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Thomas Chambrion

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# Periodic excitations of bilinear quantum systems

Thomas Chambrion\*

May 21, 2011

## Abstract

An usual way to transfer the population of a quantum system from an eigenspace of the free Hamiltonian to another is to use a periodic control law with angular frequency equal to the difference of the eigenvalues. This paper gives a theoretical explanation of the effectiveness of this widely used method. We introduce a notion of efficiency and demonstrate its interest for the design of control laws on the example of the rotation of a planar molecule.

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# 1 Introduction

## 1.1 Effective control of quantum systems

The state of a quantum system evolving on a finite dimensional Riemannian manifold  $\Omega$ , with associated measure  $\mu$ , is described by its *wave function*, that is, a point in the unit sphere of  $L^2(\Omega, \mathbf{C})$ . A system with wave function  $\psi$  is in a subset  $\omega$  of  $\Omega$  with the probability  $\int_{\omega} |\psi|^2 d\mu$ .

In the absence of interaction with the environment and with a suitable units choice, the time evolution of the wave function is given by the Schrödinger equation

$$i \frac{\partial \psi}{\partial t} = -\frac{1}{2} \Delta \psi + V(x) \psi(x, t),$$

where  $\Delta$  is the Laplace-Beltrami operator on  $\Omega$  (with suitable boundary conditions) and  $V : \Omega \rightarrow \mathbf{R}$  is a real function (usually called potential) accounting for the physical properties of the system. When submitted to an excitation by an external electric field (*e.g.* a laser), the Schrödinger equation reads

$$i \frac{\partial \psi}{\partial t} = -\frac{1}{2} \Delta \psi + V(x) \psi(x, t) + u(t) W(x) \psi(x, t), \quad (1)$$

where  $W : \Omega \rightarrow \mathbf{R}$  is a real function accounting for the physical properties of the laser and  $u$  is a real function of the time accounting for the intensity of the laser.

A natural question, with many practical implications, is whether there exists a control  $u$  that steers the quantum system from a given initial wave function to a given target wave function (controllability issue) and, more important, how to build this control law (effective design of controls).

Considerable efforts have been made by different communities to study the controllability of (1). We refer to [Tur00, Ner10, BM09, Mir09, BL10, BCCS11] and references therein for a description of the known theoretical results concerning the existence of controls steering a given source to a given target. As proved in [Ner10, MS10, PS10], approximate controllability is a generic property for systems of the type of (1).

Effective control algorithms have been obtained [WRD93, OKF98, BST08]. Most of the controls used in practice exhibit a remarkable pattern of periodic shape, with frequency corresponding to a resonance in the quantum system. They appear to work remarkably well, with little influence of the shape [BFS86]. However, to our knowledge, no theoretical proof of this effectiveness is available. The aim of the present paper is to provide a mathematically rigorous explanation of the surprising robustness of these control algorithms.

## 1.2 Framework and notations

We first reformulate the problem (1) in a more abstract framework. In a separable Hilbert space  $H$  endowed with norm  $\|\cdot\|$  and Hilbert product  $\langle \cdot, \cdot \rangle$ , we

consider the following evolution problem:

$$\frac{d\psi}{dt} = (A + uB)\psi \quad (2)$$

where  $(A, B)$  satisfies Assumption 1.

**Assumption 1.**  $(A, B)$  is a pair of linear operators such that

1.  $A$  is skew adjoint and has purely discrete spectrum  $(-i\lambda_k)_{k \in \mathbf{N}}$ ;
2. there exists an Hilbert basis  $(\phi_k)_{k \in \mathbf{N}}$  of  $H$  such that  $A\phi_k = -i\lambda_k\phi_k$  for every  $k$  in  $\mathbf{N}$ ;
3. the sequence  $(\lambda_n)_{n \in \mathbf{N}}$  takes value in  $(0, +\infty)$ , is non-decreasing and its only accumulation point is  $+\infty$ ;
4.  $B$  is skew adjoint and bounded;
5. for every  $u$  in  $\mathbf{R}$ ,  $A + uB$  is skew-adjoint,  $D(A + uB) = D(A)$  and  $D((A + uB)^2) = D(A^2)$ ;
6. there exists  $C_{A,B} \geq 0$  such that  $|\Im \langle A\psi, B\psi \rangle| \leq C_{A,B} |\langle A\psi, \psi \rangle|$  for every  $\psi$  in  $D(A)$ .

