

Introduction to Control, Coherence, and Dissipative Dynamics

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Outline

1 Modelization

- Single quantum system: wavefunction formulation
- Several quantum systems: density matrix formulation
- Simultaneous control of quantum systems
- Evolution semigroup

2 Observables

3 Numerical algorithms

- Simultaneous control of quantum systems: discretization
- Monotonically convergent algorithms
- Experimental control
- Lyapounov (tracking) approaches

4 Inversion and identification approaches

- Identifiability
- Optimal identification : implementable experimental + numerical algorithms

Single quantum system

Time dependent Schrödinger equation

$$\begin{cases} i \frac{\partial}{\partial t} \Psi(x, t) = H(t) \Psi(x, t) \\ \Psi(x, t = 0) = \Psi_0(x). \end{cases} \quad (1)$$

- $H(t) = H_0 +$ 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
 $V(x) = D_0[e^{-\beta(x-x_0)} - 1]^2 - D_0$, $\mu(x) = \mu_0 x e^{-x/x^*}$
- 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

- Motivation (I) : evolution equation for a projector
- Motivation (II) : evolution equation for a sum of projectors
- Equation for the density matrix evolution

$$\begin{cases} i \frac{\partial}{\partial t} \rho(x, t) = [H(t), \rho(x, t)] \\ \rho(x, t = 0) = \rho_0(x). \end{cases} \quad (2)$$

Simultaneous control of quantum systems

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

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

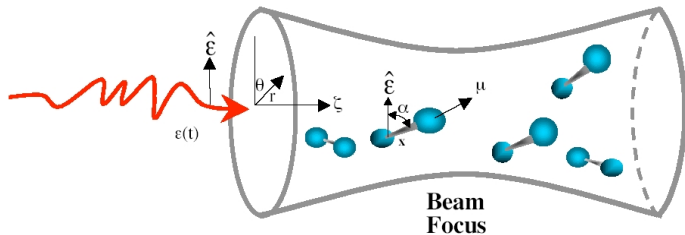
Any molecule evolves by its own Schrödinger equation

$$i\hbar \frac{\partial}{\partial t} |\Psi_{\ell}(t)\rangle = [H_0^{\ell} - \mu^{\ell} \cdot \epsilon(t)] |\Psi_{\ell}(t)\rangle$$

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 $\mu(x)\epsilon(t)u(R)$ where $R = (r, \theta, \zeta)$ characterizes the localization of the molecule in the ensemble.



Evolution semigroup

$$\begin{cases} i \frac{\partial}{\partial t} U(t) = H(t)U(t) \\ U(0) = Id. \end{cases} \quad (3)$$

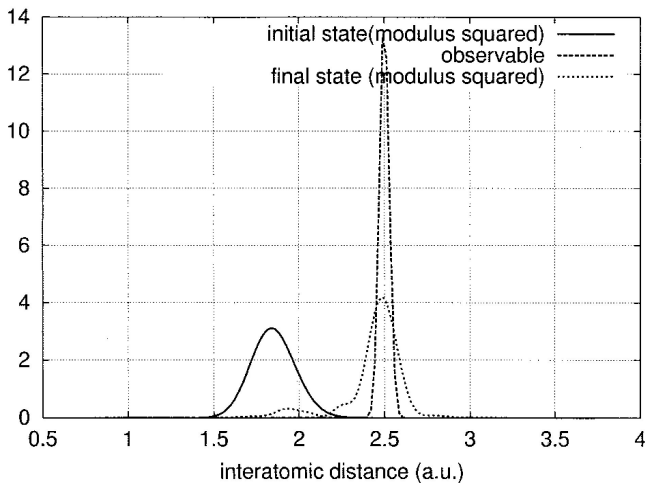
Relationship with wavefunction and density matrix versions: ...

