Approximate controllability of the Schrödinger equation with a polarizability term

Nabile Boussaïd
Laboratoire de mathématiques
Université de Franche–Comté
25030 Besançon, France
Nabile.Boussaid@univ-fcomte.fr

Marco Caponigro
Dept of Mathematical Sciences and CCIB
Rutgers University
08102 Camden, NJ, USA
marco.caponigro@rutgers.edu

Thomas Chambrion
Institut Élie Cartan de Nancy and
INRIA Nancy Grand Est
54506 Vandœuvre, France
Thomas.Chambrion@inria.fr

***Abstract*— This paper is concerned with the controllability of quantum systems in the case where the standard dipolar approximation, involving the permanent dipole moment of the system, has to be corrected by a so-called polarizability term, involving the field induced dipole moment. Sufficient conditions for controllability between eigenstates of the free Hamiltonian are derived and control laws are explicitly given. As an illustration, the results are applied to the planar rotation of the HCN molecule.**

I. INTRODUCTION

A. *Control of quantum systems*

The state of a quantum system evolving on a Riemannian manifold Ω is described by its wavefunction ψ, an element of the unit sphere of $L^2(\Omega,\mathbf{C})$. When the system is submitted to an electric field, the time evolution of the wavefunction is given by the Schrödinger equation

$$\mathrm{i}\frac{\partial\psi}{\partial t}=(-\Delta+V(x))\psi+\mu(u,x)\psi(t),\quad x\in\Omega,$$

where Δ is the Laplace-Beltrami operator on Ω, $V:\Omega\to\mathbf{R}$ is a potential describing the system in absence of control, u is the scalar (time variable) intensity of the electric field and $\mu:\Omega\times\mathbf{R}\to\mathbf{R}$ describes the effect of the external field. In the dipolar approximation we expand μ to the first order in u and we then represent $\mu(u,x)$ as $uW(x)$, where W is a real function.

Although the dipolar approximation usually gives excellent results, it is sometimes necessary (see [1], [2]) to consider a better approximation of μ involving the first two terms of its expansion in u. We approximate $\mu(u,x)$ with $uW_1(x)+u^2W_2(x)$ for two real functions $W_1(x)$ and $W_2(x)$. An example of this approximation is given by problem of orienting a rotating HCN molecule as presented in Section IV.

The aim of this work is to derive controllability properties for the controlled Schrödinger equation, by means of the dipolar term uW_1 and the polarizability term u^2W_2.

This question has already been tackled by various authors in [3], [4] (for finite dimensional approximations) and in [5] (for the infinite dimensional version of the problem, when Ω is a bounded set of $\mathbf{R}^n$ and W_1,W_2 are smooth functions). All these contributions rely on Lyapunov methods.

The novelty of our contribution is that we are able to deal with some unbounded or not continuous potentials W_1 and W_2. Moreover, when considering the physically relevant problem of transferring the quantum system from an energy level to another in the case where W_2 is bounded, all our methods are constructive and allow easy numerical simulations.

B. *Framework and notations*

We set the problem in a more abstract framework. In a separable Hilbert space H, endowed with the Hermitian product $\langle\cdot,\cdot\rangle$, we consider the following control system

$$\frac{d}{dt}\psi=(A+u(t)B+u^2(t)C)\psi,\tag{1}$$

where (A,B,C,k) satisfies Assumption 1 for some k.

Assumption 1: k is an integer and (A,B,C) is a triple of (possibly unbounded) linear operators in H such that

1) A is skew-adjoint with pure point spectrum $(-\mathrm{i}\lambda_j)_{j\in\mathbf{N}}$ with $\lambda_j\neq 0$, $\lambda_j\to\infty$;
2) for every (u_1,u_2) in $\mathbf{R}^2$, $A+u_1B+u_2C$ is essentially skew-adjoint with domain $D(A)$;
3) for every (u_1,u_2) in $\mathbf{R}^2$, $|A+u_1B+u_2C|^{k/2}$ has domain $D(|A|^{k/2})$);
4) $\displaystyle\sup_{\psi\in D(|A|^k)\setminus\{0\}}\frac{|\Re\langle|A|^k\psi,B\psi\rangle|}{|\langle|A|^k\psi,\psi\rangle|}+\frac{|\Re\langle|A|^k\psi,C\psi\rangle|}{|\langle|A|^k\psi,\psi\rangle|}<+\infty;$
5) there exist $d>0$ and $0\leq r<k$ such that $\|B\psi\|\leq d\||A|^{r/2}\psi\|$ and $\|C\psi\|\leq d\||A|^{r/2}\psi\|$ for every ψ in $D(|A|^{r/2})$.

If (A,B,C,k) satisfies Assumption 1, we define $c_{(A,B,C,k)}$ as the lower bound of the set of every real c such that for every ψ in $D(|A|^k)$, $|\Re\langle|A|^k\psi,B\psi\rangle|\leq c|\langle|A|^k\psi,\psi\rangle|$ and $|\Re\langle|A|^k\psi,C\psi\rangle|\leq c|\langle|A|^k\psi,\psi\rangle|$.

From Assumption 1, we deduce that there exists an Hilbert basis $(\phi_k)_{k\in\mathbf{N}}$ of H made of eigenvectors of A. For every j, $A\phi_j=\mathrm{i}\lambda_j\phi_j$. As A is skew-adjoint and diagonalizable, $|A|$ is self-adjoint positive and diagonalizable in the same basis as A. The eigenvalues of $|A|$ are the moduli of the eigenvalues of A. We define the k-norm of an element ψ of $D(A^k)$ as $\|\psi\|_k=\||A|^k\psi\|$. When Ω is a compact manifold and $A=\mathrm{i}\Delta$, the k-norm is equivalent to the H^{2k} norm on Ω.

