Complete controllability of quantum systems

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Sufficient conditions for complete controllability of N-level quantum systems subject to a single control pulse that addresses multiple allowed transitions concurrently are established. The results are applied in particular to Morse and harmonic-oscillator systems, as well as some systems with degenerate energy levels. Controllability of these model systems is of special interest since they have many applications in physics, e.g., Morse and harmonic oscillators serve as models for molecular bonds, and the standard control approach of using a sequence of frequency-selective pulses to address a single transition at a time is either not applicable or only of limited utility for such systems.

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. This has prompted researchers to study these systems from a control-theoretical point of view, in particular, in view of the limited success of initially advocated control schemes based largely on physical intuition in both theory and experiment [1].

One issue that arises is the question whether, or under which conditions, it is possible to control a quantum system in such a way as to achieve any physically permitted evolution of the system. Complete controllability is an important theoretical concept that also has significant practical implications. For example, it has been shown that kinematical constraints on the evolution of nondissipative quantum systems give rise to universal bounds on the optimization of observables [2] and that the practical issue of dynamical realizability of these bounds depends on the controllability of the system [3]. Controllability is also important in quantum computation as it is directly related to the question of universality of a quantum computation element [4].

It has been shown that an atomic system with N accessible energy levels, which are sufficiently separated to allow control based on frequency discrimination, is completely controllable using a sequence of frequency-selective pulses that address only a single transition at a time [5]. Although this approach is enormously useful, control based on frequency discrimination is not applicable to systems with equally spaced energy levels such as truncated harmonic oscillators, and problematic for systems with almost equally spaced energy levels such as Morse oscillators, since in this case, any external field is likely to address multiple transitions simultaneously. Moreover, systems with degenerate energy levels also present problems for this technique.

In this paper, we therefore concentrate on complete controllability of quantum systems subject to a single control field, e.g., a laser pulse or magnetic field, that addresses multiple transitions concurrently. In particular, we study the problem of controllability of a system subject to a single control, for which the interaction with the control field is determined by the dipole approximation. The systems considered are assumed to be nondecomposable, i.e., systems that can be decomposed into noninteracting subsystems are excluded. Note that such systems can never be completely controllable [3].

It will be shown in particular that a nondecomposable quantum system with dipole interaction is completely controllable with a single control pulse if there is some anharmonicity in the energy levels. It must be noted, however, that it is sufficient if, e.g., the transition frequency for the first transition is different from all the other transition frequencies. This is a much weaker condition than frequency discrimination, which requires that all the transition frequencies are sufficiently different. Furthermore, for a system with no anharmonicity, i.e., equally spaced energy levels, we demonstrate that complete controllability depends on the values of the transition dipole moments of the system, and establish sufficient criteria for complete controllability. The controllability of some systems with degenerate energy levels is also discussed and examples of systems that are not completely controllable are presented.

II. QUANTUM CONTROL SYSTEM

Given any N-level quantum system, the Hamiltonian of the unperturbed system can be written as

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

where $\{|n\rangle : n=1,\ldots,N\}$ is a complete set of orthonormal eigenstates and E_n are the corresponding energy levels of the system.

The application of external control fields perturbs the system and gives rise to a new Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}_I$, where $\hat{H}_I$ is an interaction term. In the control-linear approximation, the interaction term is of the form

$$\hat{H}_I = \sum_{m=1}^{M} f_m(t)\hat{H}_m, \tag{2}$$

where $f_m(t)$ for $m=1,\ldots,M$ are independent control fields, and the operator $\hat{H}_m$ represents the interaction of the field

$f_m(t)$ with the system. The off-diagonal elements of $\hat{H}_m$ depend on the transition dipole moments $d_{n,n'}$ for transitions between energy eigenstates. Each $\hat{H}_m$ is Hermitian since the transition dipole moments satisfy $d_{n',n} = d_{n,n'}^*$, where $d_{n,n'}^*$, is the complex conjugate of $d_{n,n'}$. In this paper, we are particularly interested in the case $M = 1$, i.e., a single control pulse, for which the interaction operator is of dipole form

$$\hat{H}_1 = \sum_{n=1}^{N-1} d_n(|n\rangle\langle n+1| + |n+1\rangle\langle n|), \quad d_n \neq 0. \quad (3)$$

Note that we exclude systems for which any of the transition dipole moments $d_n \equiv d_{n,n+1} = d_{n+1,n}$ vanish since these systems can be decomposed into noninteracting subsystems.

An arbitrary initial state of the system can be represented by a density matrix $\hat{\rho}_0$ that evolves according to the dynamical law

$$\hat{\rho}(t) = \hat{U}(t,t_0)\hat{\rho}_0\hat{U}(t,t_0)^\dagger, \quad (4)$$

where $\hat{U}(t,t_0)$ is the time-evolution operator, which satisfies the Schrödinger equation

$$i\hbar \frac{\partial}{\partial t}\hat{U}(t,t_0) = (\hat{H}_0 + \hat{H}_I)\hat{U}(t,t_0), \quad (5)$$

with initial condition $\hat{U}(t_0,t_0) = \hat{I}$.

