PHYSICAL REVIEW A VOLUME 52, NUMBER 1 JULY 1995

Optimal control of optical pulse propagation in a medium of three-level systems

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(Received 25 April 1995)

We develop an optimal control technique (OCT) in order to design a secondary optical pulse that can control the propagation of an arbitrary primary optical pulse in a medium consisting of three-level systems. The output primary pulse shape can be manipulated by designing the shape of the secondary pulse. The OCT is shown to be better than the earlier proposed pulse schemes for protecting the magnitude and shape of the primary pulse from reshaping by the medium.

PACS number(s): 42.65.Re, 32.80.Qk, 42.50.Gy, 42.25.Bs

The cooperation of two optical pulses propagating in a medium leads to a variety of quantum coherent phenomena such as simulton propagation [1], Raman solitons [2], and lasing without inversion [3]. Recently, interest has been expressed in electromagnetically induced transparency (EIT) [4], where the absorption of a primary pulse can be greatly reduced by applying a strong controlling pulse. It was shown that a superposition of the two pulses, termed as the dressed-field pulse, was unchanged during the propagation [5]. Hioe and Grobe gave general analytic solutions for a pair of solitary waves that can propagate through three- and five-level systems with their shapes invariant [6].

The matched pulse scheme (MPS) proposed by Harris [4] calls for the primary and the controlling pulses to have the same shapes. Though the absorption of the primary pulse is greatly reduced, it still suffers a front edge loss, and the shape is distorted especially when the intensity of the controlling pulse is limited. The solitary wave pairs of Hioe and Grobe vary widely in shape, but they do not cover an arbitrary pulse shape. Furthermore, the medium is required to be in particular initial states. A natural question is, given an arbitrary incident primary pulse and initial states of the sample (for example, all atoms being in their ground states), how may one design a controlling pulse so that the primary pulse suffers the least absorption and shape distortion during the propagation? We will develop an optimal control technique (OCT) to solve this problem and compare the results with the MPS.

Recently, there has been considerable interest in the control of atomic or molecular dynamics by appropriately designed laser pulses for various purposes, such as population inversion [7–9], population transfer [10,11], and selective [12] and efficient [13,14] dissociation of molecules. In all these works, the absorption and reshaping of the controlling pulse by the atomic and molecular system are neglected. This is valid for controlling the dynamics of a medium with low atomic or molecular density. In many applications such as optically driven unimolecular reactions in condensed media or the gas phase with sufficient high density, the absorption and reshaping of the controlling pulse cannot be neglected. This reshaping of the field presents a major difficulty for existing OCT codes for controlling field design. In this Rapid Communication, we extend the OCT to systematically account for pulse absorption and reshaping by incorporating the Maxwell equations, which extends the design capabilities of the OCT to optically dense media.

Recent experiments with the EIT have been performed in an atomic gas [15] where the atoms can be modeled as a three-level Λ system, shown in Fig. 1 [4,5,15]. In general, a more complex multilevel system can arise, and here we demonstrate the nature of control with a three-level Λ system. In the slowly varying amplitude approximation and the rotating-wave approximation, the Schrödinger-Maxwell equations that describe the propagation of two pulses are

$$\dot{a}_1=\frac{\Omega_p}{2}a_3, \tag{1a}$$

$$\dot{a}_2=\frac{\Omega_c}{2}a_3, \tag{1b}$$

$$\dot{a}_3=-\frac{\Gamma_3}{2}a_3-\frac{\Omega_p}{2}a_1-\frac{\Omega_c}{2}a_2, \tag{1c}$$

$$\frac{\partial\Omega_p}{\partial z'}=\frac{\Gamma_3}{2}a_3a_1, \tag{1d}$$

$$\frac{\partial\Omega_c}{\partial z'}=\frac{k_c\mu_c^2}{2k_p\mu_p^2}\Gamma_3a_3a_2. \tag{1e}$$

