Controllability of multi-partite quantum systems and selective excitation of quantum dots

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Abstract. We consider the degrees of controllability of multi-partite quantum systems, as well as necessary and sufficient criteria for each case. The results are applied to the problem of simultaneous control of an ensemble of quantum dots with a single laser pulse. Finally, we apply optimal control techniques to demonstrate selective excitation of individual dots for a simultaneously controllable ensemble of quantum dots.

1. Introduction

Control of quantum processes is essential to realize the vast potential of quantum technology ranging from applications in quantum information processing [1] to atomic and molecular physics and chemistry [2]. Although robust and efficient control of quantum phenomena remains a challenge, especially in practice, recent advances in theory and technology have made control of systems at the quantum level increasingly feasible [3].

To achieve the best possible control outcomes, it is crucial to know the degree to which a quantum system is controllable, to understand fundamental limits on control, and to find optimal ways to implement control given certain constraints. Although significant progress has been made recently with regard to refining the notions of and criteria for controllability of quantum systems [4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16], understanding constraints [17, 18, 19], and developing techniques for optimal control field design [20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31], many interesting questions remain.

Among these are the controllability and optimal control of multi-partite systems, i.e., systems comprised of $L$ distinct quantum elements such as quantum dots or molecules. The obvious questions to ask in this case include (a) whether each of
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the components is controllable individually, and (b) whether the composite system is controllable as a whole. In practice, however, one often faces more subtle questions. For instance, given an ensemble of quantum dots [32], with negligible interdot coupling but clustered too close together to allow (spatial) addressing of individual dots, when is it possible to selectively excite individual dots in the ensemble? What are appropriate notions of controllability? How can we achieve selective excitation in practice?

To address these questions, we first review existing notions of controllability for (finite-dimensional) quantum systems (Sec. 2). We then proceed to consider the application of these notions to multi-partite systems, introduce the notion of simultaneous controllability (Sec. 3), and discuss necessary and sufficient criteria for the latter notion (Sec. 4). Finally, the results will be applied to the problem of simultaneous controllability of ensembles of quantum dots (Sec. 5) and selective excitation of individual dots using optimal control (Sec. 6).

2. Notions of Controllability for Quantum Systems

We will restrict ourselves here to discussing basic notions of controllability for finite-dimensional, Hamiltonian quantum systems subject to open-loop coherent control, i.e., a system whose evolution is governed by the quantum Liouville equation

\[ i\hbar \frac{d}{dt} \hat{\rho}(t) = \hat{H}[f(t)]\hat{\rho}(t) - \hat{\rho}(t)\hat{H}[f(t)], \] (1)

where \( \hat{\rho}(t) \) is the density operator representing the state of the system, and \( \hat{H}[f(t)] \) is the total Hamiltonian, which depends on various control fields \( f = (f_1, \ldots, f_M) \), e.g.,

\[ \hat{H}[f(t)] = \hat{H}_0 + \sum_{m=1}^{M} f_m(t)\hat{H}_m. \] (2)

The strongest possible requirement in terms of controllability, sometimes referred to as complete controllability [11], is the ability to create any dynamical evolution, which for a Hamiltonian system is equivalent to the ability to dynamically generate any unitary operator in \( U(N) \), where \( N \) is the dimension of the system’s Hilbert space, by applying a suitable control field \( f(t) \).

Another common notion is mixed-state controllability, which requires that the system can be driven from any given (pure or mixed) initial state \( \hat{\rho}_0 \), to any kinematically equivalent state \( \hat{\rho}_1 = \hat{U}\hat{\rho}_0\hat{U}^T \), \( \hat{U} \) being a unitary operator in \( U(N) \), by applying a suitable control field \( f(t) \). Mixed-state controllability also implies observable controllability [14].

If the initial state of the system is known to be pure, one may instead consider the weaker requirement of pure-state controllability—which simply requires that the system can be driven from any pure initial state \( \hat{\rho}_0 = |\Psi\rangle\langle\Psi| \) to any other pure state by applying a suitable control field \( f(t) \).

