2 transition of Rubidium-87: $|g\rangle = |5^2{\rm S}_{1/2}, {\rm F}=2, m_{\rm F}=2\rangle$, $|s\rangle = |5^2{\rm S}_{1/2}, {\rm F}=1, m_{\rm F}=0\rangle$ and $|e\rangle = |5^2{\rm P}_{3/2}, {\rm F}=2, m_{\rm F}=1\rangle$. By taking into account the relevant branching ratios, we take $\gamma_{eg} = \frac{1}{12}(2\pi)6.067$ MHz and $\gamma_{es} = \frac{1}{8}(2\pi)6.067$ MHz.

We first consider an optical depth of $d = 20$ on the $|e\rangle - |g\rangle$ transition. From equation (15), we find that a suitable write control field duration is given by $\gamma_{eg}\tau_W^{\rm approx} = 0.09$. This implies a field duration of $\tau_{\rm W}^{\rm approx} \approx 29$ ns. Further considering an optical depth of $\bar{d} = 20$ on the $|e\rangle - |s\rangle$ transition and a weak write control field such that $\Omega_{\rm W}^{\rm max}\tau_{\rm W} = 0.01$, the number of write photons is $n_{\rm w} = 2 \times 10^{-4}$ within a short detection window of $\tau_{\rm d} \approx 100$ ns. We note that the ability to create pulses with a rising exponential shape with field amplitude duration as low as 20 ns has already been demonstrated in works such as [21, 22].

Subsequently, the retrieval pulse on the $|e\rangle - |g\rangle$ transition requires a Rabi frequency of $\Omega_{\rm R} \gg (2\pi)5.3$ MHz, with a predicted retrieval efficiency of 89%, essentially achieving η^* (see table 1). This compares favourably to the retrieval efficiency from a flat spin wave $\eta^{\rm off-res} = 82\%$.

5. Conclusion

In this work, we have discussed conditions for the optimal generation of single photons from spontaneous Raman processes in cold atoms.

We have first recognised that the ability to create favourable spin wave shapes can significantly improve the heralded retrieval efficiency. Since the reemission process is collective, the retrieval process benefits from all atoms participating favourably, in this case benefitting from a particular optimal spin shape. A resonant write pulse offers the option to create spatially varying waves due to its significant interaction through the sample. We have thus proposed a detailed recipe to create single photons with efficiencies that compare favourably to standard strategies utilising flat spin waves.

The recipe focuses on cases where the spin coherence time is longer than the optical coherence times and consists in first specifying the decay rates γ_{eg} and γ_{es} from the excited states and the optical depths d and $\bar{d}$ on the $|e\rangle - |g\rangle$ and $|e\rangle - |s\rangle$ transitions. Then the recipe fixes the duration of the detection window to be smaller than the shortest decoherence times, that is, the minimum of $1/\gamma_{eg}$ and $1/\gamma_{eg}$ while maintaining that the number of write photons is sufficiently low. Finally, the recipe proposes to take for the write pulse an exponential rising function in time, whose duration is given by $\tau_W^{\rm approx} = \gamma_{eg}^{-1}\left(1 + \frac{d}{2}\right)^{-1}$. To estimate the heralding rates, one can next compute the write photon number with the formula in equation (8) given the Rabi frequency of the write pulse. The readout efficiency obtained with a fast readout pulse, that is a readout pulse with a duration much smaller than the atomic coherence times, reaches the values given in figure 3 (yellow triangles) as soon as the corresponding Rabi frequency defines essentially a π-pulse. This recipe describes a convenient way to come close to the optimal efficiency of single photon sources with given optical depths based on spontaneous Raman processes. This work could help in the implementation of the first quantum repeater protocol successfully outperforming the direct transmission of photons [4].

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Appendix A. Retrieval process

A.1. Retrieval emission dynamics

We begin from the Hamiltonian $H = H_0 + V$ (see [15]), where we consider an atomic sample of length L, and a classical field sent from the $z = L$ side of the sample. Choosing $|g\rangle$ to be the energy level reference for the atomic states, we have

$$H_0 = \int \mathrm{d}\omega \hbar\omega \hat{a}_\omega^\dagger \hat{a}_\omega + \sum_{i=1}^{N} (\hbar\omega_s \sigma_{ss}^j + \hbar\omega_e \sigma_{ee}^j) \tag{17}$$

$$V = -\hbar \sum_{i=1}^{N} \Bigg(\Omega_{\mathrm{R}}\left(t - \frac{L - z_i}{c}\right) \sigma_{es}^i \mathrm{e}^{-\mathrm{i}\omega_2 t} \mathrm{e}^{+\mathrm{i}\omega_2\left(\frac{L-z_i}{c}\right)} + g\sqrt{\frac{L}{2\pi c}} \int \mathrm{d}\omega\ a_\omega \mathrm{e}^{\mathrm{i}\omega\frac{L-z_i}{c}} \sigma_{eg}^i + \mathrm{h.c.} \Bigg), \tag{18}$$

where $\sigma_{\mu\nu}^i = |\mu\rangle_i \langle\nu|$ indicates atomic level operators for the ith atom, and a_w indicates the annihilation operator for the photonic mode at frequency ω. ω_2 (ω_1) indicates the frequency of the read control (photon) field, respectively. Note that we are considering resonant pulses, so we have ω_1 (ω_2) $= \omega_e$ (ω_s). Using

$$A = \sum_{i=1}^{N} [\hbar(\omega_1 - \omega_2)\sigma_{ss}^i + \hbar\omega_1 \sigma_{ee}^i] + \hbar\omega_1 \int \mathrm{d}\omega\ \mathcal{E}_r^\dagger(z, t)\mathcal{E}_r(z, t),$$
$$U = \mathrm{e}^{-\mathrm{i}At/\hbar},$$

for the change of frame, then in the continuum limit, we obtain

$$\begin{aligned} H_{\mathrm{new}} &= U^\dagger H U - A \\ &= \int \mathrm{d}\omega\ \hbar\omega a_\omega^\dagger a_\omega - \hbar\omega_1 \int \mathrm{d}z\ \mathcal{E}_r^\dagger(z, t)\mathcal{E}_r(z, t) \\ &\quad + \frac{N}{L} \int \mathrm{d}z \{ -\hbar\Omega_{\mathrm{R}}(z, t)\sigma_{es}(z, t)\mathrm{e}^{+\mathrm{i}\omega_2\frac{L-z}{c}} + \mathrm{H.c.} \\ &\quad - g\mathcal{E}_r(z, t)\sigma_{eg}(z, t)\mathrm{e}^{+\mathrm{i}\omega_1\frac{L-z}{c}} + \mathrm{H.c.} \}, \end{aligned}$$

