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Controllability analysis of quantum systems immersed within an engineered environment

A. Grigoriu · H. Rabitz · G. Turinici

Abstract The problem of controllability of quantum systems interacting with an engineered environment, whose dynamics are described by a non-Markovian master equation is addressed. The manipulations of the dynamics is realized with both a laser field and a tailored non-equilibrium, and generally time-dependent, state of the surrounding environment. Lie algebra theory is used to characterize the structures of the reachable state sets and to prove controllability. The theoretical results are supported by examples.

Keywords Quantum control · Controllability of non-Markovian equations · Lie group controllability · Controllability in a non-compact Lie group

1 Introduction

Since the first successful laboratory experiments obtained in the 1990s [1,2], the control of quantum systems using laser fields has been subject to significant developments

A. Grigoriu (✉)
Laboratoire Jacques-Louis Lions, Université Paris-Diderot, 175, rue du Chevaleret, 75013 Paris, France
e-mail: grigoriu@ljll.univ-paris-diderot.fr

H. Rabitz
Frick Laboratory, Department of Chemistry, Princeton University, Princeton, NJ 08544, USA
e-mail: hrabitz@princeton.edu

G. Turinici
CEREMADE, Université Paris Dauphine, Pl. du Mal de Lattre de Tassigny, 75016 Paris, France
e-mail: gabriel.turinici@dauphine.fr

([1–5] etc.). An important part of the associated theoretical work has been devoted to the investigation of closed quantum systems with unitary dynamics. However, realistic physical situations include circumstances where the quantum system is not isolated, but interacting with an environment (e.g., a molecule in a solvent). For these type of systems, also called open quantum systems, one of the main characteristics is that the dynamics is non-unitary. Generic open systems may be difficult to control because the environment can contain irreversible dynamics that fights against control mechanisms. We consider in this work a particular situation when the environment can be engineered i.e. its characteristics chosen at will. The resulting circumstance is not a general open quantum system but a large closed system containing a small part of interest to the experimenter. The mathematical formulation will be identical to that used to describe an open system except that the environment is considered controllable.

For weak system-environment coupling the dynamics of the system is described by a Markovian equation involving the reduced density matrix ρ

$$\frac{d}{dt}\rho(t) = \mathcal{L}\rho(t) \tag{1}$$

with the generator $\mathcal{L}$ in Lindblad form (see [6–8]):

$$\mathcal{L}\rho = -i[H, \rho] + \sum_i \gamma_i \left(L_i \rho L_i^* - \frac{1}{2}L_i^* L_i \rho - \frac{1}{2}\rho L_i L_i^* \right), \tag{2}$$

where the first term represents the unitary part of the system dynamics, with H the Hamiltonian of the system. The second term describes the dissipative dynamics, also referred as the dissipator, dependent upon the Lindblad operators L_i. The time independent coefficients γ_i represent coupling to the environment, functioning as the relaxation rates for different decay modes of the open system. In the following for any X, we denote by X^* its adjoint.

For cases of strong system-environment coupling, structured and finite reservoirs, and/or low temperatures one often encounters non-Markovian processes, implying that the dynamics is governed by significant memory effects. A systematic approach to non-Markovian dynamics is provided by projection operator techniques [9–11]. These techniques can lead to integro-differential equations for the reduced density matrix, through application of the Nakajima–Zwanzig projection operator technique. If a time-convolution free projection operator technique [12–14] is applied, one can deduce an approximate or even exact non-Markovian, first-order differential equation for the open system reduced density matrix ρ:

$$\frac{d}{dt}\rho(t) = \mathcal{K}(t)\rho(t). \tag{3}$$

The non-Markovian character of this equation is reflected by the fact that its generator $\mathcal{K}$ depends explicitly on time and does not have the Lindblad form above.

If we require that the density matrix be Hermitian and its trace constant, the most general form of the generator $\mathcal{K}(t)$ is given by [8,13]:

$$\mathcal{K}(t)\rho = -i[H(t), \rho] + \sum_i \gamma_i(t) \left(A_i(t)\rho A_i^*(t) - \frac{1}{2} A_i(t)^* A_i(t)\rho - \frac{1}{2}\rho A_i A_i^* \right),$$

(4)

where the Hamiltonian $H(t)$, the operators $A_i(t)$ and the relaxation rates $\gamma_i(t)$ depend on time.

If the relaxation rates $\gamma_i(t)$ are positive functions, the generator $\mathcal{K}(t)$ is in Lindblad form for each fixed $t \geqslant 0$. Such processes may be called time-dependent Markovian although the corresponding dynamical map do not lead to a quantum dynamical semigroup [15]. However if the relaxation rates become negative in certain time intervals (see examples of negative rates γ_i in [13,15]), the master equation in no longer in Lindblad form and the generator is not completely positive. Therefore non-Markovian character is also related to the emergence of negative rates.

Given the diversity of models that describe the quantum system-environment interaction, the problem manipulating them becomes difficult to address, especially since environment is usually composed of a large number of degrees of freedom which are difficult to control. Despite that, research on control of such systems is strongly motivated by a large number of applications including quantum computing [16], laser cooling, quantum reservoir engineering, management of decoherence, chemical reactions and energy transfer in molecules [17].

