Outline

1. Introduction
   - Closed quantum system
   - Open quantum system

2. Control of open quantum systems

3. Control of nuclear excitations

4. Conclusions and work in progress
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1. Introduction
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Introduction

Control of quantum systems using lasers
- affects the structure of the molecules (create or break chemical bonds)
- obtain a finesse far beyond the usual macroscopic means (temperature, pressure...)

successful experiments from the 90s (G. Gerber)

Others applications of this technique
- designing logical gates in future quantum computers
- investigations of imaging by nuclear magnetic resonance - NMR
- study of protein dynamics
- molecular detection
- molecular orientation and alignment
- construction of ultra-short laser

**Figure**: control by laser
Introduction

**Figure** : Better than our photovoltaic cells (8% – 20%), plants know how to transform more than 95% of solar energy into chemical energy. A recent study (Nature 446, 782-786 2007) shows the importance of quantum effects. Source de l’image : http://www.wikipedia.org.
Introduction

**Figure:** Selective product formation using optimally-tailored strong field laser pulses
Contrôle direct et indirect des systèmes quantiques ouverts

Introduction

Figure: Examples of final states obtained by laser control
Introduction

**FIGURE:** Photograph of the H. Rabitz group laboratory at Princeton University. The apparatus scattering blue light in the foreground is a pulse shaper designed to shape pulses in the UV spectral range.
Introduction

Closed quantum system

Open quantum system

Control of open quantum systems

Control of nuclear excitations

Conclusions and work in progress
Introduction

Closed quantum system

- evolution modeled by time-dependent Schrödinger equation (TDS)

\[ i \frac{\partial}{\partial t} \Psi(t) = H(t)\Psi(t) \]

where

- \( H(t) \) is the Hamiltonian of the system Ex. : \( H_0 = -\Delta + V(x) \), unbounded domain
- \( \Psi(t) \) is the state vector

evolution on the unit sphere : \( \| \Psi(t) \|_{L^2} = 1, \forall t \geq 0 \)

solution represented in terms of unitary time evolution operator \( U(t, t_0) \)

\[
\begin{align*}
\Psi(t) &= U(t, t_0)\Psi(t_0) \\
U^\dagger(t, t_0)U(t, t_0) &= U(t, t_0)U^\dagger(t, t_0) = I.
\end{align*}
\]
Introduction

Closed quantum system

- The system is in a mixed state; it is characterized by the density matrix operator

\[ \rho(t) = \sum_k p_k |\Psi_k(t)\rangle\langle \Psi_k(t)|; \quad \sum_k p_k = 1 \]

with the properties:

- \( \rho \geq 0, \rho^\dagger = \rho, \quad tr(\rho) = 1 \)

- Equation of motion for the density matrix von Neumann or Liouville von Neumann equation

\[ i \frac{\partial}{\partial t} \rho(t) = [H(t), \rho(t)] \]

\[ \rho(t) = U(t, t_0) \rho(t_0) U^\dagger(t, t_0) \]

We denote: \([A, B] = AB - BA\)
Control quantum system

- add external interaction: $\epsilon(t)$

$$i \frac{\partial}{\partial t} \Psi(x, t) = (H_0 + \epsilon(t) H_1(x)) \Psi(x, t)$$

- $H_1$ is the dipole operator

- prove controllability
  - infinite dimensional setting Ball and al, Turinici, K. Beauchard, U. Boscain, T. Chambrion, J-M. Coron, V. Nersesyan
  - finite dimensional setting
    - Lie algebra approaches: Sussmann and Jurdjevic 1972; Lobry 1974; Ramakrishna et al. 1995; Coron 2007
    - Graph Theory Rabitz and Turinici 2001, 2003; Altafini 2002

- algorithms to find the control → optimal control theory
  - Y. Maday and G. Turinici, J. Salomon and G. Turinici, Mirrahimi and Rouchon, K. Beauchard J.M Coron, A. Grigoriu

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Open quantum system

- Quantum system is not isolated, but interacting with an environment (e.g., a molecule in a solvent)
- The total system is closed following a Hamiltonian dynamics
  \[ i \frac{\partial}{\partial t} \rho(t) = [H(t), \rho(t)] \]
- \( H(t) = H_S + H_B + H_{int} \)
- open quantum system: subsystem of the combined total system

\[ S + B \]

**Figure**: Schematic picture of an open system
Open quantum system

- The state of the subsystem $S$ can’t be represented in terms of unitary Hamiltonian evolution.

