

Photon-Photon Correlations as a Probe of Vacuum Induced Coherence Effects

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We present new experimental implications of the effects of vacuum induced coherence on the photon-photon correlation in the π -polarized fluorescence in $j = 1/2$ to $j = 1/2$ transition. These effects should be thus observable in measurements of photon statistics in for example Hg and Ba ion traps.

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I. INTRODUCTION

An early work [1] had predicted a very unusual effect of quantum interference in the problem of spontaneous emission. It was for example shown that in a degenerate V-system one could get population trapping and generation of quantum coherences in the excited states. One of the key conditions for the occurrence was that the dipole matrix elements of the two transitions from the excited states were orthogonal. The later condition is difficult to meet though very large body of theoretical literature has been devoted to the subject of vacuum induced coherences [2, 3, 4, 5, 6]. It was also suggested how the above condition on dipole matrix elements can be bypassed if we consider anisotropic vacuum [7] which for example would be the case while considering emission from excited atoms on nano particles [8]. It is clearly important to find out easily realizable systems so that experimental results can be obtained. Kiffner. et. al.[9] showed that one very important case would involve $j = 1/2$ to $j = 1/2$ transition. They calculated the spectrum of the emitted radiation and showed how vacuum induced coherences change the spectrum of the emitted radiation. There are many systems where it is easy to find $j = 1/2$ to $j = 1/2$ transition. In fact in an early experiment of Eichmann et. al.[10] such transitions in $^{198}\text{Hg}^+$ were used to examine interferences in a system of two ions. A recent work uses the $j = 1/2$ to $j = 1/2$ transition in $^{138}\text{Ba}^+$ ions [11]. While the results of Kiffner. et. al. on the spectrum are quite interesting the current experimental efforts [11, 12, 13] are more focussed on the study of photon-photon correlations. Thus an important question would be – do the vacuum induced coherences significantly affect the photon-photon correlations? This is the question we answer in affirmative.

It may be added that the photon-photon correlations have acquired new significance in the context of quantum information processing and quantum imaging as well as in interferences from independent atoms [12, 13, 14]. Thus it is important to have a consistent calculation of such correlations in situations where vacuum induced coherence (VIC) effects are important.

The organization of this paper is as follows-In Sec 2 we introduce the model and present the working

equations. In Sec 3 we calculate the photon-photon correlations both in presence and in absence of the vacuum induced interference effects. In Sec 4 we present numerical results to highlight the effects of vacuum induced coherences on photon-photon correlations. In Sec 5 we conclude with the outlook and future directions.

II. MODEL

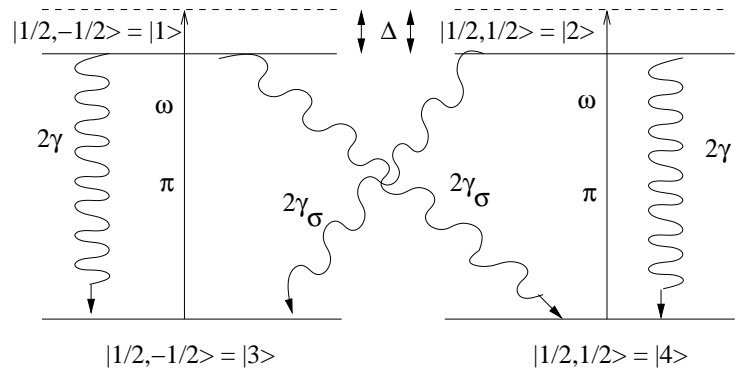


FIG. 1: Schematic diagram of a four-Level atom modelled by $j = 1/2$ to $j = 1/2$ transition

The Fig.1. shows the level scheme of a four-level atom modelled by $j = 1/2$ to $j = 1/2$ transition. This kind of level scheme is easily realizable and has already been studied, for example in $^{198}\text{Hg}^+$ [10] and $^{138}\text{Ba}^+$ [11] ions. The ground level is $6s^2S_{1/2}$ and the excited level is $6p^2P_{1/2}$. Each of these levels are two fold degenerate. The transitions $|1\rangle \leftrightarrow |4\rangle$ and $|2\rangle \leftrightarrow |3\rangle$ couple to σ^+ and σ^- polarized light respectively. The transitions $|1\rangle \leftrightarrow |3\rangle$ and $|2\rangle \leftrightarrow |4\rangle$ couple to light linearly polarized along the \mathbf{e}_z and their dipole moments are antiparallel. The spontaneous decays of the excited state to the two ground states are given by 2γ and $2\gamma_\sigma$ as shown in the figure. The electric dipole moment operator for this level scheme is defined as

$$\mathbf{d} = \sum_{ij} \mathbf{d}_{ij} A_{ij} ,$$

$$A_{ij} = |i\rangle\langle j|. \quad (i, j = 1, \dots, 4) \quad (1)$$

The non-vanishing matrix elements of the electric dipole moment operator \mathbf{d} can be found using the Wigner-Eckart theorem and are given by,

$$\begin{aligned}\vec{d}_{31} &= -\vec{d}_{42} = -\frac{1}{\sqrt{6}}\mathcal{D}\hat{e}_z, \\ \vec{d}_{41} &= \vec{d}_{32}^* = \frac{1}{\sqrt{3}}\mathcal{D}\hat{e}_-, \end{aligned} \quad (2)$$

with $\hat{e}_- = (\hat{x} - i\hat{y})/\sqrt{2}$. In Eq. (2) \mathcal{D} denotes the reduced matrix element of the dipole moment operator \mathbf{d} . The four-level system is driven by a π polarized monochromatic field of frequency ω ,

$$\mathbf{E}(t) = \vec{\mathcal{E}}_0 e^{-i\omega t} \mathbf{e}_z + c.c., \quad (3)$$