The Assumption 1.5 ensures that, for every constant  $u$  in  $\mathbf{R}$ ,  $A + uB$  generates a group of unitary propagators. Hence, for every initial condition  $\psi_0$  in  $H$ , for every piecewise constant control  $u$ , we can define the solution of (2) that we will note  $t \mapsto \Upsilon_t^u \psi_0$ .

**Proposition 2** (Well-posedness). *For every  $T$  in  $\mathbf{R}^+$ , for every  $\psi_0$  in  $H$ , the map  $u \mapsto \Upsilon_T^u \psi_0$  admits a unique continuous extension to the set  $L^1([0, T])$  of real valued integrable functions on  $[0, T]$ . If  $\psi_0$  belongs to  $D(A)$ , then for every  $u$  in  $L^1([0, T])$ , the curve  $t \mapsto \Upsilon_t^u \psi_0$  is absolutely continuous and, for almost every  $t$ ,*

$$\frac{d}{dt} \Upsilon_t^u \psi_0 = (A + u(t)B) \Upsilon_t^u \psi_0.$$

*Proof.* Since  $B$  is bounded (Assumption 1.4), the result of Proposition 2 is classical and we just give a sketch of the proof.

To begin with, the solutions of (2) can be constructed with the standard Banach fixed point theorem (see [BM09, Proposition 1.1] for an elementary proof). In the case where  $u$  is piecewise constant, this definition coincides with the one given above. This defines the map  $u \mapsto \Upsilon_{t,s}^u \psi_0$ , which, with every  $u$  in  $L^1([0, T])$ , associates the solution of (2) at time  $t$  with initial condition  $\psi_0$  at time  $s$ . In the following, we note  $\Upsilon_{t,0}^u = \Upsilon_t^u$ .

To see that  $u \mapsto \Upsilon_T^u \psi_0$  is continuous on  $L^1([0, T])$ , consider a sequence  $(u_n)_{n \in \mathbf{N}}$  converging to  $u$  in  $L^1([0, T])$ . If  $\psi_0$  belongs to  $D(A)$ , then the mappings  $t \mapsto \Upsilon_t^u \psi_0$  and  $t \mapsto \Upsilon_t^{u_n} \psi_0$  are absolutely continuous for every  $n$  and, for almost every  $t$ ,

$$\frac{d}{dt} \Upsilon_t^u \psi_0 = (A + u(t)B) \Upsilon_t^u \psi_0 \quad \frac{d}{dt} \Upsilon_t^{u_n} \psi_0 = (A + u_n(t)B) \Upsilon_t^{u_n} \psi_0.$$

In particular,  $t \mapsto \Upsilon_t^u \psi_0 - \Upsilon_t^{u_n} \psi_0$  is absolutely continuous and, for almost every  $t$ ,

$$\frac{d}{dt} (\Upsilon_t^u \psi_0 - \Upsilon_t^{u_n} \psi_0) = (A + u(t)B) (\Upsilon_t^u \psi_0 - \Upsilon_t^{u_n} \psi_0) + (u(t) - u_n(t)) B \Upsilon_t^{u_n} \psi_0.$$

Conclusion follows from Duhamel formula and unitarity of  $\Upsilon_{t,\tau}^u$ :

$$\|(\Upsilon_t^u - \Upsilon_t^{u_n})\psi_0\| = \left\| \int_0^t (u(\tau) - u_n(\tau)) \Upsilon_{t,\tau}^u B \Upsilon_\tau^{u_n} \psi_0 d\tau \right\| \leq \|u_n - u\|_{L^1} \|B\| \|\psi_0\|.$$

In the general case, where  $\psi_0 \notin D(A)$ , pick  $\tilde{\psi}_0$  in  $D(A)$  and write

$$\begin{aligned} \Upsilon_T^{u_n} \psi_0 - \Upsilon_T^u \psi_0 &= \Upsilon_T^{u_n} \psi_0 - \Upsilon_T^{u_n} \tilde{\psi}_0 + \Upsilon_T^{u_n} \tilde{\psi}_0 - \Upsilon_T^u \tilde{\psi}_0 + \Upsilon_T^u \tilde{\psi}_0 - \Upsilon_T^u \psi_0 \\ &= \Upsilon_T^{u_n} (\psi_0 - \tilde{\psi}_0) + \Upsilon_T^{u_n} \tilde{\psi}_0 - \Upsilon_T^u \tilde{\psi}_0 + \Upsilon_T^u (\tilde{\psi}_0 - \psi_0) \end{aligned}$$

