

Generation Engineering of Heralded Narrowband Colour Entangled States.

A. Zavatta

*Istituto Nazionale di Ottica (CNR-INO) Largo E. Fermi, 6 - 50125 Firenze, Italy. and
Department of Physics and Astronomy, Università di Firenze, Italy.**

M. Artoni

*Department of Engineering and Information Technology, Brescia University, Brescia, Italy.
European Laboratory for Nonlinear Spectroscopy, Firenze, Italy. and
Istituto Nazionale di Ottica (CNR-INO) Largo E. Fermi, 6 - 50125 Firenze, Italy.*

G. La Rocca

Scuola Normale Superiore and CNISM, Pisa, Italy.

(Dated: December 22, 2018)

Efficient generation of entanglement is an essential requirement for quantum communications; however, long distances can only be achieved utilising entangled states that can be efficiently mapped into matter. Hence, sources generating states with bandwidths naturally compatible with linewidths of atomic transitions are crucial. We harness the indistinguishability between two spontaneous four-wave mixing processes to achieve the heralded generation of single-photon frequency-bin entangled states. State manipulation admits entanglement and generation probability optimizations yet with negligible absorption. The scheme could also be adapted to photonic and solid interfaces.

Quantum states of light are crucial for quantum communications as well as for all emerging quantum technologies. The propagation of quantum states through optical channels is however dramatically affected by losses making quantum networks a hard task to accomplish. In spite of this, the seminal paper of Duan *et al.* [1] proposed long distance quantum communications through the entanglement of distant atomic ensembles. Since then, a growing attention has been devoted towards the development of narrowband sources of quantum states of light to efficiently map light into matter. Recently, several generation schemes based on spontaneous four-wave mixing and Raman processes in atomic ensembles have been proposed and demonstrated such as, *e.g.*, entangled photon pairs generated in trapped cold atoms [2–5] and in hot atoms vapor cells [6]. In such schemes, the quantum states can be generated with bandwidths naturally compatible with the atomic transitions, allowing a more efficient mapping of light into matter. Unfortunately, such processes are probabilistic and the generation happens at random time with very low probability. In order to circumvent this limitation, so called heralded schemes, where desired output states are announced by ancillary photons, are required to permit both further processing into quantum communication networks and quantum state characterization [7].

Within this context, it is convenient to exploit the multilevel structure of a resonant media such as atoms or atom-like systems [8–10], in order to engineer the generation of quantum states in a frequency multimode scenario [11]. In this Letter, we propose a scheme for the heralded generation of single-photon color entangled states *i.e.*, states where a single photon excitation is simultaneously shared by two distinct frequency modes

of light, exploiting indistinguishability between four-wave mixing processes. We show that narrowband colour entangled states can be engineered and efficiently generated only if some specific conditions between detunings and Rabi frequencies of pump and coupling fields are met. Propagation [12] and mapping [13] onto a quantum memory of color-entangled states through a specific atomic interface has recently been studied hence the present work represents an additional building block toward the realization of a quantum network, in which colour-entanglement is generated, manipulated and stored.

The underlying physical mechanism hinges on a four-photon spontaneous four-wave mixing process in a three-level third-order nonlinear medium where, in the presence of a pair of *weak* coupling (ω_c) and pump (ω_p) co-propagating beams, Stokes and anti-Stokes photon pairs emerge (Fig.1). The two processes, one (*A*) leading to the emission of a *Stokes* (ω_s) and an *anti-Stokes* (ω_{as}) photon pair and the other (*B*) leading to the emission of the same *Stokes* (ω_s) yet a different *anti-Stokes* ($\omega_{as'}$) photon are sketched in Fig. 1 (*blue and red lines*) and governed by the following energy conservation laws $\omega_s + \omega_{as} = \omega_p + \omega_c$ and $\omega_s + \omega_{as'} = 2\omega_p$, respectively. The main feature of the scheme is that under suitable pump and coupling conditions (*i*) the probability that the two processes *A* and *B* occur simultaneously can be made to be negligible while (*ii*) the generation of anti-Stokes photons either through process *A* or through process *B* can be made to occur with the same probability. Hence the detection of a single-photon at frequency ω_s will entail, via a state-projection measurement (Fig. 2), the emission of an anti-Stokes photon though its detection does not reveal, even in principle, at which frequency *i.e.* ω_{as} or $\omega_{as'}$ has been emitted. Such an indistinguishability results in the state