Here a_1, a_2, and $-ia_3$ are the probability amplitudes of the levels $|1\rangle$, $|2\rangle$, and $|3\rangle$, and $\Omega_p=2\mu_pE_p/\hbar$ and $\Omega_c=2\mu_cE_c/\hbar$ are the Rabi frequencies of the primary and controlling pulses. $k_{p,c}$ are wave vectors of the primary and the controlling pulses, and $\mu_{p,c}$ are the transition dipole moments (assumed to be real) between levels $|1\rangle$,$|3\rangle$, and

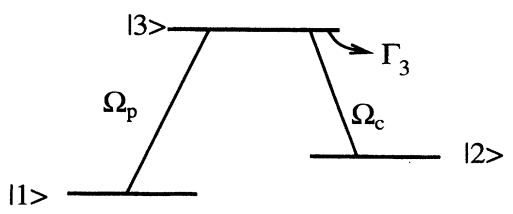

FIG. 1. A sketch of the atomic energy levels, connected by resonant primary (Ω_p) and controlling (Ω_c) pulses.

$|2\rangle,|3\rangle$. Level $|3\rangle$ decays to other states (not $|1\rangle$ and $|2\rangle$) with rate Γ_3. Note that we have used the moving frame, where the dot denotes the derivative with respect to $\tau=t-z/c$, with z,t being the spatial and time coordinates, and c being the velocity of light. z' is the pulse penetration distance measured in units of the primary pulse absorption length; i.e., $z'=\alpha z$, with $\alpha=8\pi k_p N\mu_p^2/\Gamma_3$ being the absorption coefficient of the primary pulse. Here N is the atomic density.

We have assumed that both pulses have no phase modulation and thus a_i, $\Omega_{p,c}$ are real. Phase modulation can be included by allowing a_i, $\Omega_{p,c}$ to be complex in Eq. (1). The extension of the OCT to include phase modulation is straightforward.

For a given incident primary pulse $\Omega_p(z'=0,\tau)$, we want to design the incident controlling pulse $\Omega_c(z'=0,\tau)$ so that the output primary pulse has the desired shape $\Omega_r(\tau)$ (reference pulse). For this purpose, we construct the following objective functional:

$$J=\frac{1}{2}\int_0^T d\tau\, w(\tau)[\Omega_r(\tau)-\Omega_p(L,\tau)]^2+\frac{\beta}{2}\int_0^T d\tau\,\Omega_c^2(0,\tau), \tag{2}$$

where T is the pulse duration of the incident primary pulse and L is the thickness of the sample measured in units of the primary pulse absorption length. The first term measures the deviation of the output primary pulse from the reference pulse $\Omega_r(\tau)$, with $w(\tau)>0$ being a weight function. The second term is the cost of the fluence of the controlling pulse, with $\beta>0$ being a weight factor. Since we want to match the output primary pulse with the reference pulse while keeping the cost as small as possible, we need to find the optimal incident controlling pulse $\Omega_c^{\mathrm{OC}}(0,\tau)$ that minimizes J. Lagrange multipliers λ_j, $j=1,2,3$ and $\lambda_{p,c}$ are introduced in an augmented cost functional $\bar{J}$ to include the constraint of satisfying the Schrödinger-Maxwell equations (1)

$$\begin{aligned}\bar{J}=J+\int_0^L dz'\int_0^T d\tau\Bigg[&\lambda_1\left(\dot{a}_1-\frac{\Omega_p}{2}a_3\right)+\lambda_2\left(\dot{a}_2-\frac{\Omega_c}{2}a_3\right)\\&+\lambda_3\left(\dot{a}_3+\frac{\Gamma_3}{2}a_3+\frac{\Omega_p}{2}a_1+\frac{\Omega_c}{2}a_2\right)\\&+\lambda_p\left(\frac{\partial\Omega_p}{\partial z'}-\frac{\Gamma_3}{2}a_3a_1\right)\\&+\lambda_c\left(\frac{\partial\Omega_c}{\partial z'}-\frac{k_c\mu_c^2}{2k_p\mu_p^2}\Gamma_3a_3a_2\right)\Bigg].\end{aligned} \tag{3}$$

