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Abstract: In this paper we study the controllability properties of the quantum rotational dynamics of a 3D symmetric molecule, with electric dipole moment not collinear to the symmetry axis of the molecule (that is, an *accidentally symmetric-top*). We control the dynamics with three orthogonally polarized electric fields. When the dipole has a nonzero component along the symmetry axis, it is known that the dynamics is approximately controllable. We focus here our attention to the case where the dipole moment and the symmetry axis are orthogonal (that is, an *orthogonal* accidentally symmetric-top), providing a description of the reachable sets.

Keywords: Bilinear control, controllability, distributed-parameter systems, partial differential equations, quantum angular momentum, reachable states, Schrödinger equation.

1. INTRODUCTION

Rotational dynamics of molecules plays a relevant role in quantum physics and chemistry, both from a theoretical and an experimental viewpoint (Biedenharn and Louck (1981); Varshalovich et al. (1988); Cook and Gordy (1984)).

The controllability properties of the rotational molecular dynamics is also important in several engineering applications, starting from well-established protocols such as enantio- and state- selectivity in chiral molecules (Averbukh et al. (2018); Giesen et al. (2019)), and going forward to new applications in quantum information (Yu et al. (2019)). For a general overview about molecular dynamics and controllability, see, for instance, Koch et al. (2019).

In this paper, we focus our attention on some controllability properties of the Schrödinger equation for a rotating symmetric-top molecule, driven by three orthogonally polarized electric fields. The control of the Schrödinger equation has attracted substantial interest in the last 15 years. For the finite-dimensional case, see, D'Alessandro (2008); Altafini and Ticozzi (2012) and references therein. For the control of the Schrödinger equation as a PDE see, for instance, Mirrahimi and Rouchon (2004); Beauchard and Coron (2006); Chambrion et al. (2009); Nersesyan (2010); Boscain et al. (2012, 2014, 2015, 2019); Caponigro and Sigalotti (2018)). In particular, the control of the quantum angular momentum has been studied in Boscain et al. (2012) for a planar molecule, in Boscain et al. (2014) for a linear 3D molecule.

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In Boscain et al. (2019), the controllability of a symmetric-top, driven by three orthogonally polarized electric fields, is characterized in terms of the dipole moment position fixed inside the molecule: if the dipole is either (i) collinear or (ii) orthogonal to the symmetry axis, controllability is proved to fail, since two different symmetries arise in the Schrödinger equation; if (iii) the dipole is neither collinear nor orthogonal to the symmetry axis, then approximate controllability is proved to hold using the block-wise Galerkin control condition, introduced in Boscain et al. (2019), and recalled here in Section 2, Definition 4.

Here, we describe and explain how to compute the reachable sets in case (ii), i.e., when the dipole is orthogonal to the symmetry axis of the molecule, that is, we find the invariant subspaces of the ambient Hilbert space and we apply the Galerkin control condition to prove that the Schrödinger equation is approximate controllable on those invariant subspaces.

Notice that, while the non-controllability of case (i) is somehow expected since the corresponding classical system is not controllable, the non-controllability of case (ii) is more surprising since the corresponding classical system is controllable. This is due to the fact that in case (i) there is a classical symmetry which is also a quantum one, while in case (ii) the quantum symmetry does not correspond to any classical one (Boscain et al. (2019)).

1.1 Structure of the paper

The paper is organized as follows: in Section 2 we recall the controllability and tracking results about the abstract multi-imput Schrödinger equation (Boscain et al. (2019)), in particular the block-wise Galerkin control condition (Definition 4) and the tracking Theorem 1; in Section 3 we formulate the problem of controlling the rotational

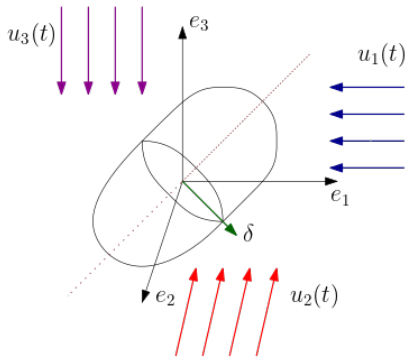

Fig. 1. Inertia ellipsoid of a symmetric-top with dipole δ orthogonal to the symmetry axis, controlled through three polarized electric fields in the directions e_1, e_2, e_3 of a fixed orthogonal frame.

dynamics of a symmetric-top molecule; in Section 4 we describe the reachable sets of our problem (Theorem 3).

2. CONTROLLABILITY OF THE MULTI-INPUT SCHRÖDINGER EQUATION

Let $l \in \mathbb{N}$, $a > 0$, and $U := [-a, a]^l$. Let $\mathcal{H}$ be an infinite-dimensional Hilbert space with scalar product $\langle \cdot, \cdot \rangle$ (linear in the first entry and conjugate linear in the second), $H, B_1, \dots, B_l$ be (possibly unbounded) self-adjoint operators on $\mathcal{H}$, with domains $D(H), D(B_1), \dots, D(B_l)$. We consider the controlled Schrödinger equation

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

where $u(t) \in U$.

