Quantum Interference in Probe Absorption: Narrow Resonances, Transparency, and Gain without Population Inversion

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We examine the absorption of a weak probe beam for a V-type atom with a closely spaced doublet and demonstrate that quantum interference between the two excitation pathways can result in very narrow resonances, transparency, and even gain without population inversion. The origin of these effects is discussed. [S0031-9007(96)02035-2]

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Following earlier work [1–3], there has been much interest in a variety of new effects which have their origin in the phenomenon of quantum interference. Examples are lasing without population inversion [3,4], electromagnetically induced transparency [5], enhancement of the index of refraction without absorption [6], and fluorescence quenching [1,2,7–10]. Quantum interference between two laser-induced channels can lead to the elimination of the spectral line at the driving laser frequency in the spontaneous emission spectrum [8], and to the existence of a dark line in the spontaneous emission from one of the excited sublevels [11]. The former effect has been observed very recently by Xia *et al.* [9] in sodium dimers, with the dipole moments of the two upper levels for spontaneous emission being parallel. We have recently shown that quantum interference can give rise to a very narrow resonance in fluorescence [10].

In this paper we investigate the effects of quantum interference in a V-type atom—a model similar to others studied previously [1,2,7,10] except for the absence of a coherent driving field. We report a variety of effects, including narrow resonances, transparency, and gain without inversion. Since experiments on this type of system have been recently reported [5,9], and experiments involving probe absorption should be easier than those detecting resonance fluorescence, for example, observation of these features may be possible with current technology.

In our model the ground state $|0\rangle$ is coupled by the vacuum modes to the closely spaced doublet $|1\rangle$, $|2\rangle$. The equations of motion of the reduced density matrix elements for the atomic variables in the frame rotating with the average atomic transition frequency $\omega_0 = (\omega_1 + \omega_2)/2$ take the form [1,10],

$$\begin{aligned}
\dot{\rho}_{jj} &= -\gamma_j \rho_{jj} - \tfrac{1}{2}\gamma_{12}(\rho_{12} + \rho_{21}), \\
\dot{\rho}_{21} &= -\tfrac{1}{2}(\gamma_1 + \gamma_2 + i2\omega_{21})\rho_{21} - \tfrac{1}{2}\gamma_{12}(\rho_{22} + \rho_{11}), \\
\dot{\rho}_{10} &= -\tfrac{1}{2}(\gamma_1 - i\omega_{21})\rho_{10} - \tfrac{1}{2}\gamma_{12}\rho_{20}, \\
\dot{\rho}_{20} &= -\tfrac{1}{2}(\gamma_2 + i\omega_{21})\rho_{20} - \tfrac{1}{2}\gamma_{12}\rho_{10},
\end{aligned} \tag{1}$$

where $\omega_{21} = E_2 - E_1$ is the level splitting between the excited sublevels, and γ_j is the spontaneous decay constant of the excited sublevel $|j\rangle (j = 1, 2)$ to the ground level $|0\rangle$. However, γ_{12} represents the effect of quantum interference resulting from the cross coupling between the transitions $|1\rangle \leftrightarrow |0\rangle$ and $|0\rangle \leftrightarrow |2\rangle$. Its value is very sensitive to the orientations of the atomic dipole polarizations. If they are parallel, then $\gamma_{12} = \gamma_M \equiv \sqrt{\gamma_1\gamma_2}$ and the interference effect is maximal, while if they are perpendicular, then $\gamma_{12} = 0$ and quantum interference disappears.

This atomic system is illuminated by a weak, frequency-tunable probe beam. Linear response theory gives the steady-state probe absorption spectrum to be [12]

$$A(\omega_p) = \Re \int_0^\infty \lim_{t\to\infty} \langle [D(t + \tau), D^\dagger(t)] \rangle e^{i(\omega_p - \omega_0)\tau}\, d\tau, \tag{2}$$

