Beyond bilinear controllability: applications to quantum control

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Abstract. Quantum control is traditionally expressed through bilinear models and their associated Lie algebra controllability criteria. But, the first order approximation are not always sufficient and higher order developpements are used in recent works. Motivated by these applications, we give in this paper a criterion that applies to situations where the evolution operator is expressed as sum of possibly non-linear real functionals of the same control that multiplies some time independent (coupling) operators.

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1. Background on quantum control

Controlling the evolution of molecular systems at quantum level has been envisioned from the very beginnings of the laser technology. However, approaches based on designing laser pulses based on intuition alone did not succeed in general situations due to the very complex interactions that are at work between the laser and the molecules to be controlled, which results e.g., in the redistribution of the incoming laser energy to the whole molecule. Even if this circumstance initially slowed down investigations in this area, the realization that this inconvenient can be recast and attacked with the tools of (optimal) control theory \cite{18} greatly contributed to the first positive experimental results \cite{2, 21, 33, 6, 5, 17, 20}.

The regime that is relevant for this work is related to time scales of the order of the femtosecond ($10^{-15}$) up to picoseconds ($10^{-12}$) and the space scales from the size of one or two atoms to large polyatomic molecules.

Historically, the first applications that were envisioned were the manipulation of chemical bonds (e.g., selective dissociation) or isotopic separation. Although initially, only few atoms molecules were investigated (di-atomics) the experiments soon were designed to treat more complex situations \cite{2} as selective bond dissociation in an organi-metallic complex $\text{CpFe(CO)}_2\text{Cl}$ ($\text{Cp}$ is the cyclopentadienyl ion) by maximizing or minimizing the quotient of $\text{CpFeCOCl}^+$ ions obtained with respect to $\text{FeCl}^+$ ions.

Continuing this breakthrough, other poly-atomic molecules were considered in strong fields. For instance, in \cite{21} the molecules are the acetone $(\text{CH}_3)_2\text{CO}$, the trifluoroacetone $\text{CH}_3\text{COCF}_3$ and the acetophenone $\text{C}_6\text{H}_5\text{COCH}_3$. Using tailored laser pulses it was shown possible to obtain $\text{CH}_3\text{CO}$ from $(\text{CH}_3)_2\text{CO}$, $\text{CF}_3$ (or $\text{CH}_3$) from $\text{CH}_3\text{COCF}_3$ but also $\text{C}_6\text{H}_5\text{CH}_3$ (toluene) from $\text{C}_6\text{H}_5\text{COCH}_3$.

But the applications of laser control do not stop here. High Harmonic Generation) \cite{7} is a technique that allows to obtain output lasers whose frequency is large integer multiples of the input pulses.

A different class of applications works in a different regime of shorter time scales and large intensity. This regime is additionally not compatible with the standard Born-Oppenheimer approximation and requires to consider both nuclei and electrons as quantum particles with entangled wavefunction \cite{4}.

In a different framework, the manipulation of quantum states of atoms and molecules allows to envision the construction of quantum computers \cite{13, 27}.

Finally, biologically related applications are also the object of ongoing research.

2. Background on controllability criteria

We start in this section to investigate the theoretical controllability results that are nowadays available for quantum systems. The evolution of the system will be described by the driving Schrödinger equation (we work here in atomic units i.e.
\[ h = 1 \]

\[
\frac{\partial}{\partial t} \Psi(t, x) = H(t) \Psi(t, x) \quad (2.1)
\]

where \( H(t) \) is the Hamiltonian of the system and \( x \in \mathbb{R}^\gamma \) the set of internal degrees of freedom. We introduce the Hilbert space structure given by the scalar product

\[
\langle f, g \rangle = \int_{\mathbb{R}^\gamma} f(x)g(x)dx \quad (2.2)
\]

where \( a + ib = a - ib \) the conjugate of a complex number.

We only consider in this paper situations when the Hamiltonian is auto-adjoint \( H(t) = H(t) \)\(^\dagger\); we denoted by \( T \) the adjoint of a operator \( T \). The auto-adjointeness of \( H \) implies that the \( L_2^2(\mathbb{R}^\gamma) \) norm of the evolving state is conserved. Indeed

\[
\frac{d}{dt} \| \Psi(x, t) \|_{L_2^2(\mathbb{R}^\gamma)} = \frac{d}{dt} \langle \Psi(x, t), \Psi(x, t) \rangle
\]

\[
= \langle \frac{d}{dt} \Psi(x, t), \Psi(x, t) \rangle + \langle \Psi(x, t), \frac{d}{dt} \Psi(x, t) \rangle
\]

\[
= \langle \frac{H(t)}{i} \Psi(x, t), \Psi(x, t) \rangle + \langle \Psi(x, t), \frac{H(t)}{i} \Psi(x, t) \rangle = 0. \quad (2.3)
\]

Thus

\[
\| \Psi(x, t) \|_{L_2^2(\mathbb{R}^\gamma)} = \| \Psi_0 \|_{L_2^2(\mathbb{R}^\gamma)}, \quad \forall t > 0,
\]

so the wave function \( \Psi(t) \), evolves on the (complex) unit sphere

\[
S = \left\{ \psi \in L^2(\mathbb{R}^\gamma) : \| \psi \|_{L_2^2(\mathbb{R}^\gamma)} = 1 \right\}.
\]

When the system evolves freely under its own internal dynamics i.e. when isolated molecules are considered, the free evolution Hamiltonian \( H_0 \) is introduced. This Hamiltonian is the sum of the kinetic part \( T \) and the potential operator \( V(x) \): \( H_0 = T + V(x) \). A prototypical example of \( T \) is the Laplace operator while for \( V(x) \) one can encounter Coulomb potential or Lennard-Jones type dependence. We obtain the following evolution in the absence of external interaction:

\[
\frac{i}{\partial t} \Psi(t, x) = H_0 \Psi(t, x) \quad (2.5)
\]

\[
\Psi(t_0, x) = \Psi_0(x).
\]

But, when the free evolution of the system does not generate a satisfactory dynamical output, an external interaction is introduced to control it. An example of external control of paramount importance is a laser source of intensity \( \epsilon(t) \in \mathbb{R}, \ t \geq 0 \).