where c.c is the complex conjugate. With this particular choice of polarization, the driving field couples only to the two antiparallel dipole moments \vec{d}_{31} and \vec{d}_{42} . The total Hamiltonian for this atom-field system is then given by

$$\mathcal{H} = \mathcal{H}_A + \mathcal{H}_I, \quad (4)$$

where the unperturbed Hamiltonian for the atom is,

$$\mathcal{H}_A = \hbar \sum_{i=1}^4 \omega_i |i\rangle \langle i|, \quad (5)$$

and the interaction Hamiltonian for this system is given by

$$\begin{aligned}\mathcal{H}_I &= -\mathbf{d} \cdot \mathbf{E}(t) \\ &= \hbar \Omega_c (|1\rangle \langle 3| - |2\rangle \langle 4|) e^{-i\omega t} + H.c., \end{aligned} \quad (6)$$

where H.c is the Hermitian conjugate and Ω_c is the Rabi frequency defined by

$$\Omega_c = \frac{\vec{d}_{42} \cdot \vec{\mathcal{E}}_0}{\hbar}, \quad (7)$$

The time evolution of this four level system is investigated by studying the density matrix equation. The spontaneous emission is included via the master equation techniques. Following the standard procedure [1] we obtain,

$$\begin{aligned}\dot{\rho} &= -\frac{i}{\hbar} [\hat{\mathcal{H}}, \rho] + \mathcal{L}\rho, \\ \mathcal{L}\rho &= -\gamma_\sigma [|1\rangle \langle 1| \rho + |2\rangle \langle 2| \rho + \rho |1\rangle \langle 1| \\ &\quad + \rho |2\rangle \langle 2| - 2|3\rangle \langle 3| \rho_{22} - 2|4\rangle \langle 4| \rho_{11}] \\ &\quad - \gamma [|1\rangle \langle 1| \rho + |2\rangle \langle 2| \rho + \rho |1\rangle \langle 1| \\ &\quad + \rho |2\rangle \langle 2| - 2|3\rangle \langle 3| \rho_{11} - 2|4\rangle \langle 4| \rho_{22}] \\ &\quad + \gamma [|4\rangle \langle 3| \rho_{21} + |3\rangle \langle 4| \rho_{12}], \end{aligned} \quad (8)$$

The last two terms in Eq. (8) arise from the vacuum induced interference and it comes as the dipole matrix

elements \vec{d}_{13} and \vec{d}_{24} are anti-parallel. In a frame rotating with the frequency of the coherent drive the density matrix equations are,

$$\begin{aligned}\dot{\tilde{\rho}}_{11} &= i\Omega_c^* \tilde{\rho}_{13} - i\Omega_c \tilde{\rho}_{31} - 2\Gamma \tilde{\rho}_{11}, \\ \dot{\tilde{\rho}}_{22} &= i\Omega_c \tilde{\rho}_{42} - i\Omega_c^* \tilde{\rho}_{24} - 2\Gamma \tilde{\rho}_{22}, \\ \dot{\tilde{\rho}}_{33} &= i\Omega_c \tilde{\rho}_{31} - i\Omega_c^* \tilde{\rho}_{13} + \gamma_\sigma \tilde{\rho}_{22} + \gamma \tilde{\rho}_{11}, \\ \dot{\tilde{\rho}}_{12} &= -i\Omega_c \tilde{\rho}_{32} - i\Omega_c^* \tilde{\rho}_{14} - 2\Gamma \tilde{\rho}_{12}, \\ \dot{\tilde{\rho}}_{13} &= -i\Delta \tilde{\rho}_{13} + i\Omega_c (\tilde{\rho}_{11} - \tilde{\rho}_{33}) - \Gamma \tilde{\rho}_{13}, \\ \dot{\tilde{\rho}}_{14} &= -i\Delta \tilde{\rho}_{14} - i\Omega_c \tilde{\rho}_{12} - i\Omega_c \tilde{\rho}_{34} - \Gamma \tilde{\rho}_{14}, \\ \dot{\tilde{\rho}}_{23} &= -i\Delta \tilde{\rho}_{23} + i\Omega_c \tilde{\rho}_{21} + i\Omega_c \tilde{\rho}_{43} - \Gamma \tilde{\rho}_{23}, \\ \dot{\tilde{\rho}}_{24} &= -i\Delta \tilde{\rho}_{24} - i\Omega_c (\tilde{\rho}_{22} - \tilde{\rho}_{44}) - \Gamma \tilde{\rho}_{24}, \\ \dot{\tilde{\rho}}_{34} &= -i\Omega_c \tilde{\rho}_{32} - i\Omega_c^* \tilde{\rho}_{14} - \gamma \tilde{\rho}_{12}, \end{aligned} \quad (9)$$

where

$$\begin{aligned}\tilde{\rho}_{ii} &= \rho_{ii} \quad ; \quad \tilde{\rho}_{ij} = \rho_{ij} e^{-i\omega t} \\ \Gamma &= (\gamma_\sigma + \gamma); \quad \Delta = \omega - \omega_{13} = \omega - \omega_{24}, \end{aligned} \quad (10)$$

The remaining equations can be generated by taking complex conjugates and using $\text{Tr}\{\rho\} = 1$. The steady state solution of Eq. (10) are found to be

$$\tilde{\rho}_{12} = \tilde{\rho}_{14} = \tilde{\rho}_{32} = \tilde{\rho}_{34} = 0, \quad (11)$$

$$\begin{aligned}\tilde{\rho}_{11} = \tilde{\rho}_{22} &= \frac{1}{2} \frac{|\Omega_c|^2}{[2|\Omega_c|^2 + \Gamma^2 + \Delta^2]}, \\ \tilde{\rho}_{33} = \tilde{\rho}_{44} &= \frac{1}{2} \frac{|\Omega_c|^2 + \Gamma^2 + \Delta^2}{[2|\Omega_c|^2 + \Gamma^2 + \Delta^2]}, \\ \tilde{\rho}_{13} = -\tilde{\rho}_{24} &= -\frac{i\Omega_c}{\Gamma + i\Delta} \left\{ \frac{1}{2} \frac{\Gamma^2 + \Delta^2}{[2|\Omega_c|^2 + \Gamma^2 + \Delta^2]} \right\}, \end{aligned} \quad (12)$$

As can be seen from Eqs. (11) and (12) the vacuum induced interference has no effect on the steady state solutions. Clearly vacuum induced coherence can show up in dynamical quantities.

