Non-classical photon pair generation in atomic vapours

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Abstract

A scheme for the generation of non-classical pairs of photons in atomic vapours is proposed. The scheme exploits the fact that the cross correlation of the emission of photons from the extreme transitions of a four-level cascade system shows anti-bunching which has not been reported earlier and which is unlike the case of the three level cascade emission which shows bunching. The Cauchy-Schwarz inequality which is the ratio of cross-correlation to the auto correlation function in this case is estimated to be $10^3 - 10^6$ for controllable time delay, and is one to four orders of magnitude larger compared to previous experiments. The choice of Doppler free geometry in addition to the fact that at three photon resonance the excitation/deexcitation processes occur in a very narrow frequency band, ensures cleaner signals.

1 Introduction

Non-classical sources of light include triggered single photon emitters [1] as well as photon pair emitters [2] where the first photon heralds the arrival of the second one. The recent elegant experiments on the generation of non-classical photon pairs have successfully reported the temporal control of emission of an anti-stokes photon by programmable time delay. Polyakov et al [3], report a violation of the classical Cauchy-Schwarz inequality in the photon correlations, and they further report that the ratio of cross-correlation to auto-correlation $R_{max} = 292 \pm 57$. All these experiments have been done on cold atoms in magneto-optical traps (MOT). Recently, this experiment has been repeated in atomic vapours at room temperature [4] and the results are comparable with the MOT results. While in the MOT set up the magnetic and quadrupole fields induces decoherence in the correlations, in the latter case the inefficient absorption/excitation due to Doppler broadening is the limiting factor.
In this paper I propose a scheme for the generation of non-classical pair of photons wherein Doppler effects are minimised even at room temperatures, and the degree of violation of the Cauchy-Schwarz inequality is very large \( \sim 10^3 - 10^6 \). The key feature we exploit here hinges on the fact that the cross correlation between the emission of the extreme transitions in a four-level cascade system shows anti-bunching, which is in contrast to the well known fact that cross-correlation of the emission from three level cascade systems shows bunching \( \text{[5, 6]} \). In addition, one can obtain very strong correlations between the photon emissions from the extreme transitions by controlling the driving fields. To achieve this I consider here a model which consists of the 4-level cascade system in Rubidium which is an extension of the well studied 3-level cascade system in the context of the absorptive as well as the dispersive property in electromagnetically induced transparency (EIT) systems \( \text{[7, 8]} \). In addition, to the 3-level cascade system consisting of \( 5s_{1/2}, 5p_{3/2} \) and \( 5d_{5/2} \) considered in the previous examples, an additional level, say, one of the other hyperfine levels of \( 5p_{3/2} \) may be included and a strong coupling introduced between different hyperfine levels of \( 5p_{1/2} \) through a radio-frequency (rf) field (see Fig1). The role of this ‘sandwich’ coupling is to change the usual ‘bunched’ emission from the cascade system to antibunched emission, with the difference that the cascade emission is interrupted by the rf field after the first emission. This control field helps in inducing strong correlations between the uppermost and the lowermost transitions. It further provides us with a handle to control the time delay between the successive emissions of photons from the extreme transitions. The combined feature of the violation of two classical inequalities namely (i) antibunching and (ii) Cauchy-Schwarz inequality in this scheme makes it very attractive. Further the problem of the absorption/emission into other frequency modes close to resonance (due to Doppler broadening) which leads to the weakening of the signals \( \text{[4]} \) is circumvented in the present scheme because of the intrinsic narrow absorption features due to atomic coherence effects induced by three-photon resonance.

The plan of the paper is as follows: In section II the model under consideration is described in detail and the equation of motion for the atom+field system is set up. In section III the relevant photon correlation functions are evaluated numerically and in some special cases analytical expressions for the correlation functions are presented, which are evaluated perturbatively. Further, these results are compared with the exact numerical results for the correlation functions.

2 The model

The system considered here consists of an ensemble of four-level ladder atoms (Fig1). This could correspond, eg., to \( 5s_{1/2}, 5p_{3/2} \) and \( 5d_{5/2} \) of Rb \( \text{[10]} \). I would like to recall here that this was exactly the scheme used by Banacloche et al \( \text{[7]} \) and Fulton et al \( \text{[9]} \) for studying EIT. Labeling the energy levels \( 5s_{1/2}, 5p_{3/2} \) as \( |1> \) and \( |2> \) respectively, level \( |3> \) may
Figure 1: Four-level system interacting with three driving fields of strengths $\Omega_1, \Omega_{rf}, \Omega_3$ and their respective detunings $\Delta_1, \Delta_{rf}$ and $\Delta_2$. $\Gamma_2, \Gamma_3, \Gamma_4$ are the decay constants of the corresponding levels.

be identified as one of the hyperfine levels of $^{5}p_{3/2}$ ($F=0,1,2,3$ of $^{87}$Rb and $F=1,2,3,4$ of $^{85}$Rb), with level separations typically of the order of $10^2 MHz$. Level $|4>$ may be chosen to be $5d_{5/2}$. The only dipole allowed transitions are $|1>\leftrightarrow |2>$, $|2>\leftrightarrow |3>$ and $|3>\leftrightarrow |4>$ in the limit of the hyperfine splittings being larger compared to the Rabi frequency [18]. Comparing this scheme with the 3-level cascade system of references [7, 9, 8], levels $|1>$, $|2>$, $|3>$ of these setups correspond respectively to levels $|1>$, $|2>$ and $|4>$ of the present model. In addition we have introduced another hyperfine level which we have labeled as $|3>$. For this system, the two transitions $|1>\rightarrow |2>$ and $|3>\rightarrow |4>$ respectively may be chosen to correspond to wavelengths 780nm and 775.8nm, as in the schemes used in references [7, 9, 8]. Apart from these two driving fields in the optical region, we consider yet another coupling between the hyperfine levels (belonging to $^{5}p_{3/2}$) $|2>$ and $|3>$ through a rf field of strength $\Omega_{rf}$. Such couplings between the hyperfine levels have been considered in the study of absorption in 4-level systems [12]. The strengths of the optical fields are denoted by $\Omega_1$ and $\Omega_3$, and the respective detunings by $\Delta_1$ and $\Delta_3$. The corresponding decay parameters are $\Gamma_2, \Gamma_3 = 6 MHz$, $\Gamma_4 \approx 1 MHz(0.97 MHz)$. To make the three photon interaction Doppler free we choose the two beams in the optical region, as already mentioned, to be counter-propagating. They couple respectively transitions $|1>\leftrightarrow |2>$ and $|3>\leftrightarrow |4>$. The optical frequencies, being nearly equal, will give null contribution to the linear Doppler shift. While, the linear Doppler shift of the rf field (in the atomic rest frame) at this frequency is very small and hence negligible. This choice for minimizing the Doppler effect is
preferable compared to the phase-matched geometry used in four-wave mixing as this offers a larger interaction volume compared to the latter.