We say the system is initially in a pure state if $\text{Tr}(\rho_0^2) = 1$. In this case, the initial state of the system can also be represented by a normalized wave function $|\psi_0\rangle$, which is either an energy eigenstate $|n\rangle$ or a superposition of energy eigenstates

$$|\psi_0\rangle = \sum_{n=1}^{N} c_n|n\rangle, \quad (6)$$

where the c_n are complex coefficients that satisfy the normalization condition $\Sigma_n c_n c_n^* = 1$ [6]. The time evolution of a pure state represented by a wave function $|\psi(t)\rangle$ is

$$|\psi(t)\rangle = \hat{U}(t,t_0)|\psi_0\rangle, \quad (7)$$

where $|\psi_0\rangle = |\psi(t_0)\rangle$ and $\hat{U}(t,t_0)$ is the time-evolution operator as defined above.

III. CRITERIA FOR COMPLETE CONTROLLABILITY

Since $\hat{H}_0 + \hat{H}_I$ is Hermitian, Eq. (5) implies that the time-evolution operator $\hat{U}(t,t_0)$ is unitary. Hence, examination of Eq. (4) reveals that only target states $\hat{\rho}(t_F)$ that are related to the initial state $\hat{\rho}_0$ by $\hat{\rho}(t_f) = \hat{U}\hat{\rho}_0\hat{U}^t$, where $\hat{U}$ is a unitary operator, are kinematically admissible. However, in general, not all of these states can actually be dynamically reached, unless the dynamical Lie group generated by $i\hat{H}_0$ and $i\hat{H}_m$, $m = 1,2,\ldots,M$, is the unitary group U(N). (See the Appendix for a discussion of this requirement.) This motivates the following.

Definition 1: A quantum system $\hat{H} = \hat{H}_0 + \hat{H}_I$ with $\hat{H}_0$ and $\hat{H}_I$ as in Eqs. (1) and (2) is completely controllable if every unitary operator U is accessible from the identity operator $\hat{I}$ via a path $\gamma(t) = \hat{U}(t,t_0)$ that satisfies Eq. (5).

Complete controllability implies that any kinematically admissible target state can be dynamically reached from the initial state by driving the system with a suitable control field. If the system is initially in a mixed state represented by a density matrix $\hat{\rho}_0$, then this means that any other kinematically admissible mixed state can be dynamically reached. Similarly, if the system is initially in a pure state represented by a normalized wave function $|\psi_0\rangle$ then complete controllability guarantees that every other pure state represented by a normalized wave-function $|\psi_1\rangle$ can be dynamically reached from the initial state. In Ref. [3] it is furthermore shown that complete controllability implies dynamical realizability of the universal kinematical bounds on the optimization of observables for (nondissipative) quantum systems.

It is apparent that if the dimension of the Lie algebra L_0 generated by the operators $\{\hat{H}_0,\ldots,\hat{H}_M\}$, or more accurately, their skew-Hermitian counterparts $\{i\hat{H}_0,\ldots,i\hat{H}_M\}$ is N^2 then L_0 is the Lie algebra of skew-Hermitian $N \times N$ matrices u(N). Ramakrishna *et al.* have shown in Ref. [7], using results by Jurdjevic and Sussmann [8], that in this case the dynamical Lie group of the system is the unitary group U(N). Noting that the dimension of u(N) is N^2 and that any Lie algebra of skew-Hermitian $N \times N$ matrices of dimension N^2 is (isomorphic to) u(N) we have, therefore, the following.

Theorem 1 (Ramakrishna et al.). A necessary and sufficient condition for complete controllability of a quantum system $\hat{H} = \hat{H}_0 + \hat{H}_I$ with $\hat{H}_0$ and $\hat{H}_I$ as in Eqs. (1) and (2) is that the Lie algebra L_0 has dimension N^2.

This theorem provides a condition for complete controllability of a quantum system that can easily be verified by computing the Lie algebra generated by $\hat{H}_0,\ldots,\hat{H}_M$ and determining its dimension.

A basic algorithm for constructing a basis for the Lie algebra L_0 in terms of iterated commutators is presented in Table I. It can be optimized to increase the speed of the computation and to improve the accuracy of the numerical results. In Table I we use $\hat{H}$ to denote a $N \times N$ matrix and using the fact that a matrix can also be interpreted as a vector, **H** for the N^2 column vector obtained by concatenating the columns of $\hat{H}$ vertically. W is a $N^2 \times N^2$ matrix whose columns $W_{:,j}$ represent the basis elements of L_0. Note that $\hat{W}_{:,j}$ is the jth basis element interpreted as $N \times N$ matrix.

To initialize W, we start with $W_{:,1} = \mathbf{H}_0$ and add $\mathbf{H}_m$ for $m = 1,\ldots,M$ provided that the additional column increases the rank of W. This guarantees that W initially consists of linearly independent generators of L_0. To construct a basis for L_0, we compute all possible commutators of the columns $W_{:,j}$ of W, interpreted as $N \times N$ matrices. Whenever a commutator is linearly independent of the columns of W, we add the commutator as a new column to W. Note that if we add a new column to W then we also have to compute the commu-

TABLE I. Algorithm to compute the Lie algebra generated by a control system $\{\hat{H}_0,\ldots,\hat{H}_M\}$.

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let $W = \mathbf{H}_0$
let $r = \text{rank}(W)$
for $m = 2, \ldots, M+1$ **do**
  if $\text{rank}([W,\mathbf{H}_m]) > r$ **then**
    append W by column vector $\mathbf{H}_m$
    $r = r+1$
  endif
endfor
let $r_o = 0$
let $r_n = \text{rank}(W)$
repeat
  for $l = r_o+1, \ldots, r_n$ **do**
    for $j = 1, \ldots, l-1$ **do**
      let $\hat{h} = [\hat{W}_{:,l}, \hat{W}_{:,j}]$
      if $\text{rank}([W,\mathbf{h}]) > r$ **then**
        append W by column vector $\mathbf{h}$
        $r = r+1$
      endif
    endfor
  endfor
  let $r_o = r_n$
  let $r_n = \text{rank}(W)$
until $r_n = r_o$ **or** $r = N^2$

TABLE II. Dimensions of the Lie algebra.

