Enhancing the controllability of quantum systems via a static field

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Abstract

We provide a sufficient condition for the controllability of a bilinear closed quantum system based on the notion of static field and weakly conically connected spectrum. More precisely, we show that if a controlled Hamiltonian with two inputs has a weakly conically connected spectrum, then, freezing one of the two inputs at almost every constant value, the obtained single-input system is controllable. The result is illustrated with two examples, enantiomer-selective excitation in a chiral molecule and the driven Jaynes–Cummings Hamiltonian.

1 Introduction

Consider a bilinear controlled closed quantum system

$$i\dot{\psi}(t) = H(u(t))\psi(t) = \Big(H_0 + \sum_{\ell=1}^{m} u_\ell(t) H_\ell\Big)\psi(t), \tag{1}$$

where $\psi(t)$ belongs to the unit sphere of a finite- or infinite-dimensional Hilbert space, H_0 is the internal Hamiltonian of the system, $H_1, \dots, H_m$ are operators describing the coupling between the system and the controls $u_1(t), \dots u_m(t)$.

The controls can account for many different physical interactions (e.g., electric or magnetic fields). Depending on the physical constraints on the control u_ℓ and on the corresponding coupling H_ℓ, it might not be possible to make $u_\ell(t)$ oscillate at the desired frequency (e.g., the frequency $E_k - E_j$ if u_ℓ is coupling the energy levels

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E_k and E_j) and actually u_ℓ should be considered almost constant. Such a situation results in having less controls on the system, since some of them should be treated as static. However, the possibility of choosing the value of such static fields can be enough to guarantee controllability.

This occurs for instance in molecular dynamics, where a constant electric field is applied to lift spectral degeneracies of the rotational drift Hamiltonian. This procedure is usually referred to as the Stark effect. Another external field can then be switched on and off, with a time-dependent frequency, to displace the state from an initial to a target condition: since the system is experiencing a static external field which is supposed to lift the degeneracies, this is now easier since it is more likely to possess a non-degenerate chain of transitions between those states. For such control strategies in the presence of a static field and a physical application we refer, e.g., to the recent work [12], dealing with the task of creating chiral vibrational wave packets in ensembles of achiral molecules. The dependence of the controllability properties of the system on the strength of the static field is studied in [12] by using graphical methods accounting for uncoupled transitions among energy levels obtained by resonant controls. Such graphical methods extend the enantio-selective controllability analysis for chiral quantum rotors proposed in [13, 17].

In this paper we propose a novel technique to study the controllability of systems subject both to time-varying and static fields, which leverages on the presence of (weakly) conical intersections in the spectrum of the Hamiltonian.

Consider a system driven by a real Hamiltonian and by two controls. In [14] we proved that if the spectrum is weakly conically connected, then the system is exactly controllable in the finite-dimensional case and approximately controllable in the infinite-dimensional one. The notion of weakly conically connected, introduced in [14], generalizes the more classical notion of conically connected spectrum, formalized in [4]. In both cases, the main assumption behind this notion is that the spectrum is discrete and that every two nearby energy levels of the system are connected by an eigenvalue intersection. For conically connected spectra these intersections must be conical, while for weakly conically connected spectra it is enough that a conical direction through the intersection exists. Moreover, in the case of weakly conically connected spectra we replace the assumption that eigenvalue intersection cannot pile up at a common value of the control by another, weaker, assumption, stating that the eigenvalue intersections have rationally unrelated germs in a suitable sense (see Definition 3.8). Both for conically connected and weakly conically connected spectra, the controllability result is reflected in an adiabatic control strategy to steer the system between two eigenstates going through eigenvalue intersections along conical directions.

The main result of the present study is that, under the assumption that the spectrum is weakly conically connected, freezing one of the two controls, then, for almost every choice of its constant value, the system is controllable (exactly for finite-dimensional systems and approximately for infinite-dimensional ones) via the other control.

The price to pay is that, since only one control can be modulated as a function of time, the system cannot be controlled via adiabatic techniques and hence the explicit expression of the non-constant control realizing the transition is not easily computable (although in principle possible via Lie–Galerkin techniques, see, for instance, [3,5,6]).

The structure of the paper is the following. In Section 2 we introduce the main assumptions on the class of systems that we are considering. In Section 3 we recall the controllability results from [14]. Section 4 contains the statement and the proof of our main result (Theorem 4.1). In Sections 5 and 6 we apply our theory on a 3-level enantio-selectivity model and on the driven Jaynes–Cummings system.

2 Assumptions

We consider the controllability of the bilinear Schrödinger equation

$$i\dot{\psi}(t) = H(u(t))\psi(t) = \Big(H_0 + \sum_{\ell=1}^{m} u_\ell(t) H_\ell\Big)\psi(t), \tag{2}$$

where $\psi(t)$ belongs to a complex Hilbert space $\mathcal{H}$, $H(u(t))$ is a self-adjoint operator on $\mathcal{H}$, and the control $u(\cdot) = \big(u_1(\cdot), \dots, u_m(\cdot)\big)$ takes values in an open and connected subset U of $\mathbb{R}^m$. We say that System (2) satisfies **(H)** if

- $H_0, \dots, H_m$ $(m \geq 2)$ are self-adjoint operators on $\mathcal{H}$ with $\dim(\mathcal{H}) < \infty$,

and that System (2) satisfies **($\mathbf{H}^\infty$)** if the following assumptions hold true:

- $\mathcal{H}$ is a separable Hilbert space with $\dim(\mathcal{H}) = \infty$,
- H_0 is a self-adjoint operator on $\mathcal{H}$ with domain $D(H_0)$ and $H_1, \dots, H_m$ $(m \geq 2)$ are symmetric operators on $D(H_0)$,
- H_0 is an operator with compact resolvent, i.e., $(H_0+i)^{-1}$ is a compact operator, and $H_1, \dots, H_m$ are Kato-small with respect to H_0 (see [7]).

Under assumption **(H)** or **($\mathbf{H}^\infty$)**, the spectrum of $H(u)$ is discrete for every $u \in U$. Let $\mathcal{I}$ denote $\{1, \dots, \dim(\mathcal{H})\}$ (respectively, $\mathbb{N}^*$) when **(H)** (respectively, **($\mathbf{H}^\infty$)**) is satisfied. For every $u \in U$, we denote by $\{\lambda_j(u)\}_{j\in\mathcal{I}}$ the increasing sequence of eigenvalues of $H(u)$, counted according to their multiplicities, and by $\{\phi_j(u)\}_{j\in\mathcal{I}}$ a corresponding sequence of eigenvectors forming an othonormal basis of $\mathcal{H}$.

3 Controllability of systems with weakly conically connected spectrum: previous results

Let us recall a classical definition of controllability for systems evolving in a finite-dimensional Hilbert space.

Definition 3.1 (Controllability in the unitary group)**.** *When $n = dim(\mathcal{H}) < \infty$, the* lift of system (2) *on $\mathcal{G} = U(n)$ (or on $\mathcal{G} = SU(n)$ in the case where $iH_0, \dots, iH_m \in su(n)$) is defined as*

$$i\dot{q}(t) = H(u(t))q(t), \tag{3}$$

where $q(\cdot)$ takes values in $\mathcal{G}$ and has the identity as initial condition. System (2) *is* exactly controllable in the unitary group *if its lift* (3) *has the property that, for every choice of initial and final state in $\mathcal{G}$, there exists an admissible trajectory of the system going from the former to the latter.*

For systems evolving in an infinite-dimensional Hilbert space the natural counterpart of the previous definition is the following.

Definition 3.2 (Approximate controllability in the unitary group)**.** *Let* **(H^∞)** *be satisfied. For every piecewise constant control $u : [0,T] \to U$, in the form of $u = \sum_{j=1}^{l} u_j 1_{(t_{j-1},t_j)}$ with $t_0 = 0$, let the* propagator *of system* (2) *associated with u be given by*

$$\Upsilon_t^u = e^{i(t-t_l)H(u_l)} \circ e^{i(t_l-t_{l-1})H(u_{l-1})} \circ \cdots \circ e^{it_1 H(u_1)}, \quad \text{for } t_l < t \leq t_{l+1}.$$

We say that system (2) *is* approximately controllable in the unitary group *if for every $k \in \mathbb{N}$, $\Upsilon : \mathcal{H} \to \mathcal{H}$ unitary, $\psi_0, \ldots, \psi_k$ in the unit sphere of $\mathcal{H}$, and every $\epsilon > 0$, there exist a time $T \geq 0$ and a piecewise constant control function $u : [0,T] \to U$ such that $\|\Upsilon_T^u(\psi_j) - \Upsilon(\psi_j)\| < \epsilon$ for every $j = 1, \ldots, k$.*

Remark 3.3. *Notice that different authors use different names for the type of controllability defined in Definition 3.2. For instance in [3] it is called* simultaneous approximate controllability *and in [8] it is called* strong controllability*.*

As explained in the introduction, this paper deals with the problem of understanding when the controllability (or approximate controllability) of the system corresponding to the Hamiltonian $H(\cdot,\cdot)$ implies that, fixing the control $u_1 = \bar{u}_1$, the system with the Hamiltonian $H(\bar{u}_1,\cdot)$ is still controllable (or approximately controllable) for almost every $\bar{u}_1$. Without additional hypotheses, this is not true in general. Consider, for instance, the system corresponding to the Hamiltonian