Recall that  $\Upsilon_T^{u_n}$  and  $\Upsilon_T^u$  are unitary operators of  $H$ , hence

$$\|\Upsilon_T^{u_n} \psi_0 - \Upsilon_T^u \psi_0\| \leq 2\|\tilde{\psi}_0 - \psi_0\| + \|\Upsilon_T^{u_n} \tilde{\psi}_0 - \Upsilon_T^u \tilde{\psi}_0\|,$$

which tends to zero as  $u_n$  tends to  $u$  and  $\tilde{\psi}_0 - \psi_0$  tends to zero.  $\square$

### 1.3 Main result

**Definition 1.** A point  $(j, k)$  of  $\mathbf{N}^2$  is said to be a *non-resonant transition* of  $(A, B)$  if (i)  $j \neq k$ , (ii)  $\langle \phi_j, B\phi_k \rangle \neq 0$  and (iii) for every  $l, m$  in  $\mathbf{N}$ ,  $|\lambda_j - \lambda_k| = |\lambda_l - \lambda_m|$  implies  $\{j, k\} = \{l, m\}$  or  $\langle \phi_l, B\phi_m \rangle = 0$ . A non-resonant transition  $(j, k)$  of  $(A, B)$  is *strongly non-resonant* if for every  $l, m$  in  $\mathbf{N}$ ,  $|\lambda_l - \lambda_m| \in \mathbf{N}|\lambda_j - \lambda_k|$  implies  $\{j, k\} = \{l, m\}$ .

**Theorem 1.** Let  $u^* : \mathbf{R}^+ \rightarrow \mathbf{R}$  be a locally integrable function. Assume that  $u^*$  is periodic with period  $T = \frac{2\pi}{|\lambda_j - \lambda_k|}$  for some non resonant transition  $(j, k)$  of  $(A, B)$ , and that

$$\int_0^T u^*(\tau) e^{i(\lambda_l - \lambda_m)\tau} d\tau = 0$$

for every  $l, m$  such that  $\{l, m\} \neq \{j, k\}$ ,  $\frac{\lambda_l - \lambda_m}{\lambda_j - \lambda_k} \in \mathbf{Z}$  and  $\langle \phi_l, B\phi_m \rangle \neq 0$ . If

$$\int_0^T u^*(\tau) e^{i(\lambda_j - \lambda_k)\tau} d\tau \neq 0,$$

then there exists  $T^* > 0$  such that the sequence  $\left( \left| \langle \phi_k, \Upsilon_{nT^*}^{\frac{u^*}{n}}(\phi_j) \rangle \right| \right)_{n \in \mathbf{N}}$  tends to 1 as  $n$  tends to infinity.

This result provides a rigorous formulation of a well-known fact: to induce a transition between levels  $j$  and  $k$  of a quantum system, one can use a periodic excitation of frequency exactly equal to the difference of the corresponding eigenvalues. In the case of a strongly non-resonant transition, for almost every shape of the control, the trajectory eventually reaches any neighborhood of the target, provided the control is small enough and has the correct frequency.

## 1.4 Content of the paper

The paper splits in two parts. The first one is a theoretical proof of Theorem 1. Some technical tools are introduced in Section 2.1. A time reparametrization (Section 2.2) allows to prove Theorem 1 and to provide an explicit expression of the time  $T^*$  (Section 2.3). In a second part, the theoretical results are tested on numerical simulations of the rotation of a planar molecule (Sections 3.2 and 3.3).

## 2 Proof of the convergence result

The strategy of the proof is inspired by [CMSB09, BCCS11] and relies upon the approximation of the original infinite dimensional system by its finite dimensional approximations.

### 2.1 Good Galerkin approximation

In this Section, we explain how to construct a good Galerkin approximation of the original system. The term “good” refers to the fact that the error made when replacing the original system by its Galerkin approximation is bounded uniformly with respect to the control  $u$ . What follows is a very simplified version of a much more general construction (valid also when operator  $B$  is not bounded or when the eigenvalues of  $A$  accumulate) presented in [BCC11].

From Assumption 1.3,  $-iA$  is a self-adjoint operator of  $H$  with positive eigenvalues. For every  $\psi$  in  $D(A)$ ,  $-iA\psi = \sum_{j \in \mathbf{N}} \lambda_j \langle \phi_j, \psi \rangle \phi_j$ . For every  $s > 0$ , the linear operator  $(-iA)^s = |A|^s$  is defined by  $(-iA)^s \psi = \sum_{j \in \mathbf{N}} \lambda_j^s \langle \phi_j, \psi \rangle \phi_j$ , for every  $\psi$  in  $D((-iA)^s)$ .