The purpose of control may be formulated as to drive the system from its initial state \( \Psi_0 \) to take a convenient dynamical path to a final state compatible with predefined requirements. The control is here the laser intensity \( \epsilon(t) \). We will come back later with details on the laser field \( \epsilon(t) \).
This laser will modify the Hamiltonian $H(t)$ of the system. A first order approximation can be considered by introducing a time-independent dipole moment operator $\mu(x)$ resulting in the dynamics:
\[
i \frac{\partial}{\partial t} \Psi(t, x) = (H_0 + \epsilon(t)\mu) \Psi(t, x)
\]
\[
\Psi(t_0, x) = \Psi_0(x).
\]
This is the so-called bi-linear framework (the control enters linearly multiplying the state), that is the object of much theoretical and numerical work in quantum control. We also review below some of the results that are available in this formulation. However, recently, higher order field dependence has been considered in different circumstances see e.g., [14, 15] for details. In these situations the Hamiltonian $H(t)$ is developed further as:
\[
H(t) = H_0 + \epsilon_1(t)\mu_1 + \epsilon_2(t)\mu_2 + \ldots + \epsilon_L(t)\mu_L.
\]
(2.7)

The question that will be of interest to us in this work is the study of all possible final states for the quantum system. This question is important in order to understand the capabilities that a laboratory experiment will be able to provide and also, in a more general setting, to accompany the introduction of new experimental protocols.

More specifically, we will show how the criteria available for bilinear control can be extended to treat the Hamiltonian (2.7) where a single control amplitude $\epsilon(t)$ appears before different coupling operators $\mu_1, \ldots, \mu_L$.

Many of the questions regarding the properties of the quantum control procedures, such as controllability, optimal control definition, etc, ... need, in order to be defined, to specify the admissible control class, i.e., the set $\mathcal{U}$ where the control $\epsilon(t)$ is allowed to vary. Among the properties that can define this admissible set, some are related to the regularity of the time-dependence ($L^2, H^1, \ldots$ etc) or of the Fourier expression (sum of sinusoidal functions multiplied by an overall envelope, etc,...) or to additional structure: e.g. piecewise continuous, piecewise constant, locally bounded ...

The choice of one or several conditions in the list above is motivated in practice by capability to reproduce that particular form or to inherent experimental restrictions (finite total laser energy/fluence, etc). As the laser technology is constantly evolving, the first class of constraints becomes less critical and thus it is realistic to consider very weak constraints on the control set, e.g. $\mathcal{U} = L^2(\mathbb{R}) \cap L^\infty_{\text{loc}}(\mathbb{R})$.

However, to treat even more general situations, we will consider in this work controls $\epsilon(t)$ that are piecewise constant, taking any value in a set $V$, which will remain fully general.

2.1. Infinite dimensional bilinear control

When compared to the finite dimensional control equations (see Section 2.2), controllability of the infinite dimensional version of the bilinear Time Dependent
Schrödinger Equation is much less understood at this time. In fact, most of the progress obtained so far takes the form of negative results, in contradiction with the positive results available in finite dimensional settings. However we see the absence of positive controllability results is rather a failure of today’s control theory tools to provide insight into controllability rather than an actual restriction. We do believe that new tools and concepts will make positive results possible.

Let us write the solution of (2.6) in the following form:

$$\Psi(t) = e^{-iH_0t}\Psi_0 - i \int_0^t \epsilon(s)e^{-iH_0(t-s)}\mu\Psi(s)ds$$

(2.8)

This formulation (see [11] for details) is granted by the properties of the operator $\mu : H^1_0(\mathbb{R}^\gamma) \rightarrow H^{-1}(\mathbb{R}^\gamma)$ which is continuous when $\mu$ is bounded; we also recall that the control $\epsilon$ can be considered bounded in both $L^\infty$ and $L^2$.

The application $\epsilon(t) \mapsto \Psi(x,t)$ possesses an important compact property which is the key of the controllability results (we refer the reader interested in details to [3, 25]):

**Lemma 2.1.** Suppose that $\mu : X \rightarrow X$ is a bounded operator and that $H_0$ generates a $C^0$ semigroup of bounded linear operators on some Banach space $X$ (e.g. $X = H^1_0(\mathbb{R}^\gamma)$). Denote for $T > 0$ and $\epsilon \in L^1([0,T])$ by $\Psi_\epsilon(x,t)$ the solution of (2.6) with control $\epsilon$. Then $\epsilon \mapsto \Psi_\epsilon$ is a compact mapping in the sense that for any $\epsilon_n$ that converges weakly to $\epsilon$ in $L^1([0,T])$ $\Psi_{\epsilon_n}$ converges strongly in $C([0,T];X)$ to $\Psi_\epsilon$.

