Highly efficient optical memory with long coherence time in cold atoms: supplementary material

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1. THE EFFECT OF FOUR-WAVE-MIXING

High-fidelity storage of quantum states requires the memory mechanism to be noiseless as well as high-efficiency. Any gain process that is intrinsic to the memory will add noise to the system and therefore degrade the quantum fidelity.

To estimate the effect of four-wave-mixing (4WM) in the gradient echo memory, we numerically solve the equations of motion for the system. The use of a bias magnetic field lifts the degeneracy of the Zeeman sub-levels. Using σ^+ polarised light for both the coupling and probe fields allows us to consider only the $|g\rangle \equiv |F=1, m_F=1\rangle$, $|s\rangle \equiv |F=2, m_F=1\rangle$ and $|e\rangle \equiv |F'=2, m_F=2\rangle$ energy levels, as illustrated in Fig. S1 (a). We take the following definitions: $\hat{\sigma}_{sg}$ is the collective operator for the coherence between the levels $|s\rangle$ and $|g\rangle$, D is the optical depth, η is the gradient of detuning due to the applied magnetic field, Γ is the population decay rate of the excited state, N is the number of atoms and γ is the ground-state decay rate. Assuming that the detuning is large relative to the fastest experimental timescale and that the population resides almost entirely in the $|g\rangle$ state, we adiabatically eliminate the excited state and write the Maxwell-Bloch equations [1, 2] as:

$$\begin{aligned} i\frac{\partial \hat{S}}{\partial \tau} &= (\eta(\tau)z + i\gamma') \hat{S} - \Gamma \sqrt{\frac{D}{2}} \left(\frac{\Omega_c^*}{2\bar{\Delta}_s} \hat{a}_s + \frac{\Omega_c^*}{2\bar{\Delta}_i} \hat{a}_i^\dagger \right), \\ -i\frac{\partial \hat{a}_s}{\partial z} &= \frac{D}{4} \frac{\Gamma}{\bar{\Delta}_s} \hat{a}_s + \sqrt{\frac{D}{2}} \frac{\Omega_c}{2\bar{\Delta}_s} \hat{S}, \\ i\frac{\partial \hat{a}_i^\dagger}{\partial z} &= \frac{D}{4} \frac{\Gamma}{\bar{\Delta}_i} \hat{a}_i^\dagger + \sqrt{\frac{D}{2}} \frac{\Omega_c}{2\bar{\Delta}_i} \hat{S}, \end{aligned}$$

where

$$\begin{aligned} \tau &= t - z/c, \quad \hat{S} = \sqrt{N} \hat{\sigma}_{sg}, \\ \hat{a}_{s(i)} &= \sqrt{\frac{2c}{\Gamma L}} e^{i\omega_0(t-z/c)} \int_{\omega} \hat{a}_{s(i)}(\omega) e^{i\omega z/c}, \\ \bar{\Delta}_{s(i)} &= \frac{i\Gamma/2 + \Delta_{s(i)}}{(\Gamma/2)^2 + \Delta_{s(i)}^2} \text{ and} \\ \gamma' &= \gamma + \frac{\Gamma}{2} \left(\frac{|\Omega_c|^2/4}{(\Gamma/2)^2 + \Delta_s^2} + \frac{|\Omega_c|^2/4}{(\Gamma/2)^2 + \Delta_i^2} \right). \end{aligned}$$

The numerical simulations are done using the software XMDS2 [3] with the parameters $D = 488$, $\Delta_s = 2\pi \times 325$ MHz, $\Gamma = 2\pi \times 5.75$ MHz, $\Delta_i = 2\pi \times 6.51$ GHz, $\Omega_c = 2\pi \times 7.06$ MHz, $\eta_{write} = 2\pi \times 1.24$ MHz, $\eta_{read} = 2\pi \times 1.32$ MHz and $\gamma = 500$ Hz. The values for optical depth, one-photon detuning, and magnetic field gradients were all measured directly under experimental conditions that corresponded to the highest efficiency recall that we obtained. The value for the coupling field Rabi frequency was indirectly measured using the ac-Stark shift of the Raman lines. The only fit parameter was the spin-wave population decay of 500 Hz, too small to be directly measured.

Figure S1 (b) shows the results of the numerical simulation (dashed red line), shown along with the experimental data (solid red line). The numerical simulation is in close agreement with the experiment, although the expected efficiency is 89% as opposed to a measured efficiency of 87%. The predicted idler field (dashed green line), shown magnified by a factor of 100 for clarity, is an indication of the magnitude of the 4WM gain in the system. We find that the total energy contained in the idler is expected to be 0.9% of the total input energy, with a gain in the output of 0.9%.

Fig. S1. (a) Energy level diagram. (b) Numerical results (solid lines) compared to experimental data (filled regions). The expected output is shown in red. The difference between including and excluding the four-wave-mixing in the model is shown by the thickness of the line. The green line is the expected idler field, magnified by a factor of 100.