III. PHOTON-PHOTON CORRELATIONS

Since the objective of this paper is to investigate the observable consequences of the vacuum induced coherence; we focus our attention on the photon statistics of the radiation emitted by our model system. We in particular will calculate photon-photon correlations as currently considerable experimental effort is on such correlations. For this we need to know how to relate the atomic properties with the statistical properties of the spontaneously emitted radiation. The answer to this question already exists in quantum theory. In fact from the existing [15] literature, we know that the positive frequency

part of the electric-field operator at a point \vec{r} in the far-field zone can be written in terms of the atomic operators as

$$\begin{aligned} \mathbf{E}^+(\vec{r}, t) &= \mathbf{E}_0^+(\vec{r}, t) - k_0^2 \sum_i \{ [\hat{R}_i \times (\hat{R}_i \times \vec{d}_{31}) A_{31}] \\ &+ [\hat{R}_i \times (\hat{R}_i \times \vec{d}_{42}) A_{42}] \\ &+ [\hat{R}_i \times (\hat{R}_i \times \vec{d}_{32}) A_{32}] \\ &+ [\hat{R}_i \times (\hat{R}_i \times \vec{d}_{41}) A_{41}] \} R_i^{-1} \\ &\times e^{-i(k_0 \vec{r} \cdot \vec{r}_i + \omega \tau)}, \end{aligned} \quad (13)$$

where $\vec{R}_i = \vec{r} - \vec{r}_i$, \vec{r} being the distance of the point of observation from the origin and \vec{r}_i being the position of the atom from the origin. Further $\tau = t - \frac{r}{c}$ is the retarded time, $k_0 = \frac{\omega_0}{c}$, $\omega_0 = \omega_{13} = \omega_{24}$, \vec{d}_{ij} is the electric dipole moment operator and the atomic operators are as defined in Eq. (1). The first term on the right of Eq. (13) is the free field term and the second term is the retarded dipole field emitted by the atom. The emitted radiation consist of different polarization components– the π and the σ polarized components. The terms A_{31} and A_{42} correspond to π polarization whereas the ones A_{32} and A_{41} correspond to σ polarization. We next calculate the photon-photon correlations and the normalized second order correlations for the emitted radiations from the π transitions of this driven four-level atom. For π polarization the relevant part of the electric field operator is given by,

$$\begin{aligned} \mathbf{E}^+(\vec{r}, t) &= \mathbf{E}_0^+(\vec{r}, t) - \left(\frac{\omega_0}{c}\right)^2 \frac{1}{r} ([\hat{n} \times (\hat{n} \times \vec{d}_{31})] |3\rangle\langle 1|_\tau \\ &+ [\hat{n} \times (\hat{n} \times \vec{d}_{42})] |4\rangle\langle 2|_\tau), \end{aligned} \quad (14)$$

In the lowest order correlation the free field term of Eq. (14) does not contribute. This can be seen directly from the definition of quantized fields [15], the fact that the field is initially in the vacuum state and the expression for the normally ordered correlation function for the field, $\langle \mathbf{E}^-(\vec{r}, t) \mathbf{E}^+(\vec{r}', t') \rangle$. Hence with no contribution from the free field term the intensity I_π of the light emitted on the π transition from the atom is ,

$$\begin{aligned} \langle I_\pi \rangle &= \langle \mathbf{E}_\pi^-(\vec{r}, t) \cdot \mathbf{E}_\pi^+(\vec{r}, t) \rangle \\ &= \left(\frac{\omega_0}{c}\right)^4 \frac{1}{r^2} \langle [\hat{n} \times (\hat{n} \times \vec{d}_{31})]^* \cdot [\hat{n} \times (\hat{n} \times \vec{d}_{31})] |1\rangle\langle 1|_\tau \\ &+ [\hat{n} \times (\hat{n} \times \vec{d}_{42})]^* \cdot [\hat{n} \times (\hat{n} \times \vec{d}_{42})] |2\rangle\langle 2|_\tau \rangle, \end{aligned} \quad (15)$$

where we have taken our origin at the location of the atom , $\vec{r} = \hat{n}r$, τ is the retarded time and we used the property $A_{ij}A_{kl} = A_{il}\delta_{kj}$. The negative frequency part of the electric field operator $\mathbf{E}^-(\vec{r}, t)$ can be found by taking the complex conjugate of the positive frequency part. Now if we assume that the point of observation lies perpendicular to both the polarization and propagation direction we have from Eq. (15)

$$\langle I_\pi \rangle = \left(\frac{\omega_0}{c}\right)^4 \frac{1}{r^2} (|\vec{d}_{31}|^2 \langle |1\rangle\langle 1| \rangle_\tau + |\vec{d}_{42}|^2 \langle |2\rangle\langle 2| \rangle_\tau), \quad (16)$$

Eq.(16) can be further simplified using Eqs. (2) and (12), where in using Eq. (12) we have assumed that observation is been made at long time limit. The final expression for I_π in the long time limit (steady state) is then,

$$\langle I_\pi \rangle^{st} = \left(\frac{\omega_0}{c}\right)^4 \frac{|\mathcal{D}|^2}{6r^2} \frac{|\Omega_c|^2}{[2|\Omega_c|^2 + \Gamma^2 + \Delta^2]}, \quad (17)$$

Eq.(17) clearly show that intensity emitted on the π transitions is not altered by vacuum induced coherences and is simply proportional to the steady state population of the excited states.