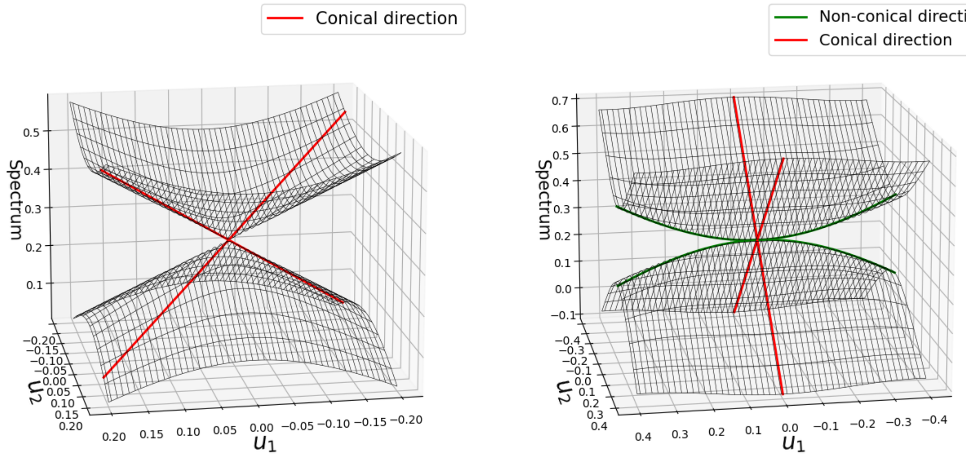
$$H(u,v) = \begin{pmatrix} 1 & u & v & 0 \\ u & 2 & 0 & 2v \\ v & 0 & 3 & u \\ 0 & 2v & u & 6 \end{pmatrix},$$

which is controllable. However, for all $\bar{v} \in \mathbb{R}$, the system with Hamiltonian $H(\cdot,\bar{v})$ is not controllable. In the following, we will prove that, if the spectrum of $H(\cdot,\cdot)$ is weakly conically connected and has rationally unrelated germs, we can deduce controllability results not only for the two-input system but also for the single-input system with the Hamiltonian $H(\bar{u}_1,\cdot)$ for almost every $\bar{u}_1$.

Definition 3.4 (Conical intersection)**.** *Let $\mathcal{I}^- = \mathcal{I} \setminus \{\dim \mathcal{H}\}$ (that is, $\mathcal{I}^- = \{1, \ldots, \dim \mathcal{H} - 1\}$ if $\dim \mathcal{H} < \infty$ and $\mathcal{I}^- = \mathcal{I}$ otherwise). Given $j \in \mathcal{I}^-$, an eigenvalue intersection $\bar{u} \in U$ between two eigenvalues $\lambda_j(\cdot)$ and $\lambda_{j+1}(\cdot)$ is said to be* conical *if it is of multiplicity two and there exist $C, \delta > 0$ such that*

$$\frac{1}{C}|t| \leq |\lambda_{j+1}(\bar{u} + t\eta) - \lambda_j(\bar{u} + t\eta)| \leq C|t|$$

for every unit direction $\eta \in \mathbb{R}^m$ and $t \in (-\delta, \delta)$.



(a) A conical intersection (b) A weakly conical intersection

Figure 1: Intersections between eigenvalues of $H(u)$ seen as functions of $u = (u_1, u_2)$, with $m = 2$: For the conical intersection, the red lines show the separation of the eigenvalues along a conical direction. For the weakly conical intersection, the red lines show the separation of the eigenvalues along a conical direction and the green lines show the separation along a non-conical direction.

See Figure 1a for an example of conical intersection.

Definition 3.5 (Conical direction)**.** *Given $j \in \mathcal{I}^-$ and an intersection $\bar{u}$ between $\lambda_j(\cdot)$ and $\lambda_{j+1}(\cdot)$, a unit vector $\eta \in \mathbb{R}^m$ is said to be a* conical direction at $\bar{u}$ *if there exist $C, \delta > 0$ such that*

$$\frac{1}{C}|t| \leq \lambda_{j+1}(\bar{u} + t\eta) - \lambda_j(\bar{u} + t\eta) \leq C|t|$$

for $t \in (-\delta, \delta)$.

Let us now recall the notion of weakly conical intersection proposed in [14], which generalizes the notion of semi-conical intersection considered in [1].

Definition 3.6 (Weakly conical intersection)**.** *A non-conical eigenvalue intersection $\bar{u} \in U$ between two eigenvalues $\lambda_j(\cdot)$ and $\lambda_{j+1}(\cdot)$, $j \in \mathcal{I}^-$, is said to be* weakly conical *if it is of multiplicity two, $\bar{u}$ is an isolated point in*

$$\{u \in U \mid \lambda_j(u) = \lambda_{j+1}(u)\},$$

and there exists at least one conical direction at $\bar{u}$.

See Figure 1b for an example of weakly conical intersection.

Definition 3.7 (Weakly conical connected spectrum)**.** *Let **(H)** or **(H$^\infty$)** be satisfied. We say that the spectrum of $H(\cdot)$ is* weakly conically connected *if each of its eigenvalue intersections is either conical or weakly conical, and, moreover, for every $j \in \mathcal{I}^-$, there exists $\bar{u}_j \in U$ such that $\lambda_j(\bar{u}_j) = \lambda_{j+1}(\bar{u}_j)$.*

Definition 3.8 (Rationally unrelated germs)**.** *Let **(H)** or **(H^∞)** be satisfied and the spectrum of $H(\cdot)$ be weakly conically connected. For a given intersection point $\bar{u} \in U$ of the spectrum of $H(\cdot)$, define*

$$\mathcal{I}(\bar{u}) := \{j \in \mathcal{I}^- \mid \lambda_j(\bar{u}) = \lambda_{j+1}(\bar{u})\}. \tag{4}$$

We say that intersections have rationally unrelated germs at $\bar{u}$ *if for every finite subset $\mathcal{J}$ of $\mathcal{I}(\bar{u})$, and for every neighborhood V of $\bar{u}$, the family of functions*

$$\Big(V \ni u \mapsto \lambda_{j+1}(u) - \lambda_j(u)\Big)_{j\in\mathcal{J}}$$

is rationally independent, meaning that if $\{\alpha_j\}_{j\in\mathcal{J}} \in \mathbb{Q}^{\mathcal{J}}$ is such that

$$\sum_{j\in\mathcal{J}} \alpha_j(\lambda_{j+1}(u) - \lambda_j(u)) = 0, \quad \forall u \in V,$$

then $\alpha_j = 0$ for all $j \in \mathcal{J}$.

Let us stress that in [14, Section 4] we proposed constructive tests for checking that an eigenvalue intersection has rationally unrelated germs. The following spectral conditions for controllability have been proven in [14].

Theorem 3.9 (Theorem 1.1 in [14])**.** *Assume that System* (2) *satisfies **(H)**, that the spectrum of $H(\cdot)$ is weakly conically connected, and that its eigenvalue intersections have rationally unrelated germs at each intersection point. Then $Lie(iH_0, \ldots, iH_m) = su(n)$ if $iH_0, \ldots, iH_m \in su(n)$ and $Lie(iH_0, \ldots, iH_m) = u(n)$ otherwise, meaning that System* (2) *is exactly controllable in the unitary group.*

Theorem 3.10 (Theorem 3.8 and Remark 3.9 in [14])**.** *Assume that System* (2) *satisfies **(H^∞)**, that the spectrum of $H(\cdot)$ is weakly conically connected, and that its eigenvalue intersections have rationally unrelated germs at each intersection point. Then System* (2) *is approximately controllable in the unitary group.*

4 Main result

From now on, let us fix $m = 2$. For every possible static value $u_1 = \bar{u}_1$ in

$$I^1 = \{u_1 \in \mathbb{R} \mid \exists u_2 \in \mathbb{R} \text{ s.t. } (u_1, u_2) \in U\} \tag{5}$$

let us consider the single-input system

$$i\dot{\psi}(t) = H(\bar{u}_1, u_2(t))\psi(t) = \Big(H_0 + \bar{u}_1 H_1 + u_2(t) H_2\Big)\psi(t), \tag{6}$$

where $u_2(\cdot)$ takes value in the set $\{u_2 \mid (\bar{u}_1, u_2) \in U\}$. Notice that for each possible static value $\bar{u}_1$, $u_2(\cdot)$ takes value in an open nonempty set, since U is open in $\mathbb{R}^2$.

Under the assumptions of Theorems 3.9 and 3.10, we can further deduce the controllability of the single-input system with Hamiltonian $H(\bar{u}_1, \cdot)$.

Theorem 4.1. *Assume that $m = 2$, that System* (2) *satisfies **(H)** (respectively, **(H^∞)**), that the spectrum of $H(\cdot)$ is weakly conically connected, and that its eigenvalue intersections have rationally unrelated germs at each intersection point. Then, for almost every $\bar{u}_1 \in I^1$ defined in* (5)*, the single-input system* (6) *is controllable (respectively, approximately controllable) in the unitary group.*

Before proving Theorem 4.1, let us show an intermediate result. Recall that $\mathcal{I}$ and $\mathcal{I}^-$ can be defined as in Section 2. For $u \in U$ and $j \in \mathcal{I}$ such that $\lambda_j(u)$ is simple, denote by $P_j(u)$ the associated eigenprojection on the space spanned by the eigenvector $\phi_j(u)$.