where ω_p is the frequency of the probe field and $D(t) = d_{10}|0\rangle\langle 1| + d_{20}|0\rangle\langle 2|$ is the component of the atomic polarization operator [13] in the direction of the probe field polarization vector $\mathbf{e}_p$, with $d_{j0} = \mathbf{d}_{j0} \cdot \mathbf{e}_p$ being the dipole moment of the atomic transition from $|0\rangle$ to $|j\rangle$. We evaluate the spectrum by utilizing the quantum regression theorem and Eqs. (1) as

$$A(\omega_p) = \Re\left[\frac{(\Gamma/2 - i\gamma_1\omega_-)(\bar{\rho}_{00} - \bar{\rho}_{11}) + (\Gamma/2 - i\gamma_2\omega_+)(\bar{\rho}_{00} - \bar{\rho}_{22}) + i\gamma_{12}(\omega_+\bar{\rho}_{21} + \omega_-\bar{\rho}_{12})}{(\frac{1}{2}\gamma_1 - i\omega_+)(\frac{1}{2}\gamma_2 - i\omega_-) - \frac{1}{4}\gamma_{12}^2}\right], \tag{3}$$

where $\omega_\pm = \omega_p - \omega_0 \pm \frac{1}{2}\omega_{21}$, $\Gamma = (\gamma_1\gamma_2 - \gamma_{12}^2)$ measures the degree of quantum interference, $\bar{\rho}_{jj}$ is the steady-state population of level $|j\rangle$, $(j = 0, 1, 2)$, and

$$\bar{\rho}_{21} = -\frac{\gamma_{12}(\bar{\rho}_{11} + \bar{\rho}_{22})}{\gamma_1 + \gamma_2 + i2\omega_{21}}, \qquad \bar{\rho}_{12} = \bar{\rho}_{21}^*, \tag{4}$$

are the steady-state atomic coherences, induced by the quantum interference effect.

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Expression (3) is our basic result and clearly demonstrates that the absorption spectrum consists of three parts, the first two originating from the direct atomic transitions $|0\rangle \rightarrow |j\rangle$, $(j = 0, 1, 2)$ and proportional to the population difference $\bar{\rho}_{00} - \bar{\rho}_{jj}$ between the states $|0\rangle$ and $|j\rangle$, while the final one stems from quantum interference between the two absorption channels, and is proportional to γ_{12}, $\bar{\rho}_{21}$, and $\bar{\rho}_{12}$. If $\gamma_{12} = 0$, the probe absorption spectrum (3) reduces to the sum of two Lorentzians with linewidths γ_1 and γ_2 located at ω_1 and ω_2, respectively. Without population inversion, it is impossible to amplify the probe field. However, if quantum interference is taken into account, this conclusion is dramatically modified.

First we consider the case $\omega_{21} = 0$; $\gamma_1 = \gamma_2 = \gamma$ and $\gamma_{12} = \gamma_M = \gamma$. Thus $\Gamma = 0$. The steady-state solutions in this situation are surprisingly dependent on the particular initial states of the atom [14], due to quantum interference. We consider the following cases.

(i) If the atom is initially in the antisymmetric state $|a\rangle = (|1\rangle - |2\rangle)/\sqrt{2}$, then the steady-state solutions are $\bar{\rho}_{00} = 0$, $\bar{\rho}_{11} = \bar{\rho}_{22} = 1/2$, and $\bar{\rho}_{21} = -1/2$.

(ii) If the atom is initially in the symmetric state $|b\rangle = (|1\rangle + |2\rangle)/\sqrt{2}$, or in the ground state $|0\rangle$, then $\bar{\rho}_{00} = 1$, $\bar{\rho}_{11} = \bar{\rho}_{22} = 0$, and $\bar{\rho}_{21} = 0$.

(iii) If the atom is initially in one of the excited doublet states $|1\rangle$, $|2\rangle$, then $\bar{\rho}_{00} = 1/2$, $\bar{\rho}_{11} = \bar{\rho}_{22} = 1/4$, and $\bar{\rho}_{21} = -1/4$.

The case (i) is the most interesting, because the absorption is zero for all frequencies of the probe beam, $A(\omega_p) = 0$, and the medium is transparent for the probe field. This effect is attributed to population trapping, and is also an example of population inversion without lasing [2]. For the case (ii), the probe absorption spectrum is a single Lorentzian with linewidth 2γ, and for the case (iii), the probe absorption spectrum is also a Lorentzian with linewidth 2γ, but the maximum value is only half the value of case (ii). This is because half the population is trapped in the state $|a\rangle$ [14,15], which is totally decoupled from the probe field.