Let us now investigate what happens incase of two time photon-photon correlations on the π transitions. The two-time photon-photon correlation for the level scheme in Fig.1 can be written as

$$\begin{aligned} \langle I_\pi(t + \tau) I_\pi(t) \rangle &= \langle \mathbf{E}_\pi^-(\vec{r}, t) \mathbf{E}_\pi^-(\vec{r}, t + \tau) : \\ &\quad \mathbf{E}_\pi^+(\vec{r}, t + \tau) \mathbf{E}_\pi^+(\vec{r}, t) \rangle \\ &= \left(\frac{\omega_0}{c}\right)^8 \frac{1}{r^4} \{ [\hat{n} \times (\hat{n} \times \vec{d}_{31})]^* \\ &\quad \cdot [\hat{n} \times (\hat{n} \times \vec{d}_{31})] \}^2 \\ &\quad \langle (|1\rangle\langle 3| - |2\rangle\langle 4|)_t (|1\rangle\langle 1| + |2\rangle\langle 2|)_{t+\tau} \\ &\quad (|3\rangle\langle 1| - |4\rangle\langle 2|)_t \rangle, \end{aligned} \quad (18)$$

The two-time correlation function which appears in Eq. (18) is to be obtained from the solution of the time-dependent density matrix equations (Eq.(9)) and the quantum regression theorem [16]. A closer look at Eq. (9) show that eight of the fifteen equations form a closed set of linear equations which can be solved to find $|1\rangle\langle 1|_{t+\tau}$, $|2\rangle\langle 2|_{t+\tau}$ and hence the term $(|1\rangle\langle 1| + |2\rangle\langle 2|)_{t+\tau}$ in Eq. (18). Before going further let us list those eight equations,

$$\begin{aligned} \dot{\tilde{\rho}}_{11} &= i\Omega_c^* \tilde{\rho}_{13} - i\Omega_c \tilde{\rho}_{31} - 2\Gamma \tilde{\rho}_{11}, \\ \dot{\tilde{\rho}}_{33} &= i\Omega_c \tilde{\rho}_{31} - i\Omega_c^* \tilde{\rho}_{13} + \gamma_\sigma \tilde{\rho}_{22} + \gamma \tilde{\rho}_{11}, \\ \dot{\tilde{\rho}}_{13} &= -i\Delta \tilde{\rho}_{13} + i\Omega_c (\tilde{\rho}_{11} - \tilde{\rho}_{33}) - \Gamma \tilde{\rho}_{13}, \\ \dot{\tilde{\rho}}_{31} &= i\Delta \tilde{\rho}_{31} - i\Omega_c^* (\tilde{\rho}_{11} - \tilde{\rho}_{33}) - \Gamma \tilde{\rho}_{31}, \\ \dot{\tilde{\rho}}_{22} &= i\Omega_c \tilde{\rho}_{42} - i\Omega_c^* \tilde{\rho}_{24} - 2\Gamma \tilde{\rho}_{22}, \\ \dot{\tilde{\rho}}_{44} &= i\Omega_c^* \tilde{\rho}_{24} - i\Omega_c \tilde{\rho}_{42} + \gamma_\sigma \tilde{\rho}_{11} + \gamma \tilde{\rho}_{22}, \\ \dot{\tilde{\rho}}_{24} &= -i\Delta \tilde{\rho}_{24} - i\Omega_c (\tilde{\rho}_{22} - \tilde{\rho}_{44}) - \Gamma \tilde{\rho}_{24}, \\ \dot{\tilde{\rho}}_{42} &= i\Delta \tilde{\rho}_{42} + i\Omega_c^* (\tilde{\rho}_{22} - \tilde{\rho}_{44}) - \Gamma \tilde{\rho}_{42}, \end{aligned} \quad (19)$$

In compact notation this equations can be written as,

$$\dot{\tilde{\rho}} = \mathcal{M} \tilde{\rho}, \quad (20)$$

where $\dot{\tilde{\rho}}$, $\tilde{\rho}$ are (8×1) column matrix and \mathcal{M} is a (8×8) square matrix. Now using the method depicted in [17] and using Eq.(19) the solution of $\langle |1\rangle\langle 1|_{t+\tau} \rangle$ and $\langle |2\rangle\langle 2|_{t+\tau} \rangle$ can be expressed in the form

$$\begin{aligned} \langle |1\rangle\langle 1|_{t+\tau} \rangle &= f_{11}(\tau) \langle |1\rangle\langle 1|_t \rangle + f_{12}(\tau) \langle |3\rangle\langle 3|_t \rangle \\ &+ f_{13}(\tau) \langle |3\rangle\langle 1|_t \rangle + f_{14}(\tau) \langle |1\rangle\langle 3|_t \rangle \\ &+ f_{15}(\tau) \langle |2\rangle\langle 2|_t \rangle + f_{16}(\tau) \langle |4\rangle\langle 4|_t \rangle \\ &+ f_{17}(\tau) \langle |4\rangle\langle 2|_t \rangle + f_{18}(\tau) \langle |2\rangle\langle 4|_t \rangle, \end{aligned} \quad (21)$$