Lemma 4.2. *Let $m = 2$, assume that System* (2) *satisfies **(H)** or **(H^∞)**, and that the spectrum of $H(\cdot)$ is weakly conically connected. Then there exists a dense subset $\bar{U}$ of U with zero-measure complement such that for each $\bar{u} \in \bar{U}$ all eigenvalues of $H(\bar{u})$ are simple and*

$$\langle \phi_j(\bar{u}), H_2 \phi_{j+1}(\bar{u}) \rangle \neq 0, \quad \forall j \in \mathcal{I}^-.$$

Proof. For each $j \in \mathcal{I}$, let us define

$$U_j = \{ u \in U \mid \lambda_j(u) \text{ is simple} \}.$$

Since the spectrum is weakly conically connected, U_j is an open and dense subset of U with discrete complement.

For every $j \in \mathcal{I}^-$, let us define

$$Z_j = \{ u \in U_j \cap U_{j+1} \mid P_j(u) H_2 P_{j+1}(u) = 0 \}.$$

Because of the analyticity of $u \mapsto P_j(u) H_2 P_{j+1}(u)$ (cf. [7]) on the open and connected set $U_j \cap U_{j+1}$, we have either that $Z_j = U_j \cap U_{j+1}$ or that Z_j has empty interior and zero measure. Let us define

$$\bar{U} = (\cap_{j \in \mathcal{I}} U_j) \setminus (\cup_{j \in \mathcal{I}^-} Z_j).$$

Notice that for all $u \in \bar{U}$, the spectrum of $H(\cdot)$ is simple and

$$|\langle \phi_j(u), H_2 \phi_{j+1}(u) \rangle| = \| P_j(u) H_2 P_{j+1}(u) \| \neq 0, \quad \forall j \in \mathcal{I}^-.$$

Using the superscript c to denote the complement of a set in U, we have

$$\bar{U}^c = (\cup_{j \in \mathcal{I}} U_j^c) \cup (\cup_{j \in \mathcal{I}^-} Z_j).$$

We need to prove that $\bar{U}^c$ has empty interior and zero measure. Since a countable union of subsets of $\mathbb{R}^m$ with zero measure has zero measure and empty interior, we are left to prove that for each $j \in \mathcal{I}^-$, the measure of Z_j is zero. This can be done by proving that $u \mapsto P_j(u) H_2 P_{j+1}(u)$ is not identically zero on $U_j \cap U_{j+1}$. Let us assume by contradiction that

$$P_j(u) H_2 P_{j+1}(u) = 0, \quad \forall u \in U_j \cap U_{j+1}. \tag{7}$$

Let us take an intersection $\bar{u} = (\bar{u}_1, \bar{u}_2) \in U$ between $\lambda_j(\cdot)$ and $\lambda_{j+1}(\cdot)$. Since $\bar{u}$ is an isolated point in U, there exists $\delta > 0$ such that

$$[\bar{u}_1 - \delta, \bar{u}_1 + \delta] \times [\bar{u}_2 - \delta, \bar{u}_2 + \delta] \setminus \{\bar{u}\} \subset U_j \cap U_{j+1}.$$

Define $W = [\bar{u}_1 - \delta, \bar{u}_1 + \delta] \times [\bar{u}_2 - \delta, \bar{u}_2 + \delta]$. For $u \in W$, let us denote by $P(u)$ the spectral projection associated with the pair of eigenvalues $\{\lambda_j(\cdot), \lambda_{j+1}(\cdot)\}$. For $u_1 \in [\bar{u}_1 - \delta, \bar{u}_1 + \delta]$, let us define $\gamma(u_1, \cdot) : [-\delta, \delta] \to W$ by $\gamma(u_1, t) = (u_1, \bar{u}_2 + t)$. Denote by $\mathcal{U}(\mathcal{H})$ the group of unitary transformations of $\mathcal{H}$ and let us define the transformation function $V : W \to \mathcal{U}(\mathcal{H})$ such that for all $u_1 \in [\bar{u}_1 - \delta, \bar{u}_1 + \delta]$, $V(u_1, \cdot)$ satisfies

$$\frac{\mathrm{d}}{\mathrm{d}t} V(\gamma(u_1, t)) = \left[\frac{\mathrm{d}}{\mathrm{d}t} P(\gamma(u_1, t)), P(\gamma(u_1, t))\right] V(\gamma(u_1, t)), \;\; V(\gamma(u_1, -\delta)) = \mathrm{Id}. \quad (8)$$

By classical results on solutions of ODEs, $V(\cdot, \cdot)$ is well defined and continuous on W.

We prove next that for each vertical segment $\gamma(u_1, \cdot)$ that does not intersect $\bar{u}$, and for each $s \in \{j, j+1\}$, the transformed projector $V(\cdot)^\dagger P_s(\cdot) V(\cdot)$ is conserved along the segment.

Let us fix for now $u_1 \in [\bar{u}_1 - \delta, \bar{u}_1) \cup (\bar{u}_1, \bar{u}_1 + \delta]$, and set $P(t) = P(\gamma(u_1, t))$, $V(t) = V(\gamma(u_1, t))$, $P_j(t) = P_j(\gamma(u_1, t))$, $P_{j+1}(t) = P_{j+1}(\gamma(u_1, t))$, $\lambda_j(t) = \lambda_j(\gamma(u_1, t))$, $\lambda_{j+1}(t) = \lambda_{j+1}(\gamma(u_1, t))$, and $H(t) = H(\gamma(u_1, t))$. By differentiating with respect to t the equation

$$P_j(t) P_{j+1}(t) = 0, \quad t \in [-\delta, \delta],$$

we obtain that

$$\dot{P}_j(t) P_{j+1}(t) = -P_j(t) \dot{P}_{j+1}(t), \qquad t \in [-\delta, \delta]. \quad (9)$$

Using the equation

$$P_s(t) H(t) = H(t) P_s(t) = \lambda_s(t) P_s(t), \qquad s \in \{j, j+1\},$$

and differentiating the equation

$$P_j(t) H(t) P_{j+1}(t) = 0, \qquad t \in [-\delta, \delta],$$

we deduce that

$$\begin{aligned} 0 &= \dot{P}_j(t) H(t) P_{j+1}(t) + P_j(t) \dot{H}(t) P_{j+1}(t) + P_j(t) H(t) \dot{P}_{j+1}(t) \\ &= \lambda_{j+1}(t) \dot{P}_j(t) P_{j+1}(t) + P_j(t) H_2 P_{j+1}(t) + \lambda_j(t) P_j(t) \dot{P}_{j+1}(t). \end{aligned}$$

By equations (7) and (9), and given that $\lambda_j(t) \neq \lambda_{j+1}(t)$, we get, for all $t \in [-\delta, \delta]$,

$$\dot{P}_j(t) P_{j+1}(t) = \frac{1}{\lambda_j(t) - \lambda_{j+1}(t)} P_j(t) H_2 P_{j+1}(t) = 0 = \dot{P}_{j+1}(t) P_j(t). \quad (10)$$

Hence,

$$\begin{aligned}\left[\dot{P}(t), P(t)\right] &= \left[\dot{P}_j(t) + \dot{P}_{j+1}(t), P_j(t) + P_{j+1}(t)\right] \\ &= \left[\dot{P}_j(t), P_j(t)\right] + \left[\dot{P}_{j+1}(t), P_{j+1}(t)\right].\end{aligned} \tag{11}$$

Moreover, by differentiating

$$P_s(t)^2 = P_s(t), \qquad s \in \{j, j+1\},$$

with respect to t, it follows that, for all $t \in [-\delta, \delta]$,

$$\dot{P}_s(t)P_s(t) + P_s(t)\dot{P}_s(t) = \dot{P}_s(t), \qquad s \in \{j, j+1\}. \tag{12}$$

For $s \in \{j, j+1\}$, by left-multiplying equation (12) by $P_s(t)$, we can deduce that

$$P_s(t)\dot{P}_s(t)P_s(t) + P_s(t)\dot{P}_s(t) = P_s(t)\dot{P}_s(t), \qquad t \in [-\delta, \delta],$$

and thus

$$P_s(t)\dot{P}_s(t)P_s(t) = 0, \qquad t \in [-\delta, \delta]. \tag{13}$$

By equations (8), (10), (11), (12), and (13), we can then obtain that, for $s \in \{j, j+1\}$ and along each $\gamma(u_1, \cdot)$,

$$\begin{aligned}\frac{\mathrm{d}}{\mathrm{d}t}\big(V(t)^\dagger P_s(t)V(t)\big) &= -V(t)^\dagger[\dot{P}(t), P(t)]P_s(t)V(t) + V(t)^\dagger \dot{P}_s(t)V(t) \\ &\quad + V(t)^\dagger P_s(t)[\dot{P}(t), P(t)]V(t) \\ &= V(t)^\dagger\Big(- \dot{P}_s(t)P_s(t) + \dot{P}_s(t) - P_s(t)\dot{P}_s(t)\Big)V(t) = 0.\end{aligned}$$

