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Cite as: J. Chem. Phys. **129**, 041105 (2008); https://doi.org/10.1063/1.2958220
Submitted: 16 April 2008 • Accepted: 24 June 2008 • Published Online: 29 July 2008

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(Received 16 April 2008; accepted 24 June 2008; published online 29 July 2008)

Quantum conditions on the control of dynamics of a system coupled to an environment are obtained. Specifically, consider a system initially in a system subspace H_0 of dimensionality M_0, which evolves to populate system subspaces H_1, H_2 of dimensionalities M_1, M_2. Then, there always exists an initial state in H_0 that does not evolve into H_2 if $M_0 > dM_2$, where $2 \leqslant d \leqslant (M_0+M_1+M_2)^2$ is the number of operators in the Kraus representation. Note, significantly, that the maximum d can be far smaller than the dimension of the bath. If this condition is not satisfied, then dynamics from H_0 that avoids H_2 can only be attained physically under stringent conditions. An example from molecular dynamics and spectroscopy, i.e., donor to acceptor energy transfer, is provided. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2958220]

The study of open quantum systems[1–3] is of great current interest, a consequence of the desire to understand and to manipulate devices that are reliant on quantum effects. Of particular interest is the control of such quantum systems, with coherent control[4,5] being a most promising approach.

The essential principle of the coherent control of atomic and molecular processes relies on the creation of multiple interfering pathways to the same final state.[4,5] Manipulation of laboratory parameters that affect these coherent pathways then allows direct control over the associated interference contributions, and hence control over the relative cross sections involved in the process. Numerous theoretical scenarios and experimental results, in a host of applications, have successfully demonstrated such control.

Despite the importance and generality of the field, there are remarkably few formal results that provide necessary and sufficient conditions under which complete control of the system dynamics is possible. These include theorems such as that of Huang–Tarn–Clark,[6] a theorem by Ramakrisnan *et al.* on the dimensionality of the Lie Algebra induced by the interaction between the system and the control field,[7] and a theorem by Shapiro and Brumer,[8] where control was shown to depend on the dimensionality of the controlled subspaces. These theorems all deal with closed systems, i.e., those that are not in contact with an environment. Real quantum systems are, however, not isolated. They often interact with a surrounding environment, which is regarded as an uncontrolled system of arbitrarily large dimension. As a result, the system is open to its environment, and proofs regarding control in open systems are becoming the focus of serious attention. For example, Ref. 9 provides formal theorems on controllability in open system dynamics treated via the Kraus representation.

In this letter, we significantly generalize a prior argument[8] for closed systems and obtain quantum control conditions in open systems. We show that the condition for a specific control problem in open systems is stronger than that in closed systems, but that the relative dimensionality of the different subspaces remains a crucial determinant of controllability. In addition, our approach gives the solution to the control problem when it is achievable, and provides a constructive method for extending various closed system proofs to the open system case.

To expose the difference between the open and closed cases, to adopt a common notation, and to provide a generalized formalism, we first reconsider the closed system.[8] Consider a system whose Hamiltonian eigenstates are partitioned into bases for three subspaces: H_0, H_1, H_2, of dimensionalities M_0, M_1, and M_2, respectively. Let the basis vectors $|i,n_i\rangle$ span the subspace H_i, where $i=0,1,2$ labels the subspace and $n_i=1,\ldots,M_i$ labels the states in H_i. The total dimension of the system is $M=M_0+M_1+M_2$. A superposition of basis vectors from H_0 is chosen as the initial state. We assume that the system initially resides in M_0 and that the state can flow into the other two subspaces under the dynamics. The question under consideration is under what conditions can one prevent dynamics into the subspace H_2, by preparing states in H_0 that go solely into H_1? To resolve this problem, Ref. 8 considered linear superpositions of states in H_0 and utilized interference between the resultant paths to H_1 and H_2 in order to control the dynamics.

Using the evolution operator U, we have

$$|\psi\rangle = U|\psi_0\rangle, \tag{1}$$

where $|\psi_0\rangle$ is the initial superposition in M_0 and $|\psi\rangle$ denotes the final state. With U represented as an $M \times M$ dimensional matrix and $|\psi\rangle$ expressed as an M column matrix, we can examine the number of simultaneous equations that need to be solved for $|\psi\rangle$ to have zero population in the rows corresponding to H_2. Using this approach, Ref. 8 obtained conditions on the relative dimensionality of the different spaces by requiring

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$$\langle 2,k_2|U|\psi_0\rangle = 0 \tag{2}$$

for all states k_2 in H_2. Specifically, they showed that if $M_0 > M_2$, it is always possible to prevent transitions from H_0 into H_2. In contrast, a large number of difficult-to-satisfy linear dependence conditions are required to prevent transitions if $M_0 \leqslant M_2$. Significantly, the result only depends on the dimensionalities M_0 and M_2 of these two subspaces.