This compactness property allows to give negative results for general bilinear controllability settings as in [3] where they were applied to the wave and rod equations. Specific statements for quantum control have been latter derived (Thm. 1 from [30] ; see also [3, 29]) and can be stated as:

**Theorem 2.2.** Let $S$ be the complex unit sphere of $L^2(\mathbb{R}^\gamma)$. Let $\mu$ be a bounded operator from the Sobolev space $X$ (e.g., $X = H^1_0(\mathbb{R}^\gamma)$) to itself and let $H_0$ generate a $C^0$ semigroup of bounded linear operators on $X$. Denote by $\Psi_\epsilon(x,t)$ the solution of (2.6). Then the set of attainable states from $\Psi_0$ defined by

$$\mathcal{AS} = \cup_{T>0}\{\Psi_\epsilon(x,T); \epsilon(t) \in L^2([0,T])\}$$

(2.9)

is contained in a countable union of compact subsets of $X$. In particular its complement $S \setminus \mathcal{AS}$ with respect to $S \cap X$ is everywhere dense on $S \cap X$. The same holds true for the complement with respect to $S$.

In a different formulation, the theorem implies that for any $\Psi_0 \in X \cap S$, within any open set around an arbitrary point $\Psi \in X \cap S$ there exists a state unreachable from $\Psi_0$ with $L^2$ controls.

**Remark 2.3.** Note that the result does not give information on the closure of the set $\mathcal{AS}$. In particular it may well be that while $\mathcal{AS}$ still has dense complement its closure be the whole space $X$. This would be the so-called approximate controllability i.e. the possibility to reach targets arbitrarily close to any given final state.
Despite some attempts in the literature, at this time there is no answer (positive or negative) to this question. Among the ingredients that make this study difficult we can mention the possibility to use arbitrary large final time $T$, the necessity to treat the continuous spectrum of the operator $H_0$ and the intrinsically unbounded domain on which the problem is posed.

To complicate even more the landscape, situations exists where the results obtained in infinite and finite dimensional representation are of different nature. We will illustrate with a classical result on the harmonic oscillator.

**Lemma 2.4.** The infinite dimensional harmonic oscillator $H_0 = -\frac{\partial^2}{\partial x^2} + x^2$, $\mu = x$ is not controllable. Moreover the set of all admissible states is a low-dimensional manifold of $L^2$.

**Proof.** Let us begin by noting that the operators $-iH_0$ and $-i\mu$ form a Lie algebra of dimension 4. Indeed, let us compute the iterated commutators of $H_0 = -\frac{\partial^2}{\partial x^2} + x^2$ and $\mu = x$:

\[
[i(-\frac{\partial^2}{\partial x^2} + x^2), ix] = 2 \frac{\partial}{\partial x} \tag{2.10}
\]
\[
[ix, \frac{\partial}{\partial x}] = -i \tag{2.11}
\]
\[
[i(-\frac{\partial^2}{\partial x^2} + x^2), i\frac{\partial}{\partial x}] = -2ix \tag{2.12}
\]

Thus the dimension of the Lie algebra = 4 and as such the system cannot be controllable (all the states are on a low dimensional manifold of $L^2$). We refer to [23] for recent contributions when the algebra of the operators $H_0$ and $\mu$ is finite dimensional.

This result is to be contrasted with additional works that show that any (spectral) truncation of the harmonic oscillator is controllable (see [26] for details).

What can be deduced from the above result is that truncating an infinite dimensional system is not always justified and care must be taken to check that the control obtained in the resulting finite dimensional approximation remain a good control for the initial, infinite dimensional system. Of course, this is not needed for situations which are inherently finite dimensional quantum systems (e.g., spins).

**2.2. Finite dimensional bilinear control**

Here our focus will be on finite dimensional systems. We introduce an orthonormal basis $D = \{\psi_i(x); i = 1,..,N\}$ for a finite dimensional space. An important example of such a space is the one spanned by the first $N$ eigenstates of the internal Hamiltonian $H_0$. This example is also motivated in bi-linear settings by the “perturbation” argument that considers the control term $\epsilon(t)\mu$ as a first order developpement of $H(t)$ around $H_0$. Note however that no concept of “smallness” is introduced in the definition of admissible controls $U$. 

Denote by \( M \) the linear space that \( D \) generates, and let \( H_{0;\alpha,b} = \langle H_0 \psi_\alpha, \psi_b \rangle \) and \( \mu_{\ell;\alpha,b} = \langle \psi_\alpha, \mu_\ell \psi_b \rangle \) be the expressions of the operators \( H_0 \) and \( \mu_\ell \) with respect to this basis, \( \ell = 1, \ldots, L \). To keep notations simple we will still denote from now on by \( H_0 \) and \( \mu_\ell \) the resulting \( N \times N \) symmetric matrices.

In the Galerkin approach, expressing the Schrödinger equation in the space \( M \) is equivalent to supposing \( \Psi(x,t) = \sum_{i=1}^N \psi_i(x) c_i(t) \).

\[
i \frac{dc(t;\epsilon;c_0)}{dt} = H_0 c(t;\epsilon;c_0) + \left[ \epsilon(t) \mu_1 + \cdots + \epsilon^L(t) \mu_L \right] c(t;\epsilon;c_0) \quad (2.13)
\]

\[
c(t = 0;\epsilon;c_0) = c_0.
\]

In the following, when no ambiguity prevents it, we will also simply denote \( c(t;\epsilon;c_0) = c(t;\epsilon;c_0) \). The finite dimensional counterpart of the norm conservation property (2.4) reads:

\[
\sum_{n=1}^N |c_n|^2 = 1.
\]

i.e., the state \( c \) evolves on the unit sphere \( S_N \) of \( \mathbb{C}^N \). The controllability can be formulated in this case as:

**Definition 2.5.** The system \((H_0,\mu_1,\ldots,\mu_L)\) is called (wavefunction) controllable, if for any two states \( c_k \in S_N \), \( k = 1,2 \) there exists a final time \( T < \infty \) and control \( \epsilon(t) \in L^2([0,T]) \) such that the solution of eqn. (2.13) starting from \( c_1 \) ends in \( c_2 \) at final time \( T \):

\[
c(T;\epsilon;c_1) = c_2.
\]

