Theory for the Optimal Control of Time-Averaged Quantities in Quantum Systems

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(Received 16 November 2001; published 18 November 2002)

We present a variational theory for the optimal control of quantum systems with relaxation over a finite time interval. In our approach, which is a nontrivial generalization of previous formulations and which contains them as limiting cases, the optimal control field fulfills a high-order Euler-Lagrange differential equation, which guarantees the uniqueness of the solution. We solve this equation numerically and also analytically for some limiting cases. The theory is applied to two-level quantum systems with relaxation, for which we determine quantitatively how relaxation effects limit the control of the system.

DOI: 10.1103/PhysRevLett.89.233003 PACS numbers: 32.80.Qk

The manipulation of quantum-mechanical systems by using ultrashort time-dependent fields represents a challenging fundamental physical problem. In the past years, a considerable amount of experimental and theoretical work was concentrated on designing laser pulses having optimal amplitude and modulation in order to control the quantum dynamics in various systems such as atoms and molecules [1], quantum dots [2], semiconductors [3], superconducting devices [4], and Bose-Einstein condensates [5].

Several theoretical studies, most of them using numerical optimization techniques, have shown that it is possible to construct optimal external fields (e.g., laser pulses) to drive a certain physical quantity, such as the population of a given state, to reach a desired value at a given time [6–8]. However, even for the simplest control problems the obtained fields have a rather complex nature and cannot be easily interpreted. Furthermore, since the optimal field is usually obtained from a system of coupled nonlinear integrodifferential equations, which are solved numerically using iterative methods, the achievement of the global extremum of the problem is not guaranteed.

Although the kind of control discussed above is relevant for many purposes, a more detailed manipulation of real systems may require the control of physical quantities over a finite time interval. The search for optimal fields able to perform such control is a highly complex problem for which no theoretical description has been given so far.

In this Letter we present for the first time an analytical theory for the control of simple quantum systems with relaxation over a finite time interval. By using a variational approach we derive a high-order differential equation from which the optimal control fields are obtained. We also determine the influence of relaxation, which limits control, and we analyze the potential applications to the manipulation of physical quantities, such as the induced photocurrent through impurities in semiconductors or the population of electronic states at metallic surfaces.

Our goal is to formulate a theory which permits one to derive an explicit differential equation to be satisfied by the optimal control field. Note that one can guess the form of such equation from general physical arguments. Since temporal coherence and memory effects are expected to be important, one should search for a differential equation containing both the shape of the external field envelope $V(t)$ and the pulse area $\theta(t) = \mu \int_{t_0}^{t} dt' V(t')$, μ being the dipole matrix element of the system. For the control of quantities over a finite time interval $[t_0, t_0 + T]$ two boundary conditions $[\theta(t_0), \theta(t_0 + T)]$ have to be fulfilled. It means that a differential equation satisfied by $\theta(t)$ must be of at least second order. If one also imposes boundary conditions for the field $[\dot{\theta}(t_0)/\mu, \dot{\theta}(t_0 + T)/\mu]$, then the differential equation for $\theta(t)$ must be at least of fourth order. We show below that under certain conditions a fourth order differential equation for $\theta(t)$ arises naturally as an Euler-Lagrange (EL) equation.

We start by considering a quantum-mechanical system being in contact with the environment and interacting with an external field $E(t) = V(t)\cos(\omega t)$. Here $V(t)$ refers to an arbitrary pulse shape and ω is the carrier frequency. The evolution of such a system obeys the quantum Liouville equation for the density matrix $\rho(t)$ with dissipative terms. The control of a time-averaged dynamical quantity of the system requires the search for the optimal shape $V(t)$ of the external field on the time interval $[0, T]$. Thus, we propose the following Lagrangian (throughout the Letter we use atomic units $\hbar = m = e = 1$):

$$L = \int_0^T A(t)\left(\frac{\partial}{\partial t} + i\hat{\mathcal{Z}}(t)\right)\rho(t)dt + \beta \int_0^T \mathcal{L}_1 dt. \quad (1)$$

Here, β is a Lagrange multiplier and $A(t)$ is a Lagrange multiplier density. The first term in Eq. (1) ensures that the density matrix satisfies the quantum Liouville equation with the corresponding Liouville operator $\hat{\mathcal{Z}}(t)$[6]. While the first term describes the dynamics of the system under the external field, the functional density