Considerable work has been done to find necessary and sufficient conditions for each of these concepts of controllability, especially in terms of the dynamical Lie algebra \( \mathcal{L} \) generated by the skew-Hermitian matrices \( i\hat{H}_m, m = 0, \ldots, M \) (see e.g. [11, 12, 13, 14, 15]). The key results can be summarized in the following theorem:

**Theorem 1. (Degrees of Controllability)** The system defined by Eqs (1), (2) is

- completely controllable if and only if \( \mathcal{L} \equiv u(N) \), the Lie algebra of skew-Hermitian \( N \times N \) matrices;
• mixed-state controllable if and only if $L \equiv \mathfrak{u}(N)$ or $L \equiv \mathfrak{su}(N)$, the Lie algebra of traceless skew-Hermitian $N \times N$ matrices;
• pure-state controllable if and only if $L$ is $\mathfrak{u}(N)$, $\mathfrak{su}(N)$, or if $N$ is even, the symplectic Lie algebra $\mathfrak{sp}(N/2)$ or $\mathfrak{sp}(N/2) \oplus \mathfrak{u}(1)$.

Noting that $\dim_{\mathbb{R}} \mathfrak{u}(N) = N^2$, $\dim_{\mathbb{R}} \mathfrak{su}(N) = N^2 - 1$ and $\dim_{\mathbb{R}} \mathfrak{sp}(N/2) = N(N + 1)/2$, these Lie-algebraic criteria allow us to easily verify the degree of controllability of a system by simply computing the dimension of the dynamical Lie algebra.

Mixed-state controllability appears to be the most important notion of controllability as it implies pure-state controllability for Hamiltonian systems, and unlike pure-state controllability, can be extended to open systems and systems subject to feedback control—although the definition needs to be modified slightly in the latter cases. Hence, we shall refer to mixed-state controllable systems simply as controllable in the following. For a discussion of notions of controllability for quantum systems subject to feedback control we refer the reader to [6]. For open systems whose evolution is governed by a dynamical semi-group we draw attention to the related concept of the set of states that are reachable from a given initial state at a certain target time by applying suitable control fields [4, 33].

3. Notions of Controllability of Multi-partite Systems

The degrees of controllability defined above obviously apply to any quantum system, including composite or multi-partite systems. However, in the latter case it is useful to introduce additional notions of controllability.

For instance, consider a system of $L$ particles or quantum units such as different molecules or quantum dots. If the Hilbert space of particle $\ell$ is $\mathcal{H}_\ell$ then the Hilbert space of the composite system is usually the tensor product space $\mathcal{H} = \mathcal{H}_1 \otimes \ldots \otimes \mathcal{H}_L$, and if $\dim \mathcal{H}_\ell = N_\ell$ then the dimension of the tensor product space is $N = N_1 \cdots N_L$. Hence, by Theorem 1 the composite system is controllable if and only if the dynamical Lie algebra generated by the system and control Hamiltonians $\hat{H}_0$ and $\hat{H}_m$, $m = 1, \ldots, M$, is $\mathfrak{u}(N)$ or $\mathfrak{su}(N)$. However, the tensor product space may be huge and not always the most appropriate Hilbert space for the system.

If the particles do not interact with each other, for example, then we can reduce the Hilbert space of the composite system to the direct sum of the Hilbert spaces $\mathcal{H}_\ell$ of the non-interacting parts rather than the tensor product. This situation arises in practice in quantum chemistry, in particular laser control of chemical reactions, where we may want to control several different types of molecules in a dilute solution, all of which interact simultaneously with an external control field such as a laser pulse, but for which intermolecular interactions are negligible [34]. The same problems are encountered for ensembles of quantum dots close enough together to prevent selective addressing of a single dot with a laser, but with negligible interdot coupling (e.g., due to different dot sizes etc.) We will refer to this case of a quantum system consisting of multiple non-interacting quantum units as decomposable or separable. Clearly, such a system is not controllable or even pure-state controllable in the strong sense defined in Sec. 2.

Appropriate notions of controllability in this case are the individual controllability of the components (separate quantum units) and their simultaneous controllability. The latter is a stronger notion than the former as individual controllability of each
unit does not imply that we can simultaneously control all units. For example, even if we could completely control each dot in a cluster of quantum dots individually, this does not necessarily mean that we can simultaneously control all the dots in the cluster with the same control pulse or pulse sequence. This naturally prompts the question as to when the components of a separable quantum system are simultaneously controllable.