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- 3 Numerical algorithms
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 - Monotonically convergent algorithms
 - Experimental control
 - Lyapounov (tracking) approaches
- 4 Inversion and identification approaches
 - Identifiability
 - Optimal identification : implementable experimental + numerical algorithms

Observables: localization

e.g. O-H bond : $O(x) = \frac{\gamma_0}{\sqrt{\pi}} e^{-\gamma_0^2(x-x')^2}$



Observables: projections on eigenstates

Other important example: projection on eigenstates
density matrix formulation $Tr(\rho O)$

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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 $\{\psi_i; i = 1, \dots, N\}$, e.g. the eigenfunctions of the H_0 : $H_0 \psi_k = e_k \psi_k$
- wavefunction written as $\Psi = \sum_{k=1}^N c_k \psi_k$
- construct the matrices ($N \times N$) associated to the operators H_0 and μ : $A_{kl} = \langle \psi_k | H_0 | \psi_l \rangle$, $B_{kl} = \langle \psi_k | \mu | \psi_l \rangle$,

Galerkin discretization of the Time Dependent Schrödinger equation

- Finite dimensional equations

$$\begin{cases} i\hbar \frac{\partial}{\partial t} c = (A - \epsilon(t)B)c \\ c(t=0) = c_0 \end{cases}$$

The system evolves on the complex unit sphere $\mathcal{S}_{\mathbb{C}}^{N-1}$ of \mathbb{C}^N : $\sum_{i=1}^N |c_i|^2(t) = 1, \forall t \geq 0$.

Simultaneous control of quantum systems: discretization

Discretization spaces $D^\ell = \{\psi_i^\ell(x); i = 1, \dots, N_\ell\}$, $N_\ell, N_\ell \geq 3$
eigenstates of H_0^ℓ

A^ℓ and B^ℓ = matrices of the operators H_0^ℓ and μ^ℓ respectively,
with respect to D^ℓ ; $N = \sum_{\ell=1}^L N_\ell$,

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

Note: the system evolves on the product of spheres

$\mathcal{S} = \prod_{\ell=1}^L \mathcal{S}_{\mathbb{C}}^{N_\ell-1}$, $\mathcal{S}_{\mathbb{C}}^{k-1}$ = complex unit sphere of \mathbb{C}^k .

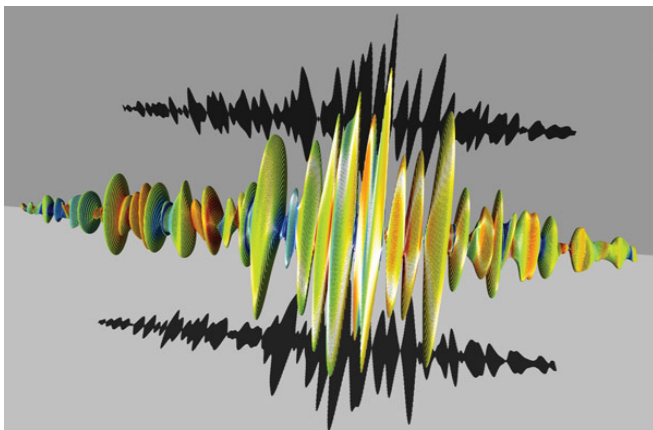


Figure: Polarization-shaped pulse, optimized for the ionization of potassium molecules. Ellipses represent the amplitude of the electric field, colours indicate different frequencies; Yaron Silberberg, Nature 430, 624-625 (2004)

Monotonic algorithms

- evaluation of the quality of a control through a objective functional to maximize

$$J(\epsilon) = 2\Re\langle\psi_{target}|\psi(\cdot, T)\rangle - \int_0^T \alpha(t)\epsilon^2(t)dt$$

$$J(\epsilon) = 2 - \|\psi_{target} - \psi(\cdot, T)\|_{L^2}^2 - \int_0^T \alpha(t)\epsilon^2(t)dt$$

$$J(\epsilon) = \langle\Psi(T)|O|\Psi(T)\rangle - \alpha \int_0^T \epsilon^2(t)dt$$

Standard optimization procedure

- construction of an extended objective functional i.e., add constraints through an *adjoint state* $\chi(x, t)$

$$J(\epsilon) = \langle \Psi(T) | O | \Psi(T) \rangle - \alpha \int_0^T \epsilon^2(t) dt$$

$$- 2\text{Re} \int_0^T \left\langle \chi(x, t), \left\{ \frac{\partial}{\partial t} + i \cdot [H_0 - \epsilon(t)\mu] \right\} \Psi(x, t) \right\rangle$$

