Effects of chirped laser pulses on nonclassical correlations and entanglement of photon pairs

C. H. Raymond Ooi

Department of Physics, Korea University, Anam-dong, Seongbuk-gu, Seoul 136–713, Republic of Korea
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We study the effects of arbitrary laser pulse excitations on quantum correlation, entanglement, and the role of quantum noise. The transient quantities are computed exactly using a method that provides exact solutions of the Langevin field operators for photon pairs produced by a double Raman atom driven by laser pulses. Short pulses with appropriate chirping, delay, and/or detuning can generate broadband photon pairs and yield results that provide insights on how to generate very large nonclassical correlation. We find that short pulses are not favorable for entanglement. Photon correlation and entanglement are favored by exclusively two different initial conditions. We find rapid variations of entanglement with pulse width and strength. Explanation is given based on the pulse area concept and the phase-sensitive nature of entanglement. Analysis of results reinforces our understanding of the two nonclassical concepts and the nature of quantum noise.

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I. INTRODUCTION

The intriguing nature of atoms and photons manifest through entanglement and quantum correlation. These nonclassical concepts carry subtly different physical meanings although they are mathematically related for the two-mode Gaussian state [1]. As pointed out by Glauber, quantum correlation exists when the density matrix is not a product form $\Pi_{i,j}(\alpha_i|\alpha_j)$ (in coherent state basis), rather in an incoherent superposition $\Sigma_{\alpha} w(\alpha)|\alpha\rangle\langle\alpha|$ or mixed state [2]. Entanglement is commonly depicted as a superposition of two mutually inverted states. The physical mechanisms that create quantum correlation and entanglement may not be the same and not fully understood.

We consider a single atom with four-level double $\Lambda$ system (Fig. 1) driven by laser pulses. This scheme is interesting for being able to generate controllable nonclassically correlated Stokes and anti-Stokes photons from an atom [3] as well as extended medium [4]. Previous works on a single atom driven by continuous wave (c.w.) lasers were based on Schrödinger’s equation [3,5], and master equation [6]. The former approach yields solutions only for c.w. lasers. The latter uses atomic correlation to obtain field correlations indirectly via quantum regression theorem [7]. A general and more transparent method for computing correlations is based on the Heisenberg-Langevin (HL) formulation, which can be extended to include spatial propagation. However, the previous studies have not considered excitations with laser pulses.

Laser pulses provide extra degrees of freedom for coherent control of nonclassicality. It is interesting to explore the effects of pulse width (duration) and other parameters such as chirp, phase, and shape on quantum correlation and entanglement of photon pairs, as well as on the role of quantum noise [8]. Nonclassical photon pairs driven by laser pulses could be made sufficiently intense and serves as a new tool for doing quantum nonlinear optics. A scheme to generate single-cycle photon pairs has been proposed [9]. For (short and intense) laser pulses, exact solutions cannot be obtained using Schrödinger’s equation. The quantum regression approach may give numerical solutions for pulses, but cannot account for spatial propagation. The ultimate goal is to compute quantum correlation for extended medium driven by arbitrary laser pulses.

As the first step toward this goal, we present a general method that gives exact transient solutions for quantum operators $\hat{a}_k$ and $\hat{a}_q$ of photon pairs in HL equations for a single atom. The solutions enable us to study the effects of various laser pulse parameters on the transient quantum correlation and entanglement of the photon pairs. We obtain interesting results that reflect the unique advantages of using pulses, thus showing the inherent advances in extending the previous studies to pulsed excitations. We find that chirped and overlapping laser pulses can give an extremely large photon correlation. In addition, short pulses tend to increase the photon correlation but reduces the entanglement. We give physical explanations that shed insights on the effects of finite spectral bandwidth on the nonclassical properties. By analyzing the main results that contain rich underlying physics, we acquire conceptual insights on (i) coherent control of nonclassical photons, (ii) the differences between nonclassical correlation and entanglement of photon pairs, and (iii) the effects of pulses on the role of quantum noise.