Therefore, for $s \in \{j, j+1\}$, $V(\cdot)^\dagger P_s(\cdot)V(\cdot)$ is conserved along each vertical segment $\gamma(u_1, \cdot)$ with $u_1 \in [\bar{u}_1 - \delta, \bar{u}_1) \cup (\bar{u}_1, \bar{u}_1 + \delta]$. For all $u_1 \in [\bar{u}_1 - \delta, \bar{u}_1) \cup (\bar{u}_1, \bar{u}_1 + \delta]$ and $u_2 \in [\bar{u}_2 - \delta, \bar{u}_2 + \delta]$, we have that

$$P_s(u_1, u_2) = V(u_1, u_2)P_s(u_1, \bar{u}_2 - \delta)V(u_1, u_2)^\dagger.$$

Since $P_s(\cdot)$ is analytic on $W \setminus \{\bar{u}\}$ and $V(\cdot)$ is continuous on W, by continuous extension, we can obtain that

$$P_s(u_1, u_2) = V(u_1, u_2)P_s(u_1, \bar{u}_2 - \delta)V(u_1, u_2)^\dagger, \quad \forall u \in W \setminus \{\bar{u}\}.$$

Consider an analytic path $\gamma : (-1, 1) \to W$ such that $\gamma(0) = \bar{u}$ and $\dot{\gamma}(0)$ is a non-zero conical direction at $\bar{u}$ for the intersection between $\lambda_j(\cdot)$ and $\lambda_{j+1}(\cdot)$. Then, for $s \in \{j, j+1\}$, we have that

$$\lim_{t\to 0^+} P_s(\gamma(t)) = \lim_{t\to 0^-} P_s(\gamma(t)) = V(\bar{u}_1, \bar{u}_2)P_s(\bar{u}_1, \bar{u}_2 - \delta)V(\bar{u}_1, \bar{u}_2)^\dagger.$$

However, by passing through a conical direction, we should have that

$$P_s(\gamma(0^-)) \neq P_s(\gamma(0^+)).$$

The contradiction is reached, concluding the proof. □

We also recall the following result.

Lemma 4.3 (Lemma 3.7 in [14])**.** *Let System* (2) *satisfy* ***(H)*** *or* ***(H$^\infty$)****. Assume that the spectrum of $H(\cdot)$ is weakly conically connected and that the eigenvalue intersections have rationally unrelated germs at every intersection point $\bar{u} \in U$. Then there exists a dense subset $\bar{U}$ of U with zero-measure complement such that if $\sum_{j=1}^{N} \alpha_j \lambda_j(\bar{u}) = 0$ with $\bar{u} \in \bar{U}$, $N \in \mathbb{N}^*$, $N \leq \dim \mathcal{H}$, and $\{\alpha_1, \ldots, \alpha_N\} \in \mathbb{Q}^N$ then $\alpha_1 = \cdots = \alpha_N$.*

We can now prove Theorem 4.1.

Proof of Theorem 4.1. By applying Lemma 4.3 we deduce that there exists a subset $\bar{U}_1$ of U which is dense and has zero-measure complement such that for all $\bar{u} \in \bar{U}_1$ the spectrum is simple and the eigenvalues $\{\lambda_j(\bar{u})\}_{j\in\mathcal{I}}$ are non-resonant (i.e. two spectral gaps $\lambda_j(\bar{u}) - \lambda_k(\bar{u})$ and $\lambda_r(\bar{u}) - \lambda_s(\bar{u})$ are different if $(j,k) \neq (r,s)$ and $j \neq k$, $r \neq s$). By applying Lemma 4.2, there exists another subset of $\bar{U}_2$ of U which is dense and has zero-measure complement such that for all $\bar{u} \in \bar{U}_2$ the spectrum is simple and

$$\langle \phi_j(\bar{u}), H_2 \phi_{j+1}(\bar{u}) \rangle \neq 0, \quad \forall j \in \mathcal{I}^-.$$

Let us define

$$\bar{U} = \bar{U}_1 \cap \bar{U}_2,$$

which is dense and has zero-measure complement in U. Note that

$$\bar{I}^1 = \left\{ u_1 \in \mathbb{R} \mid \exists u_2 \in \mathbb{R} \text{ s.t. } (u_1, u_2) \in \bar{U} \right\}$$

is dense and has zero-measure complement in I^1 (as defined at the beginning of this section). Take $\bar{u}_1 \in \bar{I}^1$ and consider the single-input system (6) for this static value. By definition of the interval $\bar{I}^1$, there exists $\bar{u}_2 \in \mathbb{R}$ such that $(\bar{u}_1, \bar{u}_2) \in U$, the eigenvalues are non-resonant at $(\bar{u}_1, \bar{u}_2)$ and, for all $j \in \mathcal{I}^-$, $\langle \phi_j(\bar{u}_1, \bar{u}_2), H_2 \phi_{j+1}(\bar{u}_1, \bar{u}_2) \rangle \neq 0$. We can deduce from [4, Proposition 11] and [4, Proposition 15] that, at every $\bar{u}_1 \in \bar{I}^1$, the single-input system (6) is controllable in the unitary group when $\dim(\mathcal{H}) < \infty$ and is approximately controllable in the unitary group when $\dim(\mathcal{H}) = \infty$. $\square$

5 Application: Enantioselective excitation in a three-level model for a chiral molecule

Chiral molecules exist in left-handed and right-handed conformations, called enantiomers. Enantiomer-selective excitation of rotational or ro-vibrational states of chiral molecules as a precursor for chiral discrimination with electric fields only is presently attracting significant interest in molecular physics and physical chemistry [10, 15, 18]. It is based on quantum interference in cyclic excitation between three quantum states, where constructive interference for one enantiomer and destructive interference for the other leads to selective population of one of the quantum states [9]. Such cyclic population transfer can be realized in chiral molecules via dipole interaction between the components of the molecular dipole moment and three time-dependent external fields

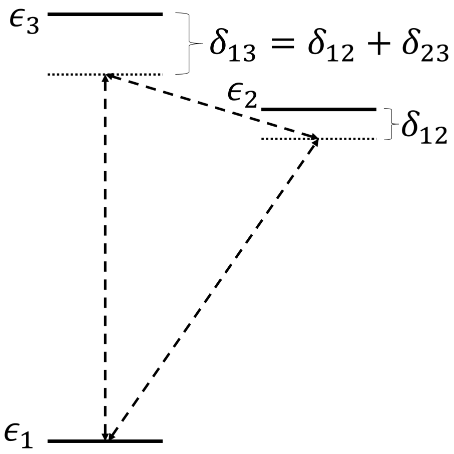


Figure 2: Three-level cyclic excitation scheme for enantiomer selective excitation of ro-vibrational states. The three states with energies ϵ_1, ϵ_2, and ϵ_3 are the ro-vibrational ground state $|0, 0_{00}\rangle$, and the states $|1, 1_{11}\rangle$ and $|1, 1_{10}\rangle$. The central frequency of the laser and microwave pulses driving the transition $i \leftrightarrow j$, $i, j = 1, 2, 3$, is detuned from the energy difference $\epsilon_j - \epsilon_i$ by δ_{ij}. Here we choose $\delta_{12} = \delta_{23}$.

with perpendicular polarization, e.g. three microwave pulses [15] or a combination of two infrared (IR) and one microwave pulse [11], and the cyclic excitation can be driven either resonantly or slightly detuned from resonance. Enantiomer-selectivity arises from the different sign of one of the Cartesian projections of the molecular dipole moment, in otherwise identical Hamiltonians for the two enantiomers. Enantiomer-selective controllability of asymmetric top molecules subject to several time-dependent controls has been analyzed using graphical methods [13, 17], showing that complete enantiomer-selective controllability can be achieved by combining at least five different combinations of polarization and frequency. Controllability analysis has also been applied to achiral molecules that can be made temporarily chiral, identifying the conditions for net chiral signals in an ensemble of molecules [12].

Here, instead, we consider the controllability for chiral molecules where the lowest state is the ro-vibrational ground state and the two other states are specific rotational states in an excitated vibrational level, as depicted in Fig. 2. The vibrational transitions can be driven by continuous wave IR radiation, with electric fields $\mathcal{E}_1 \cos(\omega_{12}t + \phi_{12})$ and $\mathcal{E}_2 \cos(\omega_{13}t + \phi_{13})$, while the pure rotational transition $2 \leftrightarrow 3$ is driven by a time-dependent microwave pulse, $\mathcal{E}_3 \cos(\omega_{23}t + \phi_{23})$, which is assumed to be phase-locked to the IR pulses [11]. The frequencies of the fields are detuned from resonance such that $\omega_{12} = \epsilon_2 - \epsilon_1 - \delta_{12}$, $\omega_{13} = \epsilon_3 - \epsilon_1 - \delta_{13}$, and $\omega_{23} = \epsilon_3 - \epsilon_2 - \delta_{23}$. Within the rotating wave approximation, this reduces the control Hamiltonian to two (quasi) static inputs and a single time-dependent control, a scenario for which enantiomer-selective controllability has not yet been studied. In this case, the Hamiltonian can be written as

$$H^{\pm} = \begin{pmatrix} -\delta_{12} & \pm H_{12} & H_{13} \\ \pm H_{12} & 0 & H_{23}(t) \\ H_{13} & H_{23}(t) & \delta_{23} \end{pmatrix}, \tag{14}$$

where the subscripts $\pm$ refer to the two enantiomers. Here, we assume that the detunings are chosen such that $\delta_{13} = \delta_{12} + \delta_{23}$ and the relative phases between the fields are set to $\phi_{12} = \phi_{23} = \phi_{13} = 0$. The Hamiltonians for the two enantiomers differ in the sign of one of the three off-diagonal elements, reflecting the sign difference of one of the dipole moment projections as mentioned above. The off-diagonal matrix elements read $H_{12} = -\mu_{12}\mathcal{E}_{12}$, $H_{13} = -\mu_{13}\mathcal{E}_{13}$, and $H_{23}(t) = -\mu_{23}\mathcal{E}_{23}(t)$, with μ_{ij} beeing the (transition) dipole moments and $\mathcal{E}_{12}$ and $\mathcal{E}_{13}$ the constant field amplitudes, while the field amplitude $\mathcal{E}_{12}(t)$ is time-dependent. If the corresponding Schrödinger equation is simultaneously controllable for H^+ and H^-, complete enantiomer-selective state transfer is possible in the setup described above.