where we have defined a real Rabi frequency $\Omega_{\mathrm{R}}(z, t) = \Omega_{\mathrm{R}}(t - \frac{L-z}{c})$ and $\mathcal{E}_{\mathrm{r}}(z, t) = \sqrt{\frac{L}{2\pi c}} \int \mathrm{d}\omega\ \mathrm{e}^{\mathrm{i}\omega_1 t} a_\omega \mathrm{e}^{\mathrm{i}(\omega-\omega_1)\frac{L-z}{c}}$. Using the field propagation equation along with the Heisenberg–Langevin equations of motion, we have in a moving coordinate frame, ignoring spinwave decoherence and the noise terms, and also considering that $\sigma_{gg} \sim 1$,

$$\begin{aligned} \partial_z \mathcal{E}_{\mathrm{r}}(z, t) &= -\frac{\mathrm{i}g\sqrt{N}}{c} P(z, t) \\ \partial_t P(z, t) &= -\gamma_{eg} P(z, t) + \mathrm{i}g\sqrt{N}\mathcal{E}_{\mathrm{r}}(z, t) + \mathrm{i}\Omega_{\mathrm{R}}(t) S(z, t) \\ \partial_t S(z, t) &= \mathrm{i}\Omega_{\mathrm{R}}(t) P(z, t), \end{aligned}$$

where $g^2N = \frac{d\gamma_{eg}c}{L}$, $P(z, t) = \sqrt{N}\sigma_{ge}(z, t)\mathrm{e}^{-\mathrm{i}\omega_1\frac{L-z}{c}}$ and $S(z, t) = \sqrt{N}\sigma_{gs}(z, t)\mathrm{e}^{-\mathrm{i}(\omega_1-\omega_2)\frac{L-z}{c}}$. In the continuum limit, the spin and field operators obey the following commutation relations

$$[\sigma_{\alpha\beta}(z, t), \sigma_{\mu\nu}(z', t)] = \frac{L}{N}\delta(z - z')(\delta_{\beta\mu}\sigma_{\alpha\nu}(z, t) - \delta_{\nu\alpha} \tag{19}$$

$$[\mathcal{E}_\mathrm{r}(z, t), \mathcal{E}_\mathrm{r}^\dagger(z, t')] = \frac{L}{c}\delta(t - t'). \tag{20}$$

Rewriting equations (A.1) in the reverse direction e.g. $\bar{\mathcal{E}}_\mathrm{r}(z', t) = \bar{\mathcal{E}}_\mathrm{r}(L - z, t) = \mathcal{E}_\mathrm{r}(z, t)$, and taking the Laplace transform from $z' \to u$ we obtain

$$\begin{aligned}
\bar{\mathcal{E}}_\mathrm{r}(u, t) &= \frac{\mathrm{i}g\sqrt{N}}{cu}\bar{P}(u, t) + \frac{1}{u}\bar{\mathcal{E}}_\mathrm{r}(z' = 0, t),\\
\partial_t\bar{P}(u, t) &= -\gamma_{eg}\bar{P}(u, t) + \mathrm{i}g\sqrt{N}\bar{\mathcal{E}}_\mathrm{r}(u, \mathrm{t}) + \mathrm{i}\Omega_\mathrm{R}(t)\bar{S}(u, t),\\
\partial_t\bar{S}(u, t) &= \mathrm{i}\Omega_\mathrm{R}(\mathrm{t})\bar{P}(u, t).
\end{aligned} \tag{21}$$

We can combine these three equations into a single differential equation, where we have ignored the boundary term $\bar{\mathcal{E}}_\mathrm{r}(z' = 0, t)$ since we send the read control field into the $z = L$ side of the atoms. On resonance ($\Delta = 0$), let $A = \gamma_{eg} + \frac{g^2N}{cu}$ and $B = \Omega_\mathrm{R}^2$ to see

$$\ddot{\bar{S}}(u, t) + A\dot{\bar{S}}(u, t) + B\bar{S}(u, t) = 0. \tag{22}$$

A.2. Fast retrieval

In the strong regime for the read control field, one requires $2|\Omega_\mathrm{R}| \gg \gamma_{eg}(1 + d)$, which implies

$$\begin{aligned}
2\Omega_\mathrm{R} &\gg \gamma_{eg}\left(1 + \frac{dz'}{L}\right)\\
\Rightarrow 2\Omega_\mathrm{R} &\gg \gamma_{eg}\left(1 + \frac{d}{Lu}\right),
\end{aligned}$$

which then yields the regime $4B \gg A^2$.

The solution for equation (22) in this regime is

$$\bar{S}(u, t) = \mathrm{e}^{-At/2}\cos(\Omega_\mathrm{R}t)C_1(u) + \mathrm{e}^{-At/2}\sin(\Omega_\mathrm{R}t)C_2(u),$$

where the initial condition implies

$$\bar{S}(u, t) = \mathrm{e}^{-At/2}\cos(\Omega_\mathrm{R}t)\bar{S}(u, t = 0).$$

One can then find the prepared polarisation in terms of the intial spin condition,

$$\begin{aligned}
\bar{P}(u, t) &= \frac{1}{\mathrm{i}\Omega_R}\partial_t\bar{S}(u, t)\\
&= \frac{\mathrm{i}}{\Omega_R}\mathrm{e}^{-\frac{A}{2}t}\left[\frac{A}{2}\cos(\Omega_R t) + \Omega_R\sin(\Omega_R t)\right]\bar{S}(u, t = 0).
\end{aligned}$$

In the limit where we have a sufficiently strong read control field ($2\Omega_\mathrm{R} \gg \frac{\pi}{2}\gamma_{eg}(1 + d)$), the π-pulse is completed quickly and we obtain a lossless preparation of $\bar{P}(u, t)$ from $\bar{S}(u, t = 0)$ in the form

$$\bar{P}(u, \tau_\mathrm{R}) = \mathrm{i}\bar{S}(u, t = 0). \tag{23}$$

Once the polarisation is prepared, we find the emission by solving for the dynamics in the absence of the laser,

$$\begin{aligned}
\partial_z\bar{\mathcal{E}}_\mathrm{r}(z, t) &= -\mathrm{i}\frac{g\sqrt{N}}{c}\bar{P}(z, t),\\
(\partial_t + \gamma_{eg})\bar{P}(z, t) &= \mathrm{i}g\sqrt{N}\bar{\mathcal{E}}_\mathrm{r}(z, t).
\end{aligned}$$