### 4. Criteria for Simultaneous Controllability

The Hilbert space of a quantum system that consists of \( L \) non-interacting subsystems can be represented as the direct sum \( \mathcal{H} = \mathcal{H}_1 \oplus \ldots \oplus \mathcal{H}_L \) of the Hilbert spaces \( \mathcal{H}_\ell \) of the independent subsystems, \( \ell = 1, \ldots, L \). Thus, with respect to a suitable basis, the state \( \hat{\rho}(t) \) and the Hamiltonians \( \hat{H}_m, 0 \leq m \leq M \), have a block-diagonal structure

\[
\hat{\rho}(t) = \text{diag}(\hat{\rho}_1(t), \ldots, \hat{\rho}_L(t)) \\
\hat{H}_m = \text{diag}(\hat{H}_{m,1}, \ldots, \hat{H}_{m,L}), \quad m = 0, 1, \ldots, M
\]

where \( \hat{\rho}_\ell(t) \) and \( \hat{H}_{m\ell} \) are \( N_\ell \times N_\ell \) matrices and \( N_\ell = \dim \mathcal{H}_\ell \). Note that we require at least two Hamiltonians, \( \hat{H}_0 \) and \( \hat{H}_1 \), but in some cases it is necessary or at least advantageous to have more control Hamiltonians. For example, given an ensemble of optically controlled quantum dots that is sufficiently large, it may be possible to focus the laser beam on a certain region, which would allow us to define a separate control Hamiltonian for each independently accessible region. In other cases, e.g., for an ensemble of quantum dots whose internal energy level structure requires control fields with different polarizations, multiple control Hamiltonians may be essential.

It is obvious from this structure of the dynamical generators in Eq. (3) that the dynamical Lie group of the system must be contained in \( U(N_1) \times \cdots \times U(N_L) \), and that maximal orbits for a generic mixed state \[35\] under the action of this Lie group are thus homeomorphic to \( U(N_1) \times \cdots \times U(N_L)/[U(1) \times \cdots \times U(1)] \), where there are \( N \) terms in the denominator. The latter is equivalent to \( SU(N_1) \times \cdots \times SU(N_L)/[U(1) \times \cdots \times U(1)] \) where we have \( N - L \) terms in the denominator. These considerations lead to the following criterion (see Appendix A):

**Theorem 2. (Simultaneous Controllability)** The independent components of a decomposable system with Hamiltonian [4] of the form [3] are simultaneously mixed-state controllable if and only if the dimension of the dynamical Lie algebra \( \mathcal{L} \) is

\[
\dim \mathcal{L} = r + \sum_{\ell=1}^L (N_\ell^2 - 1),
\]

where \( r \) is the rank of matrix \( A \) of Eq. (A.4).

This result is a generalization of the sufficient criteria in [36] for arbitrary \( M \). Specifically, note that if we have only two Hamiltonians \( \hat{H}_0 \) and \( \hat{H}_1 \) of the form [9], i.e., \( M = 1 \), then we have \( r = 0 \) if the diagonal generators \( \hat{D}_0 \) and \( \hat{D}_1 \) defined in Appendix A are both 0, \( r = 1 \) if they are linearly dependent, and \( r = 2 \) if they are linearly independent, i.e., we recover the sufficient criteria of Theorem 2 in [36]. Furthermore, our derivation in Appendix A also shows that the Lie algebra dimension condition in Theorem 2 is both necessary and sufficient, at least for generic mixed-state controllability.

Finally, a similar argument shows that a necessary and sufficient condition for the weaker notion of *simultaneous pure-state controllability*—which is sufficient if each
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Figure 1. Five-dot ensemble: If dots 1 and 4 are identical the ensemble is not simultaneously controllable if the laser is centered in position C. However, if we can create two regions A and B, which can be separately addressed by the laser, then the ensemble becomes simultaneously controllable.

subsystem is known to be in a pure quantum state initially and evolves unitarily—is that the Lie algebra \( \tilde{L} \) (see Appendix A) be a direct sum of \( L \) terms, where the \( \ell \)th terms is either \( \text{su}(N\ell) \), or if \( N\ell \) is even, \( \text{sp}(N\ell/2) \).

5. Controllability of Ensembles of Quantum Dots

Consider again the example of an ensemble of \( L \) quantum dots, each of which is itself an \( N\ell \)-dimensional quantum system. According to Theorem 1 each of the dots is individually controllable if its associated dynamical Lie algebra \( L\ell \) has (at least) dimension \( N^2\ell - 1 \), and the entire ensemble is controllable as an \( N = N_1 \times \ldots \times N_L \) dimensional composite system if the Lie algebra \( L \) of the entire system has (at least) dimension \( N^2 - 1 \). If interdot coupling is negligible then the system is decomposable, and hence not controllable as a composite system, but its components are simultaneously controllable if the Lie algebra of the system satisfies Theorem 2.

