Storing short single-photon-level optical pulses in Bose–Einstein condensates for high-performance quantum memory

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Abstract

Large-scale quantum networks require quantum memories featuring long-lived storage of non-classical light together with efficient, high-speed and reliable operation. The concurrent realization of these features is challenging due to inherent limitations of matter platforms and light–matter interaction protocols. Here, we propose an approach to overcome this obstacle, based on the implementation of the Autler–Townes-splitting (ATS) quantum-memory protocol on Bose–Einstein condensate (BEC) platform. We demonstrate a proof-of-principle of this approach by storing short pulses of single-photon-level light as a collective spin-excitation in a rubidium-BEC. For 20 ns long-pulses, we achieve an ultra-low-noise memory with an efficiency of 30% and lifetime of 15 μs. The non-adiabatic character of the ATS protocol (leading to high-speed and low-noise operation) in combination with the intrinsically large atomic densities and ultra-low temperatures of the BEC platform (offering highly efficient and long-lived storage) opens up a new avenue toward high-performance quantum memories.

1. Introduction

Atomic systems are prime candidates for long-lived storage and on-demand retrieval of optical quantum states, due to the long coherence times of their optically accessible spin-states [1, 2]. Several spin-based memory approaches have been proposed and experimentally studied in a wide range of atomic media [2], including warm [3] and cold atomic gases [4], rare-earth–ion doped solids [5], and single atoms in optical cavities [6, 7] relying on various storage protocols, such as electromagnetically-induced-transparency (EIT) [8], off-resonant Raman [9, 10], and photon-echo [11–13] techniques. To date, no combination of a platform and a protocol has been agreed upon as the ideal practical memory that concurrently features long lifetime [14, 15], efficient [16–18], fast [19, 20] and reliable operation [21–24], although these features have been demonstrated either individually or in pairs. Here, we introduce the Autler–Townes splitting (ATS) quantum memory protocol [25] on a Bose–Einstein condensate (BEC) platform as a path toward overcoming this obstacle.

BECs of alkali atoms were among the first-proposed light-storage platforms [26, 27], since a BEC’s ultralow temperature inhibits thermal diffusion and thereby offers long-term storage [28]. In addition, a BEC’s large atomic density allows for strong light–matter coupling without optical cavities or particularly large atom numbers, leading to high-efficiency and high-speed quantum memory. Despite these intrinsic advantages, there have been only a few experimental studies exploring BECs for quantum memory. Early experiments focused on the long-lived storage and coherent manipulation of optical information in the classical domain [26, 28, 29], while more recent demonstrations tested the quantum nature of these
processes [30, 31]. All of these experiments used the EIT memory protocol [1–3, 5, 8, 14, 15, 18, 23, 24], which is favorable for efficient storage of long light-pulses but not well-suited to the short-pulse/large bandwidth storage regime due to this protocol’s adiabatic nature [10, 32]. Moreover, the large optical densities and control-field powers required for a broadband EIT memory increase the impact of photonic noise processes, making reliable operation in the quantum regime difficult [33–36].

The ATS memory scheme [25] overcomes these protocol-related limitations. In contrast to the EIT scheme, the non-adiabatic (fast) character of this method allows for optimal storage of short light-pulses with substantially reduced technical demand and complexity [32], and exceptional robustness to many noise processes [36]. In this article, we present a proof-of-concept experimental implementation of the ATS protocol with a BEC to explore the unique advantages of this protocol-platform combination for a high-performance quantum memory. We demonstrate efficient and ultra-low-noise storage of single-photon-level light pulses that are one-to-two orders of magnitude shorter than those reported in EIT based BEC-memories. We also show that ATS based-storage in a BEC platform significantly outperforms its implementations in laser-cooled atoms.

2. Experimental demonstrations

In our experiments, a BEC of $^{87}$Rb atoms is prepared using standard laser- and evaporative-cooling techniques [37], with the resulting ultracold atoms held in an optical dipole trap (ODT) (see figures 1(A) and (B), and appendix A for details). By reducing the depth of the ODT, evaporative cooling drives the temperature of the atoms below the critical temperature $T_c \approx 0.5 \, \mu$K at which Bose–Einstein condensation begins (figure 1(B)). The fraction of condensed atoms ($F_{\text{BEC}}$) increases with further cooling, resulting in nearly pure BECs at $F_{\text{BEC}} \approx 0.8$ and $T = 280 \, nK$ with the atom number of $N \approx 10^5$ and characteristic spatial extent (Thomas–Fermi diameter) of $R_{TF} \approx 10 \, \mu m$. Using the trap depth as a control, we study memory operation above and below the transition temperature.

The ATS protocol is implemented using a Δ-type three-level configuration within the ‘D2’ transition of Rb atoms by addressing an excited level ($|F = 2\rangle \equiv |e\rangle$) and two ground hyperfine levels ($|F = 1\rangle \equiv |g\rangle$ and $|F = 2\rangle \equiv |s\rangle$), as shown in figure 1(C). In the storage (writing) stage, optical coherence from a weak ‘probe’ pulse (resonant with $|g\rangle \rightarrow |e\rangle$) is transferred into collective excitations between the ground levels (spin-wave mode) via a strong ‘control’ field (coupled to the $|s\rangle \rightarrow |e\rangle$) with a pulse area of $2\pi$. By reapplying the control pulse after an adjustable storage time (read-out stage), the coherence from the spin-wave is mapped back to the optical mode, resulting in reemission as an output probe, as demonstrated in figure 1(D).