If (A,B,C,k) satisfies Assumption 1, for every u in $\mathbf{R}$, $A+uB+u^2C$ generates a group of unitary propagators $t\mapsto e^{t(A+uB+u^2C)}$. By concatenation, one can define the solution of (1) for every piecewise constant u, for every initial condition ψ_0 given at time t_0. We denote this solution $t \mapsto \Upsilon^u_{t,t_0}\psi_0$.

We define PC, the set of piecewise constant functions u such that there exists two sequences $0 = t_1 < t_2 < \ldots < t_{p+1}$ and $u_1, u_2, \ldots, u_p > 0$ with

$$u = \sum_{j=1}^{p} u_j \mathbf{1}_{[t_j, t_{j+1})}.$$

Set $\tau_j = t_{j+1} - t_j$, we write $u = (u_j, \tau_j)_{1\leq j\leq p}$.

The operators B and C can be seen as infinite dimensional matrices in the basis $(\phi_j)_{j\in\mathbf{N}}$. For every $j, l \in \mathbf{N}$, we denote $b_{jl} = \langle \phi_j, B\phi_l\rangle$ and $c_{jl} = \langle \phi_j, C\phi_l\rangle$. For every N, the orthogonal projection $\pi_N : H \to H$ on the space spanned by the first N eigenvectors of A is defined by

$$\pi_N(x) = \sum_{l=1}^{N} \langle \phi_l, x\rangle \phi_l \qquad \text{for every } x \text{ in } H.$$

Let $\mathcal{L}_N$ be the range of π_N. The *compressions* of A, B and C at order N are the finite rank operators $A^{(N)} = \pi_N A_{\restriction \mathcal{L}_N}$, $B^{(N)} = \pi_N B_{\restriction \mathcal{L}_N}$ and $C^{(N)} = \pi_N C_{\restriction \mathcal{L}_N}$ respectively. The *Galerkin approximation* of (1) of order N is the system in $\mathcal{L}_N$

$$\dot{x} = (A^{(N)} + uB^{(N)} + u^2 C^{(N)})x. \tag{2}$$

A couple (j, l) in $\mathbf{N}^2$ is a *non-degenerate transition* of (A, B, C) if $|b_{jl}| + |c_{jl}| \neq 0$ and, for every m, n, $|\lambda_j - \lambda_l| = |\lambda_n - \lambda_m|$ implies $\{j, l\} = \{m, n\}$ or $\{m, n\} \cap \{j, l\} = \emptyset$ or $|b_{mn}| + |c_{mn}| = 0$.

A subset S of $\mathbf{N}^2$ is a *chain of connectedness* of (A, B, C) if there exists α in $\mathbf{R}$ such that, for every $m, n \in \mathbf{N}$, there exists a finite sequence $p_1, p_2, \ldots, p_r \in S$ such that $p_1 = m$, $p_r = n$, and $\langle \phi_{p_{l+1}}, (B + \alpha C)\phi_{p_l}\rangle \neq 0$ for every $l = 1, \ldots, r-1$. A chain of connectedness S of (A, B, C) is *non-degenerate* if every (m, n) in S is a non-degenerate transition of (A, B, C).

C. Main results

Theorem 1: Assume that (A, B, C) admits a non-degenerate chain of connectedness. Then, for every $\varepsilon > 0$, for almost every $\delta > 0$, for every ψ_0, ψ_1 in the Hilbert unit sphere of H, there exists $u_\varepsilon : [0, T_\varepsilon] \to \{0, \delta\}$ such that $\|\Upsilon^{u_\varepsilon}_{T_\varepsilon,0}\psi_0 - \psi_1\| < \varepsilon$.

Theorem 2: Assume that $(1, 2)$ is a non-degenerate transition of (A, B, C). Then, for every $\varepsilon > 0$, for almost every $\delta > 0$, there exists $u_\varepsilon : [0, T_\varepsilon] \to \{0, \delta\}$ such that

$$\|u_\varepsilon\|_{L^1} \leq \frac{5\pi}{4|b_{12} + \delta c_{12}|} \text{ and } \|\Upsilon^{u_\varepsilon}_{T_\varepsilon,0}\phi_1 - \phi_2\| < \varepsilon.$$

This result is constructive when C is bounded (see Section III-D for the construction of u).

D. Content of the paper

The first part of the paper (Section II) concerns the proof of some finite dimensional preliminary results. In Section III, we first give some consequences of Assumption 1 in term of approximation of the system (1) by its finite dimensional Galerkin approximations (Section III-A). Then, we use an infinite dimensional tracking result (Section III-B) to prove Theorems 1 and 2 (Section III-C). For the physically important case of quantum transfer between two eigenstates when C is bounded, an explicit construction of control law is proposed in Section III-D, using averaging theory. Finally, we present in Section IV a case study inspired by the rotational state of the HCN molecule.

II. FINITE DIMENSIONAL PRELIMINARY RESULTS

We consider the finite dimensional control problem in $\mathcal{L}_N = \mathrm{span}(\phi_1, \ldots, \phi_N)$

$$\dot{x} = (A^{(N)} + u(t)B^{(N)})x \tag{3}$$

Since $B^{(N)}$ is bounded, for every locally integrable u, we can define the solution (in the sense of Carathéodory) $t \mapsto X^u_{(N)}(t, t_0)x_0$ of (3) with initial condition, at time t_0, x_0 in $\mathcal{L}_N$.