$\mathcal{L}_1$ explicitly includes the description of the optimal control and is given by

$$\mathcal{L}_1(\rho, V) = \mathcal{L}_{\rm ob}(\rho) + \lambda V^2(t) + \lambda_1\left(\frac{dV(t)}{dt}\right)^2, \quad (2)$$

where λ and λ_1 are Lagrange multipliers. $\mathcal{L}_{\rm ob}(\rho)$ refers to the physical quantity (objective) to be maximized during the control time interval. The second term represents a constraint on the total energy of the control field

$$2\int_0^T E^2(t)dt \approx \int_0^T V^2(t)dt = \int_0^T \frac{\dot{\theta}^2(t)}{\mu^2}dt = E_0. \quad (3)$$

The third term corresponds to a further constraint on the properties of the pulse envelope. The requirement

$$\int_0^T \left(\frac{dV(t)}{dt}\right)^2 dt = \int_0^T \frac{\ddot{\theta}^2(t)}{\mu^2}dt \le R, \quad (4)$$

where R is a positive constant (maximal curvature). Equation (4) excludes very narrow peaks or abrupt steplike solutions, which cannot be achieved experimentally. As we show below, the constraint (4) is necessary when one needs to impose boundary conditions for the field amplitude $V(t)$.

Assuming that the density matrix $\rho(t)$ depends only on $\theta(t)$ and on the time t, one obtains an explicit expression for the functional $\mathcal{L}_1 = \mathcal{L}_1(\theta, \dot{\theta}, \ddot{\theta}, t)$. The corresponding extremum condition $\delta\mathcal{L}_1 = 0$ yields the high-order EL equation

$$-\lambda_1\frac{d^4\theta}{dt^4} + \lambda\frac{d^2\theta}{dt^2} - \frac{\mu^2}{2}\frac{\partial\mathcal{L}_{\rm ob}(\rho)}{\partial\theta} = 0. \quad (5)$$

Here, the dependence on the pulse energy and curvature is contained implicitly in the Lagrange multipliers λ and λ_1. In order to solve Eq. (5) one can assume the natural boundary conditions $\theta(0) = \dot{\theta}(0) = \dot{\theta}(T) = 0$, and $\theta(T) = \theta_T$. The choice of the constant θ_T depends on the problem. In general, the constants θ_T, R, and E_0 can be also the object of the optimization. Note that the above formulated problem is highly nonlinear with respect to the function $\theta(t)$ and in most cases can be solved only numerically.

Equation (5) is one of the main results of this Letter and provides an explicit differential equation for the control field. Note that this equation is applicable only if $\rho = \rho[\theta(t), t]$. In order to show that Eq. (5) can describe optimal control in real physical situations, we apply our theory to a two-level quantum system. This is characterized by the energy levels ϵ_1 and ϵ_2, a dipole matrix element μ, and the relaxation and dephasing constants γ_1 and γ_2, respectively. The carrier frequency of the control field is chosen to be the resonant frequency $\omega = \epsilon_2 - \epsilon_1$. Assuming that the control field satisfies the adiabatic criterion and using the rotating wave approximation we describe the dynamics of the density matrix $\rho(t)$ by the equations

$$\begin{aligned} i\frac{\partial\rho_{\ell\ell}}{\partial t} &= (-1)^\ell[\mu V(t)(\rho_{21} - \rho_{12}) - i\gamma_1\rho_{22}], \\ i\frac{\partial\rho_{12}}{\partial t} &= \mu V(t)(\rho_{22} - \rho_{11}) - i\gamma_2\rho_{12}, \end{aligned} \quad (6)$$

with $\ell = 1, 2$. Note that $\rho_{11} + \rho_{22} = 1$ and $\rho_{21} = \rho_{12}^*$. Equations (6) are used for the description of different physical processes, for instance, two-level atoms interacting with an electromagnetic field, the response of donor impurities in semiconductors to terahertz radiation [3], or the excitation of the surface into image charge states at noble metal surfaces [9]. Therefore, the initial conditions are set as $\rho_{11} = 1$, $\rho_{22} = \rho_{12} = \rho_{21} = 0$.