If each dot can be represented as a two-level system with Hamiltonian

\[
\hat{H}^{(\ell)}[f(t)] = \epsilon_{\ell} \hat{\sigma}_z + f(t) d_{\ell} \hat{\sigma}_x,
\]

where \( \hat{\sigma}_x \) and \( \hat{\sigma}_z \) are the Pauli matrices, then we can use Theorem 2 to give explicit conditions for simultaneous controllability. Clearly, the \( \ell \)th dot is controllable individually exactly if \( \epsilon_{\ell} \neq 0 \) and \( d_{\ell} \neq 0 \), as \([\hat{\sigma}_x, \hat{\sigma}_z] = -2\hat{\sigma}_y\). However, this is not sufficient for simultaneous controllability. If two or more dots in the ensemble have exactly the same system parameters, for example, then the Lie subalgebra they generate is a higher-dimensional representation of \( \text{su}(2) \) and the dots are not simultaneously controllable. In this simple case one can show (see Appendix B) that the ensemble of dots is simultaneously controllable exactly if \((\epsilon_{\ell}, d_{\ell}) \neq (\pm \epsilon_{\ell'}, \pm d_{\ell'})\) unless \( \ell = \ell' \).

However, even if two or more dots in the ensembles are identical, we can often recover simultaneous controllability if we can divide the ensemble into (possibly overlapping) regions that can be selectively addressed. For example, consider the five-dot ensemble shown in Fig. 1 where dots 1 and 4 are identical but all the other dots are distinct, i.e., \((\epsilon_1, d_1) = (\epsilon_4, d_4)\), but \((\epsilon_{\ell}, d_{\ell}) \neq (\pm \epsilon_{\ell'}, \pm d_{\ell'})\) for \( \ell \neq \ell' \) and \( \ell, \ell' \in \{1, 2, 3, 5\} \). As dots 1 and 4 have the same characteristics, \( \hat{H}^{(1)} = \hat{H}^{(4)} \), the Lie algebra generated by

\[
\hat{H}_0 = \epsilon_1 \hat{\sigma}_z \oplus \epsilon_2 \hat{\sigma}_z \oplus \epsilon_3 \hat{\sigma}_z \oplus \epsilon_1 \hat{\sigma}_z \oplus \epsilon_5 \hat{\sigma}_z
\]
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\[ \hat{H}_C = d_1 \sigma_x + d_2 \sigma_y + d_3 \sigma_z + d_4 \sigma_x + d_5 \sigma_z \]

has dimension 12, and the ensemble is not simultaneously controllable. However, if we can adjust the laser to create two regions \( A \) and \( B \), encompassing dots \( \{1, 2, 3\} \) and \( \{3, 4, 5\} \), respectively, simultaneous controllability can be recovered as the Lie algebra generated by \( \{\hat{H}_0, \hat{H}_A, \hat{H}_B\} \) with

\[
\begin{align*}
\hat{H}_0 &= \epsilon_1 \hat{\sigma}_z + \epsilon_2 \hat{\sigma}_y + \epsilon_3 \hat{\sigma}_z + \epsilon_4 \hat{\sigma}_z + \epsilon_5 \hat{\sigma}_z \\
\hat{H}_A &= d_1 \hat{\sigma}_x + d_2 \hat{\sigma}_x + d_3 \hat{\sigma}_x + 0 + 0 \\
\hat{H}_B &= 0 + 0 + d_3 \hat{\sigma}_x + d_4 \hat{\sigma}_x + d_5 \hat{\sigma}_x
\end{align*}
\]

has dimension 15, as required.

We can also apply the controllability results to dots with a more complicated internal structure. For instance, consider an ensemble of \( L \) dots with a two-fold degenerate internal ground state and an excited state in a \( \Lambda \) configuration as shown in Fig. 2. Let \( \epsilon_\ell \) denote the energy gap between the degenerate ground states and the excited state, and \( d_\ell^+, d_\ell^- \) be the dipole moments of the \( \sigma_+ \) and \( \sigma_- \) transition, respectively, for the \( \ell \)th dot. Without loss of generality we can assume \( \hat{H}_m = \bigoplus_{\ell=1}^L \hat{H}_{m,\ell} \) for \( m = 0, 1, 2, \ldots, L \), and

\[
\hat{H}_{m,\ell} = \frac{\epsilon_\ell}{3} \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & +2 \end{pmatrix}, \quad \hat{H}_{1,\ell} = d_\ell^+ \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix}, \quad \hat{H}_{2,\ell} = d_\ell^- \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}.
\]