Optimal control by a tailored non-equilibrium, and generally time-dependent, state of the surrounding environment has been addressed in the literature (see [18] for more details). The master equation for the system interacting with an environment, characterized by its distribution function $n_k(t)$ is given by

$$\frac{d}{dt}\rho(t) = -i[H, \rho(t)] + \sum_\omega \gamma_\omega(n_k(t)) \left(A_\omega \rho A_\omega^* - \frac{1}{2} A_\omega^* A_\omega \rho - \frac{1}{2}\rho A_\omega^* A_\omega \right), \quad (5)$$

where the coefficients $\gamma_\omega(t) \geqslant 0$ determine the transitions rates between energy levels with transition frequencies ω.

Control by lasers fields normally affects the system through Hamiltonian evolution and transforms pure states into pure states, while control by the environment (i.e., control by $\gamma(t)$) affects a system through dissipative dynamics and can be used to steer the system from a pure or a mixed state into mixed and in some cases pure states (a familiar example is the cooling of a thermalized quantum system, which requires coupling to a reservoir). This type of incoherent control by the environment may be combined with coherent fields to allow for simultaneous control through both the Hamiltonian and dissipative parts of the system dynamics.

The equation for a system that simultaneously interacts with an electromagnetic field $\epsilon(t)$ and an environment described by a function $\gamma(t) \geqslant 0$ generally has the form [18]:

$$\frac{d}{dt}\rho(t) = -i[H_0 + H_{eff} + \epsilon(t)H_1, \rho(t)]$$
$$+ \sum_i \gamma_i(t) \left(A_i \rho A_i^* - \frac{1}{2} A_i^* A_i \rho - \frac{1}{2} \rho A_i^* A_i \right), \quad (6)$$

where H_0 is the Hamiltonian describing the coherent part of the dynamics, H_{eff} is the effective Hamiltonian describing the unitary part of the system-environment interaction and H_1 is the dipole moment operator describing the interaction between the system and the field $\epsilon(t)$.

Although the notion of simultaneous control by both a laser and by the environment has been mentioned in the literature [18], a theoretical analysis of the controllability has not been investigated yet. The goal of this paper is to propose such an analysis in a generic situation. A fundamental question involving the system above is whether for any pair of states ρ_i and ρ_f, both positive semidefinite, Hermitian, $tr(\rho_i) = tr(\rho_f) > 0$, a control $(\epsilon(t), \gamma(t))$ exists such that $\rho(0) = \rho_i$ and $\rho(T) = \rho_f$ for some $T > 0$. If the answer is positive the system is called controllable. Since $\gamma(t)$ may also take negative values, the present work addresses the controllability analysis for situations when both $\epsilon(t)$ and $\gamma(t)$ are arbitrary real numbers. Under this hypothesis the property of complete positivity of the density matrix is lost; moreover with dynamics being non-Markovian. Note that if physical considerations require $\gamma(t)$ to have further specific properties the results of the analysis will not apply, but a practical proposal is to impose the constraints in the numerical simulations by penalization, leading to an optimal control framework.

Thus we suppose that the system follows a first-order differential Eq. (6) for the reduced density matrix ρ.

The generator for the above equation is time dependent and has the same form as $\mathcal{K}(t)$, previously defined by Eq. (4). Our main goal is to analyze the controllability (as defined above) of the Eq. (6).

Another motivation for this analysis is given by systems that are not controllable in the isolated setting ($\gamma(t) = 0$); one direction on addressing controllability can rely on the use of specially tailored environments, characterized by $\gamma(t)$, applied through the dissipative part of the dynamics. It should be noted, however, that the assumption $\gamma(t) \in \mathbb{R}$ does not imply that the propagation of an arbitrary positive matrix at time t with the help of the master Eq. (6) necessarily leads to a positive matrix for future times. It is important to mention that although we want to introduce results as close as possible to physical circumstances, the complexity of the theoretical problems addressed in the framework of controllability of quantum systems interacting with an environment is a complex topic for which we have a partial analysis.

The balance of the paper is as follows: in Sect. 2 we introduce the general framework, the main notation together with a short review of Lie algebra controllability results previously introduced in the literature. In Sect. 3 we characterize the reachable sets and we prove two controllability results. The last section is dedicated to numerical computations.

2 The problem

We consider a quantum system interacting with an environment. The system is manipulated by both a laser field of intensity $\epsilon(t)$ and a tailored non-equilibrium, and generally time-dependent rate, $\gamma(t)$ depending on the state of the surrounding environment; the dynamics is described by Eq. (6). We suppose that the quantum dynamics takes place in a finite dimensional space (e.g., either intrinsically so or because a suitable large basis set approximation has been chosen). Thus, $\rho(t)$ is a $N \times N$ complex matrix for some integer $N > 0$, H_0, H_{eff} and H_1 are real N-dimensional symmetric matrices (thus Hermitian), A_i is an arbitrary N-dimensional matrix and ϵ, γ are real time dependent functions.