**Proposition 3.** *For every  $\psi_0 \in D(A)$  and  $K > 0$ , there exists  $C_K$  such that for every  $T \geq 0$  and for every control  $u$  for which  $\|u\|_{L^1} < K$ , one has*

$$|\langle A\Upsilon_T^u(\psi_0), \Upsilon_T^u(\psi_0) \rangle| < C_K.$$

*Proof.* We will show a stronger result, valid for  $\psi_0$  in  $D(|A|^{1/2})$ .

Consider first a piecewise constant control  $u$ , and let  $x(t)$  be the solution of  $\dot{x} = (A + u(t)B)x$  with initial condition  $\psi_0$  in  $D(A^2)$ . By Assumption 1.5,  $x(t)$  belongs to  $D(A^2)$  for every  $t$ . Consider the real mapping  $f : t \mapsto |\langle Ax(t), x(t) \rangle|$ . Since  $x(0)$  belongs to  $D(A)$ ,  $f$  is differentiable almost everywhere and, for a.e.  $t$ ,

$$\begin{aligned} \frac{d}{dt} f(t) &= i \frac{d}{dt} \langle Ax(t), x(t) \rangle \\ &= i \langle Ax(t), (A + u(t)B)x(t) \rangle + i \langle A(A + u(t)B)x(t), x(t) \rangle \\ &= 2i^2 \Im \langle Ax(t), (A + u(t)B)x(t) \rangle \\ &= -2 \Im \langle Ax(t), u(t)Bx(t) \rangle, \end{aligned}$$

so that, thanks to Assumption 1.6,

$$|f'(t)| \leq 2|u(t)| |\langle Ax(t), Bx(t) \rangle| \leq C_{A,B} |u(t)| f(t).$$

Gronwall lemma implies that

$$|\langle Ax(t), x(t) \rangle| \leq e^{C_{A,B} \int_0^t |u(\tau)| d\tau} |\langle A\psi_0, \psi_0 \rangle| = e^{C_{A,B} \int_0^t |u(\tau)| d\tau} \| |A|^{\frac{1}{2}} \psi_0 \|^2. \quad (3)$$

Let  $(\psi_0^n)_{n \in \mathbf{N}}$  be a sequence in  $D(A^2)$  with limit  $\psi_0$  in  $D(|A|^{\frac{1}{2}})$ . For every  $\tilde{\psi}$  in  $D(|A|^{\frac{1}{2}})$ , for every  $t \geq 0$ ,

$$|\langle |A|^{\frac{1}{2}} \tilde{\psi}, \Upsilon_t^u(\psi_0^n) \rangle| \leq |\langle \tilde{\psi}, |A|^{\frac{1}{2}} \Upsilon_t^u(\psi_0^n) \rangle| \leq \|\tilde{\psi}\| \| |A|^{\frac{1}{2}} \Upsilon_t^u(\psi_0^n) \|,$$

which is bounded uniformly with respect to  $n$  (see (3)).

Since  $\psi \mapsto \Upsilon_t^u(\psi)$  is unitary,  $(\Upsilon_t^u(\psi_0^n))_{n \in \mathbf{N}}$  converges to  $\Upsilon_t^u(\psi_0)$  and

$$|\langle |A|^{\frac{1}{2}} \tilde{\psi}, \Upsilon_t^u(\psi_0) \rangle| \leq \|\tilde{\psi}\| e^{C_{A,B} \int_0^t |u(\tau)| d\tau} \| |A|^{\frac{1}{2}} \psi_0 \|^2.$$

Hence,  $\Upsilon_t^u(\psi_0)$  belongs to  $D(|A|^{\frac{1}{2}*}) = D(|A|^{\frac{1}{2}})$  and

$$\| |A|^{\frac{1}{2}} \Upsilon_t^u(\psi_0) \|^2 \leq e^{C_{A,B} \int_0^t |u(\tau)| d\tau} \| |A|^{\frac{1}{2}} \psi_0 \|^2.$$

If  $u$  is a general function in  $L^1$ , let  $(u_n)_{n \in \mathbf{N}}$  be sequence of piecewise function tending to  $u$  and such that  $\|u_n\|_{L^1} \leq \|u\|_{L^1}$  for every  $n$ . For every  $\psi$  and  $\tilde{\psi}$  in  $D(|A|^{\frac{1}{2}})$ , for every  $t \geq 0$ ,

$$|\langle |A|^{\frac{1}{2}} \tilde{\psi}, \Upsilon_t^{u_n}(\psi_0) \rangle| \leq |\langle \tilde{\psi}, |A|^{\frac{1}{2}} \Upsilon_t^{u_n}(\psi_0) \rangle| \leq \|\tilde{\psi}\| \| |A|^{\frac{1}{2}} \Upsilon_t^{u_n}(\psi_0) \|,$$

is uniformly bounded with respect to  $n$  and the conclusion follows from the same argument as above.  $\square$

