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In situ determination of Zeeman content of collective atomic memories

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Received 14 January 2012, in final form 1 March 2012
Published 8 June 2012
Online at stacks.iop.org/JPhysB/45/124006

Abstract

Knowledge and control of atomic Zeeman populations is necessary for the realization of usef ul, long-lived quantum memories. We propose and implement a method to determine atomic state population distributions for atomic spin waves. Zeeman composition of single atomic spin waves of a cold atomic gas, confined in a one-dimensional optical lattice, is inferred with high precision by measurements of signal–idler polarization correlations as a function of spin-wave storage time.

(Some figures may appear in colour only in the online journal)

1. Introduction

The control of an atom’s external and internal degrees of freedom is a feature of modern research in ultracold atomic physics. For applications to quantum information processing, such as quantum networks based on the idea of the quantum repeater [1–12], magneto-optically trapped and cooled atoms can be used to store information in spin-wave excitations [3, 13–16]. The latter are characterized by a grating structure induced by laser Raman scattering, superimposed on the frozen atomic ensemble. These spin waves are distinguished by their internal spin structure, being defined by pairs of states in the hyperfine doublet of the atom’s electronic ground level. A spin wave based on the \( m = 0 \leftrightarrow m = 0 \) clock transition is only second-order sensitive to ambient magnetic fields and is thus attractive for the realization of long-lived quantum memory [17–22]. Alkali atoms prepared in magneto-optical traps are normally created in an internal mixed quantum state in which population is equally distributed across the Zeeman states of one of the hyperfine ground levels. Larmor precession of magnetically sensitive Zeeman states leads to characteristic collapse and damped revivals of the optically retrieved signal [23, 24]; the damping is a result of inhomogeneous broadening of the Zeeman components due to variations of the ambient magnetic field [17]. An atomic ensemble prepared in an unpolarized magnetic distribution of one of the hyperfine ground levels exhibits this characteristic signal over tens of \( \mu \)s storage time [17].

Long storage is also challenged by thermal motion of cold atoms on ms timescales, as determined by the time for displacement across a spin-wave grating period. Optical lattice confinement augmented by compensation schemes for the lattice-induced differential ac Stark shift of the clock doublet has resulted in storage times in the 100 ms range [20–22]. In such experiments, optical pumping into the \( m = 0 \) hyperfine Zeeman state may be employed in an attempt to empty other Zeeman states and thereby reduce the excitation of magnetically sensitive spin waves that dephase in less than a ms. Another form of optical pumping, of particular interest here, may occur in protocols such as the DLCZ protocol [2], which is the standard method for creating single atomic spin-wave excitations in a quantum memory. The protocol involves Raman scattering of a sequence of weak off-resonant write laser pulses between hyperfine ground levels of an initially prepared atomic ensemble until a Raman signal photon is detected. Specifically, in the experiments to be reported here, a trial begins with a 50 ns write pulse of power \( \simeq 1 \ \mu \text{W} \) incident on \(^{87}\text{Rb} \) atoms prepared in the \(| b \rangle = |5S_{1/2}, F_b = 1 \rangle \) ground hyperfine level and detuned \(-20 \text{ MHz} \) from the \(| c \rangle = |5P_{1/2}, F_c = 1 \rangle \) excited level. If a signal photon emitted on the \(| c \rangle \leftrightarrow | a \rangle \) \((| a \rangle = |5S_{1/2}, F_a = 2 \rangle)\) transition is not detected, the write pulse is followed 700 ns
later by a 200 ns clean pulse (power 270 μW) resonant with the $|a⟩ → |c⟩$ transition. Both the write and clean pulses have a transverse Gaussian intensity profile with a $1/e^2$ waist of 230 μm. After a quiescent interval, the next trial begins; the trial period is 1.5 μs. Figure 1 shows results for the populations of the $m = -1, 0, 1$ Zeeman states, $|b, m_b⟩$; of the initially unpolarized hyperfine level $|b⟩$ over 2000 trials. These results were found by numerically integrating the optical Bloch equations for a single $^{87}$Rb atom including all the relevant hyperfine structure. A few hundred trials are sufficient to noticeably alter the population distribution, whereas typically $10^2$–$10^4$ trials are needed to generate high-quality, single (i.e. with double excitations strongly suppressed) spin-wave excitations in experiments. A conventional way to discuss the distribution of population in the $F_b$ excitations in experiments. A conventional way to discuss the spin-wave storage (i.e. with double excitations strongly suppressed) spin-wave correlations are studied as a function of spin-wave storage time suitable measurements. Specifically signal–idler polarization spectroscopy (see for example [25, 26]). In this section, we summarize the basic physical principles involved in the storage and retrieval process in $^{87}$Rb. As we are interested in extracting information on the atomic population distribution produced by the protocol that generates single atomic spin waves, the role of hyperfine structure, laser polarization and external magnetic field are important factors. A detailed theoretical treatment of the system, including the latter features, is presented in the appendix: this is written in a rather general way so as to be applicable to other alkali atoms. In this section, we summarize some of the salient features useful for understanding of polarization correlations of the detected light in the $^{87}$Rb system that are discussed in the following section.

2.2. Spin-wave storage

The spin wave evolves in time under the influence of a magnetic field. During storage, the atomic hyperfine coherences undergo Larmor precession in the magnetic field $\tau_{\text{fast}} \ll \tau_e \ll \tau_{\text{slow}}$. In the former microsecond storage regime, the atomic state depends sensitively on the instant the spin wave is written. In the longer storage time limit, we take advantage of the long coherence time of the magnetically insensitive spin waves and the relatively benign effects of atomic motion over a sub-millisecond timescale in a cold atomic rubidium gas. While here we apply this technique to an ensemble of $^{87}$Rb atoms, we point out in the appendix that this method is applicable to any ensemble of three-level atoms whose ground hyperfine levels $|b⟩$ and $|a⟩$ have total respective angular momenta, $F_a$ and $F_b$, that satisfy $F_a = F_b + 1$, and whose excited level $|c⟩$ has a total angular momentum $F_c = F_b$.

The remainder of this paper is organized as follows. Storage and retrieval of spin waves are described in section 2. Theoretical analysis for the situation of pure atomic alignment is presented in section 3. Experimental methods are described in section 4; the observed data are presented in section 5. The details of the full theoretical model are given in the appendix.
We discuss a limiting case that does not require the general Landé \( g \)-factors for levels \( |a \rangle \) and \( |b \rangle \), respectively, and \( \delta g \equiv g_a - g_b \); numerically for \(^{85}\text{Rb} \), \( g_a = -0.5018 \) and \( \delta g = -0.0022 \). The Larmor frequencies \( \omega_{0,\pm 2} \) correspond to fast elementary spin waves \( \hat{s}_{m_0,m_1}(z,t) \) (with \( m_0, m_1 = (\pm 1, \pm 1) \) or \( (0, \pm 2) \)) and \( \omega_{0,\pm 1,\pm 1} \) correspond to slow elementary spin waves \( \hat{s}_{\pm 1,\pm 1}(z,t) \).

Magnetic field inhomogeneities cause Zeeman shifts that vary in space across the atomic ensemble causing the Heisenberg operator \( \hat{s}_{m_0,m_1}(z,t) \) to pick up a spatially varying phase factor \( e^{-i\omega_{m_0,m_1}(z,t)^2/B_0} \) resulting in dephasing of the stored excitations. After storage for time \( T_s \) in an ensemble with a Gaussian density profile \( f(z) \), the elementary distributed spin wave \( \hat{s}_{m_0,m_1}(z,t) \) suffers a Gaussian dephasing factor \( e^{-\Gamma_{m_0,m_1}^2(z,t)/2} \), where \( \Gamma_{m_0,m_1} \equiv |\omega_{m_0,m_1}B'/B_0| \) and \( l \) is the sample length. The fast and slow elementary spin waves dephase in proportion to their precession rates. For short enough storage time, both fast and slow spin waves may be observed, but for longer storage periods, only the slow spin waves remain.

### 2.3. Spin-wave retrieval.

After a storage time \( T_s \), a linearly polarized read field, counterpropagating with respect to the write laser field, is applied near resonance with the \( |a \rangle \leftrightarrow |c \rangle \) transition. The read laser converts the stored atomic excitation into an idler field by Raman emission on the \( |b \rangle \leftrightarrow |c \rangle \) transition. The idler field wavevector \( \mathbf{k}_r \) is determined by the four-wave-matching condition \( \mathbf{k}_r - \mathbf{k}_v + \mathbf{k}_p - \mathbf{k}_s = 0 \); hence, the idlers are counterpropagating to the signal field in our geometry. Idler and signal photon pairs are strongly correlated in this geometry as they result from a cyclic two-photon emission process in which each atom starts and ends in the same Zeeman state. When the read laser is polarized in the \( z \)-direction, parallel to the magnetic field, the influence of dephasing on the retrieval process is straightforward to analyse. In the appendix, we discuss the more general case in which the interaction Hamiltonian is expressed in terms of eigenstates of angular momentum along the laser polarization direction. In both cases, the retrieval process can be described in terms of dark-state polarizations.