Although specific results for this setting exist \[31, 32\], a different alternative is to see (2.13) as a system posed on \( U(N) \). We introduce the evolution equation on \( U(N) \):

\[
i \frac{dU(t;\epsilon)}{dt} = \left[ H_0 + \epsilon(t) \mu_1 + \cdots + \epsilon^L(t) \mu_L \right] U(t;\epsilon) \quad (2.14)
\]

\[
U(t = 0;\epsilon) = Id.
\]

Since \( H_0 \) and \( \mu_\ell \) are symmetric matrices, \( U(t;\epsilon) \) will remain unitary for all \( t \geq 0 \). It is classical to remark then that the evolution of \( c(t;\epsilon;c_0) \) can be obtained from the evolution of \( U(t;\epsilon) \) by

\[
c(t;\epsilon;c_0) = U(t;\epsilon)c_0.
\]

In particular it follows that if the set of all attainable matrices \( U(t;\epsilon) \) is at least \( SU(N) \) then the system is controllable. This is almost a necessary condition for controllability, a notable exception being the circumstance when \( N \) is even: in this case, if the set of all attainable matrices contains \( Sp(N/2) \) then controllability still holds. We refer to \[10, 1\] for more detailed information.

\[\text{1} U(N) \text{ is the set of all } N \times N \text{ complex unitary matrices.}\]
Let us just mention that different representations of the system include the density matrix formulation with time dependent density matrix operator $\rho(t)$ satisfying

$$\frac{\partial}{\partial t} \rho(t; \epsilon; \rho_0) = [H_0 + \epsilon(t)\mu_1 + ... + \epsilon^L(t)\mu_L, \rho(t; \epsilon; \rho_0)]$$  \hspace{1cm} (2.15)

$$\rho(t = 0; \epsilon; \rho_0) = \rho_0$$

Then one can show $\rho(t; \epsilon; \rho_0) = U(t; \epsilon)\rho_0 U^\dagger(t; \epsilon)$. Controllability in this case is the possibility to steer any initial mixed state $\rho_0$ to any other state $\rho_f$ unitarily equivalent to it.

Note that the density matrix controllability is equivalent to requiring that the set of all matrices attainable from identity be at least $SU(N)$.

At a general level, the evolution equation (2.14) can be re-written as

$$\frac{dx(t; \epsilon; x_0)}{dt} = (A + \epsilon(t)B_1 + ...\epsilon^L(t)B_L)x(t; \epsilon; x_0)$$  \hspace{1cm} (2.16)

$$x(0) = x_0.$$  \hspace{1cm} (2.17)

where $x(t; \epsilon; x_0)$ belongs to a Lie group $G$ (see [10, 8, 9] for basic facts about the lie groups) and $A, B_1, ..., B_K$ to its associated Lie algebra $L(G)$. The equation above is to be taken in the usual sense (using the exponential map) when e.g., $\epsilon(t)$ is piecewise continuous/constant and in a weak sense (integral form) for general $\epsilon(t)$ (see e.g. [8] for additional details). For the quantum control problem $A = -iH_0$ and $B_\ell = -i\mu_\ell$, $G = U(N)$.

Remark 2.6. Everything that will be said in this and following sections applies with trivial modifications to the situation of several laser fields. For notational conveniences we will only give here the results for a unique laser field.

We will denote by $L_{A,B_1,...,B_k} \subset L(G)$ the Lie algebra spanned by $A, B_k$, $k = 1, ..., K$ and by $e$ the unity of $G$.

Let us now consider the set of all reachable states from an initial state $y$:

$$\mathcal{R}_t^t(y) = \{x(t; \epsilon; y) \text{ solution of } (2.16); \epsilon \in U\}.$$  \hspace{1cm} (2.18)

It is immediate to see that

$$\mathcal{R}_t^t(y) = \mathcal{R}_t^t(e)y$$  \hspace{1cm} (2.19)

and thus, describing the set $\mathcal{R}_t^t(e)$ allows to completely describe all other reachable sets. When the final time is not specified, we will denote

$$\mathcal{R}_U(y) = \cup_{t \geq 0} \mathcal{R}_t^t(y).$$  \hspace{1cm} (2.20)

The central question is to characterize $\mathcal{R}_U(e)$. When the bi-linear setting is considered i.e. $L = 1$ and we note $B = B_1$, we have the following result [19, 22]:

---

2 A $N \times N$ matrix $\rho_2$ is said unitarily equivalent to a $N \times N$ matrix $\rho_1$ if there exists $M \in U(N)$ such that $\rho_2 = M \rho_1 M^\dagger$. 

Theorem 2.7. Consider the system (2.16) defined on a Lie group $G$ with associated Lie algebra $L(G)$ containing $A$ and $B$. If $G$ is compact and the Lie algebra $L_{A,B}$ generated by $A$ and $B$ is the complete algebra $L(G)$: $L_{A,B} = L(G)$ then the set $\mathcal{R}_U(e)$ of all states states from the identity is the Lie group $G$. Moreover, there exists $0 < T < \infty$ such that $\mathcal{R}_U^{T'}(e) = G$ for all $T' \geq T$.

This gives, when applied to quantum control [24]: $(L = 1, \mu = \mu_1)$:

Theorem 2.8. If the Lie algebra $L_{-iH_0,-i\mu}$ generated by $-iH_0$ and $-i\mu$ has dimension $N^2$ (as a vector space over the real numbers) then the system (2.14) is density matrix controllable. Furthermore, if both $-iH_0$ and $-i\mu$ are traceless then a sufficient condition for the density matrix (thus wavefunction) controllability of quantum system is that the Lie algebra $L_{-iH_0,-i\mu}$ has dimension $N^2 - 1$.