Definition 1.
- We say that H has the property ($\mathbb{A}1$) if H has discrete spectrum with infinitely many distinct eigenvalues (possibly degenerate).
  Denote by $\mathcal{B}$ a Hilbert basis $(\phi_k)_{k\in\mathbb{N}}$ of $\mathcal{H}$ made of eigenvectors of H associated with the family of eigenvalues $(\lambda_k)_{k\in\mathbb{N}}$ and let $\mathcal{L}$ be the set of finite linear combination of eigenstates, that is,
  $$\mathcal{L} = \cup_{k\in\mathbb{N}} \mathrm{span}\{\phi_1, \dots, \phi_k\}.$$
- We say that $(H, B_1, \dots, B_l, \mathcal{B})$ has the property ($\mathbb{A}2$) if $\phi_k \in D(B_j)$ for every $k \in \mathbb{N}$, $j = 1, \dots, l$.
- We say that $(H, B_1, \dots, B_l, \mathcal{B})$ has the property ($\mathbb{A}3$) if
  $$H + \sum_{j=1}^{l} u_j B_j : \mathcal{L} \to \mathcal{H}$$
  is essentially self-adjoint for every $u \in U$.
- We say that $(H, B_1, \dots, B_l, \mathcal{B})$ has the property ($\mathbb{A}$) if H has the property ($\mathbb{A}1$) and $(H, B_1, \dots, B_l, \mathcal{B})$ the properties ($\mathbb{A}2$) and ($\mathbb{A}3$).

If $(H, B_1, \dots, B_l, \mathcal{B})$ has the property ($\mathbb{A}$) then, for every $(u_1, \dots, u_l) \in U$, $H + \sum_{j=1}^{l} u_j B_j$ generates a subgroup $e^{-\mathrm{i}t(H+\sum_{j=1}^{l} u_j B_j)}$ of the group of unitary operators $U(\mathcal{H})$. It is therefore possible to define the propagator $\Gamma^u(T)$ at time T of system (1) associated with a l-uple of piecewise constant controls $u(\cdot) = (u_1(\cdot), \dots, u_l(\cdot))$ by composition of flows of the type $e^{-\mathrm{i}t(H+\sum_{j=1}^{l} u_j B_j)}$. If, moreover, $B_1, \dots, B_l$ are bounded operators then the definition can be extended by continuity to every L^∞ control law (see Ball et al. (1982)).

Denote by $\mathcal{S} := \{\psi \in \mathcal{H} \mid ||\psi|| = 1\}$ the unit sphere of $\mathcal{H}$. Let $(H, B_1, \dots, B_l, \mathcal{B})$ have the property ($\mathbb{A}$).

Definition 2.
- Given $\psi_0, \psi_1 \in \mathcal{S}$, we say that ψ_1 is reachable from ψ_0 if there exist a time $T > 0$ and a piecewise constant control function $u : [0, T] \to U$ such that $\psi_1 = \Gamma^u(T)(\psi_0)$. We define
  $$\mathrm{Reach}(\psi_0) = \{\psi_1 \in \mathcal{S} \mid \psi_1 \text{ is reachable from } \psi_0\}.$$
- We say that (1) is approximately controllable if for every $\psi_0 \in \mathcal{S}$ the set $\mathrm{Reach}(\psi_0)$ is dense in $\mathcal{S}$.

We introduce the notion of module-tracker (m-tracker, for brevity) that is, a system for which any given curve can be tracked up to (relative) phases. The identification up to phases of elements of $\mathcal{H}$ in the basis $\mathcal{B} = (\phi_k)_{k\in\mathbb{N}}$ can be accomplished by the projection

$$\mathcal{M} : \psi \mapsto \sum_{k\in\mathbb{N}} |\langle \phi_k, \psi \rangle| \phi_k.$$

Definition 3. Let $(H, B_1, \dots, B_l, \mathcal{B})$ have the property ($\mathbb{A}$). We say that system (1) is an *m-tracker* if, for every $r \in \mathbb{N}$, $\psi_1, \dots, \psi_r$ in $\mathcal{H}$, $\widehat{\Gamma} : [0, T] \to U(\mathcal{H})$ continuous with $\widehat{\Gamma}(0)$ equal to the identity operator, and $\epsilon > 0$, there exists an invertible increasing continuous function $\tau : [0, T] \to [0, T_\tau]$ and a piecewise constant control $u : [0, T_\tau] \to U$ such that

$$\|\mathcal{M}(\widehat{\Gamma}(t)\psi_k) - \mathcal{M}(\Gamma(\tau(t))^u \psi_k)\| < \epsilon, \qquad k = 1, \dots, r$$

for every $t \in [0, T_\tau]$.

Remark 1. We recall that if system (1) is an m-tracker, then it is also approximately controllable, as noticed in (Boscain et al., 2014, Remark 2.9).