$$\begin{aligned}
\langle |2\rangle\langle 2|_{t+\tau} \rangle &= f_{51}(\tau)\langle |1\rangle\langle 1|_t \rangle + f_{52}(\tau)\langle |3\rangle\langle |3|_t \rangle \\
&+ f_{53}(\tau)\langle |3\rangle\langle |1|_t \rangle + f_{54}(\tau)\langle |1\rangle\langle |3|_t \rangle \\
&+ f_{55}(\tau)\langle |2\rangle\langle |2|_t \rangle + f_{56}(\tau)\langle |4\rangle\langle |4|_t \rangle \\
&+ f_{57}(\tau)\langle |4\rangle\langle |2|_t \rangle + f_{58}(\tau)\langle |2\rangle\langle |4|_t \rangle, \quad (22)
\end{aligned}$$

where the f 's are defined by

$$f_{ik}(\tau) = (e^{\mathcal{M}\tau})_{ik}, \quad (23)$$

and

$$\begin{aligned}
\mathcal{M}_{ik} &= \sum_l P_{il}\Lambda_{ll}P_{lk}^{-1} \\
(e^{\mathcal{M}\tau})_{ik} &= \sum_l P_{il}e^{\Lambda_{ll}\tau}P_{lk}^{-1}, \quad (24)
\end{aligned}$$

Here we have diagonalized the matrix \mathcal{M} with Λ being the eigenvalues and P being the corresponding eigenvectors. We now make use of the quantum regression theorem to obtain the two time correlation function as,

$$\begin{aligned}
\langle B^\dagger(t)\langle |1\rangle\langle 1| + |2\rangle\langle 2| \rangle_{t+\tau} B(t) \rangle &= F_1(\tau)\langle B^\dagger(t)|1\rangle\langle 1|_t B(t) \rangle \\
&+ F_2(\tau)\langle B^\dagger(t)|3\rangle\langle 3|_t B(t) \rangle \\
&+ F_3(\tau)\langle B^\dagger(t)|3\rangle\langle 1|_t B(t) \rangle \\
&+ F_4(\tau)\langle B^\dagger(t)|1\rangle\langle 3|_t B(t) \rangle \\
&+ F_5(\tau)\langle B^\dagger(t)|2\rangle\langle 2|_t B(t) \rangle \\
&+ F_6(\tau)\langle B^\dagger(t)|4\rangle\langle 4|_t B(t) \rangle \\
&+ F_7(\tau)\langle B^\dagger(t)|4\rangle\langle 2|_t B(t) \rangle \\
&+ F_8(\tau)\langle B^\dagger(t)|2\rangle\langle 4|_t B(t) \rangle, \quad (25)
\end{aligned}$$

where we define the operator B as, $B^\dagger(t) = (|1\rangle\langle 3| - |2\rangle\langle 4|)_t$; $B(t) = (B^\dagger(t))^\dagger$ and $F_i(\tau) = f_{1i}(\tau) + f_{5i}(\tau)$. Using this new definition of the operator in Eq. (18), the expression for the two-time photon-photon correlation becomes,

$$\begin{aligned}
\langle I_\pi(t+\tau)I_\pi(t) \rangle &= \left(\frac{\omega_0}{c}\right)^8 \frac{1}{r^4} \{[\hat{n} \times (\hat{n} \times \vec{d}_{31})]^* \\
&\cdot [\hat{n} \times (\hat{n} \times \vec{d}_{31})]\}^2 \\
&\times \langle B^\dagger(t)\langle |1\rangle\langle 1| + |2\rangle\langle 2| \rangle_{t+\tau} B(t) \rangle, \quad (26)
\end{aligned}$$

which when Eq. (25) is used, simplifies to

$$\begin{aligned}
\langle I_\pi(t+\tau)I_\pi(t) \rangle &= \left(\frac{\omega_0}{c}\right)^8 \frac{1}{r^4} \{[\hat{n} \times (\hat{n} \times \vec{d}_{31})]^* \\
&\cdot [\hat{n} \times (\hat{n} \times \vec{d}_{31})]\}^2 \\
&\times (F_2(\tau)\langle |1\rangle\langle 1|_t \rangle + F_6(\tau)\langle |2\rangle\langle 2|_t \rangle), \quad (27)
\end{aligned}$$

In the long time limit $\langle |1\rangle\langle 1|_t \rangle \equiv \tilde{\rho}_{11}(t)$ and $\langle |2\rangle\langle 2|_t \rangle \equiv \tilde{\rho}_{22}(t)$, where $\tilde{\rho}_{11}(t), \tilde{\rho}_{22}(t)$ are the steady state populations of the excited states given by Eq. (12). Now following our assumption that the point of observation lies perpendicular to both the polarization and propagation directions and substituting for $\tilde{\rho}_{11}, \tilde{\rho}_{22}$ from Eq. (12), Eq. (27) can be further simplified. The final expression for the two-time photon-photon correlation is then,

$$\begin{aligned}
G_\pi^{(2)}(\tau) &= \langle I_\pi(t+\tau)I_\pi(t) \rangle \\
&= \left(\frac{\omega_0}{c}\right)^8 \frac{|\mathcal{D}|^4}{36r^4} (F_2(\tau) + F_6(\tau)) \\
&\times \left(\frac{1}{2} \frac{|\Omega_c|^2}{[2|\Omega_c|^2 + \Gamma^2 + \Delta^2]}\right), \quad (28)
\end{aligned}$$

where we have used Eq. (2) for the dipole matrix elements. Note that $F_2(\tau)[F_6(\tau)]$ is the sum of probabilities of finding the atom in the states $|1\rangle$ and $|2\rangle$ given that at $\tau = 0$, the atom was in the state $|3\rangle[|4\rangle]$. In the limit of large τ ,

$$G_\pi^{(2)}(\tau) \rightarrow \left(\frac{\omega_0}{c}\right)^8 \frac{|\mathcal{D}|^4}{36r^4} \left(\frac{2|\Omega_c|^2}{[2|\Omega_c|^2 + \Gamma^2 + \Delta^2]}\right), \quad (29)$$

