Heralded generation of single photons entangled in multiple temporal modes with controllable waveforms

A Gogyan¹, N Sisakyan¹, R Akhmedzhanov² and Yu Malakyan¹,³

¹ Institute for Physical Research, Armenian National Academy of Sciences, Ashtarak-2, 0203, Armenia
² Institute of Applied Physics, Russian Academy of Sciences, ul. Ul’yanova 46, Nizhni Novgorod, 603950, Russia
³ Centre of Strong Field Physics, Yerevan State University, 1 A. Manukian St., Yerevan 0025, Armenia

E-mail: agogyan@gmail.com

Received 23 April 2014, revised 3 August 2014
Accepted for publication 19 August 2014
Published 2 October 2014

Abstract
Time-bin entangled single-photons are highly demanded for long distance quantum communication. We propose a heralded source of tunable narrowband single photons entangled in well-separated multiple temporal modes (time bins) with controllable amplitudes. The detection of a single Stokes photon generated in a cold atomic ensemble via Raman scattering of a weak write pulse heralds the preparation of one spin excitation stored within the atomic medium. A train of read laser pulses deterministically converts the atomic excitation into a single anti-Stokes photon delocalized in multi-time-bins. The waveforms of bins are well-controlled by the read pulse parameters. A scheme to measure the phase coherence across all time bins is suggested.

Keywords: time-bin entanglement, single photon, entanglement and quantum nonlocality

((Some figures may appear in colour only in the online journal)
do not fulfill all these requirements. In particular, for SPDC in non-linear crystals, the generation of pure SP states and controllability of the waveforms of temporal modes are main challenges. Furthermore, the photon linewidth is too broad to address atomic transitions effectively. For atomic ensembles, the repetition rate and a nonzero probability of generating more than one photon are major obstacles. Additionally, in some models, the number of SP temporal modes and delay between the bins are not easily controlled.

In this paper, we propose a scheme that is a promising candidate for high-quality sources of time-bin entangled SP states featuring the properties listed above. The proposed scheme is a heralded generation system, where, at first, similar to DLCZ protocol [16], a Stokes photon is produced via Raman scattering of a weak write pulse in an ensemble of cold lambda-atoms confined inside a hollow core of a single-mode photonic-crystal fiber (HC-PCF) (figure 1). The successful detection of the Stokes photon by two single-photon detectors D1 and D2 heralds the creation of one collective excitation in the atomic ensemble. Then, the atomic excitation is converted into one anti-Stokes photon by applying a train of phase-locked read laser pulses, the number and intensities of which are adjusted such that an individual read pulse cannot retrieve the anti-Stokes photon completely but the total conversion is highly efficient with probability of 1. This can be achieved due to the fiber enhanced atom-photon interaction and multi-atom collective interference effects [16]. As a result, the anti-Stokes photon is emitted in a well-defined spatial mode and is coherently localized in many temporal modes. Note that the control of anti-Stokes photon waveform by varying the intensity and frequency of read pulse has been demonstrated experimentally in the DLCZ scheme [17, 18].

The main limitation of our scheme is its low heralding efficiency due to low probability for the Stokes-photon emission needed to exclude multi-atom events in the collective spin excitation. Additional imperfections may result from Stokes photon losses when a heralded signal is present but no Stokes photon is detected due to detector inefficiencies. Therefore, the experimental verification of heralded creation of atomic excitation is necessary to assess the single-excitation regime for each ensemble. A convenient parameter is the cross-correlation function between the Stokes and anti-Stokes photons, the large values of which under conditions of weak Stokes generation indicate the presence of a single excitation in the medium. This protocol has been recently realized in cold atoms confined in a magneto-optical trap [20, 21] by performing sequential write trials and heralding measurements. The cross-correlation function temporal structure was reported in [19].

Our model is described in more detail in the next section. The anti-Stokes photon flux is calculated for different sets of read pulses, clearly demonstrating a well-defined dependence of time-bin waveforms on the profiles of read pulses. The experimental test to verify the coherence between anti-Stokes temporal modes is discussed in section 3. The results and conclusions are provided in section 4.