**Remark 1.** The quantity  $|\langle A \Upsilon_T^u(\psi_0), \Upsilon_T^u(\psi_0) \rangle|$  is sometimes called *expected value of the energy* in physics textbooks. Notice that its boundedness is a deep obstruction to exact controllability in  $H$  of system (2) with  $L^1$  controls.

For every  $N$  in  $\mathbf{N}$ , we define the orthogonal projection

$$\begin{aligned} \pi_N : H &\rightarrow H \\ \psi &\mapsto \sum_{j \leq N} \langle \phi_j, \psi \rangle \phi_j \end{aligned}$$

**Proposition 4.** For every  $\epsilon > 0$ ,  $K \geq 0$ ,  $n \in \mathbf{N}$ , there exists  $N \in \mathbf{N}$  such that for every control  $u$ , for every  $0 \leq s < 1$ ,

$$\|u\|_{L^1} \leq K \implies |\langle (-iA)^s (\text{Id} - \pi_N) \Upsilon_t^u(\phi_j), (\text{Id} - \pi_N) \Upsilon_t^u(\phi_j) \rangle| < \epsilon \quad (4)$$

for every  $t \geq 0$ , and  $j = 1, \dots, n$ . As a consequence, there exists  $N \in \mathbf{N}$  such that for every control  $u$ ,

$$\|u\|_{L^1} \leq K \implies \|B(\text{Id} - \pi_N) \Upsilon_t^u(\phi_j)\| < \epsilon, \quad (5)$$

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

*Proof.* Fix  $j \in \{1, \dots, n\}$ . For every  $N > 1$ , one has

$$\begin{aligned} | \langle (-iA)^s (\text{Id} - \pi_N) \Upsilon_t^u(\phi_j), (\text{Id} - \pi_N) \Upsilon_t^u(\phi_j) \rangle | &= \sum_{n=N+1}^{\infty} \lambda_n^s |\langle \phi_n, \Upsilon_t^u(\phi_j) \rangle|^2 \\ &\leq \lambda_N^{s-1} \sum_{n=N+1}^{\infty} \lambda_n |\langle \phi_n, \Upsilon_t^u(\phi_j) \rangle|^2 \\ &\leq \lambda_N^{s-1} |\langle A \Upsilon_t^u(\phi_j), \Upsilon_t^u(\phi_j) \rangle|. \end{aligned}$$

By Proposition 3, there exists  $C_K$  such that  $|\langle A \Upsilon_t^u(\phi_j), \Upsilon_t^u(\phi_j) \rangle| \leq C_K$  for every  $t$  in  $\mathbf{R}^+$  and  $u$  in the  $K$  ball of  $L^1$ . Since  $s < 1$  then  $|\langle (-iA)^s (\text{Id} - \pi_N) \Upsilon_t^u(\phi_j), (\text{Id} - \pi_N) \Upsilon_t^u(\phi_j) \rangle|$  tends to 0, uniformly with respect to  $u$ , as  $N$  tends to infinity. Equation (5) is a direct consequence of (4) since  $B$  is bounded.  $\square$

**Definition 2.** Let  $N \in \mathbf{N}$ . Denote  $\mathcal{L}_N = \text{span}(\phi_1, \dots, \phi_N)$ . The Galerkin approximates of  $A$  and  $B$  of order  $N$  are the operators  $A^{(N)} : H \rightarrow H$  and  $B^{(N)} : H \rightarrow H$  defined by

$$A^{(N)} = \pi_N A|_{\mathcal{L}_N} \quad \text{and} \quad B^{(N)} = \pi_N B|_{\mathcal{L}_N}.$$

We define the system  $(\Sigma_N)$  as

$$\dot{x} = (A^{(N)} + uB^{(N)})x \tag{\Sigma_N}$$

and denote  $X_{t,s}^{(N),u}$  (or simply  $X_t^{(N),u}$  when  $s = 0$ ) the propagator of  $(\Sigma_N)$ .