- The reduced density matrix $\rho_S$ of the open quantum system $S$

\[ \rho_S(t) = tr_B \{ U(t,t_0) \rho(t_0) U^\dagger(t,t_0) \} \]

where $U(t,t_0)$ is the time-evolution operator of the total system.

\[ \frac{\partial}{\partial t} \rho_S(t) = -itr_B [H(t), \rho(t)] \]

- Determine the equations of motion for the reduced density matrix that are used to approximate the above dynamical eq.

**Figure:** Schematic picture of an open system.
Open quantum systems

For weak system-environment coupling the dynamics is described by a Markovian equation

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

with the generator $\mathcal{L}$ in Lindblad form:

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

- The first term represents the unitary part of the system dynamics.
- $H$ is the Hamiltonian of the system
- The second term describes the dissipative dynamics (dissipator)
- $L_i$ are the Lindblad operators, $\gamma_i \geq 0$
- $\gamma_i$ represent coupling to the environment, functioning as the relaxation rates for different decay modes of the open system
- $\rho \geq 0$, $\rho^\dagger = \rho$, $tr(\rho) = \text{const}$
Open quantum systems

For strong system-environment coupling the process can be non-Markovian processes, (the dynamics is governed by significant memory effects).

\[
\frac{d}{dt}\rho(t) = \mathcal{K}(t)\rho(t).
\]

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

- \( \rho = \rho^* \), \( tr(\rho) = cst \)
- If \( \gamma_i(t) \geq 0 \) the generator \( \mathcal{K}(t) \) is in Lindblad form for each fixed \( t \geq 0 \) (time-dependent Markovian although the corresponding dynamical map do not lead to a quantum dynamical semigroup)
- If \( \gamma_i(t) < 0 \) in certain time intervals the master equation in no longer in Lindblad form and the generator is not completely positive.
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Control of open quantum systems

Add external interaction: $\epsilon(t)$ (ex. electromagnetic field)

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

where

- $H_0$ is the Hamiltonian describing the coherent part of the dynamics,
- $H_{int}$ is the effective Hamiltonian describing the unitary part of the system-environment interaction
- $H_1$ is the dipole moment operator describing the interaction between the system and the field $\epsilon(t)$
- $\rho \geq 0$, $\rho^\dagger = \rho$, $tr(\rho) = const$ for every $t > 0$
- $H_0$ and $H_1$ are Hermitian

Controllable system for any pair of states $\rho_i$ and $\rho_f$, both positive semidefinite, Hermitian, $tr(\rho_i) = tr(\rho_f) > 0$, a control $\epsilon(t)$ exists such that $\rho(0) = \rho_i$ and $\rho(T) = \rho_f$ for some $T > 0$. 
Control of open quantum systems

- **Generic open systems may be difficult to control** because the environment can contain irreversible dynamics that fights against control mechanisms.

- **N-dim Lindblad equation** ($\gamma \geq 0$ and constant) the system can be accessible, but **neither small-time controllable nor controllable in finite time** (C. Altafini 2002, 2003).

