Origin and restoration of missing interference in emission in a laser-driven V system

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We present physical reasoning for the missing interference [Zhu, Narducci, and Scully, Phys. Rev. A **52**, 4791 (1995)] in emission in a laser-driven V system. We demonstrate how the interference effects can be restored by considering an additional channel of spontaneous emission. Analytical results are given to identify the various pathways contributing to this interference. The interference terms show up in the form of dispersive contributions to the line shape. [S1050-2947(98)05607-8]

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In the past few years quantum interference among different transition pathways has become a very important tool in controlling the optical properties of matter. This includes not only the linear but also nonlinear optical properties such as various macroscopic linear and nonlinear susceptibilities [1–3]. Considerable literature exists on the understanding of interferences in the absorption and gain spectra [4,5]. For example, in a recent publication [4(b)] the origin of quantum interferences in probe absorption in various level schemes (Λ,**V**,Ξ) was analyzed. The interferences were found to be constructive or destructive depending on the level scheme. It was also discussed how the nature of interferences could be changed by changing different relaxation parameters. Since in a pumped system one does not necessarily have reciprocity [6] between emission and absorption, it becomes important to analyze the effects of relaxation parameters on interferences in the emission spectrum. Quantum interference has also been shown to be very important in the context of spontaneous emission [7] from, say, two close-lying states. The interference arises as the two pathways are created due to emission into a common vacuum of the electromagnetic field. More generally, if two or more close-lying states interact with a single bath (responsible for dissipative behavior), then the interference more or less always occurs [6].

There are exceptions, however. Zhu *et al.* [8] considered a coherently driven V system with ground level $|3\rangle$ connected to two upper levels $|1\rangle$ and $|2\rangle$ by a dipole transition. They examined the spontaneous emission spectrum on the transition $|1\rangle \rightarrow |3\rangle$ when the transition $|2\rangle \leftrightarrow |3\rangle$ was coherently driven. The coherent drive mixes strongly the levels $|2\rangle$ and $|3\rangle$ leading to new dressed states $|\psi_{\pm}\rangle$. The level separation between $|\psi_{+}\rangle$ and $|\psi_{-}\rangle$ depends on the strength and detuning of the coherent drive. They showed that the spectrum of spontaneous emission consists of two independent Lorentzians, i.e., there is no interference between two possible channels of emission, viz., $|1\rangle \rightarrow |\psi_{+}\rangle$ and $|1\rangle \rightarrow |\psi_{-}\rangle$. In quantum mechanics two transition amplitudes always interfere unless they are out of phase, thus the nonexistence of the quantum interference is surprising despite two apparent paths for emission to the state $|3\rangle$. We would like to understand why there is no interference and how the two paths can be made to interfere [9]. We demonstrate that the interference can arise if we include spontaneous decay of the state $|2\rangle$ to $|3\rangle$. Thus interference can appear from the *opening up of a new pathway due to spontaneous emission on the transition* $|2\rangle \rightarrow |3\rangle$.

We note that the emission spectra for coherently driven multilevel systems have been calculated extensively [10] and these spectra have been analyzed over a very wide range of parameters. However, most of these studies concentrated on the behavior of resonances in the spectra and were not concerned with quantum interferences, which are being discussed now [11]. Thus we analyze the spectra from the point of view of quantum interferences. We analyze the conditions under which interferences occur. We also note that the traditional secular approximation will miss such interferences and a suitable pumping mechanism with the levels of interest can restore the interferences.

The system under consideration is schematically shown in Fig. 1. The coherent driving field $\vec{E}_2 = \vec{\varepsilon}_2 e^{-i\omega_2 t} + \text{c.c.}$ with Rabi frequency $2G_2 = 2\vec{d}_{23} \cdot \vec{\varepsilon}_2 / \hbar$ acts on the transition $|2\rangle \leftrightarrow |3\rangle$. We examine the spontaneous emission on the transition $|1\rangle \leftrightarrow |3\rangle$. Let Δ_2 be the detuning of the coherent drive: $\Delta_2 = \omega_{23} - \omega_2$ with ω_{ij} representing the frequency of the transition $|i\rangle \rightarrow |j\rangle$. The level $|1\rangle$ can be pumped incoherently at the rate $2\Lambda(2\Lambda_0)$ from the state $|3\rangle$ (or externally). Let $2\gamma_2$ $(2\gamma_1)$ be the rates of emission from the level $|2\rangle$ $(|1\rangle)$ in the absence of the coherent drive. We work in the density matrix framework and we will discuss cases when either Λ or $\Lambda_0 \neq 0$. Let ω_1 be the frequency of the photon emitted on the transition $|1\rangle \leftrightarrow |3\rangle$ and let $\Delta_1 = \omega_{13} - \omega_1$. Af-

