Observation of prolonged coherence time of the collective spin wave of atomic ensemble in a paraffin coated ^{87}Rb vapor cell

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We report a prolonged coherence time of the collective spin wave of a thermal ^{87}Rb atomic ensemble in a paraffin coated cell. The spin wave is prepared through a stimulated Raman Process. The long coherence time time is achieved by prolonging the lifetime of the spins with paraffin coating and minimize dephasing with optimal experimental configuration. The observation of the long time delayed-stimulated Stokes signal in the writing process suggests the prolonged lifetime of the prepared spins; a direct measurement of the decay of anti-Stokes signal in the reading process shows the coherence time is up to 300μs after minimizing dephasing. This is one hundred times longer than the reported coherence time in the similar experiments in thermal atomic ensembles based on the Duan-Lukin-Cirac-Zoller (DLCZ) and its improved protocols. This prolonged coherence time sets the upper limit of the memory time in quantum repeaters based on such protocols, which is crucial for the realization of long-distance quantum communication. The previous reported fluorescence background in the writing process due to collision in a sample cell with buffer gas is also reduced in a cell without buffer gas.

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Quantum communication is the absolute secure method for the information transfer. At present the communication distance is limited to hundred-kilometer scale mainly due to the inevitable loss of the photon during the transfer. To tackle this problem, a model of quantum repeater [1] is introduced, combining the quantum memory, entanglement purification and entanglement swapping. In the protocols to realize it, such as the DLCZ protocol [2] and the following improved schemes [3, 4, 5], quantum memory is essential to increase the success probability of such protocols because the generation of entanglement states are probabilistic.

The quantum memory is achieved by coherent manipulation of the atomic states, such as preparing a total symmetric collective state (spin wave) by Raman scattering process [2]. The qubit can then be stored and retrieved by manipulating such state. The coherence time of this collective state will determine the memory time of the quantum repeater. Long memory time is desirable for the long-distance quantum communication. For instance, to establish entanglement of two qubits over hundred-kilometer scale, the memory time needs to be on the order of hundred-microsecond.

Several groups are presently working on the implementation of the quantum repeater based on the DLCZ and its improved protocols and have achieved many significant experimental advances [6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17]. This includes the coherent manipulation of the atomic states through Raman process in an atomic ensemble [6, 7, 8, 9, 10, 11]; entanglement of the atomic states [12, 13, 14, 15] and the realization of the building block of the quantum repeater [16, 17]. Both the thermal vapor and cold atomic ensembles are employed in those experiments. Most reported coherence time based on cold atoms is in the time scale of 10 microseconds [10, 13, 16], which was attributed mainly limited by the residual magnetic field. For thermal atoms, the bottleneck of the coherence time was reported to be the atomic diffusion out of the read laser region and the inelastic collisions between the atoms and glass wall, the reported cohererence time was only about 3μs [8].

During the preparation of this paper, a study on cold atomic ensemble which is simultaneously carried out by another group also led by Jian-Wei Pan at Heidelberg recently reported ms coherence time in cold Rb ensemble [18]. This is achieved by reducing the effect of magnetic field using 'clock state' and the dephasing caused by random atomic motion with correct detection configuration.

The thermal atomic vapor cells enjoy simple technique and better magnetic shielding, but in regular glass cells, the coherence time of the prepared spin wave is limited by the collisions between walls and atoms themselves. In the regular glass cell collisions with wall will cause the spin flip, reducing its lifetime. The time scale is on the order of tens us for a realistic pencil shaped cell (cm in dimension), which is much faster than the collision between atoms (under our temperature T = 78 ^{o}C, the atomic density is $10^{12}/cm^3$, and the mean collision time is on the order of ms). A cell with paraffin coated walls [19] would exceed this limit, since paraffin coating had been demonstrated in reducing such destructive collisions, allowing atoms to undergo many elastic wall-collisions and prolong the spin lifetime up to 1 second [20]. The paraffin-coated alkali-vapor cells had been successfully used in entanglement between atomic ensembles with continuous variables [21], and slow light experiment [22]. To our knowledge, the study on the coherence time of spin waves in a paraffin coated cell based on the DLCZ protocol and the effect of dephasing as reported by Zhao *et al.* [18] in a thermal ensemble has not been reported yet. In order to prolong the coherence time, buffer gas is often filled in the sample cell, it would slow the diffusion speed of atoms to some extent, but also brought a new problem [23]: an additional and considerable fluorescence noise signal, caused by collisional perturbation of the excited state due to buffer gas during the write (read) process, severely limited the fidelity of quantum communication.

