

# Limits of control for quantum systems: Kinematical bounds on the optimization of observables and the question of dynamical realizability

S. G. Schirmer

*Quantum Processes Group, The Open University, Milton-Keynes MK7 6AA, United Kingdom*

J. V. Leahy

*Department of Mathematics and Institute of Theoretical Science, University of Oregon, Eugene, Oregon 97403*

(Received 20 June 2000; published 11 January 2001)

In this paper we investigate the limits of control for mixed-state quantum systems. The constraint of unitary evolution for nondissipative quantum systems imposes kinematical bounds on the optimization of arbitrary observables. We summarize our previous results on kinematical bounds and show that these bounds are dynamically realizable for completely controllable systems. Moreover, we establish improved bounds for certain partially controllable systems. Finally, the question of dynamical realizability of the bounds for arbitrary partially controllable systems is shown to depend on the accessible sets of the associated control system on the unitary group  $U(N)$  and the results of a few control computations are discussed briefly.

DOI: 10.1103/PhysRevA.63.025403

PACS number(s): 32.80.Qk, 03.65.Ta, 05.30.-d, 02.20.Sv

## I. INTRODUCTION

Recent advances in laser technology have opened up new possibilities for laser control of quantum phenomena such as control of molecular quantum states, chemical reaction dynamics, or quantum computers to mention only a few. The limited success of initially advocated control schemes based largely on physical intuition in both theory and experiment [1] has prompted researchers in recent years to study these systems systematically using control theory. One of the many interesting questions that has arisen is the issue of dynamical realizability of bounds on the optimization of observables imposed by kinematical constraints, which we shall address in this paper.

The answer to this question is not just of theoretical interest, it is a matter of practical importance as well since most algorithms designed to find optimal controls [2,3] to drive a system are based on a set of differential equations providing necessary but not sufficient conditions for a global maximum, i.e., these algorithms may produce controls that steer the system to a local maximum or minimum, for instance. Independent knowledge of dynamically attainable bounds on the expectation value of the observable makes it possible to determine the effectiveness of a given control in achieving the control objective of maximizing or minimizing the expectation value of a given observable.

## II. QUANTUM STATISTICAL MECHANICS MODEL

We consider a quantum-mechanical system whose pure states form a separable Hilbert space  $\mathcal{H}$ . A mixed state is an ensemble of orthonormal pure quantum states  $|\Psi_k\rangle$  with a discrete probability distribution assigning each pure state a certain probability  $w_k$  such that  $0 \leq w_k \leq 1$  and  $\sum_k w_k = 1$ . Such a state can always be represented by a density operator  $\hat{\rho}$  on  $\mathcal{H}$  with eigenvalue decomposition

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

where  $w_k$  are the eigenvalues, and  $|\Psi_k\rangle$  the corresponding normalized eigenstates of  $\hat{\rho}$ . Unless otherwise specified, we shall use the word ‘‘state’’ in the following to refer to a mixed quantum state represented by a density operator  $\hat{\rho}$ .

If the system is Hamiltonian, the time evolution of  $\hat{\rho}$  is given by

$$\hat{\rho}(t) = \hat{U}(t, t_0) \hat{\rho}_0 \hat{U}(t, t_0)^\dagger, \quad (2)$$

where  $\hat{\rho}_0 = \hat{\rho}(t_0)$  and  $\hat{U}(t, t_0)$  is the time-evolution operator of the system satisfying the time-dependent Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} \hat{U}(t, t_0) = \hat{H} \hat{U}(t, t_0). \quad (3)$$

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

Observables are represented by Hermitian operators  $\hat{A}$  on  $\mathcal{H}$  and we define their expectation value to be the *ensemble average*

$$\langle \hat{A}(t) \rangle = \text{Tr}[\hat{A} \hat{\rho}(t)]. \quad (4)$$

The aim of controlling the system is to maximize the expectation value of a chosen observable  $\hat{A}$  at a certain target time  $t_F$  by driving the system using a set of optimal control fields  $\mathbf{f}(t) = [f_1(t), \dots, f_M(t)]$ . If the control fields are sufficiently weak, it can be assumed that the system is control linear,

$$\hat{H} = \hat{H}_0 + \sum_{m=1}^M f_m(t) \hat{H}_m, \quad (5)$$

where  $\hat{H}_0$  is the internal Hamiltonian of the system and  $\hat{H}_m, m \geq 1$ , is the interaction Hamiltonian for the field  $f_m$ .

