Controllability of the rotation of a quantum planar molecule

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***Abstract*— We consider the simplest model for controlling the rotation of a molecule by the action of an electric field, namely a quantum planar pendulum.**

This problem consists in characterizing the controllability of a PDE (the Schrödinger equation) on a manifold with nontrivial topology (the circle S^1). The drift has discrete spectrum and its eigenfunctions are trigonometric functions.

Some controllability results for the Schrödinger equation can be applied in this context. We tackle the problem by adapting the general method proposed by some of the authors in a recent paper. This requires, in particular, proving, by perturbation arguments, the non-resonance of the spectrum of the differential operator corresponding to a small constant control. The spectrum of this operator is given by the Mathieu characteristic values and its eigenfunctions are the Mathieu sinus and cosinus.

Our main result says that we have simultaneous approximate controllability separately for the even and odd components of the wave function.

I. Physical motivations and main results

Molecular orientation and alignment are well-established topics in the quantum control of molecular dynamics both from the experimental and theoretical points of view (See [7], [5] and references therein). For linear molecules driven by linearly polarized laser fields in gas phase, alignment means an increased probability direction along the polarization axis whereas orientation requires in addition the same (or opposite) direction as the polarization vector. Such controls have a variety of applications extending from chemical reaction dynamics to surface processing, catalysis and nanoscale design. A large amount of numerical simulations have been done in this domain [5] but the mathematical part remains to explore. From this perspective, the controllability problem is the first question to solve.

In other respect, the rotational molecular dynamics is one of the most important examples of quantum systems with an infinite-dimensional Hilbert space and a discrete spectrum. It appears therefore as a natural model to study the controllability and to extend the results of [3].

We focus in this paper on the control by laser fields of the rotation of a rigid linear molecule confined to a plane. This control problem corresponds to the control of the Schrödinger equation on a circle. We show the approximate controllability of this system up to a trivial symmetry corresponding to the parity of the eigenstates. The controllability is proved for arbitrarily small controls which could be interesting for practical applications. In particular, this means that there exist control strategies which bring the initial state arbitrarily close to states maximizing the molecular orientation [8].

A. The model

We consider a polar linear molecule in its ground vibronic state subject to a nonresonant (with respect to vibronic frequencies) linearly polarized laser field. Within the rigid rotor approximation, the controlled dynamics is governed by the Schrödinger equation on the sphere S^2 which can be written in units such that $\hbar = 1$ as:

$$i\frac{\partial\psi(\theta,\phi,t)}{\partial t} = (-B\Delta - \mu_0 E(t)\cos\theta)\psi(\theta,\phi,t) \quad (1)$$

where B is the rotational constant, μ_0 the permanent dipole moment, Δ the Laplacian on the sphere (called in this context the angular momentum operator), θ the polar angle between the polarization direction and the molecular axis and ϕ the azimuthal angle. The control is given by the electric field E. We neglect in this model the polarizability tensor term which corresponds to the field-induced dipole moment. This approximation is correct if the intensity of the laser field is sufficiently weak. Despite its simplicity, this equation reproduces very well the experimental data on the rotational dynamics of rigid molecules [7].

As a first step in the study of the controllability of rotating molecules, we consider a simple control problem of a linear molecule moving in a plane. This problem can be viewed as the control of the Schrödinger equation on a circle. The dynamics is ruled by the equation:

$$i\frac{\partial\psi(\theta,t)}{\partial t} = \left(-\frac{\partial^2}{\partial\theta^2} + u(t)\cos(\theta)\right)\psi(\theta,t) \quad (2)$$

which is written in normalized coordinates. The angle θ can be interpreted as the angle between the polarization direction and the molecular axis. The control field is now given by the function u. Note that such a system has already been used to understand the dynamics of molecular orientation or alignment (see, e.g., [6]). This simple model allows to solve analytically the question of approximate controllability, which explains its choice in this paper.