In our demonstrations, we use single-photon-level probe pulses with $\tau_p = 20 \, ns$ duration (at full-width-half maximum of their Gaussian temporal profile), which is shorter than the natural lifetime of the ground-to-excited-level coherence ($\tau_{eg} = 1/\gamma_{eg} = 54 \, ns$) of the Rb D2 line. Limited by our setup’s focusing ability, the probe beam diameter (at $1/e^2$) is reduced to $R_p \approx 25 \, \mu m$, which is still larger than the diameter of the atomic cloud $R_a \approx 10 \, \mu m$ for the lowest temperatures. To alleviate this size mismatch, we release the atomic cloud from the trap and allow 3.5 ms of free expansion before the storage-and-recall process, resulting in a cloud diameter comparable to that of the probe. The storage-and-recall process is then achieved using the write and read-out control fields with the same temporal profile of the probe pulses, but in a spatial mode oriented by $\theta = 110^\circ$ away from the probe beam, as depicted in figure 1(A). The retrieved probe signal is detected via time-resolved photon-counting measurements using a single-photon detector (SPD) and a time-to-digital converter (TDC), allowing the evaluation of memory performance by recording detection-vs-time histograms, as detailed in appendix B.

We characterize the performance of the ATS–BEC memory at various temperatures, ranging from above the condensation temperature (with $F_{\text{BEC}} = 0$) to well below the transition to BEC (where $F_{\text{BEC}} \rightarrow 1$), as shown in figure 2. First, memory efficiency $\eta_m$ is measured at temperatures between 1.5 $\mu$K to 280 $nK$ (corresponding to thermal and nearly pure-BEC clouds, respectively) for an average input-probe-photon number of $n_{in} = 1$ and storage time of $t_s = 200 \, ns$. We observe that efficiency increases as the temperature is reduced (figure 2(A)), due to a significant increase in peak atomic density associated with BEC (figure 2(B)). When the BEC fraction is $F_{\text{BEC}} \approx 15\%$ at $T \approx 340 \, nK$, the efficiency reaches its maximum $\eta_m = (30.2 \pm 1.5)\%$. However, further evaporation leads to a reduction in efficiency with $\eta_m = (13.0 \pm 0.9)\%$ for the nearly pure BEC at $T \approx 280 \, nK$ despite an additional increase in the peak density. We attribute the loss of efficiency to the limited ability to focus the probe beam onto a sufficiently small area at the center of the BEC, where the atomic density is largest. In the limits of $R_p \gg R_a$ or $R_p \sim R_a$ as in our demonstrations, the efficiency does not entirely follow the variation in peak atomic density (unfilled circles, figure 2(B)). Instead, it is either partly or fully governed by the atom number, which inevitably decreases during the evaporative cooling (unfilled squares, figure 2(B)). To verify this conjecture,
we repeat the efficiency measurements with larger beam size $R_p \approx 65 \mu m$ (grey squares, figure 2(A)) and find that memory efficiency decreases monotonically with temperature over the entire range and does, indeed, track the variation in atom-number. This also shows that the size mismatch between the probe and BEC is the primary limitation to reaching high efficiencies in our setup: the free expansion alleviates this mismatch at the expense of an overall reduction in the atomic density and hence efficiency.

Next, we investigate the variation of memory lifetime $\tau_m$ with respect to temperature $T$ in both the thermal and BEC regimes. We measure the efficiency of ATS memory as a function of storage time (from $t_s = 2$ to $10 \mu s$) at three different temperatures between $T = 280$ nK and $6200$ nK and determine the lifetime (defined as storage time for which efficiency decreases to $1/e$ of its original value) for each $T$. As the temperature is lowered toward the BEC regime, memory lifetime increases significantly and reaches a maximum of $\tau_m = 15 \mu s$ at $T = 280$ nK for the nearly pure BEC, as shown in figure 2(E). We attribute this observation to the combined effect of two spin-decoherence mechanisms: thermally induced atomic-diffusion (given by cloud temperature), and magnetic dephasing (due to uncancelled ambient magnetic fields). Memory lifetime is mainly limited by thermal decoherence at relatively high temperatures ($T > 3$ $\mu K$) in the thermal regime, while magnetic decoherence dominates in the BEC regime, where thermal motion is suppressed. Since thermal decoherence also increases with probe-control separation angle $\theta$, inhibiting thermal diffusion in the BEC regime provides a flexibility to use large angles ($\theta > 2^\circ$, as in our demonstrations), where low-noise operation can be realized.

To demonstrate simultaneous low-noise and broadband operation, we implement ATS memory in the nearly-pure BEC for several mean input-photon-numbers less than unity (figure 2(C) shows measurements for $\bar{n}_m \approx 0.2$). We determine the signal-to-noise ratio $\text{SNR} = (p_s - p_n)/p_n$ as a function of $\bar{n}_m$ after $t_s = 200$ ns, where $p_s$ and $p_n$ are independently measured detection probabilities for retrieved probe and noise. We measure a background-noise probability of $p_n = (6.6 \pm 1.5) \times 10^{-3}$ (inset of figure 2(C)),

![Figure 1](image-url)

**Figure 1.** Demonstration of ATS memory in BEC. (A) Schematic of experimental setup. AOM: acousto-optic modulator; NDF: neutral density filter; FC: fiber coupler; TDC: time-to-digital convertor; ODT: optical dipole trap. (B) Preparation of BEC, represented by velocity distributions for thermal, mixed, and condensed clouds (after 20 ms of free expansion) at different stages of ODT-evaporation with temperatures of $T$ and BEC fractions of $F_{\text{BEC}}$. (C) $\Lambda$-system on the D2 transition of $^{87}\text{Rb}$. (D) Storage and recall of 20 ns-long probe pulses at the single photon level with an input-mean photon number of $n_{\text{in}} = 1$ (lower panel) via write and read-out control pulses with Gaussian profiles (upper panel). The measured memory efficiency is 30% under the conditions of $T = 340$ nK and $F_{\text{BEC}} = 15\%$. 