2. Model and basic equations

The model discussed in this paper is based on our earlier works [22, 23]. It describes a cold ensemble of $N$ four-level atoms, which are trapped inside a HC-PCF of small diameter $D$ and length $L$ (see figure 1). The atoms are initially prepared in the state $\ket{1}$ by optical pumping and are strongly confined in the transverse direction inside the fiber core that prevents atom-wall collisions [24]. The write $\omega_W$ laser field with peak Rabi frequency $\Omega_W$ interacts with the atoms at the transition $1 \rightarrow 3$ and generates a single Stokes photon at the transition $3 \rightarrow 2$. Since in the forward direction all the atoms are identically and strongly coupled to the Stokes photon [16], a symmetrically distributed spin excitation is stored in the medium.
We employ the far off-resonant Raman configuration (figure 1), with large detuning $\Delta_{\text{W}} = \omega_3 - \omega_\text{W}$ that makes the system immune to spontaneous losses into field modes other than the Stokes mode, as well as to dephasing effects induced by other excited states. After a Stokes photon is successfully detected by the D1:D2 block in figure 1, a train of read $\omega_\text{R}$ laser pulses is applied at the transition $2 \rightarrow 4$ in a far off-resonant Raman configuration $\Delta_{\text{R}} = \omega_{\text{D}2} - \omega_\text{R} \neq 0$ with adjusted intensities such that the atomic spin excitation is completely converted at the transition $4 \rightarrow 1$ into a tunable anti-Stokes photon delocalized in multi time-bins (the case of two read pulses is shown in figure 1) with amplitudes that are easily controlled as they are proportional to the profiles of read laser subpulses (see equation (12)). It should be noted that the four-level scheme is more general in the sense that it allows adjustment of the atomic spin-flip operators by the input–output relation yields the photon fluxes as follows:

$$N(t) = \frac{2\pi h_0 \omega_{\text{R}}}{V} \left( \hat{a}_S + \hat{a}_S^\dagger \right) \exp (i \mathbf{k}_z z - i \omega t), \quad i = S, AS,$$  

with the quantization volume $V$ equal to the interaction volume $V = \pi D^2 L/4$. Owing to the large detunings $\Delta_{\text{W}, \text{R}}$ we adiabatically eliminated the upper states $| 1 \rangle$ and $| 4 \rangle$, which are generally different. Then, in the rotating frame approximation, the interaction Hamiltonian for the total system in terms of the atomic collective spin operators

$$S^{\dagger} = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} \sigma_{21}^{(i)}$$  

is written as [22]

$$H = \hbar \sqrt{N} \left[ G(t) S^{\dagger} \hat{a}_S - F(t) S^{\dagger} \hat{a}_{\text{AS}} \right] + \text{h.c.}$$  

In equation (2), the summation is taken over all atoms. $\hat{d}_{\text{off}}^{(i)} = | \alpha \rangle \langle \beta |$ is the atomic spin-flip operator on the basis of the two ground states $| 1 \rangle$ and $| 2 \rangle$ for the $i$th atom.

$$G(t) = g_S \frac{\Omega_\text{W}}{\Delta_{\text{W}}}(t), \quad F(t) = g_{\text{AS}} \frac{\Omega_\text{R}(t)}{\Delta_{\text{R}}},$$  

where $f_\text{W}(t)$ is the profile of the write pulse, while $\Omega_\text{R}(t)$ is defined as

$$\Omega_\text{R}(t) = \sum_{i=1}^{J} \Omega_{\text{R}}^{(i)}(t - t_i),$$  

showing that the well-separated readout pulses with peak Rabi frequencies $\Omega_i$ are localized at time moments $t_j > t_j - 1 > \ldots > t_1$ with temporal profiles $f_j(t - t_i)$ and relative delay between them much larger than their lengths $T_j$. Hereafter, for simplicity, we consider the Rabi frequency of the write pulse as real. In equations (3) and (4) the atom-photon coupling constants are given by

$$g_S = \left( \frac{2\pi h_0 \omega_{\text{R}}}{V} \right)^{1/2} \frac{\mu_3}{2}, \quad g_{\text{AS}} = \left( \frac{2\pi h_0 \omega_{\text{R}}}{V} \right)^{1/2} \frac{\mu_4}{2},$$  

where $\mu_{ij}$ the $| i \rangle \rightarrow | j \rangle$ transition dipole matrix element. Note that the Stark shifts of the ground levels $| 1 \rangle$ and $| 2 \rangle$ induced by the write and read pulses are considered smaller compared to the spectral width of the Stokes and anti-Stokes fields and can be incorporated into their frequencies.