B. The main results

In the following we denote by $\psi(T;\psi_0,u)$ the solution at time T of equation (2), corresponding to control u and with initial condition $\psi(0;\psi_0,u)=\psi_0$, belonging to the Hilbert sphere $\mathcal{S}$ of $\mathcal{H}=L^2(S^1,\mathbb{C})$.

Let us split $\mathcal{H}$ as $\mathcal{H}_e \oplus \mathcal{H}_o$, where $\mathcal{H}_e$ (resp. $\mathcal{H}_o$) is the subspace of $\mathcal{H}$ of even (resp. odd) functions. Here the parity of the function is meant with respect to the origin $\theta=0$. Notice that $\mathcal{H}_e$ and $\mathcal{H}_o$ are Hilbert spaces. Let us also denote $\psi=(\psi_e,\psi_o)$, where $\psi_e\in\mathcal{H}_e$ and $\psi_o\in\mathcal{H}_o$.

Our first result is that the system is not controllable since the norms of the even and odd parts of the wave function are conserved.

Proposition 1: For every $u\in L^\infty([0,T],\mathbb{R})$ we have $\|\psi_o(t)\|_{\mathcal{H}}=\|\psi_o(0)\|_{\mathcal{H}}$ and $\|\psi_e(t)\|_{\mathcal{H}}=\|\psi_e(0)\|_{\mathcal{H}}$.

Our main result says that we have simultaneous approximate controllability separately for the even and odd components.

Theorem 1: For every $\psi^0=(\psi_e^0,\psi_o^0)$, $\psi^1=(\psi_e^1,\psi_o^1)$ belonging to the Hilbert sphere of $L^2(S^1,\mathbb{C})$ with $\|\psi_e^0\|_{\mathcal{H}}=\|\psi_e^1\|_{\mathcal{H}}$ and $\|\psi_o^0\|_{\mathcal{H}}=\|\psi_o^1\|_{\mathcal{H}}$ and every $\varepsilon,\delta>0$, there exist $T>0$ and $u\in L^\infty([0,T],(0,\delta])$ such that $\|\psi^1-\psi(T;\psi^0,u)\|<\varepsilon$.

II. MATHEMATICAL FRAMEWORK

A. Notations and definition of solutions

Hereafter $\mathbb{N}$ denotes the set of strictly positive integers and $\mathbb{N}_0=\mathbb{N}\cup\{0\}$ denotes the set of positive integers. Definition 1 below provides the abstract mathematical framework that will be used to formulate the controllability results later applied to the Schrödinger equation (2).

Definition 1: Let $\mathcal{H}$ be a complex Hilbert space and U be a subset of $\mathbb{R}$. Let A,B be two operators on $\mathcal{H}$ with values in $\mathcal{H}$. The control system (A,B,U) is the formal controlled equation

$$\frac{d\psi}{dt}(t)=A\psi(t)+u(t)B\psi(t),\qquad u(t)\in U. \tag{3}$$

We say that (A,B,U) is a skew-adjoint discrete-spectrum control system if the following conditions are satisfied: (H1) A and B are skew-adjoint. B is a bounded operator, while A is possibly unbounded with domain $D(A)$; (H2) there exists an orthonormal basis $(\phi_n)_{n\in\mathbb{N}}$ of $\mathcal{H}$ made of eigenvectors of A.

Under the assumption (H1) well-known results assert that if $u\in L^1([0,T],U)$ then there exists a unique weak (and mild) solution $\psi(\cdot;\psi^0,u)\in\mathcal{C}([0,T],\mathcal{H})$ of (3) satisfying $\psi(0;\psi^0,u)=\psi^0$.

Moreover, if $\psi_0\in D(A)$ and $u\in\mathcal{C}^1([0,T],U)$ then $\psi(t;\psi_0,u)$ is differentiable and it is a strong solution of (3). (See [1] and references therein.)

B. Approximate controllability

It is known that, in general, exact controllability is hopeless for skew-adjoint discrete-spectrum control systems when $\mathcal{H}$ is an infinite dimensional L^2 space (see [1], [9]). Nevertheless, one may sometimes get controllability in a weaker sense.

