Analytical solution of the optimal laser control problem in two-level systems

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Abstract

The optimal control of two-level systems by time-dependent laser fields is studied using a variational theory. We obtain, for the first time, general analytical expressions for the optimal pulse shapes leading to global maximization or minimization of different physical quantities. We present solutions which reproduce and improve previous numerical results.

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Optimal laser control in quantum systems is a problem of fundamental importance for atomic, molecular, solid-state and chemical physics which has attracted much attention in the last 10 years. Any system exhibiting quantum coherence can be subject to optimal control, which basically consists in the manipulation of the quantum dynamics by external time-dependent laser fields. A general procedure for this manipulation is described by the optimal control theory (OCT) [1,2]. The time-envelope of the external field is optimized by experimental or numerical pulse-shaping techniques in order to drive the wave function $\psi(t)$ of the system to fit a target state $\phi_0$ at a particular control time $t_{\text{control}}$. Thus, the optimal shape $V_{\text{opt}}(t)$ of the external field achieves the maximization of the quantity $|\langle \phi_0 | \psi(t_{\text{control}}) \rangle|$. Since the pioneering work by Hudson and Rabitz [3], different approaches to optimal control have been proposed and experimentally applied in various contexts [1,2,4–10]. The major problem of the current theoretical description of optimal control is that the resulting equations are of high complexity and must be solved numerically. Therefore, neither the experimental realization nor the theoretical description of optimal control guarantee that the result of the optimization corresponds to the true global extremum of the control problem considered. As a functional of the pulse shape, the physical quantity to be maximized or minimized represents a hyper surface in a multidimensional space, which might exhibit many local extrema in which the experimental or numerical procedure can get trapped.

The only way to extract the global extremum from among the multiplicity of local extrema is by finding the analytical solution to the control problem. In this paper we present for the first time analytical results for the optimal pulse shapes leading to the global extrema of different optimal control problems.

We concentrate on physical situations which can be described by two-level systems. Using our approach, we give explicit pulse shapes for inducing the maximization of population transfer between two levels and for the achievement of self-induced transparency under the constraint of fixed pulse energy.

We consider a physical quantity $Q$ which is only nonzero when the system is in the excited state, and which we wish to maximized or minimized within the control interval
For this purpose an external field of optimal shape has to be applied. $Q$ can be seen as a fitness function. We write it as a time average, over the time interval $[T_0, T_1]$, of the form

$$Q = \int_{T_0}^{T_1} Q(t) dt. \quad (1)$$

$Q(t)$ is a fitness density, which is a functional of the density matrix $\rho(t)$ of the system and, consequently, also of the external field $V(t) \cos(\omega t)$.

Our approach to derive an equation for the optimal field shape is based on a Lagrangian of the form [11]

$$L = \int_{T_0}^{T_1} \mathcal{L} dt = \int_{T_0}^{T_1} \Gamma(t) \left( \frac{\partial}{\partial t} + i \hat{Z}(t) \right) \rho(t) dt + \lambda \int_{T_0}^{T_1} Q[\rho(t)] dt + \lambda_1 \int_{T_0}^{T_1} V^2(t) dt, \quad (2)$$

where $\mathcal{L}$ is the Lagrangian density, $\lambda$ and $\lambda_1$ are Lagrange multipliers and $\Gamma(t)$ a Lagrange multiplier density [12].

The first term in Eq. (2) ensures that the density matrix satisfies the quantum Liouville equation with the corresponding Liouville operator $\hat{Z}(t)$. The second term explicitly includes the description of the optimal control and refers to a physical quantity to be optimized during the control time interval. The third term in Eq. (2) represents a constraint on the total energy $E_0$ of the control field [11]. The Lagrangian (2) is the basis of our control theory, since it allows the derivation of the equations to be fulfilled by the control field.

