Engineering of heralded narrowband color-entangled states

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The efficient generation of entanglement is an essential requirement for quantum communications; however, long distances can only be achieved by utilizing entangled states that can be efficiently mapped into matter. Hence, sources generating states with bandwidths naturally compatible with the 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 generation and generation probability optimizations yet with negligible absorption. The scheme could also be adapted to photonic and solid interfaces.

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Introduction. 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, growing attention has been devoted towards the development of narrowband sources of quantum states of light to efficiently map light into atoms. 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 times with very low probabilities. In order to circumvent this limitation, so-called heralded schemes, where desired output states are announced by ancillary photons, are required to allow both further processing into quantum communication networks and quantum state characterization [7].

Within this context, it is convenient to exploit the multi-level structure of resonant media such as atoms or atomlike systems [8–10], in order to engineer the generation of quantum states in a frequency multimode scenario [11,12]. In this Rapid Communication, 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 color-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 [13] and mapping [14] onto a quantum memory of color-entangled states through a specific atomic interface has recently been studied, hence the present Rapid Communication represents an additional building block toward the realization of a quantum network, in which color 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 (ωp) and pump (ωp) copropagating beams, Stokes and anti-Stokes photon pairs emerge (Fig. 1). The two processes, one (A) leading to the emission of a Stokes (ωs) and an anti-Stokes (ωas) photon pair and the other (B) leading to the emission of the same Stokes (ωs) yet a different anti-Stokes (ω′ as) photon, are sketched in Fig. 1 (red and blue lines) and governed by the following energy conservation laws ωs + ωas = ωp + ωs and ωs + ω′ as = 2ωp, respectively. The main feature of the scheme is that under suitable pump and coupling driving 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 Stokes photon at frequency \( \omega_s \) will entail, via a state-projection measurement on the Stokes (\( \omega_s \) ) photon (Fig. 2). The spontaneous four-wave mixing processes (a) and (b) result in a small probability of generating single anti-Stokes photons at frequencies \( \omega_{as} \) through channels (a) and (b) can be made to be undistinguishable subject to a state-projection measurement on the Stokes (\( \omega_s \)) photon (Fig. 2). The atomic levels correspond to the transition \( ^5S_{1/2} \rightarrow ^5P_{1/2} \) (\( ^85\text{Rb} D_1 \) line) with \( \lambda_{51} = 795 \text{ nm} \), \( \omega_{51} = 2\pi \times 3 \text{ GHz} \), and decay rates \( \Gamma = 2\pi \times 5.75 \text{ MHz} \) and \( \gamma = 2\pi \times 10 \text{ kHz} \).

Two more processes will clearly contribute to the heralded generation of the 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 and D from concurring to the generation of the state (1). [22]

Entanglement generation. Assuming classical fields in the form of plane waves \( E_i^{\pm} = E_i e^{i k_i r - i \omega_i t} \) with \( i = \{c, p\} \) respectively for the control and pump beams, and a standard field operator

\[
\hat{A}_j(\omega) = \frac{1}{\sqrt{\pi}} \int d\omega \frac{2\hbar \omega}{c \epsilon_0 A} e^{ik_i r - i\omega t} \hat{a}_j(\omega)
\]

with \( j = \{s, as\} \), for the Stokes and anti-Stokes photons propagating with a (complex) wave vector \( k_j(\omega) \) [23–25], the effective Hamiltonian describing the photon-atom interaction can be written as

\[
\hat{H}_I = \frac{\epsilon_0 A}{4} \int_{-L/2}^{L/2} dz \left[ \chi_A^{(3)} E_p^+ E_r^+ \hat{E}_{ar} \hat{E}_{as}^+ + \chi_B^{(3)} E_p^+ E_r^+ \hat{E}_{ar}^+ \hat{E}_{as} \right.
\]