Although the results above conveniently address the situation of a bi-linear setting, we are not aware of any similar results for the general quantum control situations (2.7). In particular, we know by the result above that, if $u_1,\ldots,u_L$ are independent controls, i.e.,

$$\frac{dx(t; \epsilon; x_0)}{dt} = (A + u_1(t)B_1 + \ldots + u_L(t)B_L)x(t; \epsilon; x_0) \quad (2.21)$$

$$x(0) = x_0, \quad (2.22)$$

an equivalent condition for the controllability of the above system on its compact Lie group $G$ is that $A,B_1,\ldots,B_L$ generate the whole Lie algebra $L(G)$. But, there is no obvious way to say what will happen when the controls $u_\ell$ are not independent but related by the condition $u_\ell = \epsilon^\ell(t)$. This study is the purpose of the next section.

3. Criteria for non linear operators

In order to extend the controlability results above beyond bi-linear interaction Hamiltonians, we will introduce in this section a more general setting: we will rewrite the control equation (2.16) as

$$\frac{dx(t; \epsilon; x_0)}{dt} = (F_1(\epsilon(t))B_1 + \ldots + F_L(\epsilon(t))B_L)x(t; \epsilon; x_0) \quad (3.1)$$

$$x(0) = x_0. \quad (3.2)$$

where $F_k : V \to \mathbb{R}$ are real functionals. Note in particular that we do not impose any assumption on the regularity of the functionals $F_k$. Of course, one can recover the equation (2.16) by setting $F_k(x) = x^k$ and adding $F_0 = 1$.

In order to avoid trivialities, we will suppose in the following that

the functionals $(F_k)_{k=1}^L$ are linearly independent. (3.3)

Otherwise one may just consider a subset of functionals that are linearly independent and adjust the matrices $B_k$ accordingly. Of course, since we do not specify the set $V$ that lists all the possible control values $\epsilon$ the hypothesis above needs to
be understood in the following acception: the functionals \( F_k \) are said to be linearly dependent if there exist constants \( \lambda_1, \ldots, \lambda_L \in \mathbb{R} \) such that \( \sum_{j=1}^{L} \lambda_j F_j(v) = 0 \) for all \( v \in V \). Otherwise the functionals are said to be linearly independent.

In order to obtain the quantum controllability results, we begin in this section with a controllability criterion on compact Lie groups. These results build on classical references for bilinear controllability [19]. We give first a weak but intuitive form and then we state the fully general one.

**Theorem 3.1.** Let \((3.1)\) be a control system posed on a compact connected Lie group \( G \), with linearly independent functionals \( (F_k)_{k=1}^{L} \). Then if the Lie algebra generated by \( B_1, \ldots, B_L \) is the full Lie algebra \( L(G) \) of the group \( G \), then the system is approximatelly controllable, i.e. for any \( a, b \in G \), \( b \) is an accumulation point of the set of all states \( x(t) \) attainable from \( x(0) = a \) with admissible controls.

**Proof.** Let us begin by noting that if \( F_k \) are independent then there exist values \( e_j \in V \), \( j = 1, \ldots, L \) such that the vectors \( v(e_j) = (F_1(e_j), \ldots, F_L(e_j)) \) are linearly independent. Suppose on the contrary that this is not true. Consider then a maximal set of vectors \( v(E_1), \ldots, v(E_p) \) that are linearly independent. The matrix \( (F_k(E_j))_{k=1}^{L_p} \) has rank precisely \( p \) and thus one can extract \( p \) functionals, denoted for notational convenience \( F_1, \ldots, F_p \) such that \( \text{rank}(F_k(E_j))_{k=1}^{p+1} = p \).

Take now some functional \( F_{p+1} \) not in this set. It follows that \( \text{rank}(F_k(E_j))_{k=1}^{p+1} = p \) and as such \( \det(F_k(E_j))_{k=1}^{p+1} = 0 \) for any \( E_{p+1} \in V \). This determinant can be computed as:

\[
\det(F_k(E_j))_{k=1}^{p+1} = \lambda_1 F_1(E_{p+1}) + \ldots + \lambda_{p+1} F_{p+1}(E_{p+1}) = 0. \quad (3.4)
\]

Note that \( \lambda_k \) do not depend on \( E_{p+1} \) and that in particular

\[
\lambda_{p+1} = \det(F_k(E_j))_{k=1}^{p+1} \neq 0.
\]

Thus eq. (3.4) implies that a linear combination with at least one non-null coefficient \( \lambda_{p+1} \) exists such that \( \sum_{k=1}^{L} \lambda_k F_k(E) = 0 \) for all \( E \in V \). This is prevented by hypothesis.

We have thus proved the existence of \( e_j \in V \), \( j = 1, \ldots, L \) with the \( v(e_j) = (F_1(e_j), \ldots, F_L(e_j)) \) linearly independent. This means that \( M_j = \sum_{k=1}^{L} F_k(e_j) B_k \) are also linearly independent and span the same linear space as \( B_k, k = 1, \ldots, L \) and thus \( M_j \) span also the Lie algebra \( L(G) \). Moreover, all states \( \{e^{M}; x(0); t \in \mathbb{R}_+, j \leq L \} \) are attainable from \( x(0) \) for the control system \((3.1)\).

