# Pulse propagation in a coherently prepared multilevel medium

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We present an analytical solution of the coupled Maxwell-Bloch equations for the case of a laser-driven and coherently prepared medium with one upper state and $N$ lower states. We show that under specific conditions the medium can become either completely opaque or completely transparent to the laser pulses. We also study the potential for efficient parametric generation of $N-m$ laser pulses and show that this is possible by injecting the system with $m$ input pulses.

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The interaction of laser pulses with a coherently prepared three-level system, the so-called "phaseonium," has led to many interesting phenomena. Examples include the enhancement of the index of refraction [1], electromagnetically induced transparency [2,3], the creation of matched pulses in optically thick media [4–9], and the potential for resonantly enhanced nonlinear pulse generation [10–13]. Experimental studies have also been carried out [14–21], demonstrating the potential for applications of these phenomena. Similar phenomena also occur in coherently prepared four-level systems of various configurations [22–25].

In this Brief Report we analyze the propagation dynamics of $N$ laser pulses that interact near resonantly with the $(N+1)$-level quantum system shown in Fig. 1. The laser pulses couple the $N$ lower levels to a single excited level. Our scheme generalizes recently studied three- [7] and four-level [25] models. Here, we obtain an analytical solution of the coupled Maxwell-Bloch equations assuming an overdamped medium under weak excitation. Using this solution we show that the system can become completely opaque or completely transparent to the laser pulses depending on the initial populations and coherences. In addition we show that, by simply applying a single laser pulse, the medium can generate $N-1$ new laser pulses.

We denote the excited state by $|0\rangle$ and the lower states by $|1\rangle, |2\rangle, \ldots, |N\rangle$ and assume that each laser pulse addresses only one transition. The Hamiltonian of this system in the interaction picture and in the rotating wave and dipole approximations is given by (we use units such that $\hbar=1$)

$$\hat{H}=\sum_{n=1}^{N} \Omega_n(z,t)e^{-i(\delta_n t+k_n z)}|n\rangle\langle 0|+\text{H.c.} \qquad (1)$$

Here, $\Omega_n(z,t)=-\boldsymbol{\mu}_{n0}\cdot\boldsymbol{\varepsilon}_n\mathcal{E}_n f_n(z,t)$ is the Rabi frequency of the transition $|n\rangle\leftrightarrow|0\rangle$, with $\boldsymbol{\mu}_{n0}$ being the associated dipole transition matrix element. Also, $\delta_n=\omega_0-\omega_n-\overline{\omega}_n$ is the laser field detuning from resonance with the transition $|0\rangle\leftrightarrow|n\rangle$, with the energies of the $n$th lower level and upper level being $\omega_n$ and $\omega_0$, respectively. The laser field is described classically as a time- and spatially dependent electric field,

$$\boldsymbol{E}(z,t)=\sum_{n=1}^{N} \boldsymbol{\varepsilon}_n[\mathcal{E}_n f_n(z,t)e^{i(\overline{\omega}_n t-k_n z)}+\text{c.c.}], \qquad (2)$$

where $\overline{\omega}_n$ is the angular frequency, $k_n$ the wave number, $\boldsymbol{\varepsilon}_n$ the polarization vector, $\mathcal{E}_n$ the electric field amplitude, and $f_n(z,t)$ the dimensionless pulse envelope of each laser pulse.

We will analyze the system using a density matrix approach. In the local (retarded) frame where $\tau=t-z/c$, $\zeta=z$ the equations for the density matrix elements read

$$i\frac{\partial}{\partial\tau}\rho_{00}(\zeta,\tau)=\sum_{n=1}^{N}[-i\Gamma_{0n}\rho_{00}(\zeta,\tau)+\Omega_n^*\rho_{n0}(\zeta,\tau)-\Omega_n\rho_{0n}(\zeta,\tau)], \qquad (3)$$

$$i\frac{\partial}{\partial\tau}\rho_{nn}(\zeta,\tau)=-i\sum_{m}\Gamma_{nm}\rho_{nn}(\zeta,\tau)+i\sum_{k}\Gamma_{kn}\rho_{kk}(\zeta,\tau)+\Omega_n\rho_{0n}(\zeta,\tau)-\Omega_n^*\rho_{n0}(\zeta,\tau),\quad n=1,\ldots,N, \qquad (4)$$

$$i\frac{\partial}{\partial\tau}\rho_{n0}(\zeta,\tau)=-(\delta_n+i\gamma_{n0})\rho_{n0}(\zeta,\tau)+\Omega_n\rho_{00}(\zeta,\tau)-\sum_{m=1}^{N}\Omega_m\rho_{nm}(\zeta,\tau),\quad n=1,\ldots,N, \qquad (5)$$

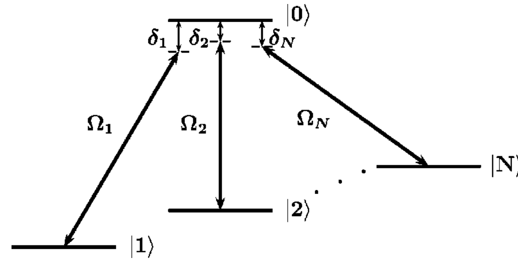


FIG. 1. Schematic diagram of the system considered. An upper level is coupled to $N$ lower levels by $N$ near-resonant laser fields.