**Figure**: The reachable sets $\mathcal{R}(\rho_i)$ for a two level system $\mathcal{L}(I) = 0$
Control of open quantum systems

- Optimal control problem (H. Rabitz et A. Pechen 2011) for $\gamma(t) \geq 0$: tailored non-equilibrium environment, characterized by its distribution function $n_k(t)$

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

the coefficients $\gamma_{\omega}(t) \geq 0$ determine the transitions rates between energy levels with transition frequencies $\omega$ ($H_0\psi_i = \epsilon_i\psi_i$, $\omega_{ij} = \epsilon_i - \epsilon_j$)

- Combine indirect control by the environment with direct control by laser A. Grigoriu, H. Rabitz, G. Turinici 2012
  - simultaneous control through both the Hamiltonian and dissipative parts of the system dynamics.
  - Control by laser: transforms pure states into pure states,
  - Control by the environment (i.e., control by $\gamma(t)$) steer the system from a pure or a mixed state into mixed and in some cases pure states.
Control of open quantum systems

The equation for a system that simultaneously interacts with an electromagnetic field \( \epsilon(t) \) and an environment described by a function \( \gamma(t) \):

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

- \( \gamma(t) \) may also take negative values (the property of complete positivity of the density matrix is lost)
- \( \rho = \rho^*, \text{tr}(\rho(t)) = \text{const} \)
- both \( \epsilon(t) \) and \( \gamma(t) \) are arbitrary real numbers;
- the dynamics is non-Markovian
- 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)

The goal is to analysis the controllability
Control of open quantum systems

- Another motivation for this analysis is given by systems that are not controllable in the isolated setting ($\gamma(t) = 0$);

$$\frac{d}{dt}\rho(t) = -i[H_0 + \epsilon(t)H_1, \rho(t)]$$

Address controllability by using specially tailored environments, characterized by $\gamma(t)$, applied through the dissipative part of the dynamics.

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

- The assumption $\gamma(t) \in \mathbb{R}$ does not imply that the propagation of an arbitrary positive matrix at time $t$ necessarily leads to a positive matrix for future times.

- We use the Lie algebraic approach to analyze the controllability.

- An advantage of this technique is that it also provides an explicit controllability criteria, which can be verified using numerical computations.
Controllability of CQS using Lie algebra approach

- The solution of

\[ i \frac{\partial}{\partial t} \Psi(t) = H(t)\Psi(t) \]

can be represented in terms of unitary time evolution operator \( U(t, t_0) \)

- We have the following evolution equation:

\[ i \frac{\partial}{\partial t} U(t) = (H_0 + \epsilon(t)H_1)U(t) \]

\[ U(t_0) = I \]

where

\[ \Psi(t) = U(t, t_0)\Psi(t_0) \]

\[ U^\dagger(t, t_0)U(t, t_0) = U(t, t_0)U^\dagger(t, t_0) = I. \]

- We work on a Lie group, compact and connected, \( G = U(N) \)

\[ U(N) = \{ U \in \mathbb{C}^{N \times N}; U^\dagger U = U^\dagger U = I \} \]

**Definition**

The state \( \Psi \) is reachable starting from an initial state \( \Psi_0 \) if it exists \( 0 < T < \infty \) and \( \epsilon(t) \) such that \( \Psi = U(t, t_0)\Psi_0 \).
Controllability of CQS using Lie algebra approach

Theorem (Jurdjevic and Sussmann ’72, Lobry ’74, Coron ’07)

Consider the system defined on the Lie group $G$ with the associated Lie algebra $L$ containing $-iH_0$ and $-iH_1$. If $L_{-iH_0, -iH_1} = L$, the set of reachable states $\mathcal{R}(\Psi_0)$ is the Lie group $G$.

Definition

The system is wavefunction controllable if for any two states $\Psi_1$ and $\Psi_2$ there exists a final time $T < \infty$ and a control $\epsilon(t)$ such that starting from the initial state $\Psi_1$ at final time $T$ we have $\Psi(T, \Psi_1) = \Psi_2$.

Definition

The system is density matrix controllable if for any two matrices $\rho_1$ and $\rho_2$ there exists a final time $T < \infty$ and a control $\epsilon(t)$ such that the solution $U(T)$ transforms $\rho_1$ into $\rho_2 : \rho_2 = U(T)\rho_1 U^\dagger(T)$.