Taking the Laplace transform from $L - z = z' \to u$ and neglecting the boundary term since it does not contribute to the photon number, we have

$$\begin{aligned}
\bar{\mathcal{E}}_\mathrm{r}(u, t) &= \mathrm{i}\frac{g\sqrt{N}}{cu}\bar{P}(u, t),\\
(\partial_t + \gamma_{eg})\bar{P}(u, t) &= \mathrm{i}g\sqrt{N}\bar{\mathcal{E}}_\mathrm{r}(u, t) = -\frac{g^2N}{cu}\bar{P}(u, t).
\end{aligned}$$

This yields the evolution of $P(u, t)$ after its preparation from $S(u, t)$,

$$\bar{P}(u,\, t) = \mathrm{e}^{-\left(\gamma_{eg} + \frac{g^2 N}{cu}\right)(t-\tau_{\mathrm{R}})} \bar{P}(u,\, \tau_{\mathrm{R}}), \tag{24}$$

and gives an emitted field of

$$\begin{aligned}\bar{\mathcal{E}}_{\mathrm{r}}(z',\, t) = {} & \mathrm{i}\frac{g\sqrt{N}}{c}\mathrm{e}^{-\gamma_{eg}(t-\tau_{\mathrm{R}})} \\ & \times \int_0^{z'} \mathrm{d}z'' J_0\left[2\sqrt{\frac{g^2 N}{c}(t - \tau_{\mathrm{R}})(z' - z'')}\right]\bar{P}(z',\, \tau_{\mathrm{R}}),\end{aligned}$$

where $J_n[x]$ refers the nth Bessel function of the first kind. Now with $z' = L - z$, we require the field at $z = 0$ for backward retrieval, and we finally obtain

$$\begin{aligned}\mathcal{E}_r(0,\, t) = {} & -\frac{g\sqrt{N}}{c}\mathrm{e}^{-\gamma_{eg} t} \\ & \times \int_0^{L} \mathrm{d}z'' J_0\left[2\sqrt{\frac{g^2 N}{c}t(L - z'')}\right]S(L - z'',\, 0),\end{aligned} \tag{25}$$

where we have used equation (23) for a lossless preparation.

A.3. Slow retrieval

In the weak regime for the read control field, one requires $2|\Omega_{\mathrm{R}}| \ll \gamma_{eg}$, which implies

$$\begin{aligned} 2\Omega_R &\ll \gamma_{eg}\left(1 + \frac{dz}{L}\right) \\ \Rightarrow 2\Omega_R &\ll \gamma_{eg}\left(1 + \frac{d}{Lu}\right),\end{aligned}$$

which then yields the regime $4B \ll A^2$.

The solution for equation (22) in this regime is

$$\bar{S}(u,\, t) = \mathrm{e}^{-\frac{1}{2}(A+\sqrt{A^2-4B})t}C_u(1) + \mathrm{e}^{-\frac{1}{2}(A-\sqrt{A^2-4B})t}C_u(2).$$

When there is no laser ($B = 0$), there should be no spinwave decay since we have considered zero spin wave decoherence, so we set $C_u(1) = 0$ and obtain

$$\bar{S}(u,\, t) = \mathrm{e}^{-\frac{1}{2}(A-\sqrt{A^2-4B})t}\bar{S}(u,\, t = 0).$$

Now, in this regime when the Rabi frequency is small, we have

$$\begin{aligned}\mathrm{e}^{-\frac{1}{2}(A-\sqrt{A^2-4B})t} &= \mathrm{e}^{-\frac{1}{2}\left(A - A\sqrt{1-\frac{4B}{A^2}}\right)t} \\ &\approx \mathrm{e}^{-\frac{B}{A}t} \\ &= \mathrm{e}^{-\frac{\Omega^2}{\gamma_{eg}\left(1+\frac{d}{Lu}\right)}t}\end{aligned}$$

This gives

$$\bar{S}(u,\, t) = \mathrm{e}^{-Kt\frac{1}{1+s/u}}\bar{S}(u,\, t = 0),$$

where $K = \frac{\Omega_{\mathrm{R}}^2}{\gamma_{eg}}$ and $s = \frac{d}{L}$. One can proceed to find $\bar{P}(u,\, \mathrm{t}) = \frac{1}{\mathrm{i}\Omega_{\mathrm{R}}}\partial_t\bar{S}(u,\, \mathrm{t})$ and $\bar{\mathcal{E}}(u,\, t) = \mathrm{i}\frac{g\sqrt{N}}{cu}\bar{P}(u,\, t)$, giving

$$\bar{\mathcal{E}}(u,\, t) = -\frac{g\sqrt{N}}{c}\frac{K}{\Omega_{\mathrm{R}}}\left[\frac{1}{u + s}\mathrm{e}^{-Kt+Kt\left(\frac{s}{s+u}\right)}\right]\bar{S}(u,\, t = 0).$$

This yields

$$\begin{aligned}\bar{\mathcal{E}}(z',\, t) = {} & -\frac{g\sqrt{N}}{c}\frac{K}{\Omega}\mathrm{e}^{-Kt} \\ & \times \int_0^{z'} \mathrm{e}^{-s(z'-z'')} I_0(2\sqrt{Kts(z' - z'')})\bar{S}(z'',\, t = 0)\mathrm{d}z''.\end{aligned} \tag{26}$$

One can then compute the retrieval efficiency from a single spin wave, and this is found to yield the optimal retrieval efficiency.

$$
\begin{aligned}
&\int_0^\infty \mathrm{d}t\ \frac{c}{L}\langle \mathcal{E}^\dagger(0,t)\mathcal{E}(0,t)\rangle \\
&= \int_0^\infty \mathrm{d}t\ \frac{d}{L^2}\frac{\Omega_R^2}{\gamma_{eg}}\mathrm{e}^{-2Kt}\int_0^L \mathrm{d}z_1''\int_0^L \mathrm{d}z_2''\ \mathrm{e}^{-\frac{d}{L}(2L-z_1''-z_2'')} \\
&\times I_0\left[2\sqrt{Kt\frac{d}{L}(L-z_1'')}\right]I_0\left[2\sqrt{Kt\frac{d}{L}(L-z_2'')}\right] \\
&\times \langle S^\dagger(L-z_1'',t=0)S(L-z_2'',t=0)\rangle \\
&= \frac{1}{L}\int_0^L \mathrm{d}z_1''\ \frac{1}{L}\int_0^L \mathrm{d}z_2''\ \frac{d}{2}\mathrm{e}^{-\frac{d}{2}\frac{(L-z_1'')+(L-z_2'')}{L}} \\
&\times I_0\left[\mathrm{d}\sqrt{\frac{L-z_1''}{L}}\sqrt{\frac{L-z_2''}{L}}\right]\langle S^\dagger(L-z_1'',t=0)S(L-z_2'',t=0)\rangle
\end{aligned}
$$

where $I_n[x]$ denotes the nth modified Bessel function of the first kind. We have made use of the fact that $I_n(z)=\mathrm{i}^{-n}J_n(\mathrm{i}z)$ and also $\int_0^\infty \mathrm{e}^{-\alpha x}J_\nu(2\beta\sqrt{x})J_\nu(2\gamma\sqrt{x})\mathrm{d}x=\frac{1}{\alpha}I_\nu\left(\frac{2\beta\gamma}{\alpha}\right)\exp\left(-\frac{\beta^2+\gamma^2}{\alpha}\right)$.

