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

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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.

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The research on generation of photon states entangled in multi-temporal modes is rapidly growing recently. This interest is due to two main reasons. First, this type of entanglement can be transferred over significantly large distances with very little decoherence [1, 2], which allows much more robust quantum communication systems in contrast to types of entanglement based on polarization-encoded qubits. Second, employment of time-bin entangled photons allows to perform linear optical quantum computing (LOQC) in a single spatial mode [3], providing scalable implementation of many-qubit protocols without creating unwieldy networks which inevitably arise in all schemes of LOQC [4, 5] due to many spatial modes.

Generation of optical pulses in distinct temporal modes has been studied experimentally in a variety of settings. The interferometric preparation of time-bin qubits of broadband photons generated in non-linear crystals from a spontaneous parametric down conversion (SPDC) has been reported in Refs. [6, 7, 8, 9]. The conversion of weak signal pulses into multi-temporal modes in room-temperature vapors has been shown under electromagnetic induced transparency conditions [10] and far-off resonant Raman schemes [11]. The generation of time-bin entangled photon pairs in the 1.5μ m band via spontaneous four-wave mixing in a cooled fiber is demonstrated in [12]. There exist different models to realize such photonic states based, for example, on the parametric interaction between two single-photon pulses in a coherent atomic ensemble [13], stimulated Raman adiabatic passage in a single quantum dot [14] or in an atom-cavity system [15]. However, each of these methods has certain disadvantages impeding their use as efficient sources of time-bin entangled photons. The good source should provide: i) a pure single-photon (SP) state without mixture from both the multiphoton and zero-photon states; ii) well separated quantum temporal modes such that one can perform spacelike separated local measurements on the modes; iii) controllable amplitudes of the temporal modes, and iv) phase coherence across all time bins. The schemes proposed so far 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 the main challenges. Besides, 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. Also, 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, which is a promising candidate for highquality 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 the probability one. This is achieved due to the fiber enhanced atom-photon interaction and multiatom collective interference effects [16]. As a result, the anti-Stokes photon is emitted in a well-defined spatial mode being 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 DLCZ scheme [17, 18].

The main limitation of our scheme is the low heralding efficiency due to low probability for Stokes-photon emission needed to exclude the multi-atom events in the collective spin excitation. The 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 in order 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 have been shown in Ref. [19]. After a single collective atomic spin excitation is heralded our system operates as a deterministic source of anti-Stokes single-photons entangled in multi-time-bins.

Our model is described more detailed in the next Section, where 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 are concluded 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). We consider the atoms are initially prepared in the state $|1\rangle$ by optical pumping and are strongly confined in the transverse direction inside the fiber core that prevents atom-wall collisions [24]. The write ω_W laser field with peak Rabi frequency Ω_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 $\Delta_W = \omega_{31} - \omega_W$ that makes the system immune to the 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 was successfully detected by the D1:D2 block in Figure 1, a train of read ω_R laser pulses is applied at the transition $2 \rightarrow 4$ in a far off-resonant Raman configuration $\Delta_R = \omega_{42} - \omega_R \neq 0$ 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, which are easily controlled being proportional to the profiles of read laser subpulses (see Eq.(12)). It should be noted that four level scheme is more general in the sense that it allows to adjust the anti-Stokes photon frequency, at the same time this does not complicate the calculations.



Figure 1. (Color online) Generation of Stokes and anti-Stokes photons by a write W (green) and read R (brown) pulses. Setup and atomic level scheme are in the inset). The case of two read pulses and, correspondingly, the delocalization of anti-Stokes photon in two possible modes (blue filled) is shown. An ensemble of cold atoms is trapped inside a HC-PCF of length L and diameter D. The Stokes photon is detected with photon counters D1 and D2.

