Control-laser-induced subnatural linewidths and quenching of spontaneous emission

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(Received 13 May 1996)

We show how control lasers lead to subnatural linewidths in spontaneous emission as well as to the quenching of the quantum noise. We trace this quenching to the dispersive contributions to the line shape. We obtain analytical expressions for linewidths that for $\wedge$ systems are half the natural linewidths with the possibility of much more reduction for detuned control laser. We further present a simple physical analysis in terms of a *two-photon* Fermi golden rule to understand cancellation of spontaneous emission. Finally, we relate our results to recent experiments. [S1050-2947(96)50111-3]

PACS number(s): 42.50.Gy, 42.50.Lc

Early publications [1,2] revealed the possibility of quantum interferences in spontaneous emission; in particular, a degenerate $\vee$ system had the possibility of coherent population trapping [1] in an excited state. In recent times there has been a revival [3–6] of interest in interference effects in spontaneous emission. The spectrum of emission has been analyzed under a variety of conditions involving excitation by laser fields. Though the spectra for $\vee$, $\wedge$, and Ξ systems were calculated in the late 1970s and early 1980s [7], one has now realized [3–5] the possibility of many new interesting results such as line narrowing, besides the possibility of establishing contact with experiments [8–10].

In this Rapid Communication we show that control lasers can lead to subnatural linewidths in spontaneous emission as well as to the quenching of spontaneous emission noise. We demonstrate how the quenching of emission noise comes about from quantum interferences. We demonstrate all this in the context of $\wedge$ systems that have been extensively studied in connection with lasing without inversion, population trapping, and a host of other problems [11]. The results we present are also relevant in the context of reported experiments [9,10] on the quenching and even cancellation of spontaneous emission. Although we present a detailed density-matrix analysis, we conclude the paper with a very simple model using the two-photon Fermi golden rule [12], which enables us to understand physically the quenching of spontaneous emission noise. We also establish a connection of our results to some recent experiments [9,10].

The details of the $\wedge$ system are fairly standard [7(b)] and we illustrate it in Fig. 1(a). Here $2G_2$ is the Rabi frequency of the control laser of frequency ω_l, 2Λ is the pumping, and $2\gamma_1$ and $2\gamma_2$ are, respectively, the rates of spontaneous emission on the two transitions. The spectrum of spontaneous emission on the transition $|1\rangle \leftrightarrow |3\rangle$ is known to be related to the two time correlation function

$$S(\omega) \equiv \int_0^\infty d\tau \langle A_{13}(t+\tau) A_{31}(t) \rangle e^{i\Delta_1 \tau} + \text{c.c.}, \tag{1}$$

where $\Delta_1 = \omega_{13} - \omega$ and $A_{\alpha\beta}(t) \equiv |\alpha\rangle\langle\beta|$ is the dipole moment operator for the transition $|\alpha\rangle \leftrightarrow |\beta\rangle$. The correlation function in Eq. (1) can be calculated using the usual density-matrix equations and the quantum regression theorem. After a canonical transformation to eliminate the optical frequencies, the density-matrix equations read

$$\begin{aligned}
\dot{\rho}_{11} &= -2(\gamma_1 + \gamma_2 + \Lambda)\rho_{11} + 2\Lambda \rho_{33} + iG_2 \rho_{21} - iG_2^* \rho_{12}, \\
\dot{\rho}_{22} &= 2\gamma_2 \rho_{11} - iG_2 \rho_{21} + iG_2^* \rho_{12}, \\
\dot{\rho}_{21} &= -(\Gamma_{21} - i\Delta_2)\rho_{21} - iG_2^* \rho_{22} + iG_2^* \rho_{11},
\end{aligned} \tag{2}$$