Appendix B. Write process

B.1. Heisenberg–Langevin equations for the atomic coherences

The goal here is to first derive the expressions for the evolution of the atomic coherences in the write process. We begin from the Hamiltonian $\bar{H}=\bar{H}_0+\bar{V}$

$$
\bar{H}_0=\int \mathrm{d}\omega\hbar\omega a_\omega^\dagger a_\omega+\sum_{i=1}^{N}(\hbar\omega_s\sigma_{ss}^j+\hbar\omega_e\sigma_{ee}^j) \tag{27}
$$

$$
\begin{aligned}
\bar{V}=-\hbar\sum_{i=1}^{N}\Big(&\Omega_{\mathrm{W}}(t-z_i/c)\sigma_{eg}^i\mathrm{e}^{-\mathrm{i}\omega_1(t-z_i/c)} \\
&+\bar{g}\sqrt{\frac{L}{2\pi c}}\int \mathrm{d}\omega\ \hat{a}_\omega \mathrm{e}^{\mathrm{i}\omega z_i/c}\sigma_{es}^i+\mathrm{H.c.}\Big).
\end{aligned} \tag{28}
$$

Using

$$
\begin{aligned}
\bar{A}&=\sum_{i=1}^{N}(\hbar\omega_s\sigma_{ss}^i+\hbar\omega_e\sigma_{ee}^i)+\hbar\omega_2\int \mathrm{d}z\ \mathcal{E}_w^\dagger(z,t)\mathcal{E}_w(z,t), \\
\bar{U}&=\mathrm{e}^{-\mathrm{i}\bar{A}t/\hbar}
\end{aligned}
$$

for the change of frame, then in the continuum limit, we obtain

$$
\begin{aligned}
\bar{H}_{\mathrm{new}}&=\bar{U}^\dagger\bar{H}\bar{U}-\bar{A} \\
&=\int \mathrm{d}\omega\ \hbar a_\omega^\dagger a_\omega-\hbar\omega_2\int \mathrm{d}z\ \mathcal{E}_w^\dagger(z,t)\mathcal{E}_w(z,t) \\
&+\frac{N}{L}\int \mathrm{d}z\{-\hbar\Omega_{\mathrm{W}}(t-z/c)\sigma_{eg}(z,t)\mathrm{e}^{\mathrm{i}\omega_1 z/c}+\mathrm{H.c.} \\
&-g\mathcal{E}_w(z,t)\sigma_{es}(z,t)\mathrm{e}^{\mathrm{i}\omega_2 z/c}+\mathrm{H.c.}\},
\end{aligned} \tag{29}
$$

where we have defined $\mathcal{E}_{\mathrm{w}}(z,t)=\sqrt{\frac{L}{2\pi c}}\mathrm{e}^{\mathrm{i}\omega_2 t}\int d\omega\ a_\omega \mathrm{e}^{\mathrm{i}(\omega-\omega_2)z/c}$.

Assuming a real Rabi frequency Ω_{W}, this yields the Heisenberg–Langevin equations as follows:

$$
\begin{aligned}
\partial_t\sigma_{se}&=-\gamma_{es}\sigma_{se}+\mathrm{i}\Omega_W\mathrm{e}^{\mathrm{i}\omega_1 z/c}\sigma_{sg} \\
&-\mathrm{i}\bar{g}\mathcal{E}_w\mathrm{e}^{\mathrm{i}\omega_2 z/c}(\sigma_{ee}-\sigma_{ss})+F_{se} \\
\partial_t\sigma_{sg}&=-\gamma_0\sigma_{sg}+\mathrm{i}\Omega_W\mathrm{e}^{-\mathrm{i}\omega_1 z/c}\sigma_{se} \\
&-\mathrm{i}\bar{g}\mathcal{E}_w\mathrm{e}^{\mathrm{i}\omega_2 z/c}\sigma_{eg}+F_{sg} \\
\partial_t\sigma_{eg}&=-\gamma_{eg}\sigma_{eg}-\mathrm{i}\Omega_W\mathrm{e}^{-\mathrm{i}\omega_1 z/c}\sigma_{gg}+F_{eg},
\end{aligned} \tag{30}
$$

where ω_1 (ω_2) indicates the frequency of the write control field (write photon field), respectively, and $\bar{g}^2N=\frac{d\gamma_{es}c}{L}$.

B.2. Creating atomic coherences

During the write process we account for possible depletion of the write laser intensity, and hence do not assume $\Omega_{\mathrm{W}}(r,t)$ to be constant throughout the sample. As a result of the laser we create coherences between the $|g\rangle-|e\rangle$

transition, which forms the initial state for the write photon field. Here we proceed to find the atomic coherences prepared as a result of our exponential shaped resonant write control field.

For a sufficiently short write control field, the dynamics of the field and the atoms can be described with the dynamics along the $|g\rangle - |e\rangle$ transition. Ignoring the noise terms on σ_{ge} and making the analogy between the classical and quantum fields on the $|g\rangle - |e\rangle$ transition,

$$\begin{aligned} c\partial_z\Omega_{\mathrm{W}}(z,\, t) &= \mathrm{i}g^2N\sigma_{ge}(z,\, t)\mathrm{e}^{-\mathrm{i}\omega_1 z/c}, \\ \partial_t\sigma_{ge} &= -\gamma_{eg}\sigma_{ge} + \mathrm{i}\Omega_{\mathrm{W}}(z,\, t)\mathrm{e}^{+\mathrm{i}\omega_1 z/c}\sigma_{gg} \\ &\approx -\gamma_{eg}\sigma_{ge} + \mathrm{i}\Omega_{\mathrm{W}}(z,\, t)\mathrm{e}^{+\mathrm{i}\omega_1 z/c}, \end{aligned} \tag{31}$$

where we have assumed that almost all atoms remain in the $|g\rangle$ level.