A. Time reparametrization

We define the mapping $\mathcal{P} : PC \to PC$ by

$$\mathcal{P}\left((u_j, \tau_j)_{1\leq j\leq p}\right) = \left(\frac{1}{u_j}, u_j\tau_j\right)_{1\leq j\leq p}$$

for every $u = (u_j, \tau_j)_{j=1}^{p}$ in PC.

The mapping $\mathcal{P}$ is a reparametrization of the time with the L^1 norm of the control. Indeed, benoting by $\widehat{X}^u_{(N)}(t, s)$ the propagator of $\dot{x} = uA^{(N)}x + B^{(N)}x$, then, for every u in PC,

$$\widehat{X}^{\mathcal{P}u}_{(N)}\left(\int_0^T |u(\tau)|\mathrm{d}\tau, 0\right) = X^u_{(N)}(T, 0).$$

Indeed, for $\alpha > 0$,

$$\exp(t(A^{(N)} + \alpha B^{(N)})) = \exp\left(t\alpha\left(\frac{1}{\alpha}A^{(N)} + B^{(N)}\right)\right).$$

B. A tracking result

Proposition 3: For every $a < 0$, $b > 0$, for every $T > 0$, for every piecewise constant function u^* with support in $[0, T]$, there exists a sequence $(u_n)_{n\in\mathbf{N}}$ of piecewise constant functions $u_n : [0, T_n] \to \{a, 0, b\}$ such that $X^{u_n}_{(N)}(T_n, 0)$ tends to $X^{u^*}_{(N)}(T, 0)$ as n tends to infinity and $\|u_n\|_{L^1} \leq \|u^*\|_{L^1}$. If, moreover, u^* is nonnegative, the sequence $(u_n)_{n\in\mathbf{N}}$ can be chosen such that u_n takes value in $\{0, b\}$ for every n.

Remark 1: The approximation result in Proposition 3 is classical and it can be obtained, for instance, with Lie groups techniques, see [6]. The novelty of Proposition 3 is that the approaching sequence $(u_n)_n$ is uniformly bounded in $L^1(\mathbf{R}, \mathbf{R})$. This point is crucial for the derivation of the infinite dimensional results of the next Section.

Proof: Define v^* as the cumulative function of $\mathcal{P}|u^*|$ vanishing at 0, that is $v^*(t) = \int_0^t \mathcal{P}|u^*|(s)\mathrm{d}s$. The solution of

$$\dot{y} = \mathrm{sign}(u^* \circ v^*)e^{-v^*(t)A^{(N)}}B^{(N)}e^{v^*(t)A^{(N)}}y,$$

with initial condition $y(0) = \psi_0$ satisfies

$$e^{v^*(t)A^{(N)}} y(t) = \widehat{X}_{(N)}^{\mathrm{sign}(u^* \circ v^*)\mathcal{P}|u^*|}(v^*(t), 0)\psi_0$$

for every $t \geq 0$.

For every u in PC, define the time-varying $N \times N$ matrix $t \mapsto M_u(t)$ whose entry (j,k) is given by

$$m_{jk} : t \mapsto \mathrm{sign}(u \circ v)(t) b_{kj} e^{\mathrm{i}(\lambda_j - \lambda_k)v(t)},$$

where v is the inverse of the non-decreasing function $t \mapsto \int_0^t |u(\tau)| \mathrm{d}\tau$.

Consider, for every $\eta > 0$ and $r \in \mathbf{R}$,

$$E_\eta(r) = \{v \in \mathbf{R} \mid |e^{\mathrm{i}(\lambda_j - \lambda_k)r} - e^{\mathrm{i}(\lambda_j - \lambda_k)v}| < \eta \quad \text{for every } 1 \leq j,k \leq N\}.$$

For every $r \in \mathbf{R}$, $E_\eta(r)$ is open and nonempty. The mapping

$$\begin{array}{llll} P: & \mathbf{T}^{N^2} & \to & \mathbf{T}^{N^2} \\ & (e^{\mathrm{i}\theta_{j,k}})_{1 \leq j,k \leq N} & \mapsto & (e^{\mathrm{i}((\lambda_j - \lambda_k)v + \theta_{j,k})})_{1 \leq j,k \leq N} \end{array}$$

is a volume preserving flow on the N^2 dimensional torus. By Poincaré recurrence theorem, for almost every point $e^{\mathrm{i}(\lambda_j - \lambda_k)v}$ in the ball centered in $e^{\mathrm{i}(\lambda_j - \lambda_k)r}$ with radius η, there exists an increasing sequence of integers $(\tau_n)_{n \in \mathbf{N}}$ such that $e^{\mathrm{i}(\lambda_j - \lambda_k)\tau_n v}$ also belongs to the ball centered in $e^{\mathrm{i}(\lambda_j - \lambda_k)r}$ with radius η for every n in $\mathbf{N}$. In other words, for every $\eta > 0$ and every $r \in \mathbf{R}$, the set $E_\eta(r)$ is not bounded from above. The same argument shows that $E_\eta(r)$ is not bounded from below.