II. THEORETICAL FORMULATION

The usual Hamiltonian for an atom in Fig. 1 interacting with radiation fields and two laser fields is

$$\hat{H} = \sum_{k,q} \left( \hat{a}_k^\dagger \hat{a}_k \right) + \sum_{p,c} \left( \hat{a}_p^\dagger \hat{a}_p \right) - \sum_{p,c} \left( \hat{a}_p^\dagger \hat{a}_p \right) \omega_p \tau_p - \sum_{p,c} \left( \hat{a}_p^\dagger \hat{a}_p \right) \frac{\Omega_p}{2} + \sum_{d} \left( \hat{a}_d^\dagger \hat{a}_d \right) \frac{\Omega_d}{2}$$

FIG. 1. (Color online) An atom with four-level double Raman scheme is driven by pump (p) and control (c) laser pulses, producing Stokes (k) and anti-Stokes (q) photons with transient quantum correlation and entanglement.
\[ \dot{\bar{\sigma}}_q(t) = -i \gamma_q \bar{\sigma}_q(t), \]

where \( \bar{\sigma}_q(t) = \bar{\sigma}_q e^{i \gamma_q t} \) and \( \bar{\sigma}_q(t) = \dot{\bar{\sigma}}_q e^{-i \gamma_q t} \).

### III. Method for Exact Solutions

Exact analytical solutions for \( \bar{\sigma}_k \) and \( \bar{\sigma}_q \) can be found from the exact solutions of \( \bar{\sigma}_{ab}(t) \) and \( \bar{\sigma}_{ca}(t) \) (since these equations are linear) but the solutions are very cumbersome and not helpful. Adiabatic approximation may simplify the solutions but it will considerably limit the range of operating parameters. We outline a method which provides exact solutions for the field operators. First, note that the time evolution of the density matrix elements can be solved numerically as \( X(t) = \langle X(t) \rangle = \mathcal{U}(t) \langle X(0) \rangle \), where \( \langle X(0) \rangle = \{ \bar{\sigma}_{ab}^{(0)}(0) \} \) where \( a \beta = a \alpha, \ldots, a b, a, \ldots, b, b, \ldots, d, d \) is the initial vector of the atomic operators, and \( \mathcal{U}(t) \) is the time-evolution matrix whose exact form can only be obtained numerically from the solutions \( \rho_{\alpha \beta} = \langle \bar{\sigma}_{\alpha \beta} \rangle \) of the density matrix equations. We then find the exact solutions

\[ \bar{\sigma}_k(t) = \bar{\sigma}_k(0) - i g_k \sum_{m=1}^{16} \left[ K_m(t) \bar{X}_m(0) + \int_0^t K_m(t') \bar{F}_m(t-t') \, dt' \right], \]

where \( K_m(t) = \int_0^t U_{14,m}(t') \, dt' \) and \( \bar{Q}_m(t) = \int_0^t U_{23,m}(t') \, dt' \). This technique for obtaining exact operator solutions is applicable to an arbitrary time dependent field interacting with an atom. It may be extended to molecules in future works.

The single operator expectation values in thermal vacuum are \( \langle \hat{a}_q(t) \rangle = -i g_q \sum_{m=1}^{16} K_m(t) \langle \bar{X}_m(0) \rangle \) and \( \langle \hat{a}_k(t) \rangle = i g_k \sum_{m=1}^{16} Q_m(t) \langle \bar{X}_m(0) \rangle \). From the solutions Eqs. (5) and (6), we can compute the expectation values for the products of two operators. For example,

\[ \langle \hat{a}_q(t) \hat{a}_k(t) \rangle = - g_k g_q \sum_{m,n=1}^{16} \left[ K_m(t) K_n(t) \langle \bar{X}_m(0) \bar{X}_n(0) \rangle \right] + \int_0^t Q_m(t-T') K_n(t-t') 2D_{mn} \langle t' \rangle \, dt', \]

