Piecewise time-independent procedure to control two-level systems

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We present a general and mathematically simple method to control two-level systems. It is based on a piecewise time-independent procedure. We assume that some parameters of an external interaction potential can be rapidly switched at specified time instants, and then kept constant during small time intervals Δt. By properly setting the parameter values, obtained from algebraic, instead of differential or integral, equations, we can drive the time evolution of arbitrary observables. We illustrate the approach with some examples and discuss important technical aspects, relevant in real concrete situations, such as the robustness of the method to errors in the control parameter values, the influence of the switching mechanism transient times, the appropriate choice for the Δt's, and so on.

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I. INTRODUCTION

The manipulation and control of quantum processes [1,2] are among the present and near future challenges in the physical and chemical sciences [3], being relevant in both fundamental and applied aspects. A great part of the excitement in the area comes from the rapid advances in the fabrication of time-domain ultrashort laser fields, actually making it possible to directly interfere in atomic and molecular [4–8], and condensed mater [9–11] systems; drive chemical reactions [12,13]; implement qubits for quantum computation [14,15], etc.

Allied to the experimental progress, a great effort has been put forth on the theoretical side. The goal is to develop rapid and accurate methods to predict how a given quantum system can be driven to a desired time evolution by means of external "tunable" potentials like electromagnetic fields. Obviously, a certain degree of physical intuition is helpful in such tasks [16]. However, years of research show that, indeed, solid mathematical analysis is necessary to established the conditions for quantum control [17–19].

From a technical point of view, there are different ways to follow. For instance, one possibility is to use the so-called closed-loop learning approach [20,21], where a real time interaction between the system and the control device provides the necessary feedback to maintain the system on the correct track. Another is to consider the control as an inverse problem [2,16,22,23], i.e., once the desired quantum state evolution is established, to determine what is the appropriate form for an external time-dependent potential leading to such an evolution.

In this contribution we propose a general procedure for quantum control, which is based on the inverse problem protocol. Nevertheless, it differs from other known methods because of its mathematical simplicity. In fact, we avoid commonly employed techniques like maximizing functionals or solving a set of time-dependent differential equations. The "trick" is to implement the control through an external potential U, which depends on a set of parameters $\{\lambda_n\}$. The λ_n's are changed to appropriate specific values at the instants t_j, and then kept constant during time intervals Δt_j. Hence, within each Δt_j we have a time-independent problem, simple to solve. By properly setting the switching times and the parameter values, we can drive the time evolution of the system. Besides the mathematical straightforwardness, the approach allows a clear picture of all the physical features involved in the control process, which sometimes are hidden in sophisticated mathematical descriptions. We should mention that a similar idea, but used in a different context, has been applied to probe black-box two-level systems in quantum process tomography [24].

Throughout the paper the derivations, results, and examples are focused on two-level systems. We do so, first, due to their great importance, since many different phenomena are described and controlled in terms of two-level systems [5,10,24–30], even in certain instances where the problem is effectively a many-level system [31–35]. Second, because in this case we can perform the control with a minimum number of parameters, two or eventually just one. Thus, we can introduce the method without the necessity of addressing a multidimensional parameter space. The protocol, however, can equally well be used in more general situations, as we briefly comment at the end of the work.

The paper is organized as follows. In Sec. II we present the control procedure. Different examples of applications are shown in Sec. III. In Sec. IV we discuss some relevant aspects, like the robustness to errors in the control parameter values, how to properly choose the time intervals Δt_j, and the generality of the approach (i.e., that it can be used to control the time evolution of any observable, regardless of the system's initial state). Final remarks are made and conclusions drawn in Sec. V.

II. THE METHOD

A. Time evolution of a piecewise time-independent two-level system

Assume a two-level system Hamiltonian H_0, with eigenvectors $\{|0\rangle=\binom{1}{0}, |1\rangle=\binom{0}{1}\}$ and eigenvalues $\{E_0, E_1\}$. Now, consider an arbitrary interacting potential U, so that $H=H_0+U$ reads (in the H_0 eigenstate basis)

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$$H = \begin{pmatrix} E_0 & u\exp(-i\varphi) \\ u\exp(i\varphi) & E_1 \end{pmatrix}. \quad (1)$$

From (1) we see that $\langle n|U|n\rangle = 0$ $(n=0,1)$, which is the case, for instance, when the unperturbed states possess defined parity and the potential is odd, a typical situation for dipolar interactions—important in quantum control. Furthermore, the actual parameters of U, e.g., the intensity and phase of an applied laser field, are directly related to $-\infty \le u \le +\infty$ and $0 \le \varphi \le 2\pi$ through $\langle 1|U|0\rangle = u\exp(i\varphi)$. This reparametrization is very useful since it allows one to discuss the method simply in terms of u and φ.

For fixed u and φ, the solution of $H|\Psi_\pm\rangle = E_\pm|\Psi_\pm\rangle$ leads to $|\Psi_\pm\rangle = A_\pm|0\rangle + B_\pm|1\rangle$, where

$$A_\pm = \frac{\pm\sqrt{2}u\exp(-i\varphi)}{\Gamma\sqrt{1 \pm (E_1 - E_0)/\Gamma}}, \quad B_\pm = \sqrt{\frac{1 \pm (E_1 - E_0)/\Gamma}{2}},$$

$$E_\pm = \frac{(E_1 + E_0) \pm \Gamma}{2}, \quad \Gamma = \sqrt{4u^2 + (E_1 - E_0)^2}. \quad (2)$$

Under the action of H the time evolution of $|\Psi(t=t_0)\rangle = C_0(t_0)|0\rangle + C_1(t_0)|1\rangle$ is given by $[\tan(\theta) = 2u/(E_0 - E_1)$, $\omega = (E_+ - E_-)/\hbar = \Gamma/\hbar]$

$$|\Psi(t)\rangle = C_0(t)|0\rangle + C_1(t)|1\rangle,$$

$$C_1^0(t) = \left\{ \left[\exp[\mp i\omega(t-t_0)]\cos^2\left(\frac{\theta}{2}\right) + \sin^2\left(\frac{\theta}{2}\right) \right] C_1^0(t_0) \mp \frac{1}{2}\{1 - \exp[\mp i\omega(t-t_0)]\}\exp(\mp i\varphi)\sin(\theta) C_0^1(t_0) \right\} \times \exp\left(-i\frac{E_\mp}{\hbar}(t-t_0)\right). \quad (3)$$

In quantum control one constantly needs to adjust an external interaction to correctly drive $|\Psi(t)\rangle$. If this potential changes continuously in time, $H = H(t)$ and hence $|\Psi(t)\rangle$ are given by the explicit time-dependent Schrödinger equation $i\hbar(\partial/\partial t)|\Psi\rangle = H(t)|\Psi\rangle$, instead of by the above type of solution. In this case, the control protocol often demands relatively advanced theoretical analysis to determine what are the best functional forms of $U(t)$, resulting in the sought behavior for the state evolution.