We consider a low-finesse cavity with damping rate $\chi = c/L$, where Stokes and anti-Stokes cavity fields at frequencies ω_1 and ω_2 are expressed, respectively, in terms of annihilation (creation) operators $\hat{a}_S(\hat{a}_S^{\dagger})$ and $\hat{a}_{AS}(\hat{a}_{AS}^{\dagger})$ as follows:

$$E_i^{(+)}(z,t) = \left(\frac{2\pi\hbar\omega_i}{V}\right)^{1/2} \hat{a}_i \exp(ik_i z - i\omega_i t), \ i = S, AS,$$
(1)

with the quantization volume V equal to the interaction volume $V = \pi D^2 L/4$. Owing to the large detunings $\Delta_{W,R}$ we adiabatically eliminate the upper states $|3\rangle$ and $|4\rangle$, which are different generally, and then, in the rotating frame approximation, the interaction Hamiltonian for the total system in terms of the atomic collective spin operators

$$S^{\dagger} = \frac{1}{\sqrt{N}} \sum_{i=1}^{N} \sigma_{21}^{(i)}, \quad S = (S^{\dagger})^{\dagger}$$
(2)

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is written as [22]

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

$$\tag{3}$$

In Eq.(2) the summation is taken over all atoms, $\sigma_{\alpha\beta}^{(i)} = |\alpha\rangle_i \langle \beta|$ is the atomic spin-flip operator in the basis of the two ground states $|1\rangle$ and $|2\rangle$ for the *i*-th atom and

$$G(t) = g_S \frac{\Omega_W}{\Delta_W} f_W(t), \tag{4}$$

$$F(t) = g_{AS} \frac{\Omega_R(t)}{\Delta_R},\tag{5}$$

where $f_W(t)$ is the profile of the write pulse, while $\Omega_R(t)$ is defined as

$$\Omega_R(t) = \sum_{i=1}^J \Omega_i f_i(t - t_i), \tag{6}$$

showing that the J well-separated readout pulses with peak Rabi frequencies Ω_i are localized at time moments $t_J > t_{J-1} \dots > t_1$ with temporal profiles $f_i(t-t_i)$ and relative delay between them much larger than their lengths T_i . Hereafter, for simplicity, we consider the Rabi frequency of the write pulse real. In Eqs.(3),(4) the atom-photon coupling constants are given by

$$g_S = \left(\frac{2\pi\omega_S}{V}\right)^{1/2} \mu_{32}, \quad g_{AS} = \left(\frac{2\pi\omega_{AS}}{V}\right)^{1/2} \mu_{41},$$
 (7)

with μ_{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 as 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

$$\frac{dn_i}{dt} = \langle \hat{a}_{i,\text{out}}^{\dagger}(t)\hat{a}_{i,\text{out}}(t)\rangle, \quad i = S, AS,$$
(8)

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 $\hat{a}_{i,\text{out}}(t) = \hat{a}_{i,\text{in}}(t) + \sqrt{2\chi}\hat{a}_i(t)$ and satisfies the commutation relation $[a_{i,\text{out}}(t), a_{i,\text{out}}^{\dagger}(t')] = [a_{i,\text{in}}(t), a_{i,\text{in}}^{\dagger}(t')] = \delta(t - t')$. 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 $\chi \gg g_{S,AS}$ leads to the following links between the field and atomic operators

$$\hat{a}_S(t) = -i\sqrt{N} \frac{G(t)}{\chi} S^{\dagger}(t) - \sqrt{\frac{2}{\chi}} \hat{a}_{S,\text{in}}(t), \qquad (9)$$

$$\hat{a}_{AS}(t) = -i\sqrt{N}\frac{F(t)}{\chi}S(t) - \sqrt{\frac{2}{\chi}}\hat{a}_{AS,\text{in}}(t), \qquad (10)$$

substitution of which into Eq.(8) with input-output relation yields for the photon fluxes

$$\frac{dn_S}{dt} = \alpha(t)[N_{\rm Sp}(t) + 1],\tag{11}$$

$$\frac{dn_{AS}}{dt} = \beta(t)N_{\rm Sp}(t),\tag{12}$$

where

$$\alpha(t) = \frac{2N}{\chi} G^2(t), \tag{13}$$

$$\beta(t) = \frac{2N}{\chi} \mid F(t) \mid^2 \tag{14}$$

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

$$N_{\rm sp}(t) = \int_{-\infty}^{t} dt' \alpha(t') \exp \int_{t'}^{t} d\tau [\alpha(\tau) - \beta(\tau) - \Gamma_{\rm tot}(\tau)]$$
(15)