Definition 2: Let (A,B,U) be a skew-adjoint discrete-spectrum control system. We say that (A,B,U) is approximately controllable if for every ψ^0,ψ^1 belonging to the Hilbert unit sphere $\mathcal{S}$ and every $\varepsilon>0$ there exists an admissible control function $u\in L^\infty([0,T],U)$ such that $\|\psi^1-\psi(T;\psi^0,u)\|<\varepsilon$.

Let, for every $n\in\mathbb{N}$, $i\lambda_n$ denote the eigenvalue of A corresponding to ϕ_n ($\lambda_n\in\mathbb{R}$). The main result of this paper is based on the following abstract controllability result (see [3]).

Theorem 2: Let $\delta>0$ and $(A,B,(0,\delta))$ be a skew-adjoint discrete-spectrum control system. If the elements of the sequence $(\lambda_{n+1}-\lambda_n)_{n\in\mathbb{N}}$ are $\mathbf{Q}$-linearly independent and if $\langle B\phi_n,\phi_{n+1}\rangle\neq 0$ for every $n\in\mathbb{N}$, then $(A,B,(0,\delta))$ is approximately controllable.

Recall that the elements of the sequence $(\lambda_{n+1}-\lambda_n)_{n\in\mathbb{N}}$ are said to be $\mathbf{Q}$-linearly independent if for every $N\in\mathbb{N}$ and $(q_1,\dots,q_N)\in\mathbf{Q}^N\setminus\{0\}$ one has $\sum_{n=1}^N q_n(\lambda_{n+1}-\lambda_n)\neq 0$.

III. ENERGY LEVELS THAT ARE COUPLED BY THE EXTERNAL FIELD

Let $\mathcal{H}=L^2(S^1,\mathbb{C})$. In the following A denotes the operator

$$A=i\partial_\theta^2:\quad \begin{array}{ccc} H^2(S^1,\mathbb{C}) & \to & \mathcal{H}\\ \psi & \mapsto & i\partial_\theta^2\psi\end{array}$$

and $B:\mathcal{H}\to\mathcal{H}$ the multiplication operator by $-i\cos\theta$.

We study in this section which eigenstates of the operator A are coupled by the external field. To this purpose we compute

$$b_{jk}=\int_{S^1}\cos(\theta)\phi_j(\theta)\phi_k(\theta)d\theta,$$

where $(\phi_j)_{j\in\mathbb{N}_0}$ denotes an orthonormal basis of eigenfunctions of A. The infinite matrix $(b_{jk})_{j,k\in\mathbb{N}_0}$ represents the operator iB in the basis $(\phi_j)_{j\in\mathbb{N}_0}$. Two eigenstates are *coupled* if $b_{jk}\neq 0$.

A possible choice of the basis $(\phi_j)_{j\in\mathbb{N}_0}$ is the following:

$$\phi_m(\theta)=\begin{cases}\frac{1}{\sqrt{2\pi}} & \text{if } m=0,\\ \frac{\cos(m\theta/2)}{\sqrt{\pi}} & \text{if } m>0 \text{ even},\\ \frac{\sin((m+1)\theta/2)}{\sqrt{\pi}} & \text{if } m \text{ odd}.\end{cases} \tag{4}$$

Therefore, a simple computation yields

$$b_{jk} = \begin{cases} \frac{1}{\sqrt{2}} & \text{if } \{j,k\} = \{0,2\}, \\ \frac{1}{2} & \text{if } |j-k| = 2 \text{ and } j,k > 0, \\ 0 & \text{otherwise.} \end{cases}$$

