Control of bilinear systems: multiple systems and perturbations

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Outline

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   • Optical and magnetic manipulation of quantum dynamics

2 Modelization
   • Single quantum system: wavefunction formulation
   • Several quantum systems: density matrix formulation
   • Simultaneous control of quantum systems
   • Evolution semigroup
   • Observables

3 Controllability
   • Background on controllability criteria
   • Simultaneous controllability of quantum systems
   • Controllability of a set of identical molecules
   • Controllability in presence of known perturbations
Motivation

Mathematical questions

Simultaneous controllability

Isolated system: \( \frac{d}{dt} x(t) = f(t, x(t), u(t)) \).

Collection of similar systems

\[
\frac{d}{dt} x_k(t) = f_k(t, x_k(t), u(t)), \quad k = 1, \ldots, K. \tag{1}
\]

Similar may mean \( f_k(t, x, u) = f(t, x, \alpha_k u), \ \alpha_k \in \mathbb{R} \). The systems differ in their interaction with the control e.g. for instance are placed spatially differently with respect to the controlling action.

Can one simultaneously control all systems with same \( u(t) \) ?
Controllability of “large perturbations”

Un-perturbed system: \( \frac{d}{dt} x(t) = f(t, x(t), u(t)) \).

Perturbed system:

\[
\frac{d}{dt} x_k(t) = f(t, x_k(t), u(t) + \delta u_k(t)), \quad k = 1, \ldots, K.
\]

Large perturbations: \( \delta u_k(t), \quad k = 1, \ldots, K \) in a fixed list (alphabet) but there is no knowledge about which \( k \) will actually occur.

Can one still control the system?
Laser SELECTIVE control over quantum dynamics

Figure: Optimized laser pulses can be used to control molecular dynamics. a, An optimized laser pulse excites benzene into the superposition state $S_0 + S_1$ with bidirectional electron motion that results in switching between two discrete Kekulé structures on a subfemtosecond timescale; b: A different optimized laser pulse excites benzene into the superposition state $S_0 + S_2$, which is triply ionic; c: The next frontier in simulating control is to excite both electronic and nuclear dynamics simultaneously. Credits: NATURE CHEMISTRY, VOL 4, FEBRUARY 2012, p 72.
Figure: Studying the excited states of proteins. F. Courvoisier et al., App.Phys.Lett.
Quantum non-demolition measurements

**Figure:** Physics Nobel prize 2012 to Haroche and Wineland "for ground-breaking experimental methods that enable measuring and manipulation of individual quantum systems. Serge Haroche and David Wineland have independently invented and developed methods for measuring and manipulating individual particles while preserving their quantum-mechanical nature, in ways that were previously thought unattainable”; [these are ] ”the first tiny steps towards building a quantum computer”.

Picture credits: Nature 492, 55 (06 December 2012)
Other applications

- EMERGENT technology
- NMR: spin interacting with magnetic fields; control by magnetic fields
- creation of particular molecular states
- fast “switch” in semiconductors
- ...

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Single quantum system

Time dependent Schrödinger equation

\[
\begin{aligned}
i \frac{\partial}{\partial t} \Psi(x, t) &= H(t) \Psi(x, t) \\
\Psi(x, t = 0) &= \Psi_0(x).
\end{aligned}
\] (3)

- \( H(t) = H_0 + \) interaction terms E.g. \( H_0 = -\Delta + V(x) \)
- \( H(t)^* = H(t) \) thus \( \|\Psi(t)\|_{L^2} = 1, \forall t \geq 0. \)
- dipole approximation: \( H(t) = H_0 - \epsilon(t) \mu(x) \)
- E.g. \( O - H \) bond, \( H_0 = -\frac{\Delta}{2m} + V \), \( m = \) reduced mass
  \( V(x) = D_0 \left[ e^{-\beta(x-x_0)} - 1 \right]^2 - D_0, \mu(x) = \mu_0 xe^{-x/x^*} \)
- higher order approximation: \( H(t) = H_0 + \sum_k \epsilon^k(t) \mu_k(x) \)
- misc. approximations (rigid rotor interacting with two-color linearly polarized pulse): \( H(t) = H_0 + (E_1(t)^2 + E_2(t)^2)\mu_1 + E_1(t)^2 \cdot E_2(t)\mu_2 \)
Several quantum systems: density matrix formulation