The master equation describing the interaction of the four-level atomic system with three driving fields in the rotating wave approximation is given by

\[
\frac{\partial \rho_{12}}{\partial t} = -(i\Delta_1 - \Gamma_2/2)\rho_{12} - i\Omega_1(\rho_{22} - \rho_{11}) + i\Omega_{rf}\rho_{13}
\]

\[
\frac{\partial \rho_{23}}{\partial t} = -(i\Delta_2 - (\Gamma_2 + \Gamma_3)/2)\rho_{23} - i\Omega_1\rho_{13} - i\Omega_{rf}(\rho_{33} - \rho_{22}) + i\Omega_3\rho_{24}
\]

\[
\frac{\partial \rho_{34}}{\partial t} = -(i\Delta_3 - (\Gamma_3 + \Gamma_4)/2)\rho_{34} - i\Omega_{rf}\rho_{24} - i\Omega_3(\rho_{44} - \rho_{33})
\]

\[
\frac{\partial \rho_{13}}{\partial t} = -(i\Delta_1 + \Delta_2 - \Gamma_3/2)\rho_{13} - i\Omega_1\rho_{23} + i\Omega_{rf}\rho_{12} + i\Omega_3\rho_{14}
\]

\[
\frac{\partial \rho_{14}}{\partial t} = -(i\Delta_1 + \Delta_2 + \Delta_3 - \Gamma_4/2)\rho_{14} - i\Omega_1\rho_{24} + i\Omega_3\rho_{13}
\]

\[
\frac{\partial \rho_{24}}{\partial t} = -(i\Delta_2 + \Delta_3 - (\Gamma_2 + \Gamma_4)/2)\rho_{24} - i\Omega_1\rho_{14} - i\Omega_{rf}\rho_{34} + i\Omega_3\rho_{23}
\]

\[
\frac{\partial \rho_{22}}{\partial t} = -\Gamma_2\rho_{22} + i\Omega_1(\rho_{21} - \rho_{12}) + i\Omega_{rf}(\rho_{23} - \rho_{33}) + \gamma_{23}\rho_{33} + \gamma_{24}\rho_{44}
\]

\[
\frac{\partial \rho_{33}}{\partial t} = -\Gamma_3\rho_{33} + i\Omega_3(\rho_{44} - \rho_{33}) - i\Omega_{rf}(\rho_{23} - \rho_{32}) - i\Omega_3\rho_{34} + \gamma_{34}\rho_{44}
\]

\[
\frac{\partial \rho_{44}}{\partial t} = -\Gamma_4\rho_{44} - i\Omega_3(\rho_{44} - \rho_{33})
\]

where \(\gamma_{ik}\) are the transition rates from the level \(|k > |i\), \(\Delta_i = \omega_{i,i+1} - \omega_i\) are the laser detunings (\(\rho_{ij} = \rho_{ji}^*\) and \(\text{Tr}\rho = 1\)). Throughout this paper we assume \(\gamma_{ii+1} = 1\) and the rest to be zero. I obtain analytical solutions by perturbatively solving these equations in the Laplace space in the special cases where the rf field strength is either i) large, or ii) small compared to the other two driving fields.

**Case (i) Large \(\Omega_{rf}\)**

In this regime we assume that \(\Omega_{rf} > \Omega_1, \Omega_3\). Treating the fields \(\Omega_i, i = 1, 3\) perturbatively, and labeling \(\psi_1 = \rho_{12}, \psi_2 = \rho_{23}, \psi_3 = \rho_{34}, \psi_4 = \rho_{13}, \psi_5 = \rho_{14}, \psi_6 = \rho_{24}, \psi_7 = \rho_{22}, \psi_8 = \rho_{33}, \psi_9 = \rho_{44}\) and \(\psi_i(k)\) to be the Laplace transform of \(\psi_i(t)\), the equation of motion in the Laplace space assumes the simple form

\[
(s + \Gamma_2)\psi_7^{(2)}(s) = \psi_7(0) - 2\Omega_{rf}Im\psi_2^{(2)}(s) +...
\]
I next consider the case when the rf field is weak. Considering the contribution of $\Omega_{rf}$ perturbatively, the second order equations are given by

\[(s + \bar{\Gamma}_2 + \bar{\Gamma}_3)\bar{\psi}_7^{(2)}(s) = 2\Omega_1 Im\bar{\psi}_1^{(1)}(s) + \bar{\psi}_8^{(2)}(s) - i\Omega_{rf}(\bar{\psi}_7^{(2)}(s) - \bar{\psi}_7^{(2)}(s)) - i\Omega_1\bar{\psi}_4^{(1)}(s) + i\Omega_3\bar{\psi}_6^{(1)}(s)\]
\[(s + \bar{\Gamma}_3)\bar{\psi}_8^{(2)}(s) = \psi_8(0) + 2\Omega_{rf} Im\bar{\psi}_2^{(2)}(s) - 2\Omega_3\bar{\psi}_3^{(1)}(s) + \bar{\psi}_9^{(2)}(s)\]
\[(s + \bar{\Gamma}_4)\bar{\psi}_9^{(2)}(s) = \psi_9(0) + 2\Omega_3 Im\bar{\psi}_3^{(1)}(s)\]

where $\Omega_{rf}$ is treated upto all orders and the fields $\Omega_{1,3}$ are treated perturbatively. For simplicity I have assumed $\Delta_{rf} = \Delta_i = 0, i = 1, 3$ and replaced $\Gamma_i/2$ by $\bar{\Gamma}_i$ everywhere. Further, the first order solutions for $\psi_1^{(1)}, \psi_3^{(1)}, \psi_4^{(1)}$ and $\psi_6^{(1)}$ are obtained by solving