yielding an SNR $\gtrsim 100$ in much of the low mean-photon-number regime ($\bar{n}_{\text{in}} < 1$), as illustrated in figure 2(D). The SNR can be as high as $42 \pm 9$, even for mean photon-numbers as small as $\bar{n}_{\text{in}} = 0.22$, which is typical in quantum photonics applications. This SNR would yield an error probability of $\mathcal{E} = 1/\text{SNR} = 0.023 \pm 0.005$, if quantum states were encoded into these photons. Our additional characterisations show that the observed residual noise comes from scattered light leaking from both ODT and control beams, which can be almost entirely eliminated with simple technical upgrades [38]. This also implies that there is no measurable noise contribution from any physical process linked to the memory operation, such as the FWM noise, demonstrating the reliability of ATS–BEC memory for short pulses at the single-photon level.

Finally, we examine the phase-preserving character of the photon storage process in our implementation (at near $T_c$) by controlling the phase evolution of the stored spin-wave. We apply a weak DC magnetic field to the ensemble such that $|g\rangle$ and $|s\rangle$ levels (forming a spin-wave coherence) are split into the Zeeman sublevels with energy/frequency differences proportional to the strength of the field [39]. In the writing stage, we then map optical coherence onto different classes of spin-waves among these Zeeman levels with the proper selection of the magnetic field orientation, and polarizations of probe and control-field (see supplementary material and References [40–42] for details). Since each class of spin-wave evolves with a different frequency, they acquire relative phase differences and thus interfere with one another, either constructively or destructively, resulting in the intensity of the recalled probe being modulated with storage time, as shown in figure 2(F). We achieve an interference visibility of $V = 62\%$ for small input photon-number $\bar{n}_{\text{in}} = 1$ (inset), while reaching up to $V = 80\%$ for large mean photon numbers due to a better magnetic-field stability with single-shot measurements. These results demonstrate the phase-preserving nature of our memory, which is a key requirement for quantum information storage.

3. Performance analyses

Looking beyond these proof-of-principle demonstrations, we find that the ATS–BEC approach is suitable for a high performance quantum memory, featuring the co-existence of highly efficient and long-lived storage together with broadband and low-noise operation. Particularly, the relaxed optical-depth demand of
Figure 3. Predicting ATS–BEC memory performance. All calculations use experimentally measured atom numbers, $T$, $F_{\text{BEC}}$, and ODT frequencies, and the solid lines between the data points are as guides to the eye. We assume that atoms remain trapped (no free-expansion) during storage and ATS memory is implemented in the backward recall scheme using the D1 transition of $^{87}\text{Rb}$. (A) Memory efficiency vs temperature, estimated for short storage times ($t_s \ll \tau_m$), memory bandwidth of $B = 170$ MHz ($\tau_p = 2.6$ ns), and probe-beam diameters $R_p = 1–25 \text{ \mu m}$. The dashed black line refers to peak optical density. (B) Bandwidth and optical depth vs temperature, estimated for an optimal ATS memory, where $d/2F = d \Gamma_{eg}/4\pi B \approx 4$. The horizontal dashed line indicates bandwidths and optical depths yielding efficiencies above 80%. (C) Memory lifetime vs temperature, for a small probe-control separation angle ($\theta$), based on the combination of decoherence effects due to thermal motion, recoil momentum and inelastic two-body collisions, as detailed in appendix D. The inset considers only the thermal memory lifetime $\tau_{\text{th}}$ (equation (6) in appendix D) for a wide range of temperatures and separation angles associated with different photonic noise levels. (D) Four-wave mixing (FWM) noise strength vs bandwidth, calculated for optimal ATS and EIT memories, as detailed in appendix C. The noise strength is normalized with respect to its minimum value corresponding to the smallest bandwidth of $10\Gamma_{eg}/2\pi$. The vertical dashed line indicates bandwidths yielding efficiencies above 85%.

the ATS protocol in conjunction with the ultra-large optical densities of the BEC platform renders near-unity efficiencies at GHz storage bandwidths. This performance can be achieved in our system by sampling the dense region of BEC with a probe-beam diameter that is significantly smaller than the Thomas–Fermi diameter ($R_p \ll R_s \approx R_{\text{TF}}$). Figure 3(A) shows the predicted memory efficiency with respect to temperature for pulses as short as $\tau_p = 2.6$ ns and probe-beam diameters smaller than used in our demonstrations (see appendix C for details). An effective optical depth of $d \approx 200$ is possible for $R_p = 1 \text{ \mu m}$ at $T = 280$ nK ($F_{\text{BEC}} \approx 0.8$), allowing a near-optimal memory efficiency of $\eta_m \geq 90\%$. We also predict the acceptance bandwidth and optical depth for an optimal ATS memory with respect to temperature, as shown in figure 3(B), confirming the feasibility of bandwidths approaching 200 MHz with large efficiencies of $\eta_m \geq 90\%$. The same performance via EIT or off-Resonant Raman memory would require an optical depth of about $d = 1000–1500$ [25, 32], which is hard to achieve even with typical BEC systems.