Let us first assume a write control field with Rabi frequency Ω_{W} that begins at $t = 0$. Taking the Laplace transforms from $t \to w$, we find

$$\partial_z\Omega_{\mathrm{W}}(z,\, w) = \mathrm{i}\frac{g^2N}{c}\sigma_{ge}(z,\, w)\mathrm{e}^{-\mathrm{i}\omega_1 z/c}, \tag{32}$$

$$\sigma_{ge}(z,\, w) = \frac{1}{w + \gamma_{eg}}[\mathrm{i}\Omega_{\mathrm{W}}(z,\, w)\mathrm{e}^{\mathrm{i}\omega_1 z/c} + \sigma_{ge}(z,\, t=0)]. \tag{33}$$

Insert equation (33) into equation (32), and use the initial condition $\sigma_{ge}(z, t = 0) = 0$ to obtain

$$\partial_z\Omega_W(z,\, w) = -\frac{g^2N}{c}\left(\frac{1}{w + \gamma_{eg}}\right)\Omega_W(z,\, w),$$

yielding

$$\Omega_{\mathrm{W}}(z,\, w) = \mathrm{e}^{-\frac{g^2N}{c}\left(\frac{1}{w+\gamma_{eg}}\right)z}\Omega_{\mathrm{W}}(z=0,\, w).$$

Insert this into equation (33) to obtain

$$\sigma_{ge}(z,\, w) = (\mathrm{i}\mathrm{e}^{\mathrm{i}\omega_1 z/c})\left[\frac{1}{w + \gamma_{eg}}\mathrm{e}^{-\frac{g^2N}{c}\frac{1}{w+\gamma_{eg}}z}\Omega_{\mathrm{W}}(z=0,\, w)\right].$$

After inverting the Laplace transform, we now shift the limits to consider a write control field with support on negative times, giving

$$\begin{aligned} \sigma_{ge}(z,\, t) = (\mathrm{i}\mathrm{e}^{\mathrm{i}\omega_1 z/c})\int_{-\infty}^{t} &\mathrm{e}^{-\gamma_{eg}(t-t_1'')} \\ &\times J_0\left[2\sqrt{\frac{\gamma_{eg}d}{L}(t - t_1'')z}\right]\Omega_{\mathrm{W}}(z=0,\, t_1'')\mathrm{d}t_1'', \end{aligned} \tag{34}$$

where $J_n(x)$ indicates the nth Bessel function of the first kind.

Thus, with an exponential write control field $\Omega_{\mathrm{W}}(0,\, t) = \Omega_{\mathrm{W}}^{\max}\mathrm{e}^{t/\tau_{\mathrm{W}}}$ sent up to $t = 0$, we evaluate the atomic coherence at $t = 0$ with the help of $\int_0^\infty \mathrm{e}^{-At}J_0[2\sqrt{Bt}]\mathrm{d}t = \frac{1}{A}\mathrm{e}^{-B/A}$ and finally obtain

$$\sigma_{ge}(z,\, 0) = \mathrm{e}^{\mathrm{i}\omega_1 z/c}\theta_0\mathrm{e}^{-\frac{\alpha z}{2}}, \tag{35}$$

where $\theta_0 = \mathrm{i}\frac{\Omega_{\max}\tau_{\mathrm{W}}}{1+\gamma_{eg}\tau_{\mathrm{W}}}$ and $\alpha/2 = d\frac{\gamma_{eg}\tau_{\mathrm{W}}}{1+\gamma_{eg}\tau_{\mathrm{W}}}\frac{1}{L}$.

B.3. Write photon emission

After the preparation of atomic coherences, we begin to see spontaneous emission from the $|e\rangle$ level. Along with the field propagation equation, the relevant Heisenberg–Langevin equations are

$$\begin{aligned} c\partial_z\mathcal{E}_{\mathrm{w}} &= \mathrm{i}\bar{g}N\mathrm{e}^{-\mathrm{i}\omega_2 z/c}\sigma_{se}(z,\, t), \\ \partial_t\hat{\sigma}_{es} &= -\gamma_{es}\sigma_{se} - \mathrm{i}\bar{g}\mathcal{E}_w\mathrm{e}^{\mathrm{i}\omega_2 z/c}(\sigma_{ee} - \sigma_{ss}) + F_{se}. \end{aligned}$$

Defining $Q^\dagger = \sqrt{N}\mathrm{e}^{-\mathrm{i}\omega_2 z/c}\sigma_{se}$, we will consider the write emission for short detection times. Using (35) we thus replace $\sigma_{ee} - \sigma_{ss}$ with its mean value at position z and $t = 0$ to obtain

$$\begin{aligned} c\partial_z\mathcal{E}_{\mathrm{w}}(z,\, t) &= \mathrm{i}\bar{g}\sqrt{N}Q^\dagger(z,\, t), \\ \partial_t Q^\dagger(z,\, t) &= -\gamma_{es}Q^\dagger(z,\, t) - \mathrm{i}\bar{g}\sqrt{N}|\theta_0|^2\mathrm{e}^{-\alpha z}\mathcal{E}_{\mathrm{w}}(z,\, t) \\ &\quad + F_Q^\dagger(z,\, t). \end{aligned} \tag{36}$$

Performing first the Laplace transform in space ($z \longrightarrow s$)

$$
\begin{aligned}
s\mathcal{E}_{\mathrm{w}}(s,t) - \mathcal{E}_{\mathrm{w}}(z=0,t) &= A\ Q^{\dagger}(s,t),\\
\partial_t Q^{\dagger}(s,t) &= -\gamma_{es} Q^{\dagger}(s,t) + B\mathcal{E}_{\mathrm{w}}(s+\alpha,t)\\
&\quad + F_Q^{\dagger}(s,t),
\end{aligned}
$$

and then in time ($t \longrightarrow \omega$), we get

$$
\begin{aligned}
s\mathcal{E}_{\mathrm{w}}(s,\omega) - \mathcal{E}_{\mathrm{w}}(z=0,\omega) &= A\ Q^{\dagger}(s,\omega),\\
Q^{\dagger}(s,\omega) &= \frac{1}{\gamma_{es}+\omega} B\mathcal{E}_{\mathrm{w}}(s+\alpha,\omega) + F_Q^{\dagger}(s,\omega) + Q^{\dagger}(s,t=0),
\end{aligned}
$$

where $A = \mathrm{i}\frac{\bar{g}\sqrt{N}}{c}$ and $B = -\mathrm{i}\bar{g}\sqrt{N}\theta_0^2$.

