Controllability of a rotating asymmetric molecule

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Structure of the talk

0)Bilinear Schrödinger equation

1)Control of quantum systems

2)Application to the control of a rotating molecule

. 2.1)Symmetries and controllability of symmetric molecules

. 2.2)Symmetries and controllability of asymmetric molecules

0) **Bilinear Schrödinger equation**

→ $\mathcal{H}$ Hilbert space (finite- or ∞-dimensional), $H_0, H_1, ..., H_l$ self-adjoint operators on $\mathcal{H}$, H_0 has discrete spectrum. We consider

$$\mathrm{i}\frac{d}{dt}\psi(t) = \Big(H_0 + \sum_{j=1}^{l} u_j(t)H_j\Big)\psi(t), \quad \psi(t) \in \mathcal{H}, \qquad (1)$$

$u = (u_1, ..., u_l) : [0, \infty) \to [-a, a]^l$ pwc control functions, $a > 0$.

→ Propagator $\Gamma^u(T)$ of (1): composition of flows $e^{-\mathrm{i}t(H_0+\sum_{j=1}^{l} u_j H_j)}$.

→ $\mathcal{S} \subset \mathcal{H}$ the unit sphere. For $\psi_0 \in \mathcal{S}$,

$$\mathrm{Reach}(\psi_0) = \{\psi \mid \exists u, T \text{ s.t. } \Gamma^u(T)(\psi_0) = \psi\}.$$

→ Equation (1) is

- **controllable** if $\mathrm{Reach}(\psi_0) = \mathcal{S}$, for any $\psi_0 \in \mathcal{S}$;
- **approximately controllable** if $\mathrm{Reach}(\psi_0)$ is dense in $\mathcal{S}$, for any $\psi_0 \in \mathcal{S}$.

1) Control of quantum systems

Criteria for finite-dimensional systems:

Theorem If $\dim \mathcal{H} = n < \infty$, (1) is controllable if

$$\mathfrak{su}(n) \subset \mathrm{Lie}\{\mathrm{i}H_0, \mathrm{i}H_1, \ldots, \mathrm{i}H_l\}.$$

→ $\{\phi_1, \ldots, \phi_n\}$, $\lambda_1 \leq \ldots \leq \lambda_n$ eigenvectors and eigenvalues of H_0.
→ $\Sigma = \{|\lambda_j - \lambda_k|, j, k = 1, \ldots, n\}$ spectral gaps of the system.
→ Reduced control Hamiltonians $\mathcal{E}_\sigma(H_j)$, for $\sigma \in \Sigma$, $j = 1, \ldots, l$

$$\langle \phi_i, \mathcal{E}_\sigma(H_j)\phi_k \rangle = \begin{cases} \langle \phi_i, H_j \phi_k \rangle, & \text{if } |\lambda_i - \lambda_k| = \sigma, \\ 0, & \text{otherwise.} \end{cases}$$

Theorem If $\dim \mathcal{H} = n < \infty$, (1) is controllable if

$$\mathfrak{su}(n) \subset \mathrm{Lie}\{\mathrm{i}H_0, \mathcal{E}_\sigma(\mathrm{i}H_j),\ \sigma \in \Sigma,\ j = 1, \ldots, l\}.$$

A criterium for ∞-dimensional systems: $\dim \mathcal{H} = \infty$,
$\{\phi_1, ..., \phi_n, ...\}$, $\lambda_1 \le ... \le \lambda_n \le ...$ eigenvectors and eigenvalues of H_0.
Define, for any $n \in \mathbb{N}$, orthogonal projection $\Pi_n : \mathcal{H} \to \mathcal{H}_n := \mathrm{span}\{\phi_1, ..., \phi_n\}$.