### 3. Analysis of pure atomic alignment \( p_1 = p_{-1} \)

We discuss a limiting case that does not require the general theory of the appendix, in which the atoms are prepared in an aligned state (zero orientation \( \mathcal{O} = 0 \), corresponding to \( p_1 = p_{-1} \)).

To compute the dynamics of the signal–idler correlations, we need to follow the fast and slow rotation of the various Zeeman spin waves over the storage period. Some bookkeeping is required to keep track of various sets of bosonic spin waves naturally associated with the write and read processes with linearly polarized laser fields. Each of these subsidiary spin waves may be written in terms of the elementary fast and/or slow Zeeman spin waves. The subsidiary spin-wave dynamics clarifies characteristic features of the signal–idler photon correlations. Features due to interference of fast and slow spin waves and their experimental observation will be discussed later.

Recall that the elementary clock \( \hat{s}_{0,0} \) and Zeeman coherences \( \hat{s}_{\pm 1,\pm 1} \) are slowly varying in time, while \( \hat{s}_{0,\pm 1} \) and \( \hat{s}_{\pm 0,\pm 2} \) are rapidly varying. We identify ‘linearly polarized’ slow and fast spin-wave annihilation operators

\[
\hat{s}_{\pm 1,1}^{\text{slow}} = \frac{1}{\sqrt{2}} (\hat{s}_{1,-1} + \hat{s}_{-1,1}) \\
\hat{s}_{\pm 1,1}^{\text{fast}} = \frac{1}{\sqrt{2}} (\hat{s}_{1,-1} - \hat{s}_{-1,1})
\]

These evolve in time as a fixed axis rotation. For example, the slow spin waves \( \hat{s}_{1,1}^{\text{slow}} \) and \( \hat{s}_{1,1}^{\text{slow}} \) evolve in a constant magnetic field on the slow frequency scale as

\[
\left\langle \hat{s}_{1,1}^{\text{slow}}(z,t) \right| e^{-i\omega_{1,1}t} \left| \hat{s}_{1,1}^{\text{slow}}(z,0) \right\rangle = \left\langle \hat{s}_{1,1}^{\text{slow}}(z,t) \right| e^{-i\omega_{1,1}t} \left| \hat{s}_{1,1}^{\text{slow}}(z,0) \right\rangle,
\]

with \( \sigma \) the second Pauli matrix. The pairs of fast spin waves \( \left( \hat{s}_{1,1}^{\text{fast}}, \hat{s}_{1,1}^{\text{fast}} \right)^T \) and \( \left( \hat{s}_{0,0}^{\text{fast}}, \hat{s}_{0,0}^{\text{fast}} \right)^T \) evolve similarly, but with faster frequencies \( \omega_{1,1} \) and \( \omega_{0,2} \), respectively. The notation is chosen so that the V/H spin waves \( \hat{s}_{1,1}^{\text{fast}} \) are retrieved, after a storage period, as V/H-polarized idler fields.

A photoelectric detection event of a signal photon with polarization \( k = H \) or \( V \) imprints a single-mode collective write-spin-wave excitation \( \hat{A}_1 \) onto the ensemble. The atomic density matrix conditioned on this detection is

\[
\hat{A}_1 = \int \mathcal{D}z \sqrt{f(z)} (\mu_{\text{w}} \hat{w}_{0,0}(z) + \sqrt{1 - \mu_{\text{w}}^2} \hat{w}_{1,1}(z)),
\]

where \( f(z) = |\bar{n}(z)|^2 \int \mathcal{D}z |\bar{n}(z)|^2, \mu_H = \sqrt{2p_0/(1 + p_0)} \) and \( \mu_V = \sqrt{2p_0/(1 + p_0)} \). The local spin-wave operators \( \hat{w}_{m,k}(z) \) (\( w \) for write) are quasi-bosonic with commutation relations \( [\hat{w}_{m,k}(z), \hat{w}_{m',k'}(z')] = i \delta_{m,m'} \delta_{k,k'} \delta(z - z') + \mathcal{O}(1/\sqrt{N}) \), and may be written explicitly, in terms of the subsidiary fast and slow spin waves,

\[
\hat{w}_{1,1} = -\frac{1}{\sqrt{2}} \left( \hat{s}_{1,1}^{\text{slow}} - \hat{s}_{1,1}^{\text{fast}} \right).
\]
are given by \( \hat{\omega}_{1,V} = -\frac{1}{\sqrt{2}} (\hat{\omega}_{\text{flow}}^{\text{slow}} + \hat{\omega}_{\text{fast}}^{\text{fast}}) \). \( \hat{\omega}_{0,H} = \hat{\omega}_{\text{fast}}^{\text{fast}} \) and \( \hat{\omega}_{0,V} = \frac{1}{2} \hat{\omega}_{0,H} \).

The detection of an H/V signal photon produces spin waves that will be read out as V/H idlers. Note that \( \hat{\omega}_{0,V} \) has a slowly varying contribution from the clock transition \( \delta_0,0 \), in addition to fast spin waves associated with \( \delta_{1,\pm 2} \), while \( \hat{\omega}_{0,H} \) has only the latter.

The retrieval dynamics are governed by the Heisenberg–Langevin equations for the orthogonally polarized idler fields, \( \hat{\phi}_H(z,t) \) and \( \hat{\phi}_V(z,t) \), which obey the equal time commutation relations \( [\hat{\phi}_H(z,t), \hat{\phi}_V(z',t)] = \delta_{z,z'} \delta(z - z') \), and those for the atomic excitations. For our particular atomic configuration, we take advantage of symmetry to arrive at a set of just three equations (see appendix A.1), which identify the hyperfine read \( \hat{r}_{m,\lambda} \) and optical \( \hat{e}_{m,\lambda} \) spin waves that couple to the retrieved field,

\[
\begin{align*}
\hat{\rho} + i \hat{\rho}_\lambda &= i \hat{\rho}_{\lambda} = i \hat{\omega}_{\lambda} \hat{e}_{\lambda}, \\
\hat{\delta} - \Gamma_{1} / 2 \hat{\delta} &= i \hat{\omega}_{\lambda} \hat{e}_{\lambda}.
\end{align*}
\]

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Here \( \Gamma_{1} \) is the spontaneous decay rate of level \( |c\rangle \), \( C_{0,\lambda} = -1 / \sqrt{2} \) and \( C_{1,\lambda} = (-1)^{h_{1/2}} / \sqrt{2} \). The relative coupling strengths of the idler field to optical coherences \( \hat{e}_{m,\lambda} \), \( m = 0, 1 \). Similarly, \( C_{0,\lambda} = \sqrt{(3 + \delta_{0,H}) / 10} \) and \( C_{1,\lambda} = \sqrt{3 / 10} \) determine the relative coupling of the optical coherences and read spin waves; \( \Omega \) is the read field Rabi frequency, \( \kappa \) is the single-photon Rabi frequency on the idler transition and \( \delta_{0,\lambda} \) is a noise operator. The collective optical coherence operators are given by \( e_{1,\lambda} = -i (\hat{\phi}_{0,\lambda}^{\dagger} - \hat{\phi}_{1,\lambda}^{\dagger} + \hat{\phi}_{1,\lambda}^{\dagger} - \hat{\phi}_{0,\lambda}^{\dagger}) \). The retrieval of an H/V signal photon produces spin waves that will be read out as V/H idlers. Note that \( \hat{\omega}_{0,V} \) has a slowly varying contribution from the clock transition \( \delta_0,0 \), in addition to fast spin waves associated with \( \delta_{1,\pm 2} \), while \( \hat{\omega}_{0,H} \) has only the latter.

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The experimental methods are as follows. A sample of \( N \sim 10^7 \) \(^{87}\)Rb atoms is prepared in the lowest energy hyperfine ground level \( |b\rangle \) with angular momentum \( \ell_b = 1 \) (inset of figure 2). We consider the Raman configuration with ground levels \( |b\rangle \) and \( |a\rangle \) (\( F_a = 2 \)) and excited level \( |c\rangle \) with energies \( \hbar \omega_{0b}, \hbar \omega_{0a} \) and \( \hbar \omega_{va}, \) respectively. The level \( |c\rangle \) is the \( 5P_{1/2} \) hyperfine level with angular momentum \( F_c = 1 \). A linearly polarized \( \hat{\omega}_{\text{write}} \) laser pulse with wavevector \( k_{\text{slow}} \) and nearly resonant on the \( D_1 \) \((|b\rangle \leftrightarrow |c\rangle) \) transition is Raman scattered to produce a signal field on the \( |a\rangle \) \( \leftrightarrow |c\rangle \) transition. The write pulse is polarized in \( x-z \) plane travels at a small angle with respect to the negative \( z \)-direction. A magnetic field produced by a pair of coils in a Helmholtz configuration is applied in the \( z \)-direction. A single transverse signal mode \( u^0(r) \), centred around the wavevector \( k_{\text{slow}} = -k_{\text{fast}} \hat{z} \), is collected. A vertically polarized read laser pulse \( \hat{\omega}_{\text{read}} \) is counter-propagating to the write pulse. The retrieval of an H/V signal photon produces spin waves that will be read out as V/H idlers. Note that \( \hat{\omega}_{0,V} \) has a slowly varying contribution from the clock transition \( \delta_0,0 \), in addition to fast spin waves associated with \( \delta_{1,\pm 2} \), while \( \hat{\omega}_{0,H} \) has only the latter.