The new cost functional $\bar{J}$ may be minimized with no constraint except the initial condition $a_i(z',\tau=0)=a_{i0}(z')$, $i=1,2,3$. The necessary conditions for a solution are that the variations of $\bar{J}$ with respect to $a_i(z',\tau)$, $\lambda_i(z',\tau)$, $\lambda_{p,c}(z',\tau)$, and $\Omega_{p,c}(z',\tau)$ vanish. This leads to Eq. (1) and

$$\dot{\lambda}_1=\frac{\Omega_p}{2}\lambda_3-\frac{\Gamma_3}{2}\lambda_p a_3, \tag{4a}$$

$$\dot{\lambda}_2=\frac{\Omega_c}{2}\lambda_3-\frac{\Gamma_3 k_c\mu_c^2}{2k_p\mu_p^2}\lambda_c a_3, \tag{4b}$$

$$\dot{\lambda}_3=-\frac{\Omega_p}{2}\lambda_1-\frac{\Omega_c}{2}\lambda_2+\frac{\Gamma_3}{2}\lambda_3-\frac{\Gamma_3}{2}\lambda_p a_1-\frac{\lambda_c k_c\mu_c^2}{2k_p\mu_p^2}\Gamma_3 a_2, \tag{4c}$$

$$\frac{\partial\lambda_p}{\partial z'}=-\frac{\lambda_1}{2}a_3+\frac{\lambda_3}{2}a_1, \tag{4d}$$

$$\frac{\partial\lambda_c}{\partial z'}=-\frac{\lambda_2}{2}a_3+\frac{\lambda_3}{2}a_2, \tag{4e}$$

as well as

$$\frac{\delta\bar{J}}{\delta\Omega_c(0,\tau)}=\beta\Omega_c(0,\tau)-\lambda_c(0,\tau)=0. \tag{5}$$

The variations of $\bar{J}$ with respect to $a_i(z',T)$ and $\Omega_{p,c}(L,\tau)$ must also vanish. This leads to the boundary conditions

$$\lambda_p(L,\tau)=[\Omega_r(\tau)-\Omega_p(L,\tau)]w(\tau), \tag{6a}$$

$$\lambda_c(L,\tau)=0, \tag{6b}$$

$$\lambda_1(z',T)=\lambda_2(z',T)=\lambda_3(z',T)=0. \tag{6c}$$

The coupled first-order partial differential equations (1), (4), and (5) with the initial and boundary conditions can be solved iteratively. The algorithm we used is as follows: (i) Choose an initial trial controlling pulse $\Omega_c(0,\tau)$ over the range $0\leq\tau\leq T$. (ii) Propagate the Schrödinger-Maxwell equations, Eq. (1), from $\tau=0$, $z'=0$ to $\tau=T$, $z'=L$, with the given initial conditions and time dependence of the two pulses at $z'=0$. (iii) Set up the boundary conditions from Eq. (6) and propagate Eq. (4) from $\tau=T$, $z'=L$ to $\tau=0$, $z'=0$. (iv) Update the new controlling pulse

$$\Omega_c'(0,\tau)=\Omega_c(0,\tau)-\alpha\frac{\delta\bar{J}}{\delta\Omega_c(0,\tau)}, \tag{7}$$

where $\delta\bar{J}/\delta\Omega_c(0,\tau)$ can be calculated from Eq. (5), and α is a positive constant. A linear search is then made for the best values of α for making $\Omega_c'(0,\tau)$ generate the minimum objective $\bar{J}$. (v) Repeat (ii)–(iv) until Ω_c' converges and hopefully deviates very little from Ω_c.

In our numerical calculations, we choose $k_c/k_p=0.6$, $\mu_c=\mu_p$, $\Gamma_3=0.5$ cm^{-1}, $L=50$, and $\beta=10^{-6}$. The atoms are assumed to be in level $|1\rangle$ initially, i.e., $a_{10}(z')=1$, $a_{20}(z')=a_{30}(z')=0$. The incident primary pulse $\Omega_p(0,\tau)$ is confined between $0<\tau<T$ and equal to $0.1\sin(t\pi/T)$ cm^{-1} with $T=5$ ns. The weight function is chosen to be $w(\tau)=\exp(-4\tau^2/T^2)$. We chose to put more weight on matching the output primary pulse $\Omega_p(L,\tau)$ and the reference pulse $\Omega_r(\tau)$ at short times, which is the most difficult to match according to our experience.