It is clear that to prove approximate controllability is sufficient to set \( a = e \) the neutral element of the group \( G \), i.e. we have to prove that the closure \( \mathcal{R}_d(e) \) (with respect to the Lie group topology) of the reachable states from identity is the whole \( G \). From the hypothesis and surjectivity of the exponential mapping this is equivalent to proving that

\[
\{e^{M}; M \in L(G)\} \subset \mathcal{R}_d(e).
\]
We will begin by noting that $\overline{R_U(e)}$ is a group. Indeed, take two elements $x(t_1;\epsilon_1;e), x(t_2;\epsilon_2, e) \in \overline{R_U(e)}$. Then, defining the control $\epsilon_{12} : [0, t_1 + t_2] \to \mathbb{R}$ by $\epsilon_{12}(t) = \epsilon_1(t)$ for all $0 \leq t \leq t_1$ and $\epsilon_{12}(t_1 + t) = \epsilon_2(t)$ for all $0 \leq t \leq t_2$ we obtain $x(t_1 + t_2; e) = x(t_2; \epsilon_2, e)x(t_1; \epsilon_1, e)$ and thus $x(t_2; \epsilon_2, e)x(t_1; \epsilon_1, e) \in \overline{R_U(e)}$. Hence $\overline{R_U(e)}$ is a semi-group which implies that $\overline{R_U(e)}$ is a semi-group too.

Let us now consider $a \in \overline{R_U(e)}$. Then $a^n \in \overline{R_U(e)}$ for any $n = 1, 2, \ldots$. Since $\overline{R_U(e)} \subseteq G$ which is a compact group, $\overline{R_U(e)}$ is compact at its turn. Then there exists a sequence, that we can take such that $n_k$ with $n_k - n_{k-1} \geq 2$, with $a^{n_k} \to b \in G$. But then $\overline{R_U(e)} \supseteq a^{n_k - n_{k-1} - 1} \to b^{-1}a^{-1}$ and thus $a^{-1} \in \overline{R_U(e)}$.

It is immediate to see that, since the solution for the control $\epsilon(t) \equiv \epsilon_j$ is $x(t; 0, e) = e^{tm_j}$ we have the inclusion $\{e^{tm_j}; t \geq 0, j \leq L\} \subseteq \overline{R_U(e)}$. Since $\overline{R_U(e)}$ is a group, we will also have $\{e^{tm_j}; t \in \mathbb{R}; j \leq L\} \subseteq \overline{R_U(e)}$. Consider now two matrices $X_1, X_2 \in L(G)$ such that

$$\{e^{tx_i}; t \geq 0\} \subseteq \overline{R_U(e)}, \quad i = 1, 2.$$ 

We invoke now the formula

$$e^{[X_1, X_2]} = \lim_{n \to \infty} \left( e^{-tX_2/\sqrt{n}} e^{-tX_1/\sqrt{n}} e^{tX_2/\sqrt{n}} e^{tX_1/\sqrt{n}} \right)^n$$

(3.5)

to conclude that

$$\{e^{[X_1, X_2]}; t \in \mathbb{R}\} \subseteq \overline{R_U(e)}.$$ 

Similarly, we use the formula $e^{t_1X_1 + t_2X_2} = \lim_{n \to \infty} \left( e^{t_1X_1/n} e^{t_2X_2/n} \right)^n$ to conclude that

$$\{e^{t_1X_1 + t_2X_2}; t_1, t_2 \in \mathbb{R}\} \subseteq \overline{R_U(e)}.$$ 

We have thus proved that the set $\{M \in L(G); e^{tM} \in \overline{R_U(e)}; \forall t \in \mathbb{R}\}$ contains $M_j, j = 1, \ldots, L$, is closed to commutation and is a real vector space. Thus it contains $L(G)$ hence the conclusion of the theorem. \hfill \Box

The Theorem above has the advantage to be both intuitive and self-contained. However it only gives approximate controllability results, which are not the strongest forms available. But, in order to obtain exact controllability more involved techniques are needed. In the literature, similar situations are treated by making use of the Chow theorem [12] and of the bi-linear control techniques [19, 28]. The criterion can be stated as follows:

**Theorem 3.2.** Let (3.3) be a control system posed on a compact connected Lie group $G$ with linearly independent functionals $F_k : V \to \mathbb{R}$, $k = 1, \ldots, L$ and piecewise constant controls $\epsilon$ taking any value in some set $V$. Then a necessary and sufficient condition for the exact controllability is that the Lie algebra $L_{B_1, \ldots, B_L}$ generated by $B_1, \ldots, B_L$ be the full Lie algebra $L(G)$ of the group $G$. 

Proof. We recall (see also end of Section 2.2) that the set of attainable states is included in the set of attainable states for the system

\[
\frac{dx(t; \epsilon; x_0)}{dt} = [u_1(t)B_1 + \ldots + u_L(t)B_L]x(t; \epsilon; x_0)
\]

(3.6)

\[x(0) = e,\]

(3.7)

whose controllability is equivalent to “\(L_{B_1, \ldots, B_L} = L(G)\)”. Thus \(L_{B_1, \ldots, B_L} = L(G)\) is a necessary condition for controllability. To prove that is also sufficient, consider as in the proof of the Theorem 3.1, the matrices \(M_j = \sum_{k=1}^L F_k(e_j)B_k, j = 1, \ldots, L\) that generate the same Lie algebra \(L_{B_1, \ldots, B_L}\). We recall that all states \(\{e^{tM_j}; t \in \mathbb{R}_+, j \leq L\}\) and all finite products of such states are attainable from the identity \(e\).