	Dim. N	2	3	4	5	6	7	8
Morse	$d_n = \sqrt{n}$	4	9	16	25	36	49	64
	$d_n = 1$	4	9	16	25	36	49	64
	$d_n = 1, d_{N-1} = 2$	4	9	16	25	36	49	64
Harmonic	$d_n = \sqrt{n}$	4	9	16	25	36	49	64
	$d_n = 1$	4	4	11	11	22	22	37
	$d_n = 1, d_{N-1} = 2$	4	9	16	25	36	49	64

tator of the matrix represented by the new column with all the matrices represented by the old columns of W. Hence, we repeat computing the commutators of the basis elements represented by W until no new columns have been added in the previous step or the rank of W reaches the maximum of N^2.

IV. CONTROLLABILITY CALCULATIONS

We implemented the algorithm presented in the previous section and computed the dimension of the dynamical Lie algebra for the system $\hat{H} = \hat{H}_0 + f(t)\hat{H}_1$ with $\hat{H}_0$ and $\hat{H}_1$ as in Eqs. (1) and (3) for various choices of the energy levels E_n and the transition dipole moments d_n. In particular, we studied the N-level harmonic oscillator with energy levels

$$E_n = n - \frac{1}{2}, \tag{8}$$

and the N-level Morse oscillator with energy levels

$$E_n = \left(n - \frac{1}{2}\right)\left[1 - \frac{1}{2}B\left(n - \frac{1}{2}\right)\right], \tag{9}$$

where B is a (usually small) positive real number. In our numerical computations we used $B = 0.0419$, which corresponds to a Morse oscillator model of the molecular bond for hydrogen fluoride [9]. We computed the dimension of the Lie algebra L_0 for systems with varying dimension N and for

different choices of the transition dipole moments d_n. The results of some of these computations are presented in Table II.

For the Morse oscillator system we observe that the dimension of L_0 is always N^2, i.e., it is completely controllable independent of the choice of the d_n (as long as the d_n are nonzero).

For the harmonic oscillator, however, the dimension of L_0 depends on the choice of the transition dipole moments. For the usual choice, $d_n = \sqrt{n}$, the dimension of L_0 is N^2, i.e., the system is completely controllable. However, if we chose all the d_n to be equal, e.g., $d_n = 1$ for $n = 1, \ldots, N-1$, then the dimension of the Lie algebra L_0 is less than N^2 and the system is therefore *not* completely controllable for $N > 2$. It is also worth noting that a slight modification of the d_n is sufficient in this case to restore complete controllability: if we choose $d_n = 1$, $n = 1, \ldots, N-2$ and $d_{N-1} = 2$ then the dimension of L_0 is again N^2.

The extensive data we gathered strongly suggested that any Morse oscillator system with nonzero transition dipole moments, i.e., $d_n \neq 0$, $n = 1, \ldots, N-1$, is completely controllable for any N, while complete controllability for a harmonic oscillator seemed to depend on the values of the transition dipole moments d_n. These observations prompted us to study the issue of controllability systematically using Lie algebra techniques.

V. RESULTS FROM LIE ALGEBRA THEORY

In order to prove our conjectures about complete controllability based on numerical evidence, a few general results from the theory of Lie algebras are required. For more detailed information about Lie algebras and Lie groups, the reader is referred to Refs. [10–13] or any other book on the subject.

We first observe that $u(N) = su(N) \oplus u(1)$, where $su(N)$ is the Lie algebra of traceless skew-Hermitian $N \times N$ matrices. If the diagonal elements $d_{m,m}$ of the interaction operators $\hat{H}_m$ for $m > 0$ are zero, as is the case in the dipole approximation, then the interaction operators $\hat{H}_m$ are represented by traceless Hermitian matrices, i.e., $i\hat{H}_m \in su(N)$ for $m > 0$. If the internal Hamiltonian $\hat{H}_0$ is traceless as well, i.e., $i\hat{H}_0 \in su(N)$, then the dynamical Lie algebra L_0 generated by $i\hat{H}_0$ and $i\hat{H}_m$ must be $su(N)$, or a subalgebra of $su(N)$, since the commutator of two traceless skew-Hermitian matrices is always a traceless skew-Hermitian ma-

trix. By our strict definition of complete controllability, a system whose dynamical Lie algebra is $su(N)$ is *not* completely controllable since its dynamical Lie group is $SU(N)$, i.e., the Lie group of unitary $N \times N$ matrices with determinant one, and $SU(N)$ is a proper subgroup of $U(N)$, the Lie group of all unitary $N \times N$ matrices. [For a discussion of the practical significance of the difference between $su(N)$ and $u(N)$ see appendix A] On the other hand, we have the following useful result.

Lemma 1. If the dynamical Lie algebra L_0 contains $su(N)$ and $\hat{H}_0$ has nonzero trace then we have $L_0 = su(N) \oplus u(1) \simeq u(N)$.