Let us assume now that there exists a self-adjoint (possibly unbounded) operator J, with domain $D(J)$, such that

- $\mathcal{L} \subset D(J)$,
- J has discrete spectrum and infinitely many distinct eigenvalues (possibly degenerate),
- $J(\mathcal{L}) \subset D(H)$ and J commutes with H on $\mathcal{L}$.

Then, there exists an Hilbert basis, which we still denote by $\mathcal{B}$, made by eigenvectors of H and J at the same time, that is, if $\phi_k^j \in \mathcal{B}$, then $H\phi_k^j = \lambda_k^j \phi_k^j$ and $J\phi_k^j = \mu^j \phi_k^j$. Denote by m_j the multiplicity of the eigenvalue μ^j, then we can label the basis $\mathcal{B}$ as $(\phi_k^j \mid j \in \mathbb{N}, k = 1, \dots, m_j)$.

Consider $\mathcal{H}_j := \mathrm{span}\{\phi_k^j \mid k = 1, \dots, m_j\}$, $j \in \mathbb{N}$, which are the eigenspaces of the operator J, and let us denote by

$$\Sigma_{j,j+1} = \{|\lambda_{k'}^{l'} - \lambda_k^l| \mid l, l' \in \{j, j+1\}, k' = 0, \dots, m_{l'}, \; k = 0, \dots, m_l\}$$

the spectral gaps in the subspace $\mathcal{H}_j \oplus \mathcal{H}_{j+1}$. Moreover, for every $j \in \mathbb{N}$, define the orthogonal projections

$$\Pi_{j,j+1} : \mathcal{H} \ni \psi \mapsto \sum_{l=j,j+1,\ k=1,\dots,m_l} \langle \phi_k^l, \psi \rangle \phi_k^l \in \mathcal{H},$$

$$\Pi_{0,j+1} : \mathcal{H} \ni \psi \mapsto \sum_{l=0,\dots,j+1,\ k=1,\dots,m_l} \langle \phi_k^l, \psi \rangle \phi_k^l \in \mathcal{H},$$

and we define $B_i^{(j,j+1)} := \Pi_{j,j+1} B_i \Pi_{j,j+1}$, and $B_i^{(j+1)} := \Pi_{0,j+1} B_i \Pi_{0,j+1}$, for every $i = 1, \dots, l$.

Next we consider the spectral gap excitation, given by the operator

$$\mathcal{E}_\sigma(M) = \\ (M_{(l,k),(l',k')}\delta_{\sigma,|\lambda^{l'}_{k'}-\lambda^l_k|})_{(l,k),(l',k')=(j,1),\dots,(j+1,m_{j+1})}$$

defined for $\sigma \geq 0$, every $j \in \mathbb{N}$ and every $(m_j + m_{j+1}) \times (m_j + m_{j+1})$ matrix M, where $\delta_{l,k}$ is the Kronecker delta. We are implicitly using the lexicographic correspondence between the sets $\{(l,k) \mid l = j, j+1, k = 1, \dots, m_l\}$ and $\{1, \dots, m_j + m_{j+1}\}$ to label the matrix M.

Denote by $\mathfrak{u}(j, j+1)$ the Lie algebra associated with the Lie group of the unitary operators on the space $\mathcal{H}_j \oplus \mathcal{H}_{j+1}$, that is, $\mathfrak{u}(j, j+1) := \mathfrak{u}(m_j + m_{j+1})$, and define

$$\Xi^0_{j,j+1} = \{(\sigma, i) \in \Sigma_{j,j+1} \times \{1, \dots, l\} \mid \exists M \in \mathfrak{u}(j, j+1) \text{ s.t.} \\ \mathcal{E}_\sigma(\mathrm{i}B^{(h)}_i) = \left[\begin{array}{c|c|c} 0 & 0 & 0 \\ \hline 0 & M & 0 \\ \hline 0 & 0 & 0 \end{array}\right] \text{ for every } h > j \ \}, \tag{2}$$

and

$$\Xi^1_{j,j+1} = \{(\sigma, i) \in \Sigma_{j,j+1} \times \{1, \dots, l\} \mid \exists M \in \mathfrak{u}(j, j+1) \text{ s.t.} \\ \mathcal{E}_\sigma(\mathrm{i}B^{(h)}_i) = \left[\begin{array}{c|c|c} \star & 0 & \star \\ \hline 0 & M & 0 \\ \hline \star & 0 & \star \end{array}\right] \text{ for every } h > j \ \}, \tag{3}$$

where M is in the block $j, j+1$. In the set $\Xi^1_{j,j+1}$ some spectral gaps resonances are allowed, while the set $\Xi^0_{j,j+1}$ is made by totally non-resonant gaps.