Theorem (Ramakrishna and al. ’05)

If the Lie algebra $L_{-iH_0, -iH_1}$ has dimension $N^2$ the system is density matrix controllable (wavefunction controllable)
Background on controllability on Lie groups

- Consider a connected but not necessarily compact Lie group $G$ with Lie algebra $L(G)$ and control system:

$$\frac{dX}{dt}(t) = X_0(X(t)) + \sum_{i=1}^{m} \epsilon_i(t)X_i(X(t)),$$

where $X_0$ and $X_i$ are right-invariant vector fields on $G$.

- Consider the set of all reachable states from $Y$ at time $t$:

$$\mathcal{R}^t(Y) = \{X(t; \epsilon; Y) \mid X(0; \epsilon; Y) = Y\}.$$

- We have:

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

where we denote by $e$ the identity of the Lie group $G$.

- Describing the set $\mathcal{R}^t(e)Y$ allows for completely describing all the other reachable sets.
Background on controllability on Lie groups

- Take the admissible controls $\epsilon_i(t)$ to be the set of all locally bounded and measurable functions.
- Consider $\mathbb{L}$ to be the Lie algebra generated by $X_0, X_1, \ldots, X_m$
- $S$ its corresponding Lie group (Lie subgroup of $G$)
- **We do not assume** that $S$ is compact

The results proved below build on a reformulation of a result by Jurdjevic and Sussmann 1972

**Jurdjevic and Sussmann 1972**

If there exists a constant control $\epsilon = (\epsilon_1, \ldots, \epsilon_m)$ and a sequence of positive numbers $\{t_n\}$ with $t_n \geq \delta > 0$, for some $\delta$, with the property that $\lim_{n \to \infty} X(t_n, \epsilon, e)$ exists and belongs to $\bar{S}$ (the closure is relative to $S$) then $\mathcal{R}(\epsilon) = \bar{S}$. 
We study the controllability of the $N$-dimensional equation

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

and $\mathcal{K}$ the generator

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

- all $\gamma_i$ are null except $\gamma_1$.
- Hamiltonian $H_{int}$ is assumed to commute with $H_0$ we redefine $H_0$ to be $H_0 + H_{int}$

The controllability analysis must deal with:

- the **loss of compactness** that arises from the non-Hermitian nature of the generator $\mathcal{K}$
- identifying the structure of the reachable states and the Lie group remains a task to be investigated.
Situation I: Hermitian operator $A$

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

\[
\begin{align*}
\mathcal{H}_0 : & \quad \rho \longrightarrow -i[H_0, \rho] \\
\mathcal{H}_1 : & \quad \rho \longrightarrow -i[H_1, \rho] \\
\mathcal{T} : & \quad \rho \longrightarrow A\rho A^* - \frac{1}{2}\rho A^* A - \frac{1}{2}A^* A\rho,
\end{align*}
\]

and rewrite equation as:

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

We introduce the sets of matrices:

\[
\Gamma_N = \{ Z \in \mathbb{C}^{N \times N} | Z = Z^* \}, \quad \Gamma_0^N = \{ Z \in \mathbb{C}^{N \times N} | Z = Z^*, \; \text{tr}(Z) = 0 \},
\]

- $\mathcal{H}_0, \mathcal{H}_1, \mathcal{T} \in \text{Lin}(\Gamma_N, \Gamma_N)$,
- $\mathcal{H}_0, \mathcal{H}_1, \mathcal{T} \in \text{Lin}(\Gamma_0^N, \Gamma_0^N)$. 
Situation I : Hermitian operator $A$

- consider $G_1$ the Lie group of one-to-one linear transformations of $\Gamma^0_N$ that contains the identity operator.
- The canonical notation of this group is $GL^+(\Gamma^0_N)$.
- $GL^+(\Gamma^0_N)$ is isomorphic to $GL^+(N^2 - 1)$, car $\Gamma^0_N$ is isomorphic to $\mathbb{R}^{N^2-1}$
- $GL^+(N^2 - 1)$ is the Lie group of invertible matrices of dimension $(N^2 - 1) \times (N^2 - 1)$ with positive determinant. In particular
- the dimension of $G_1$ is $(N^2 - 1)^2$.
- the group $G_1$ is connected but not compact.
- denote by $\text{Lie}(G_1)$ the Lie algebra of $G_1$ which is $\text{Lin}(\Gamma^0_N, \Gamma^0_N)$.