Our aim is to calculate the fluxes of Stokes and anti-Stokes photons from the medium

$$\frac{d n_i}{dt} = \alpha(t)(N_{\text{sp}}(t) + 1), \quad i = S, AS,$$  

where the annihilation operator of $i$th output photon $\hat{a}_{i,\text{out}}(t)$ is connected with the intracavity $\hat{a}_{i}(t)$ and input $\hat{a}_{i,\text{in}}(t)$ annihilation operators by the input–output relation $\hat{a}_{i,\text{out}}(t) = \hat{a}_{i,\text{in}}(t) + \sqrt{2} \hat{a}_{i}(t)$ and satisfies the commutation relation $[\hat{a}_{i,\text{in}}(t), \hat{a}_{i,\text{out}}(t)] = \hat{a}_{i,\text{in}}(t), \hat{a}_{i,\text{out}}(t')] = \delta(t - t').$ This technique is developed in our earlier work [22]. Here, we sketch the main steps leading to the final result.

The elimination of quantum fields in the bad cavity limit $\chi \gg g_{\text{AS}, S}$ leads to the following links between the field and atomic operators

$$\frac{dn_i}{dt} = \alpha(t) \left( N_{\text{sp}}(t) + 1 \right), \quad i = S, AS,$$  

where the substitution of these equations into equation (8) with input–output relation yields the photon fluxes

$$\frac{d n_i}{dt} = \alpha(t) N_{\text{sp}}(t), \quad i = S, AS,$$  

are the gains of Stokes and anti-Stokes fields, respectively, and $N_{\text{sp}} = \langle S | S \rangle$ is the number of atomic spin-wave excitations having the form [22]

$$N_{\text{sp}}(t) = \int_{-\infty}^{t} \! dt' \alpha(t') \exp \left[ \int_{t}^{t'} \! dt \left[ \beta(t) - \beta(t') - \Gamma_{\text{sp}}(t) \right] \right].$$  

Here, the total relaxation $\Gamma_{\text{sp}}(t) = \gamma_c + \Gamma_\text{W}(t) + \Gamma_\text{R}(t)$ comprises the decay rate $\gamma_c$ of atomic ground states coherence and the rates of optical pumping between the states 1 and 2 via write and read pulses.
\[ I_W(t) = \frac{\Omega_R^2}{\Delta_R^2} f_W(t) \gamma_{ST}, \quad I_R(t) = \frac{|\Omega_R(t)|^2}{\Delta_R^2} \gamma_{SI} \]  

(16)

where \( \gamma_{ij} \) is a partial decay rate of upper level \( i \) to the state \( j \), whose summation gives the spontaneous decay rate of upper states \( \gamma = \gamma_{ji} = \gamma_{ij} \).

There are two important consequences from equations (11), (12) and (15). First, for the large signal-to-noise ratio \( \alpha_r W \gg 1 \) and \( \beta W R \gg 1 \), the relaxation terms in equation (15) can be neglected. Then, the photon numbers are readily found from equations (11), (12) and (15) to be

\[ n_S(t) = N_{eq} = e^{\int_{-\infty}^t \alpha(r) \, dt} - 1, \quad t < T_W \]  

(17)

\[ n_{AS}(t) = n_S(\infty) \left( 1 - e^{-\int_{-\infty}^t \alpha(r) \, dt} \right), \quad t \geq T_W + \tau_D. \]  

(18)

This shows that, on one hand, the number of detected Stokes photons is equal to the number of spin excitations stored in the atomic medium during the write pulse of duration \( T_W \) and, on the other hand, the equation unambiguously determines the number of anti-Stokes photons retrieved from the medium at the end of the read subpulses. Here, the time delay \( \tau_D \) between the read and write laser pulses is large compared to \( T_W \) and, at the same time, is much shorter than the spin decoherence time \( \gamma^{-1} \).

Second, taking into account the orthogonality of the functions \( f(t-t_i) \) (see equation (6)), equation (12) describes the deterministic generation of the anti-Stokes photon entangled in non-overlapping temporal modes with controllable intensities. An important issue arising here is the phase-coherence of anti-Stokes photon delocalization in multiple time bins. In the next section, we suggest an experimental test to verify this coherence.