Finally, for every $\xi \in S^1 \subset \mathbb{C}$, consider the matrix operator W_ξ such that

$$(W_\xi(M))_{(l,k),(l',k')} = \begin{cases} \xi M_{(l,k),(l',k')}, & \lambda^l_k < \lambda^{l'}_{k'}, \\ 0, & \lambda^l_k = \lambda^{l'}_{k'}, \\ \bar{\xi} M_{(l,k),(l',k')}, & \lambda^l_k > \lambda^{l'}_{k'}. \end{cases} \tag{4}$$

Let

$$\nu^s_{j,j+1} := \{W_\xi(\mathcal{E}_\sigma(\mathrm{i}B^{(j,j+1)}_i)) \mid (\sigma, i) \in \Xi^s_{j,j+1}, \sigma \neq 0, \\ \xi \in S^1\}, \quad s = 0, 1. \tag{5}$$

Notice that $\nu^0_{j,j+1} \subset \nu^1_{j,j+1} \subset \mathfrak{su}(j, j+1)$.

Finally, denote by $\mathrm{Lie}(\nu^s_{j,j+1})$ the Lie algebra generated by the matrices in $\nu^s_{j,j+1}$, $s = 0, 1$, and we define

$$\mathcal{I}_{j,j+1} := \{A, [[C_1, C_2], \dots, C_n] \dots] \mid A \in \mathrm{Lie}(\nu^0_{j,j+1}), \\ C_j \in \mathrm{Lie}(\nu^s_{j,j+1}), s = 0, 1, \text{ and } \exists\, i \in \{1, \dots, n\} \text{ s.t.} \\ C_i \in \mathrm{Lie}(\nu^0_{j,j+1}), \ n \in \mathbb{N}\}.$$

Definition 4. Let $(H, B_1, \dots, B_l, \mathcal{B})$ have the property $(\mathbb{A})$. We say that (1) fulfills the *block-wise Galerkin control condition* if for every $j \in \mathbb{N}$ one has

$$\mathrm{Lie}(\mathcal{I}_{j,j+1}) = \mathfrak{su}(j, j+1).$$

Then we have the following (proved in Boscain et al. (2019))

Theorem 1. Assume that $(\mathbb{A})$ holds true. If the block-wise Galerkin control condition holds then (1) is an m-tracker. In particular, it is approximately controllable.

3. SYMMETRIC TOP MOLECULE

We recall in this section some general facts about Wigner functions and the theory of angular momentum in quantum mechanics (see, for instance, Biedenharn and Louck (1981); Varshalovich et al. (1988); Cook and Gordy (1984)).

The rotational dynamics for a molecule with moments of inertia I_1, I_2, I_3 is described by the Hamiltonian

$$H := \frac{1}{2}\Big(\frac{P_1^2}{I_1} + \frac{P_2^2}{I_2} + \frac{P_3^2}{I_3}\Big), \tag{6}$$

where the P_i are the components of the angular momentum expressed in a moving frame which diagonalizes the inertia tensor of the molecule. In (6), the P_i are seen as differential operators acting on the Hilbert space $\mathcal{H} := L^2(SO(3))$, self-adjoint with respect to the Haar measure of $SO(3)$. Note that there exists a self-adjoint operator which commutes with H for every $I_1, I_2, I_3 > 0$, namely, the angular momentum operator $J := P_1^2 + P_2^2 + P_3^2$.

In what follows, we shall restrict our attention to the symmetric-top, that is, the case $I_1 = I_2$. Under such an assumption, a closed expression for the spectrum and the eingenfunctions of H is known: we use Euler's angles $(\alpha, \beta, \gamma) \in [0, 2\pi) \times [0, \pi] \times [0, 2\pi)$ to describe the configuration space of the molecule, and with respect to this parametrization the eigenstates of the rotational Hamiltonian H and of the angular momentum J, seen as self-adjoint operators acting on $\mathcal{H}$, are given by the so-called Wigner functions ((Varshalovich et al., 1988, Chapter 4), (Biedenharn and Louck, 1981, Sections 3.8,3.9))

$$D^j_{k,m}(\alpha, \beta, \gamma) := e^{\mathrm{i}(m\alpha + k\gamma)} d^j_{k,m}(\beta), \\ j \in \mathbb{N}, \quad k, m = -j, \dots, j \tag{7}$$

where

$$d^j_{k,m}(\beta) := \\ n_{j,k,m} \sin\Big(\frac{\beta}{2}\Big)^{|k-m|} \cos\Big(\frac{\beta}{2}\Big)^{|k+m|} F\Big(\sin\Big(\frac{\beta}{2}\Big)^2\Big).$$

The function $\beta \mapsto F\Big(\sin\Big(\frac{\beta}{2}\Big)^2\Big)$ is a hypergeometric series and $n_{j,k,m}$ is a normalizing factor. As a summary, we have the following orthonormal decomposition of the Hilbert space

$$\mathcal{H} = \overline{\mathrm{span}}\{D^j_{k,m} \mid j \in \mathbb{N}, k, m = -j, \dots, j\},$$

where $\overline{\mathrm{span}}$ denotes the closure of span in $L^2(SO(3))$.