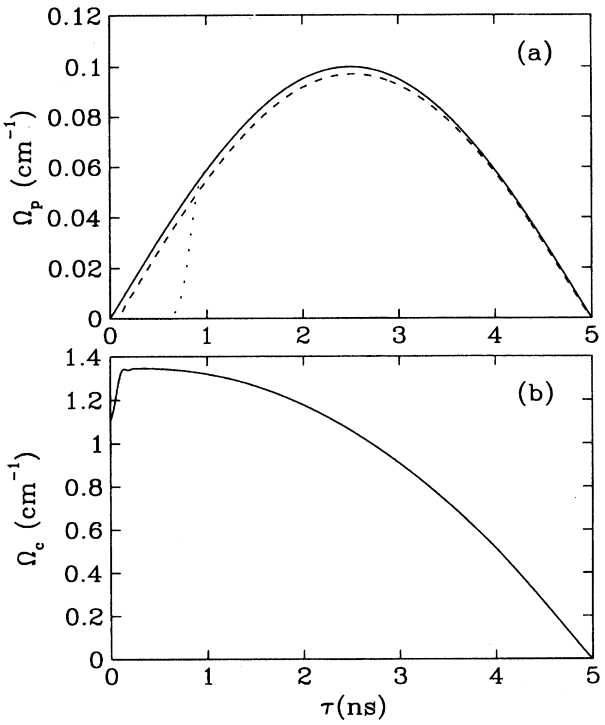

FIG. 2. (a) Incident primary pulse $\Omega_p(0,\tau)$ (solid line), outputprimary pulse from the OCT, $\Omega_p^{\mathrm{OC}}(L,\tau)$ (dashed line), and output primary pulse from the MPS, $\Omega_p^{\mathrm{MP}}(L,\tau)$ (dotted line). (b) Controlling pulse designed by the OCT, $\Omega_c^{\mathrm{OC}}(0,\tau)$.

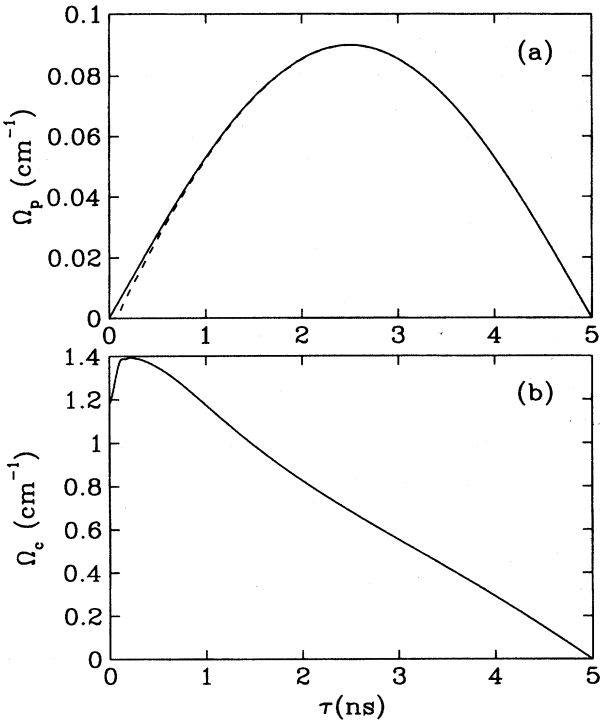

FIG. 3. (a) Reduced incident primary pulse $0.9\Omega_p(0,\tau)$ (solid line) and the output primary pulse from the OCT, $\Omega_p^{\mathrm{OC}}(L,\tau)$ (dashed line). (b) Controlling pulse designed by the OCT, $\Omega_c^{\mathrm{OC}}(0,\tau)$.

