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Intensity correlations and anticorrelations in a three-level cascade system

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Abstract. We study the intensity–intensity correlations of the radiation emitted on probe transition in a three-level cascade electromagnetically-induced transparency (EIT) scheme. Further, we show that by applying even a very weak incoherent pump, the nature of the emitted radiation can be switched from classical to non-classical without significantly affecting the probe transparency.

Keywords. Intensity correlations; two-photon coherence; non-classical light; three-level cascade system; incoherent pumping.

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1. Introduction

Study of intensity correlations of emitted radiation from multilevel systems driven by external fields, in different environments has been of tremendous interest in recent times. Several studies were done on two-photon correlations [1] in driven three-level systems in various (Λ, V and cascade) configurations. For instance, Huang *et al* [2] have studied the intensity autocorrelations in fluorescence from the upper and lower transitions of a strongly driven three-level cascade system. They found additional resonances in the upper transition which was attributed to the influence of spontaneous decay between the lower transition levels.

Ficek *et al* [3,4] investigated the influence of vacuum-induced coherences on two-photon correlations in a three-level V-type system, with perpendicular dipole moments. In their study, a DC field was applied to create an additional two-photon coherence between the two upper levels. Two-photon correlations have also been employed by Das and Agarwal [5] as a tool to probe vacuum-induced coherences. They have demonstrated interference phenomena in the second-order correlations of the fluorescence light from ion pairs. From their work, it is inferred that higher-order photon correlations could be present even in situations where first-order interferences do not appear [6]. In another study, the effect of vacuum-induced coherence on photon correlation [7] in an equispaced three-level cascade system has been reported. All these studies reveal that the two-photon correlation spectroscopy is a very effective tool in determining the nature of radiation fields emitted from the atomic system under different excitation mechanisms.

More recently, several experimental and theoretical studies [8–12] dealing with the pump–probe correlations from atomic media interacting with external fields under electromagnetically-induced transparency (EIT) [13] have also been reported. In particular, Felinto *et al* [8] have discussed the physical interpretation of the measured photon statistics of probe and pump beams interacting with a Λ-type atomic medium in EIT situation. Florez *et al* [9] reported a detailed investigation on the properties of intensity correlation spectra for laser beams interacting with Λ-type cold caesium and rubidium atoms under EIT. Their study reveals that the intensity–intensity correlation is independent of power broadening of the pump–laser field and therefore it is possible to deduce the ground-state coherence time from the measured intensity correlation spectra. Ariunbold and coworkers [10] have studied intensity correlation and anticorrelations in coherently-driven Rb-atomic vapour (Λ-configuration) and demonstrated that intensity–intensity cross-correlation between two circular polarized beams can be controlled by applying an external magnetic field. Almost all of these studies have been conducted in atomic systems modelled as Λ systems. It would also be interesting from the experimental point of view to investigate the nature of emitted radiation in the cascade-type EIT scheme with extension to the LWI configuration, the latter of which involves application of an incoherent pump. The present work deals with such a study.

For this purpose, we have considered an atomic system in a three-level cascade configuration, which can be related to a typical EIT scheme in any of the alkali atoms [13]. The lower transition of this cascade system is driven by a weak probe field while the upper transition is driven by a strong pump field. As our study does not include vacuum-induced coherence effects, we do not impose any restrictions on the orientation of the dipole moments as well as the energy level separations of the upper and lower transitions. On the other hand, our model incorporates the effect of incoherent pumping from the ground level to both the excited levels, in addition to the spontaneous emission decays.

The radiation field emitted, in the far-field zone, in both the pump and probe transitions, can be expressed in terms of atomic operators. Using the quantum regression theorem, the multitime correlation functions can be written in terms of the expectation values of the atomic operators in steady state multiplied by the function of the delayed time, which comes from the assumption of statistical stationarity of the fields.