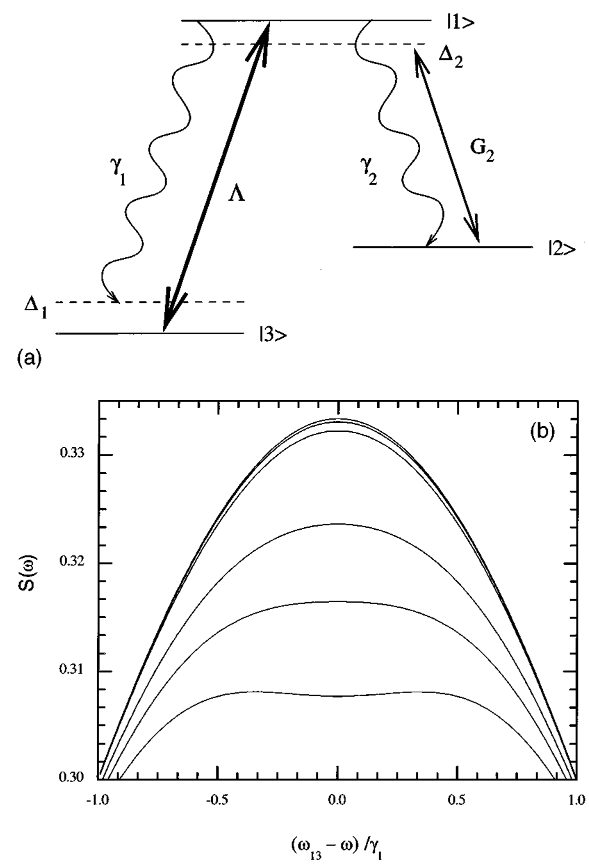

FIG. 1. (a) Schematics of the $\wedge$ system; (b) the spectrum $S(\omega)$ of spontaneous emission on the transition $|1\rangle \leftrightarrow |3\rangle$ for moderate field strengths in the limit $\gamma_2 \rightarrow 0$, $\Lambda/\gamma_1 = 1.0$ and for $G_2/\gamma_1 = 0$, 0.05, 0.1, 0.3, 0.4, and 0.5 (starting from the top curve).

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$$\dot{\rho}_{31}=-\Gamma_{31}\rho_{31}-iG_2^*\rho_{32},$$

$$\dot{\rho}_{32}=-(\Gamma_{32}+i\Delta_2)\rho_{32}-iG_2\rho_{31},$$

where $\Gamma_{\alpha\beta}$ denotes the total decay rate of the off-diagonal element $\rho_{\alpha\beta}$. For our model

$$\Gamma_{31}\equiv\gamma_1+\gamma_2+2\Lambda,\quad \Gamma_{32}=\Lambda,$$
$$\Gamma_{21}=\gamma_1+\gamma_2,\quad \Delta_2=\omega_{12}-\omega_l. \tag{3}$$

The relevant equations for the calculation of Eq. (1) are

$$\left\{\frac{d}{d\tau}+\begin{pmatrix}\Gamma_{31} & iG_2^*\\ iG_2 & \Gamma_{32}+i\Delta_2\end{pmatrix}\right\}\begin{pmatrix}\langle A_{13}(t+\tau)A_{31}(t)\rangle\\ \langle A_{23}(t+\tau)A_{31}(t)\rangle\end{pmatrix}=0, \tag{4}$$

which are to be solved subject to initial conditions

$$\langle A_{13}A_{31}\rangle=\rho_{11},\quad \langle A_{23}A_{31}\rangle=\rho_{12}. \tag{5}$$

We next discuss various interesting features that follow from the analysis of Eqs. (4) and (2).

SUBNATURAL LINEWIDTHS

Clearly the spectral characteristics of $S(\omega)$ will be determined by the zeros of the polynomial

$$P(z)=(z+\Gamma_{31})(z+\Gamma_{32}+i\Delta_2)+|G_2|^2, \tag{6}$$

i.e., by

$$z_\pm=-\tfrac{1}{2}(\Gamma_{31}+\Gamma_{32}+i\Delta_2)$$
$$\pm\tfrac{1}{2}\sqrt{(\Gamma_{32}+i\Delta_2-\Gamma_{31})^2-4|G_2|^2}. \tag{7}$$

We analyze the roots in the limit of large Rabi frequency $\sqrt{4|G_2|^2+\Delta_2^2}\equiv\Omega$. To leading order in Γ/Ω we can write the two roots of Eq. (6) as

$$z_\pm\simeq-\frac{1}{2}(\Gamma_{31}+\Gamma_{32}+i\Delta_2)\pm\frac{1}{2}i\Omega\mp\frac{1}{2}(\Gamma_{31}-\Gamma_{32})\frac{\Delta_2}{\Omega}$$
$$=\pm\frac{1}{2}i(\Omega\mp\Delta_2)-\frac{1}{2}\Gamma_{31}\left(1\pm\frac{\Delta_2}{\Omega}\right)-\frac{1}{2}\Gamma_{32}\left(1\mp\frac{\Delta_2}{\Omega}\right). \tag{8}$$