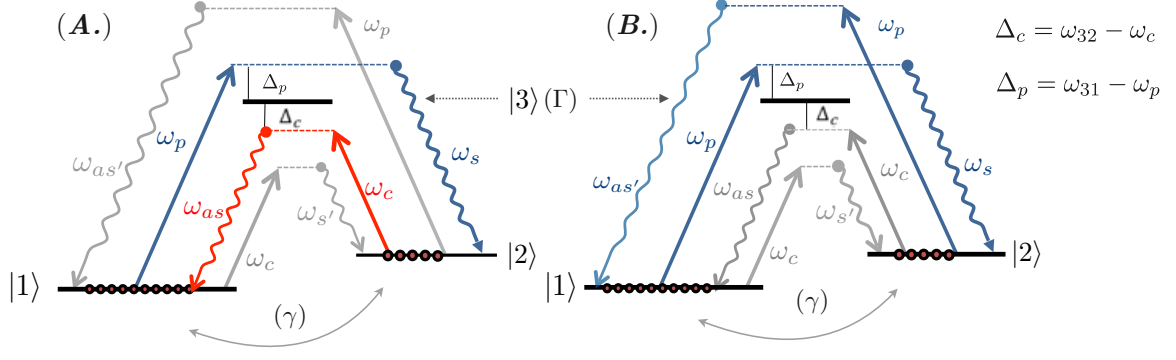


FIG. 1. **Single-photon color entanglement.** [A.] A pair of weak coupling (ω_c) and pump (ω_p) beams spontaneously scatter through the third-order nonlinearity of a three-level atom to generate an *anti-Stokes* photon ω_{as} (red) and a *Stokes* photon ω_s (blue). [B.] The different pair $\{\omega_{as'}, \omega_s\}$ of photons can be generated through spontaneous scattering off the pump (ω_p) alone. The spontaneous four-wave mixing processes A. and B. result in a small probability of generating single anti-Stokes photons at frequencies ω_{as} and $\omega_{as'}$. For certain atomic parameters ($\gamma, \Gamma, N/V$) and (driving) pump and coupling detunings ($\Delta_{p,c}$) and Rabi frequencies ($\Omega_{p,c}$) (see Appendix), the emission of one single-photon through channels A. and B. can be made to be *undistinguishable* subject to a state-projection measurement on the Stokes (ω_s) photon (Fig. 2). The sample has a density $N/V \simeq 5 \times 10^{12} \text{cm}^{-3}$, length $L = 100 \mu\text{m}$, while atomic levels correspond to the transition $5^2S_{1/2} \rightarrow 5^2P_{1/2}$ (^{85}Rb D_1 line) with $\lambda_{31} = 795 \text{nm}$, $\omega_{21} = 2\pi \times 3 \text{GHz}$ and decay rates $\Gamma = 2\pi \times 5.75 \text{MHz}$ and $\gamma = 2\pi \times 10 \text{kHz}$.

superposition,

$$\alpha |1\rangle_{\omega_{as}} |0\rangle_{\omega_{as'}} + \beta |0\rangle_{\omega_{as}} |1\rangle_{\omega_{as'}}. \quad (1)$$

It turns out that α and β essentially depend on the medium linear and nonlinear optical response hence directly tunable through the pump and coupling driving fields. This state may be envisaged as the frequency-entanglement counterpart of the familiar single-photon path entanglement [14, 15], or time-bin entanglement [16] used as important resources in quantum protocols to convey and process quantum information [17–20].

Two more processes will clearly contribute to the heralded generation of single-photon state (1), namely those involving the spontaneous generation of the different

Stokes-anti-Stokes pairs $\{\omega_{s'}, \omega_{as'}\}$ (C) and $\{\omega_{s'}, \omega_{as}\}$ (D) also sketched in Fig. 1 (gray lines). However the projection procedure above will prevent the processes {C,D} from concurring to the generation of the state (1) [21].

Entanglement generation. Assuming classical fields in the form of plane-waves $E_i^+ = E_i e^{i\mathbf{k}_i \cdot \mathbf{r} - i\omega_i t}$ with $i = \{c, p\}$, respectively for the control and pump beams, and standard field operator

$$\hat{E}_j^+ = \frac{1}{\sqrt{\pi}} \int d\omega \sqrt{\frac{2\hbar\omega}{c\epsilon_0 A}} e^{i\mathbf{k}_j(\omega) \cdot \mathbf{r} - i\omega t} \hat{a}_j(\omega) \quad (2)$$

with $j = \{s, as\}$, for the Stokes and anti-Stokes photons propagating with a (complex) wave-vector $\mathbf{k}_j(\omega)$ [22–24], the effective Hamiltonian describing the photon-atom interaction can be written as,

$$\hat{H}_I = \frac{\epsilon_0 A}{4} \int_{-L/2}^{L/2} dz [\chi_A^{(3)} E_p^+ E_c^+ \hat{E}_{as}^- \hat{E}_s^- + \chi_B^{(3)} E_p^+ E_p^+ \hat{E}_{as'}^- \hat{E}_s^- + \chi_C^{(3)} E_c^+ E_p^+ \hat{E}_{as'}^- \hat{E}_{s'}^- + \chi_D^{(3)} E_c^+ E_c^+ \hat{E}_{as}^- \hat{E}_{s'}^-] + h.c., \quad (3)$$

with the fields (negative) frequency parts E_i^- and \hat{E}_j^- computed as usual [22] and with space-time dependencies purposely omitted here. **Here, we have assumed that control and pump fields have a fixed relative phase-relationship that can be arbitrarily controlled and tuned.**

The four third-order optical nonlinear susceptibilities $\chi_l^{(3)}$ for $l = \{A, B, C, D\}$ in (3) correspond to the four spontaneous nonlinear mixing processes triggered by cou-

pling and pump. Details on the susceptibility expressions are given in the Appendix. For the sake of clarity we restrict to a nearly 1D generation geometry with Stokes and Anti-Stokes photons emitted in the z -direction and with a fixed (transverse) mode-profile cross-section A across the interaction region L . We further assume that the Stokes and the anti-Stokes are initially in the vacuum $|0\rangle_s |0\rangle_{s'} |0\rangle_{as} |0\rangle_{as'} \rightarrow |0\rangle$, so that in the weak spontaneous scattering limit [25] one has for the output state,

$$\begin{aligned}
|\Psi\rangle_{out} &\simeq \left(\mathbf{1} - \frac{i}{\hbar} \int_{-\infty}^{\infty} dt \hat{H}_I \right) |0\rangle \\
&= |0\rangle + \int d\omega \left[f_A(\omega_c + \omega_p - \omega, \omega) \hat{a}_{as}^\dagger(\omega_c + \omega_p - \omega) \hat{a}_s^\dagger(\omega) + f_B(2\omega_p - \omega, \omega) \hat{a}_{as'}^\dagger(2\omega_p - \omega) \hat{a}_s^\dagger(\omega) \right. \\
&\quad \left. + f_C(\omega_p + \omega_c - \omega, \omega) \hat{a}_{as'}^\dagger(\omega_p + \omega_c - \omega) \hat{a}_s^\dagger(\omega) + f_D(2\omega_c - \omega, \omega) \hat{a}_{as}^\dagger(2\omega_c - \omega) \hat{a}_s^\dagger(\omega) \right] |0\rangle_s |0\rangle_{s'} |0\rangle_{as} |0\rangle_{as'}. \quad (4)
\end{aligned}$$

Although states with more than one photon per frequency mode are possible (higher order processes), they are nevertheless unlikely owing to small nonlinearities of the weak spontaneous scattering [25] we examine here (4) is the generated state to a very good approximation.