Proof. Note that we can write

$$\hat{H}_0 = \frac{1}{N} \mathrm{Tr}(\hat{H}_0) \hat{I} + \hat{H}_0' \qquad (10)$$

where $i\hat{H}_0' \in su(N)$. $i\hat{H}_0$ is in L_0 by definition. Since L_0 contains $su(N)$, and hence, $i\hat{H}_0'$, it must also contain the identity matrix $i\hat{I} = (\hat{H}_0 - \hat{H}_0') N / \mathrm{Tr}(\hat{H}_0)$. Hence, noting that $\hat{I}$ generates a one-dimensional Lie algebra isomorphic to $u(1)$ we have indeed $L_0 = su(N) \oplus u(1)$. ∎

Thus, in order to show that a system is completely controllable we only need to show that $\mathrm{Tr}(\hat{H}_0) \neq 0$ and that L_0 contains $su(N)$.

To verify that L_0 contains $su(N)$ we need a complete set of generators for the Lie algebra $su(N)$. Let $\hat{e}_{n,n'}$ be the $N \times N$ matrix such that the element in the nth row and n'th column is 1 while all other elements are 0, i.e.,

$$(\hat{e}_{n,n'})_{ij} = \delta_{in} \delta_{jn}, \qquad (11)$$

where δ_{ij} is the Kronecker symbol. One can easily see that any traceless skew-Hermitian matrix must be a real linear combination of the $N^2 - 1$ basic matrices

$$\hat{e}_{n,n'}^R = \hat{e}_{n,n'} - \hat{e}_{n',n} \quad 1 \leq n \leq N-1, \ n < n' \leq N$$

$$\hat{e}_{n,n'}^I = i(\hat{e}_{n,n'} + \hat{e}_{n',n}) \quad 1 \leq n \leq N-1, \ n < n' \leq N \quad (12)$$

$$\hat{h}_n = \hat{e}_{nn} - \hat{e}_{n+1,n+1} \quad 1 \leq n \leq N-1.$$

However, verifying that L_0 contains all of the $N^2 - 1$ basis elements would be quite tedious. Fortunately, this is not necessary.

Lemma 2. The skew-Hermitian $N \times N$ matrices $\hat{e}_{n,n+1}^R$ and $\hat{e}_{n,n+1}^I$ $1 \leq n < N$, generate the Lie algebra $su(N)$.

Proof. Using the relation

$$\hat{e}_{n,n'} \hat{e}_{m,m'} = \hat{e}_{n,m'} \delta_{n'm},$$

which follows from the definition of $\hat{e}_{n,n'}$, it can be verified by direct computation that the skew-Hermitian matrices $\hat{e}_{n,n'}^R$ and $\hat{e}_{n,n'}^I$ satisfy the equations

$$[\hat{e}_{n,n'}^R, \hat{e}_{n',n''}^R] = \hat{e}_{n,n''}^R \quad n'' \neq n,$$

$$[\hat{e}_{n,n'}^R, \hat{e}_{n',n''}^I] = \hat{e}_{n,n''}^I \quad n'' \neq n, \qquad (13)$$

$$[\hat{e}_{n,n'}^R, \hat{e}_{n',n}^I] = 2i(\hat{e}_{n,n} - \hat{e}_{n',n'}),$$

and a bit of algebra therefore shows that the $2(N-1)$ elements $\hat{e}_{n,n+1}^R$ and $\hat{e}_{n,n+1}^I$ for $1 \leq n \leq N-1$ generate the entire Lie algebra $su(N)$. ∎

Hence, if L_0 contains the generators $\hat{e}_{n,n+1}^R$ and $\hat{e}_{n,n+1}^I$ for $1 \leq n \leq N-1$, then L_0 must contain $su(N)$. Thus, given a system whose energy levels are well-enough separated to permit selective control of each transition between adjacent energy levels through frequency discrimination, i.e.,

$$i\hat{H} = i\hat{H}_0 + \sum_{n=1}^{N-1} f_n(t) \cos(\mu_n t) i\hat{H}_1,$$

with $\hat{H}_0$ as in (1), $\mu_n = E_n - E_{n+1}$ and

$$i\hat{H}_n = i d_n(|n\rangle\langle n+1| + |n+1\rangle\langle n|) = d_n \hat{e}_{n,n+1}^I,$$

where $f_n(t)$ is slowly time-varying compared to $\cos(\mu_n t)$ and $d_n \neq 0$, we can conclude immediately that the Lie algebra of the system contains $su(N)$, and hence, that the system is completely controllable [if $\mathrm{Tr}(\hat{H}_0) \neq 0$]. To see this, note that the generators $\hat{e}_{n,n+1}^I$ are given and the generators $\hat{e}_{n,n+1}^R$ can be obtained by computing the commutators

$$\mu_n^{-1}[i\hat{H}_0, \hat{e}_{n,n+1}^I] = \hat{e}_{n,n+1}^R.$$

However, selective control of individual transitions between adjacent energy levels through frequency discrimination is not always possible. For instance, as pointed out earlier, it fails when the energy levels are equally spaced or degenerate, and it may not be a good approach for systems with nearly equally spaced energy levels, such as Morse oscillators. Furthermore, even if it is possible to use multiple pulses to selectively control individual transitions, one may not wish to do so. Instead, one may, for instance, wish to control the system with a single optimally shaped control pulse obtained using an efficient optimal control algorithm [14–16]

In order to establish criteria for complete controllability of N-level systems subject to a single control field that drives all permitted transitions concurrently, we need another lemma, which makes use of the dipole form of $\hat{H}_1$.