In this letter, we investigate the capability of applying the thermal atomic ensembles as candidate for quantum repeater, by studying the coherence time of the spin wave using ^{87}Rb vapor in a buffer-gas-free paraffin coated cell (P-cell). Experiments were carried out to test whether the prolonged lifetime of the spins under paraffin coating and minimizing the dephasing process could enable long coherence time in a thermal sample as well. This coherence time will provide the upper limit of the memory time in the quantum repeater. We also investigate whether the buffer-gas-free environment would reduce the collision-induced fluorescence background. Though in the DLCZ protocol, the coherent manipulation of atomic state is implemented by spontaneous Raman scattering with single excitation, stimulated Raman scattering could also be used to realize such goal, as Raymer group [24] pointed out. The coherence time would be same for both cases, and we measure it in a P-cell in the stimulated Raman region for the ease of signal detection and background distinction.

Figure 1 provides an overview of our experimental setup. The ^{87}Rb atoms are in a P-cell which is 50mm(length) by 5mm(diameter) and is heated to 78^oC. The sample cell is put in a 3-layer magnetic shielding with residual field inside 10nT. A pump laser in resonant with $|5S_{1/2}, F = 2\rangle$ to $|5P_{1/2}\rangle$ is first turned on for 50μs to pump atoms to $|5S_{1/2}, F = 1\rangle$; then the write laser whose frequency is blue shifted 1GHz with respect to the $|5S_{1/2}, F = 1\rangle$ to $|5P_{1/2}, F' = 2\rangle$ is turned on whose duration depends on the laser intensity, lasted till the stimulated Stokes signal is observed; after a controlable delay the read laser pulse of 2us whose frequency is red shifted 400MHz with respect to the $|5S_{1/2}, F = 2\rangle$ to $|5P_{1/2}, F' = 1\rangle$ is turned on. The write-read beams are counter propagating and the collection of Stokes and Anti-Stokes signals by optical fibers is also collinear. Initially we chose a skewed configuration as Braje *et al.* [23] where the two directions forms an angle $\theta = 2^o$ to minimized the background laser signal. The signals, after the polarization filter and frequency filter, were passed through a scanning Fabry-Perot (F-P) spectrometer (Finesse 200, 7.5GHz FSR, transmission 10%) and the output was fed to the single photon detector. Thus allow us to analyze the intensity of the frequency components and single out the Stokes and anti-Stokes signals. The write laser has a maximum power of 7mW and read laser of 80mW at the input face of the cell. The lasers are locked at the respective fine structures of the ^{87}Rb absorption peaks, and the frequency detuning and pulse generation are achieved by AOMs. The detection of the Stokes signal signifies the preparation of the spin wave, and the decay of the anti-Stokes signal at specified direction in the reading process at different time delay is used to measure the coherence time of the spin wave. This is essentially a Time-resolved Coherent Anti-stokes Raman Scattering (T-CARS) method.

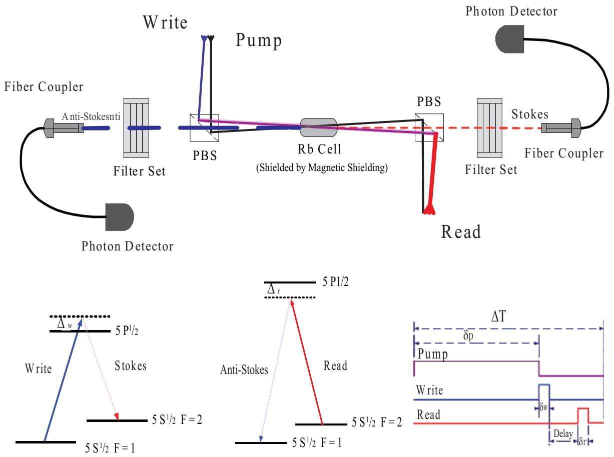

FIG. 1: Schematic of experiment for generation of atomic collective state. Off-axis, counter-propagating geometry of Braje et al.[22] is adopted in our experiment, in which write and read pulses collinearly propagate into a Rb cell in sequence and we collect the generated output fields (Stokes, Anti-Stokes), the direction of this collection forms a 2^0 angle with the direction of the write-read input light. PBS stands for polarizing beam splitters; the Filter Set is composed of selective absorption by atomic cells and narrow-band filter. The inset illustrates the relevant atomic level scheme. Δ_w =1GHz, Δ_r =400MHz

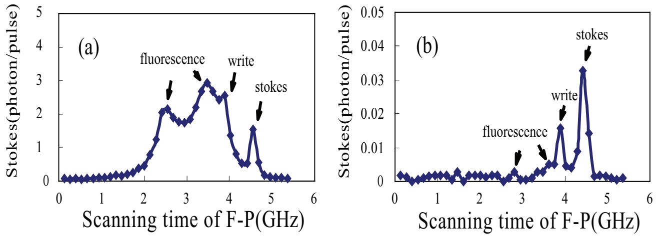

FIG. 2: Stimulated Stokes signal after a F-P. (a) is the result in a glass cell with 7 torr Neon buffer gas; (b) is the result in a P-cell with no buffer gas. The pulse width of write is 2 μs. Only polarization and narrow-band filter is applied to let the signal of the write laser appear as wavelength marker. With further frequency filter by atomic cells, the write light can be completely suppressed.