where \( \bar{Q}_k = \bar{Q}_k(t) = \langle \bar{X}_k(t) \rangle \) and \( \bar{Q}_q = \bar{Q}_q(t) = \langle \bar{X}_q(t) \rangle \) are the normal ordered and anti-Stokes photon numbers at initial time. The normal ordered and antinormal ordered diffusion coefficients are defined as \( D_{mn}(t) = \langle \bar{X}_m(t) \bar{X}_n(t) \rangle \) and \( D_{mn}^*(t) = \langle \bar{X}_m(t) \bar{X}_n(t) \rangle \), respectively. The final terms that contain the diffusion coefficients are the noise (superscript n) parts, i.e., \( \bar{Q}_k^n \) and \( \bar{Q}_q^n \), while the former terms are referred to as the boundary parts \( \bar{Q}_k^b \) and \( \bar{Q}_q^b \). We also use \( \bar{\sigma}_{ab}^{(0)}(0) = \langle \bar{\sigma}_{ab}^{(0)}(t) \rangle \) and \( \bar{\sigma}_{ca}^{(0)}(0) = \langle \bar{\sigma}_{ca}^{(0)}(t) \rangle \). We have assumed that initially the modes are statistically independent: \( \langle \hat{a}_q(0) \hat{a}_k(0) \rangle = 0 \) and are not squeezed, i.e., \( \langle \hat{a}_q^2(0) \rangle = \langle \hat{a}_k^2(0) \rangle = 0 \).

### IV. Nonclassical Quantities

The average value of the paired operators are computed to obtain mean photon numbers, photon-photon correlation, and to determine entanglement. The effects of finite pulse duration, chirping and pulse sequence on these quantities can be studied. We also analyze the effects of pulses on the contributions of the noise operators compared to the initial operators.

#### A. Nonclassical photon correlation

The solutions Eqs. (5) and (6) are linear combinations of initial field operators and atomic noise operators. Thus the two-photon correlation function can be decorrelated into terms with paired operators. The presence of nonclassical two-photon correlation between the Stokes and anti-Stokes
photon is determined by the condition \( g^{CS} > 1 \) for the Cauchy-Schwarz correlation
\[
g^{CS}(t) = \frac{\langle \hat{a}_j(t)\hat{a}_k(t)\rangle\langle \hat{a}_j(t)\hat{a}_k(t) \rangle}{\sqrt{G^{(2)}_j(t)G^{(2)}_k(t)}} = \frac{\langle \hat{a}_j(t)\hat{a}_j(t) \rangle^2 + \langle \hat{a}_j(t)\hat{a}_k(t) \rangle}{\sqrt{\langle \hat{a}_j(t)\hat{a}_j(t) \rangle^2 + \langle \hat{a}_j(t)\hat{a}_k(t) \rangle^2}}, \tag{9}
\]
where \( G^{(2)}_j(t) = \langle \hat{a}_j(t)\hat{a}_j(t) \rangle \) and we omit the \( t \) dependence to simplify the notation. Note that Eq. (9) corresponds to transient coincident (joint) two-photon detection (with no delay between the Stokes and anti-Stokes) which is mathematically related to an entanglement criteria Eq. (10) below.

B. Entanglement

The above solutions Eqs. (5) and (6) are also used to identify the presence of entanglement via \( D(t) = \langle (\Delta\hat{n})^2 \rangle + \langle (\Delta\hat{c})^2 \rangle \). This is sufficient and necessary criteria for two-mode Gaussian states. We compute the paired operators: \( \langle \hat{a}_j(t)\hat{a}_k(t) \rangle, \langle \hat{a}_j(t)\hat{a}_k(t) \rangle, \langle \hat{a}_j(t)\hat{a}_k(t) \rangle, \langle \hat{a}_j(t)\hat{a}_k(t) \rangle \) just like Eqs. (7) and (8). Note that the finite mean thermal photon numbers \( \bar{n}_q^{th} \) or \( \bar{n}_q^{th} \) effectively increases the value \( D \). Thus entanglement may not be obtained when there is a significant number of thermal photons.

Let use recall the steady state for two-photon state \( |\Phi \rangle = \sum_{k,q} C_{kq}\hat{c}_{1k}\hat{c}_{1q} \) [3]. For the Raman-EIT scheme which shows nonclassical correlation, the coefficient is inseparable \( C_{kq} \neq C_kC_q \) [5]. Thus one might expect to find entanglement in the steady state as well as in the transient. However, we find no entanglement for the Raman-EIT scheme at all times and with any control field \( \Omega \) and phase \( \phi \) of the laser. We emphasize that the inseparability of \( C_{kq} \) only implies correlation, and may not give entanglement depending on the complex phase \( \phi \) of \( \langle \hat{a}_j(t)\hat{a}_k(t) \rangle \) through [1]
\[
D = 2[1 + \bar{n}_k + \bar{n}_q + 2\sqrt{\bar{n}_k\bar{n}_q}(g^{(2)} - 1)\cos \phi_{kq}] \tag{10}
\]
which relates the quantity \( D \) with normalized correlation \( g^{(2)} = \langle \hat{a}_j(t)\hat{a}_k(t)\rangle / \bar{n}_q\bar{n}_k \).