It is usually believed that incoherent pumping would tend to decohere the system. Contrary to this intuitive understanding, it was demonstrated [14] that an appropriate rate of incoherent pumping is essential to produce an optimal steady-state value of spontaneously generated entanglement which is a signature of nonclassical behaviour. Furthermore, in a different study [15], it was found that only in the presence of incoherent pumping alone, the phase control of the dispersion, absorption and group index can be realized. Our studies also demonstrate that application of a very weak incoherent pump can significantly alter the characteristics of the emitted radiation. For suitable values of parameters, the two-photon correlation function in the probe transition oscillates between non-classical and classical behaviour, as evident from its second-order correlation function.

The organization of this paper is as follows: In §2 the mathematical formulation for obtaining the equations of motion for the atomic density matrix elements is given. This is followed by a brief description of the intensity–intensity correlation functions of the fluorescent fields in terms of the correlation functions of the atomic operators. In §3, we present a detailed analysis of the behaviour of the above system in terms of the absorption spectra and the corresponding intensity–intensity correlation function (using the convention of refs [8–12]) for the probe transition.

2. Formulation

We model the atom as a three-level system in cascade configuration. The atomic level scheme is shown in figure 1. The top level $|3\rangle$ (energy E_3) decays spontaneously with a rate $2\gamma_{32}$ to the intermediate level $|2\rangle$ (energy E_2) which decays spontaneously with a rate $2\gamma_{21}$ to the ground level $|1\rangle$ (energy E_1). The pump and the probe laser fields drive the transitions $|2\rangle$–$|3\rangle$ and $|1\rangle$–$|2\rangle$ with Rabi frequencies Ω_c and Ω_p respectively. In addition, the upper level of the probe transition $|2\rangle$ may be incoherently pumped from the ground level $|1\rangle$, at the rate Λ_{12}.

The Hamiltonian for this system in the electric dipole approximation is

$$H = H_0 - (|3\rangle\langle 2|\vec{\mu}_{32} + |2\rangle\langle 3|\vec{\mu}_{23}) \cdot (\vec{E}_c e^{-i\omega_c t} + \text{c.c.}) - (|2\rangle\langle 1|\vec{\mu}_{21} + |1\rangle\langle 2|\vec{\mu}_{12}) \cdot (\vec{E}_p e^{-i\omega_p t} + \text{c.c.}). \quad (1)$$

The equation of motion for the density operator ρ of the atomic system is

$$i\hbar \frac{\partial \rho}{\partial t} = -[H, \rho] + \mathcal{L}\rho, \quad (2)$$

where the second term accounts for the spontaneous emission from the upper levels. The equations of motion of the density matrix elements of the system after making rotating wave approximation are obtained as

$$\frac{d\rho_{11}}{dt} = -2\Lambda_{12}\rho_{11} + 2(\gamma_{21} + \Lambda_{12})\rho_{22} + i\Omega_p(\rho_{21} - \rho_{12}),$$

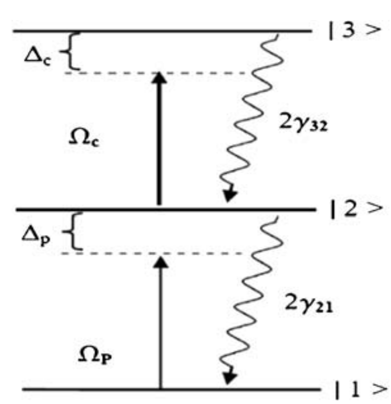

Figure 1. Three-level cascade system–energy levels and interaction scheme.

$$\frac{d\rho_{22}}{dt} = 2\Lambda_{12}\rho_{11} + 2\gamma_{32}\rho_{33} - 2(\gamma_{21} + \Lambda_{12})\rho_{22} - i\Omega_p(\rho_{21} - \rho_{12}) - i\Omega_c(\rho_{23} - \rho_{32}),$$

$$\frac{d\rho_{33}}{dt} = -2\gamma_{32}\rho_{33} + i\Omega_c(\rho_{23} - \rho_{32}),$$

$$\frac{d\rho_{12}}{dt} = -(\gamma_{21} + 2\Lambda_{12} + i\Delta_p)\rho_{12} + i\Omega_p(\rho_{22} - \rho_{11}) - i\Omega_c\rho_{13},$$