Thus according to Eq. (1) the spectrum will be determined by the complex poles

$$-i\Delta_1=z_\pm. \tag{9}$$

The resonances in the spectrum occur at

$$\Delta_1=\mp\tfrac{1}{2}(\Omega\mp\Delta_2), \tag{10}$$

with half widths

$$\frac{\Gamma_{31}}{2}\left(1\pm\frac{\Delta_2}{\Omega}\right)+\frac{\Gamma_{32}}{2}\left(1\mp\frac{\Delta_2}{\Omega}\right). \tag{11}$$

Thus the peak at $\Delta_1=(\Omega+\Delta_2)/2$ will exhibit a width which could be *much smaller than the natural linewidth if* $\Delta_2\gg2G_2$ *and if* Λ *is very small*. On the contrary, the peak at $\Delta_1=-(\Omega-\Delta_2)/2$ is broadened beyond the homogeneous linewidth. For $\Delta_2=0$, both the peaks have identical widths $(\Gamma_{31}+\Gamma_{32})/2$, which for a small rate of pumping reduces to *half* the width in the absence of the control laser [13].

The line-narrowing effect has been discussed by several authors in different contexts. In Ref. [14] Cohen-Tannoudji and Reynaud calculate the width of the dressed states and they derive the first term in Eq. (11). Thus the line-narrowing result is implicitly contained in that work. As mentioned in the Introduction the $\bigvee$ and Ξ systems have been shown to exhibit narrowing under different conditions of excitation. Zhu, Gauthier, and Mossberg [8] observed narrowing as predicted by Narducci *et al.* [6] for a driven $\bigvee$ system with one strong and one weak transition. Reference [5] reports line narrowing for ladder systems.

QUANTUM INTERFERENCES–QUENCHING BY DISPERSIVE CONTRIBUTIONS

After having discussed the possibilities of the subnatural widths, we discuss the actual spectrum of emission. We then demonstrate quenching of spontaneous emission in the region $\Delta_1\sim0$. For simplicity, we consider the case $\Delta_2=0$. The calculation shows that

$$S(\omega)=2\rho_{11}\mathrm{Re}\left\{\frac{(-i\Delta_1+\Gamma_{32}+\gamma_2)}{(\Gamma_{31}-i\Delta_1)(\Gamma_{32}-i\Delta_1)+|G_2|^2}\right\}, \tag{12}$$

which on simplification and on assuming $G_2\gg\Gamma_{ij}$ reduces to

$$S(\omega)=2\rho_{11}\mathrm{Re}\left\{\frac{iG_2+\gamma_2-\frac{1}{2}(\Gamma_{31}-\Gamma_{32})}{\left[-i\Delta_1-\left(iG_2-\frac{\Gamma_{31}+\Gamma_{32}}{2}\right)\right](2iG_2)}+\frac{-iG_2+\gamma_2-\frac{1}{2}(\Gamma_{31}-\Gamma_{32})}{(-2iG_2)\left[-i\Delta_1-\left(-iG_2-\frac{\Gamma_{31}+\Gamma_{32}}{2}\right)\right]}\right\}. \tag{13}$$

The total spectrum thus consists of a *sum of Lorentzian and dispersive contributions*. On using Eq. (3) in Eq. (13) we get

$$S(\omega)/\rho_{11}=\left\{\frac{\frac{1}{2}(\gamma_1+\gamma_2+3\Lambda)}{(\Delta_1+G_2)^2+\left(\frac{\gamma_1+\gamma_2+3\Lambda}{2}\right)^2}+\cdots\right\}$$
$$+\frac{(\gamma_2-\gamma_1-\Lambda)}{2G_2}$$
$$\times\left\{\frac{\Delta_1+G_2}{(\Delta_1+G_2)^2+\left(\frac{\gamma_1+\gamma_2+3\Lambda}{2}\right)^2}-\cdots\right\}, \tag{14}$$

where "$\cdots$" represent terms with $G_2\to-G_2$ and where

$$\rho_{11}\equiv\Lambda|G_2|^2\{3\Lambda|G_2|^2+\gamma_1|G_2|^2+\gamma_2\Lambda(\gamma_1+\gamma_2+\Lambda)\}^{-1}. \tag{15}$$