The infinite matrix $(b_{jk})_{j,k\in\mathbb{N}_0}$ has the following structure

$$\begin{pmatrix} 0 & 0 & \frac{1}{\sqrt{2}} & 0 & \cdots & & & \\ 0 & 0 & 0 & \frac{1}{2} & 0 & \cdots & & \\ \frac{1}{\sqrt{2}} & 0 & 0 & 0 & \frac{1}{2} & 0 & \cdots & \\ 0 & \frac{1}{2} & 0 & 0 & 0 & \frac{1}{2} & 0 & \cdots \\ \vdots & \ddots & \ddots & \ddots & \ddots & \ddots & \ddots & \ddots \end{pmatrix}.$$

Let $\mathcal{H}_e$ and $\mathcal{H}_o$ be the (closed) Hilbert subspaces of $\mathcal{H}$ generated by $\{\phi_m(\cdot) \mid m \text{ even}\}$ and $\{\phi_m(\cdot) \mid m \text{ odd}\}$ respectively. Notice that $\mathcal{H}_e$ is made of all functions in $\mathcal{H}$ that are even with respect to $\theta = 0$, while $\mathcal{H}_o$ is made of odd functions.

It follows from the computation above that $\mathcal{H}_e$ and $\mathcal{H}_o$ are both invariant subspaces of each operator

$$A_u = A + uB.$$

Hence, they are integral manifolds of the control system (2).

System (2) splits in two decoupled skew-adjoint discrete-spectrum control systems sharing the same control u. Proposition 1 follows.

In the following sections we show how Theorem 2 can be used to prove the approximate controllability in $\mathcal{H}_e$ and $\mathcal{H}_o$. We will then discuss how to control independently in the two spaces with the same $u = u(t)$.

IV. Controllability in $\mathcal{H}_e$ and $\mathcal{H}_o$

Since the eigenvalues of the operator $-\partial_\theta^2$ on $\mathcal{H}$ are $\{m^2\}_{m\in\mathbb{N}_0}$, then Theorem 2 does not apply neither on $\mathcal{H}$ nor on $\mathcal{H}_e$ or $\mathcal{H}_o$.

The idea is then to apply Theorem 2 for $A_\eta = -i(-\partial_\theta^2 + \eta\cos(\theta))$ and $B_\eta = B$ and for u belonging to the control set $(0, \delta - \eta]$.

This corresponds to a reparameterization of the control set that sends u into a new control $u - \eta$. The eigenvalues of A_η can be expressed in terms of Mathieu characteristic values and their asymptotic analysis can be used to prove the existence of some value of η for which the spectrum of A_η is non-resonant.

For every $\eta \in \mathbb{R}$, the operator $iA_\eta = -\partial_\theta^2 + \eta\cos(\theta)$ is self-adjoint and has discrete spectrum. We will denote by $(\lambda_m(\eta))_{m\in\mathbb{N}_0}$ the non-decreasing sequence of its eigenvalues counted according to their multiplicity and by $(\phi_m(\cdot,\eta))_{m\in\mathbb{N}_0}$ a sequence of corresponding eigenfunctions. Therefore

$$(-\partial_\theta^2 + \eta\cos(\theta))\phi_m(\theta,\eta) = \lambda_m(\eta)\phi_m(\theta,\eta). \quad (5)$$

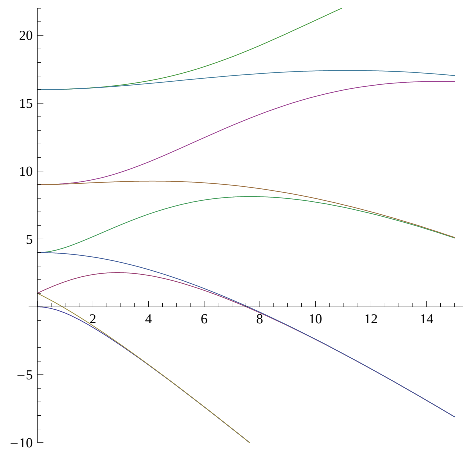

Fig. 1. Mathieu characteristic values

The relation between the functions ϕ_m and the Mathieu functions is explained below.