Substituting the second line into the first, we eliminate $Q(s,\omega)$ and are left with a boundary term in Q:

$$
\begin{aligned}
s\mathcal{E}(s,\omega) - \mathcal{E}(z=0,\omega) = \left(\frac{A}{\gamma_{es}+\omega}\right)&[B\mathcal{E}(s+\alpha,\omega)\\
&+ F_Q^{\dagger}(s,\omega) + Q^{\dagger}(s,t=0)].
\end{aligned}
$$

The following formula also holds with a shift from s to $s+\alpha$:

$$
\begin{aligned}
&(s+\alpha)\mathcal{E}_{\mathrm{w}}(s+\alpha,\omega) - \mathcal{E}_{\mathrm{w}}(z=0,\omega)\\
&= \left(\frac{A}{\gamma_{es}+\omega}\right)[B\mathcal{E}_{\mathrm{w}}(s+2\alpha,\omega) + F_Q^{\dagger}(s+\alpha,\omega) + Q^{\dagger}(s+\alpha,t=0)].
\end{aligned}
$$

By substituting $\mathcal{E}_{\mathrm{w}}(s+\alpha,\omega)$ into the previous equation we can find $\mathcal{E}(s,\omega)$ in terms of $\mathcal{E}_{\mathrm{w}}(s+2\alpha,\omega)$, and by taking the substitution into the nth step we have

$$
\begin{aligned}
\mathcal{E}_{\mathrm{w}}(s,\omega) = K(\omega)^n D(n)\mathcal{E}_{\mathrm{w}}(s+n\alpha,\omega)&\\
+ \frac{1}{B}\sum_{j=1}^{n} K(\omega)^j D(j) F_Q^{\dagger}(s+(j-1)\alpha,\omega)&\\
+ \frac{1}{B}\sum_{j=1}^{n} K(\omega)^j D(j) Q^{\dagger}(s+(j-1)\alpha,t'=0)&\\
+ [K(\omega)]^{-1}\sum_{j=1}^{n}[K(\omega)^j D(j)]\mathcal{E}_{\mathrm{w}}(z'=0,\omega),&
\end{aligned}
$$

where $K(\omega) = \frac{AB}{\gamma_{es}+\omega}$ and $D(n) = \prod_{k=0}^{n-1}\frac{1}{s+k\alpha}$.

Taking the limit of $n \longrightarrow \infty$, the first term disappears, and we proceed to perform the inverse transform $s \longrightarrow z$. With a shift in the index j, $\mathcal{L}^{-1}[D(j+1)] = \frac{1}{j!}\left(\frac{1-\mathrm{e}^{-\alpha z}}{\alpha}\right)^j$ and the shifting property of the Laplace Transform,

$$
\begin{aligned}
&\mathcal{E}_w(z,\omega)\\
&= \frac{1}{B}\sum_{j=0}^{\infty} K(\omega)^{j+1}\int_0^z \frac{1}{j!}\left(\frac{1-\mathrm{e}^{-\alpha(z-z'')}}{\alpha}\right)^j \mathrm{e}^{-j\alpha z''} F_Q^{\dagger}(z'',\omega)\,\mathrm{d}z''\\
&\quad + \frac{1}{B}\sum_{j=0}^{\infty} K(\omega)^{j+1}\int_0^z \frac{1}{j!}\left(\frac{1-\mathrm{e}^{-\alpha(z-z'')}}{\alpha}\right)^j \mathrm{e}^{-j\alpha z''} Q^{\dagger}(z'',t=0)\,\mathrm{d}z''\\
&\quad + \frac{1}{K(\omega)}\sum_{j=0}^{\infty} K(\omega)^{j+1}\frac{1}{j!}\left(\frac{1-\mathrm{e}^{-\alpha(z)}}{\alpha}\right)^j \mathcal{E}_{\mathrm{w}}(z'=0,\omega)\\
&= \frac{A}{\gamma_{es}+\omega}\int_0^z \mathrm{e}^{\left[\frac{1}{\gamma_{es}+\omega}M(z,z'')\mathrm{e}^{-\alpha z''}\right]} F_Q^{\dagger}(z'',\omega)\mathrm{d}z''\\
&\quad + \frac{A}{\gamma_{es}+\omega}\int_0^z \mathrm{e}^{\left[\frac{1}{\gamma_{es}+\omega}M(z,z'')\mathrm{e}^{-\alpha z''}\right]} Q^{\dagger}(z'',t'=0)\mathrm{d}z''\\
&\quad + \mathrm{e}^{\frac{1}{\gamma_{es}+\omega}M(z,0)}\mathcal{E}_w(z=0,\omega),
\end{aligned}
\tag{37}
$$

where $M(z',z'') = \frac{AB}{\alpha}[1-\mathrm{e}^{-\alpha(z'-z'')}]$.

Finally, noting that

$$\mathcal{L}^{-1}\left[\frac{1}{\gamma_{es}+\omega}e^{\frac{A}{\gamma_{es}+\omega}}\right] = e^{-\gamma_{es}t}[\sqrt{At}^{(-1)}I_1(2\sqrt{At}) + I_2(2\sqrt{At})],$$
$$\mathcal{L}^{-1}[e^{\frac{A}{\gamma_{es}+\omega}}] = e^{-\gamma_{es}t}\left[\sqrt{\frac{A}{t}}I_1(2\sqrt{At}) + \delta(t)\right],$$

we get

$$\begin{aligned}\mathcal{E}_w(z, t) = A\int_0^z\int_0^t e^{-\gamma_{es}(t-t_1'')}H_1[\alpha, z, z_1'', t, t_1'']F_Q^\dagger(z_1'', t_1'')dt_1''dz_1'' \\ + A\int_0^z e^{-\gamma_{es}(t)}H_1[\alpha, z, z_1'', t, 0]Q^\dagger(z_1'', 0)dz_1'' \\ + \int_0^t e^{-\gamma_{es}(t-t'')}H_2(\alpha, z, 0, t, t'')\mathcal{E}_w(0, t'')dt'' \\ + \mathcal{E}_w(0, t),\end{aligned} \tag{38}$$

where

$$H_1(\alpha, z_1, z_2, t_1, t_2) = I_0[2\sqrt{M(z_1, z_2)e^{-\alpha z_2}(t_1 - t_2)}],$$
$$H_2(\alpha, z_1, z_2, t_1, t_2) = \sqrt{\frac{M(z_1, z_2)}{t_1 - t_2}}I_1[2\sqrt{M(z_1, z_2)e^{-\alpha z_2}(t_1 - t_2)}].$$

B.4. Number of write photons

Computing the photon flux requires the commutation relations for Q and a 2-point noise correlation function involving F_Q. In a short time window τ_d where $\sigma_{ee} - \sigma_{ss}$ is not changing, and with the Einstein relations (see Ch 15.5 of [23]), the Langevin equations for system operators can be written

$$\dot{A}_\mu = D_\mu(t) + F_\mu(t). \tag{39}$$

The corresponding memoryless noise correlations for operators μ and ν are such that

$$\langle F_\mu(t')F_\nu(t'')\rangle = 2\langle D_{\mu\nu}\rangle\delta(t' - t''), \tag{40}$$

where

$$2\langle D_{\mu\nu}\rangle = -\langle A_\mu D_\nu\rangle - \langle D_\mu A_\nu\rangle + \frac{d}{dt}\langle A_\mu A_\nu\rangle. \tag{41}$$

