Nature of the quantum interference in electromagnetic-field-induced control of absorption

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(Received 28 August 1996; revised manuscript received 15 October 1996)

Various three-level schemes for electromagnetic-field-induced control of absorption are analyzed to isolate the precise nature of the quantum interference. Such interference manifests through the dispersive contributions to the absorption line shapes. Depending on the excitation scheme, the dispersive terms can be either constructive or destructive. The collisional dephasing in some cases can change the nature of interference. [S1050-2947(97)04602-7]

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

The modification of the optical properties of a medium by the application of a strong electromagnetic field has been a subject of intense activity. In particular, the linear susceptibility responsible for absorption and dispersion of the medium has been shown to behave quite differently in the region of the line center [1–6]. The absorption can be made rather small (and even zero in some cases) by making the strength of the control laser rather large [1,2]. At first sight one might argue that the control laser has shifted the positions of resonances—and hence the absorption will go as a (shift in the line position)$^{-2}$. However, for a Λ system Harris and co-workers showed that there is a quantum interference which makes absorption zero at the line center. The zero (or near zero absorption in some other cases) has tremendous possibilities in nonlinear optics [1,2,4].

The previous results were derived either by using density-matrix equations or even by using equations for amplitudes (wherever appropriate). The zero in absorption is attributed to the interference between dressed states [7]. This appears to be the most direct way of understanding the absorption zero. The equations for Schrödinger amplitudes can be written only if the decay outside the system occurs. In general, one also has to account for decay within the system as well as the dephasing processes. Thus in this paper we *trace the origin of quantum interference effects directly at the density-matrix level*. We take the results of standard calculations and express the net absorption line shape as a sum of four different contributions: (a) two absorptive contributions (b) two dispersive contributions to the absorption line shape. In the region of resonance with the dressed states only one contribution out of the four is important. However, at the line center corresponding to the bare atom resonance all four terms contribute. The absorptive contributions are positive, as expected, whereas the dispersive contributions to the absorptions line shape could be positive or negative. This depends on the particular three level scheme at hand. The interference could thus be constructive or destructive. We will demonstrate this explicitly for four different situations. In order to keep the analysis simple we will assume pump on *resonance* with the transition on which it is applied though similar conclusions can be drawn if the pump is detuned. We will also assume that the *Rabi frequency of the pump is large* compared to the relaxation parameters.

Our general result for absorption A [in dimensionless units defined by Eq. (8)], from a probe field, can be written as

$$A=\frac{\Gamma}{2}[L_W(\Delta_1-G)+L_W(\Delta_1+G)] + \frac{\beta}{G}\frac{\Gamma}{2}[D_W(\Delta_1-G)-D_W(\Delta_1+G)], \tag{1}$$

where $2G$ is the Rabi frequency of the pump and Γ is the half width of the transition on which the probe is acting. We assume, without loss of generality, that G is real and positive. In Eq. (1) Δ_1 is the probe detuning, L and D are the Lorentzians and dispersive contributions defined by

$$L_W(x)=\frac{W/\pi}{x^2+W^2}, \quad D_W(x)=\frac{x/\pi}{x^2+W^2}. \tag{2}$$

In writing this expression for A the unimportant factors have been dropped. Here β is the interference parameter which can be positive or negative.

Equation (1) can be thought of as the net contribution coming to absorption from two different channels. The two channels correspond to the two dressed states created by the strong pump. These two channels are shown explicitly in Figs. 1(e) and 1(f). If the two channels were independent, then we will obtain only the contribution $A=\Gamma/2[L_W(\Delta_1-G)+L_W(\Delta_1+G)]$. The dispersive terms are clearly the interference terms. The four terms in Eq. (1) are reminiscent of the square of the coherent sum of two amplitudes. The presence of the dispersive contribution to absorption line shape always implies the existence of phase dependent effects.