Atoms are collected and cooled in a magneto-optical trap for a period of 0.4 s. The trap laser is then detuned by up to 95 MHz below atomic resonance, the quadrupole magnetic field is turned off and the repump laser intensity is lowered for 40 ms, in order to optimize sub-Doppler cooling and loading of the optical lattice. As a result, the lattice contains about \( 10^7 \) atoms in the \( 5S_{1/2}, F = 1 \) hyperfine level (level \( |b\rangle \)), with magnetic field \( B_0 = 1.1 \) G applied along the
conditioned retrieval efficiencies, $\eta$, spin waves and $\omega$ efficiencies on storage times can be exploited to infer the 5.1. Retrieval efficiencies for aligned atoms

5. Results

20 $\mu$z

$\eta$ calculations of the write/clean sequence suggest that the write field. Detection of a signal event by D1 or D2 heralds excitation of the desired atomic spin wave, and halts the write/clean sequence. Under our experimental conditions, the fast coherence decay time $\tau_{\text{fast}} \sim 100 \mu s$, consistent with milli-Gauss variations in the magnetic field [17]. Numerical calculations of the write/clean sequence suggest that $p_0$ falls from its initial value of 1/3 to a steady state value of 0.15 within the first few milliseconds of the 72 ms protocol duration (figure 1). The alignment symmetry $p_m = p_{-m}$ is broken by the presence of the bias magnetic field, at the level of about 10% under our experimental conditions.

5. Results

5.1. Retrieval efficiencies for aligned atoms

In this section, we show how the dependence of retrieval efficiencies on storage times can be exploited to infer the populations of the Zeeman state $|b, 0\rangle$ given that the atomic populations are balanced, i.e. $p_1 = p_{-1}$. Figure 3 illustrates the sensitivity of the read-out efficiency $\eta_{\lambda\lambda'}$ on the population $p_0$ for storage times $T_s$ much less than the fast coherence decay time $\tau_{\text{fast}}$. When an H-polarized signal photon is detected, the spin waves $\tilde{w}_{0,H}$ and $\tilde{w}_{1,H}$ are imprinted on the ensemble. At $T_s = 0$ and $\omega_{0,3} T_s = 2\pi$, only atoms originating in the $m = 0$ state contribute to $\eta_{\lambda\lambda'}$. This can be traced to destructive interference between the V-polarized fast and slow spin waves originating from the $m = \pm 1$ states, $[\tilde{r}_{1,V}, \tilde{w}_{1,H}^{\dagger}]|T_s=0 = 0$. At intermediate times, the fast spin-wave rotation modulates the interference, and the read-out contributions of $\omega_{1,H}$ and $\omega_{0,H}$ are determined by the populations of the $m = \pm 1$ and $m = 0$ states. The sensitivity of this and the other three conditioned retrieval efficiencies, $\eta_{H,H}, \eta_{V,V}$ and $\eta_{V,H}$, allows us to accurately infer the population $p_0$ from experimental observations.

For $T_s \ll \tau_{\text{fast}}$, we evaluate equation (16) with $\omega_{0,2} = \omega_{0,1}$ and $\omega_{1,-1} \rightarrow 0$ to give

$$\eta = \begin{bmatrix}
(1+2p_0)^2 \sin^2(\omega_{0,2} T_s) & 3(1+(1+2p_0) \cos(\omega_{0,2} T_s)) \\
(2p_0+1)^2(2p_0+1)p_0 & 25p_0^2+3(2p_0+1) \\
(1-p_0-(1-5p_0) \cos(\omega_{0,2} T_s))^2 & 415p_0^2+3(2p_0+1)
\end{bmatrix},$$

where $\eta$ is the matrix with elements $\eta_{\lambda\lambda'}, \lambda, \lambda' \in \{H, V\}$.

Figure 3. The sensitivity of retrieval efficiencies to the population $p_0$ in an ensemble without orientation ($p_1 = p_{-1}$). The efficiency $\eta_{H,H}(T_s)$ with which an H-polarized idler photon is retrieved from the ensemble given the detection of a V-polarized signal for storage times $T_s$ much less than the fast spin-wave decoherence time. This conditioned efficiency is calculated over one oscillation period of the fast spin waves ($0 \leq \omega_{0,2} T_s/(2\pi) \leq 1$) for the range of populations $0 \leq p_0 \leq 1$.

Figure 4. The measured retrieval efficiencies $\eta_{\lambda\lambda'}(\lambda, \lambda' = H, V)$ for $T_s \ll \tau_{\text{fast}}$. Left column: without optical pumping; right column: with optical pumping. The circles, diamonds, squares and triangles are for HV, VV, HH and VH idler–signal combinations, respectively. The solid curves are theoretical, fitted according to the text.

The short-time retrieval efficiencies are shown in figure 4, left column. By fitting the matrix $\eta$ to the four measured efficiencies with a single set of three adjustable parameters (an overall amplitude, $\omega_{0,2}$ and $p_0$), we obtain $p_0 = 0.183 \pm 0.003$. The quoted uncertainty is determined by the analysis of distributions of $p_0$ generated from data sets that are Poisson-distributed around the measured means. To account for non-equal detection efficiencies, we rotate signal and idler polarizations by 90° every 20 min. The total acquisition time is 160 min. A full scan over $T_s$ (2.1–3.7 $\mu s$) is performed within 48 s to avoid slow systematic drifts.

To confirm the predicted influence of $p_0$ on the retrieval efficiencies, we optically pump with an additional laser field tuned to the $|b\rangle \leftrightarrow |c\rangle$ transition, propagating in the $x$-direction

$\tau_{\text{fast}}$
an additional independent background contribution. The solid curves represent theory, each with
and linearly polarized in the z-direction. During the optical pumping, atoms accumulate in the |b, m = 0⟩ state, whereas
the write sequence reverses this tendency. The optical pumping period, during which a cleaning field empties level 𝑎,
is 30 𝜇s, followed by 80 write/clean cycles before repeating the sequence. The measured efficiencies are shown in the bottom row of figure 4. Using the procedure described above, we extract the value 𝑝₀ = 0.47 ± 0.01. By varying the optical
pumping, we have obtained 𝑝₀ as high as 0.75.

We have not employed a completely independent method to verify the correctness of the inferred Zeeman distributions. However, as an additional check we look at the dynamics of the slow hyperfine coherences.

For 𝑡𝑠𝑙𝑜𝑤 ≪ 𝑇ᵣ ≪ 𝑡𝑠𝑙𝑜𝑤, the retrieval efficiencies can be written in the general form

\[ \eta_{Δλ}(Τ_r) = |p₀χ_{Δλ}d_{ΔλV} + p₁χ_{Δλ}(Τ_r)|^2, \]

where \( \chi_{Δλ} \) is a constant contribution from the clock spin wave and \( \chi_{Δλ}(Τ_r) \) is the amplitude of the slow spin waves \( \delta_{Δλ} \).

We note that the clock spin wave contributes only to \( \eta_{HV} \).

The probability amplitudes for Raman excitation of the clock transition interfere constructively and destructively for V- and H-polarized signal emission, respectively. Explicitly, the retrieval efficiency matrix is

\[ \eta = \frac{(1 - p₀^2) \sin^2(\omega_0 T_s) + (1 + p₀^2) \sin^2(\omega_0 T_s) + 2p₀(1 - p₀) \cos(\omega_0 T_s))}{2(1 - p₀^2) \sin^2(\omega_0 T_s) + 2p₀(1 + p₀) \sin^2(\omega_0 T_s) + 4p₀^2 + 4p₀}. \]

We note that in a limit of 𝑝₀ = 0 for storage times

\[ t_{slo\_w} \ll T_r \ll t_{slo\_w}, \]

only \( \delta_{ΔλV} \) contribute to the idler retrieval. For intermediate storage times, this pair of excitations can serve as a qubit where the Larmor spin-wave evolution approximates qubit rotation about a fixed axis.

In figure 5, we show efficiencies measured in the millisecond storage time regime, in the absence of optical
pumping. By fitting the data with the above theoretical expressions, we extract \( p₀ = 0.16 \pm 0.02 \), in agreement with the value inferred from the dynamics of the fast coherences. This value is sufficiently small that \( \eta_{HV} \) and \( \eta_{HV} \) show sinusoidal rotation, whereas \( \eta_{V} \) and \( \eta_{HV} \) are additionally modulated, as expected. The relative efficiencies at short and long times, shown in figures 4 and 5, are consistent with our theory, when adjusted at long times by an additional factor of 3/5. This factor is in agreement with our measurements of the effects of atomic motion on the clock spin-wave dynamics, cf [17].