Our goal here is still to control the system by properly tuning (in time) relevant potential parameters. However, we shall avoid technical complications like solving sets of time-dependent differential equations. So we propose a piecewise time-independent procedure. We assume that at determined instants t_j we can very rapidly switch u and φ to specific values u_j and φ_j, which are then kept constant during time intervals $\Delta t_{j+1} = t_{j+1} - t_j$ $(j=0,1,2,\ldots)$. "Very rapidly" means that the elapsed time for the transition $u_{j-1}, \varphi_{j-1} \to u_j, \varphi_j$, the transient time, is much shorter than any characteristic time of the system (see the next sections for details). Under this assumption, $|\Psi(t)\rangle$ for each Δt_{j+1} is given by (3), with

$$u = u_j, \quad \varphi = \varphi_j, \quad \text{and} \quad t_0 \to t_j. \quad (4)$$

The above prescription holds since the assumed sudden change in the parameter values allows one to obtain $|\Psi(t_j^+)\rangle$ from $|\Psi(t_j^-)\rangle$ just by projecting $|\Psi(t_j^-)\rangle$ onto the "new" eigenstates of the time-independent H in the interval $t_j < t < t_{j+1}$ [here, t_j^+ (t_j^-) means an instant right after (before) $t = t_j$].

Therefore, in the present framework the control protocol is to determine the sets of $\{t_1; t_2; \ldots\}$ and $\{u_1, \varphi_1; u_2, \varphi_2; \ldots\}$ leading to the appropriate $|\Psi(t)\rangle$.

B. Expectation values

Usually, the goal in controlling a quantum state is to obtain specific expectation values for an observable of interest, represented by an operator V. In other words, to have a certain $\tilde{S}$ for $S(t) = \langle\Psi(t)|V|\Psi(t)\rangle$ at $t = \tilde{t}$. We recall that $S(t)$ is restricted to $v_- \le S(t) \le v_+$, with $v_\pm$ the eigenvalues of the Hermitian V.

Suppose V is written in the basis $\{|0\rangle, |1\rangle\}$ (with v, v_0, v_1 real and $0 \le \alpha < 2\pi$)

$$V = \begin{pmatrix} v_0 & v\exp(-i\alpha) \\ v\exp(i\alpha) & v_1 \end{pmatrix}. \quad (5)$$

For a time-independent system described by (1)–(3), $S(t)$ reads

$$S(t) = (C_0^*(t) \quad C_1^*(t)) \begin{pmatrix} v_0 & v\exp(-i\alpha) \\ v\exp(i\alpha) & v_1 \end{pmatrix} \begin{pmatrix} C_0(t) \\ C_1(t) \end{pmatrix} = |C_0(t)|^2 v_0 + |C_1(t)|^2 v_1 + 2v\,\mathrm{Re}[C_0^*(t)C_1(t)\exp(-i\alpha)]. \quad (6)$$

Now, from the explicit form of $C_0(t)$ and $C_1(t)$ in Eq. (3), we see that the time dependence of the terms $|C_0(t)|^2$, $|C_1(t)|^2$, and $C_0^*(t)C_1(t)$ above is entirely given by the trigonometric functions $\sin[\omega(t-t_0)]$ and $\cos[\omega(t-t_0)]$. Therefore, we have

$$S(t) = V_0 + V_1\sin[\omega(t-t_0)] + V_2\cos[\omega(t-t_0)]. \quad (7)$$

Finally, the coefficients V_j in Eq. (7) can be obtained from a direct comparison with a lengthy but straightforward expansion of (6), resulting in $[C_0(t_0) = r_0\exp(i\sigma_0)$, $C_1(t_0) = r_1\exp(i\sigma_1)]$

$$\begin{aligned} V_0 = {} & (v_0 r_0^2 + v_1 r_1^2)[\cos^4(\theta/2) + \sin^4(\theta/2)] \\ & + (v_0 r_1^2 + v_1 r_0^2)\sin^2(\theta)/2 + (v_0 - v_1) r_0 r_1 \cos(\sigma_0 - \sigma_1 + \varphi) \\ & \times [\cos^2(\theta/2) - \sin^2(\theta/2)]\sin(\theta) \\ & + v r_0 r_1 \cos(\sigma_0 - \sigma_1 + \alpha)\sin^2(\theta) \\ & + v r_0 r_1 \cos(\sigma_0 - \sigma_1 + 2\varphi - \alpha)\sin^2(\theta) \\ & + v(r_0^2 - r_1^2)\cos(\varphi - \alpha)[\cos^2(\theta/2) - \sin^2(\theta/2)]\sin(\theta), \end{aligned}$$

$$\begin{aligned} V_1 = {} & v(r_0^2 - r_1^2)\sin(\varphi - \alpha)\sin(\theta) \\ & - (v_0 - v_1) r_0 r_1 \sin(\sigma_0 - \sigma_1 + \varphi)\sin(\theta) \\ & + 2v r_0 r_1 \sin(\sigma_0 - \sigma_1 + \alpha)[\cos^4(\theta/2) - \sin^4(\theta/2)], \end{aligned}$$

$$V_2 = (v_0 - v_1)(r_0^2 - r_1^2)\sin^2(\theta)/2 - v(v_0^2 - v_1^2)\cos(\varphi - \alpha)$$
$$\times[\cos^2(\theta/2) - \sin^2(\theta/2)]\sin(\theta)$$
$$- (v_0 - v_1)r_0 r_1 \cos(\sigma_0 - \sigma_1 + \varphi)$$
$$\times[\cos^2(\theta/2) - \sin^2(\theta/2)]\sin(\theta)$$
$$+ 2v r_0 r_1 \cos(\sigma_0 - \sigma_1 + \alpha)[\cos^4(\theta/2) + \sin^4(\theta/2)]$$
$$- v r_0 r_1 \cos(\sigma_0 - \sigma_1 + 2\varphi - \alpha)\sin^2(\theta). \quad (8)$$

Note that the $S(t)$ characteristic period of oscillation, $\tau = 2\pi/\omega$, depends only on the system's Rabi frequency $\omega = \sqrt{4u^2 + (E_1 - E_0)^2}/\hbar$. So fast changes in the observable expectation values are achieved by strong variations of u.

To find the specific φ, u, and $t = \tilde{t}$, leading to a certain $\tilde{S}$, we need to set $S(t) = \tilde{S}$ and then to solve Eq. (6). Although it may seem a little cumbersome, Eq. (6) is just a trigonometric polynomial, and hence straightforward to deal with numerically. The procedure to impose a whole specific "trajectory" for $S(t)$ is the following.

(1) To define the set of time intervals $\{\Delta t_j\}$. In practice, their actual choice is related to both the details of how fast the Hamiltonian parameters can be changed (e.g., determined by the external laser field time response); and how similar the obtained and ideal target trajectories must be for a specific application.

(2) Within each Δt_j, to choose a $\tilde{t}$ for which $S(\tilde{t})$ assumes the desired value $\tilde{S}$. The exact $\tilde{t}$ is not very important if Δt_j is small enough. In our simulations we always take $\tilde{t} = t_{j+1}^-$.