Thus, identifying terms in equation (36) with terms in equation (39), we make use of

$$\begin{aligned}[Q(z, t), Q^\dagger(z', t)] &= N[\sigma_{es}(z, t), \sigma_{se}(z', t)] \\ &= L\delta(z - z')|\theta_0|^2e^{-\alpha z'},\end{aligned} \tag{42}$$

then we make use of the fact that $\langle Q^\dagger(z, t)Q(z', t)\rangle$ right after our preparation of atomic coherences is zero, giving $\langle Q(z, t)Q^\dagger(z', t)\rangle = L\delta(z - z')|\theta_0|^2e^{-\alpha z'}$.

Then one obtains

$$2\langle D_{Q,Q^\dagger}\rangle = 2\gamma_{es}L|\theta_0|^2e^{-\alpha z}\delta(z - z'), \tag{43}$$

yielding

$$\langle F_Q(z, t)F_Q^\dagger(z', t')\rangle = 2\gamma_{es}L|\theta_0|^2e^{-\alpha z}\delta(z - z')\delta(t - t'), \tag{44}$$

valid when $\sigma_{ee} - \sigma_{ss}$ is not changing.

This yields a photon flux of

$$\begin{aligned}\frac{c}{L}\langle\mathcal{E}_w^\dagger(L, t)\mathcal{E}_w(L, t)\rangle = \frac{c}{L}\frac{\bar{g}^2N}{c^2}\int_0^L e^{-2\gamma_{es}t}H_1[\alpha, L, z_1'', t, 0]^2V|\theta_0|^2e^{-\alpha z_1''}dz_1'' \\ + \frac{c}{L}\frac{\bar{g}^2N}{c^2}\int_0^t\int_0^L e^{-2\gamma_{es}(t-t_1'')}H_1[\alpha, L, z_1'', t, t_1'']^2 \\ \times 2\gamma_{es}L|\theta_0|^2e^{-\alpha z_1''}dz_1''dt_1''.\end{aligned}$$

For sufficiently short detection times $\tau_d \ll \frac{1}{2\gamma_{es}}$, the noise contribution (second term) can be ignored, and furthermore when the photon number is much smaller than 1 ($\tau_d \ll \left\{\frac{\bar{g}^2N}{c}|\theta_0|^2\left(\frac{1-e^{-\alpha L}}{\alpha}\right)\right\}^{-1}$) we can consider just the leading term in the series expansion, and observe a constant flux

$$\begin{aligned}
&\frac{c}{L}\langle\mathcal{E}_{\rm w}^{\dagger}(L,\tau_{\rm d})\mathcal{E}_{\rm w}(L,\tau_{\rm d})\rangle\\
&=\frac{\bar{g}^2N}{c}|\theta_0|^2\int_0^L(I_0[2\sqrt{M[L,z_1'']\mathrm{e}^{-\alpha z_1''}\tau_{\rm d}}])^2\mathrm{e}^{-\alpha z_1''}\mathrm{d}z_1''\\
&\approx\frac{\bar{g}^2N}{c}|\theta_0|^2\int_0^L(1+2M[L,z_1'']\mathrm{e}^{-\alpha z_1''}\tau_{\rm d}+O(\tau_{\rm d}^2))\mathrm{e}^{-\alpha z_1''}\mathrm{d}z_1''\\
&=\frac{\bar{g}^2N}{c}|\theta_0|^2\frac{1-\mathrm{e}^{-\alpha L}}{\alpha}.
\end{aligned}\tag{45}$$

We therefore obtain a photon number of $\frac{\bar{g}^2N}{c}|\theta_0|^2\frac{1-\mathrm{e}^{-\alpha L}}{\alpha}\tau_{\rm d}$ within this short detection window. This is precisely the excited atom fraction multiplied by $\bar{d}\gamma_{es}\tau_{\rm d}$, since the fraction of atoms that were excited after the write pulse is $\frac{1}{L}\int_0^L\langle\sigma_{ee}(z,0)\rangle\mathrm{d}z=\frac{1}{L}\int_0^L|\theta_0|^2\mathrm{e}^{-\alpha z}\mathrm{d}z=|\theta_0|^2\frac{1-\mathrm{e}^{-\alpha L}}{\alpha L}$.

B.5. Number of prepared spins

We start with the description of the spin operator from equation (30), by defining $S=\sqrt{N}\sigma_{gs}\mathrm{e}^{-\mathrm{i}(\omega_1-\omega_2)z/c}$ and replacing $\sigma_{eg}(z,t)$ by its mean value $\theta_0^*\mathrm{e}^{-\alpha z/2}\mathrm{e}^{-\mathrm{i}\omega_1 z/c}$

$$\begin{aligned}
(\partial_t+\gamma_0)\hat{S}^{\dagger}(z,t)-F_S^{\dagger}(z,t)&=-\mathrm{i}\bar{g}\sqrt{N}\mathrm{e}^{\mathrm{i}k_{\rm w}r}\mathcal{E}_{\rm w}(z,t)\sigma_{eg}\\
&=-\mathrm{i}\bar{g}\sqrt{N}\mathcal{E}_{\rm w}(z,t)[\theta_0^*\mathrm{e}^{-\alpha z/2}].
\end{aligned}$$

Take the Laplace transform from $t\to\omega$ to see

$$(\omega+\gamma_0)S^{\dagger}(z,\omega)-S^{\dagger}(z,t=0)-F_S^{\dagger}(z,\omega)=C(z)\mathcal{E}_w(z,\omega),$$

where $C(z)=-\mathrm{i}\bar{g}\sqrt{N}\theta_0^*\mathrm{e}^{-\alpha z/2}$. Then we have

$$S^{\dagger}(z,\omega)=\frac{C(z)}{\omega+\gamma_0}\mathcal{E}_{\rm w}(z,\omega)+\frac{1}{\omega+\gamma_0}S^{\dagger}(z,t=0)+\frac{1}{\omega+\gamma_0}F_S^{\dagger}(z,\omega),\tag{46}$$

and noting that $\mathcal{L}^{-1}\left[\frac{1}{\omega+\gamma_0}\right]=\mathrm{e}^{-\gamma_0 t}$ yields

