Retrieval of phase memory in two independent atomic ensembles by Raman process

CHENG-LING BIAN1, LI-QING CHEN1,3, GUO-WAN ZHANG1, Z. Y. OU1,2(a) and WEIPING ZHANG1,3(b)

1 Quantum Institute for Light and Atoms, Department of Physics, East China Normal University
Shanghai 200062, PRC
2 Department of Physics, Indiana University-Purdue University Indianapolis - 402 N. Blackford Street,
Indianapolis, IN 46202, USA
3 State Key Laboratory of Precision Spectroscopy, Department of Physics, East China Normal University
Shanghai 200062, PRC

received 15 September 2011; accepted in final form 13 December 2011
published online 31 January 2012

PACS 42.25.Kb – Coherence
PACS 42.25.Hz – Interference
PACS 42.65.Dr – Stimulated Raman scattering; CARS

Abstract – In spontaneous Raman process in atomic cell at high gain, both the Stokes field and the accompanying collective atomic excitation (atomic spin wave) are coherent. We find that, due to the spontaneous nature of the process, the phases of the Stokes field and the atomic spin wave change randomly from one realization to another but are anti-correlated. The phases of the atomic ensembles are read out via another Raman process at a later time, thus realizing phase memory in atoms. The observation of phase correlation between the Stokes field and the collective atomic excitations is an important step towards macroscopic EPR-type entanglement of continuous variables between light and atoms.

Introduction. – Correlations in a quantum system played an important role in the test of foundation of quantum mechanics [1,2], and in the applications of quantum metrology [3] and quantum information [4]. Recently, atomic memory for correlated photons is demonstrated [5,6] based on the DLCZ scheme [7] in a collective Raman process in spontaneous emission regime for the application in quantum repeaters for long distance quantum communication.

But what is demonstrated so far is the intensity correlation between the Stokes field and the atomic excitations in the spontaneous Raman process. The phase correlation between the related optical fields and atomic medium has not been directly explored, although some phase relation is shown in the photon interference experiment for the demonstration of photon entanglement at single-photon level [8–10]. Furthermore, there is some controversy in defining the phase of a field at single-photon level [11]. Thus, the phase relation measured so far is all indirect.

It is well known that spontaneous parametric processes in the high-gain regime exhibit EPR-type quantum entanglement between quadrature-phase amplitudes in continuous variables [2,12]. With the atomic spin wave involved, the collective Raman process in atomic vapor can be considered as a parametric process. Thus, there should be similar EPR-type entanglement between the amplitudes of the Stokes field and the atomic spin wave. Since the amplitude of a field consists of intensity and phase \( A = e^{i\varphi} \sqrt{I} \), with intensity (photon) correlation demonstrated [5,6,9], we just need to show that there is a strong correlation in the phases of the atomic ensemble and the Stokes field in order to prove the amplitude correlation for a demonstration of EPR-type entanglement between the amplitudes of the Stokes field and the atomic spin wave.

In this paper, we report on an experiment in which we measure the phase difference of two spatially separated atomic spin waves created via spontaneous Raman processes in high-gain regime and directly confirm a phase anti-correlation between the Stokes field and the corresponding atomic spin wave in the collective Raman process. The phase of the atomic spin wave is retrieved after a time delay of the phase measurement of the Stokes field, thus realizing the memory of phase information of optical fields.
In fig. 1(a), two Raman pump pulses (figs. 1(a) and (b), respectively). In the first stage, we show pumping. There are two stages of operation, depicted in a lambda structure: an excited state is a collective Raman scattering process in atoms with meta-stable lower level states |g⟩, |m⟩ (insets of fig. 1). N atoms are initially prepared in the ground state |g⟩ by optical pumping. There are two stages of operation, depicted in figs. 1(a) and (b), respectively. In the first stage shown in fig. 1(a), two Raman pump pulses (W₁, W₂) start the Raman scattering to produce random Stokes fields (Swₐ, Sw₂) and the corresponding atomic spin waves \( \hat{S}_{\alpha 1,2} \equiv (1/\sqrt{N}) \sum_i |g_{1,2},i \rangle \langle m_{1,2},i | \) between two lower states in two separate atomic ensembles. We superimpose the generated Stokes fields to measure their relative phase. The second stage reads out the atomic spin waves by another Raman process as shown in fig. 1(b) and the atomic coherence is through anti-Stokes field as in the interference pattern. We look for the correlation between the two measured phases.