- Projected Hamiltonians $H_j^{(n)} = \Pi_n H_j \Pi_n$, $j = 0, \ldots, l$.
- $\Sigma_n = \{|\lambda_j - \lambda_k|, j, k = 1, ..., n\}$ spectral gaps of the projected system.
- Galerkin Spectral gaps: $\mathrm{span}\{\phi_1, ..., \phi_n\}$-preserving w.r.t higher approximations:

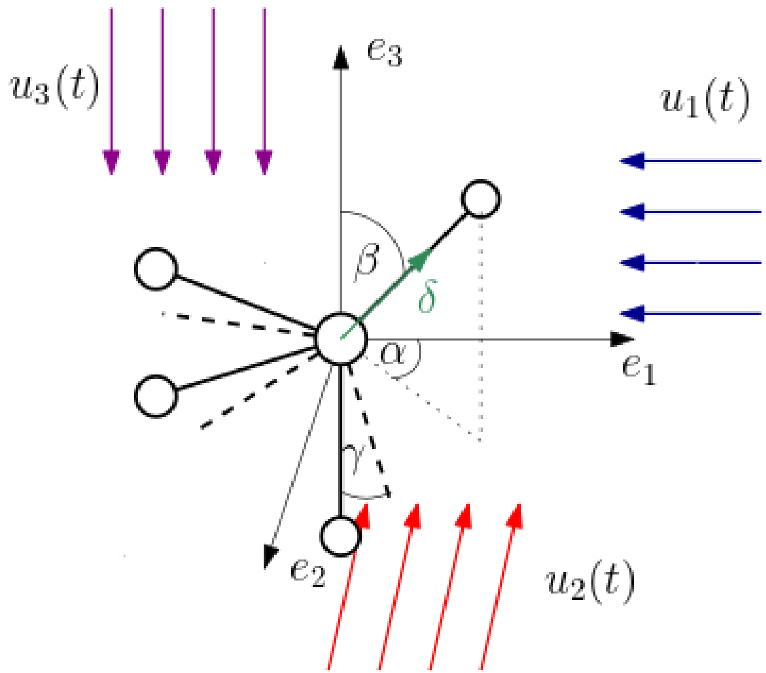
$$\Xi_n = \{(\sigma, j) \in \Sigma_n \times \{1, ..., l\} \mid \mathcal{E}_\sigma(H_j^{(N)}) = \left[\begin{array}{c|c} \mathcal{E}_\sigma(H_j^{(n)}) & 0 \\ \hline 0 & \star \end{array}\right] \text{ for every } N \ge n\}.$$

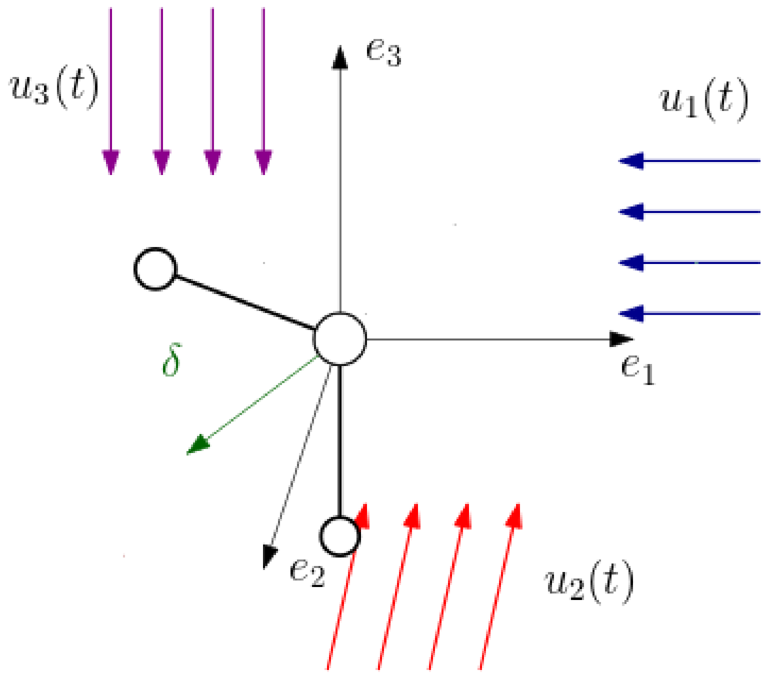
Definition Equation (1) is Lie-Galerkin if, for any $n_0 \in \mathbb{N}$, $\exists\, n \ge n_0$ s.t.

$$\mathfrak{su}(n) \subset \mathrm{Lie}\{\mathrm{i}H_0^{(n)}, \mathcal{E}_\sigma(\mathrm{i}H_j^{(n)}),\ (\sigma, j) \in \Xi_n\}.$$

Theorem[Boscain,Caponigro,Sigalotti(2014)] If (1) is Lie-Galerkin, then it is approximately controllable.