The spectrum of H is discrete and given by

$$HD^j_{k,m} = \Big(\frac{j(j+1)}{2I_2} + \Big(\frac{1}{2I_3} - \frac{1}{2I_2}\Big)k^2\Big) D^j_{k,m} \\ =: E^j_k D^j_{k,m}. \tag{8}$$

Thus, the rotational energy level E^j_k of a symmetric-top molecule is $(2j+1)$-degenerate with respect to m and also 2-degenerate with respect to k, when $k \neq 0$.

The spectrum of J is discrete and given by

$$JD^j_{k,m} = j(j+1)D^j_{k,m},$$

and the multiplicity of the eigenvalue $j(j+1)$ is $(2j+1)^2$.

To control the rotation of the molecule, we consider three electric fields orthogonally polarized in the directions

e_1, e_2, e_3 of the fixed frame. The interaction Hamiltonian between the dipole δ inside the molecule and the external electric field in the direction e_i, $i = 1, 2, 3$, is given by the Stark effect (Cook and Gordy, 1984, Chapter 10)

$$B_i(\alpha, \beta, \gamma) = -\langle R(\alpha, \beta, \gamma)\delta, e_i \rangle,$$

seen as a multiplicative self-adjoint operator acting on $L^2(SO(3))$. Thus, the controlled rotational Schrödinger equation reads

$$\begin{aligned} &\mathrm{i}\frac{\partial}{\partial t}\psi(\alpha, \beta, \gamma; t) = H\psi(\alpha, \beta, \gamma; t) + \\ &\sum_{l=1}^{3} u_l(t) B_l(\alpha, \beta, \gamma)\psi(\alpha, \beta, \gamma; t), \\ &\psi(t) \in L^2(SO(3)), u(t) \in [-a, a]^3. \end{aligned} \tag{9}$$

4. ORTHOGONAL ACCIDENTALLY SYMMETRIC TOP MOLECULE

In this section we study the controllability properties for the rotational dynamics of a symmetric-top molecule, driven by three orthogonally polarized electric fields, with electric dipole δ orthogonal to the symmetry axis of the molecule.

We denote by a_1, a_2, a_3 the moving frame, where a_3 is the symmetry axis of the molecule. In the frame a_1, a_2, a_3, the dipole δ can be written

$$\delta = (\delta_1, \delta_2, 0)^t, \quad \delta_1 \neq 0 \text{ or } \delta_2 \neq 0. \tag{10}$$

System (9) together with the assumption (10) is called the *orthogonal accidentally symmetric-top*.
By rotational symmetry around the symmetry axis, we can assume that $\delta_2 = 0$.

In Boscain et al. (2019) the non-controllability of the orthogonal accidentally symmetric-top is proved, by looking at a particular symmetry which arises in (9) when δ satisfies (10). Here, we shall describe the reachable sets of the orthogonal accidentally symmetric-top, that is, the invariant subspaces where (9) turns out to be approximately controllable when δ satisfies (10).

The symmetry which prevents the system to be controllable is more evident once one has adopted the symmetrized Wigner functions, also called the Wang functions, as eigenstates for the rotational Hamiltonian H, which are

$$\begin{cases} S^j_{k,m,\gamma} := \dfrac{1}{\sqrt{2}}(D^j_{k,m} + (-1)^\gamma D^j_{-k,m}), & k = 1, \dots, j \\ S^j_{0,m,0} = D^j_{0,m}, & k = 0 \end{cases}$$

$j \in \mathbb{N}, m = -j, \dots, j$ and $\gamma = 0, 1$.

Then we have the following result (Boscain et al. (2019)):

Theorem 2. The parity of $j+\gamma+k$ is conserved in the controlled motion of the orthogonal accidentally symmetric-top.

Since one can decompose the ambient Hilbert space $\mathcal{H} = L^2(SO(3))$ as $\mathcal{H} = \mathcal{H}_e \oplus \mathcal{H}_o$, where

$$\mathcal{H}_e := \overline{\text{span}}\{S^j_{k,m,\gamma} \mid j + \gamma + k \text{ even}\}$$

and

$$\mathcal{H}_o := \overline{\text{span}}\{S^j_{k,m,\gamma} \mid j + \gamma + k \text{ odd}\},$$

Theorem 2 says that $\mathcal{H}_e$ and $\mathcal{H}_o$ are two invariant subspaces for every propagator of (9), when $I_1 = I_2$ and $\delta_3 = 0$. We are going to show that the restriction of (9) to $\mathcal{H}_e$ or $\mathcal{H}_o$ gives an approximately controllable system.

As a technical assumption, we need the moments of inertia I_1, I_2, I_3 of the molecule to satisfy the following symmetric non-resonant hypothesis, introduced in Boscain et al. (2019)

$$I_1 = I_2, \quad I_3/(I_2 - I_3) \in \mathbb{R} \setminus \mathbb{Q}. \tag{11}$$

Theorem 3. Let I_1, I_2, I_3 satisfy (11) and δ satisfies (10).