The four “two-photon” states are generated, in general, with *different* amplitude probabilities f_l for each of the four terms $l = \{A, B, C, D\}$ (Fig. 1A). The generation of the pair $\{\omega_s, \omega_{as}\}$, *e.g.*, occurs with the probability amplitude,

$$f_A(\omega, \omega') = -i \frac{\sqrt{\omega\omega'}}{4\pi c} \chi_A^{(3)}(\omega, \omega') E_p E_c \text{sinc} \left(\frac{\Delta k_A L}{2} \right) L, \quad (5)$$

which depends on the electric field amplitudes (E_p, E_c) directly and through the susceptibility $\chi_A^{(3)}(\omega, \omega')$, and on the z -projection $\Delta k_A = (\mathbf{k}_{as} + \mathbf{k}_s - \mathbf{k}_p - \mathbf{k}_c) \cdot \hat{z}$ of the wavevector mismatch (momentum-conservation) for co-propagating pump and coupling beams. We restrict here to collecting Stokes and anti-Stokes photons pairs that are emitted with a very small angle with respect to the z -direction. Therefore, the phase-mismatch is given by:

$$\Delta k_A = \frac{\omega_{as}}{c} n_{13}(\omega_{as}) + \frac{\omega_s}{c} n_{23}(\omega_s) - \frac{\omega_p}{c} n_{13}(\omega_p) - \frac{\omega_c}{c} n_{23}(\omega_c) \quad (6)$$

where $n_{ij}(\omega) \approx 1 + \text{Re}[\chi_{ij}(\omega)]/2$ is the index of refraction calculated from the linear susceptibility $\chi_{ij}(\omega)$ associated with the $|i\rangle - |j\rangle$ two-level transition (see Appendix).

The probabilities for the three other processes B, C, D are instead obtained exchanging $c \rightarrow p$ and $as \rightarrow as'$ [to obtain $f_B(\omega, \omega')$], $c \leftrightarrow p$, $s \rightarrow s'$ and $as \rightarrow as'$ [$f_C(\omega, \omega')$], and $p \rightarrow c$ and $s \rightarrow s'$ [$f_D(\omega, \omega')$], while the same applies when computing the other mismatches $\Delta k_{B,C,D}$. It follows from (4) that the detection of a Stokes photon at frequency ω_s (Fig. 2) projects $|\Psi\rangle_{out}$ into a superposition of anti-Stokes single-photon with frequency ω_{as} and $\omega_{as'}$,

$$\begin{aligned}
|\Psi\rangle_p &= \left[{}_s\langle 0 | \hat{a}_s(\omega_s) \right] |\Psi\rangle_{out} = \\
&f_A(\omega_{as}, \omega_s) |1\rangle_{as} |0\rangle_{as'} + f_B(\omega_{as'}, \omega_s) |0\rangle_{as} |1\rangle_{as'} \quad (7)
\end{aligned}$$

which corresponds to the state sought for in (1) with (complex) coefficients $\alpha \rightarrow f_A(\omega_{as}, \omega_s)/\sqrt{\mathcal{N}}$ and $\beta \rightarrow f_B(\omega_{as'}, \omega_s)/\sqrt{\mathcal{N}}$, being $\mathcal{N} = |f_A|^2 + |f_B|^2$ (normalization). Here $|1\rangle_i = \hat{a}^\dagger(\omega_i)|0\rangle$ denotes a single-photon state at the frequency ω_i with $i = \{as, as', s, s'\}$. The degree of entanglement in $|\Psi\rangle_p$, which depends through f_A and f_B on the medium optical response (linear and nonlinear) and the wavevector mismatch can be all-optically

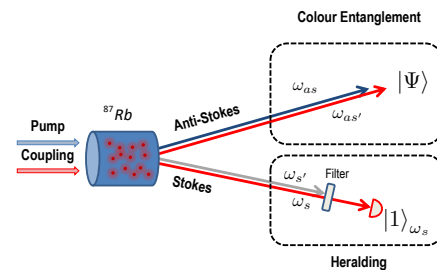


FIG. 2. **Stokes state projection.** Under specific pump and coupling drivings of a sample of cold ^{85}Rb atoms pairs of pump and coupling photons scatter via spontaneous four-wave mixing into pairs of Stokes and anti-Stokes photons $\{\omega_s, \omega_{as}\}$, $\{\omega_s, \omega_{as'}\}$ $\{\omega_{s'}, \omega_{as}\}$ and $\{\omega_{s'}, \omega_{as'}\}$. A “single” photon can be shared by two different modes ω_{as} and $\omega_{as'}$ if a Stokes photon ω_s is detected (*lower-box*). Such a conditional detection “heralds” the generation of the entangled state $|\Psi\rangle$ (*upper-box*).

controlled through pump and coupling (Fig. 1). It turns out in fact that we can span continuously from a *maximally* entangled state ($\alpha = \beta = 1/\sqrt{2}$) to a *pure* photon state [26] ($\alpha = 0$ or $\beta = 0$) directly through the Rabi frequencies $\Omega_{p,c}$ and detunings $\Delta_{p,c}$ [27]. We adopt here the negativity of the partial transpose (NPT) concept to quantify the degree of entanglement. This has been introduced in [28] and it is based on the Peres-Horodecki [29, 30] separability criterion thereby $NPT = 1$ corresponds to maximally entangled states and $NPT = 0$ to separable ones. For the specific state $|\Psi\rangle_p$ in (7) we have $NPT = 2|\alpha||\beta|$.