2) Application to the control of a rotating molecule





Free rotational dynamics:

Molecules as rigid bodies: a, b, c moving frame s.t. $A \geq B \geq C > 0$ rotational constants, configuration space $SO(3)$, rotational Hamiltonian

$$H_0 = AP_a^2 + BP_b^2 + CP_c^2,$$

P_i angular momentum, as differential self-adjoint operators on $\mathcal{H} = L^2(SO(3))$.

→ Orthogonal decomposition in harmonics:

$$L^2(SO(3)) = \overline{\mathrm{span}}\{D^j_{k,m} \mid j \in \mathbb{N},\ k, m = -j, ..., j\}.$$

→ **Symmetric-top**: $A = B$, c is the symmetry axis.

Then, $H_0 = BP^2 + (C - B)P_c^2$ with eigenvalues

$$H_0 D^j_{k,m} = \left[Bj(j+1) + (C - B)k^2\right] D^j_{k,m} =: E^j_k D^j_{k,m}.$$

→ Three families of spectral gaps: $\left|E^{j(+1)}_{k(+1)} - E^j_k\right|$.

Electric field to control the rotation:

Three orthogonal polarizations of electric field to control the system, interacting with the electric dipole $\delta = (\delta_a, \delta_b, \delta_c)$ fixed inside the molecule.

➞ e_1, e_2, e_3 resp. a, b, c fixed resp. moving frames, $R \in SO(3)$ position of the molecule, interaction Hamiltonians (bounded self-adjoint operators on $L^2(SO(3))$)

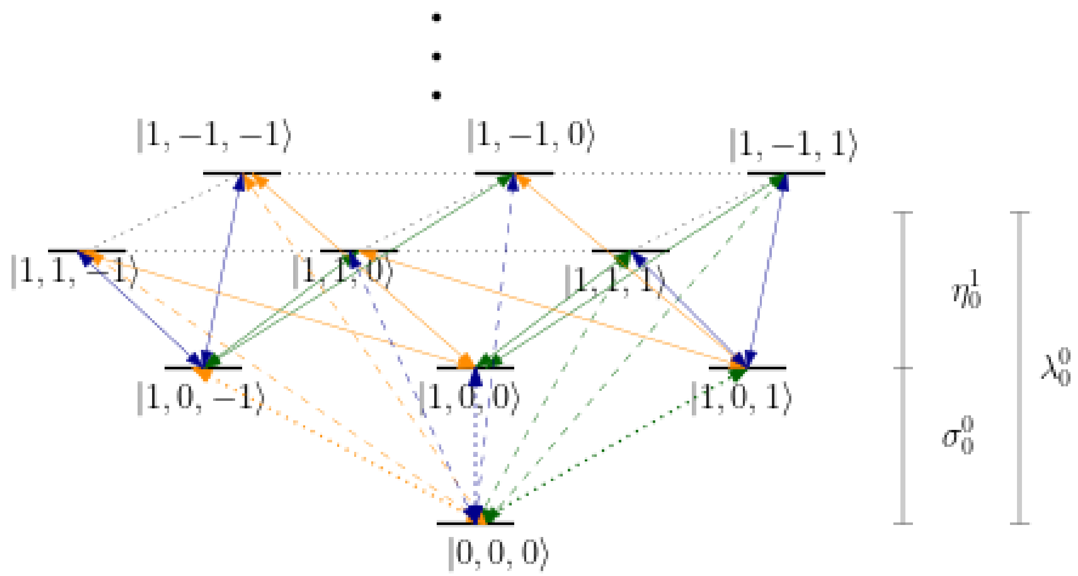
$$H_j(R, \delta) = -\langle R\,\delta, e_j \rangle, \quad j = 1, 2, 3,$$