The interaction Hamiltonian in the write process is given by [7]

\[
\hat{H}_R = \eta A_W^* \hat{a}_{Sw} \hat{S}_a + \text{h.c.},
\]

where we treat the write field as a strong classical field with an amplitude of \( A_W \). \( \hat{a}_{Sw} \) denotes the Stokes field generated in the write process and \( \hat{S}_a \) is for the atomic spin wave. The quantum evolution of the Stokes field and the atomic spin wave is given by [13]

\[
\begin{align*}
\hat{a}_{Sw}(t) &= \hat{a}_{Sw}(0) \cosh \zeta t + \hat{S}_a(0) \sinh \zeta t, \\
\hat{S}_a(t) &= \hat{S}_a(0) \cosh \zeta t + \hat{a}_{Sw}(0) \sinh \zeta t,
\end{align*}
\]

where \( \zeta \propto |A_W| \). Note that the model presented in eq. (1) and the equations of motion in eq. (2) are over-simplified without consideration of spatial propagation. A full account of traveling pulses interacting with an atomic medium can be found in ref. [14]. Nevertheless, the toy model presented here can be used to illustrate the simple physical picture in our experiment.

Fig. 1: (Color online) The conceptual diagrams for measuring phase correlation: (a) writing process; (b) reading process with two methods (Retrieval I, II). Insets: atomic levels for Raman scattering.

**Idea of the experiment.** – The conceptual sketch of the experiment is shown in fig. 1. The basic process is a collective Raman scattering process in atoms with a lambda structure: an excited state |e⟩ and two metastable lower level states |g⟩, |m⟩ (insets of fig. 1). N atoms are initially prepared in the ground state |g⟩ by optical pumping. There are two stages of operation, depicted in figs. 1(a) and (b), respectively. In the first stage shown in fig. 1(a), two Raman pump pulses (W₁, W₂) start the Raman scattering to produce random Stokes fields (Sw₁, Sw₂) and the corresponding atomic spin waves \( \hat{S}_{\alpha 1,2} \equiv (1/\sqrt{N}) \sum_i |g_{1,2},i \rangle \langle m_{1,2},i | \) between two lower states in two separate atomic ensembles. We superimpose the generated Stokes fields to measure their relative phase. The second stage reads out the atomic spin waves by another Raman process as shown in fig. 1(b) and the atomic coherence is through anti-Stokes field as in the interference pattern. We look for the correlation between the two measured phases.

The interaction Hamiltonian in the write process is given by [7]

\[
\hat{H}_R = \eta A_W^* \hat{a}_{Sw} \hat{S}_a + \text{h.c.},
\]

where we treat the write field as a strong classical field with an amplitude of \( A_W \). \( \hat{a}_{Sw} \) denotes the Stokes field generated in the write process and \( \hat{S}_a \) is for the atomic spin wave. The quantum evolution of the Stokes field and the atomic spin wave is given by [13]

\[
\begin{align*}
\hat{a}_{Sw}(t) &= \hat{a}_{Sw}(0) \cosh \zeta t + \hat{S}_a(0) \sinh \zeta t, \\
\hat{S}_a(t) &= \hat{S}_a(0) \cosh \zeta t + \hat{a}_{Sw}(0) \sinh \zeta t,
\end{align*}
\]

where \( \zeta \propto |A_W| \). Note that the model presented in eq. (1) and the equations of motion in eq. (2) are over-simplified without consideration of spatial propagation. A full account of traveling pulses interacting with an atomic medium can be found in ref. [14]. Nevertheless, the toy model presented here can be used to illustrate the simple physical picture in our experiment.