$$\frac{d\rho_{23}}{dt} = -(\gamma_{21} + \Lambda_{12} + \gamma_{32} + i\Delta_c)\rho_{23} + i\Omega_c(\rho_{32} - \rho_{22}) + i\Omega_p\rho_{13},$$

$$\frac{d\rho_{13}}{dt} = -[\gamma_{32} + \Lambda_{12} + i(\Delta_p + \Delta_c)]\rho_{13} + i\Omega_p\rho_{23} - i\Omega_c\rho_{12}, \quad (3)$$

together with $\rho_{ij} = \rho^*_{ji}$. In these equations, $\Delta_p = \omega_{21} - \omega_p$ and $\Delta_c = \omega_{32} - \omega_c$ are the frequency detunings of the probe and the pump laser fields respectively. The Rabi frequencies for the probe and the pump transitions respectively are

$$\Omega_p = \frac{\vec{\mu}_{21} \cdot \vec{E}_p}{\hbar} \quad \text{and} \quad \Omega_c = \frac{\vec{\mu}_{32} \cdot \vec{E}_c}{\hbar}$$

(which are assumed to be real with no loss of generality).

We now derive an expression for the second-order intensity–intensity correlation function in terms of the atomic variables. As is well known, the quantum statistical properties of the spontaneously emitted radiation can be expressed in terms of the atomic properties. More specifically, the positive frequency part of the electric field operator in the far-field zone can be expressed in terms of the atomic operators, $(|\beta\rangle\langle\alpha|)$, as

$$\vec{E}^{(+)}(\vec{r}, t) = \vec{E}_0^{(+)}(\vec{r}, t) + \left(\frac{\omega_0}{c}\right)^2 \frac{e^{ikr}}{r} \sum_{\alpha,\beta} e^{-ik\vec{n}\cdot\vec{R}}\, \vec{n} \times (\vec{n} \times \vec{d}_{\beta\alpha})\, (|\beta\rangle\langle\alpha|)\,, \quad (4)$$

where $\vec{r} = \vec{n}r$ and $k = \omega_0/c$. In the above equation the summation is carried over all the transitions corresponding to the possible spontaneous emission decay channels.

A general expression for the intensity–intensity correlation function, for the cascade configuration, is given as

$$\begin{aligned}\langle: I_i(t) I_j(t+\tau) :\rangle &= \langle \vec{E}_i^{(-)}(\vec{r}, t)\vec{E}_i^{(-)}(\vec{r}, t+\tau) \times \vec{E}_j^{(+)}(\vec{r}, t+\tau)\vec{E}_j^{(+)}(\vec{r}, t)\rangle \\ &= \langle(|i+1\rangle\langle i|)_t(|j+1\langle j|)_{t+\tau} \times (|j\rangle\langle j+1|)_{t+\tau}(|i\rangle\langle i+1|)_t\rangle \\ &= \langle(|i+1\rangle\langle i|)_t(|j+1\rangle\langle j+1|)_{t+\tau} \times (|i\rangle\langle i+1|)_t\rangle. \end{aligned} \quad (5)$$

In this equation, summation runs over $i, j = 1, 2$ and τ is the time delay between the signals at the two photodetectors whose outputs are correlated to give rise to the above intensity–intensity correlation. The two-time correlation function of any two arbitrary atomic operators, which are of the general form $|\beta\rangle\langle\alpha|$, denoted here by A_m and A_n for the sake of brevity, can be expressed in terms of single time expectation values, by using quantum regression theorem [16], as

$$\langle A_m(t+\tau)A_n(t)\rangle = \sum_k h_{mk}(\tau)\, \langle A_k(t)A_n(t)\rangle, \quad (6)$$

where the c-number coefficients h_{mk} are derived from the solution of the equations of motion for all the atomic operators, $A_m(t) = \sum_k h_{mk}(t)A_k(0)$.