Equations (6) are difficult to integrate, since $[\hat{\mathcal{Z}}(t), \hat{\mathcal{Z}}(t')] \neq 0$ [with $t, t' \in (0, T)$]. However, the commutators $[\hat{\mathcal{Z}}(t), \hat{\mathcal{Z}}(t')]$ become arbitrarily small under the condition [10]

$$\left|\frac{\gamma_\ell T^2}{V(t)}\left(\frac{\partial V(t)}{\partial t}\bigg|_{t'}\right)\right| \ll 1, \quad (7)$$

with $\ell = 1, 2$. In this case the approximate solution for $\rho_{22}(t)$ is

$$\begin{aligned} \rho_{22}(t) = 2\theta^2(t)F^{-1}\{&1 - \cosh(H)\exp[-(\gamma_1 + \gamma_2)t/2] \\ &+ (\gamma_1 + \gamma_2)t\sinh(H) \\ &\times \exp[-(\gamma_1 + \gamma_2)t/2]H^{-1}\}, \end{aligned} \quad (8)$$

where $H = \sqrt{[(\gamma_1 - \gamma_2)^2t^2 - 16\theta^2(t)]}/2$, and $F = \gamma_1\gamma_2t^2 + 4\theta^2(t)$. Note that this approximate solution becomes exact when $\gamma_1 = \gamma_2 = 0$ or for a control field with constant amplitude $V(t) = V_0$. Equation (8) has the form $\rho = \rho(\theta(t), t)$ which allows us to apply Eq. (5).

Now we construct the functional $\mathcal{L}_{\rm ob}(\rho) = \rho_{22}(t)/T$, so that the average occupation of the upper level $n_2 = (1/T)\int_0^T \rho_{22}(t)dt$ is the quantity to be maximized. Note, that n_2 is proportional to the observed photocharge [3] in terahertz experiments on semiconductors. The resonant tunneling charge through an array of coupled quantum dots is also proportional to n_2 [11].

It is well known that the presence of decoherence difficults optimal control [6]. From Eq. (8) one can show that for a strong control field satisfying $\gamma_{1,2}\,t/\theta(t) \ll 1$ the instantaneous population $\rho_{22}(t)$ always lies under the curve $\rho_{22}^{\max}(t) = \{1 + \exp[-(\gamma_1 + \gamma_2)t/2]\}/2$. Therefore, due to dissipative processes the following inequality holds

$$n_2 = \frac{1}{T}\int_0^T \rho_{22}(t)dt \le \frac{1}{2} + \frac{1 - \exp[-(\gamma_1 + \gamma_2)T/2]}{(\gamma_1 + \gamma_2)T} \quad (9)$$

which means that there is an absolute upper limit for the optimal control of averaged occupations in two-level systems with relaxation. Equation (9) exhibits two limiting cases: For very weak relaxation ($\gamma_{1,2}T \ll 1$) the populations can be fully inverted and remain in this state so that the maximum possible value of the controlled quantity is $n_2 \simeq 1$. In contrast, in the strong relaxation limit $\gamma_{1,2}T \simeq 1$, there is no possibility to perform coherent control on the time scale T because the system is in the saturation regime, so that the levels 1 and 2 become approximately equally occupied and $n_2 \simeq 1/2$. It is not possible to overcome the limit given by Eq. (9) within the considered model. However, the optimal pulse that satisfies Eq. (5) provides the highest possible value of n_2 for a given pulse energy, as we show below.

We have calculated the optimal $V(t)$ from the numerical integration of Eq. (5) for different values of the relaxation constants γ_1 and γ_2. We perform the determination of the optimal field for given values of E_0 and R as follows. Once we solve Eq. (5) for different values of the Lagrange multipliers we determine the parametric dependence $E_0 = E_0(\lambda, \lambda_1)$ and $R = R(\lambda, \lambda_1)$. Thus, using an interpolation procedure we obtain the searched optimal pulse with the required properties E_0 and R. In the same way we determine the optimal boundary condition for $\theta(T)$. For simplicity we consider the control interval $[0, 1]$ and set $\mu = 1$.

In Fig. 1 we show the optimal field for a two-level system without ($\gamma_{1,2} = 0$) and with relaxation ($\gamma_1 = 2\gamma_2 = 0.2$) for the same value of the pulse energy E_0. Note that in both cases the pulse maximum occurs near the beginning of the control interval. This leads to a rapid increase of the population $\rho_{22}(t)$ and therefore to a maximization of n_2. For the case $\gamma_{1,2} = 0$ the pulse vanishes when the population inversion has been achieved, whereas for $\gamma_{1,2} \neq 0$ the optimal pulse is broader $[\theta(T) > \pi/2]$. Using Eq. (8) one can estimate that in the presence of an appropriate external field $\rho_{22}(t)$ can achieve a minimal decay rate $(\gamma_1 + \gamma_2)/2 = 3\gamma_1/4$ which is smaller than the free decay of the system with rate γ_1. Therefore relaxation effects can be partially compensated by a longer application of the field during the control interval. In the inset of Fig. 1 we show the corresponding dynamics of the population $\rho_{22}(t)$ for both cases. As mentioned before, Eq. (8) is exact for $\gamma_{1,2} = 0$. However, for $\gamma_{1,2} \neq 0$ it also agrees well with the numerical solution of the Liouville equation, indicating that $V(t)$ fulfills the condition (7) on the control interval.