Although the calculations leading to figure 1 indicate that repeated write/read trials produce a slight orientation (on the order of 10%) of the atomic sample, here we have presented in situ measurements of \( p₀ \) assuming that the populations are perfectly balanced (\( p₁ = p₋₁ \)) in order to illustrate the theory presented in section 3. From the general theory presented in the appendix, we can compute the retrieval efficiencies presented in this section for arbitrary orientation. However, for the combination of field polarizations considered in this subsection, a small orientation does not result in a statistically significant effect. A non-zero orientation would best be observed by measuring conditioned retrieval efficiencies in a basis where the signal and idler are not linearly polarized. We discuss such a scenario in the following subsection.

5.2. Photon correlations for oriented and aligned atoms

The presence of atomic orientation slightly complicates the picture. Whereas the spin waves \( S_{m₀m₀} \) and \( S_{m₋₁m₋₁} \) contribute equally to the written excitations and dark-state polaritons when the populations \( p₁ \) and \( p₋₁ \) are balanced, an imbalance no longer permits one to exploit the treatment in section 3. In that section, the symmetry allowed us to identify specific optical and hyperfine coherences that coupled to either horizontally or vertically polarized light. For each polariton, there existed a corresponding dark-state polariton which the read process retrieves. In general, the two polaritons propagate at different group velocities and experience walk-off. Although we neglected walk-off in our calculation of retrieval efficiencies, identification of the dark-state polariton polarizations was essential in determining the specific linear combinations of hyperfine coherences of which they are comprised.

When the populations are unbalanced, the system still supports two distinct dark-state polaritons that correspond to idlers of orthogonal polarizations that experience walk-off. However, these polarizations are no longer linear. One can readily see that this is the case in the extreme limit where all of the atoms are initially in the state |b, 1⟩. For example, when \( p₁ = 1 \), \( p₋₁ = 0 \). Here, because \( F_r = F_o \), right circularly polarized light would propagate through the sample at speed \( c \) and not interact with the atoms, while the left circularly polarized idlers experience a diminished group velocity and can be stored in the ensemble. In that special case, detection of a signal photon can only prepare an idler polariton with left circular polarization. These polaritons, however, still contain fast and slow coherences and the retrieval efficiency would therefore oscillate with storage time, as they did when the populations were balanced. For arbitrary orientation and alignment, the idler polarizations corresponding to the dark-state polaritons supported by the ensemble must be calculated using the general formalism presented in the appendix. Detection of an idler along a certain axis then represents a detection of a superposition of the two dark-state polaritons.

When the imbalance in the population is large, the effects of the atomic orientation better manifest themselves when the signal and idler polarizations are observed in the circular basis (\( \hat{e}_+, \hat{e}_- \)) as shown in figure 6. To create the atomic orientation, we optically pump the ensemble with circularly polarized fields prior to the first
data are plotted in figure 6, resulting in inferred values of the theoretical, fitted according to the text. Left column: right-hand polarized optical pumping; right column: left-hand polarized optical pumping. The circles, diamonds, squares and triangles are for $-\ldots, +\ldots, -\ldots$ and $+\ldots$ idler–signal combinations, respectively. The solid curves are identical, and we have neglected group velocity walk-off.

write trial. Circular polarizations are observed by inserting quarter-wave plates into the signal and idler paths (see figure 2). Figure 6 shows measurement probabilities of circularly polarized idler fields conditioned on the detection of a left or right circularly polarized signal after a storage time $T_s$. The data are fit to equation (A.42) using the population $p_0$ and orientation $(p_1 - p_{-1})/2$ as adjustable parameters. Each of the two dark-state polaritons supported by the ensemble propagates at different group velocities, which, in principle, results in differing temporal envelopes $\phi_j(t)$ ($j = 1, 2$) for photons retrieved from either of these modes. For simplicity, in our calculations, we assumed that these envelopes are identical, and we have neglected group velocity walk-off. This approximation contributes to imperfections of the fits of our model to experimental data. The fits of these to the data are plotted in figure 6, resulting in inferred values of the Zeeman level populations. For the data in the left column, we performed optical pumping to more heavily populate the state $|b, 1\rangle$, while for the right column, the state $|b, -1\rangle$ was more heavily populated.

6. Conclusion

In conclusion, we have presented a method to determine Zeeman populations of an atomic sample used as a quantum memory. Repeated attempts to produce a collective excitation within the quantum memory alter the populations of the Zeeman states. By examining the retrieval efficiencies of an idler conditioned on the detection of a signal for various field polarizations and storage times, one can measure the resulting Zeeman populations in situ. The theory is in agreement with measurements on a cold atomic sample in a 1D optical lattice.

Acknowledgments

This work was supported by the Air Force Office of Scientific Research, Atomic Physics and Quantum Memories MURI Programs and the National Science Foundation. One of us (SDJ) would like to thank the EPSRC and the Leverhulme trust for support.

Appendix. Theoretical model

We consider an optically thick ensemble of $N \gg 1$ alkali atoms with ground state hyperfine levels $|a\rangle$ and $|b\rangle$. The atoms are prepared in the hyperfine ground level $|b\rangle$ with the lowest total angular momentum $F_b$. The initial single-atom density matrix is a mixed state in which an atom finds itself in the Zeeman state $|b, m\rangle$ with probability $p_m$. In alkali atoms, the second ground level $|a\rangle$ has the total angular momentum $F_a = F_b + 1$.

To generate and subsequently retrieve collective spin-wave excitations within the ensemble, we consider a $\Lambda$ configuration where all field frequencies (with the exception of those used for optical trapping) interact primarily with an excited level $|c\rangle$ with total angular momentum $F_c = F_a$. The atoms, labelled by an index $\mu = 1, \ldots, N$, are distributed in the ensemble (independent identically distributed) random positions $r_\mu$ such that the mean number density is $n(r)$.

We apply a magnetic bias field with magnitude $B$ along the z-axis, which shifts the energies of the Zeeman states $|f, m\rangle$ within a hyperfine level $|j\rangle$ by $g_j \mu_B B$ where $g_j$ is the anomalous Landé $g$ factor and $\mu_B$ is the Bohr magneton. However, imperfections in the field coils as well as residual ambient magnetic fields could result in inhomogeneities in the bias field. These inhomogeneities, as we discuss later in this appendix, introduce spatial variations in the Zeeman energy shifts, and consequently could result in dephasing of any spin waves during the storage process.

To generate a correlated state between a scattered signal photon and a stored collective atomic excitation, one shines write field propagating in the direction $\hat{k}_w$ nearly resonant on the $|b\rangle - |c\rangle$ transition with detuning $\Delta$; the write field frequency is $c k_w = (\omega_t - \omega_b) + \Delta$. For simplicity, we assume that the write field is uniform over the ensemble such that we may take its electric field at position $\mathbf{r}$

$$E_w(\mathbf{r}, t) = E_w^{(+)}(\mathbf{r}, t) + E_w^{(-)}(\mathbf{r}, t),$$

where the positive frequency component $E_w^{(+)}(\mathbf{r}, t) \equiv \hat{\epsilon}_w E_w(t) \exp(i(k_w \cdot r - c k_w t))$ and $E_w^{(-)}(\mathbf{r}, t) \equiv E_w^{(+)}(\mathbf{r}, t)$. We collect a signal field with wavenumber $k_s$ and frequency $c k_s = \omega_s - \omega_b + \Delta$ nearly resonant on the $|a\rangle - |c\rangle$ transition with transverse profile $u(r_{\perp})$, where we have neglected diffraction so that $u$ only depends on $r_{\perp} \equiv \mathbf{r} - \hat{k}_w \mathbf{k}_w$. In the paraxial and slowly varying envelope approximations, the quantized electric signal field is given by $\hat{E}_s^{(+)}(\mathbf{r}, t) + \hat{E}_s^{(-)}(\mathbf{r}, t)$ where the positive frequency component is given in the interaction picture by

$$\hat{E}_s^{(+)}(\mathbf{r}, t) = \sqrt{\frac{\hbar k_s}{2\epsilon_0}} e^{i(k_s \cdot \mathbf{r} - c k_s t)} u^*(r_{\perp}) \times \sum_{s(H,V)} \hat{\epsilon}_{s}(\mathbf{k}_s) \hat{\psi}_s(t - \mathbf{k}_s \cdot \mathbf{r} / c),$$

(A.1)

the negative frequency component $\hat{E}_s^{(-)}(\mathbf{r}, t) = \hat{E}_s^{(+)}(\mathbf{r}, t)$, and $\hat{\epsilon}_{H}(\mathbf{k})$ and $\hat{\epsilon}_{V}(\mathbf{k})$ are respectively the horizontal and vertical polarizations.
vertical polarization vectors associated with a propagation direction \( \mathbf{k} \) such that \( \{ \mathbf{e}_r(\mathbf{k}), \mathbf{e}_\perp(\mathbf{k}), \mathbf{k} \} \) form a right-handed coordinate system and \( \psi_r(t) \) are photon field operators that satisfy the approximate slowly varying bosonic commutation relations \( \{ \hat{\psi}_r(t), \hat{\psi}_r(t') \} = i \delta(t - t') \).