With vacuum input for the Stokes field and atoms in the ground state, we can calculate the correlation function

\[
\langle \hat{a}_{Sw}(t) \hat{S}_a(t) \rangle = \cosh \zeta t \sinh \zeta t,
\]

or the normalized correlation function

\[
\gamma_{a_{Sw}, S_a} = \frac{\langle \hat{a}_{Sw}(t) \hat{S}_a(t) \rangle}{\sqrt{\langle \hat{S}_a^2(t) \rangle \langle \hat{a}_{Sw}^2(t) \rangle}} = 1.
\]

Note that this quantity is different from the coherence function of \( \langle \hat{a}_{Sw}^2(t) \hat{S}_a(t) \rangle \), which vanishes here. Equation (4) indicates that the phase \( \varphi_{Sw} \) of the Stokes field and the phase \( \varphi_{S_a} \) of the atomic spin wave are anti-correlated via \( \varphi_{Sw} + \varphi_{S_a} = \text{const.} \).

To confirm the phase anti-correlation, we need to measure the phases of the Stokes field and the atomic spin wave. Since phase is a relative quantity, we use two Raman processes to compare the relevant phases. The measurement of the phase of the Stokes fields is done by superposing the two Stokes fields to observe interference pattern. This was first observed by Kuo et al. [15] in a Raman process in hydrogen cells and more recently by Chen et al. in a Rb atomic cell [16]. The interference pattern in our case is a frequency beat [16], as shown in the first part of fig. 2 (blue curve). The phases of the Stokes fields can be extracted from the beat signal (see below for more details).

For the phase of the atomic spin waves, we need to read out the atomic spin waves and superpose the two readout fields (fig. 1(b)). The traditional way to read out the atomic coherence is through anti-Stokes field as in the well-known CARS technique [17], shown as the ASR field in fig. 1(b) (Retrieval I). However, the CARS signal is typically very weak and short to exhibit any beat signal for phase measurement. Instead, we resort to an amplification feature in the same Raman process: when a seed is injected in the Raman process, it will be amplified, even though it may add in noise as spontaneous emission. Here, the seed is the atomic spin wave prepared in the first stage.
Recently, this amplification process was used to enhance Raman scattering [18]. As in any three-wave mixing process, besides the amplification of the seed wave, there is an idler field generated. It has about the equal size as the amplified field and carries the coherence of the seed. In the case here, the idler field is the Stokes field. So in the second stage of the experiment (fig. 1(b)), immediately after the first one, we send in reading pulses at similar frequency to the write lasers (Retrieval II). In order to preserve the coherence in the process, we split one laser into two reading beams. The generated Stokes fields $S_{R1}, S_{R2}$ are related to the initial atomic spin waves by [13]:

$$\hat{a}_{S_{R1}}(t) = G\hat{a}_{S_R}(0) + e^{i\varphi_R} \hat{S}_a(0)\sqrt{G^2 - 1},$$  \hspace{1cm} (5)

where $G$ is the amplitude gain of the amplifier. Then the coherence function between the two read Stokes fields is

$$\langle \hat{a}_{S_{R1}}^\dagger \hat{a}_{S_{R2}} \rangle = (G^2 - 1)\langle \hat{S}_{a1}(0)\hat{S}_{a2}(0) \rangle e^{i(\varphi_{R2} - \varphi_{R1})}.$$

So the coherence of the two atomic spin waves is directly related to the coherence of the two Stokes fields generated by the reading fields. Here the initial Stokes field $\hat{a}_S(0)$ is in vacuum and $\varphi_R$ is the phase of the reading field. We observe the interference between $S_{R1}, S_{R2}$ by mixing them. A typical interference pattern is shown as a frequency beat in the second part of fig. 2 (red line). The beat signal in the reading process is delayed from the beat signal in the writing process by a fixed duration determined by the delay between the writing and reading pulses ($\tau \sim 0.1 \mu s$ from the end of the write pulses. See the inset of fig. 2).