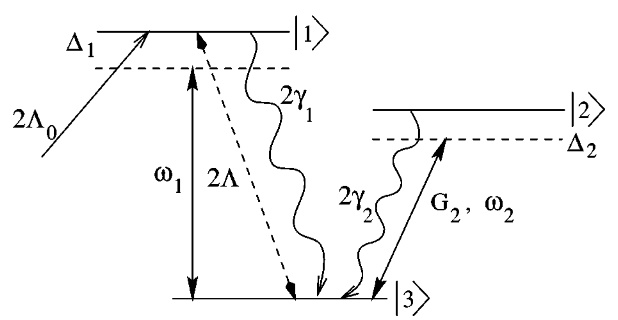

FIG. 1. Schematic diagram of the laser-driven V system under consideration. We would consider the cases either with internal pumping ($\Lambda_0 = 0$) or with external pumping ($\Lambda = 0$).

*Also at Jawahar Lal Nehru Centre for Advanced Scientific Research, Banglore, India. Electronic address: gsa@prl.ernet.in ter a canonical transformation to remove optical frequencies from the equation of motion, the density matrix equations are

$$\begin{aligned}\dot{\rho}_{11}&=-2(\gamma_1+\Lambda)\rho_{11}+2\Lambda\rho_{33}+2\Lambda_0,\\ \dot{\rho}_{12}&=-[\gamma_1+\gamma_2+\Lambda+i(\Delta_1-\Delta_2)]\rho_{12}-iG_2^*\rho_{13},\\ \dot{\rho}_{13}&=-(\gamma_1+2\Lambda+i\Delta_1)\rho_{13}-iG_2\rho_{12},\\ \dot{\rho}_{22}&=-2\gamma_2\rho_{22}+iG_2\rho_{32}-iG_2^*\rho_{23},\\ \dot{\rho}_{23}&=-(\gamma_2+i\Delta_2)\rho_{23}-iG_2(\rho_{22}-\rho_{33}),\\ \dot{\rho}_{33}&=2(\gamma_1+\Lambda)\rho_{11}+2\gamma_2\rho_{22}-2\Lambda\rho_{33}-iG_2\rho_{32}+iG_2^*\rho_{23}.\end{aligned} \tag{1}$$

The spectrum of spontaneous emission on the transition $|1\rangle\leftrightarrow|3\rangle$ is related to the dipole-dipole correlation function defined via the operators $A_{\alpha\beta}=|\alpha\rangle\langle\beta|$:

$$S(\omega_1)=\gamma_1\mathrm{Re}\left\{\lim_{t\to\infty}\int_0^\infty d\tau\langle A_{13}(t+\tau)A_{31}(t)\rangle\right\}. \tag{2}$$

We have chosen the normalization in Eq. (2) such that $S(\omega_1=\omega_{13})=1$ in the absence of the coherent drive. Using the density matrix equations (1) and the quantum regression theorem, we have calculated the spectrum. As these calculations are fairly standard, we only present the final result for $\Lambda_0=0$:

$$S(\omega_1)\equiv\rho_{11}\mathrm{Re}\left\{\frac{\gamma_1(\gamma_1+\gamma_2+\Lambda+i\Delta_2-i\Delta_1)}{|G_2|^2+(\gamma_1+\gamma_2+\Lambda+i\Delta_2-i\Delta_1)(\gamma_1+2\Lambda-i\Delta_1)}\right\}. \tag{3}$$

In Eq. (3), ρ_{11} is the steady-state solution of Eqs. (1):

$$\rho_{11}=\frac{\Lambda(\gamma_2^2+\Delta_2^2+|G_2|^2)}{[|G_2|^2(2\gamma_1+3\Lambda)+(\gamma_1+2\Lambda)(\gamma_2^2+\Delta_2^2)]}. \tag{4}$$