Therefore, the normalized two-time intensity correlation function expressed in terms of the atomic populations in steady state and the evolution operator of the density matrix [2,4] reduces to the form

$$G_{ij}(\tau) = \frac{\langle: I_i(t)I_j(t+\tau) :\rangle}{\langle|i\rangle\langle i|\rangle\langle|j\rangle\langle j|\rangle} = \frac{P_{i+1\to j}(\tau)}{P_j}. \quad (7)$$

Here P_j is the steady-state population in $|j\rangle$ and $P_{i+1\to j}(\tau)$ is the probability of population transfer, within the time interval τ, from the final state of the first emission $|i+1\rangle$, to the initial state of the second emission $|j\rangle$. These quantities can be evaluated [2,4] using eqs (3) for the density matrix elements.

Our interest here is to study the intensity–intensity correlation of the emitted radiation field in the probe transition, $G_{22}(\tau)$, as a function of different parameters. In the next section, we present numerical results for certain representative set of parameters, for the absorption profile as well as the corresponding intensity–intensity correlation function of the radiation field emitted in the probe transition.

3. Results and discussion

In this section, we present and discuss the results for probe field emission characteristics, namely the intensity–intensity correlations under electromagnetically-induced transparency (EIT) [13] and possibly lasing without inversion (LWI) [17], through incoherent pumping of the upper level ($|2\rangle$) of the probe transition.

For this purpose, the intensity correlation function $G_{22}(\tau)$ corresponding to the emission in the probe transition $|2\rangle \to |1\rangle$ along with its absorption profile are evaluated numerically as a function of various field and atomic parameters, viz. the pump field Rabi strength (Ω_c), the rate of incoherent pumping (Λ_{12}) into the upper level of the probe transition and the spontaneous decays in the probe (γ_{21}) and pump (γ_{32}) transitions.

The parameters chosen here correspond to those used in the experimental studies of EIT in three-level cascade system, for example the $3S_{1/2} \to 3P_{1/2} \to 4D_{3/2}$ transition in a sodium atom. For this transition, the level separation wavelengths are $\lambda_{21} = 589.6$ nm and $\lambda_{32} = 568.3$ nm. The corresponding spontaneous emission decay rates are $2\gamma_{21} = 2\pi$ (10 MHz) and $2\gamma_{32} = 2\pi$ (1.6 MHz) [18].

In the EIT regime, the control field Rabi frequency Ω_c is strong compared to the probe Rabi frequency Ω_p. Hence, we have fixed the probe field Rabi frequency at $\Omega_p = 0.01\gamma_{21}$ and used two typical control Rabi field strengths viz., $\Omega_c = \gamma_{21}/2$ and γ_{21} respectively in figures 2a and 2b to demonstrate the EIT characteristics of the probe field as a function of the probe detuning Δ_p. In addition, we also explore in these figures, the effect of incoherent pumping into the upper level of the probe transition ($|2\rangle$) through the ground level ($|1\rangle$), denoted by the parameter Λ_{12}.

It is seen from figures 2a and 2b that for smaller incoherent pump rates $\Lambda_{12} \ll \gamma_{21}$, there is no appreciable change in the EIT dip at the line centre and it is nearly identical to the case when there is no incoherent pumping. However, the dip vanishes as the incoherent pumping rate increases but the overall probe absorption reduces further over a wider frequency range. In order to understand the absorption reduction with increasing incoherent pumping rate Λ_{12}, we have also examined the steady-state population difference ($\rho_{22} - \rho_{11}$) between the upper and lower levels of the probe transition (figure not shown here). It is found that, for values of $\Lambda_{12} \geq 0.5\gamma_{21}$, the vanishing of EIT dip and absorption reduction occurs due to the saturation of the populations of the levels involved in the probe transition. We find no evidence of LWI in the parameter regime studied here.