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.
We associate to the evolution equation the following evolution equation on the group $G_1$:

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

- $\mathcal{H}_0^{G_1}, \mathcal{H}_1^{G_1}, \mathcal{T}^{G_1}$ elements of $\text{Lie}(G_1)$ constructed from $\mathcal{H}_0, \mathcal{H}_1$ and $\mathcal{T}$.

- by definition $\rho(t) = X(t; \epsilon, \gamma; e)\rho(0)$ (here $e$ is the identity of the Lie group $G_1$).

**Theorem (A.G, H. Rabitz, G.Turinici ’13)**

If the Lie algebra $\text{Lie}\{\mathcal{H}_0^{G_1}, \mathcal{H}_1^{G_1}, \mathcal{T}^{G_1}\} \subset \text{Lie}(G_1)$ 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

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

is density matrix controllable.
In conclusion $X$ can reach any one-to-one transformation from $\Gamma^0_N$ to itself:
For any $\rho_i$ and $\rho_f$ with

\[
\begin{align*}
\text{tr}(\rho_i) &= \text{tr}(\rho_i) \\
\rho_i - \frac{\text{tr}(\rho_i)}{N} I &\neq 0 \quad \text{and} \quad \rho_f - \frac{\text{tr}(\rho_f)}{N} I &\neq 0
\end{align*}
\]

we can find a transformation to map the non-null vector $\rho_i - \frac{\text{tr}(\rho_i)}{N} I \in \Gamma^0_N$ to the non-null vector $\rho_f - \frac{\text{tr}(\rho_f)}{N} I = \rho_f - \frac{\text{tr}(\rho_i)}{N} I \in \Gamma^0_N$ i.e., we have controllability for $\bar{\rho}$ thus for $\rho$.

**Lemma (A.G, H. Rabitz, G.Turinici ’13)**

The set of all density matrices reachable from $\rho_i$ (with $\rho_i \neq \frac{\text{tr}(\rho_i)}{N} I$ ) for $A$ being a Hermitian operator, is

\[
(\rho_i + \Gamma^0_N) \setminus \left\{ \frac{\text{tr}(\rho_i)}{N} I \right\} = \left\{ \rho_i + Z \mid Z \in \Gamma^0_N, Z \neq \frac{\text{tr}(\rho_i)}{N} I - \rho_i \right\}.
\]
Arbitrary operator $A$

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

$$G_2 := \{ X \in GL^+(\Gamma_N) \mid tr(X(Z)) = tr(Z), \forall Z \in \Gamma_N \}.$$  

isomorphic with

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

We know that $G_2$ is connected but not compact.

We will denote by $\text{Lie}(G_2)$ the Lie algebra of $G_2$ isomorphic with the set (endowed with its canonical Lie algebra structure):

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

$$\frac{d}{dt} \rho = M(\rho(t))$$

The dimension over $\mathbb{R}$ of the Lie algebra is $(N^2 - 1)N^2.$
We associate to the evolution equation the following evolution equation on the group $G_2$:

$$\frac{d}{dt} X(t) = \left( \mathcal{H}^{G_2}_0 + \epsilon(t) \mathcal{H}^{G_2}_1 + \gamma(t) T^{G_2} \right) X(t),$$

$$X(t = 0) = X_0$$

- $\mathcal{H}^{G_2}_0$ the element of $\text{Lie}(G_2)$ that is constructed from $\mathcal{H}_0$ (and the same for $\mathcal{H}_1$ and $T$)
- Then by definition $\rho(t) = X(t; \epsilon, \gamma; e)\rho(0)$.

