Electromagnetically Induced Transparency with Matched Pulses

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We show that electromagnetically induced transparency in a dense media is not a Beer's law superposition of the single atom response. When an arbitrarily shaped pulse is applied to an ensemble of population-trapped atoms, the atoms will generate a matching pulse shape on the complementary transition and, after a characteristic distance, render themselves transparent.

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The Hamiltonian describing the interaction of two atomic states which are coupled to a common third state by resonant electromagnetic fields is easily diagonalized. Of the three eigenstates, one state, denoted here by $|\psi^0\rangle$, has components of bare states $|1\rangle$ and $|2\rangle$ and not of bare state $|3\rangle$. These components have amplitudes and phases which oppose those of the driving fields, thereby decoupling and trapping population in $|\psi^0\rangle$. Because $|\psi^0\rangle$ has no component of bare state $|3\rangle$, it has a zero eigenvalue even when state $|3\rangle$ is lifetime or collisionally broadened, or is replaced by a continuum [1–3]. Many applications [4] and experiments [5] showing how optically thick transitions may be rendered nearly transparent have been reported.

In this Letter we show that an ensemble of atoms, all of which are in state $|\psi^0\rangle$ and are probed by an electromagnetic field with a time-varying envelope, establish transparency through a strong nonlinear interaction and that the transparency of an optically thick medium is not a Beer's law superposition of the independent atom response. Off resonance Fourier components of a probing pulse which, in the independent atom model, are completely absorbed; instead, after a characteristic propagation distance, experience no further absorption. The essence of this effect is the generation of matching, correctly phased frequency components on the alternate transition (Fig. 1), and therefore of temporally matched envelopes which are decoupled from the atom. In a ladder system, phase conjugate envelopes are generated and decoupled.

Dalton and Knight [6] have suggested the use of critically cross-correlated fields to enhance population trapping. Other nonlinear effects which create transparency by interference have been discussed. These include the interplay of third-harmonic generation and multiphoton ionization, and of sum-frequency generation and resonant absorption [7]. New techniques for the study of the large signal behavior of multiply resonant systems [8] have recently been developed. A theoretical study of the problem of the simultaneous propagation of different wavelength optical pulses has been given by Konopnicki and Eberly [9]. Their work is in the spirit of self-induced transparency where atoms cycle through bare state $|3\rangle$. In the present work, following an initial transient and for exactly matched pulses, bare state $|3\rangle$ is empty.

An energy level diagram for the prototype lambda system is shown in Fig. 1. We assume the ideal case for population trapping: complete metastability of states $|1\rangle$ and $|2\rangle$; lifetime decay of state $|3\rangle$ with a rate Γ_3 to states or continua which are not shown, but not to state $|1\rangle$ or $|2\rangle$; and no inhomogeneous broadening. The inset shows the absorption cross section, as a function of probe frequency, for a single atom in state $|\psi^0\rangle$. Here, only the Fourier component of an applied pulse with zero detuning has zero loss.

We assume applied electromagnetic fields

$$E_p(t) = \mathrm{Re}\{[1+f(t)]E_p \exp[j(\omega_p t + \theta_p)]\}\,,$$
$$E_c(t) = \mathrm{Re}\{[1+g(t)]E_c \exp[j(\omega_c t + \theta_c)]\}\,, \quad (1)$$

where $\omega_p = (\omega_3 - \omega_1)$, $\omega_c = (\omega_3 - \omega_2)$, and define the magnitudes of the Rabi frequencies $\Omega_p \equiv \mu_{13} E_p/\hbar$, $\Omega_c \equiv \mu_{23} E_c/\hbar$, and $\Omega_s^2 = (\Omega_p^2 + \Omega_c^2)$. To allow for ampli-