We consider now imperfect quantum interference, $\Gamma \neq 0$, when $\bar{\rho}_{00} = 1$ and $\bar{\rho}_{11} = \bar{\rho}_{22} = \bar{\rho}_{21} = 0$, independent of the initial atomic states. Figure 1 shows the probe absorption spectrum for $\omega_{21} = 0$ and different strengths of quantum interference. A very narrow resonance occurs at the atomic frequency ω_0 when the dipole moments are very nearly parallel. See, for example, $\gamma_{12} = 0.9\gamma_M$ in Fig. 1(b) and $\gamma_{12} = 0.99\gamma_M$ in Fig. 1(c). The linewidth is also dependent on the decay constants of the excited doublet. For example, the resonance for $\gamma_2 = 0.1\gamma_1$ (dashed curve) is narrower than the one for $\gamma_2 = \gamma_1$ (solid curve). However, when both moments are perpendicular [frame (a), $\gamma_{12} = 0$] or exactly parallel [frame (d), $\gamma_{12} = \gamma_M$], the narrow resonance does not occur. In the former case there is no interference at all, which results in the absorption spectrum being a sum of two independent Lorentzians with respective linewidths γ_1, γ_2, while the latter is attributed to completely destructive interference, resulting in a single broad Lorentzian with reduced height.

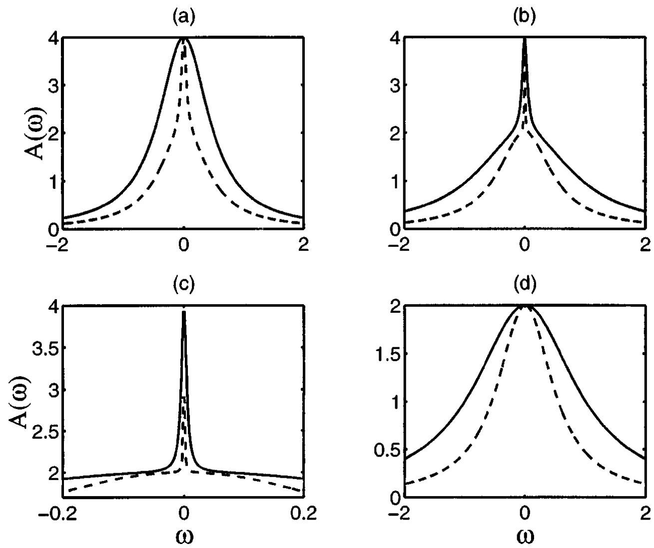

FIG. 1. The dimensionless absorption spectrum of a closed V-type atomic system with the degenerate excited doublet ($\omega_{21} = 0$), as a function of $\omega = \omega_p - \omega_0$, for various γ_{12}: (a) $\gamma_{12} = 0$, (b) $\gamma_{12} = 0.9\gamma_M$, (c) $\gamma_{12} = 0.99\gamma_M$, and (d) $\gamma_{12} = \gamma_M$, where $\gamma_M = (\gamma_1\gamma_2)^{1/2}$ is the maximum value of the interference term. The solid lines correspond to the case of $\gamma_2 = \gamma_1$, while the dashed lines show the case of $\gamma_2 = 0.1\gamma_1$. All quantities are measured in units of γ_1 in all our graphs.