As mentioned before, one of the purposes of this paper is to find analytical solutions for the "standard" optimal control problem. This means, we search for the maximization or minimization of $Q(t_{control})$ at a particular control time $t_{control}$. Note that we can treat this problem as a particular case of the theory presented above. We only need to modify the fitness density $Q$ by introducing a delta function as follows

$$Q = \int_{T_0}^{T_1} Q[\rho(t)] \delta(t - t_{control}) dt = Q[\rho(t_{control})], \quad (3)$$

where $t_{control}$ lies within the time interval $[T_0, T_1]$.

We now apply the theory described above to a two-level quantum system with energy levels $\epsilon_1$ and $\epsilon_2$, interacting with an external control field of the form $V(t) \cos(\omega t)$, where $\omega$ is
the carrier frequency. If the resonant condition \( \omega = \epsilon_2 - \epsilon_1 \) and adiabaticity criterion \(|\dot{V}\omega| \ll |V|^3 \) [13] for the control fields are satisfied, one can use the Rotating Wave Approximation (RWA) to derive the Liouville equation for the density matrix

\[
\begin{align*}
\dot{\rho}_{11} &= \mu V(t)(\tilde{\rho}_{12} - \tilde{\rho}_{21}) + i\gamma_1 \rho_{22}, \\
\dot{\rho}_{22} &= \mu V(t)(\tilde{\rho}_{21} - \tilde{\rho}_{12}) - i\gamma_1 \rho_{22}, \\
\dot{\tilde{\rho}}_{12} &= \mu V(t)(\rho_{22} - \rho_{11}) - i\gamma_2 \tilde{\rho}_{12},
\end{align*}
\] (4)

where \( \mu \) is the dipole matrix element of the two-level system and \( \gamma_1, \gamma_2 \) are relaxation and dephasing constants, respectively. Here we use the notation \( \tilde{\rho}_{12} = \rho_{12} \exp(i\omega t) \) and \( \tilde{\rho}_{21} = \rho_{21} \exp(-i\omega t) \). \( \rho_{11} \) and \( \rho_{22} \) correspond to the instantaneous occupation of the ground and excited state, respectively. Note, that \( \rho_{11} + \rho_{22} = 1 \) and \( \tilde{\rho}_{21} = \tilde{\rho}_{12}^* \). We set the initial conditions as \( \rho_{11} = 1, \rho_{22} = \tilde{\rho}_{12} = \tilde{\rho}_{21} = 0 \).

As a first application we address the phenomenon known as self-induced transparency (SIT) [14,15]. The problem consists in finding a temporal pulse shape for which a light pulse entering a material propagates without significant losses. This effect has been studied theoretically using different approaches [14,15]. However, it has never been considered so far as an optimal control problem. We show below that SIT can be viewed as the search for the optimal pulse shape for which losses during propagation are minimized.

In order to solve the problem from the perspective of optimal control, one can assume, as usual, that materials where SIT occurs are collections of inhomogeneously broadened two-level systems [14,15]. If we assume, in addition, that the material is optically thin (thus, we neglect changes of the field along the spatial axis), we can describe the phenomenon using the theory presented above and obtain analytical results. The losses of such a system are proportional to the average occupation of the upper level

\[
\Gamma_{\text{loss}} \propto Q_{22} = \int_{T_0}^{T_1} \rho_{22}(t) dt.
\] (5)

Thus, we search for the minimization of the physical quantity \( Q_{22} \), i.e., the integral of the occupation of the excited level over the time interval \([T_0, T_1]\). Since systems exhibiting SIT
are characterized by long life-times of the excited levels, we assume $\gamma_{1,2}T \ll 1$, where $T$ is a characteristic time during which the system is in the exited state. $T$ is proportional to the inverse Rabi frequency induced by the control field. This assumption permits us to neglect relaxation and dephasing effects within the control interval and write the solution for occupation of the upper level $\rho_{22}(t)$ as

$$\rho_{22}(t) = \sin^2(\theta(t)), \quad (6)$$

where the pulse area $\theta$ is defined as

$$\theta(t) = \mu \int_{t_0}^{t} dt' V(t'). \quad (7)$$

Thus, the Lagrangian density is a function of the pulse area and its first derivative $L(\theta, \dot{\theta})$ and is given by