The ensemble is simultaneously (mixed-state) controllable exactly if the Lie algebra generated by \( \{\hat{H}_m\} \) is \( \bigoplus_{\ell=1}^L \text{su}(3) \). Again, this is usually the case unless two or more dots in the ensemble are effectively identical. For example, we verified that even if \( d_\ell^+ = 1 \) and \( d_\ell^- = -1 \) for all \( \ell \), the Lie algebra for a 5-dot ensemble with \( \epsilon_\ell = 1 + \Delta \epsilon_\ell \geq 0 \) and \( \Delta \epsilon_\ell \neq \Delta \epsilon_{\ell'} \) unless \( \ell = \ell' \) had indeed dimension \( 40 = 5 \times (3^2 - 1) \). However, if two dots have the same energy gap \( \epsilon_\ell \) and the same (absolute) values of the dipole moments \( d_\ell^+ \) then simultaneous controllability is lost, e.g., if we have \( \epsilon_1 = \epsilon_4 \) and \( d_1^+ = d_4^+ \) or \( d_1^- = d_4^- \) then the Lie algebra dimension is only \( 32 = 4 \times (3^2 - 1) \). However, if \( \epsilon_1 = \epsilon_4 \) but \( d_1^+ \neq \pm d_4^+ \), for instance, then simultaneous controllability is maintained.

In the previous example, we can also ask what about controllability if we can only apply a single pulse consisting of a mixture of \( \sigma_+ \) and \( \sigma_- \) polarized light. In this case we replace \( \hat{H}_1 \) and \( \hat{H}_2 \) by the composite control Hamiltonian \( \hat{H}_C = \cos \alpha \hat{H}_1 + \sin \alpha \hat{H}_2 \) with \( \alpha \in [0, \pi/2] \), so that the single dot Hamiltonians are

\[
\begin{align*}
\hat{H}_{0,\ell} &= \frac{\epsilon_\ell}{3} \begin{pmatrix} 1 & 0 & 0 \\ 0 & -1 & 0 \\ 0 & 0 & +2 \end{pmatrix}, \quad \hat{H}_{C,\ell} = \begin{pmatrix} 0 & 0 & d_\ell^+ \cos \alpha \\ 0 & 0 & d_\ell^- \sin \alpha \\ d_\ell^+ \cos \alpha & d_\ell^- \sin \alpha & 0 \end{pmatrix}.
\end{align*}
\]
We see clearly that the system cannot be simultaneously controllable for \( \alpha = 0 \) or \( \alpha = \pi/2 \) as in this case none of dots are individually controllable, their Lie algebras being contained in \( u(2) \). However, if we assume \( d\ell - \ell = -d\ell + \ell \), which is often the case in practice, then even for \( \alpha \in (0, \pi/4) \), the Lie algebra generated by \( \hat{H}_{0,\ell} \) and \( \hat{H}_{C,\ell} \) above is a unitary representation of \( \text{so}(3) \oplus u(1) \). Hence, the dots are not individually controllable, and the ensemble is thus not simultaneously controllable. The Lie algebra of an ensemble of \( L \) (non-identical) dots of this form is a unitary representation of \( \bigoplus_{\ell=1}^{L} \text{so}(3) \oplus u(1) \). Note that \( \text{so}(3) \) is not even sufficient for pure-state controllability. If \( d\ell \neq \pm d\ell \) then the \( \ell \)th dot is generally individually controllable for \( \alpha \in (0, \pi/2) \) and any ensemble of non-identical dots would be simultaneously controllable with a mixed-polarization control pulse.

6. Optimal Control of Ensembles of Quantum Dots

Simultaneous controllability of an ensemble of non-interacting quantum dots implies in particular that it is possible to selectively excite a particular dot with a single laser pulse without the need for selective addressing. If the energy levels of the dots, and hence their resonance frequencies, are different, a standard approach would be to use frequency-selective addressing using simple, e.g., Gaussian pulses resonant with the transition frequency of the dot to be excited. However, this may be less than optimal, especially when the pulse length is to be kept to a minimum to achieve fast operations, which would be crucial in quantum information processing applications. The question therefore naturally arises whether the results could be improved using optimally shaped pulses.