For every $l > 0$, there exists $v_l^* = \sum_{j=1}^{p_l} v_{lj} \chi_{[t_{l,j}, t_{l,j+1})}$ a piecewise constant function at distance less than l of v^* for the L^∞-norm on $[0, \|u^*\|_{L^1}]$ such that the sign of $u^* \circ v_l^*$ is constant on every interval $[t_{l,j}, t_{l,j+1})$. For every $\eta > 0$, there exists a (possibly discontinuous) piecewise affine function v_l^η defined on every interval $[t_{l,j}, t_{l,j+1})$ by

$$\begin{array}{lcll} \dot{v}_l^\eta & = & 1/b & \text{if } u^*(v_{lj}) > 0, \\ \dot{v}_l^\eta & = & 1/a & \text{if } u^*(v_{lj}) < 0, \\ v_l^\eta(t) & \in & E_\eta(v_{l,j}) & \text{for } t \in [t_{l,j}, t_{l,j+1}), \end{array}$$

such that v_l^η is increasing (respectively decreasing) on $(t_{l,j}, t_{l,j+1})$ if $u^*(v_{lj}) > 0$ (respectively $u^*(v_{lj}) < 0$), see Figure 1.

By construction, the function v_l^η is injective on $(t_{l,j}, t_{l,j+1})$. Its reciprocal on $(t_{l,j}, t_{l,j+1})$ is a (possibly discontinuous) piecewise affine function, whose derivative u_l^η is a piecewise constant function taking value in $\{a, 0, b\}$ and is such that $\|u_{l,\eta}\|_{L^1} = \|u^*\|_{L^1}$.

For every t, $\int_0^t M_{u_\eta^\eta}(\tau)\mathrm{d}\tau$ tends to $\int_0^t M_{u^*}(\tau)\mathrm{d}\tau$ as η tends to zero, uniformly on every compact of $[0, +\infty)$. By [7, Lemma 8.2], the solution y_η of $\dot{y} = M_{u_\eta^\eta}(t)y$ with initial condition $y(0) = \psi_0$ tends uniformly on every compact of $[0, +\infty)$ to the solutions of $\dot{y} = M_{u^*}(t)y$ with initial condition $y(0) = \psi_0$. Hence, y_η converge toward y_{u^*} as η tends to zero. For the conclusion, we still need to show that $e^{v^*(\int_0^T |u^*(\tau)|\mathrm{d}\tau)A^{(N)}}$ can be approached by $e^{v_\eta^\eta(\int_0^T |u^*(\tau)|\mathrm{d}\tau)A^{(N)}}$. It follows as above from the Poincaré recurrence theorem. The control term is taken to be zero in the meantime, what does not affect y nor the L^1 norm of u_η^η.

Finally, notice that if $u^* \geq 0$, then $u^*(v_{l,j})$ is always non negative. Then v_η^η is increasing and u_η^η takes value in $\{0, b\}$. ■

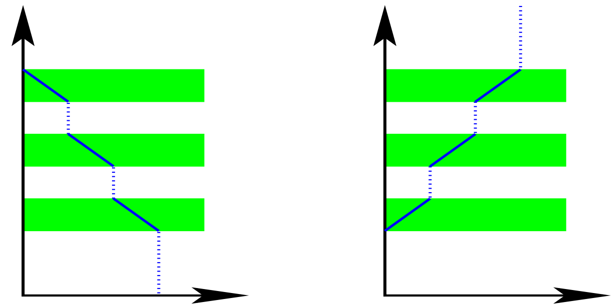

Fig. 1. Construction of the function v_l^η, when $u^*(v_{l,j}) < 0$ (left) and $u^*(v_{l,j}) > 0$ (right). The set $E_\eta(v_{l,j})$ is coloured. The piecewise affine function v_l^η is discontinuous, with derivative equal to $1/a < 0$ (left) or $1/b > 0$ (right). Notice that v_l^η is injective in both cases. The derivative u_l^η of the reciprocal function of v_l^η is piecewise affine and takes value in $\{a, 0, b\}$.

III. INFINITE DIMENSIONAL SYSTEMS

A. Weakly-coupled quantum systems

If (A, B, C, k) satisfies Assumption 1, (A, B, C) is k-*weakly-coupled*. We present here some properties of theses systems and refer to [8] for further details.

The notion of weakly-coupled systems is closely related to the growth of the $k/2$-norm $\langle |A|^k \psi, \psi \rangle$. For $k = 1$, this quantity is the expected value of the energy of the system. Next result can be found in [8, Proposition 2].

Proposition 4: Let (A, B, C, k) satisfy Assumption 1. Then, for every $\psi_0 \in D(|A|^{k/2})$, $K > 0$, $T \geq 0$, and u piecewise constant such that $\|u\|_{L^1} + \|u\|_{L^2}^2 < K$, one has $\|\Upsilon_T^u(\psi_0)\|_{k/2} \leq e^{c(A,B,C,k)K} \|\psi_0\|_{k/2}$.

Next Proposition is in [8, Proposition 4].

Proposition 5: Let k in $\mathbf{N}$ and (A, B, C, k) satisfy Assumption 1. Then for every $\varepsilon > 0$, $s < k$, $K \geq 0$, $n \in \mathbf{N}$, and $(\psi_j)_{1 \leq j \leq n}$ in $D(|A|^{k/2})^n$ there exists $N \in \mathbf{N}$ such that for every piecewise constant function u

$$\|u\|_{L^1} + \|u^2\|_{L^1} < K \Rightarrow \|\Upsilon_t^u(\psi_j) - X_{(N)}^u(t,0)\pi_N \psi_j\|_{s/2} < \varepsilon,$$

for every $t \geq 0$ and $j = 1, \ldots, n$.