 

$$i\frac{\partial}{\partial\tau}\rho_{nm}(\zeta,\tau)=(\delta_m-\delta_n-i\gamma_{nm})\rho_{nm}(\zeta,\tau)+\Omega_n\rho_{0m}(\zeta,\tau)-\Omega_m^*\rho_{n0}(\zeta,\tau),\quad n\neq m, n,m=1,\ldots,N, \tag{6}$$

with $\Sigma_{n=0}^{N}\rho_{nn}(\zeta,\tau)=1$ and $\rho_{nm}(\zeta,\tau)=\rho_{mn}^*(\zeta,\tau)$. We have assumed a closed system, i.e., there is no decay to levels outside of $(N+1)$-level manifold. We denote by $\Gamma_{nm}$ the radiative decay rate of the populations from level $|n\rangle$ to level $|m\rangle$ and by $\gamma_{nm}$ the coherence decay rate between states $|n\rangle$ and $|m\rangle$, with

$$\gamma_{nm}=\frac{1}{2}\sum_{k=1}^{N}\Gamma_{nk}+\frac{1}{2}\sum_{l=1}^{N}\Gamma_{ml}+\gamma'_{nm}, \tag{7}$$

where indices $k,l$ correspond to the states $|k\rangle$ and $|l\rangle$ to which the states $|n\rangle$ and $|m\rangle$, respectively, decay. Also, $\gamma'_{nm}$ describes the decay due to dephasing processes. Examples of dephasing processes include collisions in atomic and molecular systems or electron-electron scattering, interface roughness, and phonon scattering in semiconductor quantum well systems. We will not consider the effects of Doppler broadening. In order to study the propagation of laser pulses in this medium, the Maxwell wave equation is required, which in the slowly varying envelope and phase approximation reads

$$\frac{\partial}{\partial\zeta}\Omega_n(\zeta,\tau)=ia_n\rho_{n0}(\zeta,\tau),\quad n=1,\ldots,N. \tag{8}$$

Here, $a_n=2\pi\mathcal{N}|\boldsymbol{\mu}_{n0}|^2\overline{\omega}_n/c$ is the propagation constant for the transition $|n\rangle\leftrightarrow|0\rangle$ with $\mathcal{N}$ being the system's density.

We take the initial state of the system to be a coherent superposition of all of the lower levels,

$$|\psi(\zeta,0)\rangle=\sum_{n=1}^{N}b_n|n\rangle. \tag{9}$$

The amplitudes $b_n$ are complex in general. Such a coherent superposition can be created with the use of stimulated Raman adiabatic passage [26]. Next, we assume that the laser fields are at multiphoton resonance, i.e., they are equally detuned from resonance with the upper state $\delta_n=\delta$, with $n=1,\ldots,N$. We also assume that the coherence decay rates between the lower levels are negligibly small, i.e., $\gamma_{nm}\approx 0$, with $n\neq m$ and $n,m=1,\ldots,N$. This condition implies that the radiative decay rates $\Gamma_{nm}$ between the lower levels are essentially zero, which is quite plausible. The constants $\gamma'_{nm}$ describing the dephasing contributions to broadening could be kept small by control of the experimental conditions. If our medium is heavily damped and the laser-matter interaction is weak then the populations and coherences in the lower states manifold can be considered essentially unchanged and the following approximate expressions for the density matrix elements $\rho_{n0}(\zeta,\tau)$ can be obtained using Eqs. (3)–(6) and Eq. (9):

$$\rho_{n0}(\zeta,\tau)\approx-\frac{b_n}{\delta+i\gamma_{n0}}\sum_{m=1}^{N}b_m^*\Omega_m(\zeta,\tau),\quad n=1,\ldots,N. \tag{10}$$

As has been shown in previous studies for three- [7] and four-level systems [25], this approximation is very efficient and leads to an accurate description of the system.