In order to see the effects of quantum interference analytically, we assume γ_{12} to be close to its maximal value $\gamma_M = \sqrt{\gamma_1\gamma_2}$, so that $\Gamma \ll \gamma_s^2$ where $\gamma_s = \gamma_1 + \gamma_2$. We may then approximate the spectrum (3) as

$$A(\omega_p) \simeq \frac{1}{2}\left[\frac{\varepsilon_0^2}{(\frac{1}{2}\varepsilon_0)^2 + \omega^2} + \frac{\gamma_s^2}{\frac{1}{4}\gamma_s^2 + \omega^2}\right], \quad (5)$$

where $\omega = \omega_p - \omega_0$ and $\varepsilon_0 = \Gamma/\gamma_s \ll \gamma_s$. It consists of the superposition of a broad Lorentzian with linewidth $(\gamma_1 + \gamma_2)$ and a narrow Lorentzian with linewidth ε_0: the spectral profile shows a very sharp peak imposed on a broad one. Although it is clear from Fig. 1(a) that it is possible to obtain a narrow spectral line in the absence of quantum interference if one decay rate is much smaller than the other, the narrow resonance reported here is certainly a result of quantum interference. In principle, the resonance becomes arbitrarily narrow as the two dipole moments approach perfect alignment. For example, the widths of the solid and dashed lines in Fig. 1(c) are, respectively, 1% and 0.18% of γ_1. The expression (5) also clearly demonstrates that the quantum interference is destructive. If the dipole moments are exactly parallel, destructive interference is complete: no narrow line occurs, and the spectrum (5) is just a broad Lorentzian with linewidth $(\gamma_1 + \gamma_2)$.

For the case of $\omega_{21} \neq 0$, we first assume $\gamma_1 = \gamma_2 = \gamma$. If $\Gamma = 0$, the spectrum is composed of two Lorentzians centered at the frequency ω_0 with linewidths $\sigma_{\pm} = \gamma^2(1 - 2\varepsilon_1 \pm \sqrt{1 - 4\varepsilon_1})/2$ with $\varepsilon_1 = (\omega_{21}/2\gamma)^2$,

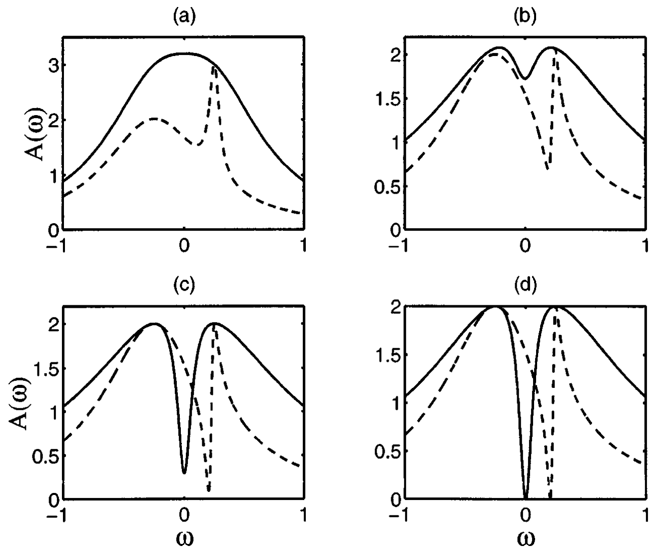

FIG. 2. Same as Fig. 1, but for the nondegenerate excited doublet, with $\omega_{21} = 0.5\gamma_1$.

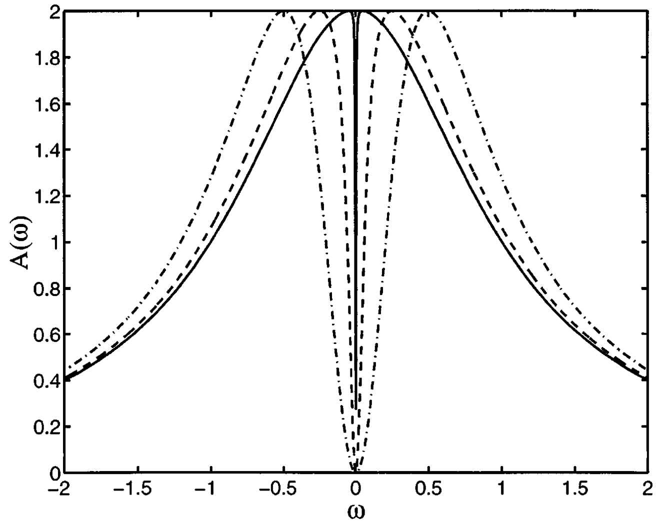

FIG. 3. Same as Fig. 1, but for $\gamma_2 = \gamma_1$, $\gamma_{12} = \gamma_M$, and different splittings: $\omega_{21} = 0.1$ (solid line), $\omega_{21} = 0.5$ (dashed line), and $\omega_{21} = 1$ (dot-dashed line).