### III. KINEMATICAL RESTRICTIONS ON THE DYNAMICS

If dissipative effects are negligible, then the quantum system is Hamiltonian and therefore the evolution of the system has to be unitary no matter how the system is driven. This observation has profound implications. Given an initial mixed state  $\hat{\rho}_0$ , the only kinematically attainable target states  $\hat{\rho}(t_F)$  are

$$\hat{\rho}(t_F) = \hat{U}(t_F, t_0) \hat{\rho}_0 \hat{U}(t_F, t_0)^\dagger, \quad (6)$$

where  $\hat{U}(t_F, t_0)$  is a unitary transformation.

This kinematical constraint on the dynamical evolution leads to bounds on the expectation value of arbitrary observables that are completely independent of the control functions and thus impose kinematical restrictions on the optimization of observables, summarized by the following two theorems [4].

*Theorem 1.* Let  $\hat{A}$  be a Hermitian operator on  $\mathcal{H}$  with eigenvalue decomposition

$$\hat{A} = \sum_{i=1}^m a_i \hat{I}(a_i), \quad (7)$$

where  $\hat{I}(a_i)$  is the projector onto the eigenspace  $E(a_i)$ , and let  $\lambda_1 \geq \lambda_2 \geq \dots \geq \lambda_N$  be the eigenvalues  $a_i$ , counted with multiplicity, and ordered in a decreasing sequence. Then we have

$$\sum_{k=1}^N \lambda_{N-k+1} w_k \leq \text{Tr}[A \hat{\rho}(t)] \leq \sum_{k=1}^N \lambda_k w_k, \quad (8)$$

provided that the weights  $w_k$  are ordered in a decreasing sequence, i.e.,  $w_1 \geq w_2 \geq \dots \geq w_k \geq \dots$ .

*Theorem 2.*  $\langle A(t_F) \rangle$  assumes its upper bound if for all  $k$  from 1 to  $m$

$$\text{span}_{j=1, \dots, d(k)} |\Psi_{r(k,j)}(t_F)\rangle = E(a_k), \quad (9)$$

and its lower bound if for all  $k$  from 1 to  $m$

$$\text{span}_{j=1, \dots, d(k)} |\Psi_{r(k,j)}(t_F)\rangle = E(a_{N-k+1}), \quad (10)$$

where  $d(k) = \dim E(a_k)$ , i.e., the dimension of the eigenspace belonging to the eigenvalue  $a_k$ , and  $r(k, j) = d(1) + \dots + d(k-1) + j$ .

Thus, to attain the kinematical maximum for an observable whose eigenvalues are distinct with multiplicity one, we need to find a unitary transformation that simultaneously maps the initial pure state with the largest probability  $w_1$  onto the eigenspace corresponding to the largest eigenvalue of  $\hat{A}$ , the initial state with the second largest probability  $w_2$  onto the eigenspace corresponding to the second largest eigenvalue of  $\hat{A}$ , and so forth. If  $\hat{A}$  is a projector onto a subspace of dimension  $d$ , then attaining the kinematical upper bound requires finding a unitary transformation that maps the  $d$  initial states with the  $d$  largest probabilities onto the eigenspace corresponding to the eigenvalue one of  $\hat{A}$ .

### IV. DYNAMICAL REALIZABILITY OF THE KINEMATICAL BOUNDS

The kinematical bounds derived above are clearly dynamically realizable if every unitary operator in  $U(N)$  is accessible from the identity operator  $\hat{I}$  via a path that satisfies the dynamical law (3), i.e., if the system is completely mixed-state controllable.

For control-linear systems, complete controllability can easily be verified numerically. Note that combining Eqs. (3) and (5) leads to

$$i\hbar \frac{\partial}{\partial t} \hat{U}(t, t_0) = \hat{H}_0 \hat{U}(t, t_0) + \sum_{m=1}^M f_m(t) \hat{H}_m \hat{U}(t, t_0). \quad (11)$$

Setting  $x(t) = \hat{U}(t, t_0)$  and

$$\mathbf{X}_m(x(t)) = -\frac{i}{\hbar} \hat{H}_m \hat{U}(t, t_0), \quad m=0, \dots, M, \quad (12)$$

Eq. (11) becomes

$$\frac{dx}{dt} = \mathbf{X}_0(x(t)) + \sum_{m=1}^M f_m(t) \mathbf{X}_m(x(t)), \quad (13)$$

which defines a control system on the Lie group  $U(N)$  of a type studied by Jurdjjevic and Sussmann [5]. From their results, the following simple algebraic conditions for complete controllability of control-linear, nondissipative quantum systems have been derived [6].