$$L = \sin^2(\theta(t)) + \lambda \frac{\dot{\theta}^2}{\mu^2}. \quad (8)$$

In Eq. (8), and also in the rest of the paper, we omit the Liouville term since we include the analytical expression (6) for the density matrix which solves the Liouville equation. The Euler-Lagrange equation derived from the above Lagrangian density reads

$$2\lambda \ddot{\theta}(t) - \mu^2 \sin(2\theta(t)) = 0. \quad (9)$$

Eq. (9) is of the second order and requires two boundary conditions. We consider for simplicity an infinitely large control interval $t \in (-\infty, \infty)$ with natural boundary conditions $\theta(-\infty) = V(-\infty) = V(+\infty) = 0$ and $\theta(+\infty) = \pi$. This condition implies that the system is excited from and de-excited to the ground-state, remaining there after the interaction with the control field. It is clear that these are the boundary conditions compatible with a minimization of $Q_{22}$ (see Fig. 1). Eq. (9) can be integrated analytically (it is mathematically equivalent to the pendulum equation). The resulting optimal field envelope $V_{opt}(t)$ is given by the expression

$$V_{opt}(t) = (\sqrt{\lambda} \cosh(t/\sqrt{\lambda/\mu^2})^{-1}. \quad (10)$$
The Lagrange multiplier $\lambda$ is determined from the normalization condition $\int_{-\infty}^{+\infty} V^2(t)dt = E_0$ for the pulse energy and is given by $\lambda = 4/(\mu E_0)^2$.

Eq. [10] represents the well known soliton solution for the pulse shape and is shown in the inset of Fig. 1. This result shows that the soliton wave does not only propagate without shape changes [16] but it also minimizes the energy losses, which are proportional to $Q_{22}$ in the limit of the weak relaxation and dephasing. This fact can be clearly shown in Fig. 1, where we show the integrated population $\int_{-\infty}^{t} \rho_{22}(t')dt'$ over the control interval for the optimal pulse shape and for a square pulse having the same area and the same energy as the optimal. From the figure it is clear that $Q_{22}$, and therefore the losses, is smaller for the soliton pulse shape.

Using other asymptotic values of the pulse area $\theta(+\infty) = N \pi$, with $N = 2, 3, \ldots$, one can immediately reproduce $2\pi, 3\pi, \ldots$ soliton solutions. These soliton shapes are the optimal pulses corresponding to different values of the pulse energy, which are, of course, higher than the energy of the shape of Eq. [10].

Now we turn to the ”standard” optimal control problem at fixed time and show how the analytical solution to the problem of maximization of an objective at a particular time $t_{control}$ arises naturally as a limiting case of our theory. If the quantity to be maximized is, for instance, the population of the upper level at time $t_{control}$, $\rho_{22}(t_{control})$, then the Lagrangian density becomes, with the help of Eq. (3)

$$L_{\delta} = \rho_{22}(t)\delta(t - t_{control}) + \lambda \dot{\theta}^2(t)/\mu^2,$$

where the delta function $\delta(t - t_{control})$ accounts for the modification of the fitness density. The optimal pulse shape can only be obtained analytically for $\gamma_1 = \gamma_2 = 0$, i.e., if relaxation and dephasing effects are neglected. In this case the corresponding Euler-Lagrange equation reads

$$2\lambda \ddot{\theta}(t) - \mu^2 \delta(t - t_{control}) \sin(2\theta(t)) = 0. \tag{12}$$

Integrating Eq. (12) one obtains the pulse area as a linear function $\theta(t) = At + B$. By substituting the boundary conditions $\theta(0) = 0, \theta(t_{control}) = \pi/2$, we find that the solution of
Eq. (12) is a field with a constant amplitude

\[ V_{opt}(t) = \frac{\pi}{2\mu t_{control}}, \quad (13) \]

with energy \( E_0 = \pi^2/(4\mu^2 t_{control}) \), measured in the time-interval \([0, t_{control}]\). This result reflects the fact that, among all pulses with area equal to \( \pi/2 \), that which minimizes the energy has \textit{time-independent shape}. It must be pointed out that this analytical solution corresponds to the global extremum of the Lagrangian as long as the RWA is applicable.