Lemma 3. If $\hat{H}_1$ has the special form (3), i.e.,

$$i\hat{H}_1 = \sum_{n=1}^{N-1} d_n \hat{e}_{n,n+1}^I, \qquad (14)$$

then it suffices to show that L_0 contains the pair of generators $\hat{e}_{12}^R$ and $\hat{e}_{12}^I$ or $\hat{e}_{N-1,N}^R$, and $\hat{e}_{N-1,N}^I$, i.e., if L_0 contains either of these two pairs of generators, then it contains all the generators of $su(N)$.

Proof. If $\hat{e}_{12}^R, \hat{e}_{12}^I \in L_0$ then

$$\hat{h}_1 = \tfrac{1}{2}[\hat{e}_{12}^R, \hat{e}_{21}^I] = i(\hat{e}_{11} - \hat{e}_{22}) \in L_0,$$

$$\hat{V}_1 = i\hat{H}_1 - d_1\hat{e}_{12}^I = \sum_{n=2}^{N-1} d_n \tilde{e}_{n,n+1}^I \in L_0.$$

where $\hat{e}_{21}^I = \hat{e}_{12}^I$. This leads to

$$[\hat{h}_1, \hat{V}_1] = d_2\hat{e}_{23}^R \in L_0,$$

$$-[\hat{h}_1, [\hat{h}_1, \hat{V}_1]] = d_2\hat{e}_{23}^I \in L_0.$$

Since $d_2 \neq 0$ by hypothesis, it follows that $\hat{e}_{23}^R, \hat{e}_{23}^I \in L_0$. Repeating this procedure $N-2$ times shows that all the generators $\hat{e}_{n,n+1}^R$ and $\hat{e}_{n,n+1}^I$ for $1 \leq n < N$ are in L_0. Similarly, we can show that $\hat{e}_{N-1,N}^R$ and $\hat{e}_{N-1,N}^I$ in L_0 implies that L_0 contains all the generators $\hat{e}_{n,n+1}^R$ and $\hat{e}_{n+1,n}^I$, $1 \leq n < N$. ∎

Thus, if $\hat{H}_1$ has the special form (3) then it suffices to show that $\hat{e}_{12}^R, \hat{e}_{12}^I \in L_0$ in order to conclude that L_0 is at least $su(N)$.

VI. COMPLETE CONTROLLABILITY FOR ANHARMONIC SYSTEMS

Let $\mu_n = E_n - E_{n+1}$ for $n = 1, \dots, N-1$ and $\hat{V} = i\hat{H}_1$.

Theorem 2. If $\mu_1 \neq 0$ and $\mu_n^2 \neq \mu_1^2$ for $n > 1$, then the dynamical Lie group of the system $\hat{H} = \hat{H}_0 + f(t)\hat{H}_1$ with $\hat{H}_0$ and $\hat{H}_1$ as defined in Eqs. (1) and (3) is at least SU(N). If, in addition, $\mathrm{Tr}(\hat{H}_0) \neq 0$, then the dynamical Lie group is U(N), i.e., the system is completely controllable.

Proof. We evaluate

$$[i\hat{H}_0, \hat{V}] = -\sum_{n=1}^{N-1} \mu_n d_n \hat{e}_{n,n+1}^R \equiv -\hat{V}',$$

$$[i\hat{H}_0, \hat{V}'] = \sum_{n=1}^{N-1} \mu_n^2 d_n \tilde{e}_{n,n+1}^I \equiv \hat{V}''.$$

Using $\hat{V}$ and $\hat{V}''$, we obtain $\hat{V}_1 \in L_0$, where

$$\hat{V}_1 \equiv \hat{V}'' - \mu_{N-1}^2 \hat{V} = \sum_{n=1}^{N-2} (\mu_n^2 - \mu_{N-1}^2) d_n \hat{e}_{n,n+1}^I.$$

Repeating the previous steps for $i\hat{H}_0$ and $\hat{V}_1$ leads to $\hat{V}_2 \in L_0$, where

$$\hat{V}_2 \equiv -[i\hat{H}_0, [i\hat{H}_0, \hat{V}_1]] - \mu_{N-2}^2 \hat{V}_1$$

$$= \sum_{n=1}^{N-3} (\mu_n^2 - \mu_{N-2}^2)(\mu_n^2 - \mu_{N-1}^2) d_n \hat{e}_{n,n+1}^I.$$

After $N-2$ iterations, we have $\hat{V}_{N-2} \in L_0$ where

$$\hat{V}_{N-2} \equiv d_1 \left[\prod_{n=2}^{N-1} (\mu_1^2 - \mu_n^2) \right] \hat{e}_{12}^I.$$

Since by hypothesis, $d_1 \Pi_{n=2}^{N-1}(\mu_1^2 - \mu_n^2) \neq 0$, this means $\hat{V}_{N-2}' = \hat{e}_{12}^I \in L_0$ and noting that $\mu_1 \neq 0$ we have

$$\hat{V}_{N-2}'' \equiv -\frac{1}{\mu_1}[i\hat{H}_0, \hat{V}_{N-2}'] = \hat{e}_{12}^R \in L_0.$$

The conclusion now follows from lemmas 1 and 3. ∎

This theorem shows that for anharmonic systems, complete controllability does not depend on the values of the transition dipole moments d_n (as long as they are nonzero). In particular, we have the following.

Corollary 1. A quantum system $\hat{H} = \hat{H}_0 + f(t)\hat{H}_1$ with $\hat{H}_0$ and $\hat{H}_1$ as in Eqs. (1) and (3), respectively, and E_n as in Eq. (9), i.e., a Morse oscillator, is completely controllable for arbitrary nonzero values of the transition dipole moments d_n.