*Theorem 3.* If the total Hamiltonian is given by Eq. (5), where  $f_m$  are independent bounded measurable control functions, and  $\dim \mathcal{H} = N < \infty$  then a necessary and sufficient condition for the system to be completely controllable is that the Lie subalgebra  $L_0$  of  $u(N)$  (the skew-Hermitian matrices) generated by  $\hat{H}_0, \dots, \hat{H}_M$  has dimension  $N^2$ , or equivalently, that the ideal  $l_0$  of  $L(U(N))$  generated by  $\hat{H}_1, \dots, \hat{H}_M$  has dimension  $N^2 - 1$ .

If the system is not control linear, i.e., the Hamiltonian depends in a nonlinear way on the control functions  $f_m$  then there is in general no simple algebraic condition to verify controllability. For systems that are not completely controllable, dynamical realizability of a particular kinematical bound depends on the set of unitary transformations  $\hat{U}(t_F, t_0) \in U(N)$  that are accessible from the identity  $\hat{I} \in U(N)$ . More precisely, it depends on whether the intersection of the set of dynamically accessible target states and the set of states for which the expectation value of the chosen observable assumes the kinematical bound is empty or not. Since the set of dynamically accessible target states consists of all density matrices satisfying Eq. (6), where  $U(t_F, t_0)$  is a unitary transformation accessible from the identity in  $U(N)$  via a path that satisfies the equation of motion (3), a crucial step towards answering the question of dynamical realizability is to determine the accessible sets for the associated control system on  $U(N)$ .

### V. DYNAMICALLY REALIZABLE BOUNDS FOR DECOUPLED SYSTEMS

Among the systems that are obviously not completely controllable are those comprised of noninteracting subsystems. We shall refer to these systems as decoupled systems.

Let us first consider a control-linear Hamiltonian system with a single control,

$$\hat{H}(f(t)) = \hat{H}_0 + f(t)\hat{V}, \quad (14)$$

where  $\hat{H}_0$  is the internal Hamiltonian of the unperturbed system and  $\hat{V}$  defines the interaction with the control field  $f(t)$ .

In this case, the system is decoupled if there exists a basis  $\mathcal{B}$  for the Hilbert space  $\mathcal{H}$  such that  $\hat{H}_0$  is diagonal and

$$\hat{V} = \hat{V}_1 \oplus \hat{V}_2 \doteq \begin{pmatrix} \hat{V}_1 & 0 \\ 0 & \hat{V}_2 \end{pmatrix}. \quad (15)$$

Let  $\mathcal{H}_1$  and  $\mathcal{H}_2$  be orthogonal subspaces of  $\mathcal{H}$  such that  $\mathcal{H} = \mathcal{H}_1 \oplus \mathcal{H}_2$  and each  $\hat{V}_i$  maps  $\mathcal{H}_i$  to itself,

$$\mathcal{H} = \mathcal{H}_1 \oplus \mathcal{H}_2, \quad \hat{V}_i : \mathcal{H}_i \rightarrow \mathcal{H}_i, \quad i = 1, 2. \quad (16)$$

It immediately follows that  $\hat{H}(f(t))$  is block-diagonal,

$$\mathcal{H}(f(t)) = \hat{H}_1 \oplus \hat{H}_2 \doteq \begin{pmatrix} \hat{H}_1 & 0 \\ 0 & \hat{H}_2 \end{pmatrix} \quad (17)$$

and maps  $\mathcal{H}_i$  to itself for  $i = 1, 2$ . Thus, the two subspaces  $\mathcal{H}_1$  and  $\mathcal{H}_2$  do not interact. Let  $\mathcal{B}_i$  be the restriction of the basis  $\mathcal{B}$  to the subspace  $\mathcal{H}_i$ ,  $\hat{P}_i$  be the projector onto the subspace  $\mathcal{H}_i$  and let  $N_i$  denote the dimension of  $\mathcal{H}_i$ .