First we report the frequency analyze of the signals during the writing process in the buffer-gas-free P-Cell. Fig 2(a) is from the regular cell with 7 torr neon buffer gas where a considerable fluorescence signal is observed besides the Stokes; Fig 2(b) is from a buffer-gas-free P-cell. It is clear that the Stokes signal in this P-cell is much purer in a sense that the fluorescence background is greatly reduced. Thus the problem of collision-induced fluorescence background as reported by Manz *et al.* [23] in the buffer gas cell is suppressed by using buffer-gas-free sample cells.

Next we measure the temporal pulse shape of the Stokes signal in the writing process. This is achieved by using a long write pulse and monitoring the Stokes signal after the F-P, set the gate (1μs long) of single photon detector at various temporal positions of the write pulse, the results are shown in Fig 3. An interesting phenomenon is observed for the first time, we discover a long-delayed stimulated Stokes at various writing powers. A Stokes signal is generated by spontaneous Raman scattering during the first microseconds after the write light is turned on, corresponding to the flat region at early time in Fig 3(a), and increased exponentially at later time to reach the stimulated Raman scattering. Fig 3(a) shows the variations of this rise time at different write powers. Depending on the power, the rise time of the simulated Stokes can be as long as 150 μs.

Phenomenon of this long-delayed stimulated Raman in P- cell is in great contrast to what we observed in a non-coated glass cell in Fig 3(b). Here the rise of the stimulated Stokes is only on the order of a few μs. The rise time is delayed at lower laser power but only about 10μs, further reduce the power would make the stimulated signal disappear. This phenomenon is the result of longer lifetime of the spins in a P-cell and can be qualitatively explained as follows: The stimulated emission happens only when the number of excited spins or the amplitude of the spin wave exceed certain threshold. The long time delayed stimulated scattering in the P-cell could only happen if there is an accumulation of the spin wave, i.e. the decay of the spin wave is slower than its generation by the write pulse. Such accumulation takes longer time at lower write intensity as long as above condition is satisfied. This long time delayed stimulated Stokes in P-cell suggests that the decaying rate of the amplitude of the spin wave is much slower than that in a regular cell, because the paraffin coating helps in protecting the prepared spin wave from being destroyed by the collision with the walls, thus making the long lifetime possible. This long lifetime is a necessary condition to achieve long coherence time of the spin wave.

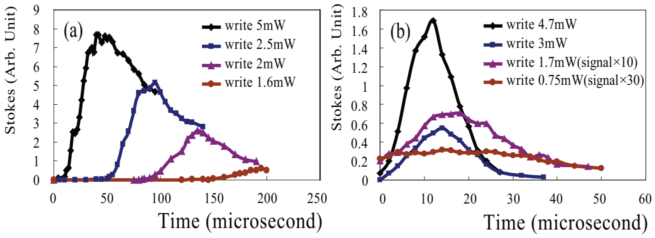

FIG. 3: (a)The long-delayed stimulated Raman Stokes signal in the paraffin-coated Rb cell. The write laser has a waist of 3mm inside the cell and its power is varied. Time zero is the turn-on of the write pulse which lasts for 200μs. (b) In the non-coated Rb cell, as the intensity of the write light decreases, the stimulated excitation transformed to spontaneous excitation without any long-delayed stimulated excitation observed. The long delayed rise of stimulated Stokes in P-cell suggests the paraffin coating helps in protecting the Rb spin from being destroyed by the collision with the wall.

Furthermore the coherence time of the spin wave generated in the writing process can be directly measured by the reading process, this is the T-CARS method as described before. The write pulse is shut off at the peak of the stimulated Stokes, which is 40μs for 5mW write power. The intensity of the anti-Stokes signal during the retrieval process is recorded and plotted against the time delay between the read and write pulses. The decay of the anti-Stokes signal is caused by decoherence of the spin wave. Fig 4(a) (write power 5mW, write beam dia.3 mm, read power 80mW, read beam dia.10mm) shows the entire decay of the anti-Stroke signal (relative intensity with respect to 0 delay between read and write) in a P-cell, which displays two exponential decays. Below 20μs is a fast decay with characterized time 10μs. The later part is a slow decay, characterized time about 80μs. This long decay time is in agreement with the results from the long-delayed stimulated Raman and suggested this long decay is resulting from the long lifetimes of the spins due to reduced inelastic collision with the paraffin coated surface. We still need to address the mechanism causing the initial fast decay in Fig 4(a).