We first choose the reference pulse as $\Omega_r(\tau)=\Omega_p(0,\tau)$; i.e., we want the output primary pulse to be identical to the incident one. In Fig. 2, we display the controlling pulse $\Omega_c^{\mathrm{OC}}(0,\tau)$ designed by the OCT [Fig. 2(b)], and the resulting output primary pulse $\Omega_p^{\mathrm{OC}}(L,\tau)$ [Fig. 2(a), dashed line]. $\Omega_p^{\mathrm{OC}}(L,\tau)$ matches the incident primary pulse $\Omega_p(0,\tau)$ [solid line in Fig. 2(a)] pretty well. Our numerical results also show that the primary pulse is almost unchanged during the propagation.

In the MPS, the controlling pulse $\Omega_c^{\mathrm{MP}}(0,\tau)$ has an envelope identical to the incident primary pulse; i.e., $\Omega_c^{\mathrm{MP}}(0,\tau)=b\Omega_p(0,\tau)$, with b being a constant. The resulting output primary pulse $\Omega_p(L,\tau)$ can fit the incident one perfectly as long as b is sufficiently large. However, in order for the controlling pulse to be practical, we have to impose a constraint on its fluence. We therefore chose b such that $\Omega_c^{\mathrm{MP}}(0,\tau)$ has the same fluence as $\Omega_c^{\mathrm{OC}}(0,\tau)$; i.e., $b^2=\int_0^T d\tau|\Omega_c^{\mathrm{OC}}(0,\tau)|^2/\int_0^T d\tau|\Omega_p(0,\tau)|^2$. The resulting output primary pulse $\Omega_p^{\mathrm{MP}}(L,\tau)$ from the MPS is shown by the dotted line in Fig. 2(a). It has a sharp rising edge and does not keep the symmetric form of the incident primary pulse. The dashed line overall matches the solid line better, which means that the OCT is better than the MPS in protecting the magnitude and shape of the primary pulse.

Since all atoms are initially in level $|1\rangle$, the absorption of the primary pulse is inevitable. Therefore the output primary pulse is always smaller than the incident one and a complete protection of its magnitude and shape is impossible. However, we can obtain a better protection of the pulse shape if

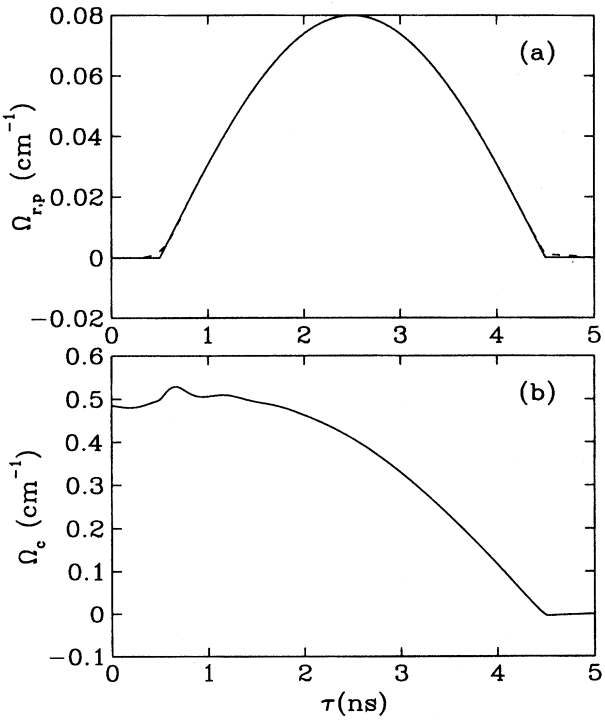

FIG. 4. (a) Reference pulse confined to be nonzero between $0.1T<t<0.9T$, $\Omega_r(\tau)=0.08\sin[(t-0.1T)\pi/(0.8T)]$ (solid line), and the output primary pulse from the OCT, $\Omega_p^{\mathrm{OC}}(L,\tau)$ (dashed line). (b) Controlling pulse designed by the OCT, $\Omega_c^{\mathrm{OC}}(0,\tau)$.