(3) To solve, at $t = \tilde{t}$, the appropriate Eq. (6) for the corresponding φ and u.

The quantum control is obtained from the calculated set of parameters φ and u. It is clear from the previous discussions that, the smaller the ratio between the transient time for each step j and the Rabi period τ, the more accurate the control. We observe that our approach contrasts with others, like those in [8,16,22] (see also [33,34] for an overview), which demand more complex numerical methods.

III. RESULTS AND EXAMPLES

Next we show the potentiality of the approach by discussing different examples. We assume in all the applications a two-level system of energies (in the atomic system of units) $E_0 = 0.323\,849$ a.u. and $E_1 = 0.323\,968$ a.u., so that $\Delta E = E_1 - E_0 = 0.119 \times 10^{-3}$ a.u., which corresponds to a natural time of $\tau_0 = 1.28$ ps. We choose these particular values just for definiteness; however, we mention that they correspond to the two excited states used to control the breaking of DH_2 molecules [5]. The quantum control is implemented by tuning the parameters u and φ as previously discussed. In laboratory conditions, for instance, our procedure could be realized by applying an external field to the system, e.g., a laser beam of variable amplitude and configured by a phase modulator [21]. The field parameters related to u and φ through the matrix element $\langle 1|U|0\rangle$ [see Sec. II A; in this case with $U = \mu\epsilon(t)$ [8,16,22,32,36],where μ is the dipole operator and $\epsilon(t)$ a piecewise time-independent electric field] should be set to the appropriate constant values during the corresponding time intervals.

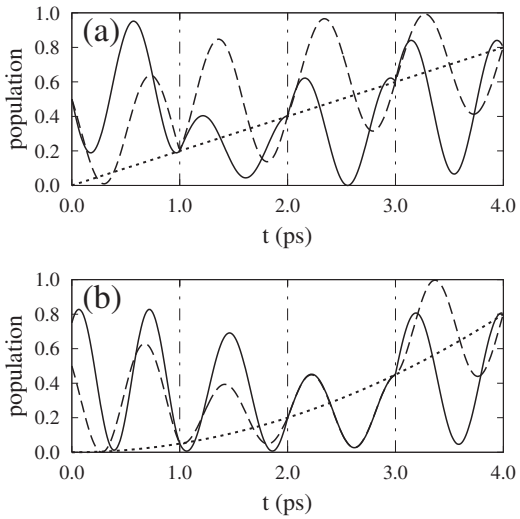

FIG. 1. The observable $S(t)$, here the population at $n=0$, controlled (solid and dashed lines) with the different sets of parameters listed in Table I. The dotted curves represent the target trajectories $S_a(t)$ (a) and $S_b(t)$ (b). In (a) the initial state is unique, but evolves according to two different sets of control parameters. In (b) the two curves represent different initial states (see main text).

A. Controlling $S(t)$ at determined t values

We start with a simple situation and discuss the control of the observable $V = |n\rangle\langle n|$ $(n = 0, 1)$ [8,37]. Thus, $S(t)$ is just the probability for the system to be found in the state n at time t. The V parameters are $v_0 = 1 - n$, $v_1 = n$, and $v = 0$. We shall drive the system evolution so that at times equal to 1, 2, 3, and 4 ps, the population of the level $n = 0$ is given by $S_a(t) = 0.2t/\Delta t$, Fig. 1(a), and $S_b(t) = 0.05(t/\Delta t)^2$, Fig. 1(b). The parameters leading to it are listed in Table I. They change only after time intervals of $\Delta t = 1$ ps. Hence, within

TABLE I. Control parameter values used in each interval of 1 ps in Fig. 1. $\bar{u} = u/(E_1 - E_0)$ and φ is in radians.

Interval	Fig. 1(a) solid line	Fig. 1(a) dashed line	Fig. 1(b) solid line	Fig. 1(b) dashed line
0–1	$\bar{u}=0.630$, $\varphi=2.649177$	$\bar{u}=0.546$, $\varphi=1.569089$	$\bar{u}=0.840$, $\varphi=0.870655$	$\bar{u}=0.630$, $\varphi=1.493502$
1–2	$\bar{u}=0.630$, $\varphi=4.896750$	$\bar{u}=0.504$, $\varphi=4.143567$	$\bar{u}=0.630$, $\varphi=0.687220$	$\bar{u}=0.630$, $\varphi=0.214842$
2–3	$\bar{u}=0.630$, $\varphi=4.048721$	$\bar{u}=0.504$, $\varphi=3.432818$	$\bar{u}=0.630$, $\varphi=1.346751$	$\bar{u}=0.630$, $\varphi=0.070247$
3-4	$\bar{u}=0.630$, $\varphi=2.567145$	$\bar{u}=0.504$, $\varphi=3.136860$	$\bar{u}=0.630$, $\varphi=0.082234$	$\bar{u}=0.630$, $\varphi=3.449264$

Δt the quantity $S(t)$ follows the dynamics of a two-level system under the action of a time-independent potential, presenting then simple Rabi oscillations.

In the first example, Fig. 1(a), we assume as the initial state $C_0=1/\sqrt{2}$ and $C_1=\exp(i\pi/4)/\sqrt{2}$ and perform the control with two different sets of u's and φ's. The existence, in certain cases, of multiple solutions is a consequence of the polynomial character of Eq. (6). It may be an advantage, since among different possibilities one can pick those that are easier to implement.

For the second example, Fig. 1(b), we have two initial states, the solid $[C_0=1/\sqrt{2},\ C_1=\exp(i\pi/4)/\sqrt{2}]$ and dashed $[C_0=\sqrt{3}/2,\ C_1=\exp(i\pi/4)/2]$ curves. Obviously, each state demands a different set of parameters (see Table I). We should observe that the quantum control can always be implemented regardless of the initial state (see the discussion in Sec. IV). For instance, the initial state $C_0=\sqrt{3}/2$, $C_1=\exp(i\pi/4)/2$, resulting in $S_b(t)$, would also lead to $S_a(t)$ at the specified t values, provided the parameters were correctly tuned.

B. Controlling $S(t)$ as a trajectory

In the previous example, the control is achieved only at certain time instants. Of course, by making the Δt_j's shorter, the number of points where $S(t)$ agrees with a given trajectory increases. However, it may seem that $S(t)$ can follow a full path only in the limit of $\Delta t_j \to 0$, i.e., for a true continuous time-dependent control. Fortunately, this is not the case. We can configure $S(t)$ reasonably close to a target trajectory by making the Δt_j's small but finite, as illustrated next. We leave to Sec. IV a more detailed discussion about how to properly choose such Δt_j intervals.

Unless otherwise mentioned, hereafter we set $\Delta t_j=\Delta t=100$ fs [38]. For V we assume (in arbitrary units) $v_0=1$, $v_1=3$, $v=\sqrt{3}$, and $\alpha=\pi/2$; thus $v_-=0$ and $v_+=4$. Once $\tau_0/\Delta t=12.8$, the time intervals are much shorter than the unperturbed system ($u=0$) natural time. Thus, the obtained $S(t)$ can be regarded as a fairly good approximation for a continuous trajectory in the τ_0 time scale. As the initial state we take the same one used in Fig. 1(a).