With an obvious abuse of notation, we will sometimes identify the operators  $A^{(N)}$  and  $B^{(N)}$  with their restrictions to the invariant space  $\text{span}_{1 \leq l \leq N} \{\phi_l\}$  and with their matrices in the basis  $(\phi_l)_{1 \leq l \leq N}$ . Entries of  $B^{(N)}$  are denoted  $(b_{l_1, l_2} = \langle \phi_{l_1}, B \phi_{l_2} \rangle)_{l_1, l_2}$ . With these identifications,  $(\Sigma_N)$  turns into a finite dimensional system in  $\mathbf{C}^N$ .

**Proposition 5** (Good Galerkin Approximation). *For every  $\epsilon > 0$ ,  $K \geq 0$ ,  $n \in \mathbf{N}$ , there exists  $N \in \mathbf{N}$  such that for every  $u \in L^1(0, \infty)$*

$$\|u\|_{L^1} \leq K \implies \|\Upsilon_t^u(\phi_j) - X_t^{(N),u} \phi_j\| < \epsilon, \tag{6}$$

for every  $t \geq 0$  and  $i = 1, \dots, n$ .

*Proof.* Let  $\epsilon > 0$ ,  $K \geq 0$ ,  $n \in \mathbf{N}$  be given. By Proposition 4, there exists  $N \in \mathbf{N}$  such that, for every  $u \in L^1(0, \infty)$  for which  $\|u\|_{L^1} \leq K$ , we have

$$\|B(\text{Id} - \pi_N) \Upsilon_t^u(\phi_j)\| < \frac{\epsilon}{K},$$

for every  $t \geq 0$  and  $j = 1, \dots, n$ . Fix  $j$  in  $1, \dots, n$  and consider  $y_j : t \mapsto \pi_N \Upsilon_t^u(\phi_j)$ . The mapping  $t \mapsto y_j(t)$  is absolutely continuous and, for almost every  $t \geq 0$ ,

$$\dot{y}_j(t) = (A^{(N)} + uB^{(N)})y_j(t) + u(t)\pi_N B(\text{Id} - \pi_N) \Upsilon_t^u(\phi_j).$$



Hence, for every  $t \geq 0$ ,

$$y_j(t) = X_t^{(N),u} \phi_j + \int_0^t X_{t,s}^{(N),u} \pi_N B (\text{Id} - \pi_N) \Upsilon_s^u(\phi_j) u(\tau) d\tau \quad (7)$$

The norm of  $t \mapsto B(\text{Id} - \pi_N) \Upsilon_t^u(\phi_j)$  is less than or equal to  $\epsilon/K$  for every  $t \geq 0$  and, since  $X_{t,s}^{(N),u}$  is unitary,  $\|y_j(t) - X_t^{(N),u}(\phi_j)\| \leq \epsilon \|u\|_{L^1} / K \leq \epsilon$ .  $\square$

## 2.2 Time reparametrization

We note  $PC$  the set of the piecewise constant functions for which there exist two sequences  $(u_j)_{1 \leq j \leq p}$  and  $(t_j)_{1 \leq j \leq p}$  with value in  $(0, +\infty)$  such that

$$u = \sum_{1 \leq j \leq p+1} u_j \chi_{[\tau_j, \tau_j + t_j)},$$

where  $\chi$  is the characteristic function and the sequence  $(\tau_j)_{1 \leq j \leq p+1}$  is defined by induction:  $\tau_1 = 0$ ,  $\tau_{j+1} = \tau_j + t_j$ . An element  $u$  of  $PC$  will be denoted  $(u_j, t_j)_{1 \leq j \leq p}$ .

The involutive mapping

$$\begin{aligned} \mathcal{P} : PC &\rightarrow PC \\ (u_j, t_j)_{1 \leq j \leq p} &\mapsto \left( \frac{1}{u_j}, u_j t_j \right)_{1 \leq j \leq p} \end{aligned}$$

performs a time reparametrization of the system (2). Indeed, introduce the control system

$$\frac{d\psi}{dt} = (u(t)A + B)\psi(t), \quad (8)$$

whose solution with initial condition  $\psi_0$  will be denoted by  $t \mapsto \tilde{\Upsilon}_t^u \psi_0$ .

**Proposition 6.** *For every  $\psi_0$  in  $H$  and every  $u$  in  $PC$ ,  $\tilde{\Upsilon}_{\int_0^t u(\tau) d\tau}^u \psi_0 = \Upsilon_t^u \psi_0$ .*

*Proof.* This follows from the equality  $e^{t(A+uB)} = e^{tu(\frac{1}{u}A+B)}$ , valid on every interval where  $u$  is constant.  $\square$

## 2.3 Proof of Theorem 1

Let  $\epsilon > 0$ ,  $u^*$  a non vanishing piecewise constant function,  $T$ -periodic,  $j < k$  two integers be given as in the hypothesis of Theorem 1.