we allow its magnitude to decrease (absorbed by the medium) a little bit. For example, we can choose $\Omega_r(\tau) = 0.9\Omega_p(0,\tau)$. After all, it is usually the pulse shape rather than its absolute magnitude that is important. The controlling pulse designed by the OCT is displayed in Fig. 3(b), and the resulting output primary pulse $\Omega_p^{OC}(L,\tau)$ is shown in Fig. 3(a) by the dashed line. The solid line in Fig. 3(a) is $0.9\Omega_p(0,\tau)$. A nearly perfect match is obtained. Note that in the MPS, the best match one can get is shown in Fig. 2(a) by the dotted line.

Since there is a no limitation on the choice of the reference pulse $\Omega_r(\tau)$ (as long as it is smaller than the incident primary pulse), one should be able to manipulate the output primary pulse shape by properly designing the controlling pulse. We give one more example where $\Omega_r(\tau)$ is confined between $0.1T<t<0.9T$ and equal to $0.08\sin[(\tau-0.1T)\pi/(0.8T)]$ cm^{-1}. This reference pulse is obtained by compressing both the magnitude and the duration of the incident primary pulse by 20%. The controlling pulse designed by the OCT is displayed in Fig. 4(b), and the resulting output primary pulse $\Omega_p^{OC}(L,\tau)$ matches $\Omega_r(\tau)$ almost perfectly [see Fig. 4(a)].

Figures 2–4 show that the controlling pulses designed by the OCT are nonzero at $\tau=0$; i.e., there is an infinitely steep rising edge, which is difficult to achieve practically. This discontinuity at $\tau=0$ is due to the fact that we confined the controlling pulse to be over the range $0\leq t\leq T$. In our numerical calculation, we have assumed that all the atoms are in level $|1\rangle$ initially. For this special initial condition, the controlling pulse, which is resonant with the $|2\rangle$-$|3\rangle$ transition, does not affect the atoms for $\tau<0$ (i.e., before the incident of the primary pulse). Therefore we can assign an arbitrary smooth rising edge that starts from some negative time to the optimal controlling pulses shown in Figs. 2–4. The creation of these smooth nanosecond controlling pulses in the laboratory should be feasible with current pulse shaping techniques [16], provided the overall bandwidth is not too large and the carrier frequency is in an accessible region.

For general initial conditions, we need to confine the controlling pulse over the range $-T_0\leq\tau\leq T$ and set the range of time integration in Eq. (2) from $-T_0$ to T. For T_0 sufficiently large, we can obtain a controlling pulse with a smooth rising edge starting from some negative time.

In summary, we develop an OCT for controlling the propagation of a primary optical pulse using a secondary controlling pulse. The absorption and reshaping of the controlling pulse by the medium is automatically incorporated in the OCT, which extends the powerful design capabilities of OCT to optically dense media. For arbitrary initial conditions of the medium and incident primary pulse shape, the OCT designs the controlling pulse to obtain an arbitrary desired output primary pulse. This procedure is especially useful for complicated primary pulse shapes where design by physical intuition fails. The OCT will provide the control pulse best able to meet the desired primary pulse objective. With a constraint on the fluence, the controlling pulse designed by the OCT is better than that of the MPS for protecting the shape and magnitude of the primary pulse. One can obtain better protection of the pulse shape if small absorptions are allowed. Figures 2 and 3 show that the peak of the optimal controlling pulse $\Omega_c^{OC}(0,\tau)$ precedes the peak of the primary pulse. This counterintuitive order can be explained as follows: the two-photon transition from $|1\rangle$ to $|2\rangle$ creates a trap state that is immune to further absorption [4]. Therefore the absorption of the primary pulse is a minimum when the two-photon transition (population transfer) is most effective, which requires that the primary and the controlling pulses be incident in counterintuitive order [10,17].

We acknowledge support from the Office of Naval Research and the U.S. Army Research Office.

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