Here the total relaxation $\Gamma_{\text{tot}}(\tau) = \gamma_c + \Gamma_W(t) + \Gamma_R(t)$ comprises the decay rate γ_c of atomic ground states coherence and the rates of optical pumping between the states 1 and 2 via write and read pulses

$$\Gamma_W(t) = \frac{\Omega_W^2}{\Delta_W^2} f_W^2(t) \gamma_{32}, \quad \Gamma_R(t) = \frac{|\Omega_R(t)|^2}{\Delta_R^2} \gamma_{41}$$
(16)

where γ_{ij} is a partial decay rate of upper level *i* to the state *j* giving in sum the spontaneous decay rate of upper states $\gamma = \gamma_i = \sum_j \gamma_{ij}$.

There are two important consequences coming out from Eqs.(11),(12) and (15). First, for the large signal-to-noise ratio $\alpha/\Gamma_W \gg 1$ and $\beta/\Gamma_R \gg 1$, the relaxation terms in Eq.(15) can be neglected. Then, the photon numbers are readily found from Eqs.(11),(12) and (15) to be

$$n_S(t) = N_{\rm Sp}(t) = e^{\int_{-\infty}^t d\tau \alpha(\tau)} - 1, \quad t \le T_W \tag{17}$$

$$n_{AS}(t) = n_S(\infty)(1 - e^{-\int_{-\infty}^t d\tau\beta(\tau)}), \quad t \ge T_W + \tau_D$$
(18)

showing that, on the one hand, the number of detected Stokes photons equals to the number of spin excitations stored in the atomic medium during the write pulse of duration T_W and, on the other hand, it unambiguously determines the number of anti-Stokes photons retrieved from the medium at the end of the read subpulses. Here we have used that the time delay τ_D between the read and write laser pulses is large compared to T_W and, at the same time, it is much shorter than the spin decoherence time γ_c^{-1} .

Second, taking into account the orthogonality of the functions $f_i(t - t_i)$ in Eq.(6), the Eq.(12) describes the deterministic generation of the anti-Stokes photon entangled in nonoverlapping 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 ⁸⁷Rb atoms with the ground states $5S_{1/2}(F = 1, 2)$ and excited states $5P_{1/2}(F = 2)$ and $5P_{3/2}(F = 2)$ as the atomic states

1, 2, and 3,4 in Figure 1, respectively. Number of atoms confined in a hollow-core fiber of the length $L \sim 3$ cm and diameter $D \sim 5\mu$ m is about $N \sim 10^4$ [25], the fields are tuned far from the one-photon resonance by $\Delta_{W,R} = 20\gamma$. Below γ_c is neglected. The durations of the write and read subpulses in Eq.(6) are taken $T_W \sim T_i \sim 1\mu$ 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 photon 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 one they are redesigned such that the temporal modes of the anti-stokes photon 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 in average only one Stokes photon is generated: $n_S(\infty) \sim 1$ or, equivalently from Eq.(17), $\int_{-\infty}^{\infty} d\tau \alpha(\tau) \sim 0.7$.



Figure 2. (Color online) Anti-Stokes photon flux as a function of time in units of γ 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). Black dotted line shows the Stokes pulse of 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.



Figure 3. (Color online) Schematic setup of coherence detection between consecutive temporal modes of anti-Stokes field. Passing the long (L) and short (S) arms of the interferometer the anti-Stokes photon interferes with itself in observing signals on the detectors D1 and D2. τ is the time separation between the modes, $|\Psi_{1,2}\rangle$ are the output states of the ani-Stokes photon, M- mirrors, BS - beam splitters, θ - phase shifter.