- Evolution equation for a projector

Let \( P_{\psi(t)} = \Psi(t)\Psi(t)^\dagger \) i.e. the projector on \( \Psi(t) \), also noted in bra-ket notation \( |\Psi(t)\rangle \langle \Psi(t)| \); then

\[
i\frac{\partial}{\partial t} P_{\psi(t)} = (i\frac{\partial}{\partial t} \Psi(t))\Psi(t)^\dagger + \Psi(t)(-i\frac{\partial}{\partial t} \Psi(t))^\dagger \tag{4}
\]

\[
(H(t)\Psi(t))\Psi(t)^\dagger + \Psi(t)(-H(t)\Psi(t))^\dagger = \tag{5}
\]

\[
[H(t), \Psi(t)\Psi(t)^\dagger] = [H(t), P_{\psi(t)}]. \tag{6}
\]
Several quantum systems: density matrix formulation

- For a sum of projectors (density matrix): $\rho(t) = \sum_k \eta_k P_{\psi^k(t)}$. By linearity the equation for the density matrix evolution

\[
\begin{cases}
  i \frac{\partial}{\partial t} \rho(x, t) = [H(t), \rho(x, t)] \\
  \rho(x, t = 0) = \rho_0(x).
\end{cases}
\] (7)

Usually $\eta_k \in \mathbb{R}_+$ define a discrete probability law with $\eta_k$ the probability to be at $t = 0$ in state $\Psi^k(0)$. This gives interpretation in terms of observables.
Simultaneous control of quantum systems

Under simultaneous control of a unique laser field \( L \geq 2 \) molecular species.

Initial state \(|\Psi(0)\rangle = \prod_{\ell=1}^{L} |\Psi_{\ell}(0)\rangle\).

Any molecule evolves by its own Schrödinger equation
\[
i\hbar \frac{\partial}{\partial t} |\psi_{\ell}(t)\rangle = \left[ H_{0}^{\ell} - \mu^{\ell} \cdot \epsilon(t) \right] |\psi_{\ell}(t)\rangle
\]
A set of identical molecules with different spatial positions

\( N \geq 2 \) identical molecules, DIFFERENT orientations, simultaneous control by one laser field.

Interaction with a field \( \mu \epsilon(t) \alpha_k \) where \( \alpha_k \) depends on \( R = (r, \theta, \zeta) \) which characterizes the localization of the molecule in the ensemble.
Evolution semigroup

\[
\begin{align*}
\left\{ \begin{array}{l}
i \frac{\partial}{\partial t} U(t) &= H(t) U(t) \\
U(0) &= \text{Id.}
\end{array} \right.
\end{align*}
\]  

(8)

Relationship with
- wavefunction formulation \( \Psi(t) = U(t) \Psi(0) \)
- density matrix version: \( \rho(t) = U(t) \rho(0) U^\dagger(t) \)
Observables: localization

Measurable quantities: \( \langle \Psi(t), O\Psi(t) \rangle \) for self-adjoint operators \( O \) (rq: phase invariance).

For density matrix: \( \rho(t) = \sum_k \eta_k P_{\psi^k(t)} \)

\[
\sum_k \eta_k \langle \psi^k(t), O\psi^k(t) \rangle = Tr(\rho O).
\] (9)

coherent with the probabilistic interpretation.

Important example: \( O = \) (sum of) projection(s) to some (eigen)state.
Observables: localization

E.g. O-H bond: \( O(x) = \frac{\gamma_0}{\sqrt{\pi}} e^{-\gamma_0^2 (x-x')^2} \)
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Single quantum system, bilinear control

Time dependent Schrödinger equation

\[
\begin{cases}
i \frac{\partial}{\partial t} \psi(x, t) = H_0 \psi(x, t) \\
\psi(x, t = 0) = \psi_0(x).
\end{cases}
\] (10)

Add external \textbf{BILINEAR} interaction (e.g. laser)

\[
\begin{cases}
i \frac{\partial}{\partial t} \psi(x, t) = (H_0 - \epsilon(t)\mu(x))\psi(x, t) \\
\psi(x, t = 0) = \psi_0(x)
\end{cases}
\] (11)

Ex.: \(H_0 = -\Delta + V(x)\), unbounded domain

Evolution on the unit sphere: \(\|\psi(t)\|_{L^2} = 1, \ \forall t \geq 0\).
Controllability

A system is **controllable** if for two arbitrary points $\Psi_1$ and $\Psi_2$ on the unit sphere (or other ensemble of admissible states) it can be steered from $\Psi_1$ to $\Psi_2$ with an admissible control.