V. RESULTS

Based on the generated Figs. 2–7, we discuss the effects of laser pulses on nonclassical photon correlation, entanglement, and the contribution of quantum noise.

A. Nonclassical photon correlation

Several interesting effects are found for excitations with ultrashort pulses. First, let us compare the results (Fig. 2) with c.w. lasers and pulsed lasers for identical and resonant
lasers: $\Delta_p=\Delta_c=0, \Omega_p=\Omega_c$ referred to as double resonant Raman (DRR). Continuous-wave lasers give a Fresnel-like oscillations between classical and quantum regimes, showing no apparent nonclassical correlation [Fig. 2(a)]. The oscillations have a period $\pi/\Omega_{pc}$ are due to optical nutation [10]. In the case of pulsed excitations, there are no oscillations due to the adiabaticity of the pulses.

1. Effects of finite pulse width

Laser pulses give nonclassical correlation much larger [Fig. 2(b)] than the correlation from c.w. laser. This is one of the main results. The correlation increases substantially for pulse width $\sigma$ shorter than $0.1/\Gamma_{ac}$ [Fig. 3(a)]. The physical explanation comes from two points: (i) a short pulse carries a large bandwidth, (ii) correlation is a measure of the amount of spectral interrelation between the Stokes and anti-Stokes photons.

2. Effects of pulse detuning

Pulses with finite detunings $\Delta_p, \Delta_c \gg \Omega_{ac}$ produce large nonclassical correlation [Fig. 3(b)]. Here, the bandwidth of the generated pulses of photons are enhanced by another mechanism (in addition to the short pulse duration); i.e., the spontaneous Raman processes from the two laser pulses. However, for the c.w. case, the correlation does not depend on the Rabi frequencies regardless of whether there is detuning or not.

3. Effects of chirping and delay of pulses

We now analyze the effects of chirping of the pulses. We consider the chirped carrier frequency of the form $\nu(t) = \nu_0 + \alpha \nu t$ ($x=p, c$). Figure 4(a) serves to remind us of the spectral content of positively chirped and negatively chirped pulses.

FIG. 4. (Color online) (a) Spectral content of chirped carrier frequency, computed from the Fourier transform of $\exp[i(\nu t + \alpha \nu t^2)]$. Effects of chirping on correlation profile for (b) coincident pulses and identical chirp, (c) coincident pulses and opposite chirp, (d) partially overlapping (pump precedes control) pulses and opposite chirp. Other parameters are the same as in Fig. 2(b).

FIG. 5. (Color online) Entanglement between the Stokes and anti-Stokes photon pairs for the DRR scheme using (a) continuous wave (c.w.) lasers, (b) coincident pulses, overlapping pulses with (c) intuitive sequence, (d) counterintuitive sequence. The overlapping pulses are shown on top of the figure. Parameters are $\sigma_{pc} = (2.1\Gamma_{ac})^{-1}, \Delta_p = 0, \Omega_p = 30\Gamma_{ac}, \gamma_0 = 0$ and $\rho_{ic}(0) = 1$. 

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When the pulses are chirped identically ($\alpha_p=-\alpha_c$), there is no significant effect on the correlation [Fig. 4(b)]. For opposite chirping ($\alpha_p=\alpha_c$) and sufficiently large $|\alpha_{p,c}|$, the correlation shows a peak. This can be understood qualitatively from Fig. 4(a). The two-spectral “wings” span an effectively broad spectrum of photons that resulted in a large correlation. The correlation can be further enhanced by displacing the control pulse slightly such that both pulses overlap partially. This yields an extremely large transient “correlation pulse.” If the pulses are entirely separated, the chirping has no enhancement effect on the correlation.

The Raman-EIT scheme has a very large $g^{CS}$ at times much smaller than the lifetime $\Gamma^{-1}$, but decays rapidly to zero for large times, corresponding to steady state photon antibunching found in previous studies [3].

