Transparency Induced via Decay Interference

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We demonstrate that decay interference from the two upper levels of a four-level system can lead to loss-free propagation of a single, short laser pulse through an absorbing medium. In contrast to recent investigations of loss-free propagation in three-level media, no second coupling laser pulse is required. [S0031-9007(99)08579-8]

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Dispersion and absorption properties of multilevel media can be modified through the introduction of a second, or coupling, laser pulse. The coherent interaction between the laser fields and the medium can result in phenomena such as electromagnetically induced transparency and matched pulse propagation [1] and the creation and propagation of "adiabatons" [2–4], a general class of solitonlike pulses that occur in adiabatically evolving systems [5]. Other remarkable properties have also been discussed [6–12]. Experimental investigations of adiabatons have been performed by Harris and co-workers [13]. As discussed by Eberly and co-workers [14], many of the above phenomena are related to coherent population trapping and the creation of "dark states" [15]. If the laser fields are applied adiabatically [5], then the medium can, under trapping conditions, establish a dark state leading to transparency in the medium.

In the above mentioned processes it is crucial to have at least two laser pulses as both are used to create the necessary coherence. In this Letter we show that interference between decay channels can make a medium transparent to a single, short laser pulse. In this scheme interference inherent in the decay is exploited so that no coupling laser field is required for transparency. This type of interference was used in the original proposal of Harris [16] for lasing without inversion and has attracted much attention with regard to fluorescence quenching and related phenomena [17–20]. As in the theory of adiabatons our scheme also relies on the adiabatic evolution of a dark state of the system depicted in Fig. 1.

In this Letter we focus on the case of an atomic medium and the decay process we consider is spontaneous emission. It should be emphasized that our analysis can be equally applied to other systems where such interference exists. Examples include spontaneous emission in molecules [21], processes involving autoionizing resonances [22], and semiconductor quantum well systems where the decay occurs via tunneling processes [23,24].

We begin our analysis with the Maxwell-Schrödinger equations of motion for the system of Fig. 1. The wave function of the system is first expanded in terms of the field-free atomic states multiplied by the corresponding space and time-dependent amplitudes $c_m(z,t)$ ($m = 0,1,2$) and $c_{3,\mathbf{k}}(z,t)$ of, respectively, the atomic and vacuum amplitudes. The laser pulse is taken to propagate in the z direction. Then $c_{3,\mathbf{k}}(z,t)$ is formally eliminated and the Weisskopf-Wigner theory of spontaneous emission applied. The result is the following set of equations in the rotating wave and the slowly varying envelope approximations (with $\hbar = 1$):

$$i \frac{\partial \mathbf{c}(\zeta,\tau)}{\partial \tau} = \mathbf{H}(\zeta,\tau)\mathbf{c}(\zeta,\tau), \qquad (1)$$

$$\Omega_1 \frac{\partial f(\zeta,\tau)}{\partial \zeta} = i\alpha_1 c_0(\zeta,\tau)c_1^*(\zeta,\tau) + i\sqrt{\alpha_1\alpha_2}\, c_0(\zeta,\tau)c_2^*(\zeta,\tau), \qquad (2)$$

where Eqs. (1) and (2) are valid in the retarded (local) time frame with $\tau = t - z/c$ and $\zeta = z$. The effective, non-Hermitian Hamiltonian is

$$\mathbf{H}(\zeta,\tau) = \begin{pmatrix} 0 & \Omega_1 f(\zeta,\tau) & \Omega_2 f(\zeta,\tau) \\ \Omega_1 f^*(\zeta,\tau) & \delta_1 - i\gamma_1/2 & -i\gamma_{12}/2 \\ \Omega_2 f^*(\zeta,\tau) & -i\gamma_{21}/2 & \delta_2 - i\gamma_2/2 \end{pmatrix}. \qquad (3)$$

Here $\Omega_m = -\mu_{0m}\mathcal{E}$ ($m = 1,2$) is the Rabi frequency of the $|0\rangle \rightarrow |m\rangle$ transition. The corresponding transition matrix elements, which we take to be real, are $\mu_{0m} = \vec{\mu}_{0m} \cdot \hat{\epsilon} = \mu_{m0}$ with $\hat{\epsilon}$ being the polarization vector of the laser field and $\mathcal{E}$ being the peak electric field