Remark 2: Notice that, in Propositions 4 and 5, the upper bound of the $|A|^{k/2}$ norm of the solution of (1) or the bound on the error between the infinite dimensional system and its finite dimensional approximation only depend on the L^1 norm of the control, not on the time.

B. An infinite dimensional tracking result

Next result can be seen as a Bang-Bang Theorem for infinite dimensional systems.

Lemma 6: Let $(A, B, 0, k)$ satisfy Assumption 1 with k in $\mathbf{N}$, T be a positive number, a, b be two real numbers such that $a < 0 < b$, u^* be a piecewise constant function with support in $[0, T]$, and ψ_0 be in H. Then, for every $\varepsilon > 0$, there exists a piecewise constant control $u_\varepsilon : [0, T_\varepsilon] \to \{a, 0, b\}$ such that $\|\Upsilon^{u_\varepsilon}_{T_\varepsilon,0}(\psi_0) - \Upsilon^{u^*}_{T,0}(\psi_0)\| < \varepsilon$, and $\|u_\varepsilon\|_{L^1} \leq \|u^*\|_{L^1}$. Moreover, if u^* is positive, then u_ε may be chosen with value in $\{0, b\}$.

Proof: Let $\varepsilon > 0$. By Proposition 5, there exists N in $\mathbf{N}$ such that, for every piecewise constant function u,

$$\|u\|_{L^1} < \|u^*\|_{L^1} \Rightarrow \|\Upsilon^u_t(\psi_0) - X^u_{(N)}(t, 0)\pi_N\psi_0\| < \varepsilon.$$

From Proposition 3, there exists $u_\varepsilon : [0, T\varepsilon] \to \{a, 0, b\}$ piecewise constant such that $\|u_\varepsilon\|_{L^1} \leq \|u^*\|_{L^1}$ and

$$\|X^{u^*}_{(N)}(T, 0) - X^{u_\varepsilon}_{(N)}(T, 0)\| < \varepsilon.$$

Then

$$\begin{aligned} &\|\Upsilon^{u_\varepsilon}_{T_\varepsilon,0}(\psi_0) - \Upsilon^{u^*}_{T,0}(\psi_0)\| \\ &\leq \quad \|\Upsilon^{u_\varepsilon}_{T_\varepsilon,0}(\psi_0) - X^{u_\varepsilon}_{(N)}(t, 0)\pi_N\psi_0\| \\ &\quad +\|X^{u_\varepsilon}_{(N)}(T_\varepsilon, 0)\pi_N\psi_0 - X^{u^*}_{(N)}(T, 0)\pi_N\psi_0\| \\ &\quad +\|\Upsilon^{u^*}_{T,0}(\psi_0) - X^{u^*}_{(N)}(T, 0)\pi_N\psi_0\| \\ &\leq \quad 3\varepsilon. \end{aligned}$$

The same proof shows that, if u^* is positive, u_ε can be chosen with values in $\{0, b\}$. ■

C. Proof of the main results

We recall results dealing with approximate controllability for bilinear systems, i.e. when $C = 0$. Their proofs are given in [9, Theorem 2.6] and [9, Proposition 2.8] respectively.

Theorem 7: Let $(A, B, 0, 0)$ satisfy Assumption 1. If there exists a non-degenerate chain of connectedness of $(A, B, 0)$ then, for every ψ_0, ψ_1 in the Hilbert unit sphere of H, for every $\varepsilon > 0$, for every $\delta > 0$, there exists $T > 0$ and a piecewise constant function $u : [0, T] \to [0, \delta]$ such that $\|\Upsilon^u(T, 0)\psi_0 - \psi_1\| < \varepsilon$.

Theorem 8: Let $(A, B, 0, 0)$ satisfy Assumption 1. If $\psi_0 = \phi_1$, $\psi_1 = \phi_2$ and $(1, 2)$ is a non-degenerate transition of $(A, B, 0)$, then for every $\varepsilon > 0$ and for every $\delta > 0$, there exists $T > 0$ and a piecewise constant function $u : [0, T] \to [0, \delta]$ such that $\|\Upsilon^u(T, 0)\psi_0 - \psi_1\| < \varepsilon$ and $\|u\|_{L^1} \leq \dfrac{5\pi}{4|b_{1,2}|}$.

We now proceed to the proof of the Theorem 1. Assume that (A, B, C, k) satisfies Assumption 1 for some k in $\mathbf{N}$ and admits a non-degenerate chain of connectedness. Then, there exists $\alpha > 0$ such that $(A, B + \alpha C, 0)$ satisfies Assumption 1 and admits a non-degenerate chain of connectedness. By analyticity, this property is true for almost every α in $\mathbf{R}$. From Theorem 7, for every ψ_0, ψ_1 in the Hilbert unit sphere of H, for every $\varepsilon > 0$, and for every $\delta > 0$, there exist $T > 0$ and a piecewise constant function $u : [0, T] \to [0, \delta]$ such that the solution of $Y : t \mapsto Y(t) \in H$ of

$$\frac{d}{dt}\psi = A\psi + u(t)(B + \alpha C)\psi$$

with initial condition $Y(0) = \psi_0$ satisfies $\|Y(T) - \psi_1\| < \varepsilon$. By Lemma 6, there exists $\tilde{u} : [0, T_{\tilde{u}}] \to \{0, \alpha\}$ such that $\|\Upsilon^{\tilde{u}}_{T_{\tilde{u}},0}\psi_0 - Y(T)\| < \varepsilon$. Thus $\|\Upsilon^{\tilde{u}}_{T_{\tilde{u}},0}\psi_0 - \psi_1\| < 2\varepsilon$. To conclude the proof of Theorem 1, it is enough to notice that, for every t, $\tilde{u}(t)B + \tilde{u}^2(t)C = \tilde{u}(t)(B + \alpha C)$ as $\tilde{u}$ takes only the values 0 and α.