Entanglement engineering. We will focus in the following on three representative situations to illustrate how entanglement can be efficiently engineered. We

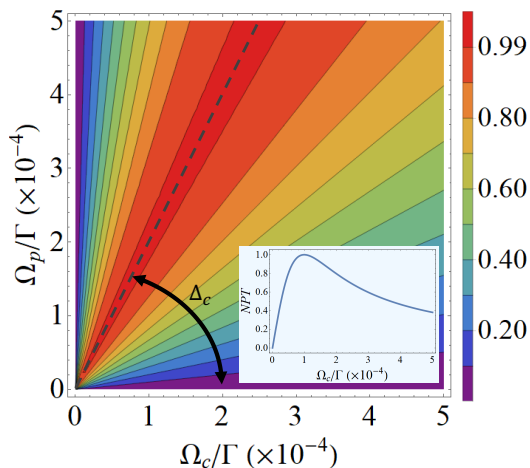


FIG. 3. **Raman-like regime.** Negativity of the partial transpose (NPT) for the state $|\Psi\rangle$ in (1) with $\Delta_p = -1$ GHz, $\Delta_c = 10$ GHz vs. varying pump and coupling intensities ($\propto \Omega_{p,c}$). For fixed driving intensities, maximum NPT values (*dashed line*) may be attained by tuning Δ_c . **Inset.** Plot section at $\Omega_p = 2 \times 10^{-4}\Gamma$: maximum entanglement is tuned through the coupling intensity ($\propto \Omega_c$).

choose a realistic set-up made of cold ^{85}Rb atoms [31], though we anticipate that the generation mechanism is general enough to be suited to non-atomic architectures as well [32–37].

We start considering the case of weak driving fields and both far detuned from resonance, *i.e.*, $\Omega_{p,c} \ll \sqrt{\gamma}\Gamma$ and $|\Delta_{p,c}| \gg \Gamma$. In these conditions, the indexes of refraction for all the frequency components are very close to unity, thus phase matching conditions are basically satisfied ($\Delta k_A, \Delta k_B \simeq 0$) over a wide range of frequencies (tens of Gigahertz) by energy conservation. This is reported in (Fig. 3) where a nearly balanced spontaneous Raman scattering regime with $NPT = 1$ over a parameters region for which $\Omega_c/\Delta_c \sim \Omega_p/\omega_{21}$. Maximum entanglement can then be directly tuned through the off-resonant coupling detuning (Δ_c) (*dash-line*), else through its intensity (Ω_c) (Fig. 3 inset), and can always be achieved at vanishing levels of absorption ($R \sim 10^{-4}$). The generation probability of the entangled state is the probability to find a single-photon state in modes ω_{as} and $\omega_{as'}$ and this is $\mathcal{P} = |\langle 1_{as} | \Psi \rangle_p|^2 + |\langle 1_{as'} | \Psi \rangle_p|^2 = |f_A|^2 + |f_B|^2$. In the spontaneous Raman regime (Fig. 3) we find values of NPT close to unity with probabilities $\mathcal{P} \sim 10^{-13}$ when background level populations are completely unbalanced ($\rho_{11}^0 \gg \rho_{22}^0$).

Anti-Stokes generation probability \mathcal{P} will improve *if* one or both driving fields are brought close to resonance though absorption at each anti-Stokes mode will become important [23]. This turns out to be an important generation regime which we will discuss below. A

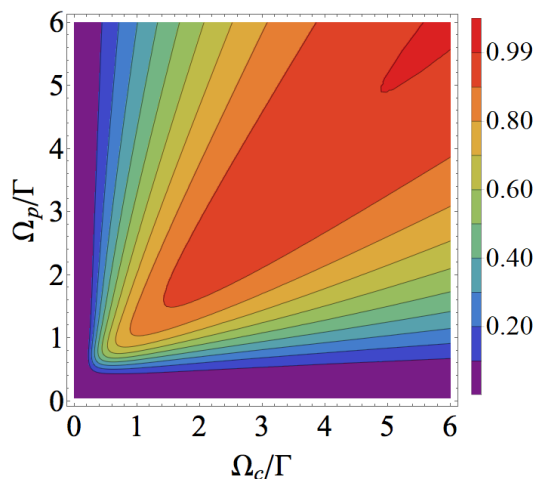


FIG. 4. **Near-resonance regime.** Same as in Fig. 3 with $\Delta_p = -\Gamma$, $\Delta_c = \Gamma$. The generation probability is almost constant $\mathcal{P} \sim 10^{-5}$ in the region in which $NPT > 0.9$, while increasing as NPT decreases. The anti-Stokes ω_{as} losses are constant with Ω_p yet varying with Ω_c from total absorption to $\lesssim 0.2\%$ ($\Omega_c = 6\Gamma$). Anti-Stokes $\omega_{as'}$ losses, constant with Ω_c , range from a $\sim 3\%$ max ($\Omega_p \simeq \Gamma$) to almost no loss at all ($\Omega_p = 6\Gamma$).

proper assessment of the negativity of the partial transpose function in the presence of absorption is in order, which is done now by adopting a standard beam splitter model [38]. According to this model, the dissipative process is formally described by means of a loss-less beam-splitter that mixes the ideal state with the vacuum, then the partial trace over the lossy channel returns the state subjected to losses. Thus, the lossy state appears to be noisy and attenuated by the reflection coefficient (R) of the beam splitter that accounts for the absorption.

We apply this losses model to each anti-Stokes mode with absorption $R(\omega)$, which is obtained from the imaginary part of the first order susceptibility $\chi_{13}(\omega)$ (see Appendix), $R(\omega) = 1 - \exp\{-\text{Im}[\chi_{13}(\omega)]\omega L/c\}$. The output state affected by absorption is then used to calculate the NPT . Hereafter, a NPT close to unity will then ensure the generation of a maximally entangled state with negligible absorption. We report in Fig. 4 the NPT behavior for small detunings $|\Delta_{p,c}| \sim \Gamma$ and driving fields such that $\sqrt{\gamma}\Gamma \lesssim \Omega_{p,c} \sim \Gamma$, showing large variations of the degree of entanglement along with losses and occurring with generation probabilities \mathcal{P} orders of magnitudes larger than in the Raman case. Maximal entanglement ($NPT \geq 0.99$) is restricted only to the high intensities region ($\Omega_{p,c} \sim 6\Gamma$) with probabilities $\mathcal{P} \sim 10^{-4}$ provided that $\rho_{11}^0 \gg \rho_{22}^0$, *i.e.*, most of the atomic background population (see Appendix) is in the lowest state $|1\rangle$. Losses, on the other hand, remain small around the high intensity region due to the *Autler-Townes* splitting and in the lower intensities region due to Electromagnetically Induced Transparency (EIT) [39].