\[(s + \bar{\Gamma}_2)\bar{\psi}_1^{(1)}(s) = i\Omega_{rf}\bar{\psi}_4^{(1)}(s) + i\Omega_1(2\bar{\psi}_7^{(0)}(s) + \bar{\psi}_8^{(0)}(s) + \bar{\psi}_9^{(0)}(s) - 1)\]
\[(s + \bar{\Gamma}_3)\bar{\psi}_4^{(1)}(s) = i\Omega_{rf}\bar{\psi}_1^{(1)}(s)\]
\[(s + \bar{\Gamma}_4)\bar{\psi}_6^{(1)}(s) = -i\Omega_{rf}\bar{\psi}_3^{(1)}(s)\]
\[(s + \bar{\Gamma}_3 + \bar{\Gamma}_4)\bar{\psi}_3^{(1)}(s) = i\Omega_3(\bar{\psi}_9^{(0)}(s) - \bar{\psi}_9^{(0)}(s)) + -i\Omega_{rf}\bar{\psi}_6^{(1)}(s)\]

The solution for $\rho_{ii}(t)$ depends on the choice of the initial condition which will be discussed in section III.

**Case (ii) Small $\omega_{rf}$.**

I next consider the case when the rf field is weak. Considering the contribution of $\Omega_{1,3}$ upto all orders and treating $\Omega_{rf}$ perturbatively, the second order equations are given by

\[(s + \bar{\Gamma}_2)\bar{\psi}_7^{(2)} = 2\Omega_1 Im\bar{\psi}_1^{(2)} + 2\Omega_{rf} Im\bar{\psi}_2^{(1)} + \bar{\psi}_8^{(2)} + \bar{\psi}_7^{(0)}\]
\[(s + \bar{\Gamma}_3)\bar{\psi}_8^{(2)} = -2\Omega_3 Im\bar{\psi}_3^{(2)} + 2\Omega_{rf} Im\bar{\psi}_2^{(1)} + \bar{\psi}_9^{(2)} + \bar{\psi}_8^{(0)}\]
\[(s + \bar{\Gamma}_4)\bar{\psi}_9^{(2)} = 2\Omega_3 Im\bar{\psi}_3^{(2)} + \bar{\psi}_9^{(0)}\]
\[(s + \bar{\Gamma}_3 + \bar{\Gamma}_4)\bar{\psi}_3^{(2)} = -i\Omega_{rf}\bar{\psi}_6^{(1)} - i\Omega_3(\bar{\psi}_9^{(2)} - \bar{\psi}_8^{(2)})\]
\[(s + \bar{\Gamma}_2)\bar{\psi}_4^{(2)} = -i\Omega_1(2\bar{\psi}_7^{(2)} + \bar{\psi}_8^{(2)} + \bar{\psi}_9^{(0)}) + i\Omega_{rf}\bar{\psi}_4^{(1)}\]

where the first order $\bar{\psi}_i$ are obtained by solving

\[(s + \bar{\Gamma}_3 + \bar{\Gamma}_2)\bar{\psi}_2^{(1)} = -i\Omega_1\bar{\psi}_4^{(1)} + i\Omega_3\bar{\psi}_6^{(1)}\]
for unit detector efficiency, where \( \sigma \) is a complete set of system operators \( \hat{O} \) and \( \Sigma \) which satisfies

\[
(s + \Gamma_3)\psi_4^{(1)} = -i\Omega_1\psi_2^{(1)} + i\Omega_3\psi_5^{(1)}
\]

\[
(s + \Gamma_2 + \Gamma_4)\psi_6^{(1)} = -i\Omega_1\psi_5^{(1)} + i\Omega_3\psi_2^{(1)}
\]

\[
(s + \Gamma_4)\psi_5^{(1)} = -i\Omega_1\psi_6^{(1)} + i\Omega_3\psi_4^{(1)}
\]

The solutions of this equation are discussed in the next section.

### 3 Correlation Function

Our interest is in determining the second order correlation function \( G_{ij}^{(2)}(r_1, t_1, r_2, t_2) \) which gives the correlation between the fluorescence signals \( I_i \) and \( I_j \) at times \( t_1 = t - r_1/c \) and \( t_2 = t - r_2/c \), and \( r_1, r_2 \) are the positions of the detectors. Here \( i,j=1,3 \) corresponds to the emission of the ground state and the upper excited state respectively. The correlation function is determined by evaluating the expectation values of the product of two-time operators \( \langle \hat{O}_{ij}^+(r_1, t_1)\hat{O}_{ij}^-(r_2, t_2)\hat{O}_{ij}^+(r_1, t_1) \rangle \) for \( i=1 \) and \( 3 \) (since we are interested only in the uppermost and the ground state transitions) where \( E^+ \) and \( E^- \) are the positive and negative frequency components of the field emitted.

The normalized temporal intensity correlation function \( g_{ij}^{(2)}(t_1, t_2) \) can be written in terms of the atomic operators in the far-zone approximation [13, 14] to be

\[
g_{ij}^{(2)}(t_1, t_2) = \langle \sigma_i^+(t_1)\sigma_j^+(t_2)\sigma_j^-(t_2)\sigma_i^-(t_1) \rangle / \langle \sigma_i^+\sigma_i^- \rangle_{ss}
\]

(5)

for unit detector efficiency, where \( \langle \sigma_i^+\sigma_i^- \rangle_{ss} \) corresponds to the steady state signal of the \( i \)th mode, \( \sigma_i^+ = |i+1 \rangle \langle i | \) and \( \sigma_i^- = |i \rangle \langle i+1 | \) being the atomic operators. The two-time expectation values are evaluated using the well known Onsager-Lax Quantum regression theorem [17] which states that if for a complete set of system operators \( \hat{A}_\mu, \mu = 1, 2...n \) the one time expectation value satisfies

\[
\langle \hat{A}_\mu(t) \rangle = \sum_\mu \mathcal{M}_{\mu\nu}(t, t') \langle \hat{A}_\nu(t') \rangle, t' < t
\]