For numerical calculations, we consider cold \(^{87}\text{Rb}\) atoms with the ground states \( 5S_{1/2}(F = 1, 2) \) and excited states \( 5P_{2/3}(F = 2) \) and \( 5P_{3/2}(F = 2) \) as the atomic states 1 and 2, and 3 and 4 in figure 1, respectively. The number of atoms confined in a hollow-core fiber of the length \( L \sim 3 \text{ cm} \) and diameter \( D \sim 5 \text{ \mu m} \) is approximately \( N \sim 10^4 \) [25], and the fields are tuned far from the one-photon resonance by \( \Delta_R R \approx 20 \text{\mu m} \). Below \( \gamma_r \) is neglected. The durations of the write and read subpulses in equation (6) are taken as \( T_W \sim T_i \sim 1 \text{\mu s} \). Here, we assume that anti-Stokes photons are retrieved and detected with efficiency close to unity, taking into account the recent progress in this area [26, 27]. In figure 2, we present the retrieved anti-Stokes photons for two sets of read pulses. In the first case, the amplitudes of three read pulses are the same (figure 2(a)), while in the second, they are redefined such that the temporal modes of the anti-Stokes photons have equal intensities (figure 2(b)). The number of anti-Stokes photons in the modes is determined by the areas of the corresponding peaks. The parameters chosen for the write pulse are such that, on average, only one Stokes photon is generated: \( n_S(\infty) \sim 1 \) or, equivalently from equation (17), \( \int_{-\infty}^{\infty} \mathrm{d} \tau \alpha(\tau) \sim 0.7 \).

**Figure 2.** Anti-Stokes photon flux as a function of time in units of \( \gamma \) in two cases of: (a) three identical read pulses (red dashed lines) and (b) equal intensities of three temporal modes of anti-Stokes field (blue solid lines). The black dotted line shows the Stokes pulse of the unit area. The sum area of anti-Stokes pulses is 0.98. In both cases the Stokes and read pulses are scaled by factors 0.5 and 0.08, respectively.

### 3. Phase coherence of atomic spin-wave conversion into multi-pulse anti-Stokes photon

The pure state of the anti-Stokes photon entangled over \( J \) temporal modes has the form

\[ |\Psi_{AS}\rangle = \sum_{j=1}^{J} C_j |j\rangle \prod_{\mu \neq j} |0\rangle. \]  

(19)

where \( |j\rangle \) and \( |0\rangle \) denote Fock states with zero and one photon, respectively, at time \( t_i \). Here, the complex amplitudes \( C_j \) with normalization \( \sum_j |C_j|^2 = 1 \) depend on the intensities and, in the ideal case, only on relative phases of readout pulses. The main requirement is that these pulses should maintain mutual coherence to preserve entanglement between temporal modes as the entanglement implies a constant phase relation between its different components. However, the collective conversion of the atomic spin excitation into anti-Stokes photons can introduce uncontrollable phases \( \varphi_{\text{random}} \) due to, for example, thermal motion of the atoms, resulting in decoherence in the state (3). Here, we suggest an experimental test to verify that coherence is preserved during the conversion process.

The single anti-Stokes photon delocalized in multi-time-bins was sent through a Franson-type interferometer with a phase shifter \( \theta \) inserted in one of the arms (figure 3). For simplicity, we considered the case when all readout pulses in equation (6) are equally separated in time. Then, the length difference of long and short arms should match the
time separation $\tau$ between two consecutive readout pulses $L_{\text{long}} - L_{\text{short}} = \epsilon \tau$ so that the outputs of the beam-splitter BS2 could interfere by observing signals on the detectors D1 and D2, showing a fringe pattern in dependence on $\theta$. As the visibility of the pattern is sensitive to random phases acquired by anti-Stokes temporal modes, this allows testing the coherence of multi-pulse conversion of the atomic spin excitation. To show this, we calculated the number of anti-Stokes photons measured by D1 and D2 detectors within a time interval much longer than the photon total duration

$$n_{\text{AS}}^{(i)} = \int_{-\infty}^{\infty} \mathrm{d}t \langle \hat{\Psi}_i(t) \hat{\Psi}_j(t) \rangle, \quad i = 1, 2, \tag{20}$$