Next let us derive the expression for two-time photon-photon correlation in absence of interference. In this case the total photon-photon correlation will be a simple addition of photon-photon correlations for radiation emitted on individual π transitions.

$$\begin{aligned}
G_\pi^{(2)}(\tau) &= \langle I_\pi(t+\tau)I_\pi(t) \rangle \\
&= \langle \mathbf{E}_\pi^-(\vec{r}, t)\mathbf{E}_\pi^-(\vec{r}, t+\tau) : \\
&\quad \mathbf{E}_\pi^+(\vec{r}, t+\tau)\mathbf{E}_\pi^+(\vec{r}, t) \rangle_{|1\rangle\langle 3|} \\
&+ \langle \mathbf{E}_\pi^-(\vec{r}, t)\mathbf{E}_\pi^-(\vec{r}, t+\tau) : \\
&\quad \mathbf{E}_\pi^+(\vec{r}, t+\tau)\mathbf{E}_\pi^+(\vec{r}, t) \rangle_{|2\rangle\langle 4|}, \quad (30)
\end{aligned}$$

$$\begin{aligned}
&= \left(\frac{\omega_0}{c}\right)^8 \frac{1}{r^4} \{[\hat{n} \times (\hat{n} \times \vec{d}_{31})]^* \\
&\cdot [\hat{n} \times (\hat{n} \times \vec{d}_{31})]\}^2 \\
&\langle |1\rangle\langle 3|_t \langle |1\rangle\langle 1| \rangle_{t+\tau} \langle |3\rangle\langle 1|_t \rangle \\
&+ \left(\frac{\omega_0}{c}\right)^8 \frac{1}{r^4} \{[\hat{n} \times (\hat{n} \times \vec{d}_{42})]^* \\
&\cdot [\hat{n} \times (\hat{n} \times \vec{d}_{42})]\}^2 \\
&\langle |2\rangle\langle 4|_t \langle |2\rangle\langle 2| \rangle_{t+\tau} \langle |4\rangle\langle 2|_t \rangle, \quad (31)
\end{aligned}$$

Finally using Eq. (21),(22) and (12) we get the photon-photon correlation in absence of interference as

$$\begin{aligned}
G_\pi^{(2)}(\tau) &= \left(\frac{\omega_0}{c}\right)^8 \frac{|\mathcal{D}|^4}{36r^4} (f_{12}(\tau) + f_{56}(\tau)) \\
&\times \left(\frac{1}{2} \frac{|\Omega_c|^2}{[2|\Omega_c|^2 + \Gamma^2 + \Delta^2]}\right), \quad (32)
\end{aligned}$$

Here $f_{12}(\tau)[f_{56}(\tau)]$ is the probability of finding the atom in the states $|1\rangle[|2\rangle]$ given that at $\tau = 0$, the atom was in the state $|3\rangle[|4\rangle]$. Eq. (32) in the limit of large τ becomes,

$$G_\pi^{(2)}(\tau) \rightarrow \left(\frac{\omega_0}{c}\right)^8 \frac{|\mathcal{D}|^4}{36r^4} \left(\frac{|\Omega_c|^2}{[2|\Omega_c|^2 + \Gamma^2 + \Delta^2]}\right), \quad (33)$$

We now further calculate the normalized photon-photon correlation corresponding to Eq. (28) and Eq. (32). The $g^{(2)}$ function gives the non-classical aspects of photon statistics.

$$g^{(2)}(t + \tau, t) = \frac{\langle I_\pi(t + \tau)I_\pi(t) \rangle}{\langle I_\pi(t + \tau) \rangle \langle I_\pi(t) \rangle} = \frac{(F_2(\tau) + F_6(\tau))\tilde{\rho}_{11}}{4\tilde{\rho}_{11}^2}, \quad (34)$$

$$\mathbf{g}^{(2)}(t + \tau, t) = \frac{\langle I_\pi(t + \tau)I_\pi(t) \rangle}{(\langle I_\pi(t + \tau) \rangle \langle I_\pi(t) \rangle)_{|1\rangle\langle 3|} + (\langle I_\pi(t + \tau) \rangle \langle I_\pi(t) \rangle)_{|3\rangle\langle 1|}} (f_{11}(\tau) + f_{33}(\tau))\tilde{\rho}_{11}$$

Here $\tilde{\rho}_{11}$ is the steady state population of the excited state given by Eq. (12) and $g^{(2)}$ [$\mathbf{g}^{(2)}$] is the normalized two time photon-photon correlation function corresponding to presence [absence] of vacuum induced interference.

IV. NUMERICAL RESULTS

In this section we present our numerical results and discuss their consequences. To begin with, we first discuss our method of computation. The decay rates of the excited states to the two ground states, $2\gamma_\sigma$ and 2γ are proportional to $|\vec{d}_{41}|^2$ and $|\vec{d}_{31}|^2$ respectively. From Eq. (2) we get, $2\gamma_\sigma \equiv \gamma_0/3$ and $2\gamma \equiv \gamma_0/6$, where γ_0 is proportional to the square of the reduced dipole matrix element. We use these values for the decays in our numerical computation and normalize all the computational parameters with respect to γ_0 . Further we use standard subroutines to diagonalize the complex general matrix \mathcal{M} and obtain complex eigenvalues and eigenvectors of the form $(\alpha + i\beta)$. For all values of detuning and Rabi

TABLE I: Eigenvalues for the diagonalized matrix \mathcal{M} corresponding to two different values of the Rabi frequency of the driving field which is on resonance with the atomic transitions.