It is worth noting that Theorem 2 also applies to some degenerate quantum systems.

Example 1. The system $\hat{H} = \hat{H}_0 + f(t)\hat{H}_1$ with

$$\hat{H}_0 = \mathrm{diag}\{E_1, E_2, \dots, E_2\}, \quad E_1 \neq E_2,$$

and $\hat{H}_1$ as in Eq. (3) is completely controllable by Theorem 2 despite the fact that energy level E_2 has multiplicity $N-1$. In fact, the proof of controllability is even simpler for this system as we have $\hat{V}'' = \mu_1^2 d_1' \hat{e}_{12}^I$ after only one step.

This example begs the question whether the system is still completely controllable if we choose

$$\hat{H}_0 = \mathrm{diag}\{\underbrace{E_1, \dots, E_1}_{N-1}, E_2\}, \quad E_1 \neq E_2,$$

instead. It is obvious that this system does not satisfy the technical condition $\mu_1 \neq 0$. However, it can easily be shown that this system is completely controllable by modifying the proof. In fact, this example is just a special case of the following.

Theorem 3. If $\mu_{N-1} \neq 0$ and $\mu_n^2 \neq \mu_{N-1}^2$ for $n < N-1$, then the dynamical Lie group of the system $\hat{H} = \hat{H}_0 + f(t)\hat{H}_1$ with $\hat{H}_0$ and $\hat{H}_1$ as defined in Eqs. (1) and (3), respectively, is at least SU(N). If, in addition, $\mathrm{Tr}(\hat{H}_0) \neq 0$, then the dynamical Lie group is U(N), i.e., the system is completely controllable.

The proof of this theorem is analogous to the proof of Theorem 2.

VII. COMPLETE CONTROLLABILITY FOR HARMONIC SYSTEMS

The condition $\mu_1^2 \neq \mu_n^2$ in the previous theorems excludes any system with equally spaced energy levels, such as a harmonic oscillator, for which we have $\mu_1 = \mu_2 = \cdots = \mu_{N-1}$. We shall assume that $\mu_1 \neq 0$ since for $\mu_1 = 0$, the system is completely degenerate with only one energy level.

In order to state our theorem, we need to introduce some technical parameters

$$v_n = \begin{cases} 2d_1^2 - d_2^2, & n = 1; \\ 2d_n^2 - d_{n-1}^2 - d_{n+1}^2, & n = 2, \dots, N-2; \\ 2d_{N-1}^2 - d_{N-2}^2 & n = N-1. \end{cases} \quad (15)$$

Observe that these technical parameters depend on the values of the transition dipole moments d_n.

Theorem 4. The dynamical Lie group for a quantum system $\hat{H} = \hat{H}_0 + f(t)\hat{H}_1$ with $\hat{H}_0$ and $\hat{H}_1$ as in Eqs. (1) and (3) and N equally spaced energy levels

$$E_n = E_1 + (n-1)\mu_1, \quad n = 1, \dots, N, \quad \mu_1 \neq 0,$$

is at least SU(N) if the parameters v_n satisfy one of the following conditions:

(1) $v_n \neq v_{N-1}$ for $1 \leq n \leq N-2$;
(2) $v_n \neq v_1$ for $2 \leq n \leq N-1$.

If, in addition, $\mathrm{Tr}(\hat{H}_0) \neq 0$, then the dynamical Lie group is U(N), i.e., the system is completely controllable.

Proof. Let $\hat{V} = i\hat{H}_1$. In this case, the element

$$\tilde{V} = -\mu_1^{-1}[i\hat{H}_0, \hat{V}] = \sum_{n=1}^{N-1} d_n \hat{e}_{n,n+1}^R,$$

is in L_0 and its sum and difference with $\hat{V}$ give rise to

$$\hat{V}_1^+ \equiv \sum_{n=1}^{N-1} d_n(\underbrace{\hat{e}_{n,n+1}^I + \hat{e}_{n,n+1}^R}_{\equiv \hat{e}_{n,n+1}^+}),$$

$$\hat{V}_1^- \equiv \sum_{n=1}^{N-1} d_n(\underbrace{\hat{e}_{n,n+1}^I - \hat{e}_{n,n+1}^R}_{\equiv \hat{e}_{n,n+1}^-}),$$

which, along with their commutator

$$\hat{V}_1^0 = \tfrac{1}{4}[\hat{V}_1^+, \hat{V}_1^{-1}]$$

$$= id_1^2 \hat{e}_{1,1} + \sum_{n=2}^{N-1} i(d_n^2 - d_{n-1}^2)\hat{e}_{n,n} - id_{N-1}^2 \hat{e}_{N,N},$$

are in L_0. Starting with $\hat{V}_1^0$ and $\hat{V}_1^+$, we have

$$[\hat{V}_1^0, \hat{V}_1^+] = \sum_{n=1}^{N-1} v_n d_n \hat{e}_{n,n+1}^+ \in L_0,$$

$$\hat{V}_2^+ \equiv [\hat{V}_1^0, \hat{V}_1^+] - v_1 \hat{V}_1^+ = \sum_{n=2}^{N-1} (v_n - v_1)d_n \hat{e}_{n,n+1}^+ \in L_0,$$

$$\hat{V}_3^+ \equiv [\hat{V}_1^0, \hat{V}_2^+] - v_2 \hat{V}_2^+$$

$$= \sum_{n=3}^{N-1} (v_n - v_2)(v_n - v_1)d_n \hat{e}_{n,n+1}^+ \in L_0,$$

$$\vdots$$

$$\hat{V}_{n-1}^+ \equiv [\hat{V}_1^0, \hat{V}_{N-2}^+] - v_{N-2}\hat{V}_{N-2}^+$$

$$= d_{N-1}\left[\prod_{n=1}^{N-2}(v_{N-1} - v_n)\right]\hat{e}_{N-1,N}^+ \in L_0.$$

Since by hypothesis, $v_{N-1} \neq v_n$ for $n = 1, 2, \dots, N-2$, we have $\hat{e}_{N-1,N}^+ \in L_0$.