We begin with a somewhat critical shape for $S(t)$, the staircaselike function of Fig. 2(a). From Eq. (6) we have $dS(t)/dt=\{V_1\cos[\omega(t-t_0)]-V_2\sin[\omega(t-t_0)]\}\omega$. Since the term between braces is bounded, abrupt changes of S are associated with $\omega=\sqrt{4u^2+(E_1-E_0)^2}/\hbar$. So, whenever we seek sharp variations of S (e.g., at the edges of the staircaselike trajectory), we need large Δu's.

For $S(t<2.5\ \text{ps})=3$, $S(2.5\le t<7.5\ \text{ps})=0.5$, $S(7.5\le t<12.5\ \text{ps})=3.5$, and $S(t\ge 12.5\ \text{ps})=1$, we perform the control in two different ways: (i) changing both u and φ; and (ii) keeping u constant and changing φ. For (i) we show in Fig. 2 the values of u and φ and the resulting $S(t)$. Observe that, each time S presents a jump, Fig. 2(a), there are more pronounced corresponding variations of u, Fig. 2(b), and of φ, Fig. 2(c).

Such strong changes of u in short times (eventually necessary, e.g., to control molecular processes in the presence of collisions [39]) may pose technical difficulties in practice. Note that u has units of energy and, for the case of an external field, u is directly related to its intensity. Thus, in our example the method would demand fast variations of the field amplitude. So another possibility is to fix u at a value never inferior to the minimum necessary to promote the largest $\Delta S/\Delta t$ transition and then to control only the phase parameter φ. This is shown in Fig. 3(a) (continuous curve), where we set $u/(E_1-E_0)=2.94$, i.e., a value 1.52 times larger than the maximum u in Fig. 2(b). In Fig. 3(c) we display $S(t)$ around a jump region. We observe some (not strong) oscillations for $S(t)$ before reaching the aimed constant value of 0.5, a behavior not seen in Fig. 2(a). This loss of quality in the control during short times is the price to be paid for using just one, namely φ, instead of two control parameters. We mention that a third possibility, to fix φ and change u, has the already mentioned drawback and gives overall poor results.

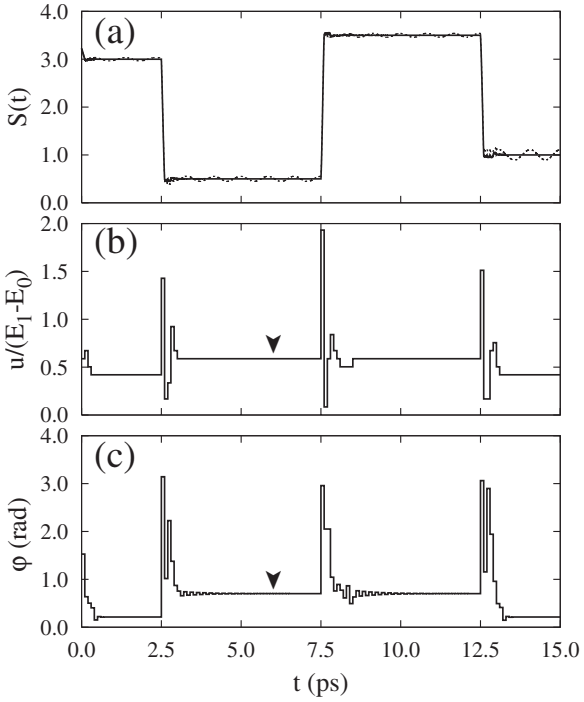

FIG. 2. (a) $S(t)$ configured as a staircaselike function by the control parameters (b) u and (c) φ. The dotted curve represents $S(t)$ for small random errors for the set $\{t_j\}$.

For a smooth $S(t)$, on the other hand, the conditions for control are more favorable. Suppose the target trajectory $S(t)=S_0+S_1\exp[-\sigma(t-T_1)^2]+S_2\exp[-\sigma(t-T_2)^2]$, i.e., basically two Gaussians centered at different instants in time. Figure 4(a) (continuous curve) displays the result for $T_1=2.5$ ps, $T_2=10$ ps, $S_0=3.0$, $S_1=-2.9$, $S_2=0.9$, and $\sigma=1.54\ \text{ps}^{-2}$. We keep $u/(E_1-E_0)=1.1$ constant and change only φ, Fig. 4(b). Note that we can set a lower value for u, compared to that in Fig. 3, even thought here the maximum and minimum of $S(t)$ are very close to the allowed extremes of $v_-=0$ and $v_+=4$. This is possible because there are no abrupt variations for $S(t)$. Furthermore, we do not have the short spurious oscillations observed in the previous case.

From Figs. 2–4, we see that during time intervals ΔT where $S(t)=\tilde{S}$ is constant, the control parameters also tend to

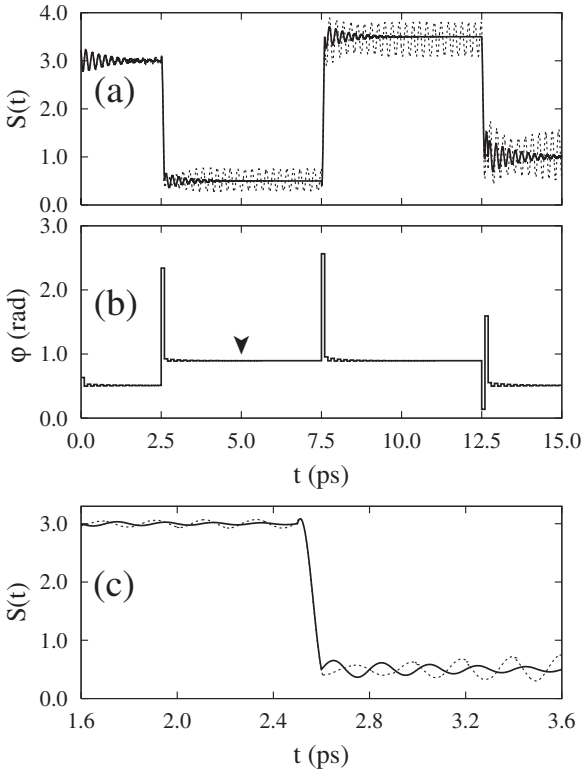

FIG. 3. (a) $S(t)$ (continuous curve) configured as in Fig. 2, but for a fixed $u/(E_1-E_0)=2.94$ and (b) a varying φ. (c) The continuous curve shows $S(t)$ around a jump region. In (a) and (c) the dotted curves represent $S(t)$ for small random deviations for the set $\{t_j\}$.