An ATS–BEC memory can also reach long lifetimes from milliseconds to a second by reducing the impact of the three major spin-decoherence mechanisms: magnetic dephasing, thermal diffusion, and internal/external dynamics of BEC. First, magnetic dephasing can be eliminated using well-mastered techniques, including a high-degree isolation from the static and time-dependent magnetic-field noise [31], spin-echo dephasing/rephasing schemes [43–45], and precise control over magnetic-insensitive Zeeman states [14, 46]. Second, thermal decoherence, which is an increasing function of both $T$ and $\theta$ [46], is already significantly reduced due to ultra-low temperatures in our evaporatively cooled system. Beyond that, as the thermal velocity is virtually zero in the nearly-pure BEC, BEC memory features much longer lifetimes than what is achievable with a purely thermal cloud, even at ultracold temperatures and small $\theta = 0.1\degree$, as shown in figure 3(C). Third, BEC-specific decoherence mechanisms are already effective only at long time scales over milliseconds, in part, because of the coherent matter-wave nature of BEC. Among these mechanisms, spatial decoherence (arising from atomic motions in the trap) can be coherently compensated using matter-wave interferometry techniques [27, 47]. However, decoherences due to recoil motion (an increasing function of $\theta$) and inelastic two-body collisions (proportional to atomic density) set an ultimate limit to memory lifetime [28, 31, 48]. Considering the combined effect of these two
mechanisms together with thermal diffusion, we predict that memory lifetime in our ultracold-Rb system can reach one hundred milliseconds (figure 3(C)).

Low-noise operation is another essential requirement that is difficult to satisfy simultaneously with long lifetimes and large bandwidths. Particularly, in the forward recall scheme (i.e. co-propagating input and retrieved probe, as in most quantum memory experiments) noise from control-leak and FWM processes can be minimized using a large-$$\theta$$ configuration [24], but this conflicts with the small-$$\theta$$ requirement for reduced thermal decoherence [46]. In comparison to laser-cooled systems ($$T \sim 10–100$$ $$\mu$$K), the small thermal-diffusion rates at ultracold temperatures ($$T \lesssim 1$$ $$\mu$$K) allow one to overcome the detriment of large-$$\theta$$ and thus provide a workable range of $$\theta \approx 4–7^\circ$$, where low-noise operation is possible while retaining a millisecond lifetime (inset of figure 3(C)). However, lifetimes of order one-hundred milliseconds still require a nearly-pure-BEC ensemble (at $$T \to 0$$) with a small $$\theta$$, which prevents decoherence due to recoil motion. On top of this, optimally efficient memory operation necessitates counter-propagating input and retrieved probe fields ("backward recall"), which cannot be implemented at arbitrarily large $$\theta$$ (see supplementary material for details). In this scenario ($$\tau_m \gg 10$$ ms, $$\theta < 0.2^\circ$$, and backward recall), low-noise operation favors small optical depths and control powers, contrasting with the high demand on these resources for a broadband memory [10, 32]. In particular, FWM noise is a significant issue, due to its exponential and quadratic dependencies on optical depth and control power, respectively (as detailed in appendix E) [33–36]. Though recent results with the ORCA protocol show excellent noise characteristics [50], the ns-storage times in this system are still quite short. When compared to adiabatic protocols like EIT and Raman, the ATS protocol offers the possibility to reduce the impact of FWM noise in the broadband regime and for spatially multimode configurations [51], due to its substantially lower requirements for these resources. Figure 3(D) compares the estimated relative strength of FWM noise vs bandwidth between the optimal implementations of the EIT and ATS protocols in our Rb-system. This prediction shows that the probability of FWM noise with ATS memory is four to five orders of magnitude less compared to that of EIT memory, for storing a few nanoseconds-long pulses at near-unity efficiencies.

4. Conclusion

In conclusion, we have experimentally demonstrated the non-adiabatic storage of single-photon-level light in a rubidium-87 BEC using the ATS protocol with a pulse duration that is 1–2 orders of magnitude shorter than those reported in previous BEC memories. Our proof-of-principle experiments and predictive analysis highlight the inherent advantages of the ATS protocol-BEC platform combination for a high-performance quantum memory, simultaneously featuring high efficiency, long lifetime, high-speed and low noise operation. In view of the recent technical progress with portable, miniaturized, and even space-based BEC experiments [52], we anticipate that this approach offers a feasible solution for large-scale ground and satellite-based quantum networks [53].

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Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

Appendix A. Experimental setup

The experimental setup (figure 1(A)) consists of ATS-memory components (including optical pulse generation and detection systems) and BEC-generation components. As part of the memory components, probe and control fields are extracted from two independent continuous-wave lasers and then temporally shaped into short pulses using acousto-optic modulators (AOMs). After attenuating the probe beam to the single-photon-level with neutral density filters (NDF) and setting the peak power of the control beam to 8 mW, both beams are coupled into single-mode optical fibers (FC), and decoupled back to free-space on a
separate bench where the BEC apparatus is located. Following polarization control with quarter-wave plates, the probe and control beam diameters are focused to a waist of 25 μm (or 65 μm) and 150 μm at the intersection of two crossed ODT beams (derived from a 1064 nm laser), using a telescope and single lens respectively (not displayed in the figure). After coupling into an optical fiber, the output probe pulses are detected using a SPD, and their arrival times are recorded on a time-to-digital converter (TDC), triggered by a function generator.