A. The Mathieu equation

Fix $a, q \in \mathbb{R}$ and let us consider the Mathieu equation:

$$(-\partial_z^2 + 2q\cos(2z))y = ay. \quad (6)$$

Solutions to this equation are the so called Mathieu cosinus and Mathieu sinus and are denoted by $C(a,q,z)$ and $S(a,q,z)$. For certain values of a, called characteristic values, Mathieu cosinus and Mathieu sinus are 2π periodic functions. More precisely for every q there exist two increasing sequences $(a_k(q))_{k\in\mathbb{N}_0}$ and $(b_k(q))_{k\in\mathbb{N}}$ such that $C(a,q,z)$ is 2π periodic if and only if $a = a_k(q)$ for some k and $S(a,q,z)$ is 2π periodic if and only if $a = b_k(q)$ for some k. The graphs of the first Mathieu characteristic values is plotted in Figure 1.

Notice that for $k \in \mathbb{N}_0$, $a_k(0) = b_k(0) = k^2$ and for $q > 0$, $k_1 \neq k_2$, the values $a_{k_1}(q)$ and $b_{k_2}(q)$ are all pairwise distinct and $a_k(q) > b_k(q)$.

Periodic cosinus and sinus of Mathieu are usually denoted by $ce_k(z,q)$ and $se_k(z,q)$. Notice that

$$C(a,0,z) = \cos(\sqrt{a}x), \quad S(a,0,z) = \sin(\sqrt{a}x). \quad (7)$$

An important point, that will be used later, is that ce_k and se_k are π-periodic if and only if k is even.

When the equation (6) is formulated on S^1, it admits a nontrivial solution if and only if a coincides with one characteristic value.

If in equation (5) we set $\theta = 2z$, we get the equation

$$(-\partial_z^2 + 4\eta\cos(2z))\phi_m(2z,\eta) = 4\lambda_m(\eta)\phi_m(2z,\eta).$$

Since θ is 2π-periodic, z is π-periodic. Hence $z \mapsto \phi_m(2z,\eta)$ is a π-periodic solution of the Mathieu

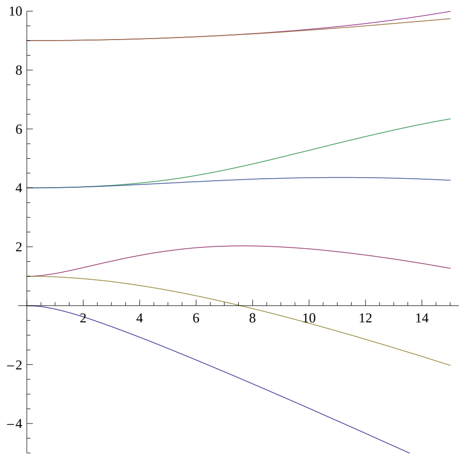

Fig. 2. Graph of $\lambda_m(\eta)$ for $m = 0, 1, 2, 3, 4, 5, 6, 7$.

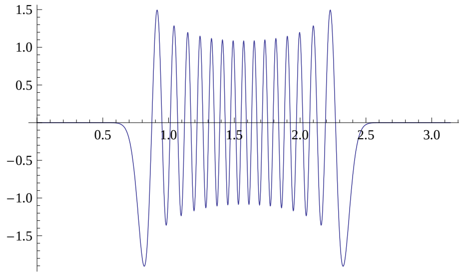

Fig. 3. Graph of $\phi_{30}(\theta, 1500)$

equation (6) with $a = 4\lambda_m(\eta)$ and $q = 2\eta$. It follows that the spectrum of iA_η is given by

$$\lambda_m(\eta) = \begin{cases} \frac{a_m(2\eta)}{4} & \text{if } m \text{ even,} \\ \frac{b_{m+1}(2\eta)}{4} & \text{if } m \text{ odd.} \end{cases}$$

Moreover,

$$\phi_m(\theta, \eta) = \begin{cases} \frac{ce_m(\theta/2, 2\eta)}{\|ce_m(\cdot/2, 2\eta)\|_{L^2(S^1)}} & \text{if } m \text{ even,} \\ \frac{se_{m+1}(\theta/2, 2\eta)}{\|se_{m+1}(\cdot/2, 2\eta)\|_{L^2(S^1)}} & \text{if } m \text{ odd.} \end{cases}$$

Notice that $\phi_m(\theta, 0) = \phi_m(\theta)$ where ϕ_m was defined in (4)

The graphs of the first eigenvalues is plotted in Figure 2. while the graph of two eigenfunctions is plotted in figures 3 and 4.