Partial derivatives

$$\frac{\delta J(\epsilon)}{\delta \epsilon} = -2\alpha\epsilon(t) - 2\text{Im} \langle \chi | \mu | \Psi \rangle (t)$$

Euler-Lagrange critical point equation

$$\begin{cases} i \frac{\partial}{\partial t} \Psi(x, t) = (H_0 - \epsilon(t)\mu)\Psi(x, t) \\ \Psi(x, t = 0) = \Psi_0(x) \end{cases}$$

$$\begin{cases} i \frac{\partial}{\partial t} \chi(x, t) = (H_0 - \epsilon(t)\mu)\chi(x, t) \\ \chi(x, t = T) = O\Psi(x, T) \end{cases}$$

$$\alpha\epsilon(t) = -\text{Im} \langle \chi | \mu | \Psi \rangle (t)$$

- Chose a numerical algorithm to update the field $\epsilon(t)$, e.g.,

$$\epsilon^{n+1} = \epsilon^n + \frac{\delta J(\epsilon^n)}{\delta \epsilon} \quad (4)$$

slow convergence \implies complicated objective functional surface

Recent works by Alfio Borzi: functional surface seems to be very flat with many almost optimal regions.

Compute the optimal field $\epsilon(t)$ (Krotov cf. Tannor et. al 1992):
 $(\chi^{k-1}, \epsilon^{k-1}, \Psi^{k-1}) \rightarrow (\chi^k, \epsilon^k, \Psi^k)$

$$\begin{cases} i \frac{\partial}{\partial t} \Psi^k(x, t) = (H_0 - \epsilon^k(t)\mu)\Psi^k(x, t) \\ \Psi^k(x, t=0) = \Psi_0(x) \end{cases} \quad (5)$$

$$\epsilon^k(t) = -\frac{1}{\alpha} \text{Im} \langle \chi^{k-1} | \mu | \Psi^k \rangle(t) \quad (6)$$

$$\begin{cases} i \frac{\partial}{\partial t} \chi^k(x, t) = (H_0 - \epsilon^k(t)\mu)\chi^k(x, t) \\ \chi^k(x, t=T) = O\Psi^k(x, T) \end{cases} \quad (7)$$

In practice solve the equations (5)-(6) by propagating the non-linear equation

$$\begin{cases} i \frac{\partial}{\partial t} \Psi^k(x, t) = (H_0 + \frac{1}{\alpha} \text{Im} \langle \chi^{k-1} | \mu | \Psi^k \rangle(t)\mu)\Psi^k(x, t) \\ \Psi^k(x, t=0) = \Psi_0(x) \end{cases} \quad (8)$$

Zhu & Rabitz formulation (1998)

$$\begin{cases} i \frac{\partial}{\partial t} \Psi^k(x, t) = (H_0 - \epsilon^k(t)\mu)\Psi^k(x, t) \\ \Psi^k(x, t=0) = \Psi_0(x) \end{cases}$$

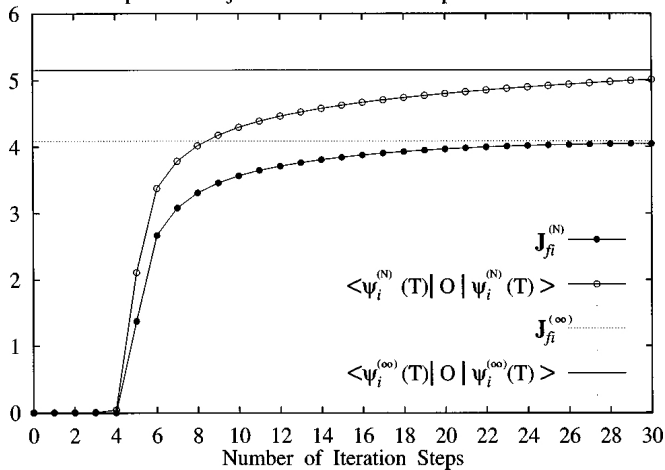
$$\epsilon^k(t) = -\frac{1}{\alpha} \text{Im} \langle \chi^{k-1} | \mu | \Psi^k \rangle (t)$$

$$\begin{cases} i \frac{\partial}{\partial t} \chi^k(x, t) = (H_0 - \tilde{\epsilon}^k(t)\mu)\chi^k(x, t) \\ \chi^k(x, t=T) = O\Psi^k(x, T) \end{cases}$$

$$\tilde{\epsilon}^k(t) = -\frac{1}{\alpha} \text{Im} \langle \chi^k | \mu | \Psi^k \rangle (t)$$