Similarly we can prove Theorem 2 using the result of Theorem 8 instead of Theorem 7.

D. Controllability between eigenstates when C is bounded

In this Section, we use averaging techniques to provide explicit expressions of control laws steering one eigenstate of the system to another.

In quantum mechanics, averaging theory has been extensively used (under the name of "Rotating Wave Approximation") since the 60's, for finite dimensional systems. It has recently been extended to the case of infinite dimensional systems [10, Theorem 1]. We recall this result in the following proposition. Let $Y_n : t \mapsto Y_n(t) \in H$ be the propagator of

$$\frac{\mathrm{d}}{\mathrm{d}t}\psi = \left(A + \frac{u}{n}B\right)\psi.$$

Proposition 9: Let $(A, B, 0, 0)$ satisfy Assumption 1. Assume that $(1, 2)$ is a non-degenerate transition of $(A, B, 0)$. Define $\mathcal{N} = \{n \in \mathbf{N} |$ there exists (l_1, l_2) with $b_{l_1,l_2} \neq 0$ and $|l_1 - l_2| = n|\lambda_1 - \lambda_2|\}$. If u is $2\pi/|\lambda_2 - \lambda_1|$-periodic and satisfies, for every n in $\mathcal{N}$,

$$\int_0^{2\pi/|\lambda_2-\lambda_1|} e^{\mathrm{i}n|\lambda_2-\lambda_1|t}u(t)\mathrm{d}t \neq 0 \quad \text{if } n = 1$$

and

$$\int_0^{2\pi/|\lambda_2-\lambda_1|} e^{\mathrm{i}n|\lambda_2-\lambda_1|t}u(t)\mathrm{d}t = 0 \quad \text{if } n > 1$$

then there exists $T^* > 0$ such that $|\langle\phi_2, Y_n(nT^*, 0)\phi_1\rangle|$ tends to 1 as n tends to infinity.

Our aim is to extend the result of Proposition 9 to the case where $C \neq 0$ is bounded.

Proposition 10: Let $(A, B, C, 0)$ satisfy Assumption 1. Assume that C is bounded and that $(1, 2)$ is a non-degenerate transition of $(A, B, 0)$. Define $\mathcal{N} = \{n \in \mathbf{N} |$ there exists (l_1, l_2) with $b_{l_1,l_2} \neq 0$ and $|l_1 - l_2| = n|\lambda_1 - \lambda_2|\}$. If u is $2\pi/|\lambda_2 - \lambda_1|$ periodic and satisfies, for every n in $\mathcal{N}$,

$$\int_0^{2\pi/|\lambda_2-\lambda_1|} e^{\mathrm{i}n|\lambda_2-\lambda_1|t}u(t)\mathrm{d}t \neq 0 \quad \text{if } n = 1$$

and

$$\int_0^{2\pi/|\lambda_2-\lambda_1|} e^{\mathrm{i}n|\lambda_2-\lambda_1|t}u(t)\mathrm{d}t = 0 \quad \text{if } n > 1$$

then there exists $T^* > 0$ such that $|\langle\phi_2, \Upsilon^{u/n}_{nT^*,0}\phi_1\rangle|$ tends to 1 as n tends to infinity.

Proof: The proof relies on the Duhamel formula. For every $t > 0$ and $n \in \mathbf{N}$,

$$\Upsilon^{u/n}_{nt,0}\phi_1 - Y_n(nt, 0)\phi_1 = \frac{1}{n^2}\int_0^{nt} u^2(s)Y_n(nt, s)C\Upsilon^{u/n}_{s,0}\phi_1\mathrm{d}s.$$

Since Y_n and Υ are unitary propagators and that u is $2\pi/|\lambda_2 - \lambda_1|$ periodic, one gets

$$\left|\Upsilon^{u/n}_{nt,0}\phi_1 - Y_n(nt,0)\phi_1\right| \leq \frac{\|C\|}{n}\left(\frac{t|\lambda_2-\lambda_1|}{2\pi}+1\right)\int_0^{\frac{2\pi}{|\lambda_1-\lambda_2|}} |u(s)|^2 \mathrm{d}s, \quad (4)$$

which, for every fixed t, tends to zero as n tends to infinity. Proposition 10 follows from Proposition 9 by taking $t = T^*$ in (4). ■

E. Controllability in higher norms

Up to now, we have considered approximate controllability of (1) in the norm of H. When $H = L^2(\Omega, \mathbf{C})$, for instance, we have convergence in L^2 norm. In this Section, we consider approximate controllability of (1) in k-norm.

Theorem 11: Let (A, B, C, k) satisfy Assumption 1 and $\delta \neq 0$ such that $(1, 2)$ is a non-degenerate transition of $(A, B + \delta C, 0)$. Then, for every $\varepsilon > 0$, for every $s < k/2$, there exists $u_\varepsilon : [0, T_\varepsilon] \to \{0, \delta\}$ such that $\|u_\varepsilon\|_{L^1} \leq \dfrac{5\pi}{4|b_{1,2} + \delta c_{1,2}|}$ and $\|\Upsilon^{u_\varepsilon}_{T_\varepsilon,0}\phi_1 - \phi_2\|_s < \varepsilon$.