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

$$|\Psi_{AS}\rangle = \sum_{j=1}^{J} C_j |1\rangle_j \prod_{i \neq j}^{J} |0\rangle_i,$$
(19)

where $|1\rangle_j$ and $|0\rangle_j$ denote Fock states with zero and one photon, respectively, at the time t_j . Here the complex amplitudes C_i 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 because 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 $\varphi_{\rm random}$ due to, for example, the thermal motion of the atoms, resulting in decoherence in the state (19). Here we suggest an experimental test to verify that the coherence is preserved during the conversion process.

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

detectors within a time interval much longer than the photon total duration

$$n_{AS}^{(i)} = \int_{-\infty}^{\infty} dt \langle \Psi_i \mid \hat{a}^{\dagger}(t)\hat{a}(t) \mid \Psi_i \rangle, \quad i = 1, 2,$$
(20)

where $| \Psi_i \rangle$, i = 1, 2, is the ani-Stokes photon state at *i*-th output port of the beamsplitter *BS2*. Omitting the vacuum part, which does not contribute to Eq. (20), these states are given by

$$|\Psi_{1,2}\rangle = \frac{1}{2} (e^{i\theta} |\Psi_{AS}^{(s)}\rangle \pm |\Psi_{AS}^{(l)}\rangle), \qquad (21)$$

where $|\Psi_{AS}^{(s,l)}\rangle$ are defined in Eq. (19) with *j*-th mode single-photon states

$$|1^{(s)}\rangle_j = \int_{-\infty}^{\infty} \Phi_j(t-t_j)\hat{a}_j^{\dagger}(t) \mid 0\rangle dt, \qquad (22)$$

$$|1^{(l)}\rangle_j = \int_{-\infty}^{\infty} \Phi_j (t - t_j - \tau) \hat{a}_j^{\dagger}(t) \mid 0\rangle dt, \qquad (23)$$

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

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

Substituting Eqs.(21) and (22), (23) into Eq.(20) and using $\hat{a}(t) = \sum_{j=1}^{J} \hat{a}_j(t)$ one obtains

$$n_{AS}^{(1,2)} = \frac{1}{2} \Big[1 \pm \sum_{j,k}^{J} \cos(\theta + \Delta \varphi_{j,k}) \mid C_j \mid \mid C_k \mid \\ \times \int_{-\infty}^{\infty} \Phi_j (t - t_j) \Phi_k (t - t_k - \tau) dt \Big],$$
(25)

where $\Delta \varphi_{j,k}$ is the relative phase between the j and k temporal modes. From this equation we recognize that the fringe pattern visibility depends on photon numbers $n_{i,k} = |C_{i,k}|^2$ in j and k modes and on the integral overlap of these modes which, according to Eq.(24), is finite 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 $\Phi_j(t - t_j) \equiv \Phi(t - t_j), j = 1, ..., J$, shown as an example in Figure 2b, suffices to illustrate the phase coherence across all time bins. With these simplifications, we have

$$n_{AS}^{(1,2)} = \frac{1}{2} \left[1 \pm \frac{1}{J} \sum_{k}^{J-1} \cos(\theta + \Delta \varphi_{k+1,k}) \right].$$
(26)

As the unknown phases that would originate from the atomic spin-wave conversion are time-independent, the photon numbers averaged over φ_{random} distribution, for real Rabi frequencies Ω_i in Eq.(6), are finally obtained as

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



Figure 4. (Color online) Averaged anti-Stokes photon number $\overline{n_{AS}^{(1)}}$ as a function of θ for three different variances of φ_{random} : 0 (blue solid line), 1.5 (red dotted line), 3 (green dashed line). Black dash-dotted line shows $\overline{n_{AS}^{(2)}}$ for zero variance.

In Figure 4 we present $\overline{n_{AS}^{(1,2)}}$ found numerically for Gaussian distribution of φ_{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 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. Such controlled scheme can be used first of all for implementation of quantum repeaters based on time-bin entangled single-photon states.

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