The function  $t \mapsto \int_0^t |u^*(\tau)| d\tau$  is non-decreasing. We denote with  $v^*$  its reciprocal function. We define also  $I = \int_0^T |u^*(\tau)| d\tau$  or, equivalently,  $v^*(I) = T$ .

By hypothesis,  $\int_0^T u^*(\tau) \exp[i(\lambda_j - \lambda_k)\tau] d\tau \neq 0$ . Define

$$K = \frac{\pi I}{2 \left| \int_0^T u^*(\tau) e^{i(\lambda_j - \lambda_k)\tau} d\tau \right| |b_{j,k}|}.$$

By Proposition 5, there exists  $N$  in  $\mathbf{N}$  such that for every  $l \leq k$ , for every  $u$ , for every  $t \geq 0$ ,

$$\|u\|_{L^1} < K \implies \|\Upsilon_t^u(\phi_l) - X_t^{(N),u}\phi_l\| < \epsilon,$$

where  $X^{(N),u}$  is the propagator associated with the control system  $(\Sigma_N)$ .

We introduce the sequence  $u_n = \frac{1}{n}u^*$ . For every  $n$  in  $\mathbf{N}$ ,  $u_n$  is a non-vanishing  $T$ -periodic function and  $\int_0^{nT} |u_n(\tau)|d\tau = \int_0^T |u^*(\tau)|d\tau$ . For every  $n$ , for every  $t$ ,

$$\mathcal{P}|u_n|(t) = n\mathcal{P}|u^*|(nt).$$

In particular,  $\mathcal{P}|u_n|$  is a  $\frac{I}{n}$  periodic function and

$$\int_0^{\frac{I}{n}} \mathcal{P}|u_n|(\tau)d\tau = \int_0^I \mathcal{P}|u^*|(\tau)d\tau = T.$$

The primitive  $v_n$  of  $\mathcal{P}|u_n|$  taking value 0 at 0,

$$v_n : t \mapsto \int_0^t \mathcal{P}|u_n|(\tau)d\tau = v^*(nt),$$

satisfies, for every  $t$  in  $\mathbf{R}^+$ ,

$$v_n\left(t + \frac{I}{n}\right) = v_n(t) + \int_0^{I/n} \mathcal{P}|u_n|(\tau)d\tau = v_n(t) + T.$$

Equivalently, one can define  $v_n$  as the reciprocal of the increasing function  $t \mapsto \int_0^t |u_n(\tau)|d\tau$ .

Let us note  $t \mapsto x_n(t) = X_t^{(N),u_n}\phi_j$  the solution of

$$\dot{x} = (A^{(N)} + u_n(t)B^{(N)})x(t) \quad (9)$$

with initial condition  $x(0) = \phi_j$ .

The set  $[0, T]$  can be written as a finite union of disjoint intervals

$$[0, T] = J_1^+ \cup J_1^- \cup \dots \cup J_p^+ \cup J_p^-,$$

such that  $u^*$  takes positive (resp. negative) values on  $J^+ = \cup_{l=1}^p J_l^+$  (resp.  $J^- = \cup_{l=1}^p J_l^-$ ). For  $t$  in  $J^-$ , (9) writes

$$\frac{dx}{dt} = (A^{(N)} + (-u_n(t))(-B^{(N)}))x(t).$$

We apply the  $\mathcal{P}$  reparametrization to the positive function  $|u^*|$  separately on every intervals in  $J^+$  and  $J^-$ . Defining the sets  $G_n^+ = (v_n)^{[-1]}(J^+) = \{l \in \mathbf{R}^+ | \exists s \in J^+, \int_0^s |u_n(\tau)|d\tau = l\}$  and  $G_n^- = (v_n)^{[-1]}(J^-) = \{l \in \mathbf{R}^+ | \exists s \in J^-, \int_0^s |u_n(\tau)|d\tau = l\}$ , we obtain the dynamics of  $y_n = x_n \circ v_n$ :

$$\frac{dy_n}{dt} = \begin{cases} (\mathcal{P}(|u_n|)(t)A^{(N)} + B^{(N)})y_n(t) & \text{for almost every } t \in G^+ \\ (\mathcal{P}(|u_n|)(t)A^{(N)} - B^{(N)})y_n(t) & \text{for almost every } t \in G^- \end{cases}, \quad (10)$$

For every  $t$ , we define  $z_n(t)$  as

$$z_n(t) = \exp\left(\left[-\int_0^t \mathcal{P}|u_n|(\tau) d\tau\right] A^{(N)}\right) y_n(t) = e^{-v_n(t)A} y_n(t).$$

Notice that, for every  $t$ , for every  $l$  in  $\mathbf{N}$ ,  $|\langle \phi_l, z_n(t) \rangle| = |\langle \phi_l, y_n(t) \rangle|$ .