In the resonance region say $\Delta_1\sim G$,

$$A\sim\frac{\Gamma}{2}L_W(\Delta_1-G), \tag{3}$$

as the remaining contributions will be of the order of $1/G^2$. However, for $\Delta_1\sim 0$ all four contributions are important. In particular, for $\Delta_1=0$ we get

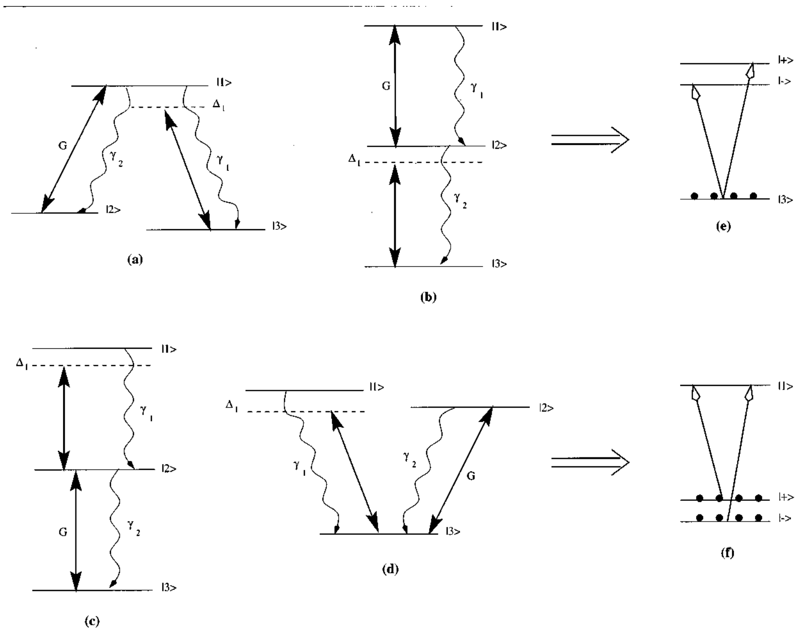

FIG. 1. Schematic representation of various level schemes. The spontaneous emission rates are $2\gamma's$; the Rabi frequency of the resonant pump is $2G$ and Δ_1 is the detuning of the probe field. The cases (a) and (b) [(c) and (d)] correspond to the case of the upper state (lower state) coherence in terms of dressed states as shown in the part (e) [(f)].

$$A=\frac{W-\beta}{G^2}. \tag{4}$$

It should be borne in mind that the dispersive contributions occur in the imaginary part of the response function. In order to see if β is positive or negative we consider several excitation schemes.

A. Λ *System*: Consider the Λ system shown in Fig. 1(a). A coherent pump on resonance acts on the transition $|1\rangle\leftrightarrow|2\rangle$ Its Rabi frequency is $2G$. The probe of frequency ω acts on the transition $|1\rangle\leftrightarrow|3\rangle$. The state $|1\rangle$ decays at the rate $2\gamma_1$ ($2\gamma_2$) to the state $|3\rangle$ ($|2\rangle$). The absorption from the probe is proportional to the imaginary part of the induced polarization on $|1\rangle\leftrightarrow|3\rangle$ transition, i.e., ρ_{13}. This can be seen from the Maxwell equation for the field E_1 in the slowly varying envelope approximation, which yields $|E_1|^2 \sim|\exp\{i\omega/c\sqrt{1+4\pi\chi(\omega)}L\}|^2$ and which for a dilute medium becomes

$$\sim\left|\exp\left\{\frac{i\omega}{c}L+\frac{2\pi i\omega\chi(\omega)L}{c}\right\}\right|^2.$$

Thus the intensity attenuation αL of the probe will be given by

$$\alpha L=\frac{4\pi\omega L}{c}\mathrm{Im}[\chi(\omega)]. \tag{5}$$

Note further that the induced polarization at the probe frequency is $nd_{31}\rho_{13}$, where n is the density of atoms. Thus $\chi(\omega)=n|d_{31}|^2\rho_{13}/\hbar(d_{13}E_1/\hbar)$ and, hence, the final result for αL is,

$$\alpha L=\frac{4\pi\omega Ln|d_{31}|^2}{\hbar c}\mathrm{Im}\left[\frac{\rho_{13}}{(d_{13}E_1/\hbar)}\right]. \tag{6}$$