λ	$\Omega = 0.5\gamma_0$	$\Omega = 3.0\gamma_0$
1	(-0.349797,-1.10904)	(-0.375000,5.99870)
2	(-0.349797,1.10904)	(-0.375000,-5.99870)
3	(-0.215794,-1.09726)	(-0.208269,5.99522)
4	(-0.215794,1.09726)	(-0.208269,-5.99522)
5	(-0.300406,0.000000)	(-0.250000,0.000000)
6	(-0.165314,0.000000)	(-0.250000,0.000000)
7	(-0.403098,0.000000)	(-0.333462,0.000000)
8	(0.000000,0.000000)	(0.000000,0.000000)

frequency used in our computation we have two pairs of complex conjugate eigenvalues and four other eigenvalues whose complex part are so small compare to the real part that these complex parts have no significant contributions. Hence these four eigenvalues can be taken to be purely real. Note that this is in contrast to the case of

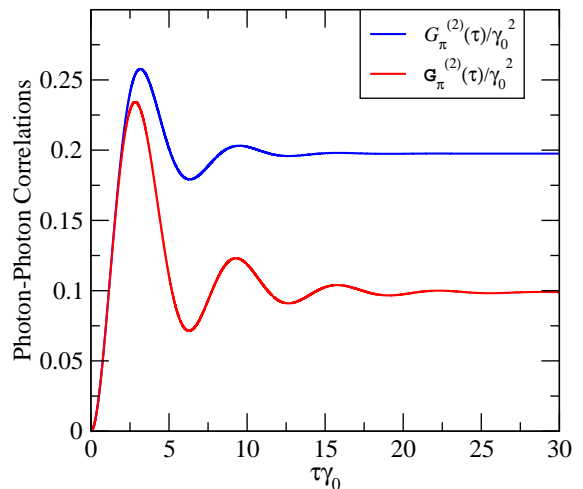


FIG. 2: Plot of two-time Photon-Photon correlation as a function of time for $\Omega = 0.5, \Delta = 0.0$. All the parameters are normalized with respect to γ_0 , where $\gamma_0 = \frac{4|\mathcal{D}|^2\omega^3}{3c^3}$. The blue and red lines correspond to photon-photon correlations in presence and absence of VIC respectively.

photon-photon correlations for the two level model where the number of eigenvalues is four [18]. The changes in the eigenvalues lead to spectral modification as discussed by Kiffner. et. al. [9]. The eigenvalues for $\Omega_c = 0.5\gamma_0$, and $\Omega_c = 3\gamma_0$ and detuning $\Delta = 0$ are listed in the Table (I). Note for example that for $\Omega_c = 3\gamma_0$ we have eigenvalues $\pm 5.99870i - 0.375$ and $\pm 5.99522i - 0.208269$. This difference in the real parts can produce a dip in the side bands in the Mollow spectrum. Next we calculate the elements f_{ij} of the 8×8 matrix $[f]$ using Eq. (23) and Eq. (24). Finally we use the elements f_{ij} corresponding to Eqs. (28),(32) and Eqs. (34),(35) to evaluate the two time photon-photon correlations and normalized photon-photon correlations in presence and absence of vacuum induced interference respectively.

The Figs. (2-4) show photon-photon correlations corresponding to Eqs. (28) and (32). The blue and red line in the figures correspond respectively, to photon-photon correlations in presence and absence of interference. The

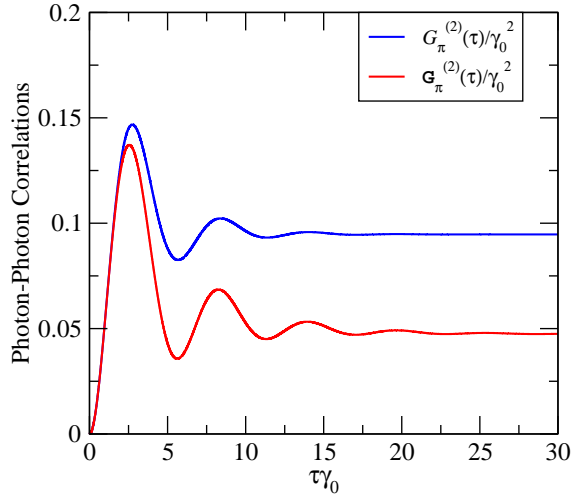


FIG. 4: Plot of two-time Photon-Photon correlation as a function of time for $\Omega = 3.0, \Delta = 0.0$. All the parameters are normalized with respect to γ_0 , where $\gamma_0 = \frac{4|\mathcal{D}|^2\omega^3}{3c^3}$.

correlations calculated in presence of interference show strong damping of the oscillations and attain an overall higher value as the time separation τ between two counts increases. The differences between $G^{(2)}$ and $\mathcal{G}^{(2)}$ are most noticeable in the limit of large time separation τ . In order to understand this we examine the distinction between $F_2(\tau) = f_{12}(\tau) + f_{52}(\tau)$ and $f_{12}(\tau)$. We recall that $f_{12}[f_{52}]$ was the probability of finding the atom in the state $|1\rangle[|2\rangle]$ given that at $\tau = 0$, it was in the state $|3\rangle$. We exhibit these probabilities in the Fig. (5). We observe that the function $f_{52}(\tau)$ starts becoming significant at the time scale of the order of γ_σ^{-1} .