- If $j + \gamma + k$ is even, then
$$\overline{\text{Reach}(S^j_{k,m,\gamma})} = \mathcal{H}_e \cap \mathcal{S}.$$
- If $j + \gamma + k$ is odd, then
$$\overline{\text{Reach}(S^j_{k,m,\gamma})} = \mathcal{H}_o \cap \mathcal{S}.$$

4.1 Proof of Theorem 3

We give a sketch of the proof when $S^j_{k,m,\gamma}$ is such that $j + \gamma + k$ is even, the computations in the case $j + \gamma + k$ odd being analogous.

From Theorem 2 we already know that $\mathcal{H}_e$ is invariant; we shall then prove that (9) is an m-tracker in the Hilbert space $\mathcal{H}_e$. To do that, we are going to show that the block-wise Galerkin control condition (Definition 4) is fulfilled by (9) restricted to $\mathcal{H}_e$ and then conclude by applying Theorem 1.

Note that the following selection rules hold in the transitions between eigenstates of H

$$\langle D^j_{k,m}, \mathrm{i}B_l D^{j'}_{k',m'} \rangle = 0, \tag{12}$$

when $|j' - j| > 1$, or $|k' - k| > 1$ or $|m' - m| > 1$, for every $l = 1, 2, 3$, as one can check, for instance, in (Cook and Gordy, 1984, Table 2.1). Let us project (9) onto the subspace of $\mathcal{H}_e$ given by

$$\mathcal{M}_j := \mathcal{H}_j \oplus \mathcal{H}_{j+1},$$

where $\mathcal{H}_l := \text{span}\{S^j_{k,m,\gamma} \mid j + \gamma + k \text{ even}, \ j = l\}$, $l \in \mathbb{N}$, are the eigenspaces of the angular momentum J seen as an operator on $\mathcal{H}_e$. The dimension of $\mathcal{M}_j$ is $(2j + 1)(j + 1) + (2(j + 1) + 1)(j + 1)$, if j is even, and $(2j+1)j + (2(j+1)+1)(j+2)$, if j is odd. Let us assume that j is even, being the computation in the case j odd analogous. We identify $\mathfrak{su}(\mathcal{M}_j)$ with $\mathfrak{su}(j, j + 1)$, that is, $\mathfrak{su}((2j + 1)(j + 1) + (2(j + 1) + 1)(j + 1))$.

Referring to the notations introduced in Section 2, we shall consider the following spectral gaps in $\mathcal{M}_j$

$$\begin{aligned} &\lambda^j_{\pm k} := |E^{j+1}_{k\pm1} - E^j_k| \\ &= \Big|\frac{j+1}{I_2} + \Big(\frac{1}{2I_3} - \frac{1}{2I_2}\Big)(\pm 2k + 1)\Big| \in \Sigma_{j,j+1}, \\ &\eta_{\pm k} := |E^{j+1}_{k\pm1} - E^{j+1}_k| \\ &= \Big|\Big(\frac{1}{2I_3} - \frac{1}{2I_2}\Big)(\pm 2k + 1)\Big| \in \Sigma_{j,j+1}, \quad k = 0, \dots, j. \end{aligned} \tag{13}$$

We remark that assumption (11) guarantees that $(\lambda^j_{\pm k}, l) \in \Xi^0_{j,j+1}$ and $(\eta_{\pm k}, l) \in \Xi^1_{j,j+1}$, for every $l = 1, 2, 3$ and $k = 0, \dots, j$.

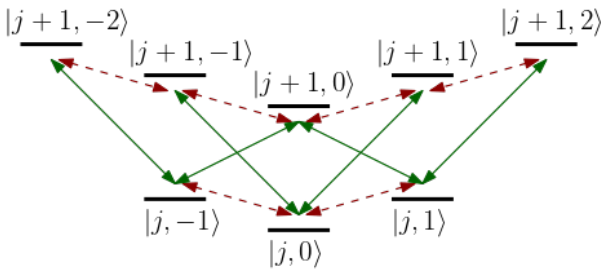

Fig. 2. Graph of the transitions associated with the frequency $\lambda^j_{\pm k}$ (solid arrows) and the frequency $\eta_{\pm k}$ (dashed arrows) between eigenstates $|j,k\rangle = |j,k,m\rangle := D^j_{k,m}$ (m is fixed).