\[
+ \chi_C^{(3)} E_p^+ E_r^+ \hat{E}_{as} \hat{E}_{as}^+ + \chi_D^{(3)} E_p^+ E_r^+ \hat{E}_{as}^+ \hat{E}_{as}^+ ] + \text{H.c.},
\]

with the fields' (negative) frequency parts \( E_i^- \) and \( \hat{E}_i^- \) computed as usual [23] and with space-time dependencies purposely omitted here. Here, we assume control and pump fields with a fixed relative phase relationship that can be arbitrarily controlled and tuned. The four third-order optical nonlinear susceptibilities \( \chi_{ij}^{(3)} \) for \( l = \{A, B, C, D\} \) in (3) correspond to the four spontaneous nonlinear mixing processes triggered by coupling and pump. Details of the susceptibility expressions are given in the Appendix. For the sake of clarity we restrict to a nearly one-dimensional 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 anti-Stokes modes are initially in the vacuum \(|0\rangle, |0\rangle_c, |0\rangle_{as}, |0\rangle_{as'} \rightarrow |0\rangle\), so that in the weak spontaneous scattering limit [26], one has for the output state,

\[
|\Psi\rangle_{\text{out}} \approx \left(1 - \frac{i}{\hbar} \int_{-\infty}^{\infty} dt \, \hat{H}_t \right) |0\rangle
\]

\[
= |0\rangle + \int d\omega f_A(\omega + \omega_p - \omega, \omega) \hat{a}_{as'}(\omega + \omega_p - \omega) \\
\times \hat{a}_{as}(\omega) + f_B(2\omega_p - \omega, \omega) \hat{a}_{as'}(\omega_p + \omega) \hat{a}_{as}(\omega) \\
+ f_C(\omega_p + \omega, \omega) \hat{a}_{as'}(\omega_p + \omega) \hat{a}_{as}(\omega) \\
+ f_D(\omega, \omega) \hat{a}_{as'}(\omega) \hat{a}_{as}(\omega) \hat{a}_{as'}(\omega) \\
\times |0\rangle, |0\rangle_c, |0\rangle_{as}, |0\rangle_{as'}.
\]  

(4)

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 [26] we examine here, hence Eq. (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. 1(a)]. The generation of the pair \(\{\alpha_s, \alpha_{as}\}\), e.g., occurs with the probability amplitude,

\[
f_A(\omega, \omega') = -i \frac{\sqrt{\omega \omega'}}{4 \pi c} \chi_A^{(3)}(\omega, \omega') E_p E_c \sin \left(\frac{\Delta k_A L}{2}\right) L,
\]

(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 projection \(\Delta k_A = (k_{as} + k_c - k_p - k_{as'}) \cdot \hat{z}\) of the wave-vector mismatch (momentum conservation) for copropagating pump and coupling beams. We restrict here to collecting Stokes and anti-Stokes photon pairs that are emitted within a very small angle with respect to the \(z\) direction. Therefore, the phase mismatch is given by

\[
\Delta k_A = -\frac{\omega_{as}}{c} n_{13}(\omega_{as}) + \frac{\omega_{as}}{c} n_{23}(\omega_s) \\
- \frac{\omega_p}{c} n_{13}(\omega_p) - \frac{\omega_c}{c} n_{23}(\omega_c),
\]

(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 the Appendix).

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

\[
|\Psi\rangle_p = \left[|\alpha_s, 0\rangle \hat{a}_{as'}(\omega_s) |\Psi\rangle_{\text{out}}\right]
\]

\[
= f_A(\omega_{as}, \omega_s) |1\rangle_{as} |0\rangle_{as'} + f_B(\omega_{as'}, \omega_s) |0\rangle_{as} |1\rangle_{as'},
\]