Note that the Rabi frequency of the probe is $2d_{13}E_1/\hbar$. We can work in the dimensionless units by rewriting Eq. (6) as

$$\alpha L=\alpha_0 L\,\mathrm{Im}\left(\frac{\rho_{13}\Gamma_{13}}{(d_{13}E_1/\hbar)}\right), \tag{7}$$

where

$$\alpha_0 L=\frac{4\pi\omega Ln|d_{31}|^2}{\hbar c\Gamma_{13}}. \tag{8}$$

Here, Γ_{13} is the line width of the bare transition $|1\rangle\leftrightarrow|3\rangle$. This is equal to the decay rate of the off-diagonal element ρ_{13} of the density matrix. We will concentrate on the dimensionless quantity

$$A=\mathrm{Im}\left(\frac{\rho_{13}\Gamma_{13}}{(d_{13}E_1/\hbar)}\right). \tag{9}$$

For Λ system the density-matrix equations lead to the well-known [1] expression

$$A=\mathrm{Re}\frac{(\Gamma_{23}+i\Delta_1)\Gamma_{13}}{G^2+(\Gamma_{13}+i\Delta_1)(\Gamma_{23}+i\Delta_1)}, \tag{10}$$

where $\Delta_1=\omega_{13}-\omega$ and Γ_{23} is the decay rate of the coherence ρ_{23}. Note that $\Gamma_{13}=\gamma_1+\Gamma_{13}^{(d)}$, $\Gamma_{23}=\Gamma_{23}^{(d)}$, where $\Gamma_{\alpha\beta}^{(d)}$ represents contributions of dephasing collisions. We exclude the possibility of direct transition between $|2\rangle\leftrightarrow|3\rangle$. The dressed states and the decay rates are identified by the poles of Eq. (10). For large G, these occur at

$$\Delta_1=\pm G \tag{11}$$

with widths

$$W=\tfrac{1}{2}(\Gamma_{13}+\Gamma_{23}). \tag{12}$$

The absorption at the origin is proportional to Γ_{23}. Note that a simple argument based on two Lorentzians located at Eq. (11) would yield a proportionality factor $\frac{1}{2}(\Gamma_{13}+\Gamma_{23})$. Clearly there is quantum interference which leads to

$$\tfrac{1}{2}(\Gamma_{13}+\Gamma_{23})\rightarrow\tfrac{1}{2}(\Gamma_{13}+\Gamma_{23})+\tfrac{1}{2}(\Gamma_{23}-\Gamma_{13})=\Gamma_{23}. \tag{13}$$

The interference parameter β is equal to

$$\beta=(\Gamma_{13}-\Gamma_{23})/2. \tag{14}$$

If there is no dephasing ($\Gamma_{23}=0$) then the *absorptive and dispersive contributions cancel each other leading to a zero in the absorption profile*. Note that in this scheme the interference effects are *destructive*. The interference vanishes if dephasing is such that $\Gamma_{13}=\Gamma_{23}$. However, if the dephasing is such that $\Gamma_{13}\gg\Gamma_{23}$, then again the interference zero is almost restored as already realized by Imamoglu and Harris [5].

B. Ladder System: Consider the ladder system of Fig. 1(b). We assume that the probe acts on the lower transition, whereas the resonant pump acts on the upper transition. The interpretation of various symbols is as in the Λ system. The absorption from the probe is given by [2]

$$A=\mathrm{Re}\frac{(\Gamma_{13}+i\Delta_1)\Gamma_{23}}{G^2+(\Gamma_{23}+i\Delta_1)(\Gamma_{13}+i\Delta_1)}, \tag{15}$$

where $\Delta_1=\omega_{23}-\omega$, $\Gamma_{23}=\gamma_2+\Gamma_{23}^{(d)}$ and $\Gamma_{13}=\gamma_1+\Gamma_{13}^{(d)}$. The absorption can be written in the form (1) with

$$W=\tfrac{1}{2}(\Gamma_{13}+\Gamma_{23}),\quad \beta=(\Gamma_{23}-\Gamma_{13})/2. \tag{16}$$

Like the Λ system one has *destructive* interference if $\Gamma_{13}=0$. The proportionality factor in Imχ for $\Delta_1=0$ is determined by the dephasing rate of the two photon off-diagonal coherence ρ_{13}. Note that even in the absence of collisional dephasing Γ_{13} will depend on the spontaneous rate of emission from the state $|1\rangle$. One can get close to a zero absorption rate only if the spontaneous emission from $|1\rangle$ is negligible. Note that the interference will disappear if

$$\Gamma_{13}=\Gamma_{23}, \tag{17}$$

which, for example, will be the case in the absence of dephasing collisions and if $\gamma_1=\gamma_2$. Note further that the sign of the interference parameter depends on the relative magnitudes of γ_1 and γ_2.