We may extract the phase information by measuring the locations ($\tau_W, \tau_R$) of the first peak in the beat signals relative to fixed reference points (A and B points in fig. 2): $\varphi_{S_{R1}} = 360 \times \tau_R/T_R, \varphi_{S_{R2}} = 360 \times \tau_W/T_W$. Here $T_W, T_R$ are the average periods of the beat signals for Stokes of write and read, respectively.

**Experiment.** – A somewhat detailed experimental sketch is shown in fig. 3, together with timing sequence as inset. The two atomic ensembles are isotopically enriched Rb-87 without buffer gas, contained in two cylindrical Pyrex cells (length and diameter are 75 mm and 19 mm, respectively). The cells are, respectively, mounted inside three-layer magnetic shielding to reduce stray magnetic fields. The cells are heated up to 72 $^\circ$C using bifilar resistive heaters. Referring to the atomic energy levels shown in fig. 1, the two lower energy levels of the lambda structure are the hyperfine splitting of the ground states: $(g, m) = 5^2S_{1/2}(F = 1, 2)$. The upper level is $|e\rangle = 5^2P_{1/2}$. The optical pumping pulses (Pump, not shown in experimental arrangement) are applied before the write pulses in order to initialize the atoms in the ground state $(g) = 5^2S_{1/2}(F = 1)$. The write pulses are from a single-frequency laser operating at 795 nm and are detuned from the Rb-87 D$_1$ line ($5^2S_{1/2} \rightarrow 5^2P_{1/2}, F' = 2$ transition) by $\Delta = 0.8$ GHz for maximum Raman gain. The duration of the two write pulses is 3 $\mu$s and the powers are 11 mW and 21 mW, respectively. After a short delay of $\tau \sim 0.1 \mu s$ from the end of the write pulses, we send in the reading pulses. They are derived from one laser by a polarization beam splitter (PBS) and have a duration of 16 $\mu$s. The powers of the reading beams ($R_{1,2}$) are 24 mW and 3.2 mW, respectively. The generated Stokes fields are orthogonal to the reading fields in polarization and can be easily separated by PBS. The Stokes fields $S_{R1}$ is combined with $S_{R2}$ via a PBS and a half wave plate (HWP$_2$). Because of the different power in the reading beams, the generated Stokes fields also differ. We use a half wave plate (HWP$_2$) to adjust their relative intensities to obtain the maximum contrast in interference. The combined field is projected to the same polarization (HWP$_1$ + PBS) before coupled into a single-mode fiber (SMF) for spatial mode clean-up and photo-detection. The Stokes fields from the write process undergo a similar arrangement. A digital scope monitors the temporal behavior of the detected signals with a typical result shown in fig. 2. The extracted phases from the two beat signals are shown in fig. 4(a).

As observed in refs. [15,16], the phases of the Stokes fields are random. Here we also observe random phases for the atomic spin waves. The results of the phase measurement are shown in fig. 4(a), where we plot $\varphi_{S_{R1}}$ vs. $\varphi_{S_{R2}}$. A strong correlation of $\varphi_{S_{R1}} - \varphi_{S_{R2}}$ = const is observed. This correlation is because both $\varphi_{S_{R1}}$ and $\varphi_{S_{R2}}$ are anti-correlated to the phase $\varphi_s$ of the atomic spin wave by $\varphi_{S_{R1}} + \varphi_{S_{R2}}$ = const and $\varphi_{S_{R1}} + \varphi_{S_{R2}}$ = const. So fig. 4(a) is an indirect confirmation of the anti-correlation relation between $\varphi_{S_{R1}}$ and $\varphi_{S_{R2}}$.

As discussed earlier and shown in fig. 1(b), we can also retrieve the phases of the atomic spin waves by anti-Stokes (Retrieval I) process even though it is quite weak. But if the read field is strong enough, the anti-Stokes field can exactly represent the atomic spin wave $\hat{S}_s$ [7,19]. For the anti-Stokes read process, we need to tune the read laser to $m \rightarrow e$ transition as shown in fig. 1(b). The anti-Stokes process will then dominate over the Stokes process. From the interference signal $AS_{R1} + AS_{R2}$, we may likewise
extract the phases $\varphi_{AS}$ of the anti-Stokes field, which is exactly the phase of the atomic spin waves.

