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Kinematical bounds on optimization of observables for quantum systems

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Upper and lower kinematical bounds for the expectation values of arbitrary observables of driven quantum systems in mixed states are derived, and criteria for their attainability established. The results are applied to the problem of maximizing the energy of a laser-driven four-level Morse oscillator model for HF, as well as a four-level harmonic-oscillator model. [S1050-2947(98)01510-8]

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I. INTRODUCTION

Recent advances in laser technology have opened up new possibilities for laser control of phenomena in the quantum regime, such as molecular dynamics or chemical reaction dynamics [1]. The limited success of initially advocated schemes based largely on physical intuition has prompted researchers in recent years to systematically study these systems using control theory. This is also the subject of this paper. We generalize the results in [2] by establishing kinematical bounds for the expectation values of arbitrary observables of driven quantum-mechanical systems. These new results are then applied to the problem of maximizing the energy for a four-level HF (hydrogen fluoride) model and a four-level harmonic-oscillator model.

II. MATHEMATICAL SETUP

We consider a quantum-mechanical system whose state space $\mathcal{H}$ is a separable Hilbert space. Any mixed state of the system can be represented by a density operator $\hat{\rho}(t)$ (acting on $\mathcal{H}$) with eigenvalue decomposition

$$\hat{\rho}(t)=\sum_k w_k|\Psi_k(t)\rangle\langle\Psi_k(t)|, \tag{1}$$

where w_k are the eigenvalues, and $|\Psi_k(t)\rangle$ the corresponding normalized eigenstates of $\hat{\rho}(t)$, which evolve in time according to the time-dependent Schrödinger equation. The eigenvalues satisfy

$$0\leqslant w_k\leqslant 1\forall\ \ k\ \ \text{and}\ \ \sum_k w_k=1, \tag{2}$$

i.e., they can be ordered in a (possibly finite) nonincreasing sequence:

$$w_1\geqslant w_2\geqslant\cdots\geqslant w_k\geqslant\cdots\geqslant 0.$$

Unless otherwise specified, we will use the word ''state'' in the following to refer to a mixed quantum state represented by a density operator $\hat{\rho}$ acting on $\mathcal{H}$. Let $\hat{\rho}(t_i)$ describe the initial state of the system. If the system is left alone, its state will evolve with time according to the quantum Liouville equation

$$\frac{\partial}{\partial t}\hat{\rho}(t)=-\frac{i}{\hbar}[\hat{H}_0,\hat{\rho}(t)], \tag{3}$$

where $\hat{H}_0$ is the Hamiltonian of the system.

The goal of controlling the system is, in general, to drive it in such a way as to maximize or minimize the expectation value

$$\langle\hat{A}(t)\rangle=\mathrm{Tr}[\hat{A}\hat{\rho}(t)] \tag{4}$$

of a given observable $\hat{A}$, e.g., the energy, at some target time $t=t_f$.

When the system is driven, i.e., subject to external control, then the time-evolution equation must be modified by adding a perturbation term $\hat{H}_1(\mathbf{f}(t))$ to the Hamiltonian $\hat{H}_0$. Hence, the controlled system obeys the quantum Liouville equation

$$\frac{\partial}{\partial t}\hat{\rho}(t)=-\frac{i}{\hbar}[\hat{H}(\mathbf{f}(t)),\hat{\rho}(t)], \tag{5}$$

where

$$\hat{H}(\mathbf{f}(t))=\hat{H}_0+\hat{H}_1(\mathbf{f}(t)) \tag{6}$$

is the total Hamiltonian, and $\mathbf{f}(t)$ (which may be multidimensional) represents a control function belonging to a class of

admissible controls $\mathcal{U}$. The minimum requirement for a control function to be admissible is that it be bounded and measurable. However, the class of admissible controls may be restricted further to reflect physical or experimental constraints.

In laser-driven applications, such as laser control of molecular dynamics, $\hat{H}_1$ is usually the dot product of an external laser field, which serves as a control function, and the electric dipole operator.