Given an observable  $\hat{A}$  on  $\mathcal{H}$ , we define the restricted observables  $\hat{A}_i = \hat{P}_i \hat{A} \hat{P}_i$  for  $i = 1, 2$ . Note that  $\hat{A}_i$  is a Hermitian operator on the subspace  $\mathcal{H}_i$ , i.e.,

$$\hat{A}_i = \hat{P}_i \hat{A} \hat{P}_i : \mathcal{H}_i \rightarrow \mathcal{H}_i \quad i = 1, 2. \quad (18)$$

Let  $\lambda_n^{(i)}$  denote the eigenvalues of  $\hat{A}_i$ , counted with multiplicity and ordered

$$\lambda_1^{(i)} \geq \lambda_2^{(i)} \geq \dots \geq \lambda_{N_i}^{(i)}. \quad (19)$$

If  $\hat{\rho}_i(t_0)$  is the density operator for subsystem  $i$ , whose matrix representation with respect to the basis  $\mathcal{B}_i$  is

$$\hat{\rho}_i(t_0) \doteq \text{diag}(w_1^{(i)}, \dots, w_{N_i}^{(i)}) \quad (20)$$

with  $w_1^{(i)} \geq w_2^{(i)} \geq \dots \geq w_{N_i}^{(i)}$ , we can apply theorem 1 to obtain bounds for the expectation value of  $\hat{A}_i$ :

$$\sum_{n=1}^{N_i} w_{N_i-n+1}^{(i)} \lambda_n^{(i)} \leq \langle \hat{A}_i(t) \rangle \leq \sum_{n=1}^{N_i} w_n^{(i)} \lambda_n^{(i)}. \quad (21)$$

Notice that the total probability for each subspace is less or equal to one, and that the sum of the subspace probabilities must equal one, i.e.,

$$p_1 = \sum_{n=1}^{N_i} w_n^{(i)} \leq 1, \quad p_1 + p_2 = 1. \quad (22)$$

If the probability for subspace  $i$  is one, then the initial ensemble is restricted to this subspace and since the subspaces do not interact, the ensemble will remain in this subspace forever, i.e.,  $p_i = 1$  for all times. In this case,  $\langle \hat{A}(t) \rangle = \langle \hat{A}_i(t) \rangle$ .

If both subspaces are initially occupied, i.e., both  $p_1$  and  $p_2$  are nonzero, then the density operator for the entire space  $\mathcal{H}$  is the direct sum of the subspace density operators  $\hat{\rho}_1(t_0)$  and  $\hat{\rho}_2(t_0)$ , i.e.,

$$\hat{\rho}(t_0) = \hat{\rho}_1(t_0) \oplus \hat{\rho}_2(t_0) \doteq \begin{pmatrix} \hat{\rho}_1(t_0) & 0 \\ 0 & \hat{\rho}_2(t_0) \end{pmatrix}. \quad (23)$$

Since  $\hat{H}$  maps each subspace to itself, we can conclude

$$\hat{\rho}(t) = \hat{\rho}_1(t) \oplus \hat{\rho}_2(t) \doteq \begin{pmatrix} \hat{\rho}_1(t) & 0 \\ 0 & \hat{\rho}_2(t) \end{pmatrix} \quad (24)$$

for  $t > t_0$  and thus it easily follows that

$$\langle \hat{A}(t) \rangle = \text{Tr}[\hat{A}_1 \hat{\rho}_1(t)] + \text{Tr}[\hat{A}_2 \hat{\rho}_2(t)]. \quad (25)$$

Since  $\hat{\rho}_i(t)$  and  $\hat{A}_i$  ( $i = 1, 2$ ) are operators on  $\hat{H}_i$ , we can apply Eq. (21). Thus we have theorem 4.

*Theorem 4.* Consider a decoupled quantum system as defined above. If  $\hat{\rho}_0$  is given by Eq. (23), then the expectation value of an observable  $\hat{A}$  is bounded by

$$\langle \hat{A}(t) \rangle \geq \sum_{n=1}^{N_1} w_n^{(1)} \lambda_{N_1-n+1}^{(1)} + \sum_{n=1}^{N_2} w_n^{(2)} \lambda_{N_2-n+1}^{(2)}, \quad (26)$$

$$\langle \hat{A}(t) \rangle \leq \sum_{n=1}^{N_1} w_n^{(1)} \lambda_n^{(1)} + \sum_{n=1}^{N_2} w_n^{(2)} \lambda_n^{(2)}, \quad (27)$$

where  $\lambda_n^{(i)}$  are the eigenvalues of the subspace observable  $\hat{A}_i$ , counted with multiplicity and ordered in a decreasing sequence. Furthermore, the upper bound is attained at  $t = t_F$  if for  $k = 1, \dots, m_1$

$$\text{span}_{j=1, \dots, d(k)} |\Psi_{r(k,j)}^{(1)}(t_F)\rangle = E(a_k^{(1)}), \quad (28)$$

where  $d(k) = \dim E(a_k^{(1)})$  and  $r(k,j) = d(1) + \dots + d(k-1) + j$ , and for  $l = 1, \dots, m_2$

$$\text{span}_{j=1, \dots, d(l)} |\Psi_{r(l,j)}^{(2)}(t_F)\rangle = E(a_l^{(2)}), \quad (29)$$

where  $d(l) = \dim E(a_l^{(2)})$  and  $r(l,j) = d(1) + \dots + d(l-1) + j$ . Similar conditions can be written down for the lower bound.