We drive the electric fields at the frequencies defined in (13) to excite the corresponding transitions between the rotational states of H. To be more precise, assumption (11) guarantees that the frequencies (13) are not resonant between each other, and thus we can write that the three polarizations driven at frequency λ^j_k, $k = 0,\dots,j$, are represented by

$$\begin{aligned}
&\mathcal{E}_{\lambda^j_{\pm k}}(\mathrm{i}B_1)\\
&= \sum_{m=-j,,j} -2c_{j,\pm k,m}\delta_1 F_{(j,k,m,\gamma),(j+1,k\pm1,m+1,\gamma)}\\
&\quad - 2c_{j,\pm k,-m}\delta_1 F_{(j,k,m,\gamma),(j+1,k\pm1,m-1,\gamma)},\\
&\mathcal{E}_{\lambda^j_{\pm k}}(\mathrm{i}B_2)\\
&= \sum_{m=-j,,j} -2c_{j,\pm k,m}\delta_1 E_{(j,k,m,\gamma),(j+1,k\pm1,m+1,\gamma)}\\
&\quad + 2c_{j,\pm k,-m}\delta_1 E_{(j,k,m,\gamma),(j+1,k\pm1,m-1,\gamma)},\\
&\mathcal{E}_{\lambda^j_{\pm k}}(\mathrm{i}B_3)\\
&= \sum_{m=-j,,j} -2d_{j,\pm k,m}\delta_1 E_{(j,k,m,\gamma),(j+1,k\pm1,m,\gamma)},
\end{aligned} \tag{14}$$

where $\gamma = 0$ if k is even, and $\gamma = 1$ if k is odd. The three fields in (14) are seen as skew-adjoint operators acting on the Hilbert space $\mathcal{M}_j$, and, with a slight abuse of notation, we write B_l instead of $B_l^{(j,j+1)} := \Pi_{\mathcal{M}_j} B_l \Pi_{\mathcal{M}_j}$, where $\Pi_{\mathcal{M}_j}$ is the orthogonal projection onto $\mathcal{M}_j$.

In (14), the matrices $E_{s,t}, F_{s,t} \in \mathfrak{su}(\mathcal{M}_j)$ are defined by $E_{s,t} = e_{s,t} - e_{s,t}$, $F_{s,t} = \mathrm{i}e_{s,t} + \mathrm{i}e_{s,t}$, where $e_{s,t}$ denotes the $(2j+1)(j+1) + (2(j+1)+1)(j+1)$-square matrix whose entries are all zero, except the one at row s and column t, which is equal to 1. Moreover, we are implicitly using the lexicographic correspondance between the sets $\{(l,k,m,\gamma) \mid l = j, j+1, k = 0,\dots,l, m = -l,\dots,l, \gamma = 0,1\}$ and $\{1,2,\dots,(2j+1)(j+1)+(2(j+1)+1)(j+1)\}$, to label the matrices in (14).

Finally, the coupling constants in (14) are given by the pairings (see, for instance, (Cook and Gordy, 1984, Table 2.1))

$$\begin{cases}
\langle D^j_{k,m}, \mathrm{i}B_1 D^{j+1}_{k+1,m\pm1}\rangle = -\mathrm{i}c_{j,k,\pm m}\delta_1\\
\langle D^j_{k,m}, \mathrm{i}B_1 D^{j+1}_{k-1,m\pm1}\rangle = -\mathrm{i}c_{j,-k,\pm m}\delta_1\\
\langle D^j_{k,m}, \mathrm{i}B_2 D^{j+1}_{k+1,m\pm1}\rangle = \pm c_{j,k,\pm m}\delta_1\\
\langle D^j_{k,m}, \mathrm{i}B_2 D^{j+1}_{k-1,m\pm1}\rangle = \pm c_{j,-k,\pm m}\delta_1\\
\langle D^j_{k,m}, \mathrm{i}B_3 D^{j+1}_{k\pm1,m}\rangle = -d_{j,\pm k,m}\delta_1,
\end{cases} \tag{15}$$

where

$$c_{j,k,m} := \frac{[(j+k+1)(j+k+2)]^{1/2}[(j+m+1)(j+m+2)]^{1/2}}{4(j+1)[(2j+1)(2j+3)]^{1/2}},$$

and

$$d_{j,k,m} := \frac{[(j+k+1)(j+k+2)]^{1/2}[(j+1)^2-m^2]^{1/2}}{2(j+1)[(2j+1)(2j+3)]^{1/2}}.$$

In the same way, one can represent the interaction operators $\mathcal{E}_{\eta_{\pm k}}(\mathrm{i}B_l)$, $l = 1,2,3$, $k = 0,\dots,j$ relative to the spectral gaps $\eta_{\pm k}$. We have