The anti-Stokes field in the Raman process depends on the atomic spin wave as well as the strength of the reading field. It is usually weak and has relatively short duration due to limited atomic excitation which results in a small atomic spin wave. Thus, it is very hard to observe multiple periods of the beat signal with the anti-Stokes fields. To extract the phases, we resort to the second method, where we measure simultaneously the individual and resultant intensities of the interfering fields. More specifically as shown in fig. 5, three detectors are used in this scheme where $D_1$, $D_2$ record the individual intensities $I_1$, $I_2$ of two interfering fields and $D_3$ records the intensity $I_{12}$ of the superposed field: $i_1 = \alpha_1 I_1, i_2 = \alpha_2 I_2, i_3 = \alpha_3 I_{12}$ Here $i_{1,2,3}$ are the photo-currents of $D_1, D_2, D_3$, respectively and $\alpha_{1,2,3}$ are the corresponding efficiencies. A single-mode fiber (SMF) is used for the superposition of the two anti-Stokes fields ($AS_{R1,2}$) in order to obtain good spatial overlap. A half wave plate (HWP) and polarization beam splitters (PBS) are used in order to adjust the relative strength of the two anti-Stokes fields. From the expression:

$$I_{12} = I_1 + I_2 + 2\sqrt{I_1 I_2} \cos \varphi, \quad (7)$$

where temporal dependence in the form of beat disappears when we set the beat frequency $\Delta \nu$ to zero by properly adjusting the powers of the reading fields, we can then extract the phase as

$$\cos \varphi = (I_{12} - I_1 - I_2) / 2\sqrt{I_1 I_2} = i_3 \sqrt{\alpha_1 \alpha_2 / \alpha_3^2} - i_2 \sqrt{\alpha_1 / \alpha_2 - i_1 \sqrt{\alpha_2 / \alpha_1}}. \quad (8)$$

The efficiency ratios $\alpha_1 / \alpha_3, \alpha_2 / \alpha_3$ can be calibrated by blocking one of the interfering fields.

To be consistent, a similar method is used to extract the phases of the Stokes fields at the same time as we extract the phases of the corresponding anti-Stokes fields. In fig. 4(b), we plot the phases of the read anti-Stokes field $\varphi_{AS}$ vs. the phases of write Stokes field $\varphi_S$. The solid line has a slope of $-1$.

**Discussion.** – The phase correlation studied here has been explored indirectly at single-photon level in some interference experiments involving entanglement between photon and atom [8–10]. Indeed, the observation of a steady interference fringe in coincidence measurement implies a steady phase correlation $\varphi_{AS} + \varphi_{SW} = \text{const}$. However, since phase is not definable at single-photon level in the sense that it cannot be measured in one realization [11], ensemble average has to be taken in these experiments. The phase correlation of $\varphi_{AS} + \varphi_{SW} = \text{const}$ makes the observation possible at single-photon level even though each phase is not definable. In our experiment, on the other hand, since we operate at macroscopic level, the phase information can be extracted in a single realization —no ensemble average is needed. So our experiment directly demonstrate the phase correlation of $\varphi_{AS} + \varphi_{SW} = \text{const}$. The preservation of the phase relationship from single-photon level to macroscopic level is due to stimulated emission. Phase measurement in single realization makes it possible to encode information in the phases of the fields for storage.

Note that the anti-phase correlation relationship in fig. 4(b) between the Stokes and the anti-Stokes field was evident in an experiment by Wang et al. [20] in a simultaneous CARS process where the write and the read fields are from the same field. In other words, there is no delay between the write and read processes. By introducing a delay in our current work, we achieved a phase memory mechanism where we first store the phase information in the atomic medium and later retrieve it. Similar delayed Raman processes were used recently to coherently store
optical pulses with unprecedented bandwidth in Cs atomic cell [21].