These fields interact with the atomic electric dipoles for the atoms labelled by an index \( \mu = 1, \ldots, N \). When one adiabatically eliminates the excited state, one arrives at the Raman scattering Hamiltonian governing the write process

\[
\hat{H}_w = \hat{V} + \hat{V}',
\]

where

\[
\hat{V} = \sum_{\mu=1}^{N} \sum_{f,g} \mathbf{E}_w(-\mathbf{r}, t) \cdot \hat{\mathbf{d}}_{f,g}^\dagger(t) \hat{\mathbf{d}}_{f,g}(t) \frac{i}{\hbar} \mathbf{F}_f \mathbf{F}_g \hat{F}_f \hat{F}_g
\]

(A.2)

dipole transition operator between two arbitrary levels \( |f\rangle \) and \( |g\rangle \) is \( \hat{\mathbf{d}}_{f,g}^\dagger(t) = \hat{\mathbf{d}}^\dagger(t) \hat{\mathbf{d}}_g(t) \), and \( \hat{\mathbf{d}}_{f,g}^\dagger(t) \) is the projection onto the level \( |f\rangle \) with the total angular momentum \( \mathbf{F}_f \) for atom \( \mu \).

We find it useful to express these dipole transition operators between the levels \( |f\rangle \) and \( |g\rangle \) in terms of a vector of Clebsch–Gordan coefficient matrices

\[
\hat{C}_{Ff,Fg} = \sum_{m=1}^{F_f+1} \xi_{f,g} \hat{C}_{Ff,Fg,m} \hat{C}_{Ff,Fg,m},
\]

(A.4)

where \( \xi_{f,g} \) and \( \hat{C}_{Ff,Fg,m} \) are the spherical basis vectors and the \( \hat{C}_{Ff,Fg,m} \) are \( (2F_f + 1) \times (2F_g + 1) \) matrices with elements

\[
[\hat{C}_{Ff,Fg,m}]_{m_1,m_2} = \hat{C}_{Ff,Fg,m_1,m_2}
\]

and \( \hat{C}_{Ff,Fg,m} \) for \( F_f, F_g \) and \( m_f = -F_f, \ldots, F_f \) and \( m_g = -F_g, \ldots, F_g \) correspond to possible magnetic quantum numbers. Between any two levels \( |f\rangle \) and \( |g\rangle \), we also define the matrix of slowly varying transition operators \( \hat{\sigma}_{f,g}^\mu \) whose elements are the transition operators between individual Zeeman states. Explicitly, these matrix elements are

\[
[\hat{\sigma}_{f,g}^\mu(t)]_{m_f,m_g} = \hat{\sigma}_{f,g}^\mu(t) = \exp(i(\omega_{f-g} - \omega_f) t) \hat{\sigma}_{f,g}^\mu(t) \text{ so that,}
\]

(A.6)

where \( \hat{\sigma}_{f,g}^\mu \) is the evolution and ultimately preservation of this position-dependent phase that is key to being able to retrieve any imparted information after some storage time. The collective spin-wave operator connecting the states \( |b, m_b\rangle \) and \( |a, m_a\rangle \) is given by

\[
\hat{S}_{m_a,m_b} = \sum_{a=1}^{N} u^*(\mathbf{r}_a) e^{i(\mathbf{k}_a - \mathbf{k}_f)} \hat{\sigma}_{b,m_a}^\mu(t) \hat{\sigma}_{m_b,m_a}^\mu(t)
\]

(A.12)

When the total number of photons scattered into either the signal field mode or other modes is much less than the number of atoms in the ensemble, by the central limit theorem, these spin-wave operators satisfy the quasi-bosonic commutation relations

\[
[\hat{S}_{m_a,m_b}, \hat{S}_{m_c,m_d}^\dagger] = \delta_{m_a,m_c} \delta_{m_b,m_d} \left( 1 + O(1/\sqrt{N}) \right).
\]

(A.13)

where \( O(1/\sqrt{N}) \) correction term is a Gaussian random variable resulting from the statistical distribution of the atomic positions. Substituting for the dipole operators in equation (A.7) into the write process interaction of equation (A.3), one finds that this interaction can be written exactly in terms of the spin waves as

\[
\hat{V}(t) = \hbar \chi \hat{F}(t) \sum_{\lambda} X_{\lambda} \hat{\psi}_{\lambda}(t) \hat{\chi}_{\lambda},
\]

(A.14)

where \( \hat{\chi}_{\lambda} \) is the collective atomic excitation imprinted by a signal photon with polarization \( \hat{\mathbf{e}}_{\lambda} \), the interaction parameter

\[
\chi = \sqrt{\frac{\delta \omega}{2 \Delta}} \sqrt{\frac{\hbar}{\Delta}} \sqrt{\int |E_w(t)|^2 \frac{\hbar}{\Delta}},
\]

(A.15)

and the temporal envelope of the emitted signal photon is

\[
\phi(t) = \frac{E_w(t)}{\int |E_w(t)|^2 dt}.
\]

(A.16)

The collective atomic operator is expressed in terms of Zeeman spin waves as

\[
\hat{\chi}_{\lambda} = \frac{\text{Tr}[P \hat{S}_{\hat{\mathbf{e}}_{\lambda} \cdot \hat{\mathbf{C}}_{Ff,Fg}] \hat{\mathbf{C}}_{Ff,Fg}] \hat{\mathbf{C}}_{Ff,Fg}]}{X_{\lambda}}.
\]

(A.17)

where \( \hat{\mathbf{C}}_{Ff,Fg} \) is the associated phase

\[
[\hat{\mathbf{C}}_{Ff,Fg}, \hat{\mathbf{C}}_{Ff',Fg'}] = \delta_{Ff,Ff'} \delta_{Fg,Fg'}
\]

(A.18)
Because light scattered into modes other than the collected signal mode leaves behind spin waves that for \( N \gg 1 \) are orthogonal to that generated by the signal [27], when one traces over the undetected signal modes, the density matrix describing the system after the write interaction is given by

\[
\hat{\rho} = \hat{U}(\chi)\hat{\rho}_0\hat{U}^\dagger(\chi),
\]

(A.19)

where the unitary operator

\[
\hat{U}(\chi) = T \exp\left( -\frac{i}{\hbar} \int dt \left( \hat{V}(t) + \hat{V}^\dagger(t) \right) \right),
\]

(A.20)

and \( T \) indicates time ordering. When the interaction parameter \( \chi \ll 1 \), as it is in the experiments under consideration, the density matrix may be written as

\[
\hat{\rho} \approx \left( 1 - i\chi \sum_k \hat{a}_k^\dagger \hat{a}_k \right) \hat{\rho}_0 \left( 1 + i\chi \sum_k \hat{a}_k^\dagger \hat{a}_k \right),
\]

(A.21)

where the single signal mode operator is given by \( \hat{a}_k \equiv \int dt \hat{\phi}^\dagger(t)\hat{\phi}_k(t) \). The emission of a signal photon of polarization \( \hat{e}_k \), leaves behind an atomic excitation in the mode \( \hat{A}_k \). The collective atomic operators satisfy the commutation relations \([\hat{A}_k, \hat{A}_k^\dagger] = 1 + O(1/\sqrt{N}) \). However, depending on the initial population distribution, the atomic excitations left behind by photons of orthogonal polarizations \( \lambda_1, \lambda_2 \) are not necessarily orthogonal. This will not affect the calculation of retrieval efficiencies based on the detection of a single excitation in the limit \( \chi \ll 1 \), however.

After a storage time \( T_s \), a classical read pulse with linear polarization \( \hat{e}_k \), travelling in the direction \( \hat{k}_s = -\hat{k}_w \), is shined on the ensemble. This read pulse is resonant on the \( |a\rangle \leftrightarrow |c\rangle \) transition. The interaction of the read field with the ensemble, in turn, leads to the emission of an idler photon with a central frequency resonant on the \( |b\rangle \leftrightarrow |c\rangle \) transition. We collect a single transverse mode of the idler field propagating in the direction \( \hat{k}_i = -\hat{k}_s \). This mode has a transverse profile conjugate to that of the signal field. The positive frequency component of the idler field is then given by

\[
\hat{E}_i^{(+)}(r, t) = \sqrt{\frac{\hbar c}{2\varepsilon_0}} e^{i(k_r \cdot r - c k_r t)} \sum_{j \in \{H,V\}} \hat{e}_j(\hat{k}_r)\hat{\phi}_j(r_i, t),
\]

(A.22)

where \( r_i \equiv \hat{k}_i \cdot r \) and the slowly varying operators \( \hat{\phi}_j(r_i, t) \) satisfy the equal time commutation relations \([\hat{\phi}_j, \hat{\phi}_k^\dagger] = \delta_{j,k}\delta_{j} \hat{\phi}_j \cdot \delta_{j} \hat{\phi}_k \).

When the length of the atomic ensemble \( L \ll |\mathbf{k}_s - \mathbf{k}_w - \mathbf{k}_f + \mathbf{k}_i|^{-1} \), and in the absence of any dephasing mechanisms during storage, the idler field is phase matched with the stored atomic excitations [27, 29].