Norm conservation: controllability is equivalent, up to a phase, to say that the projection to a target is $= 1$. 
Galerkin discretization of the Time Dependent Schrödinger equation

\[
i \frac{\partial}{\partial t} \psi(x, t) = (H_0 - \epsilon(t)\mu)\psi(x, t)
\]

- basis functions \( \{\psi_i; i = 1, \ldots, N\} \), e.g. the eigenfunctions of the \( H_0 \):
  \( H_0 \psi_k = e_k \psi_k \)

- wavefunction written as \( \psi = \sum_{k=1}^{N} c_k \psi_k \)

- We will still denote by \( H_0 \) and \( \mu \) the matrices \( (N \times N) \) associated to the operators \( H_0 \) and \( \mu : H_{0kl} = \langle \psi_k | H_0 | \psi_l \rangle, \mu_{kl} = \langle \psi_k | \mu | \psi_l \rangle, \)
Lie algebra approaches

To assess controllability of

\[ i \frac{\partial}{\partial t} \Psi(x, t) = (H_0 - \epsilon(t)\mu)\Psi(x, t) \]

construct the “dynamic” Lie algebra \( L = \text{Lie}(-iH_0, -i\mu) \):

\[
\forall M_1, M_2 \in L, \forall \alpha, \beta \in \mathbb{R} : \alpha M_1 + \beta M_2 \in L \\
\forall M_1, M_2 \in L, [M_1, M_2] = M_1 M_2 - M_2 M_1 \in L
\]

Theorem If the group \( e^L \) is compact any \( e^M\psi_0, M \in L \) can be attained.

“Proof” \( M = -iAt \) : trivial by free evolution

Trotter formula:

\[
e^{i(AB-BA)} = \lim_{n \to \infty} \left[ e^{-iB/\sqrt{n}} e^{-iA/\sqrt{n}} e^{iB/\sqrt{n}} e^{iA/\sqrt{n}} \right]^n
\]
Operator synthesis ("lateral parking")

Trotter formula: \( e^{i[A,B]} = \lim_{n \to \infty} \left[ e^{-iB/\sqrt{n}} e^{-iA/\sqrt{n}} e^{iB/\sqrt{n}} e^{iA/\sqrt{n}} \right]^n \)

\( e^{\pm iA} = \) advance/reverse ; \( e^{\pm iB} = \) turn left/right
Corollary. If $L = u(N)$ or $L = su(N)$ (the (null-traced) skew-hermitian matrices) then the system is controllable.

"Proof" For any $\Psi_0$, $\Psi_T$ there exists a "rotation" $U$ in $U(N) = e^{u(N)}$ (or in $SU(N) = e^{su(N)}$) such that $\Psi_T = U\Psi_0$.

• (Albertini & D’Alessandro 2001) Controllability also true for $L$ isomorphic to $sp(N/2)$ (unicity).

$sp(N/2) = \{ M : M^* + M = 0, M^t J + JM = 0 \}$ where $J$ is a matrix unitary equivalent to $\begin{pmatrix} 0 & I_{N/2} \\ -I_{N/2} & 0 \end{pmatrix}$ and $I_{N/2}$ is the identity matrix of dimension $N/2$. 
Results by the connectivity graph (G.T. & H. Rabitz)