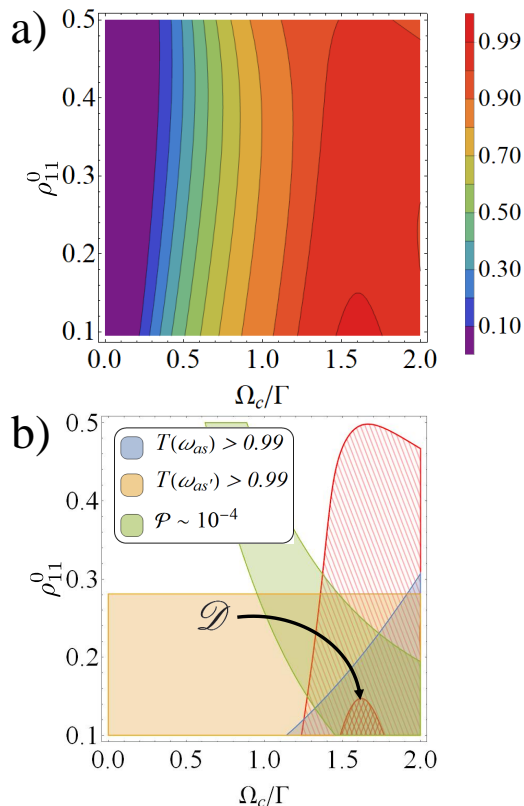


FIG. 5. **Optimal entanglement** - **a**) Same as in Fig. 3 with $\Delta_p = -\Gamma$, $\Delta_c = \Gamma$, $\Omega_p = 1.5\Gamma$ and with varying background population values ρ_{11}^0 of the lowest level [40]. **b**) Regions where the $NPT \geq 0.95$ (red line hatching) and $NPT \geq 0.99$ (red cross hatching) as in Fig. 5a. Anti-Stokes transmission (T_{as}) increases with Ω_c but strongly decreases with ρ_{11}^0 if $\Omega_c \lesssim \Gamma$. The other anti-Stokes transmission ($T_{as'}$) varies in the range $0.98 \div 1$ for $0.5 > \rho_{11}^0 > 0.1$ and regardless of Ω_c . The overlap region (\mathcal{D}) between the low absorption areas (light blue and light orange) yield optimal conditions to generate color entanglement ($NPT \geq 0.99$) with a probability $\mathcal{P} \sim 10^{-4}$.

In general, the atoms (optical) response depends on the atom's levels relative steady-state background atomic populations ρ_{jj} with $\{j = 1, 2, 3\}$ (see [40]). As there exists widely used techniques for accurate state preparation of cold ^{85}Rb samples [31] it may be worth to examine how entanglement may be engineered also through state preparation.

Unlike the Raman-like regime, where the NPT behaviour (Fig. 1) is unaffected by populations, changing the background population ρ_{11}^0 and ρ_{22}^0 of the two lower levels $|1\rangle$ and $|2\rangle$ leads (see Fig. 5a) to optimal NPT values ($NPT \geq 0.99$) at coupling strengths smaller than those of Fig. 4, with vanishing losses ($< 1\%$). A plot with all figures of merit - NPT , \mathcal{P} and absorption - is reported in Fig. 5b. Optimal color entanglement is seen to take place within an overlap region for weak drivings $\Omega_{p,c} \sim |\Delta_{p,c}| \sim \Gamma$ and a largely unbalanced back-

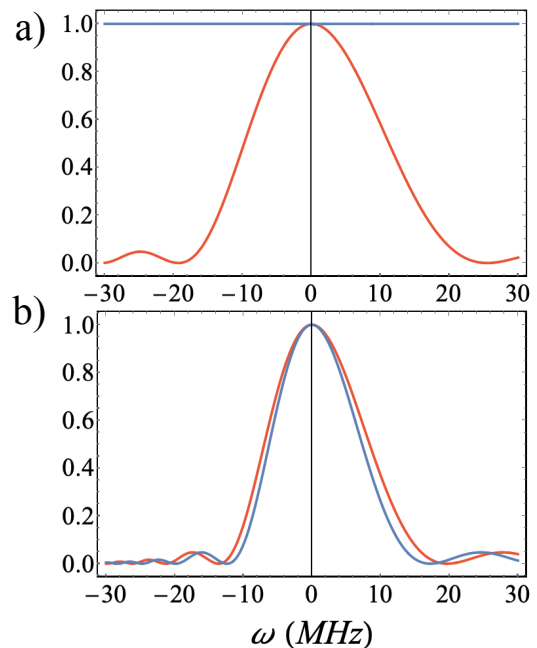


FIG. 6. **Phase-matching conditions**. The plots of $\text{sinc}^2(\Delta k_A L/2)$ (blue line) and $\text{sinc}^2(\Delta k_B L/2)$ (yellow line) show how the phase-matching conditions can be satisfied. **a**) The near-resonance parameters of Fig. 4 are used for the plots with $\Omega_p = \Omega_c = 2\Gamma$ for which the $NPT = 0.94$. **b**) Optimal entanglement conditions of Fig. 5 with $\Omega_c = 1.6\Gamma$ and $\rho_{11}^0 = 0.1$ where $NPT = 0.99$.

ground populations ρ_{11}^0 and ρ_{22}^0 of the two lower levels. From Fig. 5 it is clear how the manipulation of the lower populations yields to the optimization of generation as it guarantees almost the same performances in terms of NPT and \mathcal{P} as those of Fig. 4 yet at much lower $\Omega_{c,p}$'s which provides an advantage toward implementation of the scheme with weak driving fields.