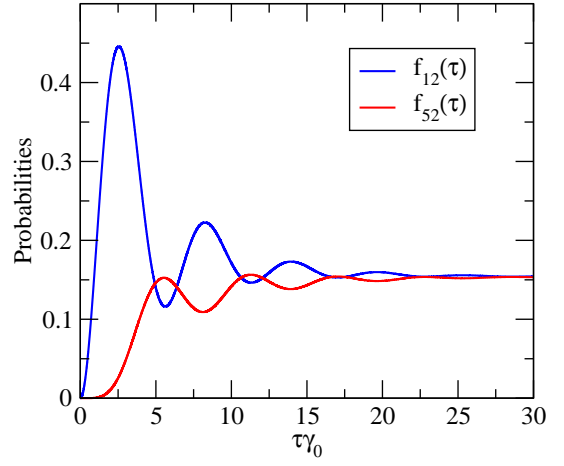


FIG. 5: Probability for finding the atom in state $|1\rangle$ (f_{12}) and $|2\rangle$ (f_{52}) given that at time $\tau = 0$ the atom was in the state $|3\rangle$.

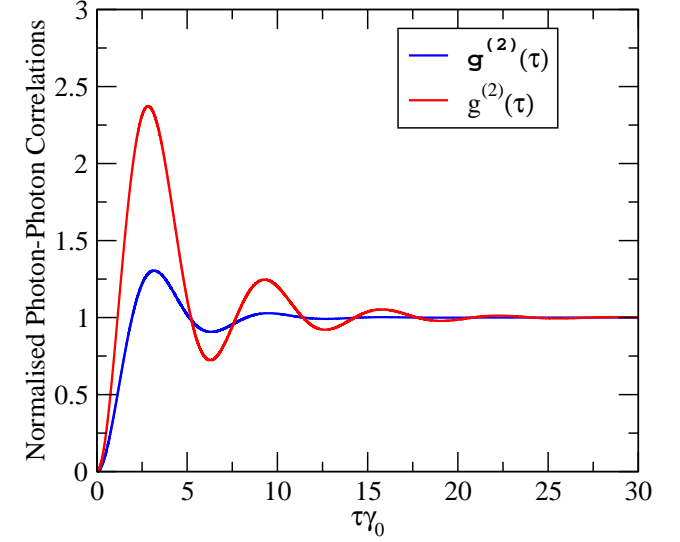
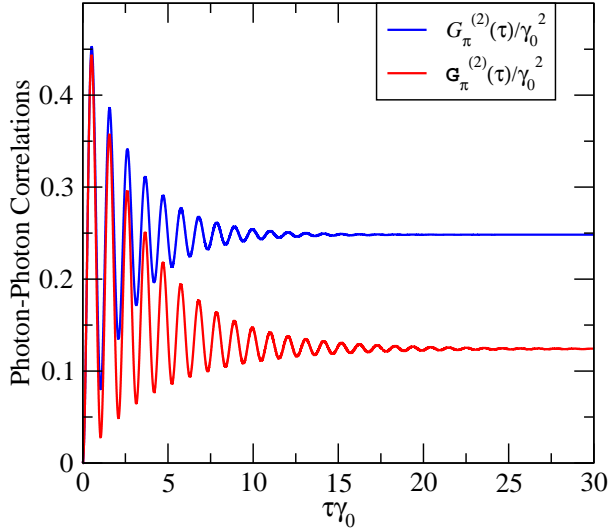


FIG. 6: Normalized photon-photon correlations plotted as a function of time, for $\Omega = 0.5, \Delta = 0.0$. All the parameters are normalized with respect to γ_0 , where $\gamma_0 = \frac{4|\mathcal{D}|^2\omega^3}{3c^3}$.

Further for large τ , f_{12} and f_{52} become comparable. The physical process that contributes to f_{52} is the following,

$$|3\rangle \xrightarrow[\pi\text{-pol}]{\text{laser}} |1\rangle \xrightarrow[\text{emission}]{\sigma\text{-photon}} |4\rangle \xrightarrow[\pi\text{-pol}]{\text{laser}} |2\rangle.$$

Similarly population can start from the state $|4\rangle$ and end up in the state $|1\rangle$ via,

$$|4\rangle \xrightarrow[\pi\text{-pol}]{\text{laser}} |2\rangle \xrightarrow[\text{emission}]{\sigma\text{-photon}} |3\rangle \xrightarrow[\pi\text{-pol}]{\text{laser}} |1\rangle.$$

We show normalized photon-photon correlations in a typical case in the Fig. (6). In case of interference we observe

stronger damping of the oscillations and an overall reduction of the $g^{(2)}$ function at shorter time scales. At long time limits $g^{(2)}(\tau \rightarrow \infty)$ is 1. Photon antibunching effect is also visible as $0 \leq g^{(2)}(0) < 1$. For shorter time scale we get $g^{(2)}(\tau) \not\leq 1$ a clear signature of the nonclassical nature of the two-time photon-photon correlations.

V. CONCLUSIONS

In conclusion we have shown that the vacuum induced coherence(VIC) do significantly affect the two-time photon-photon correlations even though they show no effect on the total steady state intensity of the radiation emitted on the π transitions. The effect of this coherence

is reflected in form of stronger damping and overall larger values of the correlation function $G^{(2)}$. The level scheme $j = 1/2 \rightarrow j = 1/2$ is easily realizable and has already been used, for example in $^{198}\text{Hg}^+$ [10] in the context of interferences produced by a system of two ions and more recently in $^{138}\text{Ba}^+$ [11] in the context of emission in presence of a mirror. In future we hope to investigate how the asymmetry in the level structures introduced by a magnetic field [19] would influence the photon-photon correlations. This might in turn give us more freedom in choosing the level structure and hence more broader choice in selecting atomic transition for experiments. Finally note that it would also be interesting to examine the VIC effects in the context of nonlinear optical experiments using $j=1/2$ to $j=1/2$ transitions.

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