In the following, we study the controllability (see the definition above) of the N-dimensional Eq. (6). Without loss of generality we consider throughout the paper only the circumstance when all γ_i are null except γ_1. This assumption is not restrictive since the same general results can be obtained for the case when more then one γ is non-null. Since the effective Hamiltonian H_{eff} is assumed to commute with H_0 we can redefine H_0 to be $H_0 + H_{eff}$ and omit H_{eff} in the sequel. Therefore, under the above hypothesis Eq. (6) becomes:

$$\frac{d}{dt}\rho = -i[H_0, \rho] - i\epsilon(t)[H_1, \rho] + \gamma(t)\left(A\rho A^* - \frac{1}{2}\rho A^* A - \frac{1}{2}A^* A\rho\right). \quad (7)$$

In the following, for simplicity, we use the notation $\mathcal{K}$ for the generator of the Eq. (7)

$$\mathcal{K}(t)\rho = -i[H_0, \rho] - i\epsilon(t)[H_1, \rho] + \gamma(t)\left(A\rho A^* - \frac{1}{2}\rho A^* A - \frac{1}{2}A^* A\rho\right). \quad (8)$$

Aspects on controllability of open quantum systems have been addressed in the literature mostly for systems (7) with γ positive and constant. The problem of kinematic state controllability of open quantum systems whose dynamics are represented by Kraus maps has been considered in [19].

The behavior of a Markovian master equation for a N-level quantum mechanics system driven by a coherent control field $\epsilon(t)$ is analyzed [20,21]. It is shown that the system can be accessible, but neither small-time controllable nor controllable in finite time.

A deterministic two stage method for engineering arbitrary pure and mixed states for quantum systems subject to the Markovian evolution is proposed in [22]. In the first stage, the system evolves under the action of a suitable optimal incoherent control. In the second stage, the system evolves under the action of a suitable coherent laser control. But in that work the focus in on the control of the dissipative regime i.e. the control by the coherent part (the laser of intensity $\epsilon(t)$ is already supposed complete), while here we on the contrary want to use $\gamma(t)$ to bring supplementary control in addition to that of $\epsilon(t)$.

In this paper we use the Lie algebraic approach (cf. [23–25]) to analyze the controllability of quantum systems those dynamics is described by Eq. (7). An advantage

of this technique is that it also provides an explicit controllability criteria, which can be verified using numerical computations.

As will be seen in the sequel, controllability analysis of systems defined by (7) must deal with the loss of compactness that arises from the non-Hermitian nature of the generator $\mathcal{K}$ defined by (8). Another problem is that we do not know *a priori* the structure of the reachable states, identifying the Lie group remains a task to be investigated.

2.1 Background on controllability on Lie groups

Let us consider a connected but not necessarily compact Lie group G with Lie algebra $L(G)$ and control system

$$\frac{dX}{dt}(t) = \mathbb{X}_0(X(t)) + \sum_{i=1}^{m} u_i(t)\mathbb{X}_i(X(t)), \qquad (9)$$

where $\mathbb{X}_0$ and $\mathbb{X}_i$ are right-invariant vector fields on G. If necessary we will denote this solution $X(t; u; Y)$ to indicate its dependence on time, controls and initial state Y. Consider the set of all reachable states from Y at time t:

$$\mathcal{R}^t(Y) = \{X(t; u; Y) | X(t; u; Y) \text{ solution of (9)}, \ X(0; u; Y) = Y\}. \qquad (10)$$

It follows to see that

$$\mathcal{R}^t(Y) = \mathcal{R}^t(e)Y. \qquad (11)$$

where we denote by e the identity of the Lie group G; thus, describing the set $\mathcal{R}^t(e)Y$ allows for completely describing all the other reachable sets. When the final time is not specified, we will use

$$\mathcal{R}(Y) = \cup_{t \geqslant 0} \mathcal{R}^t(Y). \qquad (12)$$

We take the admissible controls $u_i(t)$ to be the set of all locally bounded and measurable functions.

Consider $\mathbb{L}$ to be the Lie algebra generated by $X_0, X_1, \ldots, X_m$ and $\mathbb{S}$ its corresponding Lie group (Lie subgroup of G). We **do not assume** that $\mathbb{S}$ is compact.

The results proved below build on the following reformulation of a result in [26, Thm 6.6] (to which we refer for further details):

Theorem 1 ([26]) *If there exists a constant control $u = (u_1, \ldots, u_m)$ and a sequence of positive numbers $\{t_n\}$ with $t_n \geqslant \delta > 0$, for some δ, with the property that $\lim_{n \to \infty} X(t_n, u, e)$ exists and belongs to $\bar{\mathbb{S}}$ (the closure is relative to $\mathbb{S}$) then $\mathcal{R}(e) = \mathbb{S}$.*

3 Controllability

We define the operators $\mathcal{H}_0$, $\mathcal{H}_1$ and $\mathcal{T}$ as follows:

$$\mathcal{H}_0 : \rho \longrightarrow -i[H_0, \rho]$$
$$\mathcal{H}_1 : \rho \longrightarrow -i[H_1, \rho]$$
$$\mathcal{T} : \rho \longrightarrow A\rho A^* - \frac{1}{2}\rho A^* A - \frac{1}{2}A^* A\rho, \tag{13}$$

and rewrite Eq. (7) as:

$$\frac{d}{dt}\rho = \mathcal{H}_0\rho + \epsilon(t)\mathcal{H}_1\rho + \gamma(t)\mathcal{T}\rho. \tag{14}$$

We introduce the sets of matrices:

$$\Gamma_N = \{Z \in \mathbb{C}^{N \times N} | Z = Z^*\}, \ \Gamma_N^0 = \{Z \in \mathbb{C}^{N \times N} | Z = Z^*, \ tr(Z) = 0\}, \tag{15}$$

and recall that Γ_N (respectively Γ_N^0) has dimension N^2 (respectively $N^2 - 1$) when seen as a vector space over $\mathbb{R}$.