The phase correlation revealed here between the Stokes field and the atomic spin wave together with the intensity correlation studied in photon correlation experiment [5–7,9] implies an amplitude correlation. In fact, this type of amplitude correlation is exactly the original EPR entanglement of quadrature phase amplitudes or continuous variables entanglement associated with any three-wave mixing process [2,12]. In our case here, it is the entanglement of the quadrature-phase amplitudes of Stokes field and the atomic spin wave. Therefore, our experiment is an important step towards the demonstration of EPR-type continuous-variables entanglement between light and atomic ensemble. With two such systems, by making a projective measurement of continuous variables [14,22] on the two Stokes fields, we should be able to put the atomic spin waves in the two atomic ensembles in an entangled state, thus realizing a quantum repeater for continuous variables.

***

This work is supported by the National Basic Research Program of China (973 Program Grant No. 2011CB921604), the National Natural Science Foundation of China (Grant Nos. 11129402, 11004058, J1030309, and 10588402).

REFERENCES

[1] Clauser J. F. and Shimony A., Rep. Prog. Phys., 41 (1978) 1881.
[2] Ou Z. Y., Pereira S. F., Kimble H. J. and Peng K. C., Phys. Rev. Lett., 68 (1992) 3663.
[3] Giovannetti V., Lloyd S. and Maccone L., Science, 306 (2004) 1330.
[4] Zeilinger A., Rev. Mod. Phys., 71 (1999) S288.
[5] van der Wal C. H., Eisma M. D., Andre A., Walsworth R. L., Phillips D. F., Zibrov A. S. and Lukin M. D., Science, 301 (2003) 196.
[6] Kuzmich A., Bowen W. P., Boozer A. D., Boca A., Chou C. W., Duan L.-M. and Kimble H. J., Nature (London), 423 (2003) 731.
[7] Duan L.-M., Lukin M. D., Cirac J. I. and Zoller P., Nature, 414 (2001) 413.
[8] Matsukevich D. N. and Kuzmich A., Science, 306 (2004) 663.
[9] Chou C. W., de Riedmatten H., Felinto D., Polyakov S. V., van Enk S. J. and Kimble H. J., Nature, 438 (2005) 826.
[10] Matsukevich D. N., Chanelière T., Bhattacharya M., Lan S.-Y., Jenkins S. D., Kennedy T. A. B. and Kuzmich A., Phys. Rev. Lett., 95 (2005) 040405.
[11] Noh J. W., Fougeres A. and Mandel L., Phys. Rev. Lett., 71 (1993) 2579.
[12] Reid M. D., Phys. Rev. A, 40 (1989) 913.
[13] Raymer M. G. and Mostowski J., Phys. Rev. A, 24 (1981) 1790.
[14] Raymer M. G., J. Mod. Opt., 51 (2004) 1739.
[15] Kuo S. J., Smithey D. T. and Raymer M. G., Phys. Rev. A, 43 (1991) 4083.
[16] Chen L. Q., Bian C.-L., Zhang G.-W., Ou Z. Y. and Zhang W., Phys. Rev. A, 82 (2010) 033832.
[17] Shen Y. R., Principles of Nonlinear Optics (Wiley, New York) 1984.
[18] Chen L. Q., Zhang G.-W., Yuan C.-H., Jing J., Ou Z. Y. and Zhang W., Appl. Phys. Lett., 95 (2009) 041115.
[19] Ou Z. Y., Phys. Rev. A, 78 (2008) 023819.
[20] Wang Y. Y., Wu C., Couny F., Raymer M. G. and Benabid F., Phys. Rev. Lett., 105 (2010) 123603.
[21] Reim K. F., Nunn J., Lorenz V. O., Sussman B. J., Lee K. C., Langford N. K., Jaksch D. and Walmsley I. A., Nat. Photon., 4 (2010) 218.
[22] Furusawa A., Sorensen J. L., Braunstein S. L., Fuchs C. A., Kimble H. J. and Polzik E. S., Science, 282 (1998) 706.