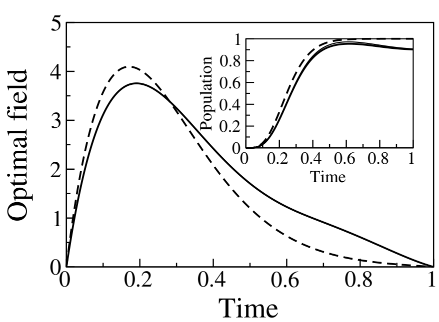

FIG. 1. Optimal control field which maximizes the time-averaged occupation n_2 in a two-level system. Dashed line: $\gamma_{1,2} = 0$. The pulse energy is $E_0 = 4.6$ and the pulse curvature $R = 182.2$. The solid line shows the optimal pulse for $\gamma_1 = 2\gamma_2 = 0.2$ with the same energy and curvature $R = 134.32$. Inset: Dynamics of the instantaneous population $\rho_{22}(t)$ for $\gamma_{1,2} = 0$ system (dashed line) and with relaxation [solid line using Eq. (8), thin solid line: numerical solution of the Liouville Eq. (6)].

The optimal pulses obtained for the system with relaxation considerably improve the value of the average occupation n_2 with respect to other pulse shapes such as Gaussian or rectangular. In Fig. 2 we show the behavior of the population $\rho_{22}(t)$ under the action of the optimal field, the best possible sine square shaped pulse, and the best possible "smooth rectangular" pulse [12] for the *same values* of E_0 and R. For the parameters used in Fig. 2 the optimal pulse yields an improvement of about 20% with respect to the best sine square pulse and of the best smooth rectangular pulse. Note that the magnitude of these improvements indeed depends on the dimensionless parameter $\gamma_1 T$ as well as on the values of E_0 and R.

In order to visualize the physics contained in the control fields of Fig. 1 one can simplify the general fourth order differential Eq. (5) and reduce it to a second order one which can be integrated analytically in some cases. For this purpose, we eliminate the constraint on the derivative of the field envelope [Eq. (4)]. Thus, the Lagrangian density $\mathcal{L}_1$ for the optimal control now has the form

$$\mathcal{L}_1 = \rho_{22}(t)/T + \lambda\dot{\theta}^2(t)/\mu^2. \tag{10}$$

For $\gamma_{1,2}T \ll 1$ one can neglect decoherence within the control interval and Eq. (8) becomes simply $\rho_{22}(t) = \sin^2[\theta(t)]$. Therefore, the corresponding EL equation is given by

$$2\lambda\ddot{\theta}(t) - \mu^2 \sin[2\theta(t)]/T = 0. \tag{11}$$

The second order differential Eq. (11) requires only two boundary conditions, for which we choose $\theta(0) = 0$ and $\theta(T) = \pi/2$. Note that this automatically leads to the unphysical initial condition $V(0) \neq 0$. However, Eq. (11)

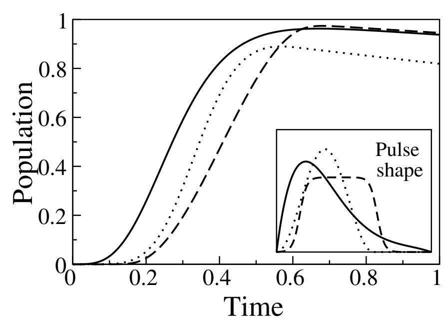

FIG. 2. Time evolution of the instantaneous population $\rho_{22}(t)$ under the action of the optimal field (solid line), the best "smooth rectangular" pulse (dashed line), and the best sine square pulse (dotted line) for the same values of $E_0 = 4.17$ and $R = 136.4$. $\gamma_1 = 2\gamma_2 = 0.1$. Inset: illustration of the three pulse shapes.