THEOREM (W. Zhu and H. Rabitz. *J. Chem. Phys.*, 109:385–391, 1998.) Suppose O is a semi-positive definite (auto-adjoint) operator. Then for any $k \geq 0$: $J(\epsilon^{k+1}) \geq J(\epsilon^k)$, i.e. there is an improvement in the functional at any iteration.

Optimized objective functional and expectation value



Other classes of algorithms (Y.Maday & G.T. 2002), other works by Y. Ohtsuki, S. Schirmer, D. Sugny, etc

$$\begin{cases} i \frac{\partial}{\partial t} \Psi^k(x, t) = (H_0 - \epsilon^k(t)\mu) \Psi^k(x, t) \\ \Psi^k(x, t = 0) = \Psi_0(x) \end{cases} \quad (9)$$

$$\epsilon^k(t) = (1 - \delta) \tilde{\epsilon}^{k-1}(t) - \frac{\delta}{\alpha} \text{Im} \langle \chi^{k-1} | \mu | \Psi^k \rangle(t) \quad (10)$$

$$\begin{cases} i \frac{\partial}{\partial t} \chi^k(x, t) = (H_0 - \tilde{\epsilon}^k(t)\mu) \chi^k(x, t) \\ \chi^k(x, t = T) = O \Psi^k(x, T) \end{cases} \quad (11)$$

$$\tilde{\epsilon}^k(t) = (1 - \eta) \epsilon^k(t) - \frac{\eta}{\alpha} \text{Im} \langle \chi^k | \mu | \Psi^k \rangle(t) \quad (12)$$

THEOREM If O is an hermitian observable semi-positive definite then, for any $\eta, \delta \in [0, 2]$ $J(\epsilon^{k+1}) \geq J(\epsilon^k)$.

$$\begin{aligned} J(\epsilon^{k+1}) - J(\epsilon^k) = & \\ & \langle \Psi^{k+1}(T) - \Psi^k(T) | O | \Psi^{k+1}(T) - \Psi^k(T) \rangle + \\ & \alpha \int_0^T \left(\frac{2}{\delta} - 1 \right) (\epsilon^{k+1} - \tilde{\epsilon}^k)^2 + \left(\frac{2}{\eta} - 1 \right) (\tilde{\epsilon}^k - \epsilon^k)^2 \end{aligned}$$

Experimental control

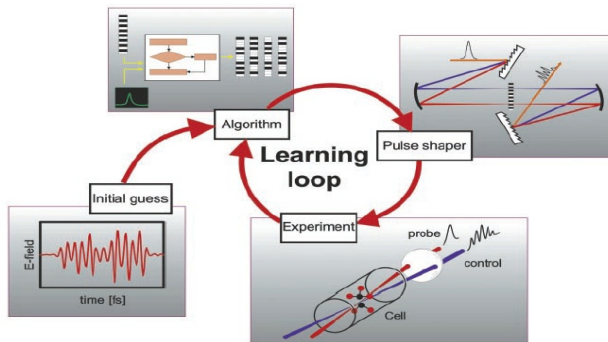


Fig. 2. A closed-loop process for teaching a laser to control quantum systems. The loop is entered with either an initial design estimate or even a random field in some cases. A current laser control field design is created with a pulse shaper and then applied to the sample. The action of the control is assessed, and the results are fed to a learning algorithm to suggest an improved field design for repeated excursions around the loop until the objective is satisfactorily achieved.

Figure: Experimental quantum control: In practice H and/or the dipole may not be known: one uses then a zero order algorithm (no gradients): genetic algorithms, Nelder-Mead simplex (C. Le Bris, H.Rabitz & G.T. PRE 2004) ... This is possible due to a high laboratory experimental repetition rate ($1\text{Hz} - 10^5\text{Hz}$ or more).