Similarly, starting with $\hat{V}^0$ and $\hat{V}_1^-$, we can also prove $\hat{e}_{N-1,N}^- \in L_0$. This implies $\hat{e}_{N-1,N}^R$ and $\hat{e}_{N-1,N}^I$ are in L_0, and hence, the conclusion follows from Lemmas 1 and 3. ∎

Example 2. The standard N-level harmonic-oscillator system $\hat{H} = \hat{H}_0 + f(t)\hat{H}_1$ with $\hat{H}_0$ and $\hat{H}_1$ as in Eqs. (1) and (3), $E_n = n - 1/2$ and $d_n = \sqrt{n}$ is completely controllable by Theorem 4, since we have

$$v_1 = v_2 = \dots = v_{N-2} = 0, \quad v_{N-1} = N,$$

i.e., the v_n satisfy $v_{N-1} - v_n = N$ for $n = 1, \dots, N-2$.

Example 3. However, the N-level harmonic-oscillator system $\hat{H} = \hat{H}_0 + f(t)\hat{H}_1$ with $\hat{H}_0$ and $\hat{H}_1$ as in Eqs. (1) and (3), $E_n = n - 1/2$ and $d_n = 1$ for $n = 1, \dots, N-1$ does not satisfy the hypothesis of Theorem 4, since we have

$$v_1 = 1, \quad v_2 = \dots = v_{N-2} = 0, \quad v_{N-1} = 1,$$

i.e., $v_{N-1} = v_1$. Numerical calculations for various N confirm that this system is indeed not completely controllable (See Table II).

For $N = 4$ one can, e.g., verify either numerically or by analyzing the Lie algebra that the simple skew-Hermitian matrix

$$\hat{e}_{12}^I = \begin{pmatrix} 0 & i & 0 & 0 \\ i & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}$$

is not in L_0 and therefore the unitary operator

$$\hat{U}(\theta) = \exp(\theta \hat{e}_{12}^I) = \begin{pmatrix} \cos(\theta) & i\sin(\theta) & 0 & 0 \\ i\sin(\theta) & \cos(\theta) & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix},$$

cannot be dynamically generated for this system for any $\theta \in (0, 2\pi)$. Thus, if the system is initially in state $\hat{\rho}_0 = \sum_{n=1}^4 w_n |n\rangle\langle n|$, then the target state

$$\hat{\rho}(t_F) = \hat{U}(\theta)\hat{\rho}_0 \hat{U}(\theta)^\dagger,$$

is generally not dynamically accessible since it is impossible to put the energy eigenstates $|1\rangle$ and $|2\rangle$ into superposition without equally mixing the states $|3\rangle$ and $|4\rangle$ of the initial ensemble.

Example 4. The N-level harmonic-oscillator system $\hat{H} = \hat{H}_0 + f(t)\hat{H}_1$ with $\hat{H}_0$ and $\hat{H}_1$ as in Eqs. (1) and (3), E_n

$=n-1/2$, $d_n=1$ for $n=1,\ldots,N-2$, but $d_{N-1}=2$, however, does satisfy the hypothesis of Theorem 4 since we have

$$v_1=1, \quad v_2=\cdots=v_{N-3}=0, \quad v_{N-2}=-3, \quad v_{N-1}=7,$$

i.e., $v_n\neq v_1$ for $n\neq 1$. Numerical calculations for various N confirm that this system is completely controllable (see Table II).

The rather surprising results of the previous two examples can be understood by analyzing the Lie algebra generated by $\hat{H}_0$ and $\hat{H}_1$ in both cases. Although the details of this analysis are beyond the scope of this paper (and will be discussed in a future paper) we would like to mention here that for a harmonic system with transition dipole moments satisfying the "symmetry" relation $d_{N-n}=\pm d_n$ for $1\leq n\leq N/2$, the transitions $n\to n+1$ and $N-n\to N+1-n$ for $1\leq n\leq N/2$ become coupled. This leads to a collapse of the Lie algebra and loss of complete controllability, which can be restored by breaking the symmetry in the transition dipole moments. This is why changing $d_{N-1}=1$ to $d_{N-1}=2$ restored controllability in the last example. In fact, changing, d_{N-1} to any value other than ± 1 would work as well.

VIII. CONCLUSION

The question of complete controllability of quantum systems using external control fields has been addressed before by various authors and it is, for instance, well known that a quantum system is completely controllable if it is possible to address a sufficiently large set of single transitions using multiple frequency-selective control pulses. However, many optimization strategies attempt to find a single control pulse that addresses all transitions concurrently to achieve the control objective. Furthermore, control based on frequency discrimination is not always possible, e.g., it is not suitable for systems with equally or almost equally spaced or degenerate energy levels.

Despite the relative importance of control strategies involving a single control pulse, sufficient criteria for complete controllability in this case have so far been missing. In this paper, we addressed this problem and established general criteria for complete controllability of quantum systems subject to a single control pulse.

