

# Broadband Quantum Memory in Atomic Barium Vapor with 95% Storage Efficiency

Kai Shinbrough,<sup>1,2,\*</sup> Benjamin D. Hunt,<sup>1,2,†</sup> Sehyun Park,<sup>3,‡</sup> Kathleen Oolman,<sup>1,2</sup>  
Tegan Loveridge,<sup>1,2</sup> J. Gary Eden,<sup>3</sup> and Virginia O. Lorenz<sup>1,2</sup>

<sup>1</sup>Department of Physics, University of Illinois Urbana-Champaign, 1110 W Green St, Urbana, IL 61801, USA

<sup>2</sup>QUIST, University of Illinois Urbana-Champaign, 1101 W Springfield Ave, Urbana, IL 61801, USA

<sup>3</sup>Department of Electrical and Computer Engineering, University of Illinois Urbana-Champaign, 306 N Wright St, Urbana, IL 61801, USA

\*kais@illinois.edu

**Abstract:** We measure  $95.6 \pm 0.3\%$  storage efficiency of ultrafast photons in a collisionally broadened barium vapor quantum memory. We measure  $31 \pm 1\%$  total efficiency, limited by control field power, and a  $0.515(6)$  ns lifetime, limited by motional dephasing. © 2023 The Author(s)

Optical quantum memory constitutes the ability to store and retrieve single photons (and quantum information encoded therein) on demand, and is a critical enabling resource for many quantum technologies. We demonstrate an atomic ensemble quantum memory in hot ( $900^\circ\text{C}$ ) atomic barium vapor with record storage efficiency of  $95.6 \pm 0.3\%$  for ultrashort photons (500 fs), an atomic-motion-limited lifetime of  $0.515(6)$  ns, and ultra-low noise. Our memory platform consists of a free-space barium (Ba) heat pipe oven with argon (Ar) buffer gas. The buffer gas enables homogeneous collisional broadening of the intermediate excited state linewidth to match the bandwidth of the stored photons, which allows for efficient memory operation in the ultrafast regime.

A schematic of our experiment is shown in Fig. 1(a). Each Ba atom possesses an internal  $\Lambda$ -type energy level system, consisting of the ground ( $|g\rangle$ )  $6s^2\ ^1S_0$  state, excited ( $|e\rangle$ )  $6s6p\ ^1P_1$  orbital state, and metastable  $6s5d\ ^1D_2$  orbital storage ( $|s\rangle$ ) state [see Fig. 1(a) inset]. The resonant wavelengths of the so-called signal ( $|g\rangle \rightarrow |e\rangle$ ) and control field ( $|e\rangle \rightarrow |s\rangle$ ) transitions are 553.5 nm and 1500 nm, respectively. Initially, all atoms thermally populate the  $|g\rangle$  state, and we tune a few-photon coherent state signal field (with duration 500 fs) and a strong control field [ $O(10\ \text{uJ})$  pulse energy, 100 fs duration] approximately 5 excited state linewidths off-resonance from the atomic transitions. This relatively small detuning eliminates linear absorption of the signal field in the absence of the control, and allows for direct measurement of the memory's storage efficiency by measuring the photon number arriving at a detector with and without the control field. The  $\sim 1510$  nm control field is derived from an 800 nm optical parametric amplifier with 1 kHz repetition rate. The weak coherent state signal field is in turn generated from the control field via sum-frequency generation (SFG) with an 877 nm laser diode. A controllable Ar buffer gas pressure is confined inside our home-built heat pipe oven with 12 inch heated region. The signal and control fields are focused in the center of the oven with waist radii  $(109 \pm 3)\ \mu\text{m}$  and  $(247 \pm 4)\ \mu\text{m}$ , respectively.

After optimizing over control field frequency, pulse area, delay (relative to the signal field), and beam pointing, we experimentally measure a storage efficiency of  $(95.6 \pm 0.3)\%$ . At an optical depth of 50, the theoretical optimal bound on our storage efficiency is  $\eta_{\text{opt}} = 95.2\%$  [1], indicating that we have saturated the optimal bound. Compared to other broadband, on-demand quantum memories in the literature, shown in Fig. 1(b), this measurement not only represents a significant improvement in efficiency in the ultra-broadband regime, but also represents the highest measured storage efficiency to our knowledge for any memory with bandwidth  $>10$  MHz. This large storage efficiency is due to the large collisionally broadened excited state linewidth in our system, which better matches the bandwidth of our ultrafast photons [2]. When considering total efficiency instead of storage efficiency, we experimentally measure a maximum of  $31 \pm 1\%$  at  $900^\circ\text{C}$ , which is limited by finite available control field power. This total efficiency is nearly a factor of 2 larger than previous THz-bandwidth quantum memories. While the majority of the increase in efficiency compared to other memories is attributable to the use of a collisionally broadened system, another effect also boosts our total efficiency: the use of near-off-resonant memory (or NORM) operation. When scanning the two-photon detuning  $\Delta$  [see Fig. 1(a) inset] at  $800^\circ\text{C}$ , we find that the largest total efficiency occurs at a small, non-zero detuning [Fig. 1(c)]. We believe this NORM operation balances reabsorption loss, which is worst on-resonance, and finite available control field power, which leads to lower efficiency at larger detuning.