In the implementation of the ATS protocol, the temporal profile of the control pulses (writing and read-out) is shaped into a Gaussian envelope (the same as that of probe pulses) with a slightly longer duration than the probe (≈ 25 ns at FWHM). Their pulse area is set to be 2π by adjusting the Rabi frequency/power of the control-field, based on the spectral measurements of Autler–Townes split lines, as described in Ref [25]. An additional characterisation of memory efficiency vs pulse area has confirmed that the pulse area of 2π results in the maximum achievable efficiency in our implementation.

We also note that although the control and probe lasers are not phase-locked to each other in our experiments, the impact of the laser-phase noise on memory performance is negligibly small due to the fact that the time scale for the storage and recall (20 ns) is much shorter than that of the typical phase fluctuations of the lasers. Similarly, the long-term frequency drift of the lasers (locked to a Rb transition) is sub-MHz which is significantly smaller than the storage bandwidth (22 MHz), and hence does not have a significant impact in our memory demonstrations.

The ultracold atoms are prepared using standard laser cooling and trapping techniques, similar to reference [37]. The sequence of these techniques begins with the preparation of cold atoms in a magneto-optical trap followed by further sub-Doppler laser cooling to temperatures down to $T \approx 50 \mu K$. Next, the atoms are loaded into a quadrupole magnetic trap for RF-induced evaporative cooling that leads to $T < 10 \mu K$. Finally, these atoms are transferred into the ODT, which has a controllable trap-depth for further evaporative cooling, and with sufficient cooling, they reach the conditions of Bose–Einstein condensation, as detailed in supplementary material.

Appendix B. Measurements

In our demonstrations, the memory performance is assessed with time-resolved photon-counting measurements for the detection of both the input-probe photons and stored-and-recalled probe photons (figure 1(A)). Each measurement period is performed during a 1 ms-detection window that follows 20 s of BEC-preparation and 3.5 ms of free-expansion. In each period, a storage-and-recall event (defined by a (figure 1(A))). Each measurement period is performed during a 1 ms-detection window that follows 20 s of measurements for the detection of both the input-probe photons and stored-and-recalled probe photons respectively. By repeating this preparation-and-measurement cycle several times from $N_{\text{cyc}} = 10$ to 300, we acquire detection-vs-time histograms for a total number of storage-and-recall events between $N_\Lambda = N_{0}N_{\text{cyc}} = 10^{4}$ and $3 \times 10^{4}$, depending on average photon-number of the input probe ($\bar{n}_{in}$) varying from 3 to 0.

The calibration of input mean-photon number is achieved with direct photon-counting measurements in the absence of an atomic cloud, following the same procedure described in our previous studies [25, 36]. In this procedure, we prepare a sequence of input probe-pulses with a number of $N_\Lambda$ and measure the number of the total detections of $n_\Lambda$. By taking into account the SPD’s detection efficiency ($\approx 0.5$) and the transmission probability ($\approx 0.2$) between the memory and SPD (due to fiber-coupling loss etc), we determine mean photon number for memory input to be $\bar{n}_{in} = (n_\Lambda/N_\Lambda) \times 10$, where the factor of 10 represents the system’s total loss after memory. In the calibration, we ensure that the mean photon number of the detected photons is sufficiently low ($\bar{n}_{in} < 0.3$) such that multi-photon detection events (associated with the Poissonian statistics of laser-light) are insignificant and the operation of the SPD is far from the saturation regime.

Appendix C. Memory efficiency and bandwdith

Here, we detail the current experimental limitations and quantitative predictions for realizing both large memory efficiency and large acceptance bandwidth by exploiting both the favorable efficiency scaling of the ATS protocol and the large optical depth of a BEC. First, we look into the optical depth and bandwidth dependence of ATS-memory [25]. In the forward-recall configuration (as in our experiments), efficiency is given by

$$\eta_t \approx (d/2F)^2 e^{-d/2F} e^{-1/F}, \quad (1)$$

where $d$ is the peak optical depth, and $F = 2\pi B/\Gamma_{eg}$ is the ATS factor that depends on the bandwidth of the probe pulse ($B = 0.44/\tau_p$ for a Gaussian profile) and the natural linewidth of the optical transition.
(Γ_{eg} = 2\gamma_{eg}) under the condition that the pulse area of the control fields are set to be 2\pi. Equivalently, $F = 0.44\pi(\tau_{eg}/\tau_{p})$ relates the duration of the probe pulse ($\tau_{p}$) relative to coherence lifetime of the optical transition ($\tau_{eg} = 1/\gamma_{eg}$).

According to this expression, the theoretical maximum efficiency is only $\eta_{f} \approx 40\%$ for the probe bandwidth used in our demonstrations ($B = 3.7\Gamma_{eg}/2\pi$), which is, in part, due to the fundamental limitation imposed by the decay time of the ground-to-excited coherence (optical decoherence), accounted for by the term $e^{-1/F}$ in equation (1). Although our experimental maximum efficiency ($\eta_{\text{m}} = 30\%$) is already close to this theoretical maximum, and can even reach it with more optical depth, efficiencies above 40\% are only possible with larger bandwidths (shorter pulses), which reduce the impact of optical decoherence ($e^{-1/F} \rightarrow 1$). Further, even with pulses much broader than the transition linewidth ($B \gg \Gamma_{eg}/2\pi$, or equivalently $\tau_{p} \ll \tau_{eg}$, such that $e^{-1/F} \approx 1$), the theoretical efficiency cannot exceed $\eta_{f} \approx 54\%$ due to the unavoidable re-absorption of the retrieved probe pulses in the storage medium. This limitation can be circumvented by implementing backward recall, for example, using counter-propagating write and read-out control pulses [10, 25]. In this case, the efficiency of ATS memory is