**Theorem (A.G, H. Rabitz, G.Turinici ’13)**

If the Lie algebra $\text{Lie}\{\mathcal{H}^{G_2}_0, \mathcal{H}^{G_2}_1, T^{G_2}\}$ generated by $\{\mathcal{H}^{G_2}_0, \mathcal{H}^{G_2}_1, T^{G_2}\}$ has dimension $(N^2 - 1)N^2$ (as a vector space over the real numbers) then the system is density matrix controllable.
Lemma (A.G, H. Rabitz, G.Turinici ’13)

The set of all density matrices reachable from $\rho_i$ for an arbitrary operator, is the set

$$\rho_i + \Gamma_0^N = \{ \rho_i + Z | Z \in \Gamma_0^N \}.$$ 

- This set contains matrices that are not necessarily positive semidefinite.
- The control, even between two states that are positive semidefinite may also use intermediary states $\rho(t)$ which are not positive semidefinite.

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.

Work in progress and perspectives:

- Numerical algorithm that penalizes, e.g. the presence of non-physical states
- The controllability analysis remains for now a conjecture if $\gamma(t)$ takes only positive values
- Controllability in infinite dimensional setting
In the following we illustrate the above theoretical results. We consider two finite-dimensional systems defined by

\[
H_0 = \begin{pmatrix}
-4 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 3
\end{pmatrix}, 
H_1 = \begin{pmatrix}
0 & -2 & 0 \\
-2 & 0 & 0 \\
0 & 0 & 0
\end{pmatrix}, 
A = \begin{pmatrix}
2 & 1 & 3 \\
1 & -1 & 0 \\
3 & 0 & -1
\end{pmatrix},
\]

and

\[
H_0 = \begin{pmatrix}
-4 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 3
\end{pmatrix}, 
H_1 = \begin{pmatrix}
0 & -2 & 0 \\
-2 & 0 & 0 \\
0 & 0 & 0
\end{pmatrix}, 
A = \begin{pmatrix}
2 & 1 & 1 \\
1 & -1 & 0 \\
3 & 0 & -1
\end{pmatrix}.
\]

- The system defined by \(H_0\) and \(H_1\) alone (i.e., with \(A = 0\)) is not controllable. The dimension of the Lie algebra generated by \(-iH_0\) and \(-iH_1\) is 4, short of \(3^2 - 1 = 8\) needed for controllability.

- Are systems defined by \(H_0, H_1\) are \(A\) controllable?
To do so we choose a parameterization such that we can write as a linear system

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

Numerically $\tilde{\mathcal{H}}_0, \tilde{\mathcal{H}}_1, \tilde{T}$ 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.

We need to numerically compute the dimension of the Lie algebra (as subalgebra of $N^2 \times N^2$ matrices) generated by $\{i\tilde{\mathcal{H}}_0, i\tilde{\mathcal{H}}_1, \tilde{T}\}$, verify:

- $A$ a Hermitian: $\dim_{\mathbb{R}}(\text{Lie}\{i\tilde{\mathcal{H}}_0, i\tilde{\mathcal{H}}_1, \tilde{T}\}) = (N^2 - 1)^2$
- $A$ is an arbitrary: $\dim_{\mathbb{R}}(\text{Lie}\{i\tilde{\mathcal{H}}_0, i\tilde{\mathcal{H}}_1, \tilde{T}\}) = (N^2 - 1)N^2$.

Since

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

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

we conclude that both systems are controllable.
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Direct control of nuclear excitation

Is it possible to directly control nuclear excitations using a laser?

Since:

- Nucleus can be effectively described by a series of quantized energy levels filled with neutrons and protons.
- Nuclei possess a series of unoccupied, energetically exited states above the stable ground into which the nucleus may be promoted through their interaction with the electromagnetic radiation.