Note that, denoting by ad_x the map $y \mapsto ad_x(y) = [x, y]$ (the adjoint representation), we also have

$$\mathcal{H}_0 = ad_{-iH_0}, \mathcal{H}_1 = ad_{-iH_1}. \tag{16}$$

Also, when $A = A^*$ then $\mathcal{T} = -\frac{1}{2}ad_A \circ ad_A$. A simple computation indicates that for any $Z \in \Gamma_N$: $\mathcal{H}_0(Z), \mathcal{H}_1(Z), \mathcal{T}(Z) \in \Gamma_N$. Moreover all three operators are linear. Thus $\mathcal{H}_0, \mathcal{H}_1, \mathcal{T} \in Lin(\Gamma_N, \Gamma_N)$, the space of linear operators from Γ_N to Γ_N (this also results from Eq. (16)). Also note that for any matrix Z:

$$tr(\mathcal{H}_0 Z) = tr(-i[H_0, Z]) = 0$$
$$tr(\mathcal{H}_1 Z) = tr(-i[H_1, Z]) = 0$$
$$tr(\mathcal{T} Z) = tr\left(AZA^* - \frac{1}{2}ZA^*A - \frac{1}{2}A^*AZ\right) = 0.$$

Thus we also have $\mathcal{H}_0, \mathcal{H}_1, \mathcal{T} \in Lin(\Gamma_N^0, \Gamma_N^0)$. In particular the trace of ρ will not change during the evolution. Moreover as we will see below, when A is Hermitian another conservation law exists: if $\rho(0) = \lambda I$ for some $\lambda \in \mathbb{R}$ then $\rho(t) = \lambda I$ for all $t \geq 0$. This motivates the following definition:

Definition 1 The evolution (14) is density matrix controllable if for any Hermitian matrices ρ_i and ρ_f with $tr(\rho_i) = tr(\rho_f) > 0$ there exists a time $t \geq 0$ and locally bounded measurable controls $\epsilon(\cdot), \gamma(\cdot)$ such that the solution of the evolution Eq. (7) starting at 0 from ρ_i reaches ρ_f at time t. When $A = A^*$ we suppose $\rho_i \neq \frac{tr(\rho_i)}{N} Id, \rho_f \neq \frac{tr(\rho_f)}{N} Id$.

The standard Lie group setting for investigating the control of an isolated quantum system works with the Lie group of unitary (or special unitary) matrices $U(N)$ (or $SU(N)$). Here, on the contrary, we work on the group of one-to-one transformations of Γ_N to Γ_N (or the same for Γ_N^0). As a numerical illustration of the difference, the dimension of $U(N)$ as a real Lie group is N^2 while the dimension of the one-to-one transformations from Γ_N to itself is N^4.

We investigate controllability results in two distinct situations: when the matrix A is Hermitian or not.

Remark 1 The particular case of A being Hermitian corresponds to unital operator $\mathcal{T}$, i.e. $\mathcal{T}I = 0$, which is a situation frequently addressed in quantum information processing.

3.1 Situation I: Hermitian operator A

Let us now consider the connected Lie group G_1 of one-to-one linear transformations of Γ_N^0 that contains the identity operator. The canonical notation of this group is $GL^+(\Gamma_N^0)$. As we do not impose any particular structure on Γ_N^0 except that of vector space, Γ_N^0 is isomorphic to $\mathbb{R}^{N^2-1}$; thus the Lie group $GL^+(\Gamma_N^0)$ is isomorphic to $GL^+(N^2-1)$, the Lie group of invertible matrices of dimension $(N^2-1) \times (N^2-1)$ with positive determinant. In particular its dimension is $(N^2-1)^2$. We know that G_1 is connected but not compact. We will denote by $Lie(G_1)$ the Lie algebra of G_1 which is therefore $Lin(\Gamma_N^0, \Gamma_N^0)$.

Denote by $\mathcal{H}_0^{G_1}$ the element of $Lie(G_1)$ that is constructed from $\mathcal{H}_0$ (and the same for $\mathcal{H}_1$ and $\mathcal{T}$). We associate to the evolution Eq. (14) the following evolution equation on the group G_1:

$$\frac{d}{dt}X(t) = \left(\mathcal{H}_0^{G_1} + \epsilon(t)\mathcal{H}_1^{G_1} + \gamma(t)\mathcal{T}^{G_1}\right)X(t),$$
$$X(t = 0) = X_0. \tag{17}$$

We will also write $X(t; \epsilon, \gamma; X_0)$ when we will need to make explicit the dependence on the parameters; of course $X(t; \epsilon, \gamma; X_0)$ belongs to the Lie group G_1. Then by definition $\rho(t) = X(t; \epsilon, \gamma; e)\rho(0)$ (here e is the identity of the Lie group G_1).