fixed values (the arrows in the figures indicate some examples). To understand this fact, note that a trivial way to get $S(t)=\tilde{S}$ is to have $|\Psi(t)\rangle=\exp[-i(E_a/\hbar)t]|\Psi_a\rangle$, with $|\Psi_a\rangle$ the eigenstate of $H(u,\varphi)$ for which $\langle\Psi_a|V|\Psi_a\rangle$ is exactly $\tilde{S}$. Hence, in these situations the method (within ΔT) progressively selects the parameters until $|\Psi(t)\rangle\rightarrow|\Psi_a\rangle$ and then locks in their values. We illustrate this by showing in Fig. 5

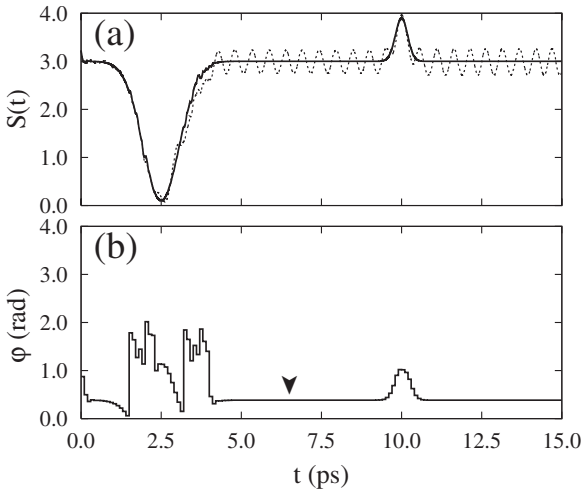

FIG. 4. (a) $S(t)$ for a fixed $u/(E_1-E_0)=1.1$ (continuous curve) and (b) a varying φ. The dotted curve represents $S(t)$ for small random errors for the set $\{t_j\}$.

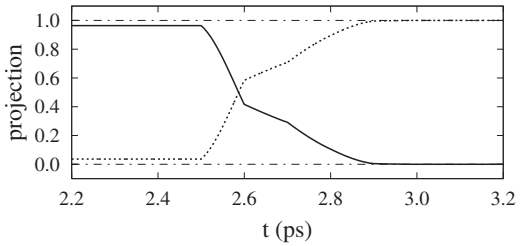

FIG. 5. Probability projections $|\langle\Psi(t)|\Psi_b\rangle|^2$ and $|\langle\Psi(t)|\Psi_a\rangle|^2$ (dotted) for $2.2<t<3.2$ ps of Fig. 2(a).

the projections $|\langle\Psi(t)|\Psi_a\rangle|^2$ and $|\langle\Psi(t)|\Psi_b\rangle|^2$ (with $|\Psi_b\rangle$ the corresponding second eigenstate of H) around $t=2.5$ ps for the example of Fig. 2. $|\Psi_a\rangle$ is the state to which $|\Psi(t)\rangle$ evolves so as to lead to $S(t)=0.5$. This transition process starts at $t=2.5$ ps, finishing 0.38 ps later. At $t=2.88$ ps, u and φ also reach the appropriate fixed values [marked by arrows in Figs. 2(b) and 2(c)]. We should mention that it is just an artifact of this particular example $|\Psi_b\rangle$ to be close to a stationary state, resulting in $S(t<2.5\ \text{ps})=3$.

IV. DISCUSSION

After presenting and exemplifying the method, we now pass to discuss some of its general technical aspects.

A. The control sensitivity to errors in the parameter values

An important point is the stability of the approach to small errors in the control parameter values. So a first issue relevant to estimating how accurate must be the switching mechanics concerns the time instants t_j at which the parameters must be changed. In our calculations we suppose the sudden approximation [see the discussion before Eq. (4)]. Thus, a delay for the transition $u_j,\varphi_j\rightarrow u_{j+1},\varphi_{j+1}$ may introduce some deviations from the target trajectory. Hence, quantifying how a "detuning" in the t_j's affects the quality of the control is also a way to probe the role played by finite transient times, which in a real situation can be short but not zero.

We test this by introducing errors δt_j for the t_j's. Each δt_j is randomly sorted within $(-\Delta t/16.66,+\Delta t/16.66)$, so the error interval width corresponds to 12% of the time intervals $\Delta t=100$ fs used in the examples. The dotted curves in Figs. 2(a), 3(a), 3(c), and 4(a) display $S(t)$ for the correct parameters tuned at times $t_j+\delta t_j$. Comparing Figs. 2(a) and 3(a), where in the former (latter) the control is performed with u and φ (just φ), we see that, as expected, the control quality is less affected by errors when both parameters are used. We also observe better results in Fig. 4(a) than in Fig. 3(a), indicating that smooth $S(t)$'s are more robust to errors.

A second situation is for small errors in the values of the control parameters themselves. We simulate it by considering $u_j+\delta u_j$ and $\varphi_j+\delta\varphi_j$, where δu_j and $\delta\varphi_j$ are taken randomly within $(-u_j/20,u_j/20)$ and $(-\varphi_j/20,\varphi_j/20)$. Hence, the error interval widths correspond to 10% of the parameters correct values u_j and φ_j. In Figs. 6(a) and 6(c) we consider the control of both u and φ and in Figs. 6(b) and 6(d) only of φ.

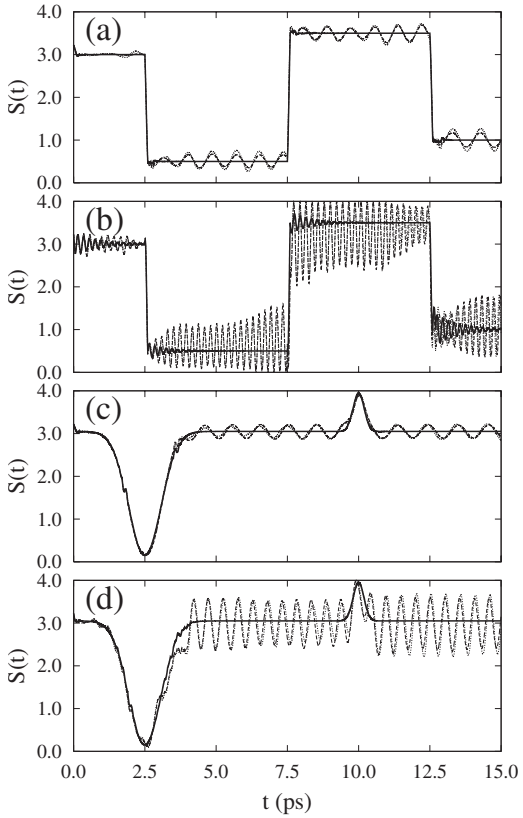

FIG. 6. $S(t)$ trajectories of the previous examples for the parameters switched at the correct t_j's and no errors for u and φ (continuous); random errors for both φ and u (dotted); and random errors only for φ (dashed). In (a) and (c) u and φ are tuned, whereas in (b) and (d) just φ is tuned.

All figures display $S(t)$ for the control performed with the exact u and φ; errors only for φ; and errors for both u and φ. We see that, as before, by controlling two parameters the deviations are much less pronounced. In fact, the worst case, especially for $5<t<11$ ps, is that of Fig. 6(b) (dashed and dotted curves). However, it is a critical example: a nonsmooth $S(t)$, controlled with just a single parameter, φ, for which there are random errors.