Let us define the connectivity graph

$$G = (V, E), V = \{\psi_1, \ldots, \psi_N\}; E = \{(\psi_i, \psi_j), i \neq j, B_{ij} \neq 0\}$$

$$A = \begin{pmatrix}
1.0 & 0 & 0 & 0 & 0 & 0 \\
0 & 1.2 & 0 & 0 & 0 & 0 \\
0 & 0 & 1.3 & 0 & 0 & 0 \\
0 & 0 & 0 & 2.0 & 0 & 0 \\
0 & 0 & 0 & 0 & 2.15 & 0
\end{pmatrix},
B = \begin{pmatrix}
0 & 0 & 0 & * & * \\
0 & 0 & 0 & * & * \\
0 & 0 & 0 & * & * \\
* & * & * & 0 & 0 \\
* & * & * & 0 & 0
\end{pmatrix}$$
Look for degenerate transitions: list all $e_{kl} = e_k - e_l$ and look for repetitions.

$$A = \begin{pmatrix}
1.0 & 0 & 0 & 0 & 0 & 0 \\
0 & 1.2 & 0 & 0 & 0 & 0 \\
0 & 0 & 1.3 & 0 & 0 & 0 \\
0 & 0 & 0 & 2.0 & 0 & 0 \\
0 & 0 & 0 & 0 & 2.15 & 0 \\
0 & 0 & 0 & 0 & 0 & 2.15
\end{pmatrix}, \quad B = \begin{pmatrix}
0 & 0 & 0 & * & * & * \\
0 & 0 & 0 & * & * & * \\
0 & 0 & 0 & * & * & * \\
* & * & * & 0 & 0 & 0 \\
* & * & * & 0 & 0 & 0
\end{pmatrix}$$

$e_{41} = 2.0 - 1.0 = 1.0$, $e_{51} = 2.15 - 1.0 = 1.15$

$e_{42} = 2.0 - 1.2 = 0.8$, $e_{52} = 2.15 - 1.2 = 0.95$

$e_{43} = 2.0 - 1.3 = 0.7$, $e_{53} = 2.15 - 1.3 = 0.85$
Theorem (G.T. & H.Rabitz 2000, C.Altafini 2001) If the connectivity graph is connected and if there are no degenerate transitions then the system is controllable.

Note: non connected = independent quantum systems.

\[
A = \begin{pmatrix}
1.0 & 0 & 0 & 0 & 0 & 0 \\
0 & 1.2 & 0 & 0 & 0 & 0 \\
0 & 0 & 1.3 & 0 & 0 & 0 \\
0 & 0 & 0 & 2.0 & 0 & 0 \\
0 & 0 & 0 & 0 & 2.15 & 0 \\
\end{pmatrix}, \quad B = \begin{pmatrix}
0 & 0 & 0 & 1 & 1 \\
0 & 0 & 0 & 1 & 1 \\
0 & 0 & 0 & 1 & 1 \\
1 & 1 & 1 & 0 & 0 \\
1 & 1 & 1 & 0 & 0 \\
\end{pmatrix}
\]
Simultaneous controllability of quantum systems

Under simultaneous control of a unique laser field $L \geq 2$ molecular species.

Initial state $|\Psi(0)\rangle = \prod_{\ell=1}^{L} |\Psi_\ell(0)\rangle$.

Total controllability $=$ can simultaneously and arbitrarily steer any state $|\Psi_\ell(0)\rangle \rightarrow |\Psi_\ell(t)\rangle$, $t \geq 0$ under the influence of only one laser field $\epsilon(t)$.

Any molecule evolves by its own Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} |\Psi_\ell(t)\rangle = [H_0^\ell - \mu^\ell \cdot \epsilon(t)] |\Psi_\ell(t)\rangle$$
Discretization spaces $D^\ell = \{ \psi^\ell_i(x); i = 1, .., N_\ell \}, \ N_\ell, N_\ell \geq 3$ eigenstates of $H^\ell_0$.

$A^\ell$ and $B^\ell = \text{matrices of the operators } H^\ell_0 \text{ and } \mu^\ell \text{ respectively, with respect to } D^\ell; \ N = \sum_{\ell=1}^{L} N_\ell,$

\[
A = \begin{pmatrix}
A^1 & 0 & \ldots & 0 \\
0 & A^2 & \ldots & 0 \\
\vdots & \vdots & \ddots & \vdots \\
0 & 0 & \ldots & A^L \\
\end{pmatrix}, \quad B = \begin{pmatrix}
B^1 & 0 & \ldots & 0 \\
0 & B^2 & \ldots & 0 \\
\vdots & \vdots & \ddots & \vdots \\
0 & 0 & \ldots & B^L \\
\end{pmatrix}.
\]