We invoke now a technique present in the proof of Thm. 3.1 of [28]: for any \(P \in \mathbb{N}\) and any multi-index \(i = (i_1, \ldots, i_r) \in \{1, \ldots, L\}^r\) denote by \(A(i, T)\) the attainable states with the sequence of operators \(i\) and total time less than \(T\):

\[A(i, T) = \left\{ \prod_{\ell=1}^r e^{t_\ell M_{i_{\ell}}}; \sum_{\ell=1}^r |t_\ell| \leq P, t_1, \ldots, t_r \in \mathbb{R} \right\}.
\]

We know by the Chow theorem that the union of the sets \(A(i, T)\) is the whole Lie group \(G\). Also, it is immediate that any \(A(i, T)\) is image of a compact set thus compact. It follows by the Baire category theorem that \(A(i, P)\) has non-empty interior for a couple \((i, P)\). For such an \(i = (i_1, \ldots, i_m)\) we introduce the mapping \(F : \mathbb{R}^m \to G\) defined by \(t = (t_1, \ldots, t_m) \mapsto F(t) = \prod_{\ell=1}^m e^{t_\ell M_{i_{\ell}}}.\) This mapping is analytic and its image is has nonempty interior. By the Sard theorem its differential \(dF(t)\) has full rank (i.e. equals the dimension of the tangent space \(TG\) of \(G\)) at least at some point \(t\) and thus in a neighborhood. But since \(dF(t)\) depends analytically on \(t\) the set of points where the rank is full is dense in \(\mathbb{R}^m\) and as such the rank is full for some \(t\) with all components strictly positive. Using a local inverse mapping theorem it follows that the image \(F(T)\) has non-empty interior where \(T\) is an open subset of \(\mathbb{R}^m\). But all points in \(F(T)\) are realisable with admissible controls and thus the set of reachable points \(R_{\mathcal{U}}(e)\) contains an open subset \(D\) of \(G\).

By the previous Theorem, \(R_{\mathcal{U}}(e)\) is a subgroup i.e. for any \(y \in R_{\mathcal{U}}(e)\) the set \(Dy\) is also reachable. Since in addition \(R_{\mathcal{U}}(e)\) is dense in \(G\) it follows that \(R_{\mathcal{U}}(e) = G.\)

\[\square\]

4. Applications to quantum control

The purpose of this section is to instantiate the results obtained previously to the specific situation of the quantum control. We will give two results, one for the density matrix formalism and the second for the wave function.

4.1. Density matrix

To consider the specific situation of the density matrix formalism, we use the results of the Section 3 for the Lie group \(U(N)\). We obtain a first
Theorem 4.1. Consider the system
\[
\begin{aligned}
i\frac{\partial}{\partial t}\rho(t;\epsilon;\rho_0) &= [H_0 + F_1(\epsilon(t))\mu_1 + \ldots + F_L(\epsilon(t))\mu_L, \rho(t;\epsilon;\rho_0)] \\
r(t = 0;\epsilon;\rho_0) &= \rho_0
\end{aligned}
\] (4.1)
and suppose that the family \(\{1, F_1, \ldots, F_L\}\) is linearly independent.

Then, when at least one matrix \(H_0, \mu_1, \ldots, \mu_L\) has nonzero trace, the equation (4.1) is density matrix controllable if and only if the Lie algebra \(L_{iH_0, i\mu_1, \ldots, i\mu_L}\) spanned by the matrices \(iH_0, i\mu_1, \ldots, i\mu_L\) is the Lie algebra \(u(N)\) of all skew-hermitian matrices or equivalently \(\dim_{R}L_{iH_0, i\mu_1, \ldots, i\mu_L} = N^2\).

Otherwise, when all matrices \(H_0, \mu_1, \ldots, \mu_L\) have zero trace, a necessary and sufficient condition for controllability is that \(L_{iH_0, i\mu_1, \ldots, i\mu_L} = su(N)\) or equivalently \(\dim_{R}L_{iH_0, i\mu_1, \ldots, i\mu_L} = N^2 - 1\).

Proof. The first part of the conclusion follows from Theorem 3.2 for the Lie group \(G = U(N)\).

When all matrices have zero trace one uses the same result for \(G = SU(N)\) noting that if two matrices \(\rho_1\) and \(\rho_2\) are unitarily equivalent \(\rho_2 = M\rho_1M^\dagger\) then there exists \(\gamma \in R\) with \(M_{su} = Me^\gamma \in SU(N)\) and \(\rho_2 = M_{su}\rho_1M_{su}^\dagger\). \(\square\)

An algorithmic verification of the above theorem can be devised as follows.

1. Test whether the functions \(\{1, F_1, \ldots, F_L\}\) are linearly independent. If the answer is yes go to next step, otherwise keep only a subset \(F_1, \ldots, F_p\) with \(\{1, F_1, \ldots, F_p\}\) linearly independent and modify the \(B_1, \ldots, B_L\) accordingly. For notational convenience we suppose all functionals are independent (\(p = L\)).

2. Construct the traceless matrices \(\tilde{H}_0 = H_0 - \frac{Tr(H_0)}{N}Id, \tilde{\mu}_1 = \mu_1 - \frac{Tr(\mu_1)}{N}Id, \ldots, \tilde{\mu}_L = \mu_1 - \frac{Tr(\mu_1)}{N}Id\). Denote by \(\mathcal{O} = \{i\tilde{H}_0, i\tilde{\mu}_1, \ldots, i\tilde{\mu}_L\}\).

3. Write any element of \(\mathcal{O}\) as a column vector and compute the rank \(r = \text{rank}(\mathcal{O})\) over the real numbers.

4. Construct all commutators \(\mathcal{C}\) of matrices in \(\mathcal{O}\) and test whether \(\text{rank}(\mathcal{O} \cup \mathcal{C}) = r\). If not, set \(\mathcal{O} := \mathcal{O} \cup \mathcal{C}\) and return to previous step.

5. Test whether \(r = N^2 - 1\). If yes the system is controllable, if not the controllability does not hold.

Even more precise results can be derived for the situation in Eqn 2.15.