Note that in the region $\Delta_1 = G_2$ the dispersive contribution is unimportant and the spectrum is well approximated by a single linewidth $(\gamma_1+\gamma_2+3\Lambda)$. However, for the region $\Delta_1 \sim 0$, all four contributions in Eq. (14) are equally important. The last two dispersive contributions are the interferences. At the line center $\Delta_1=0$, the contribution of two Lorentzians is

$$L=\frac{(\gamma_1+\gamma_2+3\Lambda)}{G_2^2}. \tag{16}$$

The interference terms from dispersive contributions lead to

$$D=\frac{(\gamma_2-\gamma_1-\Lambda)}{G_2^2}. \tag{17}$$

This can be constructive or destructive depending on the sign of $(\gamma_2-\gamma_1-\Lambda)$. The quenching of noise occurs if

$$\gamma_1+\Lambda>\gamma_2. \tag{18}$$

The contribution (16) is what one would expect from a simple argument based on detuning, i.e., on the line wings.

We next examine the form of Eq. (12) in the limit $\gamma_2 \rightarrow 0$ and for *moderate values* of G_2. The results are shown in Fig. 1(b). We see that with an increase in G_2 there is reduction in the peak height at $\Delta_1=0$. This trend is consistent with the observation of Suckewer and co-workers [10] on the quenching of spontaneous emission. The reduction further depends on relative values of G_2 and γ_1. Thus for a fixed G_2 the transition with smaller γ_1 will be most affected. This is again consistent with experimental observation [10,15].

SPONTANEOUS EMISSION CANCELLATION FROM TWO-PHOTON FERMI GOLDEN RULE

We conclude the paper by presenting a simple physical picture for the interference minimum in spontaneous emission. Our picture will be based on the generalization of the Fermi golden rule to second-order processes. Consider the somewhat simpler model shown in Fig. 2(a). Let the pump be a broadband source. In the dressed states created by the control laser the various pumping and emission processes become as shown in Fig. 2(b). The net Hamiltonian can be written in the form (all energies being measured from $|1\rangle$)

$$\begin{aligned} H=&-\hbar G_2(|1\rangle\langle 2|e^{-i\omega_l t}+|2\rangle\langle 1|e^{i\omega_l t})+E_2|2\rangle\langle 2| \\ &+E_g|g\rangle\langle g|+E_3|3\rangle\langle 3|+H_{0V}+H_{1V}+H_p, \\ H_p=&-\hbar g(|1\rangle\langle g|e^{-i\omega_g t}+\text{H.c.}) \\ H_{1V}=&-(|1\rangle\langle 3|(\vec{\mathcal{E}}_V\cdot\vec{d}_{13})+\text{c.c.}), \end{aligned} \tag{19}$$

where H_{OV} and $\mathcal{E}_V$ are, respectively, the unperturbed energy and amplitude of the vacuum of the electromagnetic field. The fast optical frequency ω_l can be eliminated by a rotating frame transformation and we can also introduce the dressed states $|\psi_\pm\rangle$. For simplicity, let us assume that $|E_2|=\hbar\omega_l$. In terms of dressed states $(|1\rangle=\alpha|\psi_+\rangle+\beta|\psi_-\rangle)$ we get

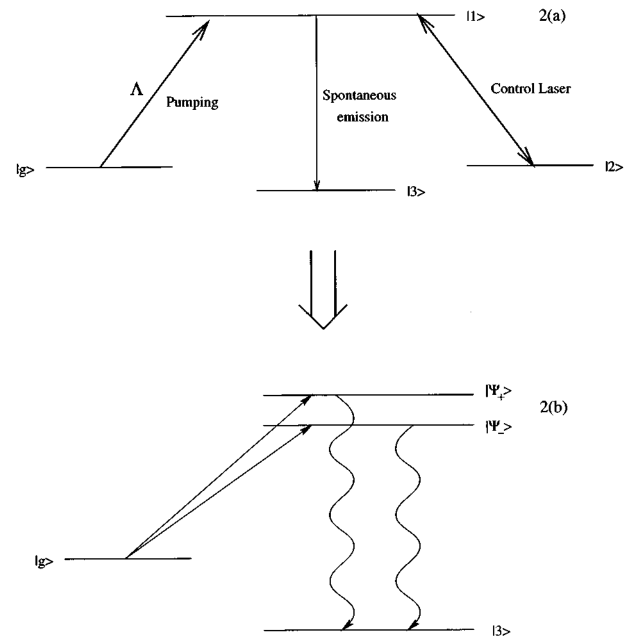

FIG. 2. Physical picture for the quenching of spontaneous emission; the two pathways for an effective second-order process in terms of dressed states are shown.