This theorem provides improved bounds for decoupled systems and it is easy to see how it can be generalized to

systems consisting of more than two noninteracting subsystems or control-linear systems with multiple controls. The improved bounds are dynamically realizable if all the subsystems are simultaneously completely controllable. While the previously mentioned condition for complete controllability can be applied to each subsystem, it is not clear whether complete controllability of all subsystems always implies complete controllability of the system as a whole. However, our computations for several decoupled systems suggest that this is the case for the systems we studied [7].

## VI. CONTROL COMPUTATIONS FOR A COUPLED PARTIALLY CONTROLLABLE SYSTEM

For partially controllable systems whose dynamics cannot be decomposed into independent subspace dynamics, the question of dynamical realizability of the kinematical bounds for a given observable cannot be answered in general. Rather it depends on the choice of the observable. Among the many computations we have done, we studied a four-level harmonic-oscillator model with unusual interaction terms,

$$\hat{H}_0 = \begin{bmatrix} 0.5 & 0 & 0 & 0 \\ 0 & 1.5 & 0 & 0 \\ 0 & 0 & 2.5 & 0 \\ 0 & 0 & 0 & 3.5 \end{bmatrix}, \quad \hat{H}_1 = f(t) \begin{bmatrix} 0 & 1 & 0 & 0 \\ 1 & 0 & 1 & 0 \\ 0 & 1 & 0 & 1 \\ 0 & 0 & 1 & 0 \end{bmatrix}.$$

The main difference of this model compared to the standard harmonic-oscillator model is that we set all the transition probabilities equal to 1 instead of  $\sqrt{n}$ . Clearly, the system is not decoupled. Yet, unlike the standard harmonic oscillator, this system is not completely controllable. In fact, it can be shown that the dimension of the associated Lie algebra drops from 16 to 11 precisely when all the transition probabilities are equal. If only one of these values is changed, complete controllability is recovered [7].

Nevertheless, our control computations maximizing (a) the energy of the system, i.e.,  $\hat{A} = \hat{H}_0$  and (b) the transition dipole moment  $\hat{A} = \hat{V}$ , assuming

$$\hat{\rho}_0 = \begin{bmatrix} 0.4 & 0 & 0 & 0 \\ 0 & 0.3 & 0 & 0 \\ 0 & 0 & 0.2 & 0 \\ 0 & 0 & 0 & 0.1 \end{bmatrix},$$

indicate that it still seems to be possible to control the system rather effectively even if all the transition probabilities are the same. The final yield after 20 iterations was 97.75% of the kinematical maximum in case (a) and 95.69% in case (b). These results suggest that further study of the dynamical realizability of the kinematical bounds for partially controllable systems is necessary.

## VII. CONCLUSION

We have shown how the kinematical constraint of unitary evolution for nondissipative quantum systems gives rise to kinematical bounds on the optimization of arbitrary observables and established general criteria for the dynamical realizability of these kinematical bounds. It has been shown in particular that the kinematical bounds are always dynamically realizable for completely controllable systems and that improved bounds can be derived for decoupled systems. Finally, we have demonstrated in Sec. VI that certain modifications of a completely controllable system may lead to a loss of complete controllability, but that even in such a case the kinematical bounds may still be approximately dynamically attainable. In the latter case, further investigation of the structure of the Lie algebra associated with the given control system is necessary to determine whether a particular kinematical bound for a partially controllable system can be dynamically attained.

- 
- [1] H. Rabitz, R. de Vivie-Riedle, M. Motzkus, and K. Kompa, *Science* **288**, 824 (2000).  
 [2] S. G. Schirmer, M. D. Girardeau, and J. V. Leahy, *Phys. Rev. A* **61**, 012101 (2000).  
 [3] W. Zhu and H. Rabitz, *J. Chem. Phys.* **109**, 385 (1998).

- [4] M. D. Girardeau, S. G. Schirmer, J. V. Leahy, and R. M. Koch, *Phys. Rev. A* **58**, 2684 (1998).  
 [5] V. Jurdjevic and H. J. Sussmann, *J. Diff. Eqns.* **12**, 313 (1972).  
 [6] V. Ramakrishna *et al.*, *Phys. Rev. A* **51**, 960 (1995).  
 [7] S. G. Schirmer (unpublished).