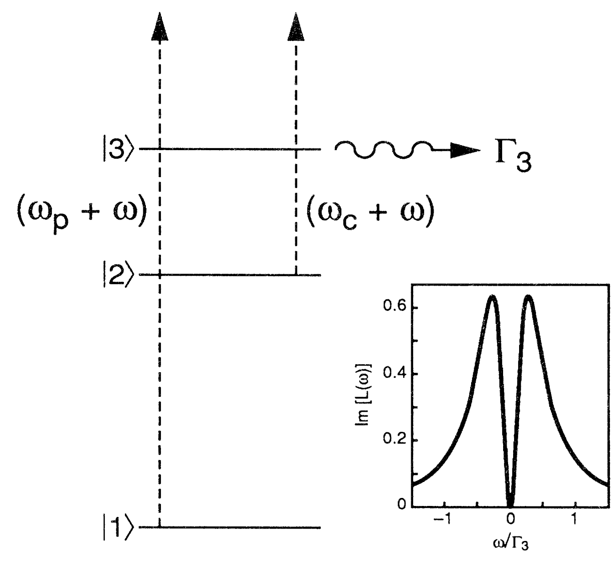

FIG. 1. Energy level diagram for prototype lambda system showing matching frequency components $\omega_p+\omega$ and $\omega_c+\omega$. Inset: The imaginary part of the single atom line shape $L(\omega)$ with $\Omega_s = \Gamma_3/2$.

tude and phase fluctuations, the quantities $f(t)$ and $g(t)$ are complex. E_p, θ_p, E_c, and θ_c are real, positive, and time invariant and the matrix elements μ_{ij} are real. The functions $f(t)$ and $g(t)$ may vary rapidly, subject only to the rotating-wave approximation.

In the basis set of the bare atom $H_0|i\rangle = \hbar\omega_i|i\rangle$, the Hamiltonian with phenomenological damping of bare state $|3\rangle$ and the dipole moment operator P are

$$H = \sum_{i=1}^{3} \hbar\omega_i|i\rangle\langle i| - j\hbar\frac{\Gamma_3}{2}|3\rangle\langle 3| - [\mu_{13}E_p(t)|1\rangle\langle 3| + \mu_{23}E_c(t)|2\rangle\langle 3| + \text{H.c.}]\,,$$
$$P = \mu_{13}|1\rangle\langle 3| + \mu_{23}|2\rangle\langle 3| + \text{H.c.} \tag{2}$$

$E_p(t)$ and $E_c(t)$ interact only with the $|1\rangle$-$|3\rangle$ and $|2\rangle$-$|3\rangle$ transitions, respectively, and counterrotating terms are neglected. We transform from the Schrödinger vector $|\psi\rangle$ to a vector $|a\rangle$ by $|a\rangle = U|\psi\rangle$, with $U = U_2U_1$,

$$U_1 = \{\exp[-j(\omega_p t + \theta_p)]\}|1\rangle\langle 1| + \{\exp[-j(\omega_c t + \theta_c)]\}|2\rangle\langle 2| + |3\rangle\langle 3|\,,$$
$$U_2 = \frac{1}{\sqrt{2}}[\sqrt{2}(\Omega_c/\Omega_s)|1\rangle\langle 1| - \sqrt{2}(\Omega_p/\Omega_s)|1\rangle\langle 2| + (\Omega_p/\Omega_s)|2\rangle\langle 1| + (\Omega_c/\Omega_s)|2\rangle\langle 2| - |2\rangle\langle 3| + (\Omega_p/\Omega_s)|3\rangle\langle 1| + (\Omega_c/\Omega_s)|3\rangle\langle 2| + |3\rangle\langle 3|] \tag{3}$$

and take $\omega_3 = 0$, so that $\omega_1 = -\omega_p$ and $\omega_2 = -\omega_c$. This transformation takes the (population-trapped) Schrödinger state

$$|\psi^0\rangle = \{(\Omega_c/\Omega_s)\exp[j(\omega_p t + \theta_p)]\}|1\rangle + \{-(\Omega_p/\Omega_s)\exp[j(\omega_c t + \theta_c)\}|2\rangle + 0|3\rangle$$

into $|a^0\rangle = U|\psi^0\rangle = |1\rangle$.

With this transformation, Schrödinger's equation is

$$\frac{d|a\rangle}{dt} + M|a\rangle = D(t)|a\rangle\,,$$
$$M = \left(\frac{\Gamma_3}{4} + j\frac{\Omega_s}{2}\right)|2\rangle\langle 2| + \left(\frac{\Gamma_3}{4} - j\frac{\Omega_s}{2}\right)|3\rangle\langle 3| - \frac{\Gamma_3}{4}|2\rangle\langle 3| - \frac{\Gamma_3}{4}|3\rangle\langle 2|\,,$$
$$D(t) = -\frac{j}{2\sqrt{2}\Omega_s}\left\{\sqrt{2}[\Omega_p^2 f_{\text{Re}}(t) + \Omega_c^2 g_{\text{Re}}(t)][|2\rangle\langle 2| - |3\rangle\langle 3|] + \sqrt{2}[\Omega_p^2 f_{\text{Im}}(t) + \Omega_c^2 g_{\text{Im}}(t)][-j|2\rangle\langle 3| + \text{H.c.}]\right\} - \frac{j\Omega_p\Omega_c}{2\sqrt{2}\Omega_s}\{[f(t) - g(t)][|1\rangle\langle 2| - |1\rangle\langle 3|] + \text{H.c.}\}\,. \tag{4}$$