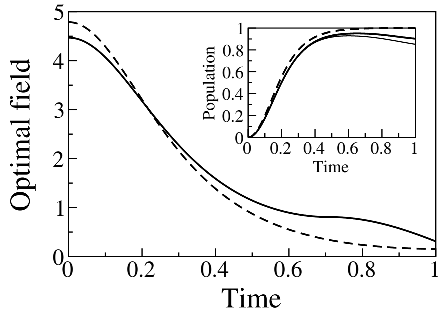

FIG. 3. Optimal control field obtained from the Lagrangian density of Eq. (10). Dashed line: $\gamma_{1,2} = 0$ [analytical solution, Eq. (12)]. The pulse energy is $E_0 = 4.8$. Solid line: optimal field for $\gamma_1 = 2\gamma_2 = 0.2$ with the same pulse energy (obtained numerically). Inset: Dynamics of the instantaneous population $\rho_{22}(t)$ for $\gamma_{1,2} = 0$ (dashed line) and with relaxation [thick solid line using Eq. (8), thin solid line: numerical solution of the Liouville Eq. (6)].

has the advantage that it can be solved analytically. The resulting field envelope is given by

$$V(t) = V(0)dn(\mu V(0)t, C), \qquad (12)$$

where dn is the Jacobian elliptic function, and $C = -[\lambda T V^2(0)]^{-1}$ is a constant of integration. Using the constraint on the pulse energy [Eq. (3)] we determine the coefficient λ.

In Fig. 3 we plot the optimal control field $V(t)$ corresponding to the Lagrangian density $\mathcal{L}_1$ [Eq. (10)] for a two-level system with and without relaxation. In both cases the field has its maximum value at $t = 0$ and exhibits a monotonic decay. As in the case of the solutions of the fourth order Eq. (5), the control field is broader for $\gamma_{1,2} \neq 0$ in order to compensate for the decay of the exited state. In the inset of Fig. 3 we plot the time evolution of the population $\rho_{22}(t)$.

From the comparison of the overall behavior of $\rho_{22}(t)$ in Figs. 1 and 3 one can conclude that the essential physics of the optimal control is already contained in the Lagrangian density $\mathcal{L}_1$ [Eq. (10)]. The boundary conditions $V(0) = V(T) = 0$ dramatically change the shape of the optimal fields, but they do not affect significantly the dynamics of the optimally controlled system.

The theory presented in this Letter is more general than optimal control theory at a given time and contains it as a limiting case. For instance, for the problem of maximization of $\rho_{22}(t)$ at time T_1 the Lagrangian density [Eq. (10)] reduces to

$$\mathcal{L}_1 = \rho_{22}(t)\delta(T_1 - t) + \lambda\dot{\theta}^2(t)/\mu^2. \qquad (13)$$

In this case the corresponding differential equation can also be integrated analytically for $\gamma_{1,2} = 0$ [10].

Finally, we can use Eq. (9) to determine the maximal possible lifetime for an image state at a Cu(111) surface which can be achieved by pulse shaping. According to Hertel *et al.* [9], those states are characterized by $\gamma_1 = 5 \times 10^{13}\ \mathrm{s}^{-1}$ and $\gamma_2 = \gamma_1/2$. Thus, our theory predicts in that case an effective decay constant $\gamma_{\mathrm{eff}} \geq (\gamma_1 + \gamma_2)/2 = 3.75 \times 10^{13}\ \mathrm{s}^{-1}$ if the system is excited by the appropriate pulse.

In summary, we presented a theory for the description of optimal control of time-averaged quantities in quantum systems with relaxation, which allows one, in contrast to previous approaches, to derive an explicit differential equation for the optimal control field. We demonstrated that due to relaxation and dephasing there is an absolute upper bound for optimal control. However, we have shown that it is still possible to design an optimal shape producing the highest achievable average occupation within the constraint of pulse energy (and eventually pulse curvature). The optimal fields arising from our approach yield a considerable improvement of the controlled quantity with respect to rectangular or Gaussian pulses of the same energy and curvature. Our approach guarantees the unique optimal solution and can be used for further investigations such as, for instance, the control of the dynamics of multilevel systems by applying n resonant external fields. In this case the corresponding pulse areas $\theta_n(t)$ should satisfy a system of n coupled EL equations.

This work has been supported by the Deutsche Forschungsgemeinschaft through SFB 450.

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[12] We model the "smooth" rectangular pulse by $V(t) = A\{\exp[\sigma(-t + t_1)] + 1\}^{-1} - A\{\exp[\sigma(-t + t_2)] + 1\}^{-1}$ and the sine square pulse by $V(t) = A\sin^2(bt)$, $t \in [0, \pi/b]$.