A given initial state $\hat{\rho}(t_i)$ can only evolve into states $\hat{\rho}(t_f)$ that are related to $\hat{\rho}(t_i)$ by

$$\hat{\rho}(t_f)=\hat{U}(t_f,t_i)\hat{\rho}(t_i)\hat{U}^{\dagger}(t_f,t_i), \tag{7}$$

where $\hat{U}(t,t_i)$ is the time-evolution operator of the control system, which satisfies the Schrödinger equation

$$\frac{\partial}{\partial t}\,\hat{U}(t,t_i)=-\frac{i}{\hbar}\,\hat{H}(\mathbf{f}(t))\hat{U}(t;t_i). \tag{8}$$

While the concrete dynamical law obeyed by $\hat{U}(t,t_i)$ depends on the control system, and thus an input function $\mathbf{f}(t)$, which is to be determined, it is clear that $\hat{U}(t_f,t_i)$ must be a unitary operator for any control system. Therefore, only target states $\hat{\rho}(t_f)$ that are related to $\hat{\rho}(t_i)$ as Eq. (7), for some unitary operator $\hat{U}(t_f,t_i)$, are *kinematically attainable*.

Kinematical attainability does not imply *dynamical reachability*, i.e., that there exists an admissible control-trajectory pair that would transfer the system from the initial state $\hat{\rho}(t_i)$ to a target state $\hat{\rho}(t_f)$ for which the expectation value of the observable assumes its (kinematical) maximum or minimum. Nevertheless, kinematical bounds impose restrictions on the dynamical evolution of expectation values of observables, and thus knowing these bounds, as well as the set of target states S_+ or S_- for which the maximum or minimum is realized, is useful.

If one tries to solve the optimal control problem of maximizing $\langle\hat{A}(t_f)\rangle$ numerically, using, e.g., a nonlinear eigensystem method as described in [3], then knowing the exact kinematical maximum, and for which states it is attained, provides a way to check the quality of the numerical solution for the control field: One (numerically) computes the trajectories as well as the time-dependent expectation value of the observable for the (numerically) obtained optimal control field. If the kinematical bound is dynamically realizable then the ratio of the computed value of $\langle\hat{A}(t_f)\rangle$ and the kinematical upper bound should approach 1, and $\hat{\rho}(t_f)$ should approach a point in S_+.

III. KINEMATICAL BOUNDS

Under certain circumstances, kinematical bounds similar to those we shall derive for finite-dimensional systems can be established for infinite-dimensional systems as well. However, since for practical computational purposes it is necessary to truncate the system to finitely many levels, we shall in the following only consider the case of a system whose state space is a Hilbert space $\mathcal{H}$ of dimension $N<\infty$.

Any real observable of a quantum-mechanical system is represented by a Hermitian operator $\hat{A}$ on $\mathcal{H}$. In the special case that $\hat{A}$ is a projector onto a subspace, the following result was proved in [2]:

Theorem 1: If $\hat{A}$ is a projector onto a subspace S of dimension d then

$$\langle\hat{A}(t)\rangle\leqslant\sum_{k=1}^{d} w_k\forall t, \tag{9}$$

and if $w_{d+1}<w_1$ then we have equality exactly if

$$\mathrm{span}_{k=1,\ldots,d}|\Psi_k(t)\rangle=S. \tag{10}$$

Furthermore, if the state space $\mathcal{H}$ has finite dimension N then

$$\langle\hat{A}(t)\rangle\geqslant\sum_{k=N-d+1}^{N} w_k\forall t, \tag{11}$$

and if $w_{N-d}>w_N$ then we have equality exactly if

$$\mathrm{span}_{k=N-d+1,\ldots,N}|\Psi_k\rangle=S. \tag{12}$$

In the case of an arbitrary observable $\hat{A}$ with eigenvalues a_i and corresponding eigenspaces $E(a_i)$, we note that (since $\mathcal{H}$ is assumed to be finite dimensional) there exists a unique eigenvalue decomposition

$$\hat{A}=\sum_{i=1}^{m} a_i\hat{I}(a_i), \tag{13}$$

where $\hat{I}(a_i)$ denotes the projector onto the eigenspace $E(a_i)$. Since $\hat{A}$ is Hermitian, all its eigenvalues are real and we may assume that they are ordered in a finite, decreasing sequence $a_1>a_2>\cdots>a_m$.

The problem of determining the kinematical maximum or minimum of the expectation value

$$\langle\hat{A}(t_f)\rangle=\mathrm{Tr}[\hat{A}\hat{U}(t_f,t_i)\hat{\rho}(t_i)\hat{U}^{\dagger}(t_f,t_i)] \tag{14}$$

at some fixed time $t=t_f$, for a given mixed initial state $\hat{\rho}(t_i)$, is equivalent to finding a unitary transformation $\hat{U}_0$ that maximizes or minimizes the value of the function

$$\hat{U}\mapsto f(\hat{U})=\mathrm{Tr}[\hat{A}\hat{U}\hat{\rho}(t_i)\hat{U}^{\dagger}], \tag{15}$$

as a function from the unitary group $\mathrm{U}(N)$ to the real numbers. f assumes both its maximum and its minimum since $\mathrm{U}(N)$ is a compact Lie group.