The monochromatic portion of the assumed fields may be eliminated by setting Ω_s in M equal to zero and leaving $D(t)$ unchanged. We observe that, if the pulse envelopes $f(t)$ and $g(t)$ are equal, then state $|a^0\rangle = |1\rangle$ is decoupled from the electromagnetic field.

In general, the pulse envelopes are different and produce a polarization on both the (bare) $|1\rangle$-$|3\rangle$ and $|2\rangle$-$|3\rangle$ transitions. To allow a model calculation, we assume that $f(t) - g(t)$ is zero until after a transient period during which $|a^0\rangle$ is established. During this period a fraction Ω_c^2/Ω_s^2 of the population of the ground state projects against $|\psi^0\rangle$ and is trapped, and a fraction Ω_p^2/Ω_s^2 is depleted from the system. When $\Omega_s \le \Gamma_3$, this transient period lasts for several units of Γ_3/Ω_s^2.

To calculate the polarization, we proceed perturbatively and replace $|a\rangle$ on the right-hand side of Eq. (4) by $|a^0\rangle$ and on the left-hand side by $|a^1\rangle$ and solve with the boundary condition $|a^1(t=0)\rangle = 0$. For the perturbative approach to be valid, $|a^0\rangle$ must not be depleted by virtual or real transitions to the other eigenstates. This imposes a limit on the magnitude (power) and integral over time (energy) of $|f(t) - g(t)|^2$. These limits depend on the spectral content of $f(t)$ and $g(t)$ and are most severe when the Fourier components of both functions are near $\Omega_s/2$; if so, $|f(t) - g(t)|^2$ must be less than $\Gamma_3^2\Omega_s^2/\Omega_c^2\Omega_p^2$, and the integral of $|f(t) - g(t)|^2$ must be less than $\Gamma_3\Omega_s^2/\Omega_c^2\Omega_p^2$. It is also required that $\Omega_p^2|f|^2 + \Omega_c^2 \times |g|^2 \ll \Omega_s^2$.

We proceed in the frequency domain and define $F(\omega)$ and $G(\omega)$ as the Fourier transforms of the envelope quantities $f(t)$ and $g(t)$. To first order of perturbation each Fourier component at the optical frequency $\omega_p + \omega$ produces a dipole moment at its own frequency and at the frequency $\omega_c + \omega$; similarly, a component at $\omega_c + \omega$ couples to itself, to $\omega_p + \omega$, and to no other Fourier components. The polarization $\langle a^0|UPU^{-1}|a^1\rangle + \text{c.c.} = P_p(t) + P_c(t)$ is

$$P_p(t) = -\frac{\pi\mu_{13}\Omega_p\Omega_c^2 N}{\Omega_s^2}\,\mathrm{Re}\left\{\int_{-\infty}^{+\infty} L(\omega)[F(\omega)-G(\omega)]\exp[j(\omega_p+\omega)t+\theta_p]d\omega\right\},$$
$$P_c(t) = -\frac{\pi\mu_{23}\Omega_c\Omega_p^2 N}{\Omega_s^2}\,\mathrm{Re}\left\{\int_{-\infty}^{+\infty} L(\omega)[G(\omega)-F(\omega)]\exp[j(\omega_c+\omega)t+\theta_c]d\omega\right\}, \qquad (5)$$
$$L(\omega) = \frac{2}{\pi}\frac{1}{\Gamma_3}\frac{2\Gamma_3\omega}{4\omega^2-\Omega_s^2-2j\Gamma_3\omega}.$$

The quantity $L(\omega)$ is the normalized line shape, $\int_{-\infty}^{+\infty}\mathrm{Im}L(\omega)d\omega = 1$, and N is the atom density. We may view the replication of the Fourier transform of $f(t)$ onto the dipole moment at ω_c as resulting from the mixing of each Fourier component with the *phased* $|1\rangle$-$|2\rangle$ transition.