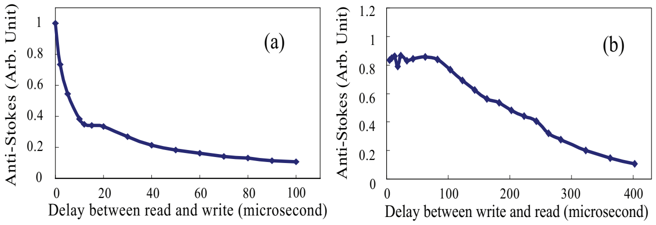

FIG. 4: Decay of Anti-Stokes V.S. delay between read and write. (a) The decay of anti-Stokes signal at $\theta = 2^o$. The diameter of the write laser is 3mm and the read is 10mm. Decay time in paraffin-coated cell is composed of fast part, which is 10μs, and slow part, which is 80μs. (b)The decay of anti-Stokes at $\theta = 0^o$. The diameter of the write laser is 6mm and the read is 10mm. It only shows a slow decay with time of 320μs.

One reason for the fast decay is caused by the diffusion of the spin wave out of the effective retrieval region covered by the reading laser, as suggested by M. D.Eisaman *et al.* [8], but this is not the major one because further studies at different write beam widths and at flatter read power distribution covering the entire cell shows the fast decay is always there under $\theta = 2^o$ detection configuration. As pointed out in the Duan *et al's* original paper [2] and investigated in detail recently by Zhao *et al* [18] in cold atom ensemble, there is a dephasing of spin wave induced by random motion of the atoms and can be easily understood in the case of single excitation.

Upon detection of the forward Stokes signal, a singly excited spin wave is created in the atomic ensemble in the form of

$$|\psi_{spin}\rangle = \frac{1}{\sqrt{N}} \sum e^{i\triangle k\cdot r_j} |1...2_j...1\rangle$$

with $\triangle k = k_w - k_s$ the wave vector of the spin wave, where k_w,k_s are those of the write laser and the Stokes. The atoms position changes to $r_j(t) = r_j + \triangle rt$ at later time and will introduce a random phase shift. This will cause a drop in the overlap between the spin waves $\langle\psi_{spin}(t)|\psi_{spin}(0)\rangle$, and thus the decay in retrieval efficiency in the reading process.

Under condition $\triangle k \cdot l << \pi$, with l the dimension of the atomic ensemble, such dephasing is negligible, but this is not the case in our skewed detection configuration. With $\theta = 2^o$, and $|\triangle k| = |k_w - k_s| \approx k_w sin\theta$, then $\lambda_{spin} \approx \lambda_w/sin\theta = 23\mu m$. This is much smaller than the size (and the maximum value of $\triangle r$) of our cell, and the dephasing caused by the random motion will be severe and the dominating factor in this case. In order to minimize the effect of dephasing, the created spin wave has to have a longer wavelength. The longest wavelength of the spin wave is achieved under collinear detection configuration with $\theta = 0^o$, where $\lambda_{spin} \approx$ 4.4cm. In fact to have the wavelength of the spin wave on the order of cm comparable to the dimension of the sample cell, the angle θ between the write and Stokes signal has to be smaller than 10^{-4} arc. So the only meaningful configuration to minimize the dephasing in our experiment is the collinear one.

Though the above argument is applied to the single excitation, it can be extended to the multi-excitation case and provides the basis for the improvement in the stimulated Raman region. A T-CARS experiment was carried out under the collinear configuration and the anti-Stokes decay is shown in Fig 4(b), the initial fast decay due to the dephasing by thermal motion disappears, only a long decay remains and the fitted (e^{-t^2/τ^2}) exponential decay time is τ ∼320 μs. This demonstrated the robust of the long wavelength of the spin wave against the dephasing and the dominant decoherence effect here becomes the spin lifetime. This result in the thermal vapor cell agrees with the recent study by Zhao *et al* in cold atom system [18].

In conclusion, we demonstrate the prolonged coherence time τ ∼320 μs of the collective spin wave in a paraffin coated ^{87}Rb cell. The coherence time is one hundred times longer than the previous reported in the thermal vapor system. In such system, the two major factors dominating the decoherence process are the lifetime of the spin and the dephasing of the spin wave due to thermal motion. The paraffin coating helps prolong the lifetime of the spin by reducing the inelastic collision with the walls which is manifested by the long delayed stimulated Stokes signal under weak write laser, and the long decay of anti-Stokes signal with T-CARS method. The dephasing can be overcome by choosing the correct detection configuration and preparing the long wavelength spin wave, this is manifested in the disappearing of the fast decay in the anti-Stokes measurement. This coherence time though measured in the stimulated region, offers at least an upper limit for memory time in quantum repeater. In our study we also demonstrated that the buffer-gas-free cell also greatly suppresses the collision-induced fluorescence background.

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