Theorem 2 *If the Lie algebra $Lie\{\mathcal{H}_0^{G_1}, \mathcal{H}_1^{G_1}, \mathcal{T}^{G_1}\} \subset Lie(G_1)$ generated by $\{\mathcal{H}_0^{G_1}, \mathcal{H}_1^{G_1}, \mathcal{T}^{G_1}\}$ has dimension $(N^2 - 1)^2$ (as a vector space over the real numbers), then the system (14) is density matrix controllable.*

Proof Without loss of generality we can suppose that $tr(H_0) = 0$ and $tr(H_1) = 0$.

As a side remark, note that since A is Hermitian we can show by computation that $\mathcal{T}$ is a Hermitian operator from Γ_N^0 to itself i.e., $\mathcal{T}^* = \mathcal{T}$. However $\mathcal{H}_0$ and $\mathcal{H}_1$ are skew-Hermitian as operators from Γ_N^0 to itself.

We consider the following change of variables:

$$\bar{\rho}(t) = \rho(t) - \frac{tr(\rho(0))}{N}I, \tag{18}$$

which implies that $\bar{\rho}(0) \in \Gamma_N^0$. However this does not imply directly that $\bar{\rho}(t) \in \Gamma_N^0$ for all $t > 0$. To prove this one recalls that A is Hermitian and $\mathcal{H}_0, \mathcal{H}_1, \mathcal{T} \in Lin(\Gamma_N^0, \Gamma_N^0)$; after invoking the Frobenius theorem for the equation:

$$\dot{\bar{\rho}} = -i[H, \bar{\rho}] + \mathcal{T}(\bar{\rho}), \tag{19}$$

one obtains that $\bar{\rho}(t) \in \Gamma_N^0$ for all $t \geq 0$.

From the above relations we conclude that $\mathcal{H}_0(\rho)$, $\mathcal{H}_1(\rho)$ and $\mathcal{T}(\rho)$ belong to $Lin(\Gamma_N^0, \Gamma_N^0)$.

The central question is to characterize $\mathcal{R}(e)$ for the system (17). We will use Theorem 1 with $G = G_1$. Then $\mathbb{L} = Lie\left\{i\mathcal{H}_0^{G_1}, i\mathcal{H}_1^{G_1}, \mathcal{T}^{G_1}\right\}$ is the Lie algebra generated by $\mathcal{H}_0^{G_1}$, $\mathcal{H}_1^{G_1}$ and $\mathcal{T}^{G_1}$.

Recall that $\mathbb{L} \subset Lie(G_1)$ and $dim_\mathbb{R}(Lie(G_1)) = dim_\mathbb{R}(Lin(\Gamma_N^0, \Gamma_N^0)) = (N^2 - 1)^2$ (dimension as vector space over $\mathbb{R}$). By hypothesis $dim_\mathbb{R}(\mathbb{L}) = (N^2 - 1)^2$ thus $\mathbb{L} = Lie(G_1)$ and $\mathbb{S} = G_1$. Take now $\epsilon = 0$, $\gamma = 0$, then Eq. (17) becomes:

$$\frac{d}{dt}X(t) = \mathcal{H}_0^{G_1}X(t),$$
$$X(t = 0) = e, \tag{20}$$

with solution $X(t) = e^{t\mathcal{H}_0^{G_1}}$; take a sequence of positive numbers $\{t_n\}$ with $t_n \geqslant \delta > 0$ for some δ (strictly positive, arbitrary but fixed); we need to prove that

$$\lim_{n \to \infty} e^{t_n \mathcal{H}_0^{G_1}} \in \bar{\mathbb{S}}. \tag{21}$$

But, $\mathcal{H}_0^{G_1}$ is a skew-Hermitian map, which means that $e^{t_n \mathcal{H}_0^{G_1}}$ belongs to the group $SO(\Gamma_N^0)$ of the special orthogonal transformations of Γ_N^0; it is known that $SO(\Gamma_N^0)$ is the maximal compact subgroup of $G_1 = GL^+(\Gamma_N^0)$.

By compactness we obtain that up to extracting a subsequence $\lim_{n \to \infty} e^{t_n \mathcal{H}_0^{G_1}}$ exists and belongs to $\bar{\mathbb{S}}$. Therefore $\mathcal{R}(e) = \mathbb{S}$. In particular X can reach any one-to-one transformation from Γ_N^0 to itself. For any ρ_i and ρ_f with $tr(\rho_i) = tr(\rho_i)$, $\rho_i - \frac{tr(\rho_i)}{N}I \neq 0$ and $\rho_f - \frac{tr(\rho_f)}{N}I \neq 0$ we can find a transformation to map the non-null vector $\rho_i - \frac{tr(\rho_i)}{N}I \in \Gamma_N^0$ to the non-null vector $\rho_f - \frac{tr(\rho_f)}{N}I = \rho_f - \frac{tr(\rho_i)}{N}I \in \Gamma_N^0$ i.e., we have controllability for $\bar{\rho}$ thus for ρ.