(6)

then the two-time expectation value takes the form

\[
\langle \hat{O}_\alpha(t')\hat{A}_\mu(t)\hat{O}_\beta(t') \rangle = \sum_\nu \mathcal{M}_{\mu\nu}(t, t') \langle \hat{O}_\alpha(t')\hat{A}_\nu(t')\hat{O}_\beta(t') \rangle, t' < t
\]

(7)

for any two system operators \( \hat{O}_\alpha \) and \( \hat{O}_\beta \). Essentially the correlation functions (two-time averages) satisfy the same equation of motion as that of the expectation values of one-time averages [14]. The c-number coefficients \( \mathcal{M}_{\mu\nu} \) are derived from the solution of the Heisenberg equation. Using the operator algebra \( \sigma_{ij}\sigma_{kl} = \delta_{jk}\sigma_{il} \) and Eqn.(5), the two-time averages for the four-level cascade
Figure 2: (Color online) The cross-correlation functions a) $g_{31}^{(2)}(\tau)$, b) $g_{32}^{(2)}(\tau)$, and c) $g_{21}^{(2)}(\tau)$ function of $\gamma \tau$ with $\Omega_1 = \Omega_3 = 4\gamma$, and $\Omega_{rf} = 20\gamma$.

The system may be expressed as [15] [16] [14]

\begin{align*}
    g_{11}^{(2)}(\tau) &= <\rho_{22}(\tau) | \rho(0) = 1 < | 1 > / \rho_{22}^{ss} \\
    g_{33}^{(2)}(\tau) &= <\rho_{44}(\tau) | \rho(0) = 3 < | 3 > / \rho_{44}^{ss} \\
    g_{31}^{(2)}(\tau) &= <\rho_{22}(\tau) | \rho(0) = 3 < | 1 > / \rho_{22}^{ss}
\end{align*}

(8)

where $\tau = t_2 - t_1$. Here $\rho_{ii}^{ss}$ denote the steady state values. The functions $g_{11}^{(2)}(\tau)$ and $g_{33}^{(2)}(\tau)$ are the auto-correlation functions of the g.s. emission ($|1 > \rightarrow |2>$) and the upper excited state emission ($|4 > \rightarrow |3>$) respectively. The cross-correlation function $g_{31}^{(2)}(\tau)$ gives the conditional probability for the emission of a photon from the $|1 > \rightarrow |2>$ transition at a time $\tau$ given an emission of a photon from the $|3 > \rightarrow |4>$ transition at time $t = 0$. Here we have used the notation of reference [14]: $<\rho_{ii}(\tau) | \rho(0) = j < | j >$ denotes the population $\rho_{ii}$ at a time $\tau$ given the initial condition that the atom is prepared such that $\rho(0) = |j > < j |$. The time dependent functions $\rho_{ij}(\tau)$ in the general case is determined numerically while in some special cases I provide the analytical solutions. I first consider the case when $\Omega_{rf}$ is large. Solving the eq.(1) and (2) in the Laplace space wherein both $\Omega_1$ and $\Omega_3$ are treated perturbatively while $\Omega_{rf}$ is considered upto all orders , the correlation functions are given by

\begin{align*}
    g_{11}^{(2)}(\tau) &= 1 + \sum_{i=1}^{5} \bar{a}_i e^{-\alpha_i \tau} \\
    g_{31}^{(2)}(\tau) &= 1 + e^{-\Gamma_{47} \tau} (\bar{b}_0 + \bar{b}_1 e^{-\alpha_1 \tau} + \bar{b}_2 e^{-\alpha_2 \tau}) + \sum_{i=3}^{5} \bar{b}_i e^{-\alpha_i \tau} \\
    g_{33}^{(2)}(\tau) &= 1 + e^{-\Gamma_{47} \tau} (\bar{c}_0 + \bar{c}_1 e^{-\alpha_1 \tau} + \bar{c}_2 e^{-\alpha_2 \tau})
\end{align*}

(9)
Figure 3: (Color online) The time delay between the emission of the photons from the extreme transitions as a function of the driving field strengths i) $\Omega_1$ with $\Omega_2 = 12\gamma$ and $\Omega_3 = 4\gamma$, ii) $\Omega_2$ with $\Omega_1 = \Omega_3 = 4\gamma$, iii) $\Omega_3$ with $\Omega_1 = \Omega_2 = 4\gamma$. 
Figure 4: (Color online) The correlation functions $g_{11}^{(2)}(\tau)$, $g_{33}^{(2)}(\tau)$, and $g_{31}^{(2)}(\tau)$. 
where \( \bar{a}_i = a_i/a_N \) \( \bar{b}_i = b_i/b_N \) and \( \bar{c}_i = c_i/c_N \) is the normalisation factors \( a_N, b_N, c_N \). The coefficients are given by

\[
\begin{align*}
 a_N &= 2\Omega^2_r \Omega^2_{rf}/\Pi^{5}_{i=1}(\alpha_i) \\
 b_N &= 2\Omega^2_r \Omega^2_{rf}/\Gamma_4(\Pi^{5}_{i=1}(\alpha_i)) \\
 c_N &= \alpha_0/\bar{\Gamma}_4 \alpha_1 \alpha_2 \\
 c_0 &= -\alpha_0/\bar{\Gamma}_4(\bar{\Gamma}_4 - \alpha_1)(\bar{\Gamma}_4 - \alpha_2) \\
 c_1 &= -\alpha_0/\alpha_2(\alpha_1 - \alpha_2)(\bar{\Gamma}_4 - \alpha_2) \\
 c_2 &= \alpha_0/\alpha_1(\alpha_1 - \alpha_2)(\bar{\Gamma}_4 - \alpha_1) \\
 c_0 &= 2\Omega^2_r \bar{\Gamma}_4 + \Gamma_4
\end{align*}
\]