Even though NPT takes into account of all the aspects connected to the state generation including phase-matching through Eq. (5), it is important to show how the phase-matching conditions Δk_A and Δk_B are simultaneously satisfied in the near-resonance conditions of Fig. 4 and Fig. 5. In the former case, perfect phase-matching occurs over a narrow bandwidth for the emission of ω_{as} while the $\omega_{as'}$ process is widely phase-matched [as shown of Fig. 6a)] since it occurs further away from level $|3\rangle$. On the other hand, in the latter case, Fig. 6b) shows that the phase-matching conditions are satisfied simultaneously over the same range of frequencies.

In conclusion, we anticipate that generation of heralded frequency-entangled single-photon states may occur through the Stokes state projection scheme of Fig. 2 whereby the detection of a single-Stokes photon in the frequency mode ω_s ($\omega_{s'}$) does not reveal, even in principle, which frequency mode ω_{as} or $\omega_{as'}$ is populated by

a single-photon. This scenario provides a new tool to develop quantum communication and information processing exploiting the frequency degree of freedom [12]. As it is often the case for non-classical effects of the electromagnetic field [41], the proposal for entanglement generation through the indistinguishability between two different spontaneous four-wave mixing processes exhibits a certain versatility. In principle, it could be adapted to atoms photonic crystal fibers interfaces [32, 33], to miniaturized (micrometer-sized) atomic vapour cells [35, 42] or to solid interfaces with crystals doped with rare-earth-metal ions [36] or with with N-V color centers [34, 37] where similar three-levels two-fold configurations exist.

We are indebted to Prof. Jin Hui Wu for many insightful discussions. Support from the Italian Ministry of Foreign Affairs (MAECI) and the National Natural Sci-

ence Foundation of China (NSFC) (Cooperative Program PGR00960), Ente Cassa di Risparmio di Firenze and the Italian Ministry of University and Research (MIUR) (QSecGroundSpace) are greatly acknowledged.

APPENDIX: Susceptibilities

Linear susceptibilities. The atomic medium response at the anti-Stokes frequencies ω_{as} and $\omega_{as'}$ is computed through a standard density matrix approach [22, 43] appropriate to the three-atomic levels configuration of Fig. 1. The diagonal and off-diagonal density matrix elements relevant, *e.g.*, to the Λ -configuration (*red*) of Fig. 1.A are computed in the limit of a “weak” anti-Stokes field that is Λ nearly-resonant with a (stronger) coupling (ω_c) through both lower ground levels $|1\rangle$ and $|2\rangle$. The resulting susceptibility can be written as,

$$\chi_{13}(\omega_{as}) = \frac{N|p_{13}|^2}{\hbar\epsilon_0} \left\{ \frac{(\omega_{31} - \omega_{as} - \Delta_c - i\gamma)}{D_{as}} \varrho_{11}^0 - \frac{\alpha_c}{1 + \alpha_c \frac{\Gamma_3}{\Gamma_2}} \frac{(\omega_{31} - \omega_{as} - i\frac{\Gamma}{2} - i\gamma)}{D_{as}} \varrho_{22}^0 \right\} \quad (8)$$

where

$$D_{as} = \left(\omega_{31} - \omega_{as} - i\frac{\Gamma}{2} \right) \left(\omega_{31} - \omega_{as} - \Delta_c - i\gamma \right) - |\Omega_c|^2 \quad (9)$$

and $\alpha_c = \frac{|\Omega_c|^2}{|\Delta_c + i\frac{\Gamma}{2}|^2}$ with $\Omega_c = \frac{\mathbf{p}_{32} \cdot \mathbf{E}_c}{\hbar}$ the coupling beam Rabi frequency. We denote by E_c the electric (real) field amplitude driving the ω_{32} transition and by \mathbf{p}_{32} the corresponding dipole moment (Fig. 1.A). Here $\Delta_c = \omega_{32} - \omega_c$ denotes the *coupling's* detuning from the resonant ω_{32} transition line while $\varrho_{11}^0 \equiv \rho_{11}^0 - \rho_{33}^0$ represents the total steady-state *background population* difference between $|1\rangle$ and $|3\rangle$ in the absence of pump and coupling and likewise for $\varrho_{22}^0 \equiv \rho_{22}^0 - \rho_{33}^0$. At thermal equilibrium the mean numbers of atoms in level $|1\rangle, |2\rangle$ and $|3\rangle$ are related by the Boltzmann's law [22, 31] so that one has, for instance, $N_2^0/N_1^0 \sim e^{-(E_2 - E_1)/k_B T}$. In particular, $N_3^0/N_1^0 \sim N_3^0/N_2^0 \sim e^{-(E_3 - E_{1,2})/k_B T} \sim 0$, a condition that is frequently matched in cold-atom experiments [31, 44]. The population difference $\varrho_{jj}^0 \equiv \rho_{jj}^0 - \rho_{33}^0 \simeq \rho_{jj}^0$ amounts to the steady-state background number density $\rho_{jj}^0 = N_j^0/N$ that one has in the absence of pump and coupling and with $N \equiv N_1^0 + N_2^0 + N_3^0 \cong N_1^0 + N_2^0$ being the total (conserved) number density. Temperature, level-degeneracy, pumping etc. are commonly used to achieve selective excitation of level $|2\rangle$ so as to vary ρ_{22}^0 [31, 45–48].

The spontaneous decay rate Γ_j of level $j = \{1, 2, 3\}$ with $\Gamma_{1,2}$ much smaller than Γ_3 , $\Gamma \simeq \Gamma_3$ and $\gamma \simeq (\Gamma_1 + \Gamma_2)/2$ [31]. In the absence of the coupling ($\alpha_c \rightarrow 0$)

the susceptibility $\chi_{13}(\omega_{as})$ recovers the well known expression for a two-level atom susceptibility whose real and imaginary parts yield the familiar dispersive and Lorentzian lineshapes [39, 49]. Likewise in the ground-state approximation [49] whereby most of the atomic background population is in the lowest state $|1\rangle$ ($\rho_{33}^0 \sim \rho_{22}^0 \sim 0$), the dominant term in (8) recovers the well known expression for a Λ three-level atom susceptibility responsible for EIT [39, 49, 50].