Consider then the same system, but where it is now open to an environment, which can be either finite or infinite dimensional, and that may, or may not, be in the presence of external fields. As is typically the case, we assume that the system can be addressed (e.g., via laser excitation) to prepare the initial state, but that the environment remains unaffected. Further, we assume that the initial system-bath density matrix $\rho_{\text{tot}}(0)=\rho(0)\otimes\rho_B(0)$ is a separable product of the initial system $\rho(0)$, usually a pure state that can be selectively prepared, and initial bath $\rho_B(0)$ density matrix.[10] The open system after evolution can be characterized by an $M\times M$ reduced density matrix ρ, defined as $\rho=\text{Tr}_B\,\rho_{\text{tot}}$, where ρ_{tot} is the final time total system-bath density matrix and Tr_B indicates a trace over the bath. Focusing on the reduced density matrix (i.e., ρ_{tot} traced over the bath) allows us to continue to use the concept of dimensionality in an open system. In general, the reduced density matrix evolves according to a linear transformation

$$\rho = \sum_{\alpha,\beta} A^{\alpha}\rho(0)B^{\beta}. \tag{3}$$

For instance, in the case of natural quantum evolution, denoting the propagator for the full (system+bath) as U, we have that

$$\rho_{\text{tot}} = U\rho_{\text{tot}}(0)U^{\dagger} = U\rho(0)\otimes\rho_B(0)U^{\dagger}, \tag{4}$$

where we have used the separability of ρ_{tot} at time zero. In general, $\rho_B(0)=\Sigma_{a,b}p_{ab}|a\rangle\langle b|$, where $|a\rangle$ and $|b\rangle$, and the $|e\rangle$ below are eigenstates of the bath Hamiltonian. By tracing Eq. (4) over the bath coordinates, we have that

$$\rho = \sum_{e,a,b} A^{a,e}\rho(0)B^{a,b,e} \tag{5}$$

with $A^{a,e}=\langle e|U|a\rangle$ and $B^{a,b,e}=p_{ab}\langle b|U^{\dagger}|e\rangle$.

Under the assumption of complete positivity,[11] Eq. (3) simplifies and the reduced density matrix evolves according to the operator-sum representation (i.e., the canonical Kraus representation), which is generic physically for the initial product density matrix,[3]

$$\rho = \sum_{\alpha=1}^{d} E^{\alpha}\rho(0)E^{\alpha\dagger}. \tag{6}$$

Here, the sum over Kraus operators[11] E^{α} is such that $\Sigma_{\alpha=1}^{d}E^{\alpha\dagger}E^{\alpha}\leqslant I$. Specifically, $\Sigma_{\alpha=1}^{d}E^{\alpha\dagger}E^{\alpha}=I$ for trace-preserving quantum operations and $\Sigma_{\alpha=1}^{d}E^{\alpha}E^{\alpha\dagger}<I$ for non-trace-preserving quantum operations such as quantum measurements. Note, significantly, that the operator-sum representation has at most M^2 Kraus operators, i.e., $2<d\leqslant M^2$ and that the operators are fixed by the given system, bath and any incident external fields.

We continue to consider below the general form [Eq. (3)], but subsequently focus on the evolution under the Kraus representation.

Note first that Eq. (3) means that the operator $\rho(0)$ can be linearly transformed into the operator ρ. If we denote the operation connecting the matrix $\rho(0)$ to ρ by V, then V, in a particular representation, is a matrix with four subscripts. That is,

$$\rho_{ts} = \sum_{t's'} V_{tst's'}\rho_{t's'}(0),$$

where t or s denotes the indices (i,n_i) of the basis vectors $|i,n_i\rangle$. Rewriting ρ as M^2 dimensional column vector denoted $\tilde{\rho}$, Eq. (6) can be equivalently rewritten as

$$\tilde{\rho} = \tilde{V}\tilde{\rho}(0), \tag{7}$$

where the explicit form of the $M^2\times M^2$ matrix $\tilde{V}$ is

$$\tilde{V} = \sum_{\alpha,\beta} A^{\alpha}\otimes B^{\beta}.$$

Equation (7) is seen to have the same form as Eq. (1), allowing the approach used earlier for a closed system to be extended to open systems. Consider then an initial reduced density matrix that is in H_0, i.e.,

$$\rho(0) = \begin{pmatrix} \rho_{M_0^2} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

where $\rho_{M_0^2}$ is an $M_0\times M_0$ matrix. In order to prevent dynamics from going into H_2, we require

$$\rho_{(2,k_2)(2,k_2)} = 0. \tag{8}$$

For the case of interest, the Kraus representation applies so that a propagated initial pure state $\rho_{M_0^2}=|\psi_0\rangle\langle\psi_0|$ becomes

$$\rho_{(2,k_2)(j,n_j)} = \sum_{\alpha=1}^{d}\langle 2,k_2|E^{\alpha}|\psi_0\rangle\langle\psi_0|E^{\dagger\alpha}|j,n_j\rangle.$$

Given that the diagonal element $\rho_{(2,k_2)(2,k_2)}$ is $\Sigma_{\alpha=1}^{d}|\langle 2,k_2|E^{\alpha}|\psi_0\rangle|^2$, satisfying $\rho_{(2,k_2)(2,k_2)}=0$ means requiring

$$\langle 2,k_2|E^{\alpha}|\psi_0\rangle = 0. \tag{9}$$

This being the case, demanding zero population in H_2 also implies that all elements $\rho_{(2,k_2)(j,n_j)}$, which include coherences with the H_2 Hilbert subspace, are also zero.