Continuing in the frequency domain we use the polarization from Eq. (5) to write slowly varying envelope equations for $F(\omega)$ and $G(\omega)$ as a function of distance [10]. We assume collinear propagation and take $k(\omega_p)$, $k(\omega_c)$, $k(\omega_p+\omega)$, and $k(\omega_c+\omega)$ as the k vectors of the respective frequencies. These k vectors include contributions to the polarization which result from the portion of the $|1\rangle$-$|3\rangle$ and $|2\rangle$-$|3\rangle$ transitions which have not been included in the rotating-wave approximation and from other transitions of the atom. With the definition $\Delta k = [k(\omega_p+\omega)-k(\omega_p)]-[k(\omega_c+\omega)-k(\omega_c)]$, the coupled propagation equations for $F(\omega)$ and $G(\omega)$ are

$$\frac{\partial F(\omega)}{\partial z}+\kappa_{ff}F(\omega)+\kappa_{fg}G(\omega)=0,$$
$$\frac{\partial G(\omega)}{\partial z}+\kappa_{gg}G(\omega)+\kappa_{gf}F(\omega)=0,$$
$$\kappa_{ff} = -j\frac{\pi\omega_p\mu_{13}^2 L(\omega)}{2c\varepsilon_0\hbar}\left(\frac{\Omega_c^2 N}{\Omega_s^2}\right),$$
$$\kappa_{fg} = -\kappa_{ff}\exp(j\Delta kz), \qquad (6)$$
$$\kappa_{gg} = -j\frac{\pi\omega_c\mu_{23}^2 L(\omega)}{2c\varepsilon_0\hbar}\left(\frac{\Omega_p^2 N}{\Omega_s^2}\right),$$
$$\kappa_{gf} = -\kappa_{gg}\exp(-j\Delta kz).$$

Since there is absorption only when ω is on the order of the larger of Ω_s or Γ_3, the dispersion caused by other transitions will usually be negligible. (The dispersive contributions of the $|1\rangle$-$|3\rangle$ and $|2\rangle$-$|3\rangle$ transitions are included within the κ_{ij}.) With $\Delta k = 0$, the solution of Eq. (6) is

$$F(\omega,z) = \frac{F(\omega,z=0)}{\kappa_{ff}+\kappa_{gg}}\{\kappa_{gg}+\kappa_{ff}\exp[-(\kappa_{ff}+\kappa_{gg})z]\}+\frac{G(\omega,z=0)}{\kappa_{ff}+\kappa_{gg}}\{\kappa_{ff}-\kappa_{ff}\exp[-(\kappa_{ff}+\kappa_{gg})z]\},$$
$$G(\omega,z) = \frac{G(\omega,z=0)}{\kappa_{ff}+\kappa_{gg}}\{\kappa_{ff}+\kappa_{gg}\exp[-(\kappa_{ff}+\kappa_{gg})z]\}+\frac{F(\omega,z=0)}{\kappa_{ff}+\kappa_{gg}}\{\kappa_{gg}-\kappa_{gg}\exp[-(\kappa_{ff}+\kappa_{gg})z]\}. \qquad (7)$$

The quantities of κ_{ff} and κ_{gg} are the independent atom propagation constants of the $|1\rangle$-$|3\rangle$ and $|2\rangle$-$|3\rangle$ transitions, respectively. [Noting that $\Omega_p^2+\Omega_c^2=\Omega_s^2$, we recognize the factors $\Omega_c^2 N/\Omega_s^2$ and $\Omega_p^2 N/\Omega_s^2$ in Eq. (6) as the populations of bare states $|1\rangle$ and $|2\rangle$, respectively.]

At a distance where, for an independent atom, a particular spectral component $F(\omega)$ or $G(\omega)$ would be absorbed it is instead approaching its asymptotic value. For any boundary condition the ratio of the asymptotic values of $F(\omega)$ and $G(\omega)$ is

$$\left.\frac{F(\omega,z)}{G(\omega,z)}\right|_{z\to\infty} = 1. \qquad (8)$$

To the extent that all spectral components would be absorbed, $f(t)=g(t)$; and state $|a^0\rangle$ is decoupled. When the normalized fields $f(t)$ and $g(t)$ are equal, the actual fields have a ratio $E_p\exp(j\theta_p)/E_c\exp(j\theta_c)$ and are therefore matched to the monochromatic components.

Figure 2 shows the nonlinear behavior as compared to the independent atom (Beer's law) behavior. Two boundary conditions are shown: $G(\omega,0)=0$ and $G(\omega,0)=F(\omega,0)\angle 135°$. The latter boundary condition is an example of how, irrespective of their initial phase, the electromagnetic fields at frequencies ω_p and ω_c will continue to interact until $F(\omega)=G(\omega)$.