Remark 2 Following the proof of Theorem 2 we can also conclude that the set of all density matrices reachable from ρ_i (with $\rho_i \neq \frac{tr(\rho_i)}{N}I$) with dynamics described by Eq. (14), for A being a Hermitian operator, is

$$(\rho_i + \Gamma_N^0) \setminus \left\{\frac{tr(\rho_i)}{N}I\right\} = \left\{\rho_i + Z \,\middle|\, Z \in \Gamma_N^0, \quad Z \neq \frac{tr(\rho_i)}{N}I - \rho_i\right\}. \tag{22}$$

3.2 Situation II: arbitrary operator A

Let us now consider the connected Lie group G_2 of one-to-one linear transformations of Γ_N that contains the identity operator and preserves the trace:

$$G_2 := \left\{ X \in GL^+(\Gamma_N) \big| tr(X(Z)) = tr(Z), \quad \forall Z \in \Gamma_N \right\}. \tag{23}$$

By construction of Γ_N we only have a canonical linear space structure, thus Γ_N can be identified with $\mathbb{R}^{N^2}$; and thus G_2 is isomorphic with

$$\left\{ X \in GL(N^2) \big| det(X) \geq 0, tr(X(Z)) = tr(Z), \quad \forall Z \in \mathbb{R}^{N^2} \right\}. \tag{24}$$

We know that G_2 is connected but not compact. We will denote by $Lie(G_2)$ the Lie algebra of G_2. Note that if $\frac{d}{dt}\rho(t) = M(\rho(t))$ then $\rho(t) = \exp(Mt)\rho(0)$. If $tr(\rho(t)) = const$ it implies $\frac{d}{dt}tr(\rho(t)) = 0$. Since $\frac{d}{dt}tr(\rho(t)) = tr(\frac{d}{dt}\rho(t))$ it follows:

$$tr(M(\rho(t))) = 0, \quad \forall \, \rho(t) \in \Gamma_N. \tag{25}$$

Since $tr(\rho)$ is a linear operation and if ρ is viewed as a vector in $\mathbb{R}^{N^2}$ then $tr(\rho) = <\alpha, \rho>$ with α a vector with "1" on the positions corresponding to ρ_{ii}, $i = 1, \ldots, N^2$ and zero elsewhere. In this case the Lie algebra is thus isomorphic with the set (endowed with its canonical Lie algebra structure):

$$\left\{ M \in \mathbb{R}^{N^2 \times N^2} \big| M^T \alpha \equiv 0_{\mathbb{R}^{N^2}} i.e. \sum_{j=1}^{N^2} M_{ji} \alpha_j = 0, \forall i = 1, \ldots, N^2 \right\} \tag{26}$$

We have thus N^2 constraints on $\mathbb{R}^{N^2 \times N^2}$, that means the dimension over $\mathbb{R}$ of the Lie algebra is $(N^2 - 1)N^2$.

Denote by $\mathcal{H}_0^{G_2}$ the element of $Lie(G_2)$ that is constructed from $\mathcal{H}_0$ (and the same for $\mathcal{H}_1$ and $\mathcal{T}$). We associate to the evolution Eq. (14) the following evolution equation on the group G_2:

$$\begin{aligned} \frac{d}{dt}X(t) &= \left(\mathcal{H}_0^{G_2} + \epsilon(t)\mathcal{H}_1^{G_2} + \gamma(t)\mathcal{T}^{G_2} \right) X(t), \\ X(t=0) &= X_0. \end{aligned} \tag{27}$$

We will also write $X(t; \epsilon, \gamma; X_0)$ when we will need to make explicit the dependence on the parameters; of course $X(t; \epsilon, \gamma; X_0)$ belongs to the Lie group G_2. Then by definition $\rho(t) = X(t; \epsilon, \gamma; e)\rho(0)$.

Theorem 3 *If the Lie algebra $Lie\{\mathcal{H}_0^{G_2}, \mathcal{H}_1^{G_2}, \mathcal{T}^{G_2}\}$ generated by $\{\mathcal{H}_0^{G_2}, \mathcal{H}_1^{G_2}, \mathcal{T}^{G_2}\}$ has dimension $(N^2 - 1)N^2$ (as a vector space over the real numbers) then the system (14) is density matrix controllable.*

Proof Without loss of generality we can suppose that $tr(H_0) = 0$ and $tr(H_1) = 0$. We did not yet prove that G_2 is indeed a Lie group: this results from Cartan's theorem that states that any closed subgroup of a Lie group is a Lie subgroup (we include G_2 in the group $GL^+(\Gamma_N)$ of one-to-one transformations on Γ_N of positive determinant). We will use the same line of proof as in Theorem 2 and invoke Theorem 1 (this time without any change of variables) for $G = G_2$. Then $\mathbb{L} = Lie\left\{i\mathcal{H}_0^{G_2}, i\mathcal{H}_1^{G_2}, \mathcal{T}^{G_2}\right\}$ is the Lie algebra generated by $\mathcal{H}_0^{G_2}$, $\mathcal{H}_1^{G_2}$ and $\mathcal{T}^{G_2}$.