The whole issue of interferences is contained in the behavior of the curly brackets in Eq. (3). In order to make the analysis physically transparent, we consider the simpler case in which the coherent drive is on resonance with the $|2\rangle\leftrightarrow|3\rangle$ transition. In this special case we derive from Eq. (3) the following expression for $S(\omega_1)$:

$$\begin{aligned}S(\omega_1)/\rho_{11}=&\frac{\gamma_1}{2}L_\Gamma(\Delta_1-\Delta_0)+\frac{\gamma_1}{2}L_\Gamma(\Delta_1+\Delta_0)\\&+\frac{(\Lambda-\gamma_2)\gamma_1}{4\Delta_0}\{D_\Gamma(\Delta_1-\Delta_0)-D_\Gamma(\Delta_1+\Delta_0)\},\end{aligned} \tag{5}$$

where $L_\Gamma(X)$ and $D_\Gamma(X)$ denote, respectively, the Lorentzian and dispersive profiles

$$L_\Gamma(X)\equiv\frac{\Gamma}{\Gamma^2+X^2},\quad D_\Gamma(X)\equiv\frac{X}{(\Gamma^2+X^2)}. \tag{6}$$

The other symbols in Eq. (5) are defined by

$$\Gamma=\gamma_1+\frac{\gamma_2}{2}+\frac{3\Lambda}{2},\quad \Delta_0=\sqrt{|G_2|^2-\frac{1}{4}(\Lambda-\gamma_2)^2}. \tag{7}$$

It is assumed that the strength of the coherent drive is such that Δ_0 is *real*. The spectrum of emission on the transition $|1\rangle\to|3\rangle$ is thus given by two "Lorentzian" contributions located at $\omega_{13}-\omega_1=\pm\Delta_0$ and two dispersive contributions. The *dispersive contributions* are the result of *quantum interferences* as can be seen from the following argument: For large Δ_0 and for spontaneous emission in the region $\Delta_1\sim\Delta_0$, Eq. (5) can be approximated by

$$S(\omega)/\rho_{11}=\gamma_1L_\Gamma(\Delta_1-\Delta_0)/2. \tag{8}$$

Thus in the frequency region $\Delta_1\sim\Delta_0$, the spontaneous emission spectrum is well approximated by a single Lorentzian with half-width Γ. For the region near the line center, i.e., $\Delta_1\sim0$, we have contributions (a) from the tails of the two Lorentzians, which are of the order of Γ/Δ_0^2 and (b) from the two dispersive terms, which are of the order Γ/Δ_0. However, the weight factor of the dispersive term is also of the order γ/Δ_0. Thus in the region of the line center, *both* Lorentzian and dispersive contributions can be of *similar magnitude*. Note further that the traditional secular approximation will *miss* the dispersive contributions in Eq. (5).

The situation considered by Zhu *et al.* corresponds to an external pumping of the state $|1\rangle$ [$\Lambda\to0$ in Eq. (5), $\Lambda_0\neq0$] and no spontaneous emission on the transition $|2\rangle\to|3\rangle$ ($\gamma_2\to0$). In these limits the spectrum becomes a *sum of two Lorentzians* as the weight factor of the dispersive terms goes to zero, and thus we recover the result of Zhu *et al.*

The value at the line center for large Δ_0 compared to Γ is given by

$$\begin{aligned}S(\omega_1=\omega_{13})/\rho_{11}&\equiv\frac{\gamma_1(\gamma_1+\gamma_2+\Lambda)}{|G_2|^2+(\gamma_1+2\Lambda)(\gamma_1+\gamma_2+\Lambda)}\\&\approx\gamma_1(\gamma_1+\gamma_2+\Lambda)/|G_2|^2.\end{aligned} \tag{9}$$

The quantum interference [γ_2-term in Eq. (9)] is *constructive*. The quantum interference arises from the opening up of a new channel for spontaneous emission. In passing we also note from Eq. (5) that the pumping ($\Lambda\neq0$) out of the state $|3\rangle$ also produces interference that is destructive in nature, though it should be borne in mind that ρ_{11} (for $\Lambda_0=0$) is a

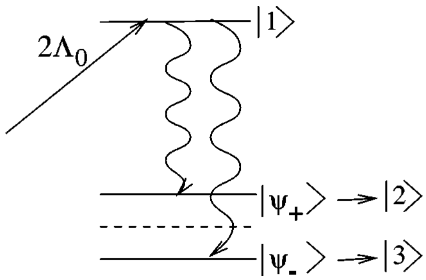

FIG. 2. Schematic illustration of the path for emission to either of the dressed states $|\psi_+\rangle$ and $|\psi_-\rangle$.

function of Λ. This constructive or destructive interference can be understood from our dressed-state analysis below.