The propagation equation (8) for the laser fields then reduce to

$$\frac{\partial}{\partial\zeta}\boldsymbol{\Omega}(\zeta,\tau)=-i\boldsymbol{K}\boldsymbol{\Omega}(\zeta,\tau), \tag{11}$$

with the general solution

$$\boldsymbol{\Omega}(\zeta,\tau)=e^{-i\boldsymbol{K}\zeta}\boldsymbol{\Omega}(0,\tau). \tag{12}$$

The matrix elements of the $N\times N$ propagation matrix $\boldsymbol{K}$ are given by $K_{nm}=\alpha_n b_n b_m^*$ with $n,m=1,\ldots,N$ and $\alpha_n=a_n/(\delta+i\gamma_{n0})$. The vector of the Rabi frequencies is given by $\boldsymbol{\Omega}(\zeta,\tau)=(\Omega_1(\zeta,\tau),\Omega_2(\zeta,\tau),\ldots,\Omega_N(\zeta,\tau))^T$. We note that $\boldsymbol{K}^l=\overline{\alpha}^{l-1}\boldsymbol{K}$, where $\overline{\alpha}=\Sigma_{n=1}^{N}\alpha_n|b_n|^2$. Therefore, from Eq. (12) we obtain

$$\begin{aligned}\boldsymbol{\Omega}(\zeta,\tau)&=\boldsymbol{\Omega}(0,\tau)+\sum_{l=1}^{\infty}\frac{(-1)^l(i\boldsymbol{K}\zeta)^l}{l!}\boldsymbol{\Omega}(0,\tau)\\&=\boldsymbol{\Omega}(0,\tau)+\frac{1}{\overline{\alpha}}\sum_{l=1}^{\infty}\frac{(-1)^l(i\overline{\alpha}\zeta)^l}{l!}\boldsymbol{K}\boldsymbol{\Omega}(0,\tau)\\&=\boldsymbol{\Omega}(0,\tau)+\frac{e^{-i\overline{\alpha}\zeta}-1}{\overline{\alpha}}\boldsymbol{K}\boldsymbol{\Omega}(0,\tau).\end{aligned} \tag{13}$$

We note that Eqs. (12) and (13) are generalizations of Eqs. (5) and (6) of Ref. [7]. The solution, Eq. (13), can be arranged in a more transparent form by introducing the complex parameter $q$,

$$q=\frac{\boldsymbol{b}^{\dagger}\boldsymbol{\Omega}(0,\tau)}{\Omega(0,\tau)}, \tag{14}$$

where

$$\Omega(0,\tau)=[\Sigma_{n=1}^{N}|\Omega_n(0,\tau)|^2]^{1/2},$$

and

$$\boldsymbol{b}=(b_1,b_2,\ldots,b_N)^T.$$

This can only be done if the initial state of the system is a pure state, as we have assumed in Eq. (9). Note that $0\leqslant|q|\leqslant 1$; therefore, $q$ can be seen as the complex analog of $\cos\theta$ in a scalar product between two real vectors $\boldsymbol{x}\cdot\boldsymbol{y}=xy\cos\theta$. In terms of $q$ the solution Eq. (13) reads

$$\mathbf{\Omega}(\zeta,\tau)=\mathbf{\Omega}(0,\tau)+\frac{e^{-i\bar{\alpha}\zeta}-1}{\bar{\alpha}}q\,\boldsymbol{\alpha}\boldsymbol{b}\Omega(0,\tau), \tag{15}$$

where $\boldsymbol{\alpha}$ is a diagonal matrix with elements $\alpha_n$, $n=1,\ldots,N$. Equation (15) presents a simple geometric interpretation of the solution: the vector $\mathbf{\Omega}(0,\tau)$ is projected on the initial state vector $\boldsymbol{b}$ of the medium. The overlap is characterized by the complex parameter $q$. Then, the vector $\boldsymbol{\alpha b}$ is rescaled by $q$ and by another $\zeta$-dependent factor and added to the input vector $\mathbf{\Omega}(0,\tau)$. We note that the pulses reach a steady state value at a rate that is determined by the $N$-photon Beer's length

$$\frac{1}{\bar{\zeta}}=-2\,\mathrm{Im}(\bar{\alpha})=2\sum_{n=1}^{N}\frac{a_n|b_n|^2\gamma_{n0}}{\delta^2+\gamma_{n0}^2}. \tag{16}$$

We can give a clear physical picture of how the parameter $q$ affects the propagation of the pulses in the medium. The coherence $\rho_{n0}(\zeta,\tau)$ in Eq. (10) can be expressed using the general solution Eq. (15):

$$\rho_{n0}(\zeta,\tau)\approx-\frac{b_n}{\delta+i\gamma_{n0}}q\Omega(0,\tau)e^{-i\bar{\alpha}\zeta}. \tag{17}$$

From this form it can be seen that $\rho_{n0}(\zeta,\tau)$ is proportional to $q$. However, this coherence appears as a source term in the Maxwell equation (8).