$$\begin{aligned} H=&E_+|\psi_+\rangle\langle\psi_+|+E_-|\psi_-\rangle\langle\psi_-|+E_g|g\rangle\langle g| \\ &+E_3|3\rangle\langle 3|+H_{OV}-\hbar g(|\alpha\psi_++\beta\psi_-\rangle\langle g|e^{-i\omega_g t}+\text{H.c.}) \\ &-[|\alpha\psi_++\beta\psi_-\rangle\langle 3|(\vec{\mathcal{E}}_V\cdot\vec{d}_{13})+\text{H.c.}]. \end{aligned} \tag{20}$$

We can now calculate the rate R for the absorption of a photon of frequency ω_g and emission of a photon of frequency ω_{ks}, momentum $\vec{k}$, and polarization s. This is a second-order process and we use a second-order Fermi golden rule [12],

$$\begin{aligned} R=&\frac{2\pi}{\hbar}\sum_{ks}\left|\frac{\langle 1_{ks},3|H_{1V}|0,\psi_+\rangle\langle 0,\psi_+|H_p|g;0\rangle}{(E_+-E_g-\hbar\omega_g)}\right. \\ &\left.+\frac{\langle 1_{ks},3|H_{1V}|0,\psi_-\rangle\langle 0,\psi_-|H_p|g;0\rangle}{(E_--E_g-\hbar\omega_g)}\right|^2 \\ &\times\delta(E_3-\hbar\omega_g+\hbar\omega_{ks}-E_g), \end{aligned} \tag{21}$$

where $|0\rangle$ and $|1_{ks}\rangle$ represent the states of the electromagnetic field with zero and one photon respectively. The matrix element in Eq. (21) is to be modified in the resonance region by taking into account appropriate damping effects. For a broadband pump with pump energy spread $\mathcal{E}$ we have to average over all pump energies $\hbar\omega_g$,

$$\begin{aligned} R=&\frac{2\pi}{\hbar\mathcal{E}}\sum_{ks}\left|\frac{\langle 1_{ks},3|H_V|0,\psi_+\rangle\langle\psi_+,0|H_p|g,0\rangle}{E_+-(E_3+\hbar\omega_{ks})}\right. \\ &\left.+\frac{\langle 1_{ks},3|H_V|0,\psi_-\rangle\langle\psi_-,0|H_p|g,0\rangle}{E_--(E_3+\hbar\omega_{ks})}\right|^2, \end{aligned} \tag{22}$$

where various matrix elements are easily read from Eq. (20). Clearly the transition rate can be zero if the matrix element in Eq. (22) can *vanish*. This clearly can happen for an appropriate frequency of the spontaneously emitted photon. Thus for a value of ω_{ks}, which in general depends on the matrix elements, *R* can become zero. For the example under consideration, interference occurs if $E_3 = \hbar\omega_{ks}$, since $E_+ = -E_-$. Thus the flourescence as a function of ω_k will exhibit a minimum that comes about from the interference between two paths as shown in Fig. 2(b). Note further that the matrix element in Eq. (22) is just the Raman matrix element. The *cancellation arises* from the energy dependence of the Raman polarizability or the dispersive nature of the interaction. Thus the simple physical picture based on the Fermi golden rule for second-order processes enables us to understand control-laser-induced interference effects and quenching of spontaneous emission. The results of a recent experiment [9] can also be understood in terms of the above two-photon picture, as the level scheme is identical to the one in Fig. 2(b).

Thus to conclude we have brought out the unusual features of spontaneous emission in $\wedge$ systems. We have shown how the subnatural linewidth can be obtained depending on the size of detuning and the strength of the control laser. We have further isolated the dispersive contributions that result in the quenching of spontaneous emission noise at the line center. Finally, the quenching is shown to follow in terms of a very simple physical picture.

ACKNOWLEDGMENTS

The author thanks S. Harris, M. O. Scully, S. Y. Zhu, K. Hakuta, S. Suckewer, and many others for enjoyable discussions over many years on the subject of quantum interferences.

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