Suppose f assumes an extremum at $\hat{U}_0\in\mathrm{U}(N)$. Letting $\hat{\chi}=\hat{U}_0\hat{\rho}(t_i)\hat{U}_0^{\dagger}$, we obtain a map

$$\hat{U}\mapsto\mathrm{Tr}(\hat{A}\hat{U}\hat{\chi}\hat{U}^{\dagger}) \tag{16}$$

that has an extremum at $\hat{U}=\hat{1}$ where $\hat{1}$ is the identity operator.

Theorem 2: If the map $\hat{U}\mapsto\mathrm{Tr}(\hat{A}\hat{U}\hat{\chi}\hat{U}^{\dagger})$ has an extremum at the identity $\hat{1}\in\mathrm{U}(N)$ then $\hat{A}$ and $\hat{\chi}$ commute.

Proof: Assume $\mathrm{Tr}(\hat{A}\hat{U}\hat{\chi}\hat{U}^{\dagger})$ is maximal when $\hat{U}=\hat{1}$ and let $\hat{U}(\tau)$ be any path in U(N) which starts at the identity. Then we must have

$$\frac{d}{d\tau}\,\mathrm{Tr}[\hat{A}\hat{U}(\tau)\hat{\chi}\hat{U}^{\dagger}(\tau)]\bigg|_{\tau=0}=0.$$

Since the trace is linear, this is equivalent to

$$\mathrm{Tr}[\hat{A}\hat{U}'(0)\hat{\chi}\hat{U}^{\dagger}(0)+\hat{A}\hat{U}(0)\hat{\chi}[\hat{U}^{\dagger}]'(0)]=0.$$

Observing that $\hat{U}(0)=\hat{1}$, and letting $(d/d\tau)\hat{U}(\tau)|_{\tau=0}=\hat{U}'(0)\equiv\hat{B}$, we have thus

$$\mathrm{Tr}(\hat{A}\hat{B}\hat{\chi}+\hat{A}\hat{\chi}\hat{B}^{\dagger})=0. \tag{17}$$

If $\hat{B}$ is the derivative of a path $\hat{U}(\tau)\subset\mathrm{U}(N)$ at $\tau=0$ then it is skew-Hermitian, and conversely, every skew-Hermitian operator $\hat{B}$ is the derivative of some path $\hat{U}(\tau)\subset\mathrm{U}(N)$ at $\tau=0$, since the Lie algebra of U(N) consists of all skew-Hermitian matrices $u(N)$. Therefore condition (17) must hold for any skew-Hermitian operator $\hat{B}$. Using $\hat{B}^{\dagger}=-\hat{B}$, we can rewrite Eq. (17) as

$$\mathrm{Tr}[(\hat{\chi}\hat{A}-\hat{A}\hat{\chi})\hat{B}]=0.$$

We will show that this condition implies $\hat{M}=[\hat{\chi},\hat{A}]=0$. Note that $\hat{M}$ is skew-Hermitian since $\hat{\chi}$ and $\hat{A}$ are Hermitian.

Choosing $\hat{B}=(b_{k\ell})$ with

$$b_{k\ell}=\begin{cases} i & \text{for } k=s,\ell=s \\ 0 & \text{otherwise}\end{cases}$$

yields

$$\mathrm{Tr}(\hat{M}\hat{B})=\sum_{j,k=1}^{N} m_{jk}b_{kj}=iM_{ss}=0.$$

This holds for $s=1$ to N. Hence, all the diagonal elements of $\hat{M}$ must vanish.