2. DECOHERENCE DUE TO CONTROL FIELD AND THE MAGNETIC FIELD GRADIENT

In our experiments, we have observed that the memory lifetime depends on the presences of the control field (Ω_c) and the magnetic field gradient (η) during the storage duration. Figure S2 shows this dependency. We compare four possible cases: (1) Ω_c off and η off, (2) Ω_c off and η on, (3) Ω_c on and η off, (4) Ω_c on and η on. The best result was obtained when both Ω_c and η are not present and the coherence time is limited by atomic motions with $\tau_g = 416 \mu s$. With the control field on (Case (3) and (4)), it is clear that the coherence time is reduced. For Case (3), the decay time is limited by the exponential characteristic time $\tau_e = 101 \mu s$. For Case (4), we observe faster decay which is limited by the Gaussian characteristic time $\tau_g = 32 \mu s$. Case (2) also shows reduced characteristic time $\tau_g = 147 \mu s$ than Case (1). Therefore, we can conclude both Ω_c and η contribute the memory lifetime.

From these experimental observations, it is obvious both Ω_c and η should be turned off during the storage time in order to extend the memory lifetime. However, understanding the decoherence mechanism in the presence of Ω_c and η is also important since the decay time both with Ω_c and η is a de facto characteristic time for multiple temporal mode storage. While the effect of control field is clear (the control field induced scattering), the effect of gradient magnetic field is less obvious and requires some modelling of the magnetic fields present in our experiment.

Fig. S2. Comparison of efficiency decay due to the control field and the magnetic field gradient: (1) $\Omega_c \rightarrow$ off, $\eta \rightarrow$ off, (2) $\Omega_c \rightarrow$ off, $\eta \rightarrow$ on, (3) $\Omega_c \rightarrow$ on, $\eta \rightarrow$ off, (4) $\Omega_c \rightarrow$ on, $\eta \rightarrow$ on. The solid lines are empirical fitting curves with Gaussian and exponential decay functions $\mathcal{E}(t) = \mathcal{E}_0 e^{-t/\tau_e} e^{-(t/\tau_g)^2}$. The angle between the probe and control in these experiments limited the maximum possible lifetime.

A. Efficiency decay model due to asymmetric magnetic field gradients

Once the probe pulse is coherently absorbed by the atoms, the collective atomic state can be written as

$$|\psi(t)\rangle = \frac{1}{\sqrt{N}} \sum_j \exp \left[i \int_0^t \delta_{in}^{(j)} dt' + i \int_{t_1}^t \delta_{out}^{(j)} dt' + i \Delta \vec{k} \cdot \vec{r}_j \right] |g_1 \dots g_N\rangle,$$

where $\delta_{in}^{(j)}$ ($\delta_{out}^{(j)}$) is to the energy shift due to the input (output) magnetic field gradient and t_1 refers to the magnetic field gradient switching time. Rephasing occurs when

$$\int_0^{t_1} \delta_{in}^{(j)} dt' + \int_{t_1}^t \delta_{out}^{(j)} dt' = 0.$$

Thus, for an ideally symmetric magnetic field gradient (i.e. $\delta_{in}^{(j)} = -\delta_{out}^{(j)}$), recall occurs when $t = 2t_1$. We consider the Zeeman shifts produced by the GEM by expanding the input and output magnetic fields up to quadratic order in z , giving spatially dependent detunings of the form $\delta_{in}(z) = \delta_{in} + \eta_{in}^{(1)} z + \eta_{in}^{(2)} z^2$ and $\delta_{out}(z) = \delta_{out} + \eta_{out}^{(1)} z + \eta_{out}^{(2)} z^2$. The GEM coils are used to generate both the offsets ($\delta_{in,out}$) and gradients by driving the two coils with different currents of the same polarity. The sign of first order gradient $\eta^{(1)}$ gradient is reversed by swapping the values of current through the coils, i.e. $\eta_{in}^{(1)} = -\eta_{out}^{(1)}$. Considering the GEM coils geometry and the ratio between currents through the coils used in the experiment, we found that the sign of second order gradient is not swapped i.e. $\eta_{in}^{(2)} = \eta_{out}^{(2)}$.