Note: the system evolves on the product of spheres $S = \prod_{\ell=1}^{L} S_{\mathbb{C}}^{N_\ell - 1}, S_{\mathbb{C}}^{k-1} = \text{complex unit sphere of } \mathbb{C}^k.$
Theorem (B. Li, G. T., V. Ramakhrishna, H. Rabitz. 2002, 2003): If

\[ \dim_{\mathbb{R}} \operatorname{Lie}(-iA, -iB) = 1 + \sum_{\ell=1}^{L} (N_{\ell}^2 - 1), \]

then the system is controllable (the dimension of \( \operatorname{Lie}(-iA, -iB) \) is computed over \( \mathbb{R} \)). Moreover, if the system is controllable, there exists a time \( T > 0 \) such that any target can be reached at or before time \( T \) (and thereafter for all \( t > T \)), i.e. for any \( c_0 \in S \) et \( t \geq T \) the set of reachable states from \( c_0 \) is \( S \).
Controllability of a set of identical molecules

\( N \geq 2 \) identical molecules, placed under simultaneous control of only one laser field. The molecules have DIFFERENT orientations.
Controllability of a set of identical molecules

Linear case:

\[
\begin{align*}
\frac{d}{dt} x_1 &= Ax_1 + Bu(t), \quad x_1(0) = 0 \\
\frac{d}{dt} x_2 &= Ax_2 + 2Bu(t), \quad x_2(0) = 0.
\end{align*}
\]

Then for any control \( u(t) \) we have \( x_2(t) = 2x_1(t) \), thus there are restrictions on the attainable set; **no simultaneous control in the linear case.**
Controllability of a set of identical molecules

Interaction with the control field is $\mu \epsilon(t) \alpha_k$ where $\alpha_k$ depends on the localization $R = (r, \theta, \zeta)$ of the molecule in the ensemble.

Theorem (GT & H. Rabitz, PRA 2004): Suppose $|\alpha_k| \neq |\alpha_j|$, $H_0$ is with non-degenerate transitions and graph of $\mu$ is connected. Then if one (arbitrary) molecule is controllable then the whole (discrete) ensemble is.
Controllability of a set of identical molecules

Interaction with the control field is $\mu \epsilon(t) \alpha_k$ where $\alpha_k$ depends on the localization $R = (r, \theta, \zeta)$ of the molecule in the ensemble.

Theorem (GT & H. Rabitz, PRA 2004): Suppose $H_0$ is with non-degenerate transitions and that the connectivity graph of the system is connected but not bi-partite. Then if a molecule is controllable then the whole (discrete) ensemble is.
Controllability of a set of identical molecules

Figure: The target yield $P(x)$ for different orientations of $x = \cos(\theta)$. The yield $P(x)$ from a field optimized at $x = 1$ produced the quality control index $Q(\epsilon) = 49\%$. The yield $P(x)$ for full ensemble control of was required to optimize a sample of $M = 31$ orientations uniformly distributed over the interval $[-1, 1]$. The resultant quality index is $Q(\epsilon) = 85\%$ (G.T. and H. Rabitz. PRA 2004)
Other results from the literature:

- control of PDE: T. Chambrión, K. Beauchard, P. Rouchon: mostly for $A + \alpha_k u(t)B$; recent works by M. Morancey, V. Nersesyan

- finite-dimensional, mostly for spin systems: C. Altafini, N. Khaneja: $\alpha_k A + u(t)B$
Controllability in presence of known perturbations: joint work with M. Belhadj, J. Salomon, C. Lefter, B. Gavrilei

\[ \frac{d}{dt} x = (A + u(t)B)x. \]  

(15)

What if \( u(t) \) is submitted to a random perturbation in a predefined (discrete) list \( \{\delta u_k, k = 1, ..., K\} \)?