We would next like to analyze the situation physically. We examine different pathways contributing to emission. For a field on resonance, the coherent part of the interaction is

$$H_c = -\hbar G_2(|2\rangle\langle 3| + |3\rangle\langle 2|). \tag{10}$$

Thus instead of working with bare states $|2\rangle$ and $|3\rangle$, we work with the dressed states

$$H_c|\psi_\pm\rangle = \pm\hbar G_2|\psi_\pm\rangle, \quad |\psi_\pm\rangle = (|2\rangle \mp |3\rangle)/\sqrt{2}. \tag{11}$$

The matrix elements of ρ in the dressed basis satisfy

$$\dot{\rho}_{1\pm} = \pm iG_2\rho_{1\pm} - \left(\frac{\gamma_2 - \Lambda}{2}\right)\rho_{1\mp} - (\Gamma + i\Delta_1)\rho_{1\pm}, \tag{12}$$

$$\dot{\rho}_{++} = (\gamma_1 + \Lambda)\rho_{11} - \frac{\Lambda}{2}(\rho_{++} + \rho_{--} - \rho_{+-} - \rho_{-+}) - \frac{\gamma_2}{2}(\rho_{++} - \rho_{--}). \tag{13}$$

Consider first the case when the system is externally pumped $(\Lambda = 0, \Lambda_0 \neq 0)$. Then Eqs. (12) and (13) reduce to

$$\dot{\rho}_{1\pm} = \pm iG_2\rho_{1\pm} - (\Gamma + i\Delta_1)\rho_{1\pm} - \frac{\gamma_2}{2}\rho_{1\mp}, \tag{14}$$

$$\dot{\rho}_{++} = \gamma_1\rho_{11} - \frac{\gamma_2}{2}(\rho_{++} - \rho_{--}). \tag{15}$$

In the limit $\gamma_2 \to 0$, the *coupling between the coherences*, i.e., the coupling of $\rho_{1\pm}$ to $\rho_{1\mp}$ is *missing*. Besides the terms like $(\gamma_2/2)\rho_{--}$ in Eq. (15), *terms responsible for transitions between the dressed states* $|\psi_\pm\rangle$ are *missing*. Thus Fig. 2 describes the physical situation in which the states $|\psi_\pm\rangle$ *do not interact* with each other. Thus there is only a *single pathway* for each transition $|1\rangle \to |\psi_+\rangle$ (or $|1\rangle \to |\psi\rangle$) and therefore we do *not* see any *quantum interference* in the spectrum of spontaneous emission.

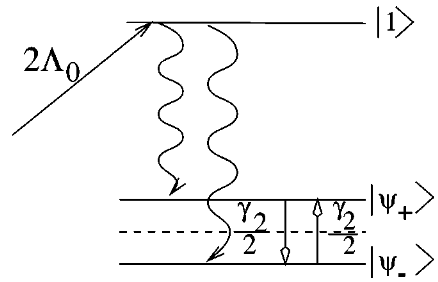

FIG. 3. Schematic illustration of the extra pathway created by spontaneous emission on the transition $|2\rangle \to |3\rangle$ for populating *any* of the two dressed states.

The situation is different when $\gamma_2 \neq 0$. The states $|\psi_+\rangle$ and $|\psi_-\rangle$ continue to interact with each other, i.e., the transitions take place between $|\psi_+\rangle$ and $|\psi_-\rangle$. This is shown schematically in Fig. 3. Because of the *final state interaction*, one now has two pathways for emission to any of the dressed states, say $|\psi_+\rangle$,

$$|1\rangle \overset{\gamma_1}{\to} |\psi_+\rangle,$$

$$|1\rangle \overset{\gamma_1}{\to} |\psi_-\rangle \overset{\gamma_2}{\to} |\psi_+\rangle. \tag{16}$$

Thus spontaneous emission on the transition $|2\rangle \to |3\rangle$ opens up *a new pathway*, making quantum interference possible.

In general the cross coupling between different coherences and between coherences and populations is very important in the determination of the spectrum of emission, as the spectrum is determined by the two-time correlation function $\langle A_{13}(t+\tau)A_{31}(t)\rangle$, which in turn is determined by $\langle A_{1+}(t+\tau)A_{31}(t)\rangle$ and $\langle A_{1-}(t+\tau)A_{31}(t)\rangle$. Clearly these correlations will satisfy equations analogous to Eq. (12). The cross-coupling term is responsible for the interference. Note the coefficient of this term $(\gamma_2 - \Lambda)$, which is the same as the one appearing in Eq. (5). Thus terms beyond the secular approximation in the dressed-state analysis enable us to understand the existence and nature of the interference effect.

In conclusion, we have shown how the inclusion of spontaneous emission on the laser-driven transition in a V system provides a new pathway leading to quantum interference. We show that the interference itself manifests in the form of dispersive contributions to the emission line shape. The strength of such dispersive contributions is strongly dependent on the Einstein *A* coefficient of the laser-driven transition.

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