The susceptibility $\chi_{13}(\omega'_{as})$ for the other anti-Stokes mode “ ω'_{as} ” can be computed using the same approach when adapted to the levels Λ -configuration (*blue*) of Fig. 1.B.

Non-linear susceptibilities. The creation of photons at frequency ω_{as} , *e.g.*, occurs through a third-order mixing process by which the *three* different weak beams $\{\omega_p, \omega_c, \omega_s\}$ interact to generate another weak beam at frequency $\omega_{as} = \omega_p + \omega_c - \omega_s$, as sketched in Fig. 1.A. The relevant third-order nonlinear susceptibility can be computed through standard perturbative expansion methods [51, 52] so that under the assumption that just only one (excited) energy-level $|3\rangle$ contributes to intermediate resonances, the susceptibility (resonant part) comprises two terms, each proportional to the steady-state background population difference ρ_{11}^0 and ρ_{22}^0 of the two lower levels.

-
- * alessandro.zavatta@ino.it
- [1] L.-M. Duan, M. D. Lukin, J. I. Cirac, and P. Zoller, “Long-distance quantum communication with atomic ensembles and linear optics,” *Nature* **414**, 413–418 (2001).
 - [2] P. Kolchin, S. Du, C. Belthangady, G. Y. Yin, and S. E. Harris, “Generation of narrow-bandwidth paired photons: Use of a single driving laser,” *Phys. Rev. Lett.* **97**, 1–4 (2006).
 - [3] B. Srivathsan, G. K. Gulati, B. Chng, G. Maslennikov, D. Matsukevich, and C. Kurtsiefer, “Narrow Band Source of Transform-Limited Photon Pairs via Four-Wave Mixing in a Cold Atomic Ensemble,” *Phys. Rev. Lett.* **111**, 123602 (2013).
 - [4] K. Liao, H. Yan, J. He, S. Du, Z.-M. Zhang, and S.-L. Zhu, “Subnatural-Linewidth Polarization-Entangled Photon Pairs with Controllable Temporal Length,” *Phys. Rev. Lett.* **112**, 243602 (2014).
 - [5] Z. Han, P. Qian, L. Zhou, J. F. Chen, and W. Zhang, “Coherence time limit of the biphotons generated in a dense cold atom cloud,” *Sci. Rep.* **5**, 9126 (2015).
 - [6] C. Shu, P. Chen, T. K. A. Chow, L. Zhu, Y. Xiao, M.M.T. Loy, and S. Du, “Subnatural-linewidth biphotons from a Doppler-broadened hot atomic vapour cell,” *Nat. Commun.* **7**, 12783 (2016).
 - [7] A. MacRae, T. Brannan, R. Achal, and a. I. Lvovsky, “Tomography of a High-Purity Narrowband Photon from a Transient Atomic Collective Excitation,” *Phys. Rev. Lett.* **109**, 033601 (2012).
 - [8] E. Arimondo, “Laser phase spectroscopy in closed-loop multilevel schemes,” *Appl. Phys. B* **122**, 293 (2016).
 - [9] A. Raczynski, J. Zaremba, and S. Zielińska-Kaniasty, “Electromagnetically induced transparency and storing of a pair of pulses of light,” *Phys. Rev. A* **69**, 043801 (2004).
 - [10] A. Raczynski, J. Zaremba, and S. Zielińska-Raczynska, “Pulse propagation in a medium optically dressed by three fields forming a triangular loop configuration,” *J. Opt. Soc. Am. B* **32**, 1229–1236 (2015).
 - [11] S. Wengerowsky, S. K. Joshi, F. Steinlechner, H. Hübel, and R. Ursin, “An entanglement-based wavelength-multiplexed quantum communication network,” *Nature* **564**, 225–228 (2018).
 - [12] A. Zavatta, M. Artoni, D. Viscor, and G. La Rocca, “Manipulating Frequency-Bin Entangled States in Cold Atoms,” *Sci. Rep.* **4**, 3941 (2014).
 - [13] D. Viscor, V. Ahufinger, J. Mompert, A. Zavatta, G. C. La Rocca, and M. Artoni, “Two-color quantum memory in double- Λ media,” *Phys. Rev. A* **86**, 053827 (2012).
 - [14] O. Morin, J.-D. Bancal, M. Ho, P. Sekatski, V. D’Auria, N. Gisin, J. Laurat, and N. Sangouard, “Witnessing Trustworthy Single-Photon Entanglement with Local Homodyne Measurements,” *Phys. Rev. Lett.* **110**, 130401 (2013).
 - [15] F. Monteiro, V. Caprara Vivoli, T. Guerreiro, A. Martin, J. D. Bancal, H. Zbinden, R. T. Thew, and N. Sangouard, “Revealing genuine optical-path entanglement,” *Phys. Rev. Lett.* **114**, 1–5 (2015).
 - [16] A. Zavatta, M. D’Angelo, V. Parigi, and M. Bellini, “Remote preparation of arbitrary time-encoded single-photon ebits,” *Phys. Rev. Lett.* **96**, 020502 (2006).
 - [17] E. Lombardi, F. Sciarrino, and F. Popescu, S. and De Martini, “Teleportation of a vacuum-one-photon qubit,” *Phys. Rev. Lett.* **88**, 070402 (2002).
 - [18] D. Salart, O. Landry, N. Sangouard, N. Gisin, H. Herrmann, B. Sanguinetti, C. Simon, W. Sohler, R. T. Thew, A. Thomas, and H. Zbinden, “Purification of single-photon entanglement,” *Phys. Rev. Lett.* **104**, 180504 (2010).
 - [19] C. W. Chou, H. de Riedmatten, D. Felinto, S. V. Polyakov, S. J. van Enk, and H. J. Kimble, “Measurement-induced entanglement for excitation stored in remote atomic ensembles,” *Nature* **438**, 828–832 (2005).
 - [20] K. S. Choi, H. Deng, J. Laurat, and H. J. Kimble, “Mapping photonic entanglement into and out of a quantum memory,” *Nature* **452**, 67–71 (2008).
 - [21] Conversely, the detection of a single-photon $\omega_{s'}$ will prevent the processes $\{A, B\}$ from concurring to the generation of $|\Psi\rangle$.
 - [22] R. Loudon, *The quantum theory of light* (Clarendon Press, Oxford, 2000).
 - [23] M. Artoni and R. Loudon, “Propagation of nonclassical light through an absorbing and dispersive slab,” *Phys. Rev. A* **59**, 2279–2290 (1999).
 - [24] M. Artoni and R. Loudon, “Quantum theory of optical-pulse propagation through an amplifying slab,” *Phys. Rev. A* **57**, 622–628 (1998).
 - [25] S. Du, J. Wen, and M. H. Rubin, “Narrowband biphoton generation near atomic resonance,” *J. Opt. Soc. Am. B* **25**, C98 (2008).
 - [26] We easily recover the single-photon case [7] for $\alpha = 0$ i.e. when $E_c = 0$.
 - [27] The relative phase of f_A and f_B in (7) can be controlled by adjusting the pump and coupling relative phase.
 - [28] J. Lee, M. S. Kim, Y. J. Park, and S. Lee, “Partial teleportation of entanglement in a noisy environment,” *J. Mod. Opt.* **47**, 2151 (2000).
 - [29] A. Peres, “Separability Criterion for Density Matrices,” *Phys. Rev. Lett.* **77**, 1 (1996).
 - [30] M. Horodecki, P. Horodecki, and R. Horodecki, “Separability of mixed states: necessary and sufficient conditions,” *Phys. Lett. A* **223**, 1–8 (1996).
 - [31] C. Foot, *Atomic Physics*, 1st ed. (Oxford University Press, USA, 2005) section 7.6.
 - [32] V. Venkataraman, K. Saha, P. Londero, and A. L. Gaeta, “Few-photon all-optical modulation in a photonic band-gap fiber,” *Phys. Rev. Lett.* **107**, 193902 (2011).
 - [33] M. Bajcsy, S. Hofferberth, T. Peyronel, V. Balic, Q. Liang, A. S. Zibrov, V. Vuletic, and M. D. Lukin, “Laser-cooled atoms inside a hollow-core photonic-crystal fiber,” *Phys. Rev. A* **83**, 063830 (2011).
 - [34] M. W. Doherty, N. B. Manson, P. Delaney, F. Jelezko, J. Wrachtrup, and L. C. L. Hollenberg, “The nitrogen-vacancy colour centre in diamond,” *Phys. Rep.* **528**, 1–45 (2013).
 - [35] J.-M. Rost, “Atomic vapour microcells: Tubes for quantum electronics,” *Nat. Photonics* **4**, 74–75 (2010).
 - [36] Q. Y. He, Y. Xue, M. Artoni, G. C. La Rocca, J. H. Xu, and J. Y. Gao, “Coherently induced stop-bands in resonantly absorbing and inhomogeneously broadened doped crystals,” *Phys. Rev. B* **73** (2006).
 - [37] Jin-Hui Wu, G. C. La Rocca, and M. Artoni, “Controlled light-pulse propagation in driven color centers in diamond,” *Phys. Rev. B* **77**, 113106 (2008).
 - [38] T. Kiss, U. Herzog, and Ulf Leonhardt, “Compensation