The roots in the exponent are given by the conjugate pairs

\[
\alpha_{1,2} = -\bar{\Gamma}_2 - \bar{\Gamma}_3 \pm i\phi; \phi = \sqrt{4\Omega^2_r - (\bar{\Gamma}_2 - \bar{\Gamma}_3)^2}
\]

and the cubic roots

\[
\begin{align*}
 \alpha_3 &= -\frac{2}{3\alpha} (\bar{\Gamma}_2 + \bar{\Gamma}_3) - 4\Omega^2_{rf} + \frac{\phi}{4} \\
 \alpha_{4,5} &= -\frac{2}{3\alpha} (\bar{\Gamma}_2 + \bar{\Gamma}_3) + 12(1 \pm i\sqrt{3})\Omega^2_{rf} - \frac{1 \pm i\sqrt{3}}{\alpha}
\end{align*}
\]

where

\[
\alpha = \frac{3}{4}(3\Omega^2_r(\bar{\Gamma}_2 + \bar{\Gamma}_3) + \frac{1}{\phi} \sqrt{324(\bar{\Gamma}_2 + \bar{\Gamma}_3)^2\Omega^4_{rf} + 6912\Omega^6_{rf}}) \frac{1}{2}
\]

Note that the coefficients \( c_i \) satisfy the condition

\[
\sum_{i=0}^{2} c_i + c_N = 0
\]

The coefficients \( a_i, b_i, i = 1, 5 \) are functions of \( \alpha_i \) and since the expressions are very lengthy we have not listed them here. However, the solution for \( \rho_{ii}(\tau) \) in the Laplace space which are displayed in the Appendix, yield an important identity amongst the coefficients

\[
\begin{align*}
 a_N + \sum_{i=0}^{5} a_i &= 0 \\
 b_N + \sum_{i=0}^{5} b_i &= 0
\end{align*}
\]

While the conditions for the coefficients \( a_i \) and \( c_i \) reflect the antibunching nature of the auto-correlation function, the condition for \( b_i \) implies the anti-bunching nature of the cross correlation function \( g_{31}^{(2)}(\tau) \). This contrasting feature which
is exhibited by a 4-level ladder system is unlike the behaviour of the cross correlation functions $g_{23}^{(2)}(\tau)$ in a 3-level cascade system which is non-zero at $\tau = 0$. A comparison of the various cross-correlation functions of a 4-level cascade system will be discussed in detail shortly.

The expressions in Eqn.(9) for the correlation functions indicate that the decoherence of $g_{33}^{(2)}(\tau), g_{31}^{(2)}(\tau)$ is controlled by the decay constants $\bar{\Gamma}_2, \bar{\Gamma}_3$ and $\bar{\Gamma}_4$ while the dephasing of the g.s. autocorrelation function is due to $\bar{\Gamma}_2$ and $\bar{\Gamma}_3$ only. The imaginary part in the exponents indicate that the the strength of the correlation oscillates with $\Omega_{rf}$.

In the limit of weak $\Omega_{rf}$ the solutions for the correlation function are given by

$$g_{11}^{(2)}(\tau) = 1 - e^{-\bar{\Gamma}_2 \tau} (\cos(2\Omega_1 \tau) - d_1 \sin(2\Omega_1 \tau))$$

$$g_{31}^{(2)}(\tau) = 1 + e^{-\bar{\Gamma}_2 \tau} (x_0 + x_1 \sin(\Omega_1 \tau) + x_2 \cos(\Omega_1 \tau) + \sum_{i=3}^{9} x_i e^{-\bar{\alpha}_i \tau})$$

$$g_{33}^{(2)}(\tau) = 1 + \sum_{i=0}^{6} y_i e^{-\bar{\alpha}_i + \tau}$$

(11)

where $d_1 = \bar{\Gamma}_2 d_0 / 2\Omega_1$, with $d_0 = 2\Omega_1^2 / (4\Omega_2^2 + \bar{\Gamma}_2^2)$. The roots $\bar{\alpha}_i, i = 3, 5$ are obtained by replacing $\bar{\Gamma}_2 \rightarrow \bar{\Gamma}_3, \bar{\Gamma}_3 \rightarrow \bar{\Gamma}_4$ and $\Omega_{rf} \rightarrow \Omega_3$ in $\alpha_i$. Further

$$\bar{\alpha}_{4,7} = - (\bar{\Gamma}_2 + \bar{\Gamma}_3 + \bar{\Gamma}_4) \mp \sqrt{\phi_3 + 2\phi_1 \phi_2}$$

$$\bar{\alpha}_{8,9} = - (\bar{\Gamma}_2 + \bar{\Gamma}_3 + \bar{\Gamma}_4) \mp i\sqrt{\phi_3 - 2\phi_1 \phi_2}$$

where

$$\phi_3 = (4(\Omega_1^2 + \Omega_3^2) - (\bar{\Gamma}_2^2 + \bar{\Gamma}_3^2 + \bar{\Gamma}_4^2 - 2\bar{\Gamma}_3 \bar{\Gamma}_4))$$

$$\phi_1 = \sqrt{(4\Omega_1^2 - \bar{\Gamma}_2^2)}$$

$$\phi_2 = \sqrt{4\Omega_3^2 - (\bar{\Gamma}_3 - \bar{\Gamma}_4)^2}$$

In this regime, the g.s. autocorrelation function resembles the second order correlation function of the usual two-level system. This is intuitively obvious since the upper level coupling is weak. Hence the dephasing of this function depends on $\bar{\Gamma}_2$. The strength of the correlation oscillates with $\Omega_1$ as shown by equation (10). While the dephasing of $g_{33}^{(2)}(\tau)$ and $g_{31}^{(2)}(\tau)$ depends on $\bar{\Gamma}_i, i = 2, 4$. The strength of the correlation now depends on $\Omega_1$ and $\Omega_3$.

It follows from the Eqn.(10) that $g_{11}^{(2)}(\tau) = 0$ for $\tau = 0$. Again, it may be verified using the solutions for $\rho_{44}, \rho_{00}, \rho_{33}$ and $\rho_{02} > \rho_{00}, \rho_{33}$ in the Laplace space given in the Appendix, that the auto-correlation function $g_{33}^{(2)}(\tau)$ and the cross-correlation function $g_{31}^{(2)}(\tau)$ are zero at $\tau = 0$. Since the expressions for $x_i$ and $y_i$ are too lengthy, we list all the solutions in the Laplace space in the Appendix. Thus, in either of the regimes, I would like to emphasize that the function $g_{31}^{(2)}(\tau) = 0$ at $\tau = 0$. This is further evident from the numerical results which are solved exactly and valid for all strengths of the
coupling fields (Fig 4c). Note that the cross-correlation is very strong for \( \gamma \tau < 1 \). Whenever the emission of the two photons occurs for time delay smaller than the lifetime of the atom, it necessarily means that both the photons belong to the same cascade emission. Hence the photons show very strong correlation for times smaller than \( \gamma^{-1} \). For larger time delay between the emissions of the two photons decoherence weakens the correlation.