The read and idler fields interact with the atoms via the electric dipole interactions. In the rotating wave approximation, the interaction potential is given by

\[
\hat{H}_I = \sum_{\mu=1}^N \left( \hat{V}_\mu^\mu + \hat{V}_r^\mu + \hat{V}_i^\mu \right) + \text{h.c.},
\]

(A.23)

where the interaction of atom \( \mu \) with the idler field is \( \hat{V}_i^\mu = -\hat{E}_i^{(+)}(r_i, t) \cdot \hat{d}_{\mu,(b)}^i \) and \( \hat{V}_r^\mu = -\text{e}^{i(k_r \cdot r - c k_r t)}\hat{E}_i(t) \cdot \hat{d}_{\mu,(a)}^i \) is the interaction with the read field. The interaction with the undetected field modes \( \hat{V}_r^\mu \) accounts for spontaneous emission.

Before examining the propagation dynamics of the idler field within the ensemble, let us look in detail at the form of the read field interaction. From equation (A.7), one can immediately write this interaction in terms of transitions between Zeeman states as

\[
\hat{V}_r^\mu = \hbar \Omega(t) \sum_{a=1}^1 \left( \xi_{\mu}^a \cdot \hat{e}_r \right) \left( \hat{\sigma}_\mu^a \cdot \hat{K} \right)
\]

(A.24)

\[
= \hbar \Omega(t) e^{i(k_r \cdot r - c k_r t)} \sum_{a=1}^1 \left( \xi_{\mu}^a \cdot \hat{e}_r \right)
\]

\[
\times \sum_{\mu=0} F_{\mu} \sum_{m=-F_\mu}^{F_\mu} \bar{C}_{\mu m} F_m |c, m + \alpha\rangle |a, m\rangle,
\]

(A.25)

where \( \Omega(t) \equiv c_{\tilde{q}, \mu} E_r(t) / \hbar \) is the read field Rabi frequency, and we have defined the matrix \( \mathcal{K} \equiv \xi_{\mu}^a \cdot \mathcal{C}_{F_\mu,F_\mu} \). When the read field polarization is oriented along the \( z \)-axis, this interaction takes on a relatively simple form in which the read field couples a state in the excited level \( |c, m\rangle \) to single ground state \( |a, m\rangle \). One can recover a similar simplification for an arbitrary linearly polarized read field by expressing this potential in terms of eigenstates of the angular momentum along the polarization direction. Such a state within a level \( \{f\} \) can be expressed as \( |f, m\rangle = \sum_m \bar{D}_{f m} |f, m\rangle \), where \( \bar{D}_{f m} \) is a matrix element of the appropriate rotation operator acting on a level with angular momentum \( F_f \). Explicitly,

\[
\hat{V}_r^\mu = \hbar \Omega(t) \sum_{m=-F_\mu}^{F_\mu} \bar{C}_{\mu m} F_m |c, m\rangle |a, m\rangle.
\]

(A.26)

Expressed in this way, one readily sees that the states \( |\tilde{a}, \pm F_\mu\rangle \) are dark with respect to the read field, and hence, that spin waves involving these states cannot be accessed during the read process. On the other hand, for the atomic level configuration under consideration, every excited state \( |c, m\rangle \) \( (-F_\mu \leq m \leq F_\mu) \) is resonantly coupled to a ground state \( |\tilde{a}, m\rangle \) via the read field. The presence of this resonant coupling for every excited Zeeman level, as we will see later, permits the formation of electromagnetically induced transparency for a resonant idler field. Thus, comparing the expressions for \( \hat{V}_r \) in equations (A.24) and (A.26), one can deduce that

\[
\mathcal{K} = \mathcal{D}_{F, F}^r \xi_{\mu}^a \mathcal{D}_{F, F}^r \mathcal{D}_{F, F}^r.
\]

(A.27)

In the weak idler limit, where the probability of any one atom to be displaced from its ground state is negligible (i.e. the populations \( \{\tilde{a}_{\mu,\epsilon,m},\{\tilde{a}_{\mu,\epsilon,m}\} \ll 1 \) ), the interaction Hamiltonian (equation (A.23)) leads to the following Heisenberg–Langevin equations describing the propagation of the idler field:

\[
\left( \frac{d}{dt} + c \hat{k}_i \cdot \hat{V} \right) \hat{\phi} = i\sqrt{\hbar} \hat{e}^{\dagger} \mathcal{D}_{F,k_i}^{\dagger} \mathcal{D}_{F,k_i} \frac{\mathcal{L}}{2} \hat{e} + i\Omega \hat{\mathcal{K}} + \hat{\zeta},
\]

(A.28a)

\[
\left( \frac{d}{dt} + \frac{\Gamma}{2} \right) \hat{\phi} = i\sqrt{\hbar} \left( \mathcal{P}^{1/2} \mathcal{C}_{F,k_i}^{\dagger} \hat{\phi} \right) + i\hat{\Omega} \hat{\mathcal{K}} + \hat{\zeta},
\]

(A.28b)

\[
\frac{d}{dt} \hat{\phi} = i\Omega \hat{\mathcal{K}},
\]

(A.28c)
where \( \delta(z,t) \) and \( \hat{\epsilon}(z,t) \) are the matrices of hyperfine and optical local spin waves respectively with the matrix elements

\[
\delta_{\mu_1 \mu_2}(r_i, t) \equiv \sum_{\mu=1}^{N} u^*(\mu, r_i) \ e^{-i(k_r - k_i)r_i} \frac{\delta(r_i - r_{\mu})}{\sqrt{\rho_\mu}} \hat{a}_{\mu_1 \mu_2}^{\mu},
\]

(A.29a)

and

\[
\hat{\epsilon}_{\mu_1 \mu_2}(r_i, t) \equiv \sum_{\mu=1}^{N} u(\mu, r_i) \ e^{-i(k_r - k_i)r_i} \frac{\delta(r_i - r_{\mu})}{\sqrt{\rho_\mu}} \hat{a}_{\mu_1 \mu_2}^{\mu},
\]

(A.29b)

Thus, in the adiabatic limit, the propagating idler field obeys the propagation equation

\[
\left( \frac{\partial}{\partial t} + c \hat{\mathbf{k}} \cdot \mathbf{V} \right) \hat{\phi} = -\frac{n_0}{\Omega^2} \text{Tr}[\mathcal{R} \mathcal{R}^T] \cdot \frac{\partial}{\partial t} \hat{\phi}.
\]

(A.35)

The vector of matrices \( \mathcal{R} \) plays the same role as the ratios of Clebsch–Gordan coefficients played in the dark-state polariton formed from a circularly polarized control field described in [23, 24]. The quantity \( \text{Tr}[\mathcal{R} \mathcal{R}^T] \) is a dyadic tensor accounting for the back action of the hyperfine spin-wave coherences on the propagating field, since the trace is over internal atomic degrees of freedom. Because \( \mathcal{R} \) contains the projection \((1 - \hat{\mathbf{k}} \hat{\mathbf{k}}^T)\) onto the subspace spanned by the polarization vectors, \( \text{Tr}[\mathcal{R} \mathcal{R}^T] \) has three spatial eigenvectors: one is \( \hat{\mathbf{k}} \), with zero eigenvalue, and the other two correspond to field polarizations labelled by \( \tilde{\psi}_j \) \((j = 1, 2)\). The component of the idler with polarization \( \tilde{\psi}_j \) obeys the propagation equation

\[
\left( \frac{\partial}{\partial t} + V_j \hat{\mathbf{k}} \cdot \mathbf{V} \right) \tilde{\psi}_j = 0,
\]

(A.36)

where \( \tilde{\psi}_j = \tilde{\psi}_j^* \cdot \tilde{\phi} \), and the group velocity

\[
V_j \equiv \frac{c}{|\Omega|^2 + n_0 \kappa|\Omega^2 + n_0 \kappa|^2 \text{Tr}[\mathcal{R}_j \mathcal{R}_j^T]},
\]

(A.37)

where \( \mathcal{R}_j \equiv \tilde{\psi}_j^* \cdot \mathcal{R} \). In this adiabatic limit, equation (A.32) suggests that the propagation dynamics is described by the dark-state polaritons

\[
\hat{\psi}_j(r_i, t) \equiv \frac{\Omega_j^0 \tilde{\psi}_j - \sqrt{n_0} \kappa \text{Tr}[\mathcal{R}_j \mathcal{R}_j^T]^{1/2}}{\sqrt{|\Omega|^2 + n_0 \kappa^2 \text{Tr}[\mathcal{R}_j \mathcal{R}_j^T]]}}
\]

(A.38)

which propagate through the ensemble with group velocities \( V_j \) and obey the quasi-bosonic commutation relations \(|\psi_j(r_i, t), \tilde{\psi}_j (r'_i, t)\rangle = \delta_{j,j'} \delta(r_i - r'_i) + O(1/\sqrt{N})\). The dark-state polariton for each eigen-polarization contains contributions from specific linear combinations of hyperfine coherences. Furthermore, since the group velocity may be different for each polarization, a pulse propagating through may experience walk-off with the two components propagating at different group velocities.