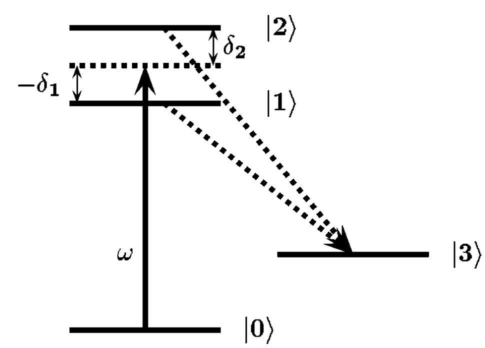

FIG. 1. A schematic diagram of the system under consideration. The ground state $|0\rangle$ is coupled to the excited states $|1\rangle$ and $|2\rangle$ by a laser field. The excited states decay to a common state $|3\rangle$.

strength of the laser field. The laser detuning from resonance with the state $|m\rangle$ is $\delta_m = \omega_{m0} - \omega$ ($m = 1, 2$), where ω_{m0} is the transition frequency between the ground state and the excited state $|m\rangle$. The spontaneous decay rate of state $|m\rangle$ is γ_m, and interference occurs through the term $\gamma_{12} = \gamma_{21} = p\sqrt{\gamma_1\gamma_2}$, where $p = \vec{\mu}_{13} \cdot \vec{\mu}_{32}/|\vec{\mu}_{13}|\,|\vec{\mu}_{32}|$ denotes the alignment of the two spontaneous emission dipole matrix elements. If these matrix elements are parallel, then $p = 1$ and the system exhibits maximum quantum interference, while if $p = 0$, no interference occurs. The pulse envelope is $f(\zeta, \tau)$ and $\alpha_m = 2\pi\mathcal{N}\mu_{0m}^2\omega/c$ ($m = 1, 2$) is the absorption coefficient of the medium having atomic density $\mathcal{N}$. In the derivation of Eq. (2) we assume that $\vec{\mu}_{01} \cdot \vec{\mu}_{20} = \mu_{01}\mu_{20}$, i.e., that these two matrix elements are parallel.

The conditions for trapping in this four-level system have been recently discussed by Zhu and Scully [17] (see also [20]). By searching for a zero root of the characteristic equation of $\mathbf{H}(\zeta, \tau)$ they showed that steady state population trapping can occur if

$$\delta_1\Omega_2^2 + \delta_2\Omega_1^2 = 0, \qquad (4)$$

$$\gamma_2\Omega_1^2 + \gamma_1\Omega_2^2 - 2p\sqrt{\gamma_1\gamma_2}\,\Omega_1\Omega_2 = 0. \qquad (5)$$

Equations (4) and (5) are satisfied when [25]

$$p = 1, \qquad \sqrt{\gamma_2}\,\Omega_1 = \sqrt{\gamma_1}\,\Omega_2, \qquad (6)$$

and the laser frequency is tuned such that

$$\delta_1 = -\frac{\gamma_1\omega_{21}}{(\gamma_1 + \gamma_2)}, \qquad (7)$$

with $\omega_{21} = \delta_2 - \delta_1$. The eigenstate corresponding to the zero eigenvalue of $\mathbf{H}(\zeta, \tau)$ is

$$|\psi_{\rm dark}(\zeta, \tau)\rangle = N(\zeta, \tau)\left[\frac{\sqrt{\gamma_1\gamma_2}\,\omega_{21}}{\gamma_1 + \gamma_2}|0\rangle + \Omega_2 f^*(\zeta, \tau)|1\rangle - \Omega_1 f^*(\zeta, \tau)|2\rangle\right], \qquad (8)$$

where $N(\zeta, \tau)$ is the normalization factor. This eigenstate is adiabatically connected to the ground state. If the laser pulse is applied adiabatically and the population trapping conditions (6) and (7) are fulfilled, then

$$\frac{c_1(\zeta, \tau)}{c_2(\zeta, \tau)} = -\frac{\Omega_2}{\Omega_1}. \qquad (9)$$

Using the general relation $\alpha_1/\alpha_2 = \Omega_1^2/\Omega_2^2$ together with the above result, Eq. (9), in Eq. (2) for the pulse envelope we find that

$$\frac{\partial f(\zeta, \tau)}{\partial \zeta} = 0, \qquad (10)$$

with the consequence that $|f(\zeta, \tau)|^2 = |f(0, \tau)|^2$ for all ζ. If the atomic medium evolves adiabatically in the laser field, the pulse propagates without loss or dispersion.