Comparison of various cross-correlation functions

For easy comparison with the 3-level cascade system, we consider the most general 4-level system and label the emission from the \( |i + 1 \rangle \rightarrow |i \rangle \) as the \( i^{th} \) mode. The cross-correlation functions \( g_{ij}^{(2)}(\tau), i \neq j \) which are proportional to \( < \rho_{j+1,j+1}(\tau) > \rho(0)=|i>|<i| \) listed in the Appendix, give the probability of emission of photon in the \( j^{th} \) mode at a time \( \tau \) given the emission of a photon in the \( i^{th} \) mode at \( t=0 \). The function \( g_{21}^{(2)}(\tau) \) corresponds to the usual cross-correlation function of the 3-level cascade system and is nonzero at \( \tau = 0 \). Again \( g_{32}^{(2)}(\tau) \) is also nonzero at \( \tau = 0 \). This is easily understood by looking at the solutions in the Laplace space (see Appendix). Both \( g_{32}^{(2)}(\tau) \) and \( g_{21}^{(2)}(\tau) \) which are respectively proportional to \( < \rho_{32}(\tau) > \rho(0)=|3>|<2| \) and \( < \rho_{22}(\tau) > \rho(0)=|2>|<2| \) have contribution from terms like \( s^2/(s + \alpha_3)(s + \alpha_4)(s + \alpha_2) \) in the Laplace space the inverse transform of which has the form \( \sum_i A_i e^{-\alpha_i \tau} \) where \( A_i = \alpha_i^2/((\alpha_i-\alpha_j)(\alpha_i-\alpha_k)), i \neq j \neq k, i, j, k = 3, 5 \). Clearly, at \( \tau = 0 \), the \( \sum_{i=3}^{5} A_i \neq 0 \). I would like to recall here similar results due to Loudon \[5\] for a 3-level cascade system, where \( g_{21}^{(2)}(\tau) \) is nonzero at \( \tau = 0 \). On the other hand, the distinguishing feature \( g_{31}^{(2)}(\tau \rightarrow 0) = 0 \) is due to the absence of such terms. Thus, while \( g_{32}^{(2)}(\tau) \) and \( g_{21}^{(2)}(\tau) \) both show bunching, \( g_{31}^{(2)}(\tau) \) is zero at \( \tau = 0 \). The exact numerical solutions which are valid for all strengths of the coupling fields, is illustrated in Fig 2.

To understand the underlying mechanism behind this feature consider the limit \( \Omega_i f \rightarrow 0 \) and \( \Omega_i, i = 1, 2 \rightarrow 0 \). In this limit the correlation function \( g_{31}^{(2)}(\tau) \) is proportional to \( e^{-((\Gamma_3/2)\tau)} \). i. e. the correlation for the two photon emission pathway \( |3 \rangle \rightarrow |2 \rangle \rightarrow |1 \rangle \) depends on \( e^{-((\Gamma_3/2)\tau)} \). Likewise, the cross-correlation for the emission pathway \( |4 \rangle \rightarrow |3 \rangle \rightarrow |2 \rangle \rightarrow |3 \rangle \rightarrow |2 \rangle \rightarrow |1 \rangle \) is proportional to \( e^{-((\Gamma_3/2)\tau)} \) and hence both these correlations are nonzero at \( \tau = 0 \). On the other hand the \( g_{31}^{(2)}(\tau) = 2/((\Gamma_3-\Gamma_2)(e^{-((\Gamma_3/2)\tau)} - e^{-((\Gamma_3/2)\tau)}) \). This implies that the cross correlation of the photons emitted by the extreme transitions gets contribution from both the two-photon pathways. At \( \tau = 0 \) this function vanishes. In other words, there is an exact cancellation of emission due to the two pathways \( |4 \rangle \rightarrow |3 \rangle \rightarrow |2 \rangle \rightarrow |3 \rangle \rightarrow |2 \rangle \rightarrow |1 \rangle \) at \( \tau = 0 \). Thus the antibunching nature of \( g_{31}^{(2)}(\tau) \) is intrinsic to the four-level cascade system and is independent of the driving field strengths. Since this implies a finite time delay for the emission of the second photon and since the offset is provided by the strength of the driving fields (Fig 4c), in principle one has a handle on the control of this time delay \( \tau_d \). Fig 3 shows the variation of \( \tau_d \) with \( \Omega_i, i = 1, 3 \).
where \( \tau_d \) is the time delay at which the cross-correlation peaks. It is clear from Fig3 that larger the field strengths, smaller the time delay between the emissions. As \( \Omega_2 \) or \( \Omega_3 \) is increased the uppermost level gets populated much faster and hence the peak of the cross-correlation function occurs at earlier \( \tau_d \) (This is reflected in Fig4c (iii) and (iii) for \( \Omega_{rf} = 10\gamma \) and 20\( \gamma \)). While increasing \( \Omega_1 \) increases the population in level \( |2 > \) till saturation. Since this does not affect the cross-correlation significantly, the variation of \( \tau_d \) with \( \Omega_1 \) is not as significant as with \( \Omega_{2,3} \). The control will be better with the use of pulsed excitation with programmable time delay (of the order of a few \( \gamma \’s \)) between successive excitations [2, 3]. This provides an advantage over the other schemes like the photon down conversion or the usual three level cascade emission where the emission of both the photons is almost simultaneous (bunched) and hence there is no way of delaying the emission of the second photon after the first photon is emitted.

I would like to recall here that antibunching is the violation of the classical inequality: \( g^{(2)}(0) \geq g^{(2)}(\tau) \). This inequality is satisfied by ‘classical fields’, i.e., the Glauber-Sudarshan phase-space function corresponding to the states of the field has positive distribution. Violation of these inequalities would imply that the distribution function corresponding to the state is not well behaved (not positive definite) and are non-classical. Having demonstrated the antibunching nature of \( g_{31}^{(2)}(\tau) \) in this section, I next discuss the violation of yet another classical inequality namely, the Cauchy-Schwarz inequality.