This work represents the first atomic ensemble quantum memory at THz-bandwidths to our knowledge; all previous THz-bandwidth quantum memories have used phononic storage states in either solids or molecular gases.

<sup>†</sup>Present address: Department of Physics, University of Colorado Boulder, 390 UCB, 325 Broadway, Boulder, Colorado 80305, USA;

<sup>‡</sup>Present address: Department of Physics and Astronomy, Rice University, 6100 Main St, Houston, Texas 77005, USA.

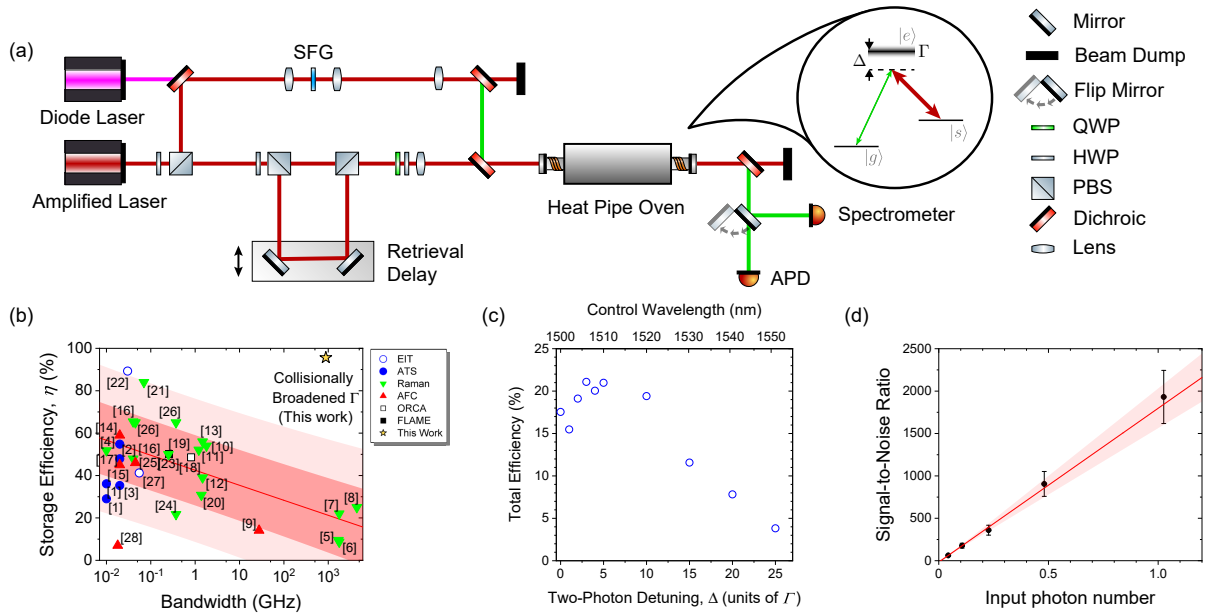


Fig. 1. (a) A schematic of the quantum memory experiment. (b) Storage efficiency as a function of memory bandwidth for published quantum memories and our memory. Numbered citations are given in Ref. [4]. (c) Total memory efficiency at 800 °C as a function of two-photon detuning, showing optimal memory efficiency at finite, near-resonant detuning. (d) Signal-to-noise ratio of our memory measured as a function of input photon number.

The use of an atomic metastable state yields a significantly longer memory lifetime, experimentally measured to be 0.515(6) ns in our case, and a time-bandwidth product of  $1031 \pm 14$ . Our memory lifetime is limited by thermal motion of the atoms and may be extended in future work with the use of a sensor state [3].

Lastly, we measure the noise performance of our memory. The signal-to-noise ratio (SNR), defined as the ratio of retrieved photon number to the number of noise photons overlapping with the retrieved field, is shown in Fig. 1(d) as a function of average input photon number. We achieve an average of  $\langle n_{\text{noise}} \rangle = (3.8 \pm 0.6) \times 10^{-5}$  noise photons per pulse, an SNR at 1 input photon per pulse of  $SNR = (8.2 \pm 1.3) \times 10^3$ , a single photon fidelity of  $F = 1 - 1/(SNR + 1) = 0.99988(2)$ , and a  $\mu_1$  parameter of  $\mu_1 = \langle n_{\text{noise}} \rangle / \eta = (1.2 \pm 0.2) \times 10^{-4}$ , where  $\eta$  is the total memory efficiency. This noise performance is significantly better than almost all broadband atomic quantum memories, and is due to the large ground-storage state splitting in Ba, which is larger than the control field frequency, and which therefore eliminates four-wave-mixing noise to first order and additionally allows for large spectral suppression of leaked control field photons.

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