Under the phase-matching conditions \( |k_r + k_i - (k_{r_0} - k_i)|L \ll 1 \) where \( L \) is the spatial extent of the ensemble, one can express the spin waves imprinted in the write process in terms of the spin waves retrieved by the read process as

\[
\hat{S} = \int dr \sqrt{f(r_i)} \delta(r_i, t).
\]

(A.39)

where \( f(r_i) \equiv \tilde{n}(r_i)/\int dz \tilde{n}(r_i) \). The spatial distribution of the spin waves imprinted in the write process determines the idler mode annihilation operators \( \hat{\gamma}_j(t) \equiv \int dr \hat{\psi}_j(r_i, t) \). The read process, in the adiabatic limit, results in the transfer of the dark-state polariton modes to single modes in the detected idler fields \( \hat{\gamma}_j \rightarrow \hat{a}_j \), where \( \hat{a}_j \equiv \int dt \hat{\psi}_j^*(t) \hat{\psi}_j(t) \), where \( \hat{\psi}_j(t) \) is the annihilation operator for a photon with polarization \( \tilde{\psi}_j \) arriving at the idler detection apparatus at time \( t \).

From equation (A.19), one sees that the atomic density operator conditioned on the detection of a signal with polarization \( \tilde{\epsilon}_k \) is \( \hat{A}_{k} \cdot \hat{\rho} \hat{A}_{k}^* \). The retrieval efficiency of an idler photon with polarization \( \tilde{\psi}_j \) given the detection of a
signal of polarization $\lambda_i$ is therefore $\langle \hat{\Upsilon}_i(T) \hat{\Upsilon}_j(T) \rangle / (\hat{\Lambda}_i \hat{\Lambda}_j)$. Using the conditioned atomic density operator, this reduces to [17, 35]

$$\langle [\hat{\Upsilon}_i(T), \hat{\Lambda}_i] \rangle^2. \quad \text{(A.40)}$$

More generally, the detection of an idler photon with polarization $\hat{\epsilon}_i$, conditioned on the detection of a signal with polarization $\hat{\epsilon}_j$, is given by

$$\eta_{i,j}(T) = \frac{\int dt \langle \hat{\psi}_j(t) \hat{\psi}_i(t) \rangle}{\langle \hat{\Lambda}_j \hat{\Lambda}_j \rangle}, \quad \text{(A.41)}$$

where $\hat{\psi}_i(t) = \sum \hat{\epsilon}_i \cdot \hat{v}_i \hat{\psi}_j(t)$. The polariton propagation dynamics imply that

$$\eta_{i,j}(T) = \sum \langle \hat{\epsilon}_i \cdot \hat{v}_i \rangle ||[\hat{\Upsilon}_j(T), \hat{\Lambda}_j]||^2$$

$$+ 2 \Re \left\{ \langle \hat{\epsilon}_i,\hat{v}_i \rangle \langle \hat{\Upsilon}_i(T), \hat{\Lambda}_i \rangle^* \right\} \times [\hat{\Upsilon}_j(T), \hat{\Lambda}_j]$$

$$\text{(A.42)}$$

In the configuration we considered in this manuscript, the idler propagated in a direction parallel to the applied magnetic field, $\hat{k} = \hat{z}$. During the storage process, the stored hyperfine coherences $\hat{s}_{ad}(z, t)$ evolve under the influence of the magnetic field $B_2$ applied along the $z$-axis, precessing at the Landé frequency $\omega_{ad} = (\mu_B B_2 / h) [g_{s}(m_s + m_p) - \delta g m_p]$ where $g_s$ and $g_p$ are the Landé factors for levels $|a\rangle$ and $|b\rangle$, respectively, and $\delta g \equiv g_a / g_b$. The retrieval efficiencies (equation (A.42)) involve linear combinations of commutators $[\hat{S}_{s,a}(\hat{\Upsilon}(T), \hat{\Lambda}_i \hat{\Lambda}_i]$, each of which contains a phase factor $\exp(i \omega_{ad} T)$. These superpositions of terms oscillating at various frequencies lead to collapses and revivals of the retrieval efficiency with storage time [23, 24]. Additionally, however, imperfections in the field coils as well as residual ambient magnetic fields result in inhomogeneities in the magnetic field across the sample, which we model as a gradient in $B$ in the $z$-component of the field. In the presence of this gradient, the $\hat{s}_{ad}(z, T)$ constituents of $\hat{s}_{ad}(T)$ pick up a spatially varying phase factor $e^{-i M_m \cdot \hat{z} B z / B_0}$, which can also be viewed as a temporal change in wave number $\Delta k_m \equiv \omega_{ad} T B / B_0$

$$\hat{s}_{ad}(T) = e^{-i M_m \cdot \hat{z} B z / B_0} \int \sqrt{f(z)} e^{-i \Delta k_m \cdot z} \hat{s}_{ad}(z). \quad \text{(A.43)}$$

In evaluating the commutators, each spin wave contributes a term with a spatial Fourier transform factor $\int dz f(z) e^{-i \Delta k_m \cdot z}$. Evaluating the commutators appearing in equation (A.42), one finds

$$[\hat{\Upsilon}_i(T), \hat{\Lambda}_i] = \sum \langle \hat{\epsilon}_i \cdot \hat{v}_i \rangle \langle 0, z \rangle \cdot \hat{\psi}_i(T)$$

$$\times [\hat{\psi}_i(T), \hat{\Lambda}_i]$$

$$\text{X}_i \sqrt{\text{Tr} (\hat{P} \hat{P}^* \hat{P}_i)} \text{ (A.44)}$$

where $\hat{S}_{ad}(T)$ is determined by the time evolution of $\hat{s}_{ad}(T)$ as given in equation (A.43) and is given explicitly by

$$\hat{S}_{ad}(T) = e^{-i M_m \cdot \hat{z} B z / B_0} \int \sqrt{f(z)} e^{-i \Delta k_m \cdot z} \hat{s}_{ad}(z). \quad \text(A.45)$$

For a Gaussian density profile $f(z)$, this leads to a Gaussian decay $e^{-z^2 / \Delta B^2}$, with $\Delta B \equiv B_1$, where $l$ is the sample length.

A.1. Pure atomic alignment

In the first part of this appendix, we derived a general framework to describe the creation and retrieval of collective hyperfine excitations from an ensemble of alkali atoms. Now, we turn our attention to the special case in which the atomic populations are perfectly aligned, i.e. $p_m = p_{-m}$, that we described in the text. Here, the idler field propagates in the $z$-direction and the detected signal that heralds the creation of the stored atomic excitations is collected from the negative $z$-direction. By exploiting symmetry relations of Clebsch–Gordan coefficients, and by virtue of symmetric populations, horizontal and vertical idler fields couple to balanced superpositions of optical coherences, $\hat{e}_{m,\lambda}$ and $\hat{e}_{-m,\lambda}$, and hyperfine coherences $\hat{s}_{m,\lambda}$ and $\hat{s}_{-m,\lambda}$. For $^8\text{Rb}$, we then recover the Heisenberg–Langlevin equations (equation (14)). These equations suggest that when $p_1 = p_{-1}$, there are two dark-state polaritron operators corresponding to emission of an idler or either a horizontal or vertical polarization. This is consistent with the dark-state polariton operators obtained directly from the more general treatment: horizontal and vertical dark-state polaritons propagate at different velocities providing a group velocity walk-off.

To understand which optical and hyperfine coherences the linearly polarized signals couple to, we explicitly evaluate the equation of motion for the field. We begin by noting that since $\hat{k}_i = \hat{k}_f$, the Clebsch–Gordan coefficient matrices satisfy the relations $[p^{\lambda}_k]_{m,m} = -[p^{\lambda}_k]_{-m,-m}$. In the specific setup described in figure 2, and with $p_1 = p_{-1}$, we can take the trace of equation (A.24a) to find the equations of motion for $\varphi_{\lambda}$, $\varphi_{\lambda}^\dagger \cdot \hat{\varphi}_{\lambda}(\lambda \in \{H, V\}$, $\Sigma_{\lambda} = \hat{\Sigma}_{\lambda} \equiv (\xi_{m,\lambda} \geq \xi - \xi_{-1,\lambda}) / \sqrt{2},$ $\Sigma_{\lambda} = \hat{\Sigma}_{\lambda} \equiv (\xi_{m,\lambda} = \xi - \xi_{-1,\lambda}) / \sqrt{2},$

$$\frac{\partial \varphi_{\lambda}}{\partial t} + c \frac{\partial \varphi_{\lambda}}{\partial z} = \frac{i}{\hbar} [\hat{h}_1, \varphi_{\lambda}]$$