$$\eta_{fb} \approx \left(1 - e^{-d/2F}\right)^2 e^{-1/F}. \tag{2}$$

This expression dictates that near-unity memory efficiencies $\eta \geq 90\%$ necessitate both sufficiently large bandwidths $B \geq 14\Gamma_{eg}/2\pi$ and optical depths $d \geq 90$ such that $d/2F \geq 4$, which also highlights the inherent broadband character of the ATS protocol. Given that large optical depths are readily available with the BEC platform, we can experimentally achieve such large efficiencies in the backward scheme simply by meeting the following technical requirements: (i) sampling the dense region of BEC with a probe-beam diameter that is smaller than the Thomas–Fermi diameter ($R_{p} \ll R_{TF}$), and (ii) reducing the probe pulse duration, which also requires using the D1 line in Rb due to the larger hyperfine splitting in the excited state manifold ($\approx 0.8$ GHz).

Second, to verify this conjecture, we predict the optical depth of our ultracold system based on its experimental characterisations (see supplementary material). The effective value of the optical depth depends on the spatial intensity profile of probe as well as the spatial-density profile of the atomic cloud, which is determined by $T$ and ODT parameters. This value can be numerically extracted from Beer’s absorption law

$$d = -\ln \left[\frac{\iint I_{\text{out}}(x, y) \, dx \, dy}{\iint I_{\text{in}}(x, y) \, dx \, dy}\right], \tag{3}$$

where $I_{\text{in}}(x, y)$ and $I_{\text{out}}(x, y)$ are the transverse intensity profiles of the input and transmitted probe, propagating along the z-direction. Assuming that the input probe beam is characterized by a Gaussian profile, $I_{\text{in}}(x, y)$ and $I_{\text{out}}(x, y)$ are then given by

$$I_{\text{in}}(x, y) = I_{0} \exp \left(-\frac{2x^2}{w_{x}^2}\right) \exp \left(-\frac{2y^2}{w_{y}^2}\right), \tag{4}$$

$$I_{\text{out}}(x, y) = I_{0} \exp \left[-\frac{3\lambda^2 \chi}{2\pi} \int_{0}^{L} \rho(x, y, z) \, dz\right], \tag{5}$$

where $\{R_{px}, R_{py}\} \equiv \{2w_{x}, 2w_{y}\}$ are the beam diameters along $\{x, y\}$ axes, $I_{0}$ is the peak intensity of the input probe, $\lambda$ is resonant wavelength, $\chi$ is the strength of the atomic transition including the degeneracy factor, $\rho(x, y, z)$ is the density distribution of the atomic cloud, involving thermal and BEC components, and $L$ is the effective length of the cloud along the probe-propagation direction. In this way, as presented in figure 3(B), we calculate $d$ for a given $R_{p}$ as well as $T$ and ODT frequencies, both of which determine the density distributions of BECs and thermal clouds (see supplementary information for further details). This calculation shows that large optical depths ($d \sim 200$) are already available in our BEC-system for small probe-beam diameters $R_{p} \sim 1$ µm. Using these $d$ values in equation (2), we predict high memory efficiencies exceeding 90\% for the storage of a few nanoseconds long pulses, as shown in figures 3(A) and (B). Finally, we note that small probe diameters near the diffraction limit can be obtained using confocal microscopy setups, which have been implemented in several applications such as trapping of single neutral atoms [54, 55].

**Appendix D. Memory lifetime**

In this section we detail the limitations of memory lifetime in our demonstrations, and show our predictive calculations for long-lived storage.
In our experiments, storage times are limited to the microseconds timescale due to the decoherence of collective spin excitations. We find that this decoherence is mainly governed by the combined effect of thermal diffusion and magnetic dephasing; the impact of the other decoherence mechanisms (including inelastic collisions [27] and recoil motion [27, 29, 31]) are predicted to be considerable only at much longer timescales (on the order of milliseconds). The detriment of the thermal diffusion is two-fold: the loss of the atoms due to their dispersive motion out of the interaction cross-section, and the loss of spatial coherence (initially set by the probe and control-wave vectors during writing). While the former is expected to be observable at millisecond and greater time scales, the latter is considerable at much shorter times due to the non-zero probe-control separation angle. In such a configuration, a spatially periodic phase pattern is imprinted as the stored spin-wave with a spatial period of \( \lambda_{\text{sw}} = 2\pi /|\kappa_{\text{sw}}| \), where \( \kappa_{\text{sw}} = k_p - k_c \) is imposed by conservation of momentum (phase-matching condition), involving the wavevectors of the probe (\( k_p \)) and control (\( k_c \)) beams [46]. Since this phase-grating can be partially or completely erased as a result of the atomic diffusion, thermal-memory lifetime \( (\tau_{\text{th}}) \) depends on \( \theta \) (determining the spatial period) and \( T \) (determining the diffusion rate), expressed by

\[
\tau_{\text{th}} = \frac{\lambda_{\text{sw}}}{2\pi v_{\text{th}}} \approx \frac{\lambda}{4\pi \sin(\theta/2)} \sqrt{\frac{m}{k_B T}},
\]

where \( v_{\text{th}} = \sqrt{k_B T/m} \) is the mean thermal speed, \( m \) is the mass of an atom, and \( k_B \) is the Boltzmann constant. Given that the difference between the wavelength (\( \lambda \)) of the probe and control fields is very small (\( |k_p| \approx |k_c| \)), \( \lambda_{\text{sw}} \) is nearly equal to \( \lambda/[2 \sin(\theta/2)] \).