➞ Controlled **Schrödinger equation**, $\psi(\cdot, t) \in L^2(SO(3))$:

$$\mathrm{i}\frac{\partial}{\partial t}\psi(R, t) = \Big(H_0 + \sum_{j=1}^{3} u_j(t) H_j(R, \delta) \Big) \psi(R, t), \tag{2}$$

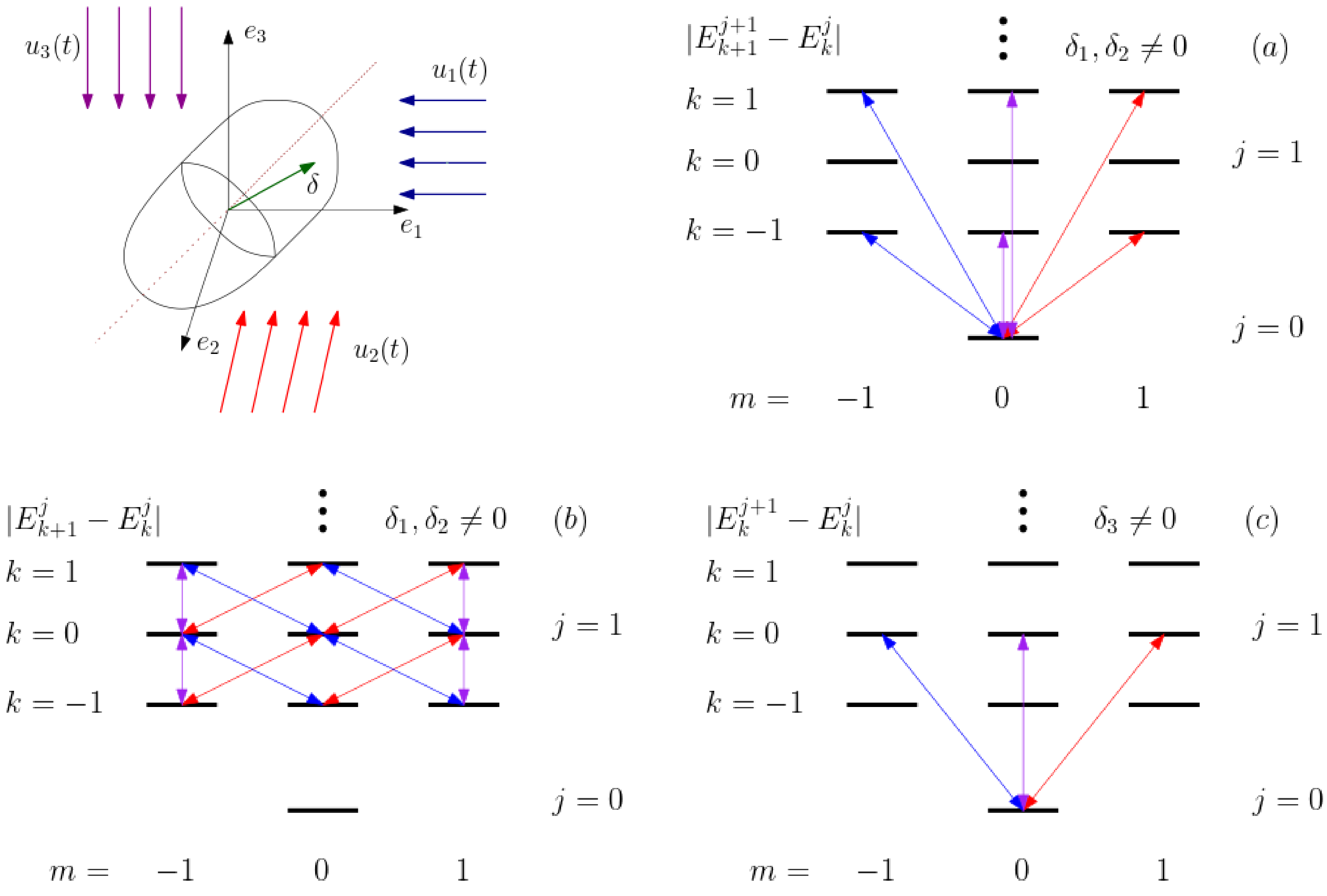
$u = (u_1, u_2, u_3) : [0, \infty) \to [-a, a]^3$ pwc control functions, $a > 0$.