The intensity correlation function $G_{22}(\tau)$ of the probe transition, corresponding to the same parameters as in figures 2a and 2b is illustrated in figures 3a and 3b respectively. These figures reveal that incoherent pumping Λ_{12} has a significant influence on the emission characteristics of the probe field. In the absence of incoherent pumping ($\Lambda_{12} = 0$), we find that within a correlation time $\tau \sim 1/\gamma_{21}$, the correlation function rapidly enters the classical domain wherein $G_{22}(\tau) > 0$ (exhibiting correlations) and remains classical for all later times. On the other hand, even a very weak incoherent pumping causes the correlation function to exhibit anticorrelations [8,17] which is a signature of non-classical behaviour. This can clearly be seen from the curves for non-zero values of Λ_{12} ranging from $0.001\gamma_{21}$ to $0.1\gamma_{21}$. Note also that, for the same values of Λ_{12}, there is hardly any variation in the EIT profile as can be seen from figures 2a and 2b. At higher values of $\Lambda_{12} \geq \Omega_c$, the correlation function re-enters the classical regime, wherein it exhibits correlations again.

Comparison between the probe intensity correlation function, for two different values of the control field strength, plotted in figures 3a and 3b shows that for the larger of the two values of the control field, namely $\Omega_c = \gamma_{21}$, the functions exhibit stronger correlations before settling into the steady state of uncorrelated random light. In both cases, it is observed that the re-entry

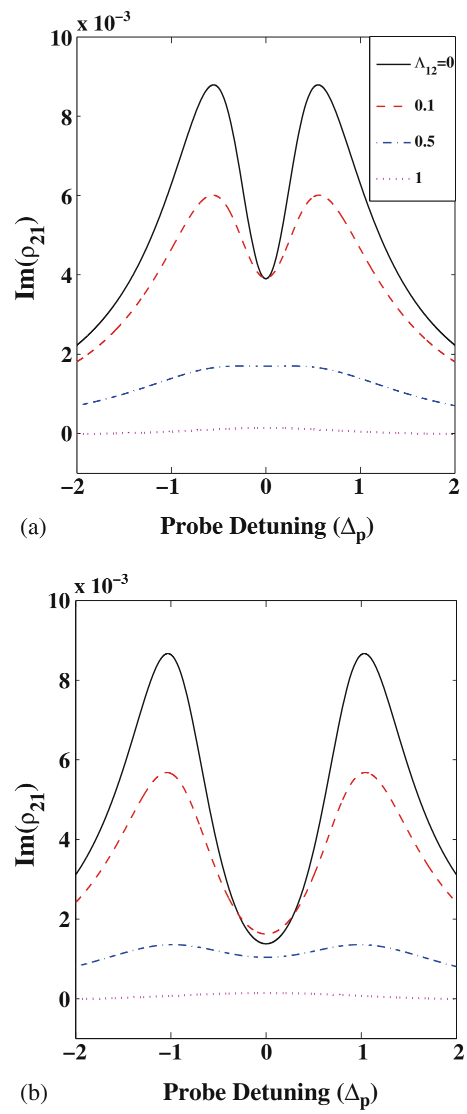

Figure 2. Imaginary part of ρ_{21}. (**a**) $\Omega_c = 0.5$ and (**b**) $\Omega_c = 1$ for unequal spontaneous emission rates $\gamma_{32} = 0.16$. The values of incoherent pumping Λ_{12} for each curve are shown in the legend. The probe field Rabi frequency $\Omega_p = 0.01$. All parameters (Δ_p, Ω_c, Ω_p, γ_{32} and Λ_{12}) used here and in subsequent figures are in units of γ_{21}.

into the non-classical regime, wherein it exhibits anti-correlations (for a range of values of the time delay τ) occurs whenever $\Lambda_{12} \geq \Omega_c$. The correlation function also is seen to exhibit oscillations for this particular value of the control field. This oscillatory behaviour can be understood as resulting from the Rabi oscillations of the populations of the levels participating in the transition. These Rabi oscillations essentially populate and depopulate the upper level of the probe transition periodically giving rise to oscillatory behaviour in the correlation function.

To further clarify the influence of spontaneous decay rates in the system on the intensity–intensity correlation characteristics, we have also considered equal decay rates ($\gamma_{21} = \gamma_{32}$), as it may be relevant for many atomic systems.