Remark 5.1. *Notice that in equation* (14)*, the trace of $H^{\pm}$ is not necessarily zero. However, we can always choose $\delta_{12} = \delta_{23}$ in order for the trace to be zero. In the following, we will only study the case where $H^{\pm}$ is traceless.*

We investigate the controllability of the system consisting of the three-level drift Hamiltonian

$$H_0 = \begin{pmatrix} E_1 & 0 & 0 \\ 0 & E_2 & 0 \\ 0 & 0 & E_3 \end{pmatrix},$$

where $E_1 < E_2 < E_3$ and $E_1 + E_2 + E_3 = 0$, and set

$$H_u = \begin{pmatrix} 0 & 1 & 0 \\ 1 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix}, \quad H_v = \begin{pmatrix} 0 & 0 & 1 \\ 0 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix} \quad H_w = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 0 & 1 \\ 0 & 1 & 0 \end{pmatrix}.$$

Notice that $iH_0, iH_u, iH_v, iH_w \in \mathfrak{su}(3)$. For $\mathbf{u} = (u, v, w) \in \mathbb{R}^3$, define

$$\begin{aligned} H^+(\mathbf{u}) &= H_0 + uH_u + vH_v + wH_w, \\ H^-(\mathbf{u}) &= H_0 - uH_u + vH_v + wH_w. \end{aligned}$$

For $(\bar{u}, \bar{v}) \in \mathbb{R}^2$, let us consider the two systems

$$\begin{aligned} i\frac{\mathrm{d}}{\mathrm{d}t}\psi^+(t) &= H^+(\bar{u}, \bar{v}, w(t))\psi^+(t), \\ i\frac{\mathrm{d}}{\mathrm{d}t}\psi^-(t) &= H^-(\bar{u}, \bar{v}, w(t))\psi^-(t). \end{aligned} \tag{15}$$

Proposition 5.2. *Assume that the control $w(\cdot)$ takes values in $\mathbb{R}$. Then for almost every $(\bar{u}, \bar{v}) \in \mathbb{R}^2$, each system in equation* (15) *is controllable.*

Proof. Let us fix $\bar{v} \in \mathbb{R}$ and define $H_{\bar{v}}(\cdot, \cdot)$ as

$$H_{\bar{v}}(u, w) = H^{+}(u, \bar{v}, w).$$

Since $E_1 < E_3$, we can introduce the angle

$$\theta = \arctan\left(\frac{2\bar{v}}{E_3 - E_1}\right), \tag{16}$$

and the unitary transformation

$$P(\theta) = \begin{pmatrix} \cos\left(\frac{\theta}{2}\right) & 0 & -\sin\left(\frac{\theta}{2}\right) \\ 0 & 1 & 0 \\ \sin\left(\frac{\theta}{2}\right) & 0 & \cos\left(\frac{\theta}{2}\right) \end{pmatrix}.$$

Define the transformed Hamiltonian

$$\tilde{H}_{\bar{v}}(\tilde{u}, \tilde{w}) = P(\theta) H_{\bar{v}}(u, w) P^{\top}(\theta) = \begin{pmatrix} \tilde{E}_1 & \tilde{u} & 0 \\ \tilde{u} & E_2 & \tilde{w} \\ 0 & \tilde{w} & \tilde{E}_3 \end{pmatrix},$$

where

$$\begin{aligned} \tilde{u} &= \cos\left(\frac{\theta}{2}\right) u - \sin\left(\frac{\theta}{2}\right) w, \\ \tilde{w} &= \sin\left(\frac{\theta}{2}\right) u + \cos\left(\frac{\theta}{2}\right) w, \\ \tilde{E}_1 &= \frac{E_1 + E_2 - \sqrt{(E_1 - E_2)^2 + 4\bar{v}^2}}{2}, \\ \tilde{E}_3 &= \frac{E_1 + E_2 + \sqrt{(E_1 - E_2)^2 + 4\bar{v}^2}}{2}. \end{aligned} \tag{17}$$

Denote by $\tilde{\lambda}_1(\tilde{u}, \tilde{w}) \leq \tilde{\lambda}_2(\tilde{u}, \tilde{w}) \leq \tilde{\lambda}_3(\tilde{u}, \tilde{w})$ the eigenvalues of $\tilde{H}_{\bar{v}}(\tilde{u}, \tilde{w})$ and by $\lambda_1(u, w) \leq \lambda_2(u, v) \leq \lambda_3(u, v)$ those of $H_{\bar{v}}(u, w)$. Evidently, the spectrum of $\tilde{H}_{\bar{v}}(\tilde{u}, \tilde{w})$ is conically connected with conical intersections between $\tilde{\lambda}_1$ and $\tilde{\lambda}_2$ at

$$(\tilde{u}, \tilde{w}) = \left(\pm\sqrt{(\tilde{E}_3 - \tilde{E}_1)(\tilde{E}_3 - E_2)}, 0\right)$$

and conical intersections between $\tilde{\lambda}_2$ and $\tilde{\lambda}_3$ at

$$(\tilde{u}, \tilde{w}) = \left(0, \pm\sqrt{(\tilde{E}_3 - \tilde{E}_1)(E_2 - \tilde{E}_1)}\right).$$

Then we can deduce that the spectrum of $H_{\bar{v}}(u, w)$ is conically connected with conical intersections between λ_1 and λ_2 at

$$(u, w) = \pm\sqrt{(\tilde{E}_3 - \tilde{E}_1)(\tilde{E}_3 - E_2)}\left(\cos\left(\frac{\theta}{2}\right), -\sin\left(\frac{\theta}{2}\right)\right), \tag{18}$$

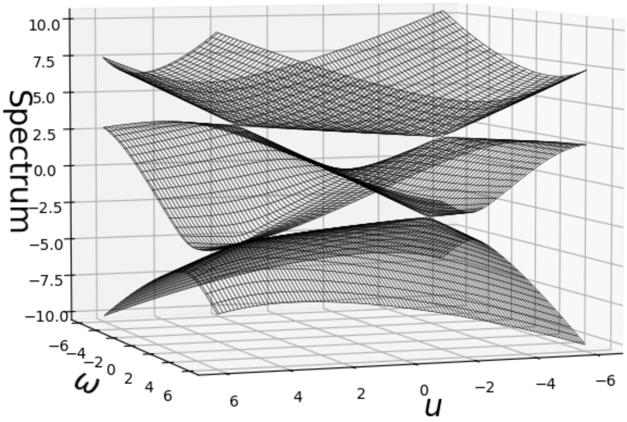


Figure 3: Case $(E_1, E_2, E_3) = (-1.5, 0.5, 1)$ and $\bar{v} = 3$: the spectrum of $H^+(u, \bar{v}, w)$ as a function of (u, w) is conically connected

and conical intersections between λ_2 and λ_3 at

$$(u, w) = \pm\sqrt{(\tilde{E}_3 - \tilde{E}_1)(E_2 - \tilde{E}_1)}\left(\sin\left(\frac{\theta}{2}\right), \cos\left(\frac{\theta}{2}\right)\right). \tag{19}$$

See Figure 3 for an illustrative example when $(E_1, E_2, E_3) = (-1.5, 0.5, 1)$ and $\bar{v} = 3$. By Theorem 4.1, we deduce that for almost every $\bar{u} \in \mathbb{R}$, the system characterized by $H_{\bar{v}}(\bar{u}, w)$ is controllable with $w(\cdot)$, which is true for all $\bar{v} \in \mathbb{R}$. Then we can conclude that for almost every $(\bar{u}, \bar{v}) \in \mathbb{R}^2$, the system characterized by $H^+(\bar{u}, \bar{v}, w)$ in equation (15) is controllable with $w(\cdot)$.

A similar reasoning shows that the system characterized by $H^-(\bar{u}, \bar{v}, w)$ in equation (15) is also controllable. □

For $\mathbf{u} = (u, v, w) \in \mathbb{R}^3$, denote by $\lambda_1^+(\mathbf{u}) \leq \lambda_2^+(\mathbf{u}) \leq \lambda_3^+(\mathbf{u})$ the eigenvalues and by $\phi_1^+(\mathbf{u}), \phi_2^+(\mathbf{u}), \phi_3^+(\mathbf{u})$ the unitary eigenstates of $H^+(\mathbf{u})$ and by $\lambda_1^-(\mathbf{u}) \leq \lambda_2^-(\mathbf{u}) \leq \lambda_3^-(\mathbf{u})$ and $\phi_1^-(\mathbf{u}), \phi_2^-(\mathbf{u}), \phi_3^-(\mathbf{u})$ those of $H^-(\mathbf{u})$.