Equation (9) has a nontrivial solution if $M_0 > dM_2$, and control via initial state preparation is achievable no matter what the form or dynamics of the d-dimensional Kraus operators. Further, this equation provides the initial state that allows for the desired control. However, this control condition is, as expected, far more stringent in the open system than in the closed system case, where $d=1$. Note, however, that the bath effects are still limited. The minimum d can be[12] as small as 2, and the maximum d is M^2, which can be far smaller than the dimensionality of the bath.

If $M_0 \leqslant dM_2$, control via initial state preparation is also far more difficult in the open system than in the (already

difficult) closed system case. Define a $dM_2 \times M_0$ dimensional matrix W with matrix elements $W_{(2k,\alpha),(0n)} = \langle 2,k|E^{\alpha}|0,n\rangle$. In the case where $M_0 \leqslant dM_2$, the rank of W is equal to M_0 unless all $M_0 \times M_0$ dimensional submatrices of W are singular. Hence, nontrivial solutions to Eq. (9) exist if $\det(W^{(k)}_{M_0,M_0})=0$, where (k) numbers all of the submatrices. This condition also implies that a set of columns of W is linearly dependent. Hence, control is possible for a *specific* class of W, expected to be difficult to obtain physically.

Note that the definition of the subspaces H_i and associated dimensions M_i can, in some instances, also be manipulated. For example, in bound state systems (such as that in the example below) these subspaces can be defined by accessing only specific system eigenstates using a pulsed laser field. This facility might prove additionally useful in attempting to satisfy the $M_0 > dM_2$ bound of this theorem.

These results allow numerous applications. Consider, for example, electronic energy transfer from donor to acceptor molecules, where both may be part of one larger molecule. These systems are ubiquitous, ranging from relatively small systems[13] to large structures such as carotenoid-to-bacteriochlorophyll energy transfer in photosynthesis.[14] The most interesting cases take place in condensed matter environments, i.e., open systems. Studies of the dynamics and spectroscopy of such systems can be carried out by laser excitation of the system from a lower electronic manifold (here H_0) to the donor (here H_1). Electronic energy transfer from the donor (H_1) to the acceptor (H_2) is then measured. Here, the subspaces H_i and associated dimensions M_i are determined by a combination of the molecular state densities and the width of the laser pulses that prepare a preliminary superposition of states on H_0 and that subsequently excite the system into H_1 and H_2.

In some systems of interest, excitation from H_0 to the donor is contaminated by partial excitation of the acceptor as well, with a concomitant reduction in the quality of the data on the subsequent electronic energy transfer dynamics. A considerable improvement would result from being able to excite H_1 with reduced population transfer to H_2 from H_0. Results of the open system theorem indicate that this is (a) difficult to achieve physically if $M_0 \leqslant dM_2$, and (b) attainable if $M_0 > dM_2$. In the latter case, one could ensure significantly reduced, and ideally zero, acceptor population at the target time. The extent to which this is achievable is dependent on the particular system; specific systems of this type will be the subject of future study.

In summary, the quantum conditions obtained above are completely general, applying to both systems that are controlled, as well as to uncontrolled system evolution. The result establishes an important inequality between the dimension of the subspace M_2 which we desire not to populate, the initial subspace M_0 from which dynamics evolves, and the dimensionality of the Kraus representation. It is a dynamics-independent property of d-dimensional Kraus evolution. As long as the geometric condition $M_0 \geqslant dM_2$ is satisfied, control via initial state preparation is achievable. Further, the fact that control can be achieved via initial state preparation, in the presence of an environment under the prescribed conditions, is useful for control applications in realistic systems.

Two supplementary remarks are in order. First, this theorem places emphasis on the importance of the dimensionality d of the Kraus representation, known to satisfy $2 \leqslant d \leqslant (M_0+M_1+M_2)^2$. Hence, these results should motivate further studies to determine the actual d for realistic systems. Second, we note that an analogous method to that described above can be used to extend other closed system proofs, such as those of Refs. 6 and 7, to open systems.

We thank Professor R. Alicki for discussions on Kraus operators, and Professor G. D. Scholes for comments on donor-acceptor dynamics. This work was supported by NSERC Canada.

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