where $\langle \hat{\Psi}_i(t) \rangle$, $i = 1, 2$, is the anti-Stokes photon state at $i$-th output port of the beam-splitter BS2. Omitting the vacuum part, which does not contribute to equation (20), these states are given by

$$\hat{\Psi}_{1,2} = \frac{1}{2} \left( e^{i\Phi} \hat{\Psi}_{1,2}^{(\text{AS})} \pm e^{-i\Phi} \hat{\Psi}_{1,2}^{(\text{AS})} \right), \tag{21}$$

where $\hat{\Psi}_{1,2}^{(\text{AS})}$ are defined in equation (19) with $j$-th mode single-photon states

$$\hat{\Psi}_j(t) = \int_{-\infty}^{\infty} \hat{\Omega}_j(t - t_j) \hat{\Psi}^\dagger_j(t) \mathrm{d}t, \tag{22}$$

and

$$\hat{\Psi}_j(t) = \int_{-\infty}^{\infty} \hat{\Omega}_j(t - t_j) \hat{\Psi}^\dagger_j(t) \mathrm{d}t, \tag{23}$$

respectively. Here, the real functions $\Phi_j(t - t_j)$ form an orthonormal set of temporal modes localized around $t = t_j$:

$$\int_{-\infty}^{\infty} \Phi_j(t - t_j) \Phi_k(t - t_k) \mathrm{d}t = \delta_{jk}. \tag{24}$$

Substituting equations (21)–(23) into equation (20) and using $\hat{\Psi}(t) = \sum_{j=1}^{J} \hat{\Psi}_j(t)$,

$$n_{\text{AS}}^{(1,2)} = \frac{1}{2} \left[ 1 \pm \sum_{j=1}^{J} \cos(\theta + \Delta \phi_{j,k}) C_i \left| C_k \right| \right] \times \int_{-\infty}^{\infty} \Phi_j(t - t_j) \Phi_k(t - t_k - \tau) \mathrm{d}t. \tag{25}$$

is obtained where $\Delta \phi_{j,k}$ is the relative phase between the $j$ and $k$ temporal modes. From this equation, we recognized that the fringe pattern visibility depends on photon numbers $n_{j,k} = |C_i|\left| C_k \right|^2$ in $j$ and $k$ modes and on the integral overlap of these modes, which, according to equation (24), is nonzero only if $j = k + 1$. For our purposes, the simplest case of equal photon numbers $n_j = 1/J$ in all modes and identical mode functions $\Phi_j(t - t_j) \equiv \Phi(t - t_j), j = 1, \ldots, J$, shown as an example in figure 2(b), suffices to illustrate the phase coherence across all time bins. With these simplifications, we have

$$n_{\text{AS}}^{(1,2)} = \frac{1}{2} \left[ 1 \pm \frac{1}{J} \int_{-\infty}^{\infty} \cos \left( \theta + \Delta \phi_{j,k+1} \right) \right]. \tag{26}$$

As the unknown phases that would originate from the atomic spin-wave conversion are time-independent, the photon numbers averaged over $\phi_{\text{random}}$ distribution, for real Rabi frequencies $\Omega_j$ in equation (6), were finally obtained as

$$n_{\text{AS}}^{(1,2)} = \frac{1}{2} \left[ 1 \pm \frac{1}{J} \int_{-\infty}^{\infty} \cos \left( \theta + \phi_{\text{random}} \right) \right]. \tag{27}$$

In figure 4, we present $n_{\text{AS}}^{(1,2)}$ found numerically for Gaussian distribution of $\phi_{\text{random}}$ with zero mean value and different variances demonstrating the dependence of interference contrast on phase scattering. The more this variation, the smaller the visibility. Therefore, the proposed method to observe the interference between temporal modes with maximal visibility $(J - 1)/J$ can be used to test the coherence of multi-time-bins entanglement of a single anti-Stokes photon, which is deterministically generated from a stored single atomic spin-excitation.
4. Conclusions

In conclusion, we have proposed a highly efficient heralded source of anti-Stokes single-photons entangled in multiple temporal modes. The source is based on the heralded creation of one atomic spin excitation followed by deterministic conversion of the latter into a single anti-Stokes photon that is delocalized in multi-time-bins. With experimentally verified heralded creation of a single atomic spin excitation, the source clearly provides high purity of single-photon states. The waveforms of anti-Stokes temporal modes are controlled by the shape of read laser pulses, while the phase coherence across all time bins can be experimentally verified by the suggested mechanism. This controlled scheme can be used first of all for implementation of quantum repeaters based on time-bin entangled single-photon states.