Lemma 5.3. *There exists a dense subset U of $\mathbb{R}^3$ with zero-measure complement such that for all $\boldsymbol{u} = (u, v, w) \in \mathbb{R}^3$,*

$$\begin{aligned} &\lambda_2^+(\boldsymbol{u}) - \lambda_1^+(\boldsymbol{u}) \neq 0, \quad \left\langle \phi_1^+(\boldsymbol{u}), H_w \phi_2^+(\boldsymbol{u}) \right\rangle \neq 0, \\ &\lambda_2^+(\boldsymbol{u}) - \lambda_1^+(\boldsymbol{u}) \notin \left\{ \lambda_k^-(\boldsymbol{u}) - \lambda_j^-(\boldsymbol{u}) \mid 1 \leq j < k \leq 3 \right\}. \end{aligned} \tag{20}$$

Proof. Let us fix $\bar{v} \in \mathbb{R}^*$. As noticed in the proof of Proposition 5.2, the spectrum of $H(u, \bar{v}, w)$ as a function of $(u, w) \in \mathbb{R}^2$ is conically connected. Then, by Lemma 4.2, there exists a dense subset $V_1(\bar v)$ of $\mathbb{R}^2$ such that for all $(u,w) \in V_1(\bar v)$,

$$\lambda_2^+(u,\bar v,w) - \lambda_1^+(u,\bar v,w) \neq 0,$$
$$\left\langle \phi_1^+(u,\bar v,w), H_w \phi_2^+(u,\bar v,w)\right\rangle \neq 0.$$

Take a conical intersection $(\bar u,\bar w)$ between $\lambda_1^+(\cdot,\bar v,\cdot)$ and $\lambda_2^+(\cdot,\bar v,\cdot)$ as given in equation (18), with $\theta \in (-\pi,\pi)$ defined in equation (16) and $(\tilde E_1, \tilde E_3)$ defined in equation (17). Since $\bar v \neq 0$, it can be checked that $(\bar u,\bar w)$ cannot be an eigenvalue intersection of the spectrum of $H^-(\cdot,\bar v,\cdot)$, which means that

$$\lambda_k^-(\bar u,\bar v,\bar w) - \lambda_j^-(\bar u,\bar v,\bar w) \neq 0, \qquad \text{for } 1 \leq j < k \leq 3.$$

Then, by the analyticity of the spectra of $H^+(\cdot,\bar v,\cdot)$ and $H^-(\cdot,\bar v,\cdot)$, we can deduce that there exists a dense subset $V_2(\bar v)$ of $\mathbb{R}^2$ with zero-measure complement such that for all $(u,w) \in V_2(\bar v)$

$$\lambda_2^+(u,\bar v,w) - \lambda_1^+(u,\bar v,w) \notin \left\{\lambda_k^-(u,\bar v,w) - \lambda_j^-(u,\bar v,w) \mid 1 \leq j < k \leq 3\right\}.$$

Finally, we can conclude the proof by defining

$$U = \cup_{\bar v \in \mathbb{R}^*} \{\bar v\} \times \left(V_1(\bar v) \cap V_2(\bar v)\right).$$

□

Lemma 5.4. *There exists a dense subset U of $\mathbb{R}^3$ with zero-measure complement such that, for all $\boldsymbol{u} \in U$, there exists no Lie algebra isomorphism $f : \mathfrak{su}(3) \to \mathfrak{su}(3)$ such that $f\left(iH^+(\boldsymbol{u})\right) = iH^-(\boldsymbol{u})$ and $f\left(iH_w\right) = iH_w$.*

Proof. By Lemma 5.3, there exists a dense subset U of $\mathbb{R}^3$ with zero-measure complement such that the conditions in (20) are true for all $\mathbf{u} \in U$. Notice that for $p \in \mathbb{N}$,

$$\begin{aligned}\mathrm{ad}^{2p}_{iH^+(\mathbf{u})}(iH_w) = \sum_{j,k=1}^{3} i\big(-(\lambda_j^+(\mathbf{u}) - \lambda_k^+(\mathbf{u}))^2\big)^p \\ \left\langle \phi_j^+(\mathbf{u}), H_w \phi_k^+(\mathbf{u})\right\rangle \phi_j^+(\mathbf{u}) \left(\phi_k^+(\mathbf{u})\right)^\dagger, \\ \mathrm{ad}^{2p}_{iH^-(\mathbf{u})}(iH_w) = \sum_{j,k=1}^{3} i\big(-(\lambda_j^-(\mathbf{u}) - \lambda_k^-(\mathbf{u}))^2\big)^p \\ \left\langle \phi_j^-(\mathbf{u}), H_w \phi_k^-(\mathbf{u})\right\rangle \phi_j^+(\mathbf{u}) \left(\phi_k^+(\mathbf{u})\right)^\dagger.\end{aligned}$$

Take a polynomial $P(X) = \sum_{p=0}^m a_p X^p$ such that $P(x) = 0$ if $x \in \{0, -(\lambda_1^-(\mathbf{u}) - \lambda_2^-(\mathbf{u}))^2, -(\lambda_1^-(\mathbf{u}) - \lambda_3^-(\mathbf{u}))^2, -(\lambda_2^-(\mathbf{u}) - \lambda_3^-(\mathbf{u}))^2\}$ and $P(x) \neq 0$ otherwise. Then

$$\sum_{p=0}^m a_p \mathrm{ad}^{2p}_{iH^-(\mathbf{u})}(iH_w) = 0,$$

and, according to equation (20),

$$\sum_{p=0}^{m} a_p \mathrm{ad}^{2p}_{iH^+(\mathbf{u})}(iH_w) \neq 0.$$

Assume by contradiction that there exists a Lie algebra isomorphism $f : \mathfrak{su}(3) \to \mathfrak{su}(3)$ such that $f(iH^+(\mathbf{u})) = iH^-(\mathbf{u})$ and $f(iH_w) = iH_w$. Then we have

$$\begin{aligned} f\left(\sum_{p=0}^{m} a_p \mathrm{ad}^{2p}_{iH^+(\mathbf{u})}(iH_w)\right) &= \sum_{p=0}^{m} a_p \mathrm{ad}^{2p}_{f(iH^+(\mathbf{u}))}(f(iH_w)) \\ &= \sum_{p=0}^{m} a_p \mathrm{ad}^{2p}_{iH^-(\mathbf{u})}(iH_w) = 0. \end{aligned}$$

The contradiction is reached since $f^{-1}(0) = 0$. □

Lemma 5.5. *There exists a dense subset V of $\mathbb{R}^2$ with zero-measure complement such that for each $(u, v) \in V$ there exists no Lie algebra isomorphism $f : \mathfrak{su}(3) \to \mathfrak{su}(3)$ such that $f(iH^+(u, v, 0)) = iH^-(u, v, 0)$ and $f(iH_w) = iH_w$.*

Proof. Assume by contradiction that there exists $V^c \subset \mathbb{R}^2$ with non-zero measure such that for each $(u, v) \in V^c$ there exists a Lie algebra isomorphism f satisfying $f(iH^+(u, v, 0)) = iH^-(u, v, 0)$ and $f(iH_w) = iH_w$. Then by the linearity of f, for all $w \in \mathbb{R}$, we have $f(iH^+(u, v, w)) = iH^-(u, v, w)$ and $f(iH_w) = iH_w$. Then such isomorphism exists for all

$$\mathbf{u} = (u, v, w) \in V^c \times \mathbb{R},$$

and $V^c \times \mathbb{R}$ is a subset of $\mathbb{R}^3$ with non-zero measure. This is in contradiction with Lemma 5.4, concluding the proof. □

Proposition 5.6. *For almost every $(\bar{u}, \bar{v}) \in \mathbb{R}^2$, the two systems in equation* (15) *are simultaneously controllable with control $w(\cdot)$.*

Proof. By Proposition 5.2, there exists a dense subset V_1 of $\mathbb{R}^2$ with zero-measure complement, such that for all $(\bar{u}, \bar{v}) \in V_1$, each system in equation (15) is controllable with control $w(\cdot)$. Moreover by Lemma 5.5, there exists a dense subset V_2 of $\mathbb{R}^2$ with zero-measure complement such that for all $(\bar{u}, \bar{v}) \in V_1$, there exists no Lie algebra isomorphism $f : \mathfrak{su}(3) \to \mathfrak{su}(3)$ such that $f(iH^+(\bar{u}, \bar{v}, 0)) = iH^-(\bar{u}, \bar{v}, 0)$ and $f(iH_w) = iH_w$. Since $V = V_1 \cap V_2$ is a dense subset of $\mathbb{R}^2$ with zero measure complement, the conclusion follows from Lemma 3 in [2]. □

Applying the proven result to the scenario of enantiomer-selective controllability subject to two continuous wave IR fields and one time-dependent microwave pulse, modeled in the rotating wave approximation, we find controllability for $\delta_{12} = \delta_{23}$. For almost all static values $\bar{u}$ and $\bar{v}$ in (15) (i.e. $(\bar{u}, \bar{v})$ in a dense subset of $\mathbb{R}^2$ with zero-measure complement), with $\bar{u}$ and $\bar{v}$ denoting Rabi frequencies $\Omega_{12} = \mu_{12}\mathcal{E}_{12}$ and $\Omega_{13} = \mu_{13}\mathcal{E}_{13}$, the time-dependent control $w(t)$ yields enantiomer-selective controllability.