Linear systems:

\[ \frac{d}{dt} x_1 = Ax_1 + Bu(t), \quad x_1(0) = 0. \]  

(16)

\[ \frac{d}{dt} x_2 = Ax_2 + B[u(t) + \alpha], \quad x_2(0) = 0. \]  

(17)

The dynamics of \( x_2(t) - x_1(t) \) is not influenced by the control:

\[ \frac{d}{dt}(x_2 - x_1) = A(x_2 - x_1) + B\alpha, \quad x_2(0) - x_1(0) = 0. \]  

(18)

Thus: no control of perturbations in the linear case.
Controllability in presence of known perturbations: joint work with M. Belhadj, J. Salomon, C. Lefter, B. Gavrilei

Theorem (M. Belhadj, J. Salomon, GT 2013)

Suppose the bi-linear system on $SU(N)$

$$\frac{d}{dt}x = (A + u(t)B)x$$ \hspace{1cm} (19)

is such that the Lie algebra generated by $[A, B]$ and $B$ is the whole Lie algebra $su(N)$. Then for any distinct $\alpha_k \in \mathbb{R}$, $k = 1, \ldots, K$, the collection of systems

$$\frac{d}{dt}x_k = (A + [u(t) + \alpha_k]B)x, \quad k = 1, \ldots, K,$$ \hspace{1cm} (20)

is controllable.
Controllability in presence of known perturbations: joint work with M. Belhadj, J. Salomon, C. Lefter, B. Gavrilei

Theorem (M. Belhadj, J. Salomon, GT 2013)

Consider the collection of control systems on $SU(N)$:

\[
\begin{align*}
\frac{dY_k(t)}{dt} &= \left\{ A + (u(t) + \delta_k u(t))B \right\} Y_k(t), \\
Y_k(0) &= Y_{k,0} \in SU(N).
\end{align*}
\]

Suppose that there exists $0 < t_1 < t_2 < \infty$ such that $\delta_k u(t) = \alpha_k$ (constant) $\forall t \in [t_1, t_2]$. Then there exists $T_{A,B,\alpha_1,\ldots,\alpha_K}$ such that if $t_2 - t_1 \geq T_{A,B,\alpha_1,\ldots,\alpha_K}$ the collection of systems (21) is simultaneously controllable at any time $T \geq t_2$. 
Controllability in presence of known perturbations: joint work with M. Belhadj, J. Salomon, C. Lefter, B. Gavrilei

Different model:

\[
\begin{aligned}
\frac{dZ_k(t)}{dt} &= AZ_k(t) + [u(t) + \alpha_k]\xi(t)BZ_k(t), \\
Z_k(0) &= Z_{k,0} \in SU(N),
\end{aligned}
\]  

(22)

Controls: \(u(t), \xi(t)\) with \(\xi(t) \in \{0, 1\}, \forall t \geq 0\) (\(t \mapsto \xi(t)\) measurable).

**Theorem**

*The system (22) is simultaneously controllable if and only if \(\mathbb{L}_{A,B} = su(N)\).*
Controllability in presence of known perturbations: extensions and perspectives

- slowly varying perturbations, (beyond piecewise constant )

- control of more non-linear situations:
  Rigid rotor interacting with linearly polarized pulse:

\[
i \frac{\partial}{\partial t} \Psi(x, t) = \left[ H_0 + u(t)\mu_1 + u(t)^2\mu_2 + u(t)^3\mu_3 \right] \Psi(x, t).
\] (23)

Rigid rotor interacting with two-color linearly polarized pulse:

\[
i \frac{\partial}{\partial t} \Psi(x, t) = \left[ H_0 + (E_1(t)^2 + E_2(t)^2)\mu_1 + E_1(t)^2 \cdot E_2(t)\mu_2 \right] \Psi(x, t).
\] (24)
Controllability in presence of known perturbations: extensions and perspectives

- Control of rotational motion: time dependent Schrödinger equation $\theta, \phi = \text{polar coordinates}$:

\[ i\hbar \frac{\partial}{\partial t} |\psi(\theta, \phi, t)\rangle = (B\hat{J}^2 - \vec{u}(t) \cdot \vec{d}) |\psi(\theta, \phi, t)\rangle \]

\[ |\psi(0)\rangle = |\psi_0\rangle \]

- Numerics (joint works with M. Belhadj, J. Salomon, C. Lefter, B. Gavriilei): several approaches: monotonic algorithms, Lyapunov approaches, multi-criterion optimization. In general it is difficult to find the field robust to control.