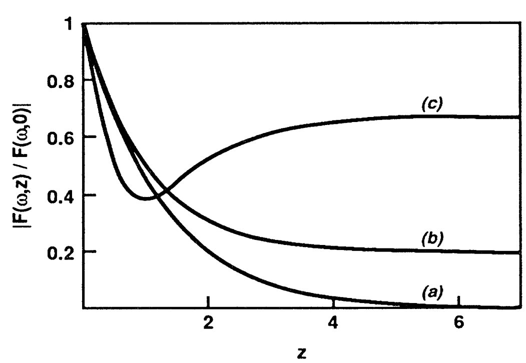

FIG. 2. $|F(\omega,z)/F(\omega,0)|$ vs distance for (a) independent atoms with absorption coefficients $\kappa_{ff}=0.89$ and $\kappa_{gg}=0$; and coupled atoms with $\kappa_{ff}=0.8$ and $\kappa_{gg}=0.2$ and boundary conditions (b) $G(\omega,0)=0$ and (c) $G(\omega,0)=F(\omega,0)\angle 135°$.

We next summarize the results for a ladder system where state $|2\rangle$ is above state $|3\rangle$. In Eq. (4), $g(t)$ becomes $g^*(t)$. In a ladder system the condition for decoupling of state $|a^0\rangle$ is $f(t)=g^*(t)$. In Eq. (6), the variable $G(\omega)$ becomes $G^*(-\omega)$; the quantities κ_{ff} and κ_{fg} remain the same, but κ_{gg} and κ_{gf} change sign. When the absolute value of the real part of κ_{gg} is less than that of κ_{ff}, then at large z, $g^*(t)\rightarrow f(t)$. When the absolute value of the real part of κ_{gg} is greater than that of κ_{ff}, there is exponential gain on both transitions and the matched condition is not attained.

Equation (7) applies when state $|3\rangle$ is replaced by a symmetric *structureless* continuum. The κ_{ij} of Eq. (6), expressed in terms of the photoionization cross sections σ_p and σ_c and the golden rule ionization rates W_p, W_c, and $W_s=W_p+W_c$ of bare states $|1\rangle$ and $|2\rangle$ to the continuum, are

$$\kappa_{ff}=-j(W_p/W_s)\sigma_p NL(\omega),\quad \kappa_{fg}=-\kappa_{ff}\exp(j\Delta kz)\,,$$

$$\kappa_{gg}=-j(W_c/W_s)\sigma_c NL(\omega),\quad \kappa_{gf}=-\kappa_{gg}\exp(-j\Delta kz)\,,$$

where $L(\omega)=j\omega/(2\omega-jW_s)$.

This transparency results from the special property of $|\psi^0\rangle$, which causes what are normally considered as nonlinear terms to be as important as the linear terms. If, at fixed Ω_p, one reduces Ω_c, population is forced to bare state $|2\rangle$, thereby reducing both the linear and nonlinear contributions to the polarization at $f(t)$ in the same ratio. At sufficiently small Ω_c/Ω_s this behavior must fail, because of both the neglect of inhomogeneous broadening and the failure of the rotating-wave approximation. The latter failure occurs because at small Ω_c/Ω_s, and therefore small population of state $|1\rangle$, other transitions contribute to the polarization at $f(t)$.

The question of the preparation of state $|\psi^0\rangle$ has not been adequately addressed. In order to have a reasonably simple mathematical model, we have assumed that the time-varying fields $f(t)$ and $g(t)$ are superimposed onto monochromatic components, as discussed earlier, which prepare $|\psi^0\rangle$. It seems likely that this method of preparation is unnecessarily restrictive; for example, following Carroll and Hioe [11], state $|\psi^0\rangle$ might be produced by allowing the leading edge of a $|2\rangle$-$|3\rangle$ pulse to exceed that of a $|1\rangle$-$|3\rangle$ pulse until after $|\psi^0\rangle$ is formed.

This work substantially modifies our understanding of population trapping in dense media. We have shown that electromagnetically induced transparency in dense media may not be considered as a Beer's law superposition of the single atom response. One application may be in transmission of pulsed radiation through lossy or ionizing media. Here, nonlinear optical techniques could be used to prepare matched pulses at the two transition frequencies. Amplitude and phase distortion of these pulses, as well as distortion produced by the media, will tend to be removed by the phenomena described here.

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