**Violation of the Cauchy-Schwarz Inequality**

The other signature for the non-classical nature of the emitted fields is the violation of Cauchy-Schwarz inequality [6]. The degree of violation is determined by the ratio \( R = (g_{31}^{(2)}(\tau))^2 / g_{33}^{(2)}(0)g_{11}^{(2)}(\tau), \ R \leq 1 \) for a classical source. One needs to optimize the emissions in such a way that one achieves a large \( R_{max} \). In the present scheme, this condition essentially boils down to demanding sub-poissonian statistics in the \( g_{ii}^{(2)}(\tau) \) for \( i = 1, 3 \) -which ensures that the photons are emitted at spaced out intervals - in addition to large values for \( g_{31}^{(2)}(\tau) \). The function \( g_{ii}^{(2)}(\tau) \) can be tailored to show sub-poissonian statistics by making the coupling field weak ( \( \Omega_1 < \bar{\Gamma}_2 \) ) . The auto-correlation function \( g_{33}^{(2)}(\tau) \) shows sub-poissonian statistics even for \( \Omega_3 > \bar{\Gamma}_4 \) since the population of the excited levels is smaller. Fig4 shows the correlation functions and the ratio \( R \) for various parameter values. Here, the dimensionless variable \( t = \gamma \tau \) where \( \gamma = 2\pi MHz \). The function \( g_{31}^{(2)}(\tau) \) increases with \( \Omega_{rf} \) (Fig4 (c)), while both the auto-correlation functions decrease with \( \Omega_{rf} \) (Fig4 (a) and (b)). On the other hand, the time delay for the occurrence of peak value of \( g_{i1}^{(2)}(\tau) \) is smaller for larger values of \( \Omega_{rf} \). This means that, in principle, the occurrence of \( R_{max} \) can be delayed/advanced by decreasing/increasing the coupling field strengths - implying a trade-off between the value of \( R_{max} \) and the time delay for the occurrence of the same. Note that \( R_{max} = 10^3 - 10^6 \) as shown in Fig4 (d) and increases with \( \Omega_{rf} \). It is noteworthy that the oscillatory behaviour of \( R \) for large \( \Omega_{rf} \) provides different time delays at which the violation is maximum.
Lastly, the merit in this scheme is that the absorption profile being very narrow (of the order of the natural line-width) \([19]\), the excitation/deexcitation of the g.s. and the upper excited state occurs within a very narrow frequency band. This is because of the three photon resonance which induces atomic coherence. It has been predicted earlier \([11]\) that when a three-level cascade system showing EIT is coupled to a third driving field, the ground state (g.s.) shows a very narrow absorption within the EIT window at three photon resonance; the Doppler integrated absorption does not suffer much change in this geometry. This type of narrow absorption features and three-peak absorption, in four-level systems, have been subsequently reported experimentally \([12]\). An insight into this behavior is revealed by the study of the single atom dynamics \([20]\): when the middle transition coupling is strong (\(\Omega_{rf}\) in this case) the four level atom behaves like a two level atom spanned by levels \(|1>\) and \(|4>\) hence inducing a strong atomic coherence between these two levels. A steady state analysis \([19]\) further reveals that in an ensemble this feature manifests as a dominant contribution of the third order nonlinear susceptibility (due to the strong atomic coherence) as compared to the linear susceptibility. This third order nonlinear susceptibility is responsible for the efficient transfer of population to the uppermost level by inducing a narrow absorption in the ground state. The role of \(\Omega_{rf}\) is not only to transfer the population to level \(|4>\) but the strong atomic coherence it introduces is responsible for the strong correlation between the photons emitted by the extreme transitions.

The absorption features for the model under consideration is very similar, and since the absorption occurs within an EIT window, there is an absorption free zone \([19]\) for detunings larger than the line width of the central peak, and detunings less than the FWHM of the EIT window. i.e. for \(15MHz > |\Delta| > 6MHz\). This ensures cleaner signals and a reduction in the noise due to the absorption and emission into other modes close to the resonant frequency which is a major advantage in this system. I would like to mention here that if the coupling between the levels \(|2>−|3>\) were in the visible region instead of radio-frequency region then the absorption of the \(|3>−|4>\) transition diminishes due to Doppler averaging \([11]\). Even though this does not affect the antibunching nature of \(g_{31}(\tau)\), the strength of the correlation function would become weaker. Hence the signature for the violation of the Cauchy-Schwarz inequality may also become weaker.

To summarize, I propose a model for generating non-classical pairs, wherein Doppler effects are negligible at room temperatures, and also the absorption/emission proceeds in a narrow frequency band. The violation of the Cauchy-Schwarz inequality is orders of magnitude larger than the experimentally reported data, so far, and therefore could provide very clear signals in the near future. Another interesting feature is the antibunching nature of the cross-correlation between the emission of photons from the extreme transition which occurs because of the cancellation of emission due to different two-photon pathways. Thus the photon pair generated in this scheme shows the violation of two of the classical inequalities.
Appendix

The solutions for the density matrix elements in the Laplace space for various initial conditions and both the strong coupling of $\Omega_{rf}$ and weak $\Omega_{rf}$ are listed below.