$$\text{(A.46)}$$

$$= \hat{h}_1 \Sigma_{\lambda} = \Sigma_{\lambda} \text{ (A.47)}$$

where $C_{\omega \lambda} = C_{\omega \lambda}^{*} F_{\lambda} = -1 / \sqrt{2}$ and $C_{\omega \lambda} = (-1)^{k,n} C_{\omega \lambda}^{*} F_{\lambda} = (-1)^{k,n} / \sqrt{2}$ are the relative coupling strengths of the idler field to optical coherences $\hat{e}_{m,\lambda}$. $\Omega$ is the real field Rabi frequency, $k$ is the single-photon Rabi frequency on the idler transition and $\xi_{0,\lambda}$ is a noise operator. The collective optical coherence operators are given by $\hat{e}_{m,\lambda} = (e^{i \omega_{\lambda} \xi_{0,\lambda}} - (-1)^{k,n} \hat{e}_{-m,\lambda}) / \sqrt{2}$. The horizontally polarized fields couple to optical coherences of the form $\hat{e}_{m,\lambda} = (\hat{e}_{m,\lambda} + \hat{e}_{-m,\lambda}) / \sqrt{2}$ and $\hat{e}_{m,\lambda} = (-i \hat{e}_{m,\lambda} - \hat{e}_{-m,\lambda}) / \sqrt{2}$. To see how these optical coherences evolve, we use equation (A.28b). Owing to the symmetries of Clebsch–Gordan coefficients between the levels $|e\rangle$ and $|a\rangle$, $[p^{\lambda}_k]_{m,m} = [p^{\lambda}_k]_{m,-m}$, and because $\mathcal{K} = \hat{\mathcal{K}} \cdot C_{\omega \lambda} = -i C_{\omega \lambda}^{*} + C_{\omega \lambda} \hat{e}_{m,\lambda} / \sqrt{2}$, the matrix
governing the coupling between these levels satisfies the symmetry relation \( K_{m_\lambda m_\lambda} = K_{-m_\lambda -m_\lambda} \). So the symmetric and antisymmetric combinations of optical coherences that respectively couple to the horizontal and vertical idler fields obey the equations of motion
\[
\left( \frac{\partial}{\partial t} + \Gamma \right) \hat{\xi}_{m_\lambda m_\lambda}^{(2)} = i \kappa \sqrt{n_{m_\lambda}} \hat{n}_{m_\lambda} \sum_{\mu=\pm 1} (\mu)^{m_\lambda \mu} \left[ C_{\mu \lambda}^{(2)} \right]_{m_\lambda m_\lambda} + i \Omega \sum_{m_\lambda \mu \neq m_\lambda \mu} F_{\mu \lambda} \sqrt{2} \left( \hat{\xi}_{m_\lambda m_\lambda} - (\mu)^{m_\lambda \mu} \hat{\xi}_{-m_\lambda -m_\lambda} \right) K_{m_\lambda m_\lambda}^{*}.
\]
(A.48)

Specifically, the coherences \( \hat{\xi}_{1,1,\lambda} \) for each polarization associated with atoms originating in the states \( |b, \pm 1 \rangle \) couple to linear combinations of the fast and slow coherences \( \xi_{b,1,1,0}^{(2)} \) and \( \hat{\xi}_{b,1,1,0}^{(2)} \). Similarly, for the atoms originating in the Zeeman state \( |b, 0 \rangle \), the collective coherence \( \hat{\xi}_{b,1,1,0} \) couples to the fast spin waves \( \xi_{b,0,0,0}^{(2)} \), but \( \xi_{b,0,0,0}^{(2)} \) has contributions from both \( \xi_{b,0,0,0}^{(2)} \) and the clock coherence \( \xi_{b,0,0,0} \). Only the horizontal fields couple to the clock coherence because the paths leading from \( |b, 0 \rangle \) to \( |a, 0 \rangle \) perfectly cancel when the idler is vertically polarized. For the coherences \( \hat{\xi}_{m_\lambda,1,\lambda} \) with \( m \in \{0, 1\} \) and \( \lambda \in \{H, V\} \), we have
\[
(\partial_t + \Gamma_e/2) \hat{\xi}_{m_\lambda,1,\lambda} = i \kappa \sqrt{n_{m_\lambda}} \hat{n}_{m_\lambda} \hat{\xi}_{m_\lambda,1,\lambda} + i \Omega C_{m_\lambda,1,\lambda} \hat{\xi}_{m_\lambda,1,\lambda} + \hat{\xi}_{m_\lambda,1,\lambda},
\]
(A.49)

where \( C_{0,1} = \sqrt{(3 + \delta_{0,1})/10} \) and \( C_{1,0} = \sqrt{3/10} \) determine the relative coupling of the optical coherences and read spin waves; \( \theta_{m_\lambda,1} \) is a noise operator. The read spin-wave operators are expressed in terms of the same fast and slow coherences imprinted in the write process, \( \hat{\xi}_{b,1,1,0} = (\hat{\xi}_{1,1,0}^{(2)} + \hat{\xi}_{1,1,0}^{(2)})/\sqrt{2} \), \( \hat{\xi}_{b,0,0,0} = \hat{\xi}_{b,0,0,0}^{(2)} \) and \( \hat{\xi}_{b,0,0,0} = \sqrt{3/2} \xi_{b,0,0,0}^{(2)} - i \xi_{b,0,0,0}^{(2)}/2 \).

From equation (A.28), we arrive at the equations of motion for the read spin waves
\[
\partial_t \hat{\xi}_{m_\lambda,1,\lambda} = i \kappa C_{m_\lambda,1,\lambda} \hat{\xi}_{m_\lambda,1,\lambda}.
\]
(A.50)

From these equations of motion for balanced populations, one can directly construct the dark-state polaritons associated with horizontally and vertically polarized idlers (equation (15)). To verify that \( \hat{x} \) and \( \hat{y} \) are indeed eigen-polarizations of the system associated with dark-state polaritons that propagate at distinct group velocities, we can examine the adiabatic diabatic matrices \( \mathcal{R}_\lambda = \mathbf{e}_\lambda^* \cdot \mathbf{R} \).

Explicitly, for \(^{87}\text{Rb} \) with \( F_s = F_c = 1 \) and \( F_a = 2 \), these matrices are given by
\[
\mathcal{R}_H = \begin{pmatrix}
0 & -i \sqrt{3} C_{0,H} / 2 C_{0,H} & 0 \\
-i C_{1,H} / 2 C_{1,H} & 0 & -i C_{1,H} / 2 C_{1,H} \\
0 & -i C_{0,H} / 2 C_{0,H} & 0 \\
i C_{1,H} / 2 C_{1,H} & 0 & -i C_{1,H} / 2 C_{1,H} \\
0 & -i \sqrt{3} C_{0,H} / 2 C_{0,H} & 0
\end{pmatrix},
\]
(A.51)

where these matrices are arranged such that the index \( m_b \) increases from \(-1\) on the left to \( 1 \) on the right, and \( m_b \) increases from \(-2\) on the top row to \( 2 \) on the bottom row. In general, in a sample with both polarization and alignment, the spatial eigenvectors associated with each dark-state polaritons are the eigenvectors of the dyadic tensor
\[
\mathbf{T}_{[\mathbf{R}_H \mathbf{R}_V]^{\dagger}} = \mathbf{T}_{[\mathbf{R}_H \mathbf{R}_H]^{\dagger}} \mathbf{i} \hat{x} + \mathbf{T}_{[\mathbf{R}_V \mathbf{R}_V]^{\dagger}} \mathbf{i} \hat{y} + \mathbf{T}_{[\mathbf{R}_V \mathbf{R}_V]^{\dagger}} \mathbf{i} \hat{x},
\]
(A.53)

where in terms of the atomic populations, the various tensor components are
\[
\mathbf{T}_{[\mathbf{R}_H \mathbf{R}_H]^{\dagger}} = p_0 \left| C_{0,H} \right|^2 + p_1 + p_2 \left| C_{1,H} \right|^2 / 2 C_{1,H}^2
\]
(A.54)
\[
\mathbf{T}_{[\mathbf{R}_H \mathbf{R}_V]^{\dagger}} = (1 - p_1 - p_2) \left| C_{1,H} \right|^2 / 2 C_{1,H}^2
\]
(A.55)
\[
\mathbf{T}_{[\mathbf{R}_V \mathbf{R}_H]^{\dagger}} = - (p_1 - p_2) \left| C_{1,H} \right|^2 / 2 C_{1,H}^2
\]
(A.56)
\[
\mathbf{T}_{[\mathbf{R}_V \mathbf{R}_V]^{\dagger}} = p_0 \left| C_{0,V} \right|^2 + p_1 + p_2 \left| C_{1,V} \right|^2 / 2 C_{1,V}^2.
\]
(A.57)

When the populations are equal, both the tensor components \( \mathbf{i} \hat{x} \) and \( \mathbf{i} \hat{y} \) are identically zero, and the spatial eigenvectors of the dark-state polaritons correspond to horizontal and vertical polarizations.

Because the idler polariton polarizations are horizontal and vertical when the populations are balanced, we chose to investigate the efficiency with which a horizontally or vertically polarized idler is retrieved given the detection of the horizontally or vertically polarized signal. Applying the treatment of appendix A to \(^{87}\text{Rb} \) with balanced populations, we obtain the expressions for the written spin waves given in section 3.

References

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