Two interesting cases are readily obtained from Eq. (15). First, if the vector $\boldsymbol{\alpha b}$ has equal components, $\alpha_n b_n=\alpha_m b_m$ with $n,m=1,\ldots,N$, and so does the vector $\mathbf{\Omega}(0,\tau)$, $\Omega_n(0,\tau)=\Omega_m(0,\tau)$ with $n,m=1,\ldots,N$, we obtain $\Omega_n(\zeta\rightarrow\infty,\tau)=0$, i.e., all pulses are completely absorbed by the medium. However, if $q=0$, i.e., the scalar product $\boldsymbol{b}^\dagger\mathbf{\Omega}(0,\tau)$ is equal to zero, then $\mathbf{\Omega}(\zeta,\tau)=\mathbf{\Omega}(0,\tau)$, so the pulses do not interact with the medium and propagate intact as in free space. In this case the initial state Eq. (9) of the material is a dark state with respect to these pulses [27]. Therefore, with appropriate control of the initial state the medium can become completely opaque or transparent to the laser pulses.

Let us now discuss the possibility of parametric generation in our model. Let the first $m$ pulses be nonvanishing at the input $\Omega_k(0,\tau)=\Omega_k(\tau)$, $k=1,\ldots,m$, while the rest is identically zero, $\Omega_l(0,\tau)=0$, $l=m+1,\ldots,N$. Then, Eq. (15) reduces to

$$\Omega_k(\zeta,\tau)=\Omega_k(\tau)+\frac{e^{-i\bar{\alpha}\zeta}-1}{\bar{\alpha}}q\,\alpha_k b_k\,\Omega(0,\tau),$$
$$k=1,\ldots,m, \tag{18}$$

$$\Omega_l(\zeta,\tau)=\frac{e^{-i\bar{\alpha}\zeta}-1}{\bar{\alpha}}q\,\alpha_l b_l\,\Omega(0,\tau),$$
$$l=m+1,\ldots,N. \tag{19}$$

Therefore, via a coherent internal generation process, a medium coherently prepared in all of the lower levels will generate $N-m$ additional fields from the applied $m$ fields. This will occur only if the coherences between the lower levels of the system are significantly large. In particular, using the definition of the parameter $q$ in the right hand side of Eq. (19) it can be seen that the Rabi frequency of a pulse generated on a specific transition is proportional to the weighted sum of the corresponding initial coherences. In the case of a pure initial state such as the one described by Eq. (9) the coherences are maximal. Note that the parametric generation can be blocked if $q=0$. The parametric generation process works even for $m=1$, i.e., when a single input field is applied. Then the system generates $N-1$ additional output fields. The behavior of the generated $N-m$ pulses is reminiscent of the matched pulse formation of Ref. [4].

An important special case of the above equations is obtained if we take a unique superposition for the lower levels with $b_k=\exp(i\varphi_k)/\sqrt{N}$, $k=1,\ldots,N$. In this case, the general solution in Eq. (15) becomes

$$\mathbf{\Omega}(\zeta,\tau)=\mathbf{\Omega}(0,\tau)+\frac{e^{-i\bar{\alpha}\zeta}-1}{\bar{\alpha}}\tilde{\Omega}(0,\tau)\boldsymbol{\beta},$$

$$\tilde{\Omega}(0,\tau)=\frac{1}{N}\sum_{n=1}^{N}e^{-i\varphi_n}\Omega_n(0,\tau), \tag{20}$$

$$\boldsymbol{\beta}=(\alpha_1 e^{i\varphi_1},\ldots,\alpha_N e^{i\varphi_N})^T.$$

Here, the phase weighted average $\tilde{\Omega}(0,\tau)$ of the incoming pulses plays a similar role as does $q$ in the previous formulas. In the case of parametric generation, the relative amplitudes of the generated fields are determined by the parameters $\alpha_l$, with $l=m+1,\ldots,N$.

In summary, we have studied the propagation dynamics of $N$ laser pulses interacting with an $(N+1)$-level quantum system whereby $N$ lower states are coupled through a single excited state. We have assumed that the system is initially prepared in a coherent superposition of the $N$ lower levels and have derived a general analytic solution for the laser pulses that holds for an overdamped system under weak excitation. This solution has been used for the determination of conditions of complete absorption or transparency of the laser pulses. We have also addressed the problem of parametric generation in this medium and have shown that $N-m$ new laser pulses can be obtained by the injection of $m$ input pulses. The parametric generation process works even if there is only a single input laser field.

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