### B. Entanglement

Figure 5 shows the entanglement for different pulse sequences for the DRR scheme.

#### 1. Favorable initial condition

The symmetrical initial condition $p_c(0)=p_b(0)=0.5$ that gives quantum correlation does not give entanglement. We obtain entanglement with a different initial condition $p_c(0)=1$. This initial condition ensures that the Stokes photon is produced first before an anti-Stokes photon since there is little or no population in level $b$. But a more important point that explains entanglement is that, the state of the atom which generates the anti-Stokes photon carries quantum information about the Stokes photon. The simultaneous excitations by lasers ensures that both Stokes and anti-Stokes are driven with the same phase. This explains how identical lasers configuration and $p_c(0)=1$ can generate entangled photon pairs.

#### 2. Effect of pulses on entanglement

The increased spectral bandwidth due to the pulses may be favorable for the correlation [see Fig. 2(b)] but not for the entanglement. Coincident pulses give entanglement while partially overlapping pulses in both intuitive and counterintuitive sequences do not give entanglement.

Figure 6 illustrates how the entanglement parameter $D$ varies with pulse width $\sigma_{p,c}$ and laser field $\Omega_{p,c}$ for the DRR scheme. The oscillatory feature implies that the presence of entanglement could be very sensitive to the variations of $\sigma_{p,c}$.
and $\Omega_{pc}$. We have also plotted the $D$ vs $\Omega_{pc}$ and time using a different pulse width of $\sigma_{pc}=15\Gamma_{ac}$ and our analysis shows that the oscillations in the transient entanglement can be understood via the pulse area concept. Previous work shows that $D$ is phase sensitive [1]. Thus the entanglement would be affected by the amplitude $\sigma_{pc}\Omega_{pc}$ of the pulse area which manifests as a phase.

C. Quantum noise

Figures 7(a) and 7(c) show the Stokes photon number for single Raman transition with only the pump field. As expected, the contribution of quantum noise is significant for resonant pump field [Fig. 7(a)] but negligible for spontaneous Raman (weak and far detuned pump) [Fig. 7(c)]. This shows that quantum noise is important and contributes to the photon numbers when the field is resonant and gives rise to spontaneous emission.

If a pump pulse is used instead, the Stokes photon number increases significantly but the proportion of the noise contribution is reduced [Fig. 7(b)]. This can be understood: the pump pulse corresponds to a range of frequencies, but only the resonant frequency contributes significantly to the noise through spontaneous emission while most frequencies that are off-resonant undergo spontaneous Raman process which gives very little noise.

When a resonant c.w. control field is applied (in addition to the pump field) we have anti-Stokes photons in the Raman-EIT (electromagnetic induced transparency) scheme ($\Delta_p=\Omega_p$, $\Delta_c=0$) [4]. The contribution of the noise to the Stokes photon number becomes significant despite the large pump detuning [see Fig. 7(d)]. This seems surprising, but can be explained as due to the noise in population level $c$ due to the spontaneous emission noise of the anti-Stokes photons.

VI. CONCLUSIONS

To summarize, we have presented a method to obtain exact solutions of photon operators that include quantum noise. The method enables exact computation of various transient quantities that involve products of photon operators such as photon correlation and entanglement. The results enable us to study the effects of pulsed excitations on the nonclassical photon correlation and entanglement. Short laser pulses produce much larger quantum correlation compared to c.w. lasers. Very large correlation can be obtained for finite detuning of the pulses. Most importantly, we have shown how the laser pulse parameters like chirping, duration, and pulse sequence can give an extremely large nonclassical photon correlation. Chirping of the pump and the control lasers in an opposite manner increases the correlation. The correlation increases further for partially overlapping pulses. We provided explanations based on the spectral content and large bandwidth of the laser pulses that conspire to enhance the photon correlation. We learned that certain initial conditions of the atom can generate transient entanglement but not correlation, and vice versa. Identical laser pulses with the initial condition $\rho_{cc}(0)=1$ can generate entangled photon pairs. We apply the pulse area concept to explain the oscillations of the transient entanglement with pulse width and the laser field. We have also found that the laser pulses tend to reduce the contribution of quantum noise to the photon number. All these results provide conceptual insights that show the significance of pulsed excitations on quantum properties.