which depend on the doublet splitting. Interestingly, one Lorentzian has a negative weight, and at line center, $A(\omega_0) = 0$: transparency occurs. The effect of quantum interference on the absorption spectrum with the splitting $\omega_{21} = 0.5\gamma_1$ is shown in Fig. 2. The spectrum is very broad in the absence of interference [Fig. 2(a)], while if quantum interference is taken into account, there is a hole bored into the broad spectrum. The stronger the interference, the deeper the hole. For maximal quantum interference, transparency occurs at the average atomic transition frequency ω_0. See Fig. 2(d).

The width of the interference-induced hole is also dependent on the doublet splitting ω_{21}. For $\varepsilon_1 \ll \gamma_s$, the absorption spectrum is approximately

$$A(\omega_p) \simeq 2\left[\frac{\gamma^2}{\gamma^2 + \omega^2} - \frac{(\omega_{21}^2/4\gamma)^2}{(\omega_{21}^2/4\gamma)^2 + \omega^2}\right]. \tag{6}$$

The width of the hole represented by the Lorentzian with negative weight can be very narrow for $\omega_{21} \ll \gamma$. We exhibit the dependence of the width of the interference-induced hole on ω_{21} in Fig. 3. For $\omega_{21} = 0.1\gamma_1$, illustrated by the solid line, the hole linewidth is only 0.25% of γ_1.

For the general case of ω_{21}, $\Gamma \neq 0$, $\gamma_1 \neq \gamma_2$, the absorption spectrum, shown by the dashed lines in Fig. 2 for $\gamma_2 = 0.1\gamma_1$, is asymmetric. A strongly dispersive profile occurs around the atomic transition frequency $\omega_2 = \omega_0 + \omega_{21}/2$ for $\Gamma \neq 0$. Figure 2(d), where interference is maximal, also shows probe transparency at frequency $\omega_p - \omega_0 \simeq 0.2\gamma_1$. Generally, transparency only occurs for maximal quantum interference, $\gamma_{12} = \gamma_M$, and then at the frequency

$$\omega_T = \omega_0 + \frac{\omega_{21}(\gamma_1 - \gamma_2)}{2(\gamma_1 + \gamma_2)}. \tag{7}$$

The formula (3) also permits a qualitative insight into gain without population inversion due to quantum interference in this simple system. With $\gamma_1 = \gamma_2 = \gamma$, the value of the probe absorption spectrum at the average atomic transition frequency ω_0 is

$$A(\omega_p = \omega_0) = \frac{2\Gamma(\bar\rho_{00} - \bar\rho_{11}) + 2\Gamma(\bar\rho_{00} - \bar\rho_{22}) - 2\gamma_{12}^2\omega_{21}^2(\bar\rho_{11} + \bar\rho_{22})/(\gamma^2 + \omega_{21}^2)}{\Gamma + \omega_{21}^2}. \tag{8}$$

The first two terms result from the usual absorption transitions $|0\rangle \rightarrow |1\rangle$ and $|0\rangle \rightarrow |2\rangle$, and describe amplification of the probe beam if population inversion could be achieved. Since they are also proportional to Γ, quantum interference reduces the magnitude of the usual contributions. For maximal interference, $\Gamma = 0$, these two terms disappear. However, the last term originates from quantum interference and is always negative—it promotes probe amplification. The interference contribution is nonzero only when there is some population in the doublet and it is nondegenerate. Assuming $\omega_{21} \neq 0$, $\bar\rho_{11} = \bar\rho_{22} \neq 0$, and $\bar\rho_{00} - \bar\rho_{11} > 0$ (no population inversion), we find that $A(\omega_p = \omega_0) < 0$, and we have amplification of the probe beam due to quantum interference, when γ_{12} satisfies

$$\gamma_{12}^2 > \frac{\gamma^2(\gamma^2 + \omega_{21}^2)}{\gamma^2 + \eta\omega_{21}^2}, \quad \text{with } \eta = \frac{\bar\rho_{00}}{\bar\rho_{00} - \bar\rho_{11}}. \tag{9}$$

We plot the absorption spectrum for $\omega_{21} = \gamma$ in Fig. 4, where the stationary populations are phenomenologically taken to be $\bar\rho_{00} = 0.8$, $\bar\rho_{11} = \bar\rho_{22} = 0.1$. The spectral profiles are qualitatively similar to those shown in Fig. 2.