Acknowledgments

This research was conducted in the scope of European Union Seventh Framework Programme Grants No. GA-295025-IPERA and No.609534-SECURE-R2I, and ANSEF Grant PS-opt 3201. We acknowledge additional support from the International Associated Laboratory (CNRS-France and SCS Armenia) IRMAS.

References

[1] Gisin N, Ribordy G, Tittel W and Zbinden H 2002 Rev. Mod. Phys. 74 145
[2] Takesue H, Harada K-I, Tamaki K, Fukuda H, Tsuchizawa T, Watanabe T, Yamada K and Itabashi S-I 2010 Opt. Express 18 16777
[3] Humphreys P C, Metcalf B J, Spring J B, Moore M, Jin X-M, Barbieri M, Kolthammer W S and Walmsley I A 2013 Phys. Rev. Lett. 111 150501
[4] Knill E, LaFlamme R and Milburn G J 2001 Nature 409 46
[5] Kok P, Munro W J, Nemoto K, Ralph T C, Dowling J P and Milburn G J 2007 Rev. Mod. Phys. 79 135
[6] Brendel J, Gisin N, Tittel W and Zbinden H 1999 Phys. Rev. Lett. 82 2594
[7] Zavatta A, D’Angelo M, Parigi V, Bellini M 2006 Phys. Rev. Lett. 96 020502
D’Angelo M, Zavatta A, Parigi V and Bellini M 2006 Phys. Rev. A 74 052114
[8] Rossi A, Vallone G, De Martini F and Mataloni P 2008 Phys. Rev. A 78 012345
[9] Takeda S, Mizuta T, Fuwa M, Yoshikawa J I, Yonezawa H and Furusawa A 2013 Phys. Rev. A 87 043803
[10] Novikova I, Phillips N B and Gorshkov A V 2008 Phys. Rev. A 78 021802
[11] Reim K F et al 2012 Phys. Rev. Lett. 108 263602
[12] Takesue H 2006 Opt. Express 14 3453
[13] Sisakyan N and Malakyan Yu 2007 Phys. Rev. A 75 063831
[14] Pathak P K and Hughes S 2011 Phys. Rev. B 83 245301
[15] Vasilev G S, Ljunggren D and Kahn A 2010 New J. Phys. 12 063024
[16] Duan L M, Lukin M D, Cirac J I and Zoller P 2001 Nature 414 413
[17] Balic V, Braje D A, Kolchin P, Yin G Y and S E Harris S E 2005 Phys. Rev. Lett. 94 183601
[18] Mendes M S, Saldanha P L, Tabosa J W R and Felinto D 2013 New J. Phys. 15 075030
[19] Polyakov S V, Chou C W, Felinto D and Kimble H J 2004 Phys. Rev. Lett. 93 263601
[20] Chou C W, Polyakov S V, Kuzmich A and Kimble H J 2004 Phys. Rev. Lett. 92 213601
[21] Matsukevich D, Chanelière T, Jenkins S, Lan S-Y, Kennedy T and Kuzmich A 2006 Phys. Rev. Lett. 97 013601
[22] Sisakyan N and Malakyan Yu 2005 Phys. Rev. A 72 043806
[23] Petrosyan Sh and Malakyan Yu 2013 Phys. Rev. A 88 063817
[24] Bajcsy M, Hofferberth S, Balic V, Liang Q, Zibrov A S, Vuletic V, Zibrov A S, Vuletic V and Lukin M D 2009 Phys. Rev. Lett. 102 203902
[25] Bajcsy M, Hofferberth S, Peyronel T, Balic V, Liang Q, Zibrov A S, Vuletic V and Lukin M D 2011 Phys. Rev. A 83 063830
[26] Simon J, Tanji H, Thompson J K and Vuletic V 2007 Phys. Rev. Lett. 98 183601
[27] Eisaman M D, Fan J, Migdall A and Polyakov S V 2011 Rev. Sci. Instrum. 82 071101