**Figure:** Nine lowest excited eigenstates of $^{153}_{63}$Eu (Europium)
Modeling

- Validate the laser-nuclear interaction within the dipole approximation

\[ i\hbar \frac{d}{dt} |\psi(t)\rangle = (H_0 - \mu_1 \epsilon_1(t) - \mu_2 \epsilon_2(t)) |\psi(t)\rangle \]

- $H_0$ the internal Hamiltonian (the nucleus is a $N$ dimensional system)
- $\mu_1$ et $\mu_2$ the electric and magnetic dipole operator corresponding to the electric $\epsilon_1(t)$ and magnetic $\epsilon_2(t)$ control

→ State controllability of nuclear quantum systems

There exist controls $\epsilon_1(t)$ et $\epsilon_2(t)$ that are capable of steering the system from an initial state $|\psi(0)\rangle = |\psi_i\rangle$ towards a final state $|\psi(T)\rangle = |\psi_f\rangle$ at a final time $T$.

→ Analysis of stability and robustness upon seeking optimal control fields

→ Problem

- Lack of coherent photon sources matching typical nuclear transition energies and the enormous laser intensities required
- transition energies are generally on the order of $10^3 - 10^5 \text{eV}$
- currently photon energies of at most $\hbar \omega = 10^4 \text{eV}$
Indirect control methods of nuclear excitation

use of Bremsstrahlung radiation produced by laser-generated plasma
- Very high maximum photon energy
- Allows the full range of nuclear excitations (photodissociation and fission)

**Figure:** Generating Bremsstrahlung radiation

**Objective:**
- feasibility of indirectly driven nuclear dynamics

*I. Wong, A. Grigoriu, H. Rabitz 2013 work in progress*
- the evolution equation
- optimal control problem
- Analyse the controllability of such systems.
The evolution equation

\[
\frac{d}{dt}\rho_S(t) = -\frac{i}{\hbar}[H_S, \rho_S(t)] + \mathcal{D}_{E1}(\rho_S(t)) + \mathcal{D}_{M1}(\rho_S(t)),
\]

with the electric and magnetic dipole interaction dissipators having a conveniently symmetrical form:

\[
\mathcal{D}_{E1}(\rho_S) = \sum_{\omega > 0} \gamma^+ (\omega) \left( 2\tilde{A}(\omega)\rho_S\tilde{A}^\dagger (\omega) - \tilde{A}^\dagger (\omega)\tilde{A}(\omega)\rho_S - \rho_S\tilde{A}^\dagger (\omega)\tilde{A}(\omega) \right) \\
+ \sum_{\omega > 0} \gamma^- (\omega) \left( 2\tilde{A}^\dagger (\omega)\rho_S\tilde{A}(\omega) - \tilde{A}(\omega)\tilde{A}^\dagger (\omega)\rho_S - \rho_S\tilde{A}(\omega)\tilde{A}^\dagger (\omega) \right),
\]

\[
\mathcal{D}_{M1}(\rho_S) = \sum_{\omega > 0} \xi^+ (\omega) \left( 2\tilde{C}(\omega)\rho_S\tilde{C}^\dagger (\omega) - \tilde{C}^\dagger (\omega)\tilde{C}(\omega)\rho_S - \rho_S\tilde{C}^\dagger (\omega)\tilde{C}(\omega) \right) \\
+ \sum_{\omega > 0} \xi^- (\omega) \left( 2\tilde{C}^\dagger (\omega)\rho_S\tilde{C}(\omega) - \tilde{C}(\omega)\tilde{C}^\dagger (\omega)\rho_S - \rho_S\tilde{C}(\omega)\tilde{C}^\dagger (\omega) \right).
\]

The sum is extended over all energy eigenvalues \( \varepsilon' \) and \( \varepsilon \) of \( H_S \) with a fixed (transition) energy difference \( \hbar \omega \).
Laser control of nuclear excitations