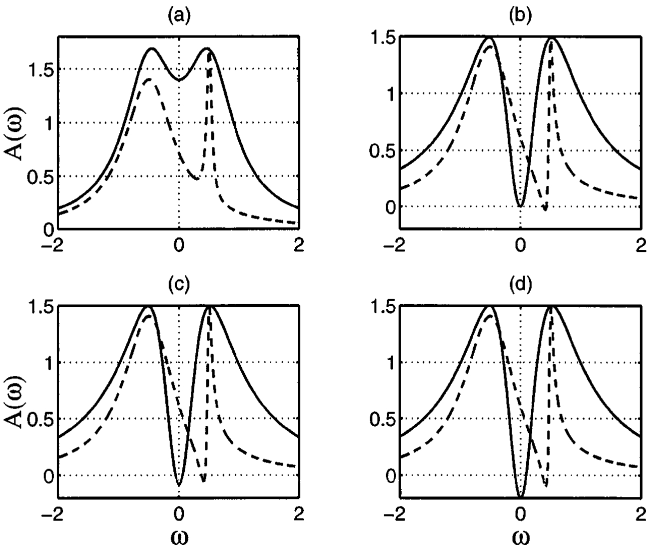

FIG. 4. The dimensionless absorption spectrum for $\omega_{21} = \gamma_1$ with $\bar{\rho}_{11} = \bar{\rho}_{22} = 0.1$, $\gamma_1 = \gamma_2$ (solid lines) and $\gamma_2 = 0.1\gamma_1$ (dashed lines). In (a) $\gamma_{12} = 0$, in (b) $\gamma_{12} = 0.966\gamma_M$, in (c) $\gamma_{12} = 0.98\gamma_M$, and in (d) $\gamma_{12} = \gamma_M$.

However, a significant difference is that gain for the probe beam around the frequency $\omega_p = \omega_T$ occurs when the quantum interference is sufficiently strong. For example, in the case of $\gamma_1 = \gamma_2$, shown by the solid lines in Fig. 4, transparency occurs for $\gamma_{12} = 0.966\gamma_M$ at the probe frequency $\omega_p = \omega_0$ [frame 4(b)], with amplification for $\gamma_{12} > 0.966\gamma_M$, e.g., $\gamma_{12} = 0.98\gamma_M, \gamma_M$ in Figs. 4(c) and 4(d), respectively.

It is worth emphasizing that for the closed V-type system with a nondegenerate excited doublet, described by Eqs. (1), the steady-state solution can only be the ground state: $\bar{\rho}_{00} = 1$, $\bar{\rho}_{11} = \bar{\rho}_{22} = 0$. Thus no gain is possible because the contribution of the quantum interference to the probe amplification is also proportional to the populations of the excited doublet. However, if we extend our model to include a fourth atomic level $|f\rangle$, where the direct transition between $|0\rangle$ and $|f\rangle$ is forbidden, the situation is different. If the additional level is coupled to the excited doublet by a coherent field [3,8,9], the atom may be partially populated to the excited doublet in the steady state. Furthermore, if the separation between $|f\rangle$ and the doublet $\{|1\rangle, |2\rangle\}$ is much greater than that between $|0\rangle$ and the doublet, the effect of the additional level $|f\rangle$ on atomic absorption under the levels $\{|0\rangle, |1\rangle, |2\rangle\}$ to be probed may be omitted, and our Eq. (3) for the absorption spectrum is thus still valid. Gain without population inversion, but due to quantum interference is possible.

It may be possible to observe these effects in systems similar to those in which interference experiments have recently been conducted [5,9].

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