In particular, we showed that most anharmonic, nondecomposable quantum systems are completely controllable using a single control that drives all the transitions concurrently, independent of the values of the transition dipole moments d_n. For quantum systems with equally spaced energy levels, we demonstrated that complete controllability depends on the values of the transition dipole moments d_n and derived conditions that guarantee complete controllability.

We verified that the standard truncated harmonic oscillator with transition dipole moments $d_n=\sqrt{n}$ satisfies these conditions and gave examples of harmonic systems that do not satisfy the conditions. In the latter case, we also checked by direct computation of the Lie algebra that they are not completely controllable and showed that there are certain unitary operators that cannot be dynamically realized for these systems.

APPENDIX: CONTROLLABILITY AND THE QUESTION OF TRACE

All the theorems about complete controllability presented in this paper require that the trace of $\hat{H}_0$ be nonzero. The mathematical necessity of this hypothesis is obvious: a set of traceless skew-Hermitian $N\times N$ matrices cannot generate all of u(N) but at most su(N). Hence, assuming that the interaction terms $\hat{H}_m$, $m>0$, are represented by traceless skew-Hermitian matrices, if $\hat{H}_0$ is traceless as well, then the dynamical Lie group can be at most SU(N), the set of unitary matrices with determinant one, which is a proper subgroup of U(N). However, our definition of complete controllability requires that all unitary matrices be dynamically accessible.

Nevertheless, the trace condition is physically somewhat disturbing since the energy levels of a physical system are generally only determined up to a constant, and hence, the trace of $\hat{H}_0$ seems physically rather insignificant as one can always make it either zero or nonzero by shifting the energy levels of the system by a constant. We shall attempt to resolve this apparent conflict by showing that the difference between SU(N) and U(N) is only a phase factor.

Let the initial state of the system be represented by the normalized wave function $|\psi_0\rangle$. If the dynamical Lie group of the system is U(N) then any other pure state represented by normalized wave function $|\psi_1\rangle$ is dynamically reachable since given any two normalized wave functions, there always exists a (not necessarily unique) unitary transformation $\hat{U}$ such that $|\psi_1\rangle=\hat{U}|\psi_0\rangle$ and we can find a path $\gamma(t)=\hat{U}(t,t_0)$ in U(N) such that $\gamma(t_0)=\hat{I}$ and $\gamma(t_F)=\hat{U}$. Since the determinant of a unitary operator is a complex number of modulus 1, we can write $\det[\gamma(t)]=e^{i\phi(t)}$. Noting that $\det(\alpha A)=\alpha^N\det(A)$ where N is the size of A, we see immediately that $\tilde{\gamma}(t)\equiv e^{-i\phi(t)/N}\gamma(t)$ has determinant 1

$$\det[\tilde{\gamma}(t)]=(e^{-i\phi(t)/N})^N\det[\gamma(t)]=e^{-i\phi(t)}e^{i\phi(t)}\equiv 1,$$

and thus defines a path in SU(N). Furthermore,

$$\tilde{\gamma}(t_F)|\psi_0\rangle=e^{-i\phi/N}\hat{U}|\psi_0\rangle=e^{-i\phi/N}|\psi_1\rangle,$$

i.e., $\tilde{\gamma}(t_F)|\psi_0\rangle$ and $|\psi_1\rangle$ differ only by a phase factor. Hence, if the dynamical Lie group is SU(N) then we loose control over the phase of the state, otherwise there is no difference.

Thus, for practical applications that do not require phase control, one need not worry about the trace. For instance, if the goal of controlling the system is to maximize the expectation value of an observable $\hat{A}$ at a target time t_F,

$$\langle\hat{A}(t_F)\rangle=\langle\psi(t_F)|\hat{A}|\psi(t_F)\rangle,$$

then clearly the phases of the target states are irrelevant as they are cancelled out by computing the expectation value anyway.

Moreover, if the initial state is given by a density matrix $\hat{\rho}_0$ then any target state $\hat{\rho}(t_F)$ that is dynamically accessible via a path in U(N) is also dynamically accessible via a path in SU(N). To see this, let $\gamma(t) = \hat{U}(t,t_0)$ be a path in U(N) such that

$$\hat{\rho}(t_F) = \hat{U}(t_F,t_0)\hat{\rho}_0\hat{U}(t_F,t_0)^t. \tag{A1}$$

Again, we have $\det[\gamma(t)] = e^{i\phi(t)}$ and $\tilde{\gamma}(t) = e^{-i\phi(t)/N}\gamma(t)$ de-

fines a path in SU(N) that is equivalent to $\gamma(t)$ since [17]

$$\begin{aligned}\hat{\rho}(t) &= \gamma(t)\hat{\rho}_0\gamma(t)^\dagger = e^{i\phi(t)/N}\tilde{\gamma}(t)\hat{\rho}_0\tilde{\gamma}(t)^\dagger e^{-i\phi(t)/N} \\ &= \tilde{\gamma}(t)\hat{\rho}_0\tilde{\gamma}(t)^\dagger,\end{aligned}$$

i.e., the phase factors $e^{-i\phi(t)/N}$ cancel out completely.

However, there are some applications of control in quantum computation where it is important to have phase control and when SU(N) is not adequate. Therefore, we have chosen to require the dynamical Lie group to be U(N) for complete controllability.

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[17] Note that if A is unitary then $\det(A^\dagger) = [\det(A)]^{-1}$, i.e., $\det[\gamma(t)^\dagger] = e^{-i\phi(t)}$.