**Strong $\Omega_{rf}$**

For the initial condition $\rho(0) = |3><3|$, the relevant $\rho_{ij}(\tau)$ in the Laplace space is given by

$$\mathcal{L}(\rho_{22}(\tau)) = \tilde{\psi}_7(s) = \frac{1}{d_3}[(s + \bar{\Gamma}_2 + \bar{\Gamma}_3 + 2\Omega_3^2)(1 + \frac{2\Omega_3^2}{d_4(s + \bar{\Gamma}_4)}(1 - s - \bar{\Gamma}_4)(s + \bar{\Gamma}_2 + \bar{\Gamma}_4)) + \frac{4\Omega_3^2\Omega_3^2}{d_4}(1 - s - \bar{\Gamma}_3)]$$

where $d_4 = \Pi_{i=1}^2(s + \bar{\Gamma}_4 + \alpha_i), d_3 = \Pi_{i=3}^2(s + \alpha_i), d_2 = \Pi_{i=1}^2(s + \alpha_i)$

$$\mathcal{L}(\rho_{33}(\tau)) = \tilde{\psi}_8(s) = \frac{1}{d_3}[(s^2 + \bar{\Gamma}_2^2 + \bar{\Gamma}_2\bar{\Gamma}_3 + s(\bar{\Gamma}_3 + 2\bar{\Gamma}_2) + 2\Omega_3^2(1 + \frac{2\Omega_3^2}{d_4(s + \bar{\Gamma}_4)}(s + \bar{\Gamma}_2 + \bar{\Gamma}_4)) + (1 - s - \bar{\Gamma}_4)) + \frac{2\Omega_3^2\Omega_3^2}{d_4}(s + \bar{\Gamma}_2)]$$

For the initial condition $\rho(0) = |2><2|$ we have

$$\mathcal{L}(\rho_{22}(\tau)) = \tilde{\psi}_7(s) = \frac{1}{d_3}[(s^2 + \bar{\Gamma}_2^2 + s(\bar{\Gamma}_2 + 2\bar{\Gamma}_3) + \bar{\Gamma}_2\bar{\Gamma}_3 + 2\Omega_3^2) + \frac{1}{sd_2}(2\Omega_3^2\Omega_3^2(s - 1) - 2\Omega_1^2(s + \bar{\Gamma}_3)(s^2 + \bar{\Gamma}_3^2 + s(\bar{\Gamma}_2 + 2\bar{\Gamma}_3) + 2\Omega_3^2 + \bar{\Gamma}_3\bar{\Gamma}_2))]$$

For the initial condition $\rho(0) = |1><1|$ we get

$$\mathcal{L}(\rho_{22}(\tau)) = \tilde{\psi}_7(s) = \frac{2\Omega_3^2}{sd_2d_3}\left[\Omega_3^2 + (s + \bar{\Gamma}_3)((s + \bar{\Gamma}_3 + \bar{\Gamma}_2)(s + \bar{\Gamma}_3) + \Omega_2^2))\right]$$

**Weak $\Omega_{rf}$**

For the initial condition $\rho(0) = |3><3|$ we have

$$\mathcal{L}(\rho_{22}(\tau)) = \tilde{\psi}_7(s) = \frac{1}{d_3}\left[\frac{2\Omega_3^2}{s} + 2\Omega_1\Omega_2C_1 - 2(s + \bar{\Gamma}_2)\Omega_2C_2 + \frac{4\Omega_3^2}{d_3p}(-\Omega_3^2 + \Omega_2\Omega_3((s + \bar{\Gamma}_3)C_3 - 2\Omega_3C_2))\right]$$

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$$+ \frac{1}{(s+\Gamma_2)d_{4p}} \left( \Gamma_2^2 + s(s-2\Omega_2^2) + 2\Gamma_2(s-\Omega_1^2) \right)$$
$$(1 + 2\Omega_2 C_2)(s^2 + \Gamma_1^2 + \Gamma_3(s + \Gamma_4) +$$
$$2\Gamma_4 s + 2\Omega_2^2) + 2C_3\Omega_2\Omega_3(s + \Gamma_4 - 1))$$

$$\mathcal{L}(\rho_{33}(\tau)) = \bar{\psi}_3(s)$$
$$= \frac{1}{d_{3p}}[(1 + 2\Omega_2 C_2)(s^2 + \Gamma_3^2 + \Gamma_3(s + \Gamma_4) +$$
$$2\Omega_2^2 + 2s\Gamma_4) + 2\Omega_2\Omega_3\Omega_3(s + 2\Gamma_4 + 1)]$$

$$\mathcal{L}(\rho_{44}(\tau)) = \psi_4(s)$$
$$= \frac{2\Omega_3}{d_{4p}}[\Omega_3(1 + 2\Omega_2 C_2) - \Omega_2\Omega_3(s + \Gamma_4)]$$

For the initial condition $\rho(0) = |2><2|$ we have

$$\mathcal{L}(\rho_{22}(\tau)) = \bar{\psi}_7(s)$$
$$= \frac{1}{d_{2p}}[2\Omega_2^2] + 2\Omega_1\Omega_2 C_1 - (s + \Gamma_2)(1 -$$
$$2\Omega_2 C_2) + \frac{4\Omega_1^2\Omega_2^2}{d_{3p}}(((s + \Gamma_3)C_3 - 2\Omega_3 C_2))$$
$$+ \frac{1}{(s+\Gamma_2)d_{3p}}(\Gamma_2^2 + s(s - 2\Omega_2^2) + 2\Gamma_2(s - \Omega_1^2)$$
$$(2\Omega_2 C_2(s^2 + \Gamma_3^2 + \Gamma_3(s + \Gamma_4) + 2\Gamma_4 s$$
$$+ 2\Omega_3^2 + 2C_3\Omega_2\Omega_3(s + \Gamma_4 - 1)))]$$

For the initial condition $\rho(0) = |1><1|$ we have

$$\mathcal{L}(\rho_{22}(\tau)) = \psi_7(s) = 2\Omega_2^2/(sd_{2p})$$

where

$$C_1 = -\Omega_1\Omega_2[(s + \Gamma_4)(s + \Gamma_4 +$$
$$\Gamma_2) + \Omega_1^2 - \Omega_2^2]/d_{4p}$$

$$C_2 = -\Omega_2[(s + \Gamma_4)(s + \Gamma_3)(s + \Gamma_4 + \Gamma_2) +$$
$$\Omega_2^2(s + \Gamma_2 + \Gamma_4) + (s + \Gamma_3)\Omega_3^2]/d_{4p}$$

$$C_3 = \Omega_2\Omega_3[(s + \Gamma_3)(s + \Gamma_4) +$$
$$\Omega_2^2 - \Omega_3^2]/d_{4p}$$

Here $d_{4p} = \Pi_{i=6}^{9}(s + \bar{\alpha}_i)$, $d_{3p} = \Pi_{i=3}^{5}(s + \bar{\alpha}_i)$ and $d_{2p} = (s + \Gamma_2^2 + 4\Omega_2^2$).

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