The operators \( \vec{A}(\omega) \) and \( \vec{C}(\omega) \) are projections of the electric dipole operator \( \vec{D} \) and the magnetic dipole operator \( \vec{M} \) onto the Hilbert space \( \mathcal{H}_S \) of the nucleus:

\[
\vec{A}(\omega) \equiv \sum_{\epsilon' - \epsilon = \hbar \omega} \Pi(\epsilon) \vec{D} \Pi(\epsilon'),
\]
\[
\vec{C}(\omega) \equiv \sum_{\epsilon' - \epsilon = \hbar \omega} \Pi(\epsilon) \vec{M} \Pi(\epsilon').
\]

and we have defined

\[
\gamma^{\pm}(\omega) = \frac{\omega^3}{6\epsilon_0 \pi \hbar c^3} \left[ N(\omega, t) + \frac{(1 \pm 1)}{2} \right], \quad \xi^{\pm}(\omega) = \frac{1}{c^2} \gamma^{\pm}(\omega).
\]

- \( \Pi(\epsilon) \) represents the projection operator onto the eigenspace of the eigenvalue \( \epsilon \).
- \( N(\omega_k, t) \) represents the average number of photons in a mode with frequency \( \omega_k \) at time \( t \).
Laser control of nuclear excitations

The optimal control problem

- Use a genetic algorithm that searches
  - through the space of relevant parameters $N(\omega_n, t)$ and $T$ (final target time)
  - the optimal values which allow for the nucleus to evolve from its initial state $\rho_i$ at $t = 0$, typically set to be the ground state, to a predefined target state $\rho_T$ at a target time $t = T$

- This fitness function is defined as

$$F = \sum_{n=1}^{k} (\rho_{nn,k} - \rho_{nn,T})^2,$$

where $\rho_{nn}$ represent the diagonal elements of the density matrix.

- Example:

$$\rho_i = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix}, \quad \rho_T = \begin{pmatrix} 0.6 & 0 & 0 & 0 \\ 0 & 0.2 & 0 & 0 \\ 0 & 0 & 0.1 & 0 \\ 0 & 0 & 0 & 0.1 \end{pmatrix}.$$
Laser control of nuclear excitations

The four lowest eigenstates of $^{181}$Ta (Tantale)
Data was taken off the ENSDF online database
The nuclear Hamiltonian is (energies in units of keV):

$$H_0 = \begin{pmatrix}
0 & 0 & 0 & 0 \\
0 & 6.2 & 0 & 0 \\
0 & 0 & 136.3 & 0 \\
0 & 0 & 0 & 158.5
\end{pmatrix}.$$

The corresponding electric and magnetic dipole matrices (electric dipole moment in units of $10^{-37} \times C \cdot m$; magnetic dipole moment in units of $10^{-27} \times J/T$)

$$\vec{D} = \begin{pmatrix}
0 & 6.82 & 0 & 0 \\
6.82 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0
\end{pmatrix}, \quad \vec{M} = \begin{pmatrix}
0 & 0 & 4.98 & 0 \\
0 & 0 & 0 & 6.52 \\
4.98 & 0 & 0 & 0 \\
0 & 6.52 & 0 & 0
\end{pmatrix}.$$
Laser control of nuclear excitations

**Figure**: Left: Convergence towards the target $\rho_T$; Right Convergence of the fitness function $F$
Outline

1. Introduction
   - Closed quantum system
   - Open quantum system

2. Control of open quantum systems

3. Control of nuclear excitations

4. Conclusions and work in progress
Conclusions and work in progress

**Conclusions**
- Controllability analysis for a non-Markovian equation

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

A.G., H. Rabitz, G. Turinici
*Controllability analysis of quantum systems immersed within an engineered environment*, accepted J. Math. Chem 2013

- Direct control of nuclear excitations

- Indirect control of nuclear excitations
  A.G. I. Wong H. Rabitz
  *Incoherent Control of Nuclear Excitations* to be submitted

**Perspectives**
- Numerical algorithm that penalizes, e.g. the presence of non-physical states
- The controllability analysis remains for now a conjecture if \( \gamma(t) \) takes only positive values
- Controllability in infinite dimensional setting
Contrôle direct et indirect des systèmes quantiques ouverts

Conclusions and work in progress