Recall that $\mathbb{L} \subset Lie(G_2)$ and $dim_{\mathbb{R}}(Lie(G_2)) = (N^2 - 1)N^2$ (dimension as vector space over $\mathbb{R}$). By hypothesis $dim_{\mathbb{R}}(\mathbb{L}) = (N^2 - 1)N^2$ thus $\mathbb{L} = Lie(G_2)$ and $\mathbb{S} = G_2$. Take now $\epsilon = 0$, $\gamma = 0$, then Eq. (27) becomes:

$$\frac{d}{dt}X(t) = \mathcal{H}_0^{G_2}X(t),$$
$$X(t = 0) = e, \tag{28}$$

with solution $X(t) = e^{t\mathcal{H}_0^{G_2}}$; take a sequence of positive numbers $\{t_n\}$ with $t_n \geqslant \delta > 0$ for some δ (strictly positive, arbitrary but fixed); we need to prove that

$$\lim_{n \to \infty} e^{t_n \mathcal{H}_0^{G_2}} \in \bar{\mathbb{S}}. \tag{29}$$

But $\mathcal{H}_0^{G_2}$ is a skew-Hermitian map which means that $e^{t_n \mathcal{H}_0^{G_2}}$ belongs to the group $SO(\Gamma_N)$ of special orthogonal transformations of Γ_N which is a subgroup of G_2. Since $SO(\Gamma_N)$ is compact it implies that up to extracting a subsequence $\lim_{n \to \infty} e^{t_n \mathcal{H}_0^{G_2}}$ exists and is orthogonal. All orthogonal transformations $e^{t_n \mathcal{H}_0^{G_2}}$ preserve the trace thus $\lim_{n \to \infty} e^{t_n \mathcal{H}_0^{G_2}}$ exists, is orthogonal and trace preserving and hence an element of $\bar{\mathbb{S}}$. Therefore $\mathcal{R}(e) = \mathbb{S}$. In particular X can reach any trace preserving one-to-one transformation from Γ_N to itself. For any ρ_i and ρ_f with $tr(\rho_i) = tr(\rho_i)$ we can find a trace preserving one-to-one transformation to map $\rho_i \in \Gamma_N$ to $\rho_f \in \Gamma_N$ i.e. we have controllability for ρ.

Remark 3 From the proof of Theorem 3 we obtain that the set of all density matrices reachable from ρ_i with dynamics described by Eq. (14), for A arbitrary operator, is the set

$$\rho_i + \Gamma_N^0 = \left\{\rho_i + Z | Z \in \Gamma_N^0\right\}. \tag{30}$$

It should be noted that in particular this set contains matrices that are not necessarily positive semidefinite. This means that the control, even between two states that are positive semidefinite may also use intermediary states $\rho(t)$ that are not physical. Should such a circumstance appear in practice, a numerical algorithm that penalizes, e.g. the presence of non-physical states, may be used. The result above indicates that an initial guess that offers perfect control but with a possible non-physical trajectory, can always be subject to such an algorithm.

Remark 4 A straightforward extension is to consider the circumstance when several non-null controls $\gamma_2, \ldots$ are present; the theoretical results can be proved in the same manner.

Remark 5 In this paper two controllability results are proved for $\gamma(t) \in \mathbb{R}$ but not necessarily positive [13]. However physical considerations may impose γ to be positive (see [6] for more details). Previous controllability results cannot be used due to the loss of compactness. Therefore the controllability analysis of the system defined by (7), for $\gamma(t) \geq 0$, remains for now a conjecture and leads to the question: are Theorems 2 and 3 true if $\gamma(t)$ takes only positive values?

4 Application

In the following we illustrate the theoretical results introduced in the above section. For this purpose consider two finite-dimensional systems defined by

$$H_0 = \begin{pmatrix} -4 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 3 \end{pmatrix}, \quad H_1 = \begin{pmatrix} 0 & -2 & 0 \\ -2 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad A = \begin{pmatrix} 2 & 1 & 3 \\ 1 & -1 & 0 \\ 3 & 0 & -1 \end{pmatrix}, \quad (31)$$

and

$$H_0 = \begin{pmatrix} -4 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & 3 \end{pmatrix}, \quad H_1 = \begin{pmatrix} 0 & -2 & 0 \\ -2 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad A = \begin{pmatrix} 2 & 1 & 1 \\ 1 & -1 & 0 \\ 3 & 0 & -1 \end{pmatrix}. \quad (32)$$

First note that the system defined by H_0 and H_1 alone (i.e., with $A = 0$) is not controllable as the dimension of the Lie algebra generated by $-i H_0$ and $-i H_1$ is 4, short of $3^2 - 1 = 8$ needed for controllability. Also note that for the system defined by (31) A is a symmetric matrix. We want to verify if systems (31) and (32) are controllable i.e., verify the hypotheses of Theorem 2 and Theorem 3.