B. Asymptotic development of Mathieu characteristic functions for η large

Let us recall some facts on the dependence of $\lambda_j(\eta)$ and $\phi_j(\theta, \eta)$ on η.

It is well known that they are analytic with respect to η. Moreover for every $j \in \mathbb{N}$, for η sufficiently

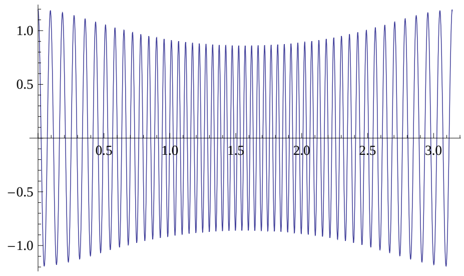

Fig. 4. Graph of $\phi_{100}(\theta, 1500)$

large, the following expansion of λ_j in Laurent series holds true

$$\lambda_j(\eta) = \sum_{k=0}^{\infty} P_k(j) \left(\frac{1}{\sqrt{\eta}}\right)^{k-2}, \tag{8}$$

where each P_k is a real polynomial of degree k. See [4].

The expansion (8) is relevant to our arguments even if the control set $(0, \delta]$ is small. This is because of the analytic dependence on η.

C. Non-resonance of the perturbed spectrum

We prove in this section that the spectrum of A_η is nonresonant for η in a subset of full measure of $\mathbb{R}^+$.

Lemma 1: Let $N \in \mathbb{N}$, $z_1, \ldots, z_N \in \mathbb{Z}$ and assume that $\sum_{m=1}^{N} z_m \lambda_m(\eta) = 0$ for every η. Then $z_1 = \cdots = z_N = 0$.

Proof: Taking the first N terms of the expansion of the resonance relation $\sum_{m=1}^{N} z_m \lambda_m(\eta) = 0$ according to (8), we get

$$\sum_{k=0}^{N-1} \left\{ \left(\sum_{m=1}^{N} z_m P_k(m) \right) \eta^{\frac{2-k}{2}} \right\} = O(\eta^{\frac{4-N}{2}}).$$

Hence,

$$\sum_{m=1}^{N} z_m P_j(m) = 0, \qquad j = 0, \ldots, N-1. \tag{9}$$

Fix $\bar{m} \in \{0, \ldots, N-1\}$ and define

$$\bar{P}(x) = \frac{1}{\bar{m}} \Pi_{m=0}^{\bar{m}-1}(x-m) \Pi_{\bar{m}+1}^{N-1}(x-m).$$

Notice that $\bar{P}$ is a polynomial of degree $N-1$ and therefore it can be written as linear combination of $P_0, \ldots, P_{N-1}$ (recall that $\deg(P_k) = k$).

Let $c_0, \ldots, c_{N-1} \in \mathbb{R}$ be such that

$$\bar{P} = \sum_{k=0}^{N-1} c_k P_k.$$

Then

$$\begin{aligned}\bar{z} &= \sum_{m=1}^{N} z_m \bar{P}(m) \\ &= \sum_{m=1}^{N} z_m \sum_{k=0}^{N-1} c_k P_k(m) \\ &= \sum_{k=0}^{N-1} c_k \sum_{m=1}^{N} z_m P_k(m) \\ &= 0\end{aligned}$$

where the last equality follows from (9). ∎

From Lemma (1) and the analyticity of $\lambda_m(\eta)$ with respect to η it follows that, given a finite nonzero sequence of rational coefficients $q_1, \ldots, q_N$, the relation $\sum_{m=1}^{N} q_m \lambda_m(\eta) = 0$ holds for at most countably many $\eta \in \mathbb{R}$. Since the set of possible choices of $N, q_1, \ldots, q_N$ is countable, then the spetrum of A_η is nonresonant except for countably many η.