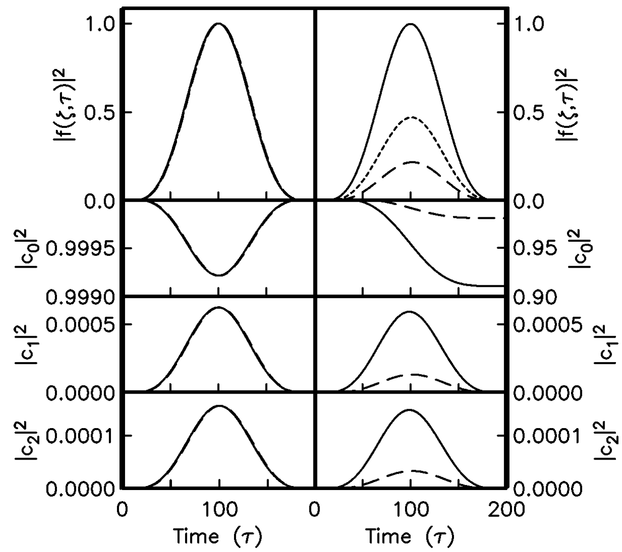

FIG. 2. The magnitude squared of the pulse envelope and the atomic populations as a function of τ for different values of ζ, with $\zeta = 0$ (solid curves), $\zeta = \zeta_{\rm max}/2 = 25$ (dotted curves), and $\zeta = \zeta_{\rm max} = 50$ (broken curves). For clarity the $\zeta = 25$ results have been omitted in the population plots. The left-hand plots are for $p = 1$ while on the right $p = 0$. The initial pulse was taken to be $f(\zeta = 0, \tau) = \sin^2(\pi\tau/\tau_p)$, with τ_p being the pulse duration. The parameters used, in units of γ_1, were as follows: $\tau_p = 200$, $\Omega_1 = 1/5$, $\Omega_2 = 2/5$, $\gamma_2 = 4$, $\alpha_1 = 1$, $\alpha_2 = 4$, $\omega_{21} = 40$, and $\delta_1 = -8$.

We show in Fig. 2 the magnitude squared of the pulse envelope at different positions in the medium as a function of τ, and the corresponding populations of the field-free atomic states. The results were obtained by solving numerically Eqs. (1) and (2) for an adiabatically applied laser pulse. For the results on the left, the trapping conditions given by Eqs. (6) and (7) are fulfilled. In the upper left-hand plot, it can be seen that the propagation of the pulse is both loss- and dispersion-free, as predicted. To demonstrate explicitly the effect of interference caused by the decay of the states $|1\rangle$ and $|2\rangle$, we show on the right in Fig. 2, the evolution of the system when $p = 0$, for the same atomic parameters. Evidently, the pulse is strongly attenuated by the medium and will ultimately be completely absorbed. The transparency of the medium to the laser field does not rely on approximations regarding the laser intensity. A strong

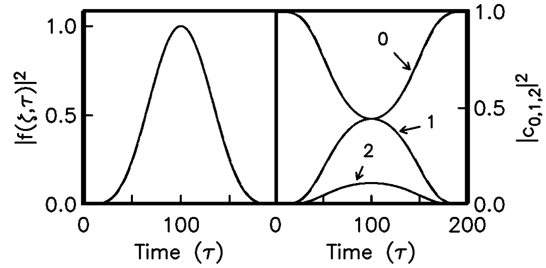

FIG. 3. The same as in Fig. 2 (with $p = 1$), however, with $\Omega_1 = 8$ and $\Omega_2 = 16$.

laser pulse will also propagate intact in our medium as is shown in Fig. 3. Indeed, the adiabaticity requirements are more readily fulfilled under strong excitation.