To address this question, we consider an ensemble of five quantum dots, modelled as two-level systems with energy differences \( \epsilon\ell \) of 1.32, 1.35, 1.375, 1.38 and 1.397 eV, respectively. If interdot coupling is negligible, the internal system Hamiltonian is

\[
\hat{H}_0 = \text{diag}(\hat{H}_{0,1}, \ldots, \hat{H}_{0,5}), \quad \hat{H}_{0,\ell} = \epsilon\ell \hat{\sigma}_z/2, \tag{5}
\]

and the Hamiltonian that describes the coupling to the external driving field is

\[
\hat{H}_1 = \text{diag}(\hat{H}_{1,1}, \ldots, \hat{H}_{1,5}), \quad \hat{H}_{1,\ell} = d\ell \hat{\sigma}_x, \tag{6}
\]

where \( d\ell \) is the dipole coupling of the \( \ell \)th dot to the field. Even if we assume, for simplicity, that all the dipole couplings are equal \( d\ell = 1 \) for \( \ell = 1, \ldots, 5 \), the dots are still simultaneously controllable, and in particular we can selectively excite a single dot without spatial addressing.

Fig. 3(a) shows the evolution of the ground and excited state populations of the dots if we simply apply a \( \pi \)-pulse, resonant with the transition frequency of the first dot, with Gaussian envelope \( A(t) = q\sqrt{\pi} \exp\left[-q^2(t - t_f/2)^2\right] \), where \( q = 4/t_f \) and the target time is \( t_f = 200 \) time units (\( \approx 130 \) fs). Although the pulse achieves almost 100% population transfer from the ground to the excited state for the target dot, it also leads to significant unwanted excitation of energetically adjacent dots. This effect tends to become more pronounced the shorter the pulses, and the smaller the differences in the transition frequencies of the dots.

Fig. 3(b) shows that we can considerably improve the results in this case using shaped pulses. The shaped pulse still achieves near perfect excitation of the target dot but considerably reduces the overall excitation of the other dots. Most importantly though, while there is some remaining transient excitation of the other dots, the shaped pulse ensures that the populations of the excited states return to (almost) zero at
the target time, except for the target dot, for which the excited state population is almost 1.

The pulse shown in Fig. 3(b) was obtained using an iterative optimal control algorithm similar to [30]. The starting point for the algorithm was the Gaussian pulse shown in Fig. 3. The observable to be optimized was chosen to be \( \hat{A} = \text{diag}(\hat{P}, Q, Q, Q, Q) \), where \( \hat{P} = |1\rangle\langle 1| \) is the projection onto the upper level and \( \hat{Q} = -\hat{P} \) to reflect our objective to at once maximize the excited state population of the first dot, while minimizing the excitation of all other dots. Note that this is not the only possible choice of the observable but the results of the algorithm depend significantly on the choice of the target observable. For example, choosing \( \hat{A}' = \text{diag}(\hat{P}, \hat{0}, \hat{0}, \hat{0}, \hat{0}) \) would be a bad choice and unlikely to improve the results because the initial Gaussian pulse does accomplish the objective of achieving 100% population transfer for the target dot, and the populations of the other dots do not affect the expectation value of \( \hat{A}' \), whence there would be no reason for the algorithm to alter the pulse shape in an attempt to suppress the off-resonant excitation of the other dots.

7. Conclusion

We have considered various notions of controllability for quantum systems and their application to multi-partite systems. In particular, we have investigated the degree of simultaneous controllability of the components of a decomposable system by means of global control, and given necessary and sufficient criteria for the simultaneous mixed-state and pure-state controllability of the components. The results have been applied to the problem of simultaneous controllability of quantum dots with negligible interdot coupling, and in particular the problem of selective excitation of ensembles of quantum dots with a laser pulse that is applied to the entire ensemble. We have also shown, for a simple model, that shaped pulses derived using optimal control theory may offer substantial improvements in selectivity compared to Gaussian pulses.