$$\begin{aligned}
&\mathcal{E}_{\eta_{\pm k}}(\mathrm{i}B_1)\\
&= \sum_{l=j,j+1,m=-l,,l-1} \mp 2h_{l,\pm k,m}\delta_1 F_{(l,k,m,\gamma),(l,k\pm1,m+1,\gamma')}\\
&\quad \sum_{l=j,j+1,m=-l+1,,l} \pm 2h_{l,\pm k,-m}\delta_1 F_{(l,k,m,\gamma),(l,k\pm1,m-1,\gamma')},\\
&\mathcal{E}_{\eta_{\pm k}}(\mathrm{i}B_2)\\
&= \sum_{l=j,j+1,m=-l,,l-1} \pm 2h_{l,\pm k,m}\delta_1 E_{(l,k,m,\gamma),(l,k\pm1,m+1,\gamma')}\\
&\quad \sum_{l=j,j+1,m=-l+1,,l} \pm 2h_{l,\pm k,-m}\delta_1 E_{(l,k,m,\gamma),(l,k\pm1,m-1,\gamma')},\\
&\mathcal{E}_{\eta_{\pm k}}(\mathrm{i}B_3)\\
&= \sum_{l=j,j+1,m=-l,,l} \pm 2q_{l,\pm k,m}\delta_1 E_{(l,k,m,\gamma),(l,k\pm1,m,\gamma')},
\end{aligned} \tag{16}$$

where $\gamma = 0, \gamma' = 1$ if k is even, and $\gamma = 1, \gamma' = 0$ if k is odd. The coupling constants in (16) are given by the pairings (see, for instance, (Cook and Gordy, 1984, Table 2.1))

$$\begin{cases}
\langle D^j_{k,m}, \mathrm{i}B_1 D^j_{k+1,m\pm1}\rangle = \mp\mathrm{i}h_{j,k,\pm m}\delta_1\\
\langle D^j_{k,m}, \mathrm{i}B_1 D^j_{k-1,m\pm1}\rangle = \pm\mathrm{i}h_{j,-k,\pm m}\delta_1\\
\langle D^j_{k,m}, \mathrm{i}B_2 D^j_{k+1,m\pm1}\rangle = h_{j,k,\pm m}\delta_1\\
\langle D^j_{k,m}, \mathrm{i}B_2 D^j_{k-1,m\pm1}\rangle = -h_{j,-k,\pm m}\delta_1\\
\langle D^j_{k,m}, \mathrm{i}B_3 D^j_{k\pm1,m}\rangle = \pm q_{j,\pm k,m}\delta_1,
\end{cases} \tag{17}$$

where

$$h_{j,k,m} := \frac{[j(j+1)-k(k+1)]^{1/2}[j(j+1)-m(m+1)]^{1/2}}{4j(j+1)},$$

and

$$q_{j,k,m} := \frac{[j(j+1)-k(k+1)]^{1/2}m}{2j(j+1)}.$$

We now introduce the family

$$\begin{aligned}
\mathcal{F}^0_j :=& \{\mathcal{E}_{\lambda^j_{\pm k}}(\mathrm{i}B_l), W_{\mathrm{i}}(\mathcal{E}_{\lambda^j_{\pm k}}(\mathrm{i}B_l)), \mid l = 1,2,3,\\
& k = 0,\dots,j\} \subset \nu^0_{j,j+1}
\end{aligned}$$

of the interaction operators excited at non-resonant frequencies, and we denote by $\mathrm{Lie}(\mathcal{F}^0_j)$ the Lie algebra generated by the matrices in $\mathcal{F}^0_j$; finally, let us introduce the new family

$$\begin{aligned}
\mathcal{F}_j :=& \{A, \left[W_\xi(\mathcal{E}_{\eta_{\pm k}}(\mathrm{i}B_l)), B\right] \mid A, B \in \mathrm{Lie}(\mathcal{F}^0_j),\ l = 1,2,3,\\
& \xi = 1, \mathrm{i}, \quad k = 0,\dots,j\} \subset \mathcal{I}_{j,j+1},
\end{aligned}$$

the sets of operators $\nu^0_{j,j+1}$ and $\mathcal{I}_{j,j+1}$ being defined in Section 2.

Then, direct Lie bracket computations can be applied to prove that $\mathrm{Lie}(\mathcal{F}_j) = \mathfrak{su}(\mathcal{M}_j)$, for every $j \in \mathbb{N}$ (see Boscain et al. (2019) for similar calculations).

5. CONCLUSIONS

We have recalled in a general framework a sufficient condition for the approximate controllability of the bilinear Schrödinger equation, which remarkably fits the problem of controlling the rotational molecular dynamics, due to the discrete quantization of the model and the existence of the angular momentum invariant j-subspaces. Then, we have applied the abstract result on a concrete model, namely, to compute the reachable states of an orthogonal accidentally symmetric molecule.

More in detail, we have found that the configuration Hilbert space of the molecule splits into two subspaces, and in each subspace we have shown how three orthogonally polarized electric fields are sufficient to control the dynamics.

This result is important because it evidences an explicit obstruction in controlling the motion of the molecule, which was not expected since it does not have classical counterpart in the rigid body dynamics. Nevertheless it tells that, starting with an initial data prepared in one of the two invariant subspaces, the molecule's rotational state can be controlled inside the given subspace.

Hence, we could expect several applications of this model both in numerical simulations and in real experiments of quantum chemistry and physics which involve rotational states of these particular class of molecules.

Furthermore, as a challenging research problem, the next step would be the computation of the reachable sets for an asymmetric molecule.

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