While seeming restrictive, the condition given by Eq. (6) can be satisfied for a variety of systems. This can be seen as follows: suppose states $|1\rangle$ and $|2\rangle$ are a result of an interaction which dresses two "bare" states. If only one of these two bare states is coupled to the ground state by a dipole allowed transition and the other is metastable, then the off-diagonal damping terms arise naturally and Eq. (6) is automatically satisfied. Such a situation occurs in a number of different contexts. Experimental investigations of such effects include states mixed by spin-orbit interaction (the interference observed in sodium dimers occurs due to this coupling [21]), the application of a dc-electric field (this was demonstrated by Hakuta *et al.* [26] who studied second harmonic generation in atomic hydrogen) and by tunneling transitions in semiconductor quantum wells [23,24]. In addition, Hahn *et al.* [22] showed that Eq. (6) is satisfied to good accuracy when the upper two levels are autoionizing states in zinc.

Having considered the adiabatic evolution of the system, we now investigate the influence of nonadiabatic effects. To obtain an analytic result we assume that the laser-atom interaction is weak, so that $c_0(\zeta,\tau) \approx 1$. From Eq. (1) we derive approximate solutions for $c_m(\zeta,\tau)$ ($m = 1,2$). Keeping terms in the solution up to $\partial f^*/\partial\tau$ we obtain, under population trapping conditions (6) and (7),

$$c_1(\zeta,\tau) = \frac{(\gamma_1+\gamma_2)\Omega_2}{\sqrt{\gamma_1\gamma_2}\,\omega_{21}} f^* - \frac{(\gamma_1+\gamma_2)^4\Omega_2 + 2i(\gamma_1+\gamma_2)^2\Omega_2\gamma_2\omega_{21}}{2\gamma_1^{3/2}\gamma_2^{3/2}\omega_{21}^3}\frac{\partial f^*}{\partial\tau}, \tag{11}$$

$$c_2(\zeta,\tau) = -\frac{(\gamma_1+\gamma_2)\Omega_2}{\gamma_2\omega_{21}} f^* + \frac{(\gamma_1+\gamma_2)^4\Omega_2 - 2i(\gamma_1+\gamma_2)^2\Omega_2\gamma_1\omega_{21}}{2\gamma_1\gamma_2^2\omega_{21}^3}\frac{\partial f^*}{\partial\tau}. \tag{12}$$

We substitute Eqs. (11) and (12) into Eq. (2) with the result

$$\frac{\partial f(\zeta,\tau)}{\partial\zeta} + \frac{1}{v}\frac{\partial f(\zeta,\tau)}{\partial\tau} = 0, \tag{13}$$

where $v = (\gamma_1^2\gamma_2\omega_{21}^2)/[\alpha_1(\gamma_1+\gamma_2)^3]$ is the group velocity of the laser pulse in the retarded frame. This means that as a first correction, nonadiabatic effects lead only to a reduction in the group velocity. This is illustrated in the plots on the left in Fig. 4, obtained by numerical integration of Eqs. (1) and (2). The pulse propagates without absorption but with a group velocity in the retarded frame that is in agreement with our analytic result.

As nonadiabatic effects become stronger, absorption of the pulse also occurs. This can be seen in the right-hand plots in Fig. 4. Here the population of the ground state does not return to unity at the end of the pulse. The pulse profile in the upper right-hand plot now exhibits absorption but still propagates for many absorption lengths with a substantial change in its group velocity and width. When $p = 0$ the pulse exhibits almost instantaneous attenuation, with the maximum value of the magnitude squared of the pulse envelope decaying by 1 order of magnitude at $\zeta = 2$. As the pulse propagates further into the medium, the rate at which the pulse is absorbed decreases and the laser-atom interaction becomes more adiabatic. Ultimately, adiabatic evolution is established and the pulse propagates intact with a reduced group velocity, as in the results on the left. This has been verified by numerical calculations.

Insight into the nonadiabatic evolution of the system can be obtained by considering the extreme case of the propagation of a step pulse in the medium. It is now possible to obtain the long time solutions of Eq. (1) under the population trapping conditions given by Eqs. (6) and (7). For $\zeta = 0$ and using Eq. (8) we have

$$|c_0(\zeta=0,\tau\to\infty)|^2 = \frac{1}{N^4}\frac{\omega_{21}^4\gamma_1^2\gamma_2^2}{(\gamma_1+\gamma_2)^4}, \tag{14}$$

$$|c_1(\zeta=0,\tau\to\infty)|^2 = \frac{1}{N^4}\frac{\omega_{21}^2\gamma_1\gamma_2\Omega_2^2}{(\gamma_1+\gamma_2)^2}, \tag{15}$$

$$|c_2(\zeta=0,\tau\to\infty)|^2 = \frac{1}{N^4}\frac{\omega_{21}^2\gamma_1^2\Omega_2^2}{(\gamma_1+\gamma_2)^2}, \tag{16}$$