In our experimental configuration with the large probe-control separation angle (\( \theta = 110^\circ \)), the impact of thermal diffusion becomes significant at relatively high temperatures (\( T > 3 \mu K \)) and hence dominates over magnetic dephasing in this regime. For example, we expect the thermal decay time constant to be \( \tau_{\text{th}} = 3.2 \mu s \) (equation (6)) at \( T = 6.2 \mu K \), which is close to the experimentally measured memory lifetime for the same temperature (4.5 \( \mu s \)). However, in the BEC regime where \( T < 0.5 \mu K \), observed memory lifetimes are significantly shorter than those predicted from thermal decoherence, indicating that magnetic dephasing is the dominant decoherence mechanism at ultralow temperatures.

Based on these characteristics, we describe the observed storage time (\( t_s \)) dependence of memory efficiency for a given cloud-temperature as,

\[
\eta_m(t_s) = \eta(t_{\text{th}}) e^{-\left(t_s-t_{\text{th}}\right)/\tau_{\text{mag}}} \left( F_{\text{BEC}} + F_{\text{th}} e^{-\left(t_s-t_{\text{th}}\right)^2/\tau_{\text{th}}^2} \right),
\]

where \( F_{\text{BEC}} \) and \( F_{\text{th}} = 1 - F_{\text{BEC}} \) are the fractions of BEC and thermal atoms in the cloud, respectively, and \( \eta(t_{\text{th}}) \) is memory efficiency measured for the shortest storage time, \( t_{\text{th}} \). In this simplified model, the Gaussian term with the characteristic decay time of \( \tau_{\text{th}} \) describes diffusion-induced decoherence (equation (6)) only for thermal atoms, as the BEC atoms are considered to be free from thermal motion. The common exponential factor with the decay-time constant of \( \tau_{\text{mag}} \) corresponds to decoherence due to magnetic dephasing of spin excitations across whole ensemble. We note that in typical cold-atom experiments with mm-size ensembles, magnetic dephasing is generally characterized by a Gaussian decay-function (with a time-constant exhibiting an inverse linear dependence on the length of the cloud), instead of the exponential decay term in equation (7). We attribute this difference to the fact that in our system, the spatial extent of our atomic ensembles is much smaller and hence exhibits more sensitivity to variations of ambient magnetic field on micrometer length scales. Moreover, as the atomic cloud falls from the ODT during the 1 ms storage-recall cycle in our single-photon-level measurements, it experiences additional position-dependent magnetic field variations across the free-fall distance, which is comparable to size of the cloud.

Using this decoherence model, we fit our results from the measurements of storage time vs efficiency to equation (7) (figure 2(E)), which shows a reasonable agreement with data taken at different cooling temperatures. In the fitting procedure, for a given \( T \), we fix the parameters of \( F_{\text{BEC}}, F_{\text{th}} \) (extracted from independent sets of temperature measurements) and \( \tau_{\text{th}} \) (calculated from equation (6) for \( \theta = 110^\circ \)) such that \( \tau_{\text{mag}} \) is a single free parameter that is evaluated from experimental data. We find that \( \tau_{\text{mag}} \) tends to be larger with lower cloud-temperatures because of the fact that the spatial extension of atomic clouds become smaller and thus less susceptible to the variations of ambient magnetic fields.

We also note that the extracted value of \( \tau_{\text{mag}} \) exhibits a variation on long timescales (over several days), which we attribute to changes in the conditions of ambient magnetic-field. For instance, we find that \( \tau_{\text{mag}} \) varies between 5 and 7 \( \mu s \) for a cloud at \( T = 340 \text{ nK} \), depending on day-to-day optimization of bias magnetic field. Figure 2(E) represents one of the best data sets, obtained after a careful bias-field optimization, yielding magnetic dephasing constant of \( \tau_{\text{mag}} = (7.0 \pm 2.5, 7.0 \pm 0.0, 16.5 \pm 2.8) \mu s \) with 1/e
memory-lifetime of $\tau_m = (4.5 \pm 2.5, 7.8 \pm 1.0, 15.8 \pm 2.8) \mu s$ for $T = (6200, 340, 280) \text{nK}$, respectively. In addition, we occasionally observe that memory efficiency drifts by about 10–30% of the typical $\eta_m$ (in part due to instability of the atom-number of prepared clouds in hours-long time scales), which seems to be more pronounced in measurements for short storage times ($t_s < 2 \mu s$) carried out for a BEC cloud.

Finally, we show the details of our predictions for the ultimately achievable memory lifetimes ($\tau_m$) in our current system (figure 3(C)). These predictions are based on our experimentally realised clouds (characterised by $T$, $F_{\text{BEC}}$ and $F_{\text{th}}$) at a probe-control separation angle $\theta$, under the assumption that certain decoherence effects such as magnetic dephasing, BEC-spatial decoherence, and inhomogeneous ac-Stark shifts due to the ODT are eliminated by technical means. In this case, we consider the impact of three major decoherence mechanisms: (i) thermal motion, (ii) recoil motion, and (iii) inelastic two-body collisions. The effects of thermal- and recoil-motion induced decoherence are separately described by Gaussian decays of memory efficiency with characteristic times of $\tau_{\text{th}}$ (equation (6)) and $\tau_{\text{rec}} = R_p \lambda m / [2h \sin(\theta/2)]$ [30, 31], respectively. While thermal-motion induced decoherence is not applicable to the BEC part of atomic cloud, the effect of recoil-motion is ignored for the thermal part (as being significantly dominated by thermal decoherence) in our regime of interest. Furthermore, decoherence due to inelastic two-body collisions in BEC is characterised by an exponential decay of memory efficiency with a decay time of $\tau_{\text{col}} = m / [4h I_m(a_w) \rho_B]$, where $I_m(a_w)$ is the imaginary part of scattering length for the two component Rb–BEC (in $|g\rangle$ and $|s\rangle$), and $\rho_B$ is the peak density of BEC [27, 28, 48]. As the atomic density in a thermal cloud is substantially smaller than a BEC, $\tau_{\text{col}}$ is considered to be negligibly small for the thermal portion of the cloud.