The memory efficiency can be estimated by calculating the overlap between the collective spin states, that is given as

$$\begin{aligned} |\langle \psi(0) | \psi(t) \rangle|^2 &= \left| \frac{1}{N} \sum_j \exp \left[i \eta^{(2)} z_j^2 t \right] \right|^2 \\ &= \left| \int_{-L/2}^{L/2} dz \sqrt{f(z)} \exp \left[i \eta^{(2)} z^2 t \right] \right|^2, \end{aligned}$$

where $f(z) \equiv \sigma_{gs}(z) / \int dz \sigma_{gs}(z)$ is the spin wave distribution function, and we used $\eta_{in}^{(1)} = -\eta_{out}^{(1)}$, $\eta^{(2)} \equiv \eta_{in}^{(2)} = \eta_{out}^{(2)}$ and $t = 2t_1$. For a Gaussian input pulse, $f(z) \propto \exp(-z^2/2\sigma^2)$. Then, the model yields the efficiency decay

$$\mathcal{E}(t) = \frac{\mathcal{E}_0 |\text{erf}[\sqrt{1-i\zeta t/(4\chi)}]|^2}{\sqrt{\zeta^2 t^2 + 1}} \exp(-\Gamma_{sc} t),$$

where $\zeta \equiv 4\eta^{(2)}\sigma^2$, $\chi \equiv \sigma/L$, the initial efficiency is normalised by $\mathcal{E}_0$, and the control field induced scattering is taken into account as well.

This fitting model shows good agreement with the measured data shown in Fig. 3 of the main text with $\chi = 0.3, 0.35, 0.4, 0.5$ and $\zeta/2\pi = 12, 21, 36, 82$ kHz for $\Omega_c/2\pi = 5.19, 6.14, 7.06, 9.50$ MHz, respectively, where the control field induced scattering rates Γ_{sc} are fixed with the calculated values.

3. CONDITIONS USED FOR QUANTUM STORAGE DATASETS

In order to collect the large data ensembles required for generating Wigner functions and the carrying out the T - V analysis, the conditions for the experiment had to be modified. The detuning of the Raman transition that gave the best efficiency was 325 MHz. For short periods the laser was stable enough that we could set this frequency and collect data showing high efficiency and long storage times, as presented in sections 5 and 6 of the main text. For the hours of data collection required for the quantum measurements, however, we had to lock the laser frequency to maintain experimental stability, otherwise the variability of the data swamped the quantum noise.

Using an acousto-optic modulator (AOM) to provide a frequency offset, we could lock the Raman detuning. The frequency range of the AOM was restricted, however, and this reduced the maximum achievable Raman detuning. The non-optimal detuning resulted in a reduction of the storage efficiency in the quantum data. In one set of data we used 160 MHz, and in another we drove the AOM harder and pushed this out to 200 MHz.

The total effective detection efficiency of the pulses released from the memory is important for the T - V parameterisation. The net detection efficiency was calculated from the transmission of the spatial filter (90%) the detector quantum efficiency (98%), the homodyne fringe visibility (95%) and the quantum noise penalty [4] inherent in heterodyne detection systems (50%). Limited separation between the shot-noise and the dark-noise of the detection system adds noise to the measurements that will skew T - V data. This can be treated as an effective amount of additional loss in our analysis. Under most circumstances, the dark-noise clearance can be improved simply by increasing the amount of local oscillator power in the heterodyne system. In our case this was challenging as the local oscillator was created by filtering out one sideband of the modulation created by fibre-coupled electro-optic modulator [5]. The power throughput of this modulator was limited and simply increasing the power leads to long-term performance degradation of the modulator. For some of the data (125 μ s storage) we did over-drive this modulator to improve the dark noise performance of the system, but we could not operate in this manner for all measurements.

In table S1 we detail conditions used when taking our quantum data.

$\langle n \rangle$	t (μ s)	Δ (MHz)	η_m (%)	DN (dB)	η_d (%)	m
0.02	10	160	73	2.5	17	2000
0.25	125	200	53	4.0	24	4000
1.4	10	160	73	2.5	17	2000
20	10	160	73	2.5	17	2000

Table S1. Quantum data collection parameters. $\langle n \rangle$ = mean photon number per pulse; t = storage time; Δ = Raman detuning; η_m = memory efficiency; DN = shot-noise to dark-noise separation; η_d = total effective detection efficiency (including the effect of the dark-noise) and m = number of pulses in the measurement ensemble.

4. MULTIMODE STORAGE

We verified the multimode operation of the memory by storing and recalling 20 pulses, as shown in Fig. S3 (a). A lower storage efficiency is evident from the leakage of the input through the memory. This is the result of using a weaker control field for storage to reduce loss due to control field scattering. The magnitude of the gradient was also increased so that shorter input pulses could be stored. This further reduces the effective optical depth of the Raman transition and lowers the absorption efficiency.

Fig. S3. Storage and recall of 20 pulses in a first-in-last-out configuration. (a) The input pulses (red) and output from the memory (blue). (b) The efficiency of each of the retrieved pulses (red dots). The solid line is a decay model fit that contains exponential and Gaussian decay terms with time constants of 186 μ s and 106 μ s, respectively.

Figure S3 (b) shows the efficiency of each recalled pulse. The

pulses are recalled in a first-in-last-out (FILO) ordering that results in uneven losses across the pulse train, varying from 26% to 5% with an overall efficiency of 14%. The solid line in Fig. S3 (b) shows a decay curve fit that includes both exponential and Gaussian decays with $1/e$ time constants of 186 μs and 106 μs , respectively.

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