If

$$b_{k\ell}=\begin{cases} 1 & \text{for } k=s,\ell=r \\ -1 & \text{for } k=r,\ell=s \\ 0 & \text{otherwise}\end{cases}$$

then

$$\mathrm{Tr}(\hat{M}\hat{B})=\sum_{j,k=1}^{N} m_{jk}b_{kj}=m_{rs}-m_{sr}=m_{rs}+m_{rs}^{*}=2\,\mathrm{Re}(m_{rs})=0,$$

and if

$$b_{k\ell}=\begin{cases} i & \text{for } k=s,\ell=r \\ i & \text{for } k=r,\ell=s \\ 0 & \text{otherwise}\end{cases}$$

then

$$\mathrm{Tr}(\hat{M}\hat{B})=\sum_{j,k=1}^{N} m_{jk}b_{kj}=i(m_{rs}+m_{sr})=i(m_{rs}-m_{rs}^{*})=-2\,\mathrm{Im}(m_{rs})=0.$$

Since this holds for $r,s=1$ to N, all off-diagonal elements of $\hat{M}$ must be zero. Hence $\hat{M}$ vanishes identically and $\hat{A}$ and $\hat{\chi}$ commute.

Theorem 3: Let $\hat{A}$ be a Hermitian operator on $\mathcal{H}$ with eigenvalue decomposition (13), where $\lambda_1\geqslant\lambda_2\geqslant\cdots\geqslant\lambda_N$ be the eigenvalues a_i, counted with multiplicity, and ordered in a decreasing sequence. Then we have

$$\sum_{k=1}^{N}\lambda_{N-k+1}w_k\leqslant\mathrm{Tr}[\hat{A}\hat{\rho}(t)]\leqslant\sum_{k=1}^{N}\lambda_k w_k. \tag{18}$$

Proof: By the previous theorem a necessary condition for $\langle\hat{A}(t)\rangle$ to have an extremum at time t_f is that $\hat{A}$ and $\hat{\chi}=\hat{\rho}(t_f)$ commute. But if they commute then they can be simultaneously diagonalized. Hence

$$\mathrm{Tr}[\hat{A}\hat{\rho}(t_f)]=\sum_{k=1}^{N}\lambda_k w_{\sigma(k)},$$

where σ is a permutation of the w_k's. It is now obvious that

$$\sum_{k=1}^{N}\lambda_{N-k+1}w_k\leqslant\mathrm{Tr}[\hat{A}\hat{\rho}(t_f)]\leqslant\sum_{k=1}^{N}\lambda_k w_k.$$

Theorem 4: $\langle A(t_f)\rangle$ assumes its upper bound if for all $k=1$ to m

$$\mathrm{span}_{j=1,\ldots,d(k)}|\Psi_{r(k,j)}(t_f)\rangle=E(a_k), \tag{19}$$

and its lower bound if

$$\mathrm{span}_{j=1,\ldots,d(k)}|\Psi_{r(k,j)}(t_f)\rangle=E(a_{N-k+1}), \tag{20}$$

where $d(k)=\dim E(a_k)$ and $r(k,j)=d(1)+\cdots+d(k-1)+j$.

If the w_k are all different then the if's in the previous theorem are really ''if and only if's.''

Proof:

$$\langle A(t)\rangle=\mathrm{Tr}(\hat{A}\hat{\rho}(t))=\sum_j w_j\langle\Psi_j(t)|\hat{A}\Psi_j(t)\rangle$$

$$=\sum_j w_j\left\langle\Psi_j(t)\middle|\sum_i a_i\hat{I}(a_i)\Psi_j(t)\right\rangle$$

$$=\sum_j w_j\sum_i a_i\|\hat{I}(a_i)\Psi_j(t)\|^2$$

$$=\sum_i a_i\sum_j w_j\|\hat{I}(a_i)\Psi_j(t)\|^2=\sum_i a_i f_i(t),$$

where we let

$$f_i(t) \equiv \sum_j w_j \|\hat{I}(a_i)\Psi_j(t)\|^2.$$

The kinematical upper bound will be realized at $t=t_f$ if

$$f_i(t_f) = \sum_{j=1}^{d(i)} w_{r(i,j)}$$

for $i=1$ to m, which is the case if

$$\mathrm{span}_{j=1,\ldots,d(i)} |\Psi_{r(i,j)}(t_f)\rangle = E(a_i)$$

for $i=1$ to m by theorem 2.

The kinematical lower bound will be realized if

$$f_{N-i+1}(t_f) = \sum_{j=1}^{d(i)} w_{r(i,j)}$$

for $i=1$ to m, which is the case if

$$\mathrm{span}_{j=1,\ldots,d(i)} |\Psi_{r(i,j)}(t_f)\rangle = E(a_{N-i+1})$$

for $i=1$ to m by theorem 2.