Under these conditions, the combined effect of the decoherence mechanisms on memory efficiency $\eta$ is described by

$$\eta_m(t_s) = \eta(0) \left[ F_{\text{BEC}} \left( e^{-t_s/\tau_{\text{th}}} \right) \left( e^{-t_s/\tau_{\text{rec}}} \right) + (1 - F_{\text{BEC}}) \left( e^{-t_s/\tau_{\text{col}}} \right) \right],$$

where $t_s$ is storage time, and $\eta(0)$ is memory efficiency for $t_s = 0$. The memory lifetime shown in figure 3(C) is defined as the characteristic time at which the memory efficiency drops to $(1/e)$ of $\eta(0)$.

We note that cold atoms in an optical lattice are an alternative memory platform to reach long storage lifetimes, since the atomic motion and collisions are, to a significant extent, eliminated in this system. On the other hand, the implementation of a highly efficient lattice-based memory is challenging, as this system has much smaller atomic density compared to a BEC. This limitation could be circumvented using an optical resonator (at the expense of an additional technical complexity [49]), which may constraint memory bandwidth.

**Appendix E. Four-Wave mixing noise**

FWM noise is one of the main limitations toward developing reliable broadband spin-wave memories. The probability of FWM noise depends on two general factors: (i) geometric relationships between the wavevectors of probe and control fields, and (ii) memory resources including optical depth and control-field power along with system specific parameters.

In general, the geometric factor is characterised by the angular separation $\theta$ between the probe and write control field. For the forward recall scheme, FWM noise is detrimental to memory for any $\theta < \theta_{\text{FWM}}$, where the phase-matching conditions for the FWM process are satisfied, as detailed in supplementary material. Here, $\theta_{\text{FWM}}$ is the threshold angle given by $2 \sin^{-1}(\sqrt{\lambda/[8\pi L])}$, for an effective medium length of $L$ along the propagation direction of the probe field. By choosing $\theta > \theta_{\text{FWM}}$, FWM noise can be significantly reduced, albeit at the expense of larger thermal-diffusion-induced decoherence and non-optimal spin-wave retrieval associated with forward recall.

The resource dependence of FWM is characterised by a ‘noise strength’ parameter, which is proportional to the probability of FWM noise corrupting the memory, as detailed in references [33, 34]. The FWM noise-strength is determined by optical depth $d$, peak Rabi frequency of the control field $\Omega_c$, and system-specific parameters (e.g. $\gamma_{eg}$ in a $\Lambda$-type three-level system) as in the following,

$$S_{\text{FWM}} \propto \Omega_c^4 \sinh \left( \frac{\zeta d \gamma_{eg}}{2 \Delta_{eg}} \right)^2,$$

where $h\Delta_{eg}$ is the energy difference between the ground levels of the $\Lambda$-system, and $\zeta = \Omega_c / \Omega'_c$ is the ratio of the Rabi frequency from $|s\rangle \rightarrow |e\rangle$ to the one from $|g\rangle \rightarrow |e\rangle$. Since the required $d$ and $\Omega_c$ for implementing an optimal broadband memory are proportional to the memory bandwidth ($B > \Gamma_{eg}/2\pi$) with certain proportionality constants specific to the memory protocol, FWM noise strength strongly depends on bandwidth and the employed protocol. Furthermore, backward recall scheme, which is required
for achieving optimal efficiency for any memory protocol, does not allow suppression of FWM noise and hence, the resource dependence of the employed protocol becomes the crucial factor in determining the impact of FWM noise. In view of this, the ATS protocol is advantageous due to its favorable resource scaling in the broadband operation regime, as compared to adiabatic memories, such as EIT and off-resonant Raman protocols. In figure 3(D), we make a comparison of FWM noise-strength ($\omega_{\text{FWM}}$) between the ATS and EIT protocols for implementation of an optimal broadband memory in our $^{87}$Rb system (featuring $\Delta_{a} / 2\pi = 6.83$ GHz and $\zeta \approx 1.33$). In this comparison, we calculate $\omega_{\text{FWM}}$ using equation (9) for a bandwidth range of $10(\Gamma_{eg} / 2\pi) < B < 40(\Gamma_{eg} / 2\pi)$, requiring optical depths of $d_{\text{ATS}} = 8 \times (2\pi B / \Gamma_{eg})$ and $d_{\text{EIT}} = 50 \times (2\pi B / \Gamma_{eg})$ as well as peak Rabi frequencies of $\Omega_{\text{ATS}} = 1.5 \times (2\pi B)$ and $\Omega_{\text{EIT}} = 4 \times (2\pi B)$ for optimal ATS and EIT memories, based on their non-adiabatic and adiabatic operation conditions, respectively (see reference [32] for details). These results show that in this bandwidth range, corresponding to probe pulse durations between 1.9–7.3 ns in our Rb system, the probability of FWM noise associated with an optimal ATS memory is 4–5 orders of magnitude smaller than that associated with an optimal EIT memory. Consequently, the ATS protocol offers a favorable option for the realization of long-lived broadband quantum memories featuring both high-speed and faithful operation.

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