Theorem 4.2. Consider the development of the interaction Hamiltonian \(H = H_0 + \epsilon(t)\mu_1 + \ldots + \epsilon^L(t)\mu_L\) resulting in the following evolution equation
\[
\begin{aligned}
i\frac{\partial}{\partial t}\rho(t;\epsilon;\rho_0) &= [H_0 + \epsilon(t)\mu_1 + \ldots + \epsilon^L(t)\mu_L, \rho(t;\epsilon;\rho_0)] \\
r(t = 0;\epsilon;\rho_0) &= \rho_0
\end{aligned}
\] (4.2)
\[3\] Some optimizations are possible at this point as only new commutators are generally needed to be computed. We do not enter into details here.
Then, when at least one matrix \( H_0, \mu_1, \ldots, \mu_L \) has nonzero trace, the equation (4.2) is density matrix controllable if and only if the Lie algebra \( L_{iH_0, i\mu_1, \ldots, i\mu_L} \) spanned by the matrices \( iH_0, i\mu_1, \ldots, i\mu_L \) is the Lie algebra \( u(N) \) of all skew-hermitian matrices or equivalently \( \dim_\mathbb{R} L_{iH_0, i\mu_1, \ldots, i\mu_L} = N^2 \).

Otherwise, when all matrices \( H_0, \mu_1, \ldots, \mu_L \) have zero trace, a necessary and sufficient condition for controllability is that \( L_{iH_0, i\mu_1, \ldots, i\mu_L} = su(N) \) or equivalently \( \dim_\mathbb{R} L_{iH_0, i\mu_1, \ldots, i\mu_L} = N^2 - 1 \).

4.2. Wave function

To derive results for the wave function of the same nature as the two criterions above one has to analyse the transitive subsets of \( U(N) \). We recall that a subset \( A \subset U(N) \) is called transitive when for any two vectors \( a, b \) on the unit sphere of \( \mathbb{C}^N \) there exists a matrix \( X \in A \) with \( b = Xa \). For the situation of quantum control, such a study is available in the literature [1]. To be able to state the corresponding result for this specific situation here, we introduce the centralizer \( C_Gz \) of an element \( z \in G \) which is defined as the set of all elements that commute with \( z \):

\[
C_Gz = \{ x \in G : xz = zx \}.
\]

We also define \( P = i \cdot \text{diag}(1, 0, \ldots, 0) \in U(N) \).

**Theorem 4.3.** Consider the system

\[
\frac{dc(t; \epsilon; c_0)}{dt} = [H_0 + (1(t)) \mu_1 + ... + F_L(\epsilon(t)) \mu_L] c(t; \epsilon; c_0)
\]

(4.3)

with \( \|c_0\| = 1 \). Suppose that the family \( \{1, F_1, \ldots, F_L\} \) is linearly independent and denote by \( L_{iH_0, i\mu_1, \ldots, i\mu_L} \) the Lie algebra spanned by the matrices \( iH_0, i\mu_1, \ldots, i\mu_L \).

Then the equation (4.3) is (wave function) controllable if and only if

\[
\dim_\mathbb{R} L_{iH_0, i\mu_1, \ldots, i\mu_L} - \dim(L_{iH_0, i\mu_1, \ldots, i\mu_L} \cap C_GP) = 2N - 2.
\]

In particular a sufficient condition for controllability is that

\[
\dim_\mathbb{R} L_{iH_0, i\mu_1, \ldots, i\mu_L} = N^2.
\]

**Proof.** The proof follows from arguments in [1]. \( \square \)

The following procedure allows to implement the above criteria:

1. Test whether the functions \( \{1, F_1, \ldots, F_L\} \) are linearly independent. If the answer is yes go to next step, otherwise keep only a subset \( F_{i_1}, \ldots, F_{i_p} \) with \( \{1, F_{i_1}, \ldots, F_{i_p}\} \) linearly independent and modify the \( B_1 \ldots B_L \) accordingly. For notational convenience we suppose all functionals are independent i.e. \( p = L \).
2. Denote \( \mathcal{O} = \{iH_0, i\mu_1, \ldots, i\mu_L\} \).
3. Write any element of \( \mathcal{O} \) as a column vector and compute the rank \( r = \text{rank}(\mathcal{O}) \) over the real numbers.
4. Construct all commutators $C$ of matrices in $O^4$ and test whether $\text{rank}(O \cup C) = r$. If not, set $O := O \cup C$ and return to previous step.

5. Extract from $O$ the matrices that commute with $P$ and compute the rank $d$ of this ensemble over $\mathbb{R}$. Test whether $r - d = 2N - 2$. If yes the system is controllable, if not the controllability does not hold.

We also obtain

**Theorem 4.4.** Consider the development of the interaction Hamiltonian $H = H_0 + \epsilon(t)\mu_1 + \ldots + \epsilon^L(t)\mu_L$ resulting in the following evolution equation

$$i\frac{dc(t; \epsilon; c_0)}{dt} = [H_0 + \epsilon(t)\mu_1 + \ldots + \epsilon^L(t)\mu_L] c(t; \epsilon; c_0)$$

(4.4)

$c(t = 0; \epsilon; c_0) = c_0$.

with $\|c_0\| = 1$. Denote by $L_{iH_0,i\mu_1,\ldots,i\mu_L}$ the Lie algebra spanned by the matrices $iH_0,i\mu_1,\ldots,i\mu_L$. Then the equation (4.4) is (wave function) controllable if and only if

$$\dim_{\mathbb{R}}L_{iH_0,i\mu_1,\ldots,i\mu_L} - \dim(L_{iH_0,i\mu_1,\ldots,i\mu_L} \cap C_P) = 2N - 2.$$

In particular a sufficient condition for controllability is that

$$\dim_{\mathbb{R}}L_{iH_0,i\mu_1,\ldots,i\mu_L} = N^2.$$

**Remark 4.5.** All the above results basically state that controllability with linearly independent functionals of a single control $\epsilon$ is true whenever the same equation, but with completely independent controls, is controllable.

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4Here again, optimizations are possible.
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