Proof: Hence the kinematical maximum of $\langle \hat{A}(t)\rangle$ is determined by the eigenvalues a_k of $\hat{A}$, as well as the eigenvalues w_k of $\hat{\rho}(t_i)$, and it is attained at time t_f if the $d(1)$ eigenstates $|\Psi_k(t_f)\rangle$ of $\hat{\rho}(t)$ with the greatest statistical weights $w_1,\ldots,w_{d(1)}$ span the $d(1)$-dimensional eigenspace corresponding to the biggest eigenvalue λ_1 of $\hat{A}$, and if the next $d(2)$ eigenstates of $\hat{\rho}(t)$ corresponding to the next largest statistical weights $w_{d(1)+1},\ldots,w_{d(1)+d(2)}$ span the $d(2)$-dimensional eigenspace corresponding to the second largest eigenvalue λ_2 of $\hat{A}$, and so forth.

IV. APPLICATION: MAXIMIZING THE ENERGY

As an example for molecular quantum control, we consider a diatomic molecule model with N discrete vibrational energy levels E_n corresponding to independent states $|n\rangle$ of the system. Together, the states $\{|n\rangle : n=1,\ldots,N\}$ form a complete orthonormal basis of the state space $\mathcal{H}$. The unperturbed Hamiltonian is thus

$$\hat{H}_0 = \sum_{n=1}^{N} E_n |n\rangle\langle n|. \tag{21}$$

The interaction Hamiltonian of the driven system is $\hat{H}_1 = f(t)\hat{V}$, where $f(t)$ is an external laser field that serves as a control function, and $\hat{V}$ is the transition operator, which we choose to be of the dipole form

$$\hat{V} = \sum_{n=1}^{N-1} d_n(|n\rangle\langle n+1| + |n+1\rangle\langle n|). \tag{22}$$

We consider mixed initial states of the system that are statistical mixtures of eigenstates of $\hat{H}_0$, i.e., we assign each pure state $|n\rangle$ a probability w_n with $0 \leq w_n \leq 1$, where w_n of some energy levels may be 0. Hence, the initial state is represented by the diagonal density operator

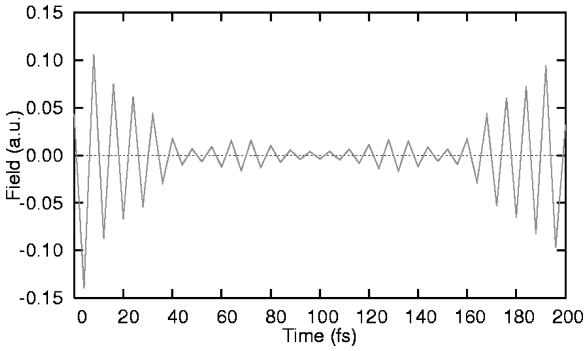

FIG. 1. Optimal pulse, Morse oscillator.

$$\hat{\rho}(0) = \sum_{n=1}^{N} w_n |n\rangle\langle n|, \tag{23}$$

and $\Sigma_{n=1}^{N} w_n = 1$, since the total probability for the system to be found in any of these eigenstates must be 1.

Our goal is to determine the laser field $f(t)$ so as to maximize the expectation value of the observable

$$\hat{A} = \hat{H}_0 = \sum_{n=1}^{4} E_n |n\rangle\langle n|, \tag{24}$$

which corresponds to the energy of the system at a certain target time $t=t_f$.

In the following examples the optimal electric field $f(t)$ of the laser pulse is computed using a nonlinear eigensystem method [3]. Optimization is carried out for a fixed pulse length $t_f = 200$ fs and a sequence of increasing pulse fluences

$$\epsilon_p = \int_0^{t_f} F^2(t)\,dt, \tag{25}$$

where $F(t) = p_{12} f(t)/\hbar$, and p_{12} is the $1 \rightarrow 2$ transition dipole moment.

A. Morse oscillator model

In our computations for a HF molecule, we used a four-level Morse oscillator model as described in [4]. The vibrational energy levels are determined by

$$E_n = \hbar\omega_0 \left(n - \frac{1}{2}\right)\left[1 - \frac{1}{2}\left(n - \frac{1}{2}\right)B\right] \tag{26}$$

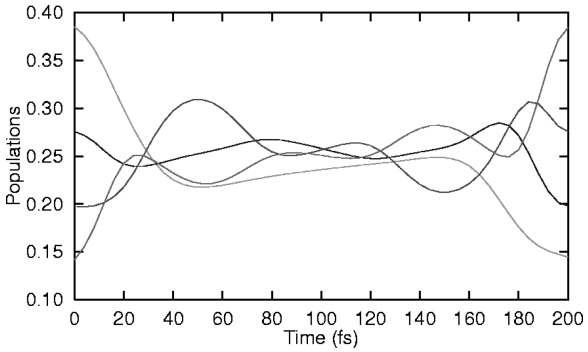

FIG. 2. Populations, Morse oscillator.

