Storage of Ultra-Broadband Pulses in Hot Atomic Barium Vapor

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Abstract: We demonstrate the potential for an ultra-broadband quantum memory in hot atomic barium vapor using an off-resonance Raman interaction. It may enable storage of THz-bandwidth photons for high-speed quantum information processing in the telecom range.

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The storage of single photons in a quantum memory enables the synchronization of photons from different sources, which is a required functionality in long distance quantum communication as well as local quantum operations. Quantum memories are also crucial for non-deterministic single photon sources to convert photon output to on-demand, dramatically improving their scalability. They have been demonstrated using many different methods including electromagnetically induced transparency, photon echo, atomic frequency comb and off-resonance Raman. Here we introduce an ultra-broadband quantum memory using the off-resonance Raman protocol in hot atomic barium vapor.



Fig. 1. (a) Schematic of energy levels in barium. (b) Experimental setup. M, mirror; DM, dichroic mirror; L, lens; G, diffraction grating; B, beam block; IF, interference filter.

The advantage of the off-resonance Raman-type quantum memory [1] is its capability to store broadband photons. Consider a A-level system shown in Fig. 1 (a). The signal and control fields are both detuned from excited state $|2\rangle$. When both fields are spatially and temporally overlapped, the strong control field will enable a two-photon transition between the ground state $|1\rangle$ and metastable state $|3\rangle$ that matches the frequency of the signal, which will then be stored and mapped onto a so-called spin-wave that corresponds to all possible superpositions of one atom in the storage state $|3\rangle$ and all other atoms in the ground state. When the control field is reapplied to the spin-wave excitation, the signal will be deterministically released. The bandwidth of the control field determines the storage bandwidth that can be broad thanks to the off-resonant interaction, limited mainly by the energy splitting between states $|1\rangle$ and $|3\rangle$. This eliminates the needs to engineer the single-photon source to match the exact transition. An important figure of merit of a quantum memory is the time-bandwidth product (ratio of memory lifetime to pulse duration), which represents the number of processes that can be performed before retrieval. While the natural lifetime of the storage state $|3\rangle$ in atomic barium is ~ 0.25 s, the anticipated lifetime in hot atomic barium vapor is around 1 ns due to the motion of atoms across the spin-wave wavelength. However, the large energy splitting between ground and storage states of ~ 340 THz enables storage of $< \sim 100$ fs photons, leading to a time-bandwidth product > 1000. The large energy splitting also leads to minimal thermal population in the storage state, giving very little noise in single-photon operation, and prohibiting noise from other four-wave mixing processes. To date, researchers have shown storage of GHz-bandwidth photons in atomic systems and THz-bandwidth photons in molecular and solid state systems [2], but not broadband storage in the telecom range. Barium has a transition between state $|2\rangle$ (6s6p¹P₁) and $|3\rangle$ (6s5d¹D₂) at telecom wavelengths, making it feasible for telecom photon storage if one prepares state $|1\rangle$ as the storage state.

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A schematic of the experimental setup is shown in Fig. 1 (b). The signal pulses, centered at 560 nm with 200 fs duration and 80 MHz repetition rate from an optical parametric oscillator (OPO) (Inspire HF100), are attenuated to provide coherent state signal photons to demonstrate storage. An optical pulse shaper is built for the signal arm such that we can optimize the storage efficiency by shaping the signal pulse (e.g. change its bandwidth or duration). The idler pulse centered at 1550 nm from the OPO is used as the control field; it is sent to an erbium-doped fiber amplifier (EDFA) (PriTel FA-35) to increase the pulse energy to 22.5 nJ. The control field is passed through a delay stage and combined with the signal spatially and temporally. They are focused into a heat-pipe oven containing barium atoms and 500 Torr Argon buffer gas. The beam size is measured and adjusted to ensure the signal has good overlap with the control field at the center of the oven. The Rayleigh length, which determines the interaction length, is about 4 cm. The oven is heated above 800°C. The signal can be easily separated from the control field using a dichroic mirror and is measured by a fast photodetector (PDA10A).



Fig. 2. (a) The signal absorption as a function of time delay between the signal and control field. The background may come from the stray light detected by the lock-in. (b) Storage efficiency vs. pulse energy of the control field for a density of $3.7 \times 10^{19} \text{ m}^{-3}$. (c) Storage efficiency vs. atomic barium density for a control field energy of 22.5 nJ. The data in (b) and (c) were taken at different times and may exhibit small variations for the same parameters due to slight changes in alignment.

We measure the absorption of the signal due to interaction with the control field by mechanically chopping the signal and control fields at different frequencies f_1 and f_2 , respectively, and detecting the signal incident on the photodiode with a lock-in amplifier locked to $f_1 + f_2$. We scan the time delay between the signal and control field. The signal absorption for various time delays is plotted in Fig. 2 (a). The storage efficiency of the barium memory at zero delay can be calculated as the signal detected at $f_1 + f_2$ divided by the signal detected at f_1 . The current optimal efficiency is about 0.4% for a control field energy of 22.5 nJ and atomic vapor density of $5.1 \times 10^{19} \text{ m}^{-3}$. We also measure the efficiency for various control pulse energies and atomic densities, shown in Fig. 2 (b) and (c), indicating their influence on the memory efficiency. As a next step we are improving the efficiency by increasing the pulse energy of the control field. To achieve this, we pick pulses from the 80 MHz pulse train and send them to the amplifier so that for a given amount of average power the pulse energy can be greatly enhanced by the ratio of the original repetition rate to the picked pulse frequency.

In summary, we experimentally demonstrate the potential for an ultra-broadband quantum memory in hot atomic barium vapor based on off-resonance Raman interaction. The broadband feature enables the use of ultrashort pulses for fast quantum operation. It also permits spectral pulse shaping to optimize the efficiency as well as fidelity. With the potential to store broadband telecom photons, we believe this memory should find broad interest and add to the toolbox of future quantum applications.

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