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Appendix A. Proof of theorem 2

Assume \( \hat{H}[f(t)] \) of the form (2), and \( \hat{\rho}(t) \) and \( \hat{H}_m \) of the form (3). For \( m = 0, \ldots, M \) and \( \ell = 1, \ldots, L \), set \( \alpha_{m,\ell} = \text{Tr}(\hat{H}_{m,\ell}) \) and define the diagonal generators

\[
\hat{D}_m = \text{diag}(\alpha_{m,1} \hat{I}_N, \ldots, \alpha_{m,L} \hat{I}_N).
\]  
(A.1)

where \( \hat{I}_\ell \) is the identity matrix in dimension \( \ell \), and the trace-zero generators

\[
\hat{H}_m = \text{diag}(\hat{H}_{m,1}, \ldots, \hat{H}_{m,L}),
\]  
(A.2)

where the blocks on the diagonal are given by

\[
\hat{H}_{m,\ell} = \hat{H}_{m,\ell} - \frac{\alpha_{m,\ell} \hat{I}_\ell}{N_\ell}.
\]  
(A.3)
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Figure 3. Selective excitation of dot 1: control field (in units of $10^5$ V/m) and evolution of the populations and observable for (a) a frequency-selective Gaussian control pulse, and (b) an optimally shaped control pulse. The expectation value of the observable corresponds to the population of the excited state of the target dot minus the sum of the populations of the excited states of all other dots. The Gaussian pulses (a) accomplishes the goal of exciting the first dot but also leads to significant excitation of the other dots, especially the dot energetically closest to the target dot, at the target time, while the shaped pulse (b) ensures that the excited state populations of all dots except the target dot return to (almost) zero at the target time.
The diagonal elements $D_m$ commute with $\dot{\rho}(t)$ for all $m = 0, \ldots, M$. Hence, the orbits of $\dot{\rho}(t)$ generated by $\tilde{H}[f(t)]$ and $\tilde{H}[f(t)] = \tilde{H}_0 + \sum_{m=1}^{M} f_m(t) \tilde{H}_m$ are identical.

Let $\mathcal{L} = \mathcal{L}(\{\tilde{H}_m\})$ be the Lie algebra generated by the trace-zero skew-Hermitian matrices $i\tilde{H}_m$. Due to the structure of the generators, the Lie algebra $\mathcal{L}$ must be a subalgebra of $\bigoplus_{\ell=1}^L su(N_\ell)$, and it follows from classical results \cite{35} that the orbits of a generic mixed state \cite{35} will be maximal if and only if $\mathcal{L} = \bigoplus_{\ell=1}^L su(N_\ell)$.

Now let $\mathcal{L} = \mathcal{L}(\{\tilde{H}_m\})$ be the Lie algebra generated by $i\tilde{H}_m$, and $\mathcal{L}_D = \mathcal{L}(\{\tilde{D}_m\})$ be the Lie algebra generated by the $M + 1$ diagonal (skew-Hermitian) matrices $i\tilde{D}_m$. Since the diagonal generators $i\tilde{D}_m$ commute with the trace-zero matrices $\tilde{H}_m$, i.e., $[D_m, H_{m'}] = 0$ for all $m, m' = 1, 2, \ldots, M$, we have

$$\mathcal{L} = \mathcal{L} \oplus \mathcal{L}_D \quad (A.4)$$

i.e., the Lie algebra $\mathcal{L}$ is the direct sum of the Lie algebras $\mathcal{L}$ and $\mathcal{L}_D$. Noting that the dimension of $su(N_\ell)$ is $N_\ell^2 - 1$ and the dimension of $\mathcal{L}_D$ is equal to the rank of the matrix

$$A = \begin{pmatrix}
\alpha_{0,1} & \cdots & \alpha_{0,L} \\
\vdots & & \vdots \\
\alpha_{M,1} & \cdots & \alpha_{M,L}
\end{pmatrix} \quad (A.5)$$

where $r = \text{rank}(A) \leq \min(M + 1, L)$, we obtain the Lie algebra dimension condition of Theorem \ref{thm:controllability} as a necessary and sufficient condition for simultaneous (mixed-state) controllability.

Appendix B. Controllability analysis for two uncoupled two-level systems

To make the difference between individual and simultaneous controllability explicit, consider two non-interacting two-level systems simultaneously driven by a coherent control field $f(t)$. We can write the Hamiltonian of the composite system as $\tilde{H} = \tilde{H}_0 + f(t) \tilde{H}_1$, where

$$\tilde{H}_0 = \text{diag}(E^{(1)}_0, E^{(1)}_1) \oplus \text{diag}(E^{(2)}_0, E^{(2)}_1)$$

$$\tilde{H}_1 = d_1 \hat{\sigma}_x \oplus d_2 \hat{\sigma}_x.$$

$\tilde{H}_0$ can further be split into a trace-zero part $\tilde{H}_0' = \epsilon_1 \hat{\sigma}_z \oplus \epsilon_2 \hat{\sigma}_z$, where $\epsilon_\ell = (E^{(\ell)}_1 - E^{(\ell)}_0)/2$, and a diagonal part $\tilde{D}_0 = \sigma_1 \hat{I} \oplus \sigma_2 \hat{I}$, where $\alpha_\ell = (E^{(\ell)}_0 + E^{(\ell)}_1)/2$. If $\epsilon_\ell = 0$ or $d_\ell = 0$ then subsystem $\ell$ is not even individually controllable. Hence, we shall assume that $\epsilon_\ell \neq 0$ and $d_\ell \neq 0$ for $\ell = 1, 2$.