B. Generality of the method regarding the initial state $|\Psi(t_0)\rangle$ and the observable V

In the explicit applications we discuss only two observables V and a few shapes for $S(t)$. However, in testing the method, we have been able to control different paths $S(t)$ for many different V's, always with good numerical accuracy. So a natural question is if the present approach allows us to drive $S(t)$ to any desired trajectory [under the restriction $v_- \leq S(t) \leq v_+$], regardless of the operator V and the problem initial state $|\Psi(t_0)\rangle$. Due to our specific type of protocol, this is equivalent to asking if, by setting the Hamiltonian parameters and choosing an appropriate instant $t_0 \leq \tilde{t} \leq t_0+\Delta t$, we can make $S(t=\tilde{t})$ assume any value $\tilde{S}$. We show in the Appendix that this is the case; thus the method can be used to control two-level systems in arbitrary contexts.

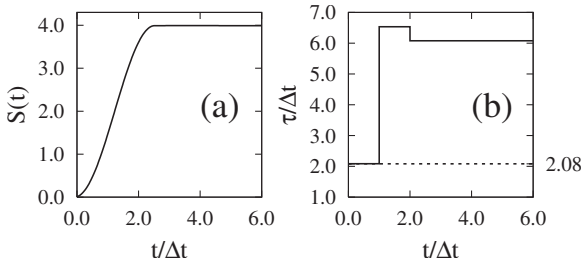

FIG. 7. (a) Control of $S(t)$, aimed to be the constant $\tilde{S}=3.99$, by tuning both u and φ. Here $\Delta t=100$ fs and the initial state is such that $\langle\Psi(t_0=0)|V|\Psi(t_0=0)\rangle=0.01$. (b) Ratio $\tau/\Delta t$ as function of time.

To demonstrate the generality of the method's building block procedure, i.e., to perform a parametric local control at each time interval Δt_j, we have considered in the Appendix the most unfavorable situation. We have found that, to control a completely general case, we should assume $\Delta t \geq \tau$. In most examples throughout the work we set $\Delta t=\tau_0/12.8$ (recall that $\tau_0 \geq \tau$), apparently in contradiction with this condition. We observe that in the Figs. 2–4, the chosen initial state $|\Psi(t_0)\rangle$ already gives $S(t_0)$ around the desired value $S(\tilde{t})=\tilde{S}$ for $t_0 \leq \tilde{t} \leq t_0+\Delta t$. Actually, the figures reflect this fact since the transients for $t \approx t_0=0$ are almost imperceptible, e.g., in Fig. 2(a) seen just as a very small spike for $S(t \approx 0)$. The important point is that, when controlling the time evolution of $S(t)$, if for each time interval Δt_{j+1} we already start with a state $|\Psi(t_j)\rangle$ close to the target state $|\Psi(\tilde{t})\rangle$, then the conditions in the Appendix can be relaxed. Obviously, such "closeness" of states at each t_j is automatically satisfied when we perform the control for a smooth $S(t)$. In these cases there are no important restrictions for the values of Δt_j, which can be small, limited only by external aspects, like the control tuning mechanisms.

However, if we cannot select the initial states [40] and also $S(t)$ has rapid variations or even jumps as in Figs. 2 and 3, we still can use short time intervals, provided u can assume large values. The method then naturally selects u so as to make the ratio $\tau/\Delta t$ appropriate for the control. We illustrate it with a simple example, assuming $S(t)$ to be the numerical constant 3.99, i.e., almost the maximum allowed value of $v_+=4$. We set $\Delta t=100$ fs and perform the control, shown in Fig. 7(a), using both u and φ. As the initial state we take $C_0=0.840\,745$ and $C_1=0.541\,431\exp(-i\pi/2)$; thus $\langle\Psi(t_0)|V|\Psi(t_0)\rangle=0.01$ is very close to the lowest possible value of $v_-=0$. In the Appendix we show analytically that, to obtain $\tilde{S}=v_+$ within Δt, from an initial state leading to $S(t_0)=v_-$, we need $\tau/\Delta t=2$. Figure 7(b), displaying $\tau/\Delta t$ as function of time, is in agreement with such results. Indeed, during the first control step $t/\Delta t<1$, we have $u/\Delta E=3.02$, a high value for u, making τ to be short enough to meet the time condition, here of $\tau/\Delta t=2.08$. Note, furthermore, that for $t/\Delta t>2$, u has already decreased to $u \approx \Delta E$.

Finally, if we cannot freely change u or even set it to a high constant value as in Fig. 3, then we can make the Δt_j small in the smooth regions of $S(t)$, but need to assume larger time intervals at the beginning of the control process

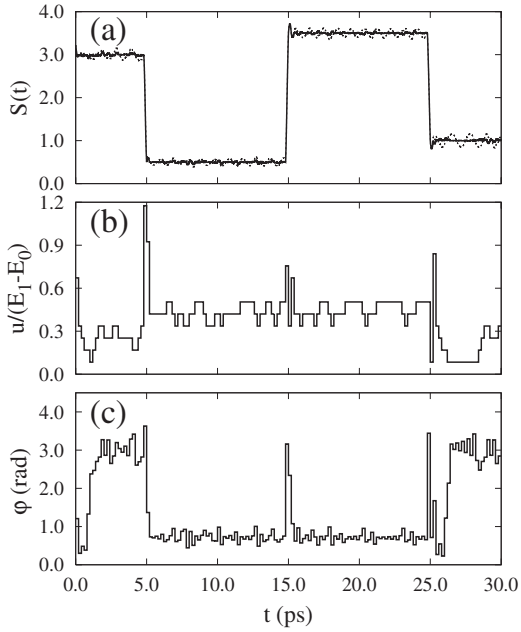

FIG. 8. Same as in Fig. 2, but for $\Delta t=200$ fs.

and around the strong variation regions of the target trajectory.

C. Connection with laser field parameters

The proposed scheme is based on a control of "bare" parameters. So it is of fundamental importance to know how they are related to actual external potential parameters, e.g., of a laser field, produced in laboratory conditions. Below we address two points related to this issue.

The first is that the method assumes a fast transient time τ_{sw} for the parameter switching. Recently, a quantum control experiment has been reported [38] with a field pulse of $\tau_{sw}=100$ fs. In fact, the present state of the art in femtosecond pulse shaping (see, for instance, Refs. [41]) implies that as "sudden" one should consider changes typically taking place within this time scale. For our previous examples and a laser field of $\tau_{sw}=100$ fs, we find that $\tau_{sw}/\tau_0 \approx 0.08$ is relatively small, therefore satisfying the method's main assumption [see the discussion just before Eq. (4) in Sec. II A].