(7)

which corresponds to the state sought for in (1) with (complex) coefficients \(\alpha \rightarrow f_A(\omega_{as}, \omega_s)/\sqrt{N}\) and \(\beta \rightarrow f_B(\omega_{as'}, \omega_s)/\sqrt{N}\), being \(N = |f_A|^2 + |f_B|^2\) (normalization). Here, \(|1\rangle_{as} = \hat{a}_{as}(\omega_s) |0\rangle\) denotes a single-photon state at the frequency \(\omega_i\) 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 wave-vector mismatch (momentum conservation) for copropagating pump and coupling beams. We restrict here to collecting Stokes and anti-Stokes photon pairs that are emitted within 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_{as}}{c} n_{23}(\omega_s) \\
- \frac{\omega_p}{c} n_{13}(\omega_p) - \frac{\omega_c}{c} n_{23}(\omega_c),
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The anti-Stokes $\omega_{as}$, $\rho_{as,c}$, decreasing. The anti-Stokes $\omega_{as}$ vary with $\omega_{as}$. Thus, the lossy state appears to be noisy and splitter that mixes the ideal state with the vacuum, then the probability to find a single-photon state in modes $\omega_{as}$ is close to unity with probabilities $\rho_{as,c}$, constant with $\Omega_r$, range from a $\sim 3\%$ maximum ($\Omega_r \simeq \Gamma$) to almost no loss at all ($\Omega_r = 6\Gamma$).

$10^{-4}$). The generation probability of the entangled state is the probability to find a single-photon state in modes $\omega_{as}$ and $\omega_{as}'$, and this is $\mathcal{P} = |\langle 1_{as} | \Psi \rangle_p|^2 + |\langle 1_{as} | \Psi \rangle_p|^2 = |f_a|^2 + |f_a'|^2$.

In the spontaneous Raman regime (Fig. 3) we find values $\mathcal{P} \sim 10^{-13}$ when background field 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 crucial [24]. This turns out to be an important generation regime which we will discuss below. A proper assessment of the negativity of the first-order susceptibility $\chi_{13}(\omega_{as,as})$ is obtained from the imaginary part of $\chi_{13}(\omega_{as,as})L/c$. The output state affected by absorption is then used to calculate the $NPT$. Hereafter, a $NPT$ close to unity will 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 magnitude larger than in the Raman case. Maximal entanglement ($NPT \geq 0.99$) is restricted only to the high-intensity region ($\Omega_{p,c} \sim 6\Gamma$) with probabilities $\mathcal{P} \sim 10^{-2}$ provided that $\rho_{11}^0 \gg \rho_{22}^0$, i.e., most of the atomic background population (see the 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-intensity region due to electromagnetically induced transparency (EIT) [40].

In general, the sample optical response depends on the atom’s levels steadystate background atomic populations $\varrho_{ij}$ with $j = \{1, 2, 3\}$ (see Ref. [41]). As accurate state preparation techniques for cold $^{87}$Rb samples are widely used [32], it may be worth examining how entanglement may be engineered also through state preparation.

Unlike the Raman-like regime, where the $NPT$ behavior (Fig. 1) is unaffected by populations, changing the background population $\rho_{11}^0$ and $\rho_{22}^0$ of the two lower levels $|1\rangle$ and $|2\rangle$ leads instead [see Fig. 5(a)] to optimal $NPT$ values equal or better than 0.99 at coupling strengths smaller than those of Fig. 4, with vanishing losses ($<1\%$). A plot with all

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 it increases as $NPT$ decreases. The anti-Stokes $\omega_{as}$ losses are constant with $\Omega_r$ yet varying with $\Omega_r$. Hereafter, a splitter model [39]. According to this model, the dissipative generation regime which we will discuss below. A proper assessment of the negativity of the partial transpose function in the presence of absorption is in order, which is done here by adopting a standard beamsplitter model [39]. According to this model, the dissipative process is formally described by means of a lossless 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 reflection of the beam splitter that accounts for absorption.