Proof: If $s = 0$, the result is just a rewriting of Theorem 2. To conclude for general s, we use a classical interpolation argument: if a sequence $(x_n)_n$ in H tends to zero for the r_1-norm and is bounded for the r_2-norm, then $(x_n)_n$ tends to zero for the $\frac{r_1+r_2}{2}$-norm. Proposition 4 and the uniform bound on the L^1 norm of the control ensures that $\Upsilon^u(t, 0)\psi_0$ is bounded in s-norm for every $s < k$. ■

IV. EXAMPLE: ALIGNMENT DYNAMICS OF HCN

A. Modeling

We consider a linear molecule with fixed length and center of mass. This is the case for instance of the HCN molecule considered in [2]. We assume that the molecule is constrained to stay in a fixed plane and that its only degree of freedom is the rotation, in the plane, around its center of mass. The state of the system at time t is described by a point $\theta \mapsto \psi(t, \theta)$ of $L^2(\Omega, \mathbf{C})$ where $\Omega = \mathbf{R}/2\pi\mathbf{Z}$ is the one dimensional torus. The Schrödinger equation reads

$$\mathrm{i}\frac{\partial \psi}{\partial t} = -\Delta\psi + u(t)\cos(\theta)\psi + u^2(t)\cos(2\theta)\psi \quad (5)$$

where Δ is the Laplace-Beltrami operator on Ω. The self-adjoint operator $-\Delta$ has purely discrete spectrum $\{k^2, k \in \mathbf{N}\}$. All its eigenvalues are double but zero which is simple. The eigenvalue zero is associated with the constant functions. The eigenspace associated with the double eigenvalue k^2 for $k > 0$ is spanned by the two eigenfunctions $\theta \mapsto \frac{1}{\sqrt{\pi}}\cos(k\theta)$ and $\theta \mapsto \frac{1}{\sqrt{\pi}}\sin(k\theta)$. The Hilbert space $H = L^2(\Omega, \mathbf{C})$ splits in two subspaces H_e and H_o, the spaces of even and odd functions of H respectively. The spaces H_e and H_o are invariant under the dynamics of (5), hence no global controllability is to be expected in H.

B. Theoretical analysis

We consider the restriction of (5) to the space H_o. The function $\phi_k : \theta \mapsto \sin(k\theta)/\sqrt{\pi}$ is an eigenvector of the skew-adjoint operator $A = \mathrm{i}\Delta_{\restriction H_o}$. The family $(\phi_k)_{k\in\mathbf{N}}$ is an Hilbert basis of H_o. Here, B and C are the restriction to H_o of the multiplication by $-\mathrm{i}\cos(\theta)$ and $-\mathrm{i}\cos(2\theta)$ respectively. The skew-adjoint operators B and C are bounded, we can then consider controls that are in the intersection $L^1([0,+\infty), \mathbf{R}) \cap L^2([0,+\infty), \mathbf{R})$ and not necessarily piecewise constant.

The $N \times N$ matrices in the basis $(\phi_j)_{1\leq j\leq N}$ of the compressions of A, B and C of order N restricted to H_o are

$$A^{(N)} = -\begin{pmatrix} \mathrm{i} & 0 & \cdots & 0 \\ 0 & 4\mathrm{i} & \ddots & \vdots \\ \vdots & \ddots & \ddots & 0 \\ 0 & \cdots & 0 & N^2\mathrm{i} \end{pmatrix},$$

$$B^{(N)} = -\mathrm{i}\begin{pmatrix} 0 & 1/2 & 0 & \cdots & 0 \\ 1/2 & 0 & 1/2 & \ddots & \vdots \\ 0 & \ddots & 0 & \ddots & 0 \\ \vdots & \ddots & 1/2 & 0 & 1/2 \\ 0 & \cdots & 0 & 1/2 & 0 \end{pmatrix},$$

$$\text{and } C^{(N)} = -\mathrm{i}\begin{pmatrix} -1/2 & 0 & 1/2 & 0 & \cdots \\ 0 & 0 & 0 & 1/2 & \ddots \\ 1/2 & 0 & 0 & \ddots & \ddots \\ 0 & 1/2 & 0 & 0 & \ddots \\ \vdots & \ddots & \ddots & \ddots & \ddots \end{pmatrix}.$$

The system (A, B, C, k) satisfies Assumption 1 for every k in $\mathbf{N}$ ([8, Section III-C]).

Let us illustrate Theorem 2 with the transition between the first and the second eigenlevel. The transition $(1, 2)$ is non-degenerate for the system $(A, B + \alpha C, 0)$ for every α in $\mathbf{R}$. Hence, it is possible to induce, with arbitrary precision, a transition from the first to the second eigenlevel while using controls taking value in $\{0, \alpha\}$ for any $\alpha > 0$. Following Proposition 10, we consider control laws of the form $u_k : t \mapsto \cos^k(3t)/10$ for k in $\mathbf{N}$. When k is even, $\int_0^{2\pi/3} \cos^k(t)e^{\mathrm{i}3t}\mathrm{d}t = 0$. Hence, we will consider odd k only.