D. Connectedness for the operator B on $\mathcal{H}_e$ and $\mathcal{H}_o$

In order to apply Theorem 2 to (2) on $\mathcal{H}_e$ and $\mathcal{H}_o$ we are left to prove that for some η for which the spectrum of A_η is nonresonant,

$$\langle B\phi_m(\cdot,\eta), \phi_{m+2}(\cdot,\eta)\rangle \neq 0$$

for every $m \in \mathbb{N}_0$. This is a consequence of the analytic dependence of $\phi_m(\cdot,\eta)$ on η and the fact that

$$\langle B\phi_m(\cdot,0), \phi_{m+2}(\cdot,0)\rangle \neq 0,$$

as proved in Section III. We have proved the approximate controllability among wave functions with the same parity.

V. Simultaneous control

To conclude the proof of Theorem 2, we need an argument of simultaneous independent controlability. The following result is a slight adaptation of [3, Theorem 2.4] (see also [2]).

Theorem 3: Let H_1 and H_2 be two Hilbert spaces with respective Hilbert bases $(\phi_j^1)_{j\in\mathbb{N}}$ and $(\phi_j^2)_{j\in\mathbb{N}}$. Let $(A_1, B_1, [0,\delta))$ and $(A_2, B_2, [0,\delta))$ be two skew-adjoint discrete-spectrum control systems on H_1 and H_2 respectively such that A_i is diagonal in the basis $(\phi_j^i)_{j\in\mathbb{N}}$ for $i = 1, 2$. If the concatenation of the spectra of A_1 and A_2 is a $\mathbf{Q}$-linearly independent family and if for every j in $\mathbb{N}$, $i = 1, 2$, $\langle B\phi_j^i, \phi_{j+1}^i\rangle \neq 0$, then the two systems $(A_1, B_1, [0,\delta))$ and $(A_2, B_2, [0,\delta))$ are simultaneously approximately controllable, that is, for every (ψ_0^1, ψ_0^2) and (ψ_1^1, ψ_1^2) in $H_1 \times H_2$ such that $\|\psi_0^i\| = \|\psi_1^i\|$ $i = 1, 2$, for every $\delta, \epsilon > 0$, there exists an admissible control $u \in L^\infty([0,T], [0,\delta))$ steering the system $(A_i, B_i, [0,\delta))$ from ψ_0^i to an ϵ-neighborhood of ψ_1^i, $i = 1, 2$.

Fix η such that the spectrum of $-\partial_\theta^2 + \eta\cos(\theta)$ is non-resonant and each $B_e^{(n)}$, $B_o^{(n)}$ is connected. We define $A_{\eta,e}$ and $B_{\eta,e}$ (resp.$A_{\eta,o}$ and $B_{\eta,o}$) on a subdomain of $\mathcal{H}_e$ (resp.$\mathcal{H}_o$) as the restrictions of A_η and B_η to $\mathcal{H}_e$ (resp.$\mathcal{H}_o$). The two skew-adjoint discrete-spectrum control systems $(A_{\eta,e}, B_{\eta,e}, [0,\delta))$ and $(A_{\eta,o}, B_{\eta,o}, [0,\delta))$ satisfy the hypotheses of Theorem 3. Theorem 2 follows.

VI. Conclusion

In this paper we consider the problem of controlling the orientation of a planar molecule by the action of a constant (in space) external field. We characterize completely the controllability properties of this system. Namely we proved that the system is not approximately controllable since the state space is the direct sum of two nontrivial integral manifolds (the spaces of even and odd wave functions). In a sense, this happens to be the only obstacle to controllability, since the even and the odd part of the wave function can be controlled simultaneously, keeping constant their relative norm.

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