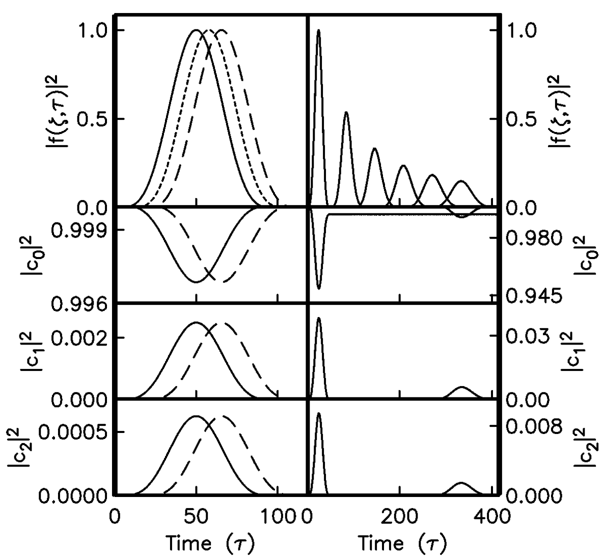

FIG. 4. The left-hand plots are the same as in Fig. 2, however, with $p = 1$, $\omega_{21} = 20$, $\delta_1 = -4$, $\tau_p = 100$, and $\zeta_{\max} = 200$. On the right-hand side $p = 1$, $\omega_{21} = 5$, $\delta_1 = -1$, $\tau_p = 50$, and $\zeta_{\max} = 250$. The propagating pulse is shown in steps of $\zeta_{\max}/5$. The amplitudes are shown for $\zeta = 0$ and $\zeta = \zeta_{\max}$.

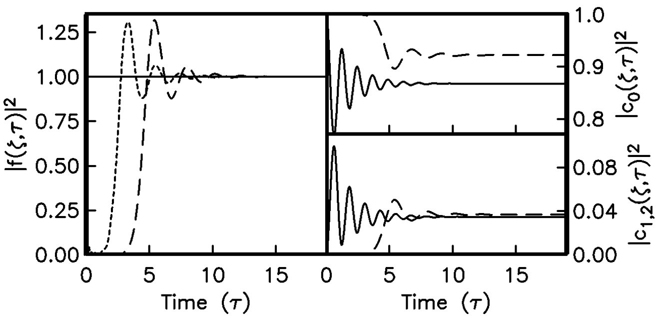

FIG. 5. The same as in Fig. 2 (with $p = 1$), however, now with a step laser pulse. The parameters used, in units of γ_1, were as follows: $\Omega_1 = 1$, $\Omega_2 = 1$, $\gamma_2 = 1$, $\alpha_1 = 1$, $\alpha_2 = 1$, $\omega_{21} = 10$, and $\delta_1 = -5$. The populations in two upper levels are the same.

where

$$N = \left[\frac{\gamma_1 \gamma_2 \omega_{21}^2}{(\gamma_1 + \gamma_2)^2} + \Omega_2^2 \left(1 + \frac{\gamma_1}{\gamma_2} \right) \right]^{1/2} . \tag{17}$$

Therefore after a certain transient period, which the system requires to establish the dark state, the atomic populations at the front of the medium will stabilize to the values given by Eqs. (14)–(16). Inside the medium, a similar transient period is evident, in which the atomic populations oscillate and the pulse experiences absorption and dispersion until the dark state is established. Equation (9) is then satisfied and the medium becomes transparent to the pulse. This is illustrated in Fig. 5 where the propagation of a step laser pulse and the atomic populations are shown at different positions in the medium.

In this Letter we have shown that interference arising from decay processes can make a four-level medium transparent to a short laser pulse. This type of transparency occurs under population trapping conditions in this system and no additional laser field is required. This latter fact distinguishes this work from recent propagation studies in multilevel atoms [1–4,6–14]. Apart from a fundamental interest in pulse-preserving propagation in dielectric media, an interesting aspect in our case is that the required coherence is maintained by the presence of dissipation.

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