6 Application: Driven Jaynes–Cummings Hamiltonian

The Jaynes-Cummings Hamiltonian is a paradigmatic model in quantum optics. It describes a two-level system coupled to a quantum harmonic oscillator where the coupling is sufficiently weak to allow for the rotating wave approximation. Experimental realizations include atoms or molecules interacting with a quantized mode of the electromagnetic field in a cavity or internal states of trapped ions which are coupled to the quantized motion in the ion trap. Without external drive, the model is analytically solvable.

Here we consider, in the Hilbert space $\mathcal{H} = L^2(\mathbb{R}) \otimes \mathbb{C}^2$, the driven Jaynes–Cummings system

$$i\dot{\psi}(t) = H_{\mathrm{JC}}(u(t))\psi(t), \tag{21}$$

where the control $u(\cdot)$ (typically a classical electromagnetic field) is piecewise constant and takes values in $\mathbb{R}$. The driven Jaynes–Cummings Hamiltonian is defined as

$$H_{\mathrm{JC}}(u) = \omega\left(a^\dagger a + \frac{1}{2}\right) + \frac{\Omega}{2}\sigma_z + g(a\sigma_+ + a^\dagger\sigma_-) + u(a^\dagger + a),$$

where $\omega, \Omega > 0$ and $g \in \mathbb{R}$ are scalar parameters of the system. Here, following the classical physical notation, we have omitted the evident tensor products, $a^\dagger$ and a represent the creation and annihilation operators for the harmonic oscillator, that is,

$$a^\dagger = \frac{1}{\sqrt{2}}\left(x - \frac{\partial}{\partial x}\right), \quad a = \frac{1}{\sqrt{2}}\left(x + \frac{\partial}{\partial x}\right), \tag{22}$$

σ_z is the Pauli matrix $\left(\begin{smallmatrix}1 & 0\\ 0 & -1\end{smallmatrix}\right)$, whose eigenvectors are denoted by e_1, e_{-1}, and σ_- and σ_+ are given by

$$\sigma_- = |e_{-1}\rangle\langle e_1| = \begin{pmatrix}0 & 0\\ 1 & 0\end{pmatrix}, \quad \sigma_+ = |e_1\rangle\langle e_{-1}| = \begin{pmatrix}0 & 1\\ 0 & 0\end{pmatrix}.$$

We can deduce from the results of the previous sections the following result on the controllability of the driven Jaynes–Cummings system.

Theorem 6.1. *For almost every $\Omega, \omega > 0$ and $g \in \mathbb{R}$, the driven Jaynes–Cummings system* (21) *is approximately controllable in the unitary group.*

This result, under a slightly different assumption, has already been proven in [16]. Here we propose an alternative proof relying on weak conical connectedness.

Before proving Theorem 6.1, let us first consider the two-input Hamiltonian

$$\begin{aligned} H(u_1, u_2) =& H_0 + u_1 H_1 + u_2 H_2 \\ =& \omega\left(a^\dagger a + \frac{1}{2}\right) + \frac{\Omega}{2}\sigma_z + u_1(a\sigma_+ + a^\dagger\sigma_-) + u_2(a^\dagger + a). \end{aligned} \tag{23}$$

The associated bilinear Schrödinger equation is given by

$$i\dot{\psi}(t) = H(u(t))\psi(t) = H(u_1(t), u_2(t))\psi(t). \tag{24}$$

Here $u(\cdot)$ is a piecewise constant function that takes its values in $U = \mathbb{R}^2$. It is important to notice that $H(g,u) = H_{\mathrm{JC}}(u)$. The parameter g can thus be considered as a static control for system (24). In the following, we will first prove that the spectrum of $H(u_1,u_2)$ is weakly conically connected and has rationally unrelated germs at each intersection point. Then we will apply Theorem 4.1 to conclude the proof of Theorem 6.1.

Let us notice that H_1, H_2 are infinitesimally small with respect to H_0 in the sense of Kato [7]. The eigenfunctions of $a^\dagger a$ are the Hermite functions

$$|n\rangle = \Phi_n(\cdot) = \left(x \mapsto \frac{1}{\sqrt{2^n n! \sqrt{\pi}}} h_n(x) e^{-\frac{x^2}{2}}\right), \quad n \in \mathbb{N},$$

where h_n denotes the nth Hermite polynomial. An analysis of the spectrum of H_0 can be conducted as in [16]. The eigenvectors of H_0 can be obtained by tensorization, namely,

$$\begin{aligned} H_0|n\rangle \otimes e_1 = E^0_{(n,1)}|n\rangle \otimes e_1 &= \Big(\omega(n+\frac{1}{2}) + \frac{\Omega}{2}\Big)|n\rangle \otimes e_1, \\ H_0|n\rangle \otimes e_{-1} = E^0_{(n,-1)}|n\rangle \otimes e_{-1} &= \Big(\omega(n+\frac{1}{2}) - \frac{\Omega}{2}\Big)|n\rangle \otimes e_{-1}. \end{aligned} \tag{25}$$

Lemma 6.2. *Assume that Ω/ω is irrational. Then for every $u_1 \in \mathbb{R}$ and $u_2 \in \mathbb{R}^*$, all eigenvalues of $H(u_1,u_2)$ are non-degenerate.*

Proof. Consider $u_1 \in \mathbb{R}$ and $u_2 \in \mathbb{R}^*$. Define $\delta = \frac{u_2}{\omega}$ and the translated creation and annihilation operators

$$\tilde{a} = a + \delta, \quad \tilde{a}^\dagger = a^\dagger + \delta.$$

The Hamiltonian H can be then written in the translated form

$$\begin{aligned} H(u_1,u_2) = \omega&\left((\tilde{a}-\delta)^\dagger(\tilde{a}-\delta) + \frac{1}{2}\right) + \frac{\Omega}{2}\sigma_z \\ &+ u_1\big((\tilde{a}^\dagger - \delta)\sigma_- + (\tilde{a}-\delta)\sigma_+\big) \\ &+ u_2(\tilde{a}^\dagger + \tilde{a}) - 2u_2\delta \\ = \omega&\left(\tilde{a}^\dagger\tilde{a} + \frac{1}{2} + \delta^2\right) + (u_2 - \delta\omega)(\tilde{a}^\dagger + \tilde{a}) \\ &+ \frac{\Omega}{2}\sigma_z \quad + u_1(\tilde{a}^\dagger\sigma_- + \tilde{a}\sigma_+) - u_1\delta\sigma_x - 2u_2\delta \\ = \omega&\left(\tilde{a}^\dagger\tilde{a} + \frac{1}{2}\right) + \frac{\Omega}{2}\sigma_z + u_1(\tilde{a}^\dagger\sigma_- + \tilde{a}\sigma_+) \\ &- u_1\delta\sigma_x + (\omega\delta^2 - 2u_2\delta). \end{aligned} \tag{26}$$

Here σ_z denotes the Pauli matrix $\left(\begin{smallmatrix} 0 & 1 \\ 1 & 0 \end{smallmatrix}\right)$. Notice that the eigenfunctions of $\tilde{a}^\dagger\tilde{a}$ are given by the translated Hermite functions

$$|\tilde{n}\rangle = \hat{D}^\dagger(\delta)|n\rangle, \tag{27}$$

where $\hat{D}$ is the displaced operator defined as

$$\hat{D}(\alpha) = \exp(\alpha a^\dagger - \alpha^* a), \qquad \alpha \in \mathbb{C}. \tag{28}$$

Define another Hamiltonian

$$\tilde{H}(\tilde{u}_1, \tilde{u}_2) = \omega\left(\tilde{a}\tilde{a}^\dagger + \frac{1}{2}\right) + \frac{\Omega}{2}\sigma_z + \tilde{u}_1(\tilde{a}^\dagger\sigma_- + \tilde{a}\sigma_+) + \tilde{u}_2\sigma_x. \tag{29}$$

Notice that the eigenvalues of $\tilde{H}(\tilde{u}_1, \tilde{u}_2)$ are non-degenerate if $\tilde{u}_1 \neq 0$ and $\tilde{u}_2 \neq 0$ [14] and that

$$H(u_1, u_2) = \tilde{H}(u_1, -u_1\delta) + \omega\delta^2 - 2u_2\delta.$$

When $u_1 = 0$, we have $H(u_1, u_2) = \tilde{H}(0, 0) + \omega\delta^2 - 2u_2\delta$. Since Ω/ω is irrational, the eigenvalues of $H(u_1, u_2)$ are non-degenerate. When $u_1 \neq 0$, $H(u_1, u_2) = \tilde{H}(u_1, -u_1\delta) + \omega\delta^2$ and its eigenvalues are non-degenerate since $u_1 \neq 0$ and $u_1\delta \neq 0$. We can deduce that the eigenvalues of $H(u_1, u_2)$ with $u_2 \neq 0$ are non-degenerate. □

Proposition 6.3. *For almost every $\Omega, \omega > 0$, the spectrum of $H(\cdot, \cdot)$ is weakly conically connected and the eigenvalue intersections are non-overlapping.*

Proof. Consider the Hamiltonian $h(g) := H_0 + gH_1$, $g \in \mathbb{R}$, and define $\Delta = \Omega - \omega$. Set, moreover, δ to be the symbol $+$ if $\Delta \geq 0$ and the symbol $-$ if $\Delta < 0$. According to the spectrum analysis conducted in [16], the eigenvalues of $h(g)$ are given by

$$\begin{aligned} E_{n,\nu}(g) &= \omega(n+1) + \nu\frac{1}{2}\sqrt{\Delta^2 + 4g^2(n+1)}, \\ &\qquad\qquad \text{for } n \in \mathbb{N}, \nu \in \{+,-\}, \\ E_{-1,\delta}(g) &= \frac{\Delta}{2}. \end{aligned}$$

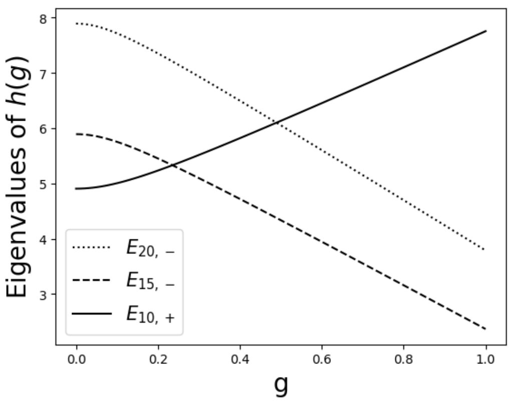
Notice that, for $n \in \mathbb{N}$ and $\nu \in \{+,-\}$, the asymptotic slope of $g \mapsto E_{n,\nu}(g)$ for $g \gg 1$ is $\nu\sqrt{n+1}$.