We apply this loss model to each anti-Stokes mode whose absorption $\mathcal{R}(\omega_{as,as})$ is obtained from the imaginary part of the first-order susceptibility $\chi_{13}(\omega_{as,as})$ (see the Appendix), i.e., $\mathcal{R}(\omega_{as,as}) = 1 - \exp[\text{Im} \chi_{13}(\omega_{as,as})]/(\omega_{as,as}L/c)$. The output state affected by absorption is then used to calculate the $NPT$. Hereafter, a $NPT$ close to unity will 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-intensity region ($\Omega_{p,c} \sim 6\Gamma$) with probabilities $\mathcal{P} \sim 10^{-2}$ provided that $\rho_{11}^0 \gg \rho_{22}^0$, i.e., most of the atomic background population (see the 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-intensity region due to electromagnetically induced transparency (EIT) [40].

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figures of merit—$NPT$, $\mathcal{P}$, and absorption $\mathcal{R}$—is reported in Fig. 5(b). Optimal color entanglement is seen to take place within an overlap region for weak drivings $\Omega_{p,c} \sim |\Delta_{p,c}| \sim 1 \Gamma$ and largely unbalanced background populations $\rho_{01}^{(1)}$ and $\rho_{02}^{(2)}$ 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 $\omega_{s}$'s, which provides an advantage toward the implementation of the scheme with weak driving fields.

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

In conclusion, we anticipate that the 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 $\omega_{s}$ ($\omega_{p,c}$) does not reveal, even in principle, which frequency mode $\omega_{as}$ or $\omega_{as'}$ is populated by a single photon. This scenario provides a tool to develop quantum communication and information processing by exploiting the frequency degree of freedom [13]. As it is often the case for nonclassical effects of the electromagnetic field [42], 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 atom photonic crystal fiber interfaces [33,34], to miniaturized (micrometer-sized) atomic vapor cells [36,43], or to solid interfaces with crystals doped with rare-earth-metal ions [37], or with NV color centers [35,38] where similar three-level twofold configurations exist.

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APPENDIX: SUSCEPTIBILITES

Linear susceptibilities. The atomic medium response at the anti-Stokes frequencies $\omega_{as}$ and $\omega_{as'}$ is computed through a standard density matrix approach [23,44] appropriate to the level configurations of Fig. 1. The diagonal and off-diagonal density matrix elements relevant, e.g., to the $A$ configuration (red) of Fig. 1(a) are computed in the limit of a “weak” anti-Stokes field at frequency $\omega_{as}$ (nearly) two-photon resonant with a “strong” coupling ($\omega_{c}$) through both lower ground levels $|1\rangle$ and $|2\rangle$. The relevant anti-Stokes susceptibility can approximately be written as,

$$
\chi_{13}(\omega_{as}) = \frac{N |p_{13}|^2}{\hbar \epsilon_0} \left\{ \frac{\rho_{00}^{(1)} - \rho_{02}^{(2)} - \Delta_{c} - i\gamma}{D_{as}} \rho_{11}^{(1)} - \frac{\alpha_{c}}{1 + \alpha_{c}^{2} \gamma^{2}} \frac{\rho_{31}^{(1)} - \rho_{21}^{(2)} - \Delta_{c} - i\gamma}{D_{as}} \rho_{22}^{(1)} \right\},
$$

where

$$
D_{as} = \left( \rho_{31}^{(1)} - \rho_{21}^{(2)} - i\gamma \right) \left( \rho_{31}^{(1)} - \rho_{21}^{(2)} - \Delta_{c} - i\gamma \right) - \Omega_{as}^{2}.
$$