A comparison of the EIT feature in both cases reveals the following. For unequal spontaneous decay rates, we have already seen from figures 2a and 2b that the EIT feature is more pronounced with absorption reduction of around 90% at the line centre. On the other hand, we find that for equal spontaneous decays (see figures 4a and 4b), the absorption reduction at the line centre is very small when $\Lambda_{12} = 0$. Furthermore, inclusion of moderate incoherent pumping, for this case, does not give rise to significant change in the EIT feature, whereas at higher values of Λ_{12}, there is reduction

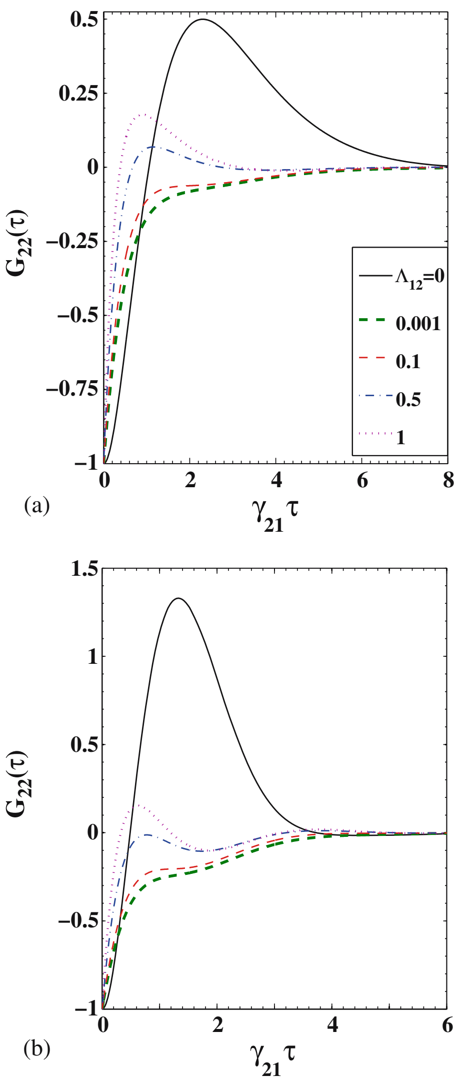

Figure 3. The probe field intensity correlation function $G_{22}(\tau)$. (**a**) $\Omega_c = 0.5$ and (**b**) $\Omega_c = 1$ for unequal spontaneous emission rates $\gamma_{32} = 0.16$ and for the values of Λ_{12} as shown in the legend. Other parameters are the same as in figure 2.

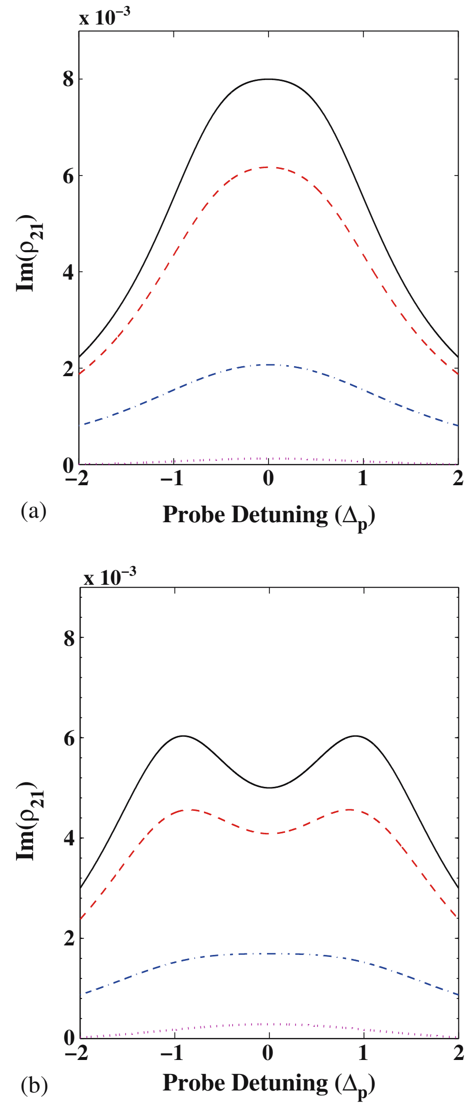

Figure 4. Imaginary part of ρ_{21}. (**a**) $\Omega_c = 0.5$ and (**b**) $\Omega_c = 1$ for equal spontaneous emission rates $\gamma_{32} = \gamma_{21}$. Other parameters are the same as in figure 2.

in overall absorption over a wider range of frequencies due to the saturation of level populations as seen previously for unequal decay rates.