To do so we choose a parameterization such that we can write (14) as a linear system

$$\frac{d}{dt}\tilde{\rho} = -i\tilde{\mathcal{H}}_0\tilde{\rho} - i\epsilon(t)\tilde{\mathcal{H}}_1\tilde{\rho} + \gamma(t)\tilde{\mathcal{T}}\tilde{\rho} \quad (33)$$

Numerically $\tilde{\mathcal{H}}_0, \tilde{\mathcal{H}}_1, \tilde{\mathcal{T}}$ are $N^2 \times N^2$ dimensional matrices and $\tilde{\rho}$ is a $N^2 \times 1$ vector. For the Hamiltonian part of the dynamics this is known as the Liouville equation in the adjoint representation.

In order to analyze the controllability we need to numerically compute the dimension of the Lie algebra (as subalgebra of $N^2 \times N^2$ matrices) generated by $\{i\tilde{\mathcal{H}}_0, i\tilde{\mathcal{H}}_1, \tilde{\mathcal{T}}\}$, which we denote by $Lie\{i\tilde{\mathcal{H}}_0, i\tilde{\mathcal{H}}_1, \tilde{\mathcal{T}}\}$ and verify for A a Hermitian N-dimensional matrix if $dim_{\mathbb{R}}(Lie\{i\tilde{\mathcal{H}}_0, i\tilde{\mathcal{H}}_1, \tilde{\mathcal{T}}\}) = (N^2 - 1)^2$ or when A is an arbitrary if $dim_{\mathbb{R}}(Lie\{i\tilde{\mathcal{H}}_0, i\tilde{\mathcal{H}}_1, \tilde{\mathcal{T}}\}) = (N^2 - 1)N^2$. For the system in (31) numerical computations give the result

$$dim_{\mathbb{R}}(Lie\{i\tilde{\mathcal{H}}_0, i\tilde{\mathcal{H}}_1, \tilde{\mathcal{T}}\}) = 64, \quad (34)$$

and for the system (32)

$$dim_{\mathbb{R}}(Lie\{i\tilde{\mathcal{H}}_0, i\tilde{\mathcal{H}}_1, \tilde{\mathcal{T}}\}) = 72. \tag{35}$$

We conclude that both systems are controllable.

References

1. A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle, G. Gerber, Science **282**, 919 (1998)
2. R. Judson, H. Rabitz, Phys. Rev. Lett. **68**, 1500 (1992)
3. P. Brumer, M. Shapiro, Acc. Chem. Res. **22**, 407 (1989)
4. R.J. Levis, G. Menkir, H. Rabitz, Science **292**, 709 (2001)
5. T. Weinacht, J. Ahn, P. Bucksbaum, Nature **397**, 233 (1999)
6. H.P. Breuer, F. Petruccione, *The Theory of Open Quantum Systems* (Clarendon Press, Oxford, 2006)
7. V. Gorini, A. Frigerio, M. Verri, A. Kossakowski, E.C.G. Sudarshan, Rep. Math. Phys. **13**, 149 (1978)
8. V. Gorini, A. Kossakowski, E.C.G. Sudarshan, J. Math. Phys. **17**, 821 (1976)
9. F. Haake, Tract. Mod. Phys. **66** (1973)
10. S. Nakajima, Prog. Theor. Phys. **20**, 948 (1958)
11. R. Zwanzing, J. Chem. Phys. **33**, 1338 (1960)
12. T. Arimitsu, F. Shibata, J. Phys. Soc. Jpn. **49**, 891 (1980)
13. H.P. Breuer, Phys. Rev. A **70**, 012 (2004)
14. A. Royer, Phys. Lett. A **315**, 335 (2003)
15. H.P. Breuer, E.M. Laine, J. Piilo, Phys. Rev. Lett. **103**, 210–401 (2009)
16. I.L. Chuang, R. Laflamme, P.W. Shor, W.H. Zurek, Science **270**, 1633 (1995)
17. H. Rabitz, G. Turinici, E. Brown, in *Computational Chemistry, Special Volume (C. Le Bris Editor) of Handbook of Numerical Analysis*, vol X, ed. by P.G. Ciarlet (Elsevier, Amsterdam, 2003), pp. 833–887
18. A. Pechen, H. Rabitz, Phys. Rev. A **73**, 062–102 (2006)
19. R. Wu, A. Pechen, C. Brif, H. Rabitz, J. Phys. A **40**, 40–5681 (2007)
20. C. Altafini, J. Math. Phys. **44**(6), 2357 (2003)
21. C. Altafini, Phys. Rev. A **70**, 062–321 (2004)
22. A. Pechen, Phys. Rev. A **84**, 042–106 (2011)
23. C. Altafini, J. Math. Phys. **43**(5), 2051 (2002)
24. M. Dahleh, H. Rabitz, V. Ramakrishna, A. Pierce, M.A. Salapaka, Phys. Rev. A **51**(2), 960 (1995)
25. H. Rabitz, G. Turinici, J. Phys. A **36**, 2565 (2003)
26. V. Jurdjevic, H.J. Sussmann, J. Differ. Equ. **12**, 313 (1972). doi:10.1016/0022-0396(72)90035-6