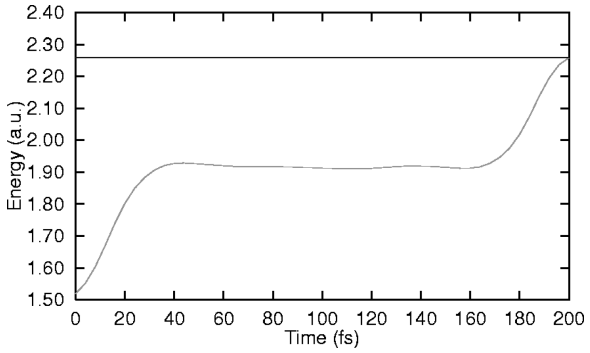

FIG. 3. Energy, Morse oscillator.

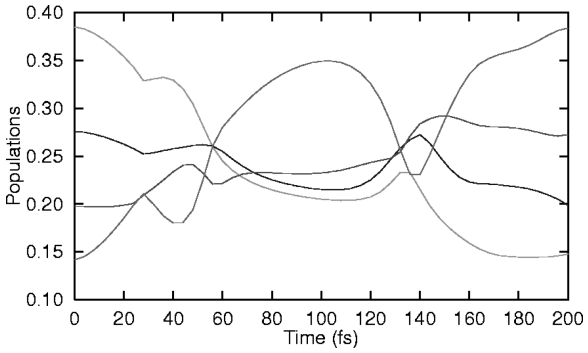

FIG. 5. Populations, harmonic oscillator.

for $n=1$ to 4, where $\omega_0=7.8\times10^{14}$ s^{-1} and $B=0.0419$, and the dipole moments are

$$d_n=0.097n^{1/2}\ \text{D}, \quad n=1,\ldots,3. \tag{27}$$

The initial ensemble is $|n\rangle=|1\rangle,\ldots,|4\rangle$ and the statistical weights of the unperturbed states are given by a Boltzmann distribution

$$w_n=\text{const}\times e^{-E_n/kT} \ \text{ with } \ kT=E_4-E_1, \tag{28}$$

i.e., concretely $w_1=0.3850$, $w_2=0.2758$, $w_3=0.1976$, and $w_4=0.1416$.

Theorem 3 gives the kinematical upper bound

$$\langle\hat{H}_0(t_f)\rangle\leqslant\sum_{i=1}^{4} w_iE_i=2.2592\hbar\omega_0, \tag{29}$$

which is exactly attained if the population of level E_1 is w_4, that of level E_2 is w_3, that of E_3 is w_2, and that of E_4 is w_1 by theorem 4.

We verified that this system is (completely) controllable according to theorem 4.1 in [5], or corollary 1 (chapter 3) in [6], if the class of admissible controls consists of all bounded measurable functions. (For a proof of the original result on controllability see [7].) Therefore any kinematically attainable state is dynamically reachable for some bounded measurable control $f(t)$. Although not every bounded measurable function is a physically reasonable control, and our optimization program required in addition the pulse fluence to be constant during each optimization step, we expect that it should be possible to approximate the kinematical upper bound closely with a physically admissible control field $f(t)$ for a suitably chosen, increasing sequence of fixed pulse fluences.

Figure 1 shows the computational results for the optimal pulse. The evolution of the energy level populations, shown in Fig. 2 for the final fluence 0.25 fs^{-1}, demonstrates that the initial eigenstate $|1\rangle$ evolves into a state closely approximating eigenstate $|4\rangle$ at the target time, $|2\rangle$ into $|3\rangle$, $|3\rangle$ into $|2\rangle$, and $|4\rangle$ into $|1\rangle$. Notice that this inversion of the ordering of the populations implies a strongly off-diagonal form of $\hat{\rho}(t)$ at intermediate times. The final populations for energy levels 1 through 4 are 0.1445, 0.1979, 0.2758, and 0.3834, respectively, agreeing with the initial populations (except for inversion) to within 2%. The energy curve is shown in Fig. 3. The final energy was $\langle\hat{H}_0(200)\rangle=2.2579\hbar\omega_0$ or about 99.94% of the kinematical upper bound.