C. Numerical simulations

Proposition 5 claims that the error done when replacing the original infinite dimensional system by the Galerkin approximation of order N tends to zero as N tends to infinity. To estimate this error, one can use a method similar the one used in [8, Section IV-C]. If $\int_0^T |u(\tau)|\mathrm{d}\tau < K_1$ and $\int_0^T |u(\tau)|^2\mathrm{d}\tau < K_2$, then, with a computation similar to [8, Lemma 11], one has

$$|\langle\phi_j, \Upsilon^u_{T,0}\phi_1\rangle| \leq \frac{(K_1+K_2)^N}{N!} \text{ for } j = 2N+1, 2N+2.$$

TABLE I

NUMERICAL RESULTS

Control law u	n	Time T	Error $1-\left\|\langle\phi_2, X^{u/n}_{(62)}(T,0)\phi_1\rangle\right\|$
$t\mapsto\cos(3t)$	1	6.80	$< 2.76\ 10^{-1}$
	10	62.30	$< 3.13\ 10^{-3}$
	30	187.95	$< 3.48\ 10^{-4}$
$t\mapsto\cos^3(3t)$	1	7.85	$< 2.30\ 10^{-1}$
	10	83.25	$< 2.77\ 10^{-3}$
	30	250.80	$< 3.09\ 10^{-4}$
$t\mapsto\cos^5(3t)$	1	9.95	$< 2.57\ 10^{-1}$
	10	100.00	$< 3.16\ 10^{-3}$
	30	301.05	$< 3.52\ 10^{-4}$

Hence, by [8, Equation (5)],

$$\|\pi_N \Upsilon^u_{T,0}\phi_1 - X^u_{(2N)}(T,0)\phi_1\| \leq \frac{(K_1+K_2)^{N+1}}{N!}.$$

All the controls we consider are such that $K_1 < 4$ and $K_2 < 4$. This is enough to guarantee that the error made when considering the Galerkin approximation of order 62 instead of the original infinite dimensional system is less than $\varepsilon = 10^{-5}$ for initial data the first eigenstate.

Simulations are straightforward with a standard desktop computer. We sum up some results in Table I.

V. CONCLUSIONS AND FUTURE WORKS

A. Conclusions

In this paper, we present a general approximate controllability result for infinite dimensional quantum systems when some polarizability term has to be considered in addition to the standard dipolar one. For the important case of transfer between two eigenstates of the free Hamiltonian, simple periodic control laws may be used. All our results are constructive. Numerical simulations on an physical example support our theoretical results.

B. Future Works

Many questions concerning the controllability of infinite dimensional quantum systems are still open. Among many other topics, one can cite the extension of the controllability results to systems involving higher power of the control, or the existence (and the estimation) of a minimal time needed to steer a quantum system from a given source to a given neighborhood of a given target.

VI. ACKNOWLEDGMENTS

It is a pleasure for the third author to thank Karine Beauchard for interesting discussions about the methodology used in [5].

This work has been partially supported by INRIA Nancy-Grand Est.

Second and third authors were partially supported by French Agence National de la Recherche ANR “GCM” program “BLANC-CSD”, contract number NT09-504590. The third author was partially supported by European Research Council ERC StG 2009 “GeCoMethods”, contract number 239748.

REFERENCES

[1] C. Dion, A. Bandrauk, O. Atabek, A. Keller, H. Umeda, and Y. Fujimura, “Two-frequency ir laser orientation of polar molecules. Numerical simulations for hcn.” *Chem. Phys. Lett.*, no. 302, pp. 215–223, 1999.

[2] C. Dion, O. Atabek, and A. Bandrauk, “Laserinduced alignment dynamics of hcn : Roles of the permanent dipole moment and the polarizability.” *Phys. Rev.*, no. 59, 1999.

[3] J.-M. Coron, A. Grigoriu, C. Lefter, and G. Turinici, “Quantum control design by lyapunov trajectory tracking for dipole and polarizability coupling,” *New. J. Phys.*, vol. 10, no. 11, 2009.

[4] A. Grigoriu, C. Lefter, and G. Turinici, “Lyapunov control of schrödinger equation:beyond the dipole approximations.” in *In Proc of the 28th IASTED International Conference on Modelling, Identification and Control*, 2009, pp. 119–123.

[5] M. Morancey, “Explicit approximate controllability of the Schrödinger equation with a polarizability term,” 2011, arXiv:1110.2860v1.

[6] Y. L. Sachkov, “Controllability of invariant systems on Lie groups and homogeneous spaces,” *J. Math. Sci. (New York)*, vol. 100, no. 4, pp. 2355–2427, 2000, dynamical systems, 8.

[7] A. A. Agrachev and Y. L. Sachkov, *Control theory from the geometric viewpoint*, ser. Encyclopaedia of Mathematical Sciences. Berlin: Springer-Verlag, 2004, vol. 87, control Theory and Optimization, II.

[8] N. Boussaïd, M. Caponigro, and T. Chambrion, “Weakly-coupled systems in quantum control,” arXiv:1109.1900v1. [Online]. Available: http://hal.archives-ouvertes.fr/hal-00620733/en/

[9] U. Boscain, M. Caponigro, T. Chambrion, and M. Sigalotti, “A weak spectral condition for the controllability of the bilinear Schrödinger equation with application to the control of a rotating planar molecule,” *Comm. Math. Phys.*, vol. 311, no. 2, pp. 423–455, 2012. [Online]. Available: http://dx.doi.org/10.1007/s00220-012-1441-z

[10] T. Chambrion, “Periodic excitations of bilinear quantum systems,” *Automatica J. IFAC*, vol. 48, no. 9, pp. 2040–2046, 2012.