and $\alpha_{c} = \frac{|\Omega_{as}|^{2}}{|\Delta_{as} + \gamma|^{2}}$ with $\Omega_{as} = \hbar \epsilon_{as} E_{as} / \hbar$ the coupling beam Rabi frequency. We denote by $E_{as}$ the electric (real) field amplitude driving the $\omega_{as}$ transition and by $p_{13}$ the corresponding dipole moment [Fig. 1(a)]. Here, $\Delta_{c} = \omega_{c} - \omega_{as}$ denotes the coupling’s detuning from the resonant $\omega_{as}$ transition line while $\rho_{00}^{(1)} - \rho_{02}^{(2)}$ represents the total steady-state background population difference between $|1\rangle$ and $|3\rangle$ in the absence of pump and coupling and likewise for $\rho_{22}^{(1)} - \rho_{22}^{(2)}$. At thermal equilibrium the number of atoms in levels $|1\rangle$, $|2\rangle$ and $|3\rangle$ are related by the Boltzmann’s law [23,32] so that one has, for instance, $N_{10}^{(1)} / N_{10}^{(2)} \sim e^{-(\omega_{as} - \omega_{c}) / k_{B} T}$. In particular, $N_{30}^{(1)} / N_{30}^{(2)} \sim N_{30}^{(1)} / N_{30}^{(2)} \sim e^{-(\omega_{as} - \omega_{c}) / k_{B} T} \sim 0$, a condition that is frequently matched in cold-atom experiments [32,45]. The population difference $\rho_{31}^{(1)} - \rho_{21}^{(2)} - \Delta_{as}$ amounts to the steady-state background number density $\rho_{00}^{(1)} = N_{00}^{(1)} / N$ that
one has in the absence of pump and coupling and with $N \equiv N_0^0 + N_2^0 + N_3^0 \cong N_0^0 + N_2^0$ being the total number density. Temperature, level-degeneracy, pumping, etc., are commonly used to achieve selective excitation of level $[2]$ so as to vary $\rho_{22}^0$ [32,46–49].

The spontaneous decay rate $\Gamma_j$ of level $j = \{1, 2, 3\}$ with $\Gamma_{12}$ much smaller than $\Gamma_3$, $\Gamma \cong \Gamma_3$, and $\gamma \cong (\Gamma_1 + \Gamma_3)/2$ [32]. In the absence of the coupling ($\alpha_c \rightarrow 0$), the susceptibility $\chi(\omega_{as})$ recovers the well-known expression for a two-level atomic susceptibility whose real and imaginary parts yield the familiar dispersive and Lorentzian line shapes [40,50]. Likewise in the ground-state approximation [50] whereby most of the atomic background population is in the lowest state $\vert 1 \rangle$ ($\rho_{33}^0 \sim \rho_{22}^0 \sim 0$), the dominant term in (A1) recovers the well-known expression for a $\Lambda$-three-level atomic susceptibility responsible for EIT [40,50,51].

The susceptibility $\chi(\omega_{as})$ for the other anti-Stokes mode $\omega_{as}$ can be computed using the same approach when adapted to the $\Lambda$-levels configuration (blue) of Fig. 1(b).

Nonlinear susceptibilities. The creation of photons at frequency $\omega_{as}$, 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 [52,53] so that under the assumption that just one (excited) energy level [3] contributes to intermediate resonances, the susceptibility (resonant part) comprises two terms, each proportional to the steady-state background population difference $\rho_{11}^0$ and $\rho_{22}^0$ of the two lower levels.


[22] Conversely, the detection of a single photon $\omega_s$ will prevent the processes $\{A, B\}$ from concurring to the generation of $|\Psi\rangle$.


We easily recover the pure single-photon case [7] with \( \alpha = 0 \) setting \( E_c = 0 \).

The relative phase of \( f_A \) and \( f_B \) in (7) can be controlled by adjusting the pump and coupling relative phase.


C. Foot, Atomic Physics, 1st ed. (Oxford University Press, New York, 2005), Sec. 7.6.


The population difference \( \rho_{0j}^{0j} = \rho_{0j}^{0j} - \rho_{03}^{03} \approx \rho_{0j}^{0j} \) amounts to the steady-state background number density \( \rho_{0j}^{0j} = N_{0j}^j / N \) that one has in the absence of pump and coupling and with \( N \approx N_{0j}^j + N_{03}^3 \) being the total (conserved) number density.