C. Ladder system: We continue with our discussion of ladder system but with the pump and probe transitions switched [Fig. 1(c)]. The pump is on resonance and there is no probe absorption in the absence of the pump. The absorption profile essentially has the form (cf. Ref. [8])

$$A\equiv\frac{G^2}{(\Gamma_{23}\gamma_2+2G^2)}\mathrm{Re}\frac{(\Gamma_{13}+\gamma_2+i\Delta_1)\Gamma_{12}}{G^2+(i\Delta_1+\Gamma_{13})(i\Delta_1+\Gamma_{12})}. \tag{18}$$

Note the explicit dependence of this absorption profile on the rate of spontaneous emission from state $|2\rangle$. This is in addition to what is contained in the decay rates $\Gamma_{\alpha\beta}$ of coherences, i.e., off-diagonal elements. The net line shape is given by Eq. (1) with

$$W=\frac{1}{2}(\Gamma_{13}+\Gamma_{12})\rightarrow\gamma_1+\frac{\gamma_2}{2}, \tag{19}$$

$$\beta\equiv\frac{1}{2}(\Gamma_{12}-\Gamma_{13}-2\gamma_2)\rightarrow-\frac{\gamma_2}{2}, \tag{20}$$

where the last results follow in the absence of dephasing. In contrast to cases A and B, the interference parameter β is negative. Thus the quantum interference is *constructive*. The interference vanishes if $\gamma_2\rightarrow 0$.

D. V System: Finally we examine the nature of interference in the V system, as shown in Fig. 1(d). The pump is on resonance with the transition $|2\rangle\leftrightarrow|3\rangle$. The probe is on $|1\rangle\leftrightarrow|3\rangle$ transition. The absorption from the probe in given by [4,9]

$$A\equiv\frac{(G^2+\gamma_2\Gamma_{23})}{(2G^2+\gamma_2\Gamma_{23})}\mathrm{Re}\frac{\left[\Gamma_{12}+\gamma_2\left(\dfrac{G^2}{G^2+\gamma_2\Gamma_{23}}\right)+i\Delta_1\right]\Gamma_{13}}{G^2+(i\Delta_1+\Gamma_{12})(i\Delta_1+\Gamma_{13})}. \tag{21}$$

The situation is similar to the one in case C. The absorption line shape can be expressed in the form (1) with

$$W=\frac{1}{2}(\Gamma_{12}+\Gamma_{13})\rightarrow\gamma_1+\frac{\gamma_2}{2}, \tag{22}$$

$$\beta=\frac{1}{2}(\Gamma_{13}-\Gamma_{12}-2\gamma_2)\rightarrow\frac{-3\gamma_2}{2}, \tag{23}$$

where the last result is in the absence of dephasing collisions. The quantum interference is *constructive*. Note further that if $\gamma_2\approx 0$, then the interference term vanishes.

The above analysis clearly shows that there is real distinction between cases A, B and C, D. In cases A and B (C and D) the pump acts with initially unoccupied (occupied) levels. The dressed state picture is perhaps more appealing, however, one must remember that one has to work beyond secular approximation to see the role of quantum interferences as such an approximation will miss the dispersive contributions in Eq. (1). In the dressed state picture one has the situation shown in Figs. 1(e) and 1(f). Clearly the nature of interference is different, depending upon the existence of upper state or lower state coherence [10]. Note further, that the transitions $|3\rangle\leftrightarrow|\pm\rangle$ are not independent because of the radiative coupling (for an explicit result, see Ref. [11]). Note that in cases 1(a), 1(b) (1(c), 1(d)) the states $|\pm\rangle$ are unoccupied (occupied). This is an important source of distinction in the two cases.

In conclusion we have identified the precise nature of interference at the line center. As a measure of interference we introduced a quantitative parameter β and presented explicit results for it in terms of spontaneous emission decay rates and various dephasing parameters. By changing collisional parameters as well as spontaneous emission rates, the sign of β can be changed.

The author thanks M.O. Scully for interesting discussions on atomic coherence effects.

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