It is important recall that Eq. (13) is a new result and should not be confused with the trivial fact that a \( \pi/2 \) pulse, when limited to a constant amplitude, produces complete inversion.

In Fig. 2 we compare the analytical result of Eq. [13] with a numerical solution obtained by Zhu et al. using OCT [2]. In Ref. [2], the optimal field to induce population inversion between two levels of the Morse potential at a particular time was calculated using an iterative numerical technique to integrate the OCT equations. The obtained field consists of a single frequency (resonant with the difference of the level-energies) and a time-dependent amplitude, shown as a dashed line in Fig. 2. In order to reproduce the same physical situation and use the same parameters as in Ref. [2] we calculated the dipole matrix element \( \mu \) as \( \mu = \langle \psi_0 | \hat{\mu} | \psi_1 \rangle \), where \( \hat{\mu}(r) = \mu_0 e^{-r/r_0} \), \( \mu_0 = 3.088 \), and \( r_0 = 0.6 \). \( \psi_0(r) \) and \( \psi_1(r) \) correspond to the ground and the first excited state eigenfunctions of the Morse potential, which is given by \( V(r) = D_0(\exp(-\beta(r-r^*)) - 1)^2 - D_0 \), with \( D_0 = 0.1994 \), \( \beta = 1.189 \) and \( r^* = 1.821 \) [2]. Thus, we used Eq. [13] to determine the magnitude of the constant optimal amplitude. Our analytically calculated optimal field, which corresponds in fact to the true global extremum, is shown as a thin solid line in Fig. 2. Note that the shape obtained by Zhu et al. is close to that of the optimal field obtained by us. However, it is clear that it does not correspond to the global extremum of the problem. Moreover, the numerically determined shape shows a slight asymmetry, which gives rise to a broadening of its Fourier spectrum, i.e., to a less effective coupling to the two-level system.

This example shows that our analytical approach can be used to check the ability of
different numerical methods to avoid local extrema.

Summarizing, we have obtained analytical solutions for the optimal shape of external fields to control populations in two-level systems over finite time-intervals and at particular control times. Our obtained optimal shapes constitute the global extrema of the control problem, in contrast to previous numerical solutions. Our results can be used as a basis to solve optimal control problems in materials which are well described by collections of two-level systems.

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Since we neglect changes of the field along the spatial variable one can treat the Eq. [9] as the sine-Gordon equation in the limit of the optically thin media.
FIG. 1. Integrated occupation of the upper level $\int_{-\infty}^{t} \rho_{22}(t')dt'$ for the optimal (soliton) pulse (solid line) and for a square pulse (dashed line), shown in the inset figure. Note that $Q_{22} = \int_{-\infty}^{\infty} \rho_{22}(t')dt'$ is proportional to the losses $\Gamma_{\text{loss}}$. Inset figure: pulse shape for the optimal control field $V_{\text{opt}}(t)$ to achieve self-induced transparency through minimization of the losses in a two-level system (solid line), and for a square pulse having the same pulse area and energy (dashed line).
FIG. 2. Thin solid line: analytical solution for the optimal control field $V_{opt}(t) \cos(\omega t)$, with energy $E_0 = \pi^2/(4\mu^2 t_{control})$ (see text) to produce inversion of the population at $t_{control} = 30000$ (a.u.). The optimal amplitude $V_{opt}(t)$ is time-independent [see Eq. (13)]. Dashed line: numerical result $V_{oct}(t)$ for the field amplitude for the same problem and the same system-parameters obtained in Ref. 2 using optimal control theory.