- of losses in photodetection and in quantum-state measurements,” *Phys. Rev. A* **52**, 2433 (1995).
- [39] M. Scully and S. Zubairy, *Quantum Optics*, 1st ed. (Cambridge University Press, 1997).
- [40] The population difference $\varrho_{jj}^0 = \rho_{jj}^0 - \rho_{33}^0 \simeq \rho_{jj}^0$ amounts to the steady-state background number density $\rho_{jj}^0 = N_j^0/N$ that one has in the absence of pump and coupling and with $N \cong N_1^0 + N_2^0$ being the total (conserved) number density.
- [41] M. Artoni and J. L. Birman, “Non-classical states in solids and detection,” *Opt. Commun.* **104**, 319–324 (1994).
- [42] T. Baluktsian, C. Urban, T. Bublath, H. Giessen, R. Löw, and T. Pfau, “Fabrication method for microscopic vapor cells for alkali atoms,” *Opt. Lett.* **35**, 1950–1952 (2010).
- [43] E. Arimondo, “Coherent population trapping in laser spectroscopy,” in *Progress in Optics*, Vol. 35, edited by E. Wolf (Elsevier, 1996) pp. 257 – 354.
- [44] S. Giorgini, L. P. Pitaevskii, and S. Stringari, “Theory of ultracold atomic fermi gases,” *Rev. Mod. Phys.* **80**, 1215–1274 (2008).
- [45] D.G.C. Jones, *Atomic Physics*, 1st ed. (Chapman & Hall, London, UK, 1997).
- [46] L. Allen and D. G. C. Jones, *Principles of Gas Lasers*, 1st ed. (Butterworths, London, UK, 1967).
- [47] I. Bloch, “Ultracold quantum gases in optical lattices,” *Nat. Phys.* **1**, 23 (2005).
- [48] I. Bloch, J. Dalibard, and S. Nascimbène, “Quantum simulations with ultracold quantum gases,” *Nat. Phys.* **8**, 267 (2012).
- [49] P. W. Milonni, *Fast Light, Slow Light and Left-Handed Light*, 1st ed. (Taylor & Francis, 2004).
- [50] S. Harris, “Electromagnetically induced transparency,” *Phys. Today* **50**, 36–42 (1997).
- [51] H. Lotem, R.T. Lynch, and N. Bloembergen, “Interference between raman resonances in four-wave difference mixing,” *Phys. Rev. A* **14**, 1748–1755 (1976).
- [52] S. A. J. Druet, B. Attal, T. K. Gustafson, and J. P. Taran, “Electronic resonance enhancement of coherent anti-stokes raman scattering,” *Phys. Rev. A* **18**, 1529–1557 (1978).