The diagonal generator $\tilde{D}_0$ is not relevant for our controllability analysis. The traceless generators $W_1 = i\tilde{H}_0'/\epsilon_1, W_2 = i\tilde{H}_1/d_1$ give rise to the following Lie algebra:

$$W_3 = [W_2, W_1]/2 = -i(\hat{\sigma}_y \oplus ab\hat{\sigma}_y)$$

$$W_4 = [W_3, W_1]/2 = -i(\hat{\sigma}_x \oplus a^2b\hat{\sigma}_x)$$

$$W_5 = [W_3, W_2]/2 = i(\hat{\sigma}_z \oplus ab^2\hat{\sigma}_z)$$

$$W_6 = [W_4, W_1]/2 = i(\hat{\sigma}_y \oplus a^3b\hat{\sigma}_y)$$

$$W_7 = [W_3, W_4]/2 = -i(\hat{\sigma}_z \oplus a^3b^2\hat{\sigma}_z)$$

where $a = \epsilon_2/\epsilon_1$ and $b = d_2/d_1$. Combining these terms yields

$$W_2 + W_4 = 0 \oplus (1 - a^2)b i\hat{\sigma}_x$$

$$a^2W_2 + W_4 = (a^2 - 1) i\hat{\sigma}_x \oplus 0$$
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\[ W_3 + W_6 = 0 + ab(a^2 - 1) i\sigma_y \]
\[ a^2 W_3 + W_6 = (1 - a^2) i\sigma_y \oplus 0 \]
\[ W_5 + W_7 = 0 + ab^2(1 - a^2) i\sigma_z \]
\[ a^2 W_5 + W_7 = (a^2 - 1) i\sigma_z \oplus 0 \]

This shows that if \( a, b \neq 0, \pm 1 \) then the relevant Lie algebra \( \hat{\mathcal{L}} \) of the system is \( \text{su}(2) \oplus \text{su}(2) \).

For \( \alpha = \pm 1 \) we can similarly show that \( V_k = W_k \) for \( k = 1, 2, 3 \), \( V_4 = W_4 \) and
\[ V_5 = [V_2, V_4]/2 = \hat{\sigma}_y \oplus ab^3 \hat{\sigma}_y \]
\[ V_6 = [V_4, V_5]/2 = i(\hat{\sigma}_x \oplus b^3 \hat{\sigma}_x) \]
\[ V_7 = [V_2, V_5]/2 = i(\hat{\sigma}_z + ab^4 \hat{\sigma}_z). \]

Combining these terms leads to
\[ V_6 - V_2 = 0 \oplus b(b^2 - 1)i\hat{\sigma}_x \]
\[ b^2 V_2 - V_6 = (b^2 - 1)i\hat{\sigma}_x \oplus 0 \]
\[ V_5 + V_3 = 0 \oplus ab(b^2 - 1)\hat{\sigma}_y \]
\[ b^2 V_3 - V_5 = (b^2 - 1)\hat{\sigma}_y \oplus 0 \]
\[ V_7 - V_4 = 0 \oplus ab^2(b^2 - 1)i\hat{\sigma}_z \]
\[ b^2 V_4 - V_7 = (b^2 - 1)i\hat{\sigma}_z \oplus 0 \]

This shows that if \( a \neq 0, b \neq 0, \pm 1 \) then the relevant Lie algebra \( \hat{\mathcal{L}} \) of the system is still \( \text{su}(2) \oplus \text{su}(2) \), and the two subsystems are therefore simultaneously controllable.

However, if \( a = \pm 1 \) and \( b = \pm 1 \) then \( V_4 = 2V_1, V_5 = V_3, V_6 = V_2, V_7 = V_1 \), i.e., Lie algebra is three-dimensional, and since \( V_3 = [V_2, V_1] \), a representation of \( \text{su}(2) \). The non-interacting two-level systems are therefore individually but not simultaneously controllable.

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