Nevertheless, a possible difficulty for a concrete realization of the discussed examples is the assumed time step of $\Delta t=100$ fs, for which the desired condition of $\tau_{sw}<\Delta t$ does not hold. In this case a continuous variation of the potential parameters within τ_{sw} has an effect similar to that of introducing errors in the values of the t_j's, exemplified in Sec. IV A. This probably would lead to a less accurate control than, for instance, those seen as continuous lines in Figs. 2(a) and 3(a). A way to overcome the problem is to use longer Δt's. In Fig. 8 we show the same situation as in Fig. 2, but for $\Delta t=200$ fs. The control is still very good and even errors in the t_j values [dotted curve in Fig. 8(a)] do not introduce strong perturbations. Note that, since the error interval width (12% of $\Delta t=200$ fs) is about $\tau_{sw}/4=25$ fs, we may expect to have experimentally a reasonable good control. Unfortunately, by increasing Δt the protocol accuracy becomes more sensitive to deviations in u_j and φ_j. Observe in Fig. 8 that, during the control process, the changes in the parameter values occur more frequently, if compared to Fig. 2. Indeed, in Fig. 8 we do not see such large plateaus as in Fig. 2. Hence, the accumulated error (due to eventual deviations δu_j and $\delta\varphi_j$) will be more pronounced. Thus, in a real situation there is a compromise between how fast the transients are and how accurately the parameter values can be tuned. But, of course, with advances in the fabrication of lasers with shorter τ_{sw}, this feature loses part of its relevance.

The second point is related to the laser amplitudes necessary in concrete applications. An important aspect is if we are dealing with an N-level systems, but the control can be performed by manipulating just two of them [31–35]. In such cases, the field peak values will determine the validity of a two-level description for the quantum control of an N-level systems. Obviously, this is not a concern when we have effectively only two levels, e.g., no other levels are accessible under the experimental conditions [10,24,27] (for a more technical analysis see also Refs. [42]).

To estimate the magnitudes of the fields, we recall the discussion in the beginning of Sec. III, and, as usual in the dipolar approximation, neglect possible spatial variations of ϵ. We can write then

$$u = |\langle 1|U|0\rangle| = |\langle 1|\mu\epsilon|0\rangle| \approx |\epsilon|\mu_{10}, \tag{9}$$

where μ_{10} is the modulus of the dipole matrix element. Therefore, as a function of time the field amplitude has exactly the same profile [e.g., like those seen in Figs. 2(b) and 3(b)] as u. Moreover, we observe that in many examples [9,35,43] the quantity μ_{10} is typically of the order of 1 a.u. So in atomic units ϵ_{max} is numerically around u_{max}. For explicit values consider Fig. 2(b). Since we have that $u_{max}/\Delta E=2$, then $\epsilon_{max}=2.3\times10^{-4}$ a.u. For comparison, in the original paper controlling the DH_2 molecule [5], the maximum control field peak is 1.9×10^{-4} a.u.; hence our method demands similar field amplitudes.

Finally, in certain instances one may wish to decrease the laser intensities. In our model it can be done by allowing larger time steps, as directly observed in Fig. 8, where we use $\Delta t=200$ fs. Note that $u_{max}/\Delta E=1.2$, in contrast to $u_{max}/\Delta E=2$ of Fig. 2(b), i.e., a diminishing of 40%. Thus, the method allows a certain flexibility in setting the maximum field peaks, provided we can afford to lose some accuracy in the control process.

V. REMARKS AND CONCLUSION

We have proposed a very simple method to control the time evolution of quantum states and consequently the expectation values of relevant operators. The main idea is to change, only at appropriate time instants, the parameter values of an external interaction. So we are left with a successive set of time-independent systems. Of course, the idea of using piecewise analytic solutions to solve either time-independent [44] or time-dependent [45] Schrödinger equations, in the context of atomic and molecular physics, is not new. However, only recently has it been considered in quantum control, e.g., in quantum process tomography [24] and in

tailoring average moments of wave packets [46]. Here we apply this approach to a full tracking control.

Here the solution of the inverse problem, i.e., to obtain the external potential (e.g., an electric field) leading to the desired time evolution, is relatively simple since we need to solve a set of time-independent algebraic, instead of differential or integral, equations. Regarding this observation, a few comments comparing ours to more conventional schemes are in order. For this, we briefly summarize the optimal and inverse (or tracking) control methods below (for an overview of different methods see, e.g., Ref. [18]).

The optimal method is based on a functional J_1 involving the wave function and the target time evolution, e.g., $S(t)$. The minimization of this functional results in the field for the sought control: to maximize the expectation value of a determined operator (within a certain time interval), to make the system wave function follow a predefined evolution [8], etc. Usually, a second functional J_2 (a penalty cost term) is also introduced. Its role is to avoid solutions for which the field presents undesired features like divergences. The joint minimization of J_1+J_2 represents then a balance between finding the field for the exact target control and keeping the field fluence down. The method's drawback, however, is its demand for computationally intensive iterative optimization procedures [16] (although more efficient algorithms are being reported in the literature [34,47]).

The inverse method follows the standard viewpoint of inverse problems. The output $S(t)$ is specified and the input, the external potential, must be found. A differential equation for $S(t)$, whose order depends on the characteristics of the observable operator V and the interaction potential U [16], is obtained. From it we can write down the desired U in terms of the time derivatives of $S(t)=\langle\Psi(t)|V|\Psi(t)\rangle$. But U appears explicitly in the Schrödinger equation, which determines the evolution of $|\Psi(t)\rangle$. Thus, we end up with a set of coupled nonlinear differential equations and numerical methods are necessary. One important aspect is that one imposes only the target evolution of $S(t)$. No restrictions are set on the external potential (the electric field). Hence, in some instances the method gives singular solutions. In such cases, allied to the inverse procedure, a special treatment for handling the field divergences must be considered [22].

From the above, the mathematical simplicity of the piecewise time-independent parametric control becomes obvious. It deals with linear algebraic equations and does not invoke a recursive procedure in time, even when extended to N-level systems (see below). Moreover, the method always leads to regular solutions. A simple and intuitive way to understand this last fact is to realize that imposing a trajectory $S(t)$ along a full continuous time interval can be too restrictive, demanding singular external fields. In the present scheme we relax this imposition. We set the control only at a single instant $\tilde{t}$ within each time interval Δt_j. As already discussed, for appropriate time scales the obtained $S(t)$ can fairly enough be considered a continuous path. Since we have a certain flexibility to choose the time parameter values, we always are able to find finite solutions for Eq. (6).

As a last comparison, we shall also cite the local control theory (LCT), developed by Tannor, Kosloff and others [32,36,48]. In this method, recently reviewed in Ref. [49], the external perturbation on a system is determined within each time instant [49], so as to influence a target observable. Therefore, our method and the LCT have the same common feature of not demanding information on later time dynamics—through forward-backward iteration—typical of global optimization approaches. However, technically they are different. In the local control theory one considers time-dependent external pulses and then writes down the problem Schrödinger equation. From it one derives rate relations, given how a level energy or its population varies in time. By properly choosing the external field at each t (a function of the time-dependent wave equation at the same t), it is possible to change monotonically the energy of certain states, lock the population of a set of levels, cool the system internal degrees of freedom (e.g., vibrational modes of molecules), etc. [49]. Our method strikes in a different direction. First, we focus and control the relevant expectation values directly, rather than their time variations. So our method may be easier to apply if one needs a specific shape for $S(t)$, whereas the LCT may be more appropriate if only monotonic variations (or no variations at all) of the observables are targeted. Second, our method requires a different character for the external field, which in the LCT is basically a train of pulses. So the experimental setup necessary for each implementation might be different.