Lyapounov (tracking) approaches

keep decreasing $\|\psi(t) - \phi_0\|^2 \dots$

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Inversion and identification approaches

here μ and/or H_0 and/or $\psi(0)$ are unknown, need to be recovered from measurements

Identifiability

Theorem (C. Le Bris, M. Mirrahimi H. Rabitz, G.T.; 2006)
Consider two finite dimensional Hamiltonians H_1, H_2 and two dipole moments μ_1, μ_2 , with Ψ_1, Ψ_2 , solutions of :

$$i\dot{\Psi}_k = (H_k + \epsilon(t) \mu_k) \Psi_k$$

Suppose $\forall t \geq 0, \forall \epsilon(t) \in L^2$

$$|\langle \Psi_1(t) | e_i \rangle|^2 = |\langle \Psi_2(t) | e_i \rangle|^2 \quad i = 1, \dots, N, \quad (13)$$

($\{e_i\}_{i=1}^N$ = canonical basis of \mathbb{C}^N).

Assume for H_1, H_2 :

- ① Equations are controllable
- ② H_1 and H_2 have the same eigenvalues λ_j .
- ③ $\lambda_{i_1} - \lambda_{j_1} \neq \lambda_{i_2} - \lambda_{j_2}$ for $(i_1, j_1) \neq (i_2, j_2)$.
- ④ $\langle \phi_i | \mu | \phi_i \rangle = 0, \quad i = 1, \dots, N$.
- ⑤ there does not exist a subspace of dimension one or two spanned by the vectors $\{e_i\}$, which remains invariant during the free evolution ($\epsilon \equiv 0$) of the first system (H_1 and μ_1).

Then, there exist $\{\alpha_i\}_{i=1}^N$ such that, for all $1 \leq i, j \leq N$,

$$(\mu_1)_{ij} = e^{i(\alpha_i - \alpha_j)} (\mu_2)_{ij}, \quad (H_1)_{ij} = e^{i(\alpha_i - \alpha_j)} (H_2)_{ij}. \quad (14)$$

Optimal identification implementable experimental + numerical algorithms

- 1 Objective functional $J(\epsilon, \mu, V)$ the distance between the measures with the field $\epsilon(t)$ and the numerical simulation with the potential V , dipole μ and field ϵ .

“Canonical” formulation of the inversion problem : solve

$$\min_{V, \mu} \int_{L^2} J(\epsilon, \mu, V) d\epsilon(t) \quad (15)$$

BUT: space L^2 too large ... introduce a **discriminating field approach** that allows to distinguish between the admissible candidates.

- 2 **Inversion problem**: $S(\epsilon) = \{\mu, V; J(\epsilon, \mu, V) \leq \text{tolerance}\}$.
 $m(\epsilon) =$ a measure of $S(\epsilon)$.
- 3 **Optimization problem**: minimize $m(\epsilon)$. The solution $\bar{\epsilon}$ is the *discriminating field*; $S(\bar{\epsilon}) =$ set of possible solutions
 In practice: $m(\epsilon) =$ diameter, $S(\epsilon)$ not explicitly computed (!).

Algorithms can use fields coming from the identifiability theory.

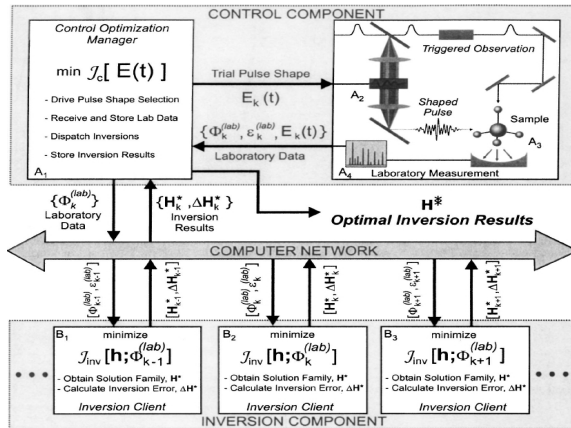


Figure: Optimal identification machine : implementable experimental + numerical algorithms (J.M. Geremia & H. Rabitz JCP, 2003)