Figures 5a and 5b reveal that the intensity–intensity correlation function remains largely non-classical for the range of pump Rabi field strengths we have studied, for equal spontaneous decay rates. The correlation function is seen to exhibit sub-Poissonian characteristics mostly, except for the largest value of control Rabi field of $\Omega_c = \gamma_{21}$ considered here, in which case there is a small window of cross-over into the classical regime from $\tau = 2.5$ to 4 (where it is slightly greater than 0). For times beyond this window of classicality, the correlation function is seen to approach the steady-state behaviour of uncorrelated random field.

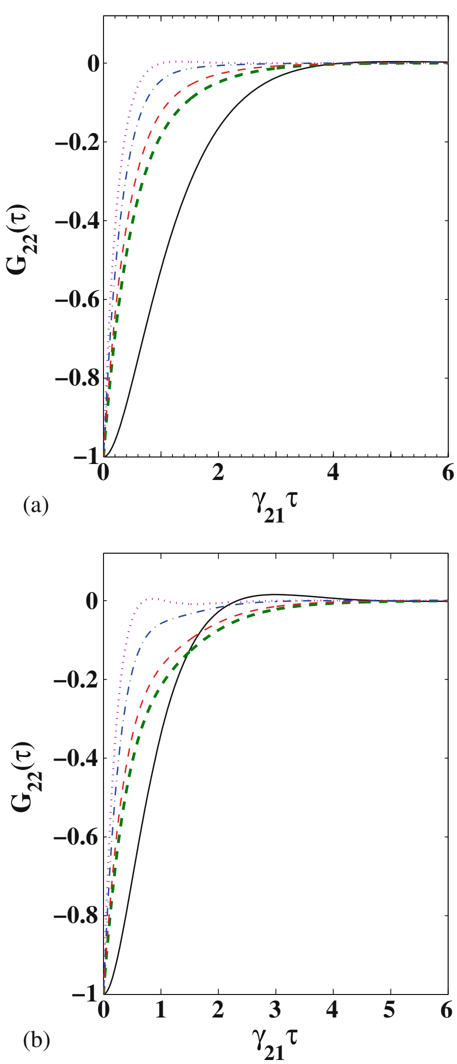

Figure 5. The probe field intensity correlation function $G_{22}(\tau)$. (**a**) $\Omega_c = 0.5$ and (**b**) $\Omega_c = 1$ for equal spontaneous emission decay rates $\gamma_{32} = \gamma_{21}$. Other parameters are the same as in figure 3.

4. Conclusion

In conclusion, we have investigated the role of incoherent pumping on photon statistics of the probe transition in a three-level cascade EIT configuration. Both equal and unequal spontaneous emission decay rates from upper levels, the latter of which corresponds to the actual experimental conditions in alkali systems were considered. Our study demonstrates that although the incoherent pumping into the upper level of the probe transition does not markedly alter the probe absorption (EIT) characteristics, it is seen to modify the second-order correlation function of the probe field significantly. Even a small amount of incoherent pumping causes the emitted radiation to change from classical (in the absence of incoherent pump) to non-classical nature.

The absorption reduction at higher values of incoherent pumping is found to occur due to saturation of the populations of the levels participating in the transition.

The present study clearly demonstrates that a comprehensive understanding of the incoherent pumping can be attained from a study of the intensity–intensity correlation function rather than looking at the absorption profile alone. Such a study may not only lead to a better understanding of the role of different processes but also provide us with a means of controlling the probe emission characteristics by manipulating the system and field parameters.

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