We have discussed the method in the context of two-level systems, interesting for their own sake. However, the approach can be extended to a more general situation of N levels. In this case the main equation for the control is a direct generalization (but of course a much more complicated version) of Eq. (6). Furthermore, if for a full control a multiparameter potential $U(\lambda_1,\ldots,\lambda_2)$ is necessary, then the analysis of how generally we can apply the method and what are the conditions for the control requires extending the type of discussions in the Appendix to a multidimensional parameter space. It is feasible, nevertheless, since there are different efficient techniques to classify the allowed dynamical outcomes of a system in terms of its parameter "landscape," used, for instance, in such complex systems as biological ones [50]. Similar techniques are already employed in closed-loop learning quantum control for as many as 130 independent parameters (see, e.g., [51] and references therein).

Finally, we observe that we have used the fact that a two-level Hamiltonian is exactly diagonalized [cf. Eqs. (2) and (3)] to obtain an explicit analytical dependence of $S(t)$ on the external potential parameters. So all the numerical work is reduced to solving $S(t)=\tilde{S}$. This is not the case for N-level systems (actually, for three levels a closed solution is also possible [52]), for which an $N\times N$ Hermitian matrix, analogous to Eq. (1), must be numerically diagonalized. In this case, the piecewise time-independent parametric control method is implemented through two steps. For each time interval Δt_j, we first set the parameter values, diagonalize the Hamiltonian, and then find numerically the N coefficients $C_n(t)$ to be inserted into the appropriate extension of Eq. (6). Second, we determine within Δt_j how close $|S(t)-\tilde{S}|$ is to zero, thus deciding if new parameter values (chosen based on

the numerical evaluation of $\nabla_\lambda S$) should be tested again for that Δt_j. Since for, let us say, 100 states a numerical calculation of H is really fast in any personal computer, our method is still easy to handle, demanding relatively low computational time for tens of levels.

Presently we are applying our procedure to control three-[53] and four-[54] level systems. The results will be reported in due course.

ACKNOWLEDGMENTS

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APPENDIX

Here we shall demonstrate that for an arbitrary V, if we correctly fix the parameters u and φ in the Hamiltonian H of Eq. (1) and properly set a $t=\tilde{t}$ within the time interval $(t_0, t_0+\Delta t)$, we can obtain any specified value $v_- \leq \tilde{S} \leq v_+$ for $S(t=\tilde{t})$, regardless of the initial state $|\Psi(t_0)\rangle$.

We start by observing that, under the action of H and for $t_0 < t < t_0+\Delta t$, the time evolution of the initial state $|\Psi(t_0)\rangle = |\Psi_0\rangle$ is given by

$$|\Psi(t)\rangle = \exp\left(-\frac{i}{\hbar}H(t-t_0)\right)|\Psi_0\rangle. \tag{A1}$$

Within Δt the eigenstates of H, $\{|\pm\rangle\}$, are a good basis for the problem. Nevertheless, we always can expand $|\Psi(t)\rangle$ in the basis of eigenstates of V, $\{|v_\pm\rangle\}$. In particular, for $t=\tilde{t}$, suppose $|\Psi(\tilde{t})\rangle = c_-|v_-\rangle + c_+|v_+\rangle$. Since $|c_-|^2+|c_+|^2=1$, we have $\tilde{S}=\rho(\tilde{t})v_+ + [1-\rho(\tilde{t})]v_-$, with $0 \leq \rho(\tilde{t}) = |c_+|^2 \leq 1$. Therefore, if somehow we can make $\rho(\tilde{t})$ assume any value between 0 and 1, we can set $\tilde{S}$ as we wish.

Let us define

$$\rho(t) = |\langle v_+|\Psi(t)\rangle|^2, \tag{A2}$$

so that $\rho(\tilde{t}) = |c_+|^2$. Next, up to unimportant overall phases we can write $(0 \leq a, b \leq 1,\ 0 \leq \alpha, \beta < 2\pi)$

$$|v_+\rangle = \sqrt{a}\exp(i\alpha)|+\rangle + \sqrt{1-a}|-\rangle,$$

$$|\Psi_0\rangle = \sqrt{b}\exp(i\beta)|+\rangle + \sqrt{1-b}|-\rangle. \tag{A3}$$

Thus, defining $\phi = (t-t_0)\omega - (\beta-\alpha)$, from Eqs. (A1) and (A3) we have for Eq. (A2)

$$\rho(t) = 2ab + 1 - (a+b) + 2\sqrt{ab(1-a)(1-b)}\cos(\phi). \tag{A4}$$

To state that we are able to perform the quantum control for any possible observable V and initial state is equivalent to stating that we can control ρ assuming $|v_+\rangle$ and $|\Psi_0\rangle$ are completely arbitrary vectors.

Now, by inspecting Eq. (2), one quickly realizes that, first, if we have total freedom to choose u and φ, then the eigenstates of H can be made equal (up to global phases) to any vector of the two-level Hilbert space. Hence, one of the two projection parameters in (A4), say $a=|\langle +|v_+\rangle|^2$, can be set to any allowed value. Second, if we properly choose $t=\tilde{t}$, so that $\phi(\tilde{t})$ assumes any value within $[0, \pi]$, then the cosine function in Eq. (A4) can span the whole numerical interval $[-1, 1]$. This is possible whenever $\Delta t\omega \geq \pi+\gamma$, for γ given by $\beta-\alpha$ (mod π). So a sufficient condition to fully control ϕ, independent of $\beta-\alpha$, is to have $\Delta t \geq \tau$. However, more favorable phase differences may lead to shorter time intervals (see below).

Finally, it is straightforward to show that Eq. (A4), for an arbitrary b, can assume any value between 0 and 1, provided we can freely vary $\cos(\phi)$ and a, which is always possible from the above results. Thus, the generality of the control procedure is established.

As an application we particularize Eq. (A4) to a situation for which the control is somewhat extreme. Suppose we target $\tilde{S}=v_+$, so that $\rho(\tilde{t})=1$. Obviously, the most "difficult" initial state in this case is $|\Psi(t_0)\rangle = |v_-\rangle$, where then $b=1-a$ and $\beta=\alpha+\pi$. Thus, Eq. (A4) reduces to

$$\rho(t) = 4a(1-a)\sin^2[(t-t_0)\omega/2]. \tag{A5}$$

Imposing $\rho(\tilde{t})=1$ above, we find $a=1/2$, $\tilde{t}=t_0+\Delta t$, and a minimum value for the time interval of $\Delta t = \tau/2$.

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