To relate these eigenvalues to those of H_0 in (25), we distinguish between three situations. At $g = 0$, we have

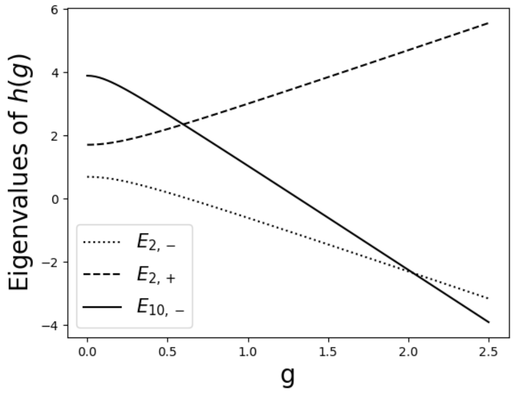
$$\begin{aligned} &\begin{cases} E_{(n,+)}(0) = E^0_{(n,1)} & \text{for } n \geq 0, \\ E_{(n,-)}(0) = E^0_{(n+1,-1)} & \text{for } n \geq -1, \end{cases} && \text{if } \Delta > 0, \\ &\begin{cases} E_{(n,+)}(0) = E^0_{(n+1,-1)} & \text{for } n \geq -1, \\ E_{(n,-)}(0) = E^0_{(n,1)} & \text{for } n \geq 0, \end{cases} && \text{if } \Delta < 0, \\ &E_{(n,\nu)}(0) = E^0_{(n,1)} = E^0_{(n+1,-1)} \quad \text{for } v \in \{+,-\} && \text{if } \Delta = 0, \end{aligned}$$

where, in the last line, $n \geq -1$ if $\nu = +$ and $n \geq 0$ if $\nu = -$. Take $(n, \nu) \in (\mathbb{N} \times \{+,-\}) \cup \{(-1, \delta)\}$.

If $\nu = +$, there exists $m \in \mathbb{N}$ such that $E_{m,-}(0) > E_{n,\nu}(0)$. Since $E_{m,-}(\cdot)$ is decreasing, $E_{n,\nu}(\cdot)$ is increasing and they have uniformly nonzero slope for $g \gg 1$, we have that $E_{m,-}(g) = E_{n,\nu}(g)$ for some $g > 0$. If $\nu = -$ and $n \geq 1$, then for all



(a) Case $(n,\nu) = (10,+)$



(b) Case $(n,\nu) = (10,-)$

Figure 4: Plot of some eigenvalues $E_{(n,\nu)}(\cdot)$ when $(\omega,\Omega) = (2/5,\sqrt{2})$. This illustrative example shows that, for each eigenvalue $E_{n,\nu}(\cdot)$, it is possible to find another eigenvalue $E_{m,\mu}(\cdot)$ such that $E_{n,\nu}(g) = E_{m,\mu}(g)$ at some positive g. For $(n,\nu) = (10,+)$, we can choose, for example, $(m,\mu) = (15,-)$ or $(m,\mu) = (20,-)$. Similarly, for $(n,\nu) = (10,-)$, we can choose $(m,\mu) = (2,-)$ or $(m,\mu) = (2,+)$.

$(m,\mu) \in (\mathbb{N} \times \{+,-\}) \cup \{(-1,\delta)\}$ such that $E_{(m,\mu)}(0) < E_{n,\nu}(0)$, there exists $g \in \mathbb{R}$ such that $E_{(n,\nu)}(g) = E_{(m,\mu)}(g)$. On the other hand, if $\nu = -$ and $n \in \{-1,0\}$, then, for each (m,μ) such that $m \geq 1$ and $\nu = -$, there exists $g \in \mathbb{R}$ such that $E_{(n,\nu)}(g) = E_{(m,\mu)}(g)$. If $\nu = -$, then, for each $(m,\mu) \in (\mathbb{N} \times \{+,-\}) \cup \{(-1,\delta)\}$ such that $E_{(m,\mu)}(0) < E_{n,\nu}(0)$, there exists $g \in \mathbb{R}$ such that $E_{(n,\nu)}(g) = E_{(m,\mu)}(g)$. Illustrative examples are given in Figure 4. Take $(n,\nu),(m,\mu) \in (\mathbb{N} \times \{+,-\}) \cup \{(-1,\delta)\}$. If there exists $g \in \mathbb{R}$ such that $E_{(n,\nu)}(g) = E_{(m,\mu)}(g)$, then $x = g^2/\omega^2$ must solve the polynomial equation

$$x^2 - 2(m+n+2)x + (m-n)^2 - \left(\frac{\Delta}{\omega}\right)^2 = 0.$$

We claim that if $(\Delta/\omega)^2$ is irrational then for every $g \in \mathbb{R}$ there exists at most one pair of distinct eigenvalues $E_{(n,\nu)}(\cdot)$ and $E_{(m,\mu)}(\cdot)$ that intersect at g. Indeed, assume that there exist two pairs of distinct eigenvalues $\{E_{(n,\nu)}(\cdot), E_{(m,\mu)}(\cdot)\}$ and $\{E_{(n',\nu')}(\cdot), E_{(m',\mu')}(\cdot)\}$ intersecting at $g = \bar{g}$, with $m \geq n$ and $m' \geq n'$. Hence, $\bar{x} = (\bar{g}/\omega)^2$ satisfies

$$\bar{x}^2 - 2(m+n+2)\bar{x} + (m-n)^2 - \left(\frac{\Delta}{\omega}\right)^2 = 0, \tag{30}$$

$$\bar{x}^2 - 2(m'+n'+2)\bar{x} + (m'-n')^2 - \left(\frac{\Delta}{\omega}\right)^2 = 0. \tag{31}$$

It is evident from any of these two equations that $\bar{x} \notin \mathbb{Q}$. By subtracting (31) from (30), we obtain that

$$2(m+n-m'-n')\bar{x} = (m-n)^2 - (m'-n')^2.$$

Since $\bar{x} \notin \mathbb{Q}$, we deduce that $m = m'$ and $n = n'$. It can be simply checked that, in addition, $\nu = \nu'$ and $\mu = \mu'$. Thus the two pairs of eigenvalues are equal.

We can deduce that, if $(\Delta/\omega)^2$ is irrational, the spectrum of $h(\cdot) = H(\cdot, 0)$ is connected by non-overlapping eigenvalue intersections between eigenvalues. It can also be proven that the intersecting eigenvalues on $u_2 = 0$ are not tangent to each other. Moreover, by Lemma 6.2 (Ω/ω is irrational since $(\Delta/\omega)^2$ is irrational), all the eigenvalues are simple when $u_2 \neq 0$. Hence all eigenvalue intersections on $u_2 = 0$ are weakly conical with conical direction $\eta = (1, 0)$. We can conclude that for almost every $\Omega, \omega > 0$ the spectrum of $H(\cdot, \cdot)$ is weakly conically connected and the eigenvalue intersections are not overlapping. □

Proof of Theorem 6.1. By Proposition 6.3, for almost every $\Omega, \omega > 0$, the spectrum of $H(\cdot, \cdot)$ is weakly conically connected and has rationally unrelated germs at each intersection point (since the eigenvalue intersectionsare non-overlapping). Since, moreover, the driven Jaynes–Cummings Hamitonian in system (21) coincides with $H(g, u)$, then the conclusion follows from Theorem 4.1. □

Our result shows that for almost every (i.e., in a dense subset with zero-measure complement in $\mathbb{R}$) atomic transition frequency ω, harmonic oscillator frequency Ω, and (vacuum) Rabi frequency g, the corresponding Jaynes-Cummings model (21) driven by the single time-dependent input $u(t)$, which is the amplitude of the resonant drive, is approximately controllable in the unitary group. In other words, this means that any unitary operation can be realized with arbitrary precision at the expense of more complex control.

7 Conclusion

In this paper, we established a spectral condition for the controllability of quantum systems subject to a static field and a time-dependent control. The key assumption is that the spectrum of a two-input quantum system is connected by weakly conical intersections. We applied our result to two physical examples: a model of enantioselective excitation and the driven Jaynes–Cummings model. These examples highlight how our result can be applied to study the controllability of more general scenarios: either by transforming the control of a multi-input system into the control via a static field and a time-dependent control, or by formally interpreting physical parameters in the system as static fields.

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