B. Harmonic-oscillator model

We performed similar computations for a four-level truncated harmonic-oscillator model with the same energy levels E_1 and E_2 as Eq. (26) but energy levels E_3 and E_4 adjusted slightly to achieve equal spacing of adjacent energy levels. The dipole moments d_n and statistical weights w_n were chosen to be the same as for the HF model, and the goal was again to maximize the energy of the system.

Theorem 3 predicts the kinematical upper bound

$$\langle\hat{H}_0(t_f)\rangle\leqslant\sum_{i=1}^{4} w_iE_i=2.3192\hbar\omega_0, \tag{30}$$

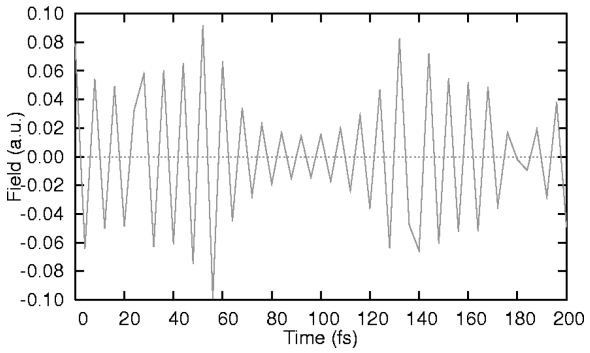

FIG. 4. Optimal pulse, harmonic oscillator.

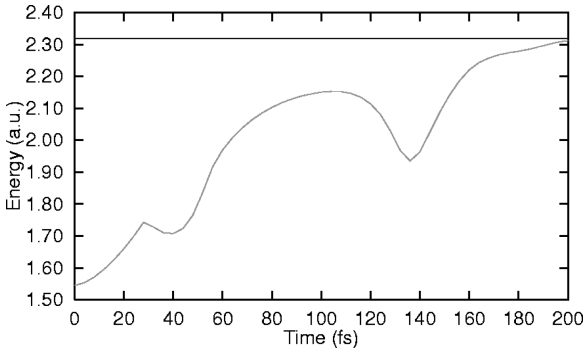

FIG. 6. Energy, harmonic oscillator.

which is by theorem 4 exactly attained if the population of level E_1 is w_4, that of level E_2 is w_3, that of E_3 is w_2, and that of E_4 is w_1.

We verified that the dimension of the Lie algebra (over the reals) generated by $\hat{H}_0$ and $\hat{V}$ is less than 16, the maximal dimension for the system, and thus this system is not (completely) controllable according to theorem 4.1 in [5], or corollary 1 (chapter 3) in [6]. Nevertheless, the computational results strongly suggest that the kinematical upper bound for this particular choice of observable can still be closely approximated dynamically. Future research on dynamical reachability and the structure of the set of accessible target states would therefore be interesting.

The final populations for energy levels 1 through 4 are 0.1478, 0.1987, 0.2726, and 0.3838, respectively, agreeing with the result predicted by the theorem to within 4%. The final energy is $\langle \hat{H}_0(200) \rangle = 2.3119 \hbar \omega_0$ or about 99.68% of the kinematical upper bound. The numerical results for the optimal pulse, the evolution of the populations, and the observable for the final fluence 0.28 fs^{-1}, are shown in Figs. 4, 5, and 6.

[1] W. Warren, H. Rabitz, and M. Dahleh, Science **259**, 1581 (1993).
[2] M. D. Girardeau, M. Ina, S. Schirmer, and T. Gulsrud, Phys. Rev. A **55**, R1565 (1997).
[3] M. Ina, Ph.D. thesis, University of Oregon, 1996 (unpublished).
[4] S. Chelkowski, A. Bandrauk, and P. B. Corkum, Phys. Rev. Lett. **65**, 2355 (1990).
[5] V. Ramakrisna *et al.*, Phys. Rev. A **51**, 960 (1995).
[6] A. G. Butkovskiy and Y. I. Saimolenko, *Control of Quantum-Mechanical Processes and Systems* (Kluwer Academic, Dordrecht, 1990).
[7] V. Jurdjevic and H. J. Sussmann, J. Diff. Eqns. **12**, 313 (1972).