3D spectral graph of a symmetric molecule



Transitions at spectral gaps $\lambda_0^0 := |E_1^1 - E_0^0|$, $\sigma_0^0 := |E_0^1 - E_0^0|$, and $\eta_0^1 = |E_1^1 - E_0^1|$ between the eigenstates $|j,k,m\rangle := D_{k,m}^j$, driven by H_1 (green arrows), H_2 (orange arrows), and H_3 (blue arrows). Same-shaped arrows correspond to equal spectral gaps.

State transitions induced by three polarizations at three spectral gaps



Symmetries and controllability of symmetric molecules:

Theorem[Boscain,Sigalotti,P.(2020)] Let $A = B > C > 0$ and $B/C \notin \mathbb{Q}$, then

- (i)$\delta = (0, 0, \delta_c) \Rightarrow$ (2) is not controllable;
- (ii)$\delta = (\delta_a, \delta_b, 0) \Rightarrow$ (2) is not controllable;
- (iii)δ different than (i) and (ii) $\Rightarrow$ (2) is approx. controllable.

Remark Non-controllability of cases (i) and (ii) follows from the existence of explicit conserved quantities. In particular, (i) is also a classical symmetry (that is, $\langle \psi, P_c \psi \rangle$), while (ii) is only quantum.

Idea of the proof of (iii) Use

$$\mathcal{X}_j := \{\mathrm{i}H_0, \mathcal{E}_{\omega_k^j}(\mathrm{i}H_l), \omega \in \{\lambda, \eta, \sigma\}, l = 1, 2, 3\}$$

and prove: (i) $\mathrm{Lie}\{\mathcal{X}_j\} = \mathfrak{su}(\mathcal{H}_j)$ and (ii) $(\omega_k^j, l) \in \Xi_j, \forall \omega \in \{\lambda, \eta, \sigma\}, l = 1, 2, 3$, for all $j \in \mathbb{N}, k = -j, \dots, j$. Conclude by applying Theorem[B,C,S(2014)].

Symmetries and controllability of asymmetric molecules:

Theorem[P.(2021)] Let $A > B > C > 0$, then

- (i)$\delta \in \{(\delta_a, 0, 0), (0, \delta_b, 0), (0, 0, \delta_c)\} \Rightarrow$ (2) is not controllable;
- (ii)δ different than (i) $\Rightarrow$ (2) is approx. controllable for a.e. A, B, C.

Remark Non-controllability of cases (i) follows from the existence of explicit conserved quantities, which are only quantum.

Idea of the proof of (ii)

→ $H_0 = H_0^{\text{symm}} + bV$ analytic perturbation of symmetric top rotational Hamiltonian, where $b \in [-1, 0]$ asymmetry parameter.

→ Apply Theorem[B,S,P(2020)] to the evolution associated with H_0^{symm}.

→ Controllability holds at $b = 0 \Rightarrow$ Controllability holds for a.e. $b \in [-1, 0]$.

References

For more details on the presentation,

[Boscain,Pozzoli,Sigalotti, *Classical and quantum controllability of a rotating symmetric molecule.* SIAM, J. Control Optim.,**59** (2021)]

For an application in quantum physics,

[Leibscher,Pozzoli,Pérez,Schnell,Sigalotti,Boscain,Koch: *Complete controllability despite degeneracy: Quantum control of enantiomer-specific state transfer in chiral molecules.* Submitted (arXiv: 2010.09296) (2020)]

Thank you for your attention !