$$\begin{aligned}
S^{\dagger}(z,t)=C(z)&\int_0^t\mathrm{e}^{-\gamma_0(t-t')}\mathcal{E}_w(z,t')\mathrm{d}t'\\
&+\mathrm{e}^{-\gamma_0 t}S^{\dagger}(z,t=0)+\int_0^t\mathrm{e}^{-\gamma_0(t-t')}F_S^{\dagger}(z,t')\mathrm{d}t',
\end{aligned}\tag{47}$$

where the field expression $\mathcal{E}$ from the previous subsection is required. Ignoring terms that do not show up in the normal ordered $\langle S^{\dagger}S\rangle$, we have

$$\begin{aligned}
S^{\dagger}(z,t)=C(z)&\int_0^t\mathrm{d}t'\mathrm{e}^{-\gamma_0(t-t')}\\
&\times\left(\int_0^{t'}\mathrm{d}t''\mathrm{e}^{-\gamma_{es}(t'-t'')}H_2(\alpha,z,0,t',t'')\mathcal{E}_{\rm w}(0,t'')+\mathcal{E}_{\rm w}(0,t')\right).
\end{aligned}\tag{48}$$

Computing $\langle S^{\dagger}S\rangle$ requires the commutator $[\mathcal{E}_{\rm w}(z,t),\mathcal{E}_{\rm w}^{\dagger}(z',t')]=L\delta[z-z'-c(t-t')]$ and yields 4 terms. In the short time window where one can ignore the atomic dephasing $\left(\tau_{\rm d}\ll\frac{1}{2\gamma_0},\frac{1}{2\gamma_{es}}\right)$, and also where the photon number is much smaller than 1 $\left(\tau_{\rm d}\ll\left\{\frac{\bar{g}^2N}{c}|\theta_0|^2\left(\frac{1-\mathrm{e}^{-\alpha L}}{\alpha}\right)\right\}^{-1}\right)$, only one term dominates (the term independent of H_2). The number of spins is then equivalent to the photon number

$$\frac{1}{L}\int_0^L\langle S^{\dagger}(z,\tau_{\rm d})S(z,\tau_{\rm d})\rangle\mathrm{d}z\approx\frac{\bar{g}^2N}{c}|\theta_0|^2\frac{1-\mathrm{e}^{-\alpha L}}{\alpha}\tau_{\rm d}.$$

Appendix C. Phase matching

By assuming the retrieval process to perform retrieval from the exact same spin wave function $S(z,t)$ that has been created by the write pulse, we have assumed the degeneracy of the two metastable states $|g\rangle$ and $|s\rangle$. In general, the metastable states could have different energies which would lead to a read process from $S(z,t)\mathrm{e}^{2\mathrm{i}(\omega_e-\omega_s)z/c}$. However, this effect is negligible in the regime $|\omega_e-\omega_s|\frac{L}{c}\ll 1$.

Appendix D. Second order coherence

D.1. Multi-pair two-mode squeezing

To ensure that a single photon source is single mode in all degrees of freedom, one would have to verify that the outgoing emission is not produced in a combination of modes. In the system that we consider, one cause for multi-mode emission are multiple two mode squeezing processes occurring during the initial write process. Thereafter, the subsequent retrieval process yields a read photon in more than one mode. Here, we include a short section to explain how the unconditional autocorrelation measurement ($g^{(2)}(0)$) scales with the number of driven mode pairs [20], and this allows one to verify that no higher-number two mode squeezing processes have occured.

Let us first consider the state created by K vacuum squeezing processes

$$\rho_{\rm multi} = (1-p)^K\left[e^{\sqrt{p}\sum_{m=1}^K a_m^\dagger b_m^\dagger}|\Omega\rangle\langle\Omega|e^{\sqrt{p}\sum_{n=1}^K a_n b_n}\right], \tag{49}$$

where $|\Omega\rangle$ indicates the vacuum in all modes. Now consider a detector that sees all the K modes, giving a number operator of the form

$$\hat{N}_K = \sum_{c=1}^K a_c^\dagger a_c. \tag{50}$$

This yields

$$\mathrm{Tr}\left[\sum_{c,d=1}^K a_c^\dagger a_d^\dagger a_c a_d \rho_{\rm multi}\right] = \left(\frac{p}{1-p}\right)^2(K^2+K), \tag{51}$$

$$\mathrm{Tr}\left[\sum_{c=1}^K a_c^\dagger a_c \rho_{\rm multi}\right] = \frac{p}{1-p}K, \tag{52}$$

giving the unconditional autocorrelation of the a modes

$$g^{(2)}_{\rm multi} = \frac{\left\langle \sum_{c,d=1}^K a_c^\dagger a_d^\dagger a_c a_d\right\rangle}{\left\langle \sum_{c=1}^K a_c^\dagger a_c\right\rangle^2} = 1 + \frac{1}{K}, \tag{53}$$

which leads us to check if our photon field is indeed consistent with that coming from a single-pair two-mode squeezing process.

We thus compute the unconditional autocorrelation function of the read photon field at time t

$$g^2_{\rm read} = \frac{\langle \mathcal{E}_r^\dagger(0,t)\mathcal{E}_r^\dagger(0,t)\mathcal{E}_r(0,t)\mathcal{E}_r(0,t)\rangle}{\langle \mathcal{E}_r^\dagger(0,t)\mathcal{E}_r(0,t)\rangle^2},$$

In the regime we consider, where we have a short detection time and a fast readout, developing the numerator of the $g^{(2)}$ function leads to the term

$$\begin{aligned}&\langle \mathcal{E}_{\rm w}(0,t_a)\mathcal{E}_{\rm w}(0,t_b)\mathcal{E}_{\rm w}^\dagger(0,t_c)\mathcal{E}_{\rm w}^\dagger(0,t_{\rm d})\rangle\\ &=\langle \mathcal{E}_{\rm w}(0,t_a)\left[\mathcal{E}_{\rm w}^\dagger(0,t_c)\mathcal{E}_{\rm w}(0,t_b) + \frac{L}{c}\delta(t_b-t_c)\right]\mathcal{E}_{\rm w}(0,t_{\rm d})\rangle\\ &=\left(\frac{L}{c}\right)^2\delta(t_a-t_c)\delta(t_b-t_{\rm d}) + \left(\frac{L}{c}\right)^2\delta(t_a-t_{\rm d})\delta(t_b-t_c),\end{aligned}$$

which yields

$$g^2_{\rm read} = \frac{2\langle \mathcal{E}_r^\dagger(0,t)\mathcal{E}_r(0,t)\rangle^2}{\langle \mathcal{E}_r^\dagger(0,t)\mathcal{E}_r(0,t)\rangle^2} = 2$$

as it should, since we assume a mono-mode emission ($K=1$). Here we have used equation (25) and the leading term of equation (48).

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