Finally, we define the time varying  $N \times N$  matrix  $M_n$  equal to

$$M_n : t \mapsto \text{sg}(u_n \circ v_n) e^{-v_n(t)A^{(N)}} B^{(N)} e^{v_n(t)A^{(N)}}.$$

From (10), one deduces the dynamics of  $z_n$ , valid for almost every  $t$  in  $G_n^+ \cup G_n^-$ :

$$\frac{dz_n}{dt} = M_n(t) z_n(t). \quad (11)$$

We denote  $Y_t^n$  the propagator associated with (11). Notice that, for every  $k$ , the mapping  $t \mapsto \langle \phi_k, z_n \rangle$  is Lipschitz continuous with Lipschitz constant  $\|B\phi_k\|$ .

**Lemma 7.** *Let  $M^\dagger$  be the constant  $N \times N$  matrix whose entries are, for  $1 \leq l_1, l_2 \leq N$ ,  $m_{l_1, l_2}^\dagger = (b_{l_1, l_2}/I) \int_0^I \exp(i(\lambda_{l_1} - \lambda_{l_2})v^*(\tau)) d\tau$  if  $T(\lambda_{l_1} - \lambda_{l_2}) \in 2\pi\mathbf{Z}$  and zero else. Then, for every  $t$  in  $\mathbf{R}^+$ , the sequence  $(Y_t^n)_n$  tends to  $\exp(tM^\dagger)$  as  $n$  tends to infinity.*

*Proof of Lemma 7.* For every  $t$  in  $[0, I]$ , for every  $n$  in  $\mathbf{N}$ , defining the integer  $s = \lfloor \frac{tn}{I} \rfloor \in (\frac{tn}{I} - 1, \frac{tn}{I}]$ ,

$$\begin{aligned} \int_0^t \text{sg}(u_n \circ v_n) e^{i(\lambda_{l_1} - \lambda_{l_2})v_n(\tau)} d\tau = \\ \int_0^{sI/n} \text{sg}(u_n \circ v_n) e^{i(\lambda_{l_1} - \lambda_{l_2})v_n(\tau)} d\tau + \int_{sI/n}^t \text{sg}(u_n \circ v_n) e^{i(\lambda_{l_1} - \lambda_{l_2})v_n(\tau)} d\tau. \end{aligned}$$

Observe that  $\left| \int_{sI/n}^t \text{sg}(u_n \circ v_n) e^{i(\lambda_{l_1} - \lambda_{l_2})v_n(\tau)} d\tau \right| \leq I/n$  tends to zero, uniformly with respect to  $t$ , as  $n$  tends to infinity.

For every  $l_1, l_2$  such that  $T(\lambda_{l_1} - \lambda_{l_2}) \notin 2\pi\mathbf{Z}$ ,

$$\begin{aligned} & \left| \int_0^{sI/n} \text{sg}(u_n \circ v_n) e^{i(\lambda_{l_1} - \lambda_{l_2})v_n(\tau)} d\tau \right| \\ & \leq \left| \sum_{m=1}^s \int_0^{I/n} \text{sg}(u_n \circ v_n) e^{i(\lambda_{l_1} - \lambda_{l_2})(v_n(\tau) + mT)} d\tau \right| \\ & \leq \left| \int_0^{I/n} \text{sg}(u_n \circ v_n) e^{i(\lambda_{l_1} - \lambda_{l_2})v_n(\tau)} \sum_{m=1}^s e^{i(\lambda_{l_1} - \lambda_{l_2})mT} d\tau \right| \\ & \leq \left( \frac{2}{|1 - \exp(iT(\lambda_{l_1} - \lambda_{l_2}))|} \right) \frac{I}{n} \end{aligned}$$

tends to zero, uniformly with respect to  $t$ , as  $n$  tends to infinity.

If  $T(\lambda_{l_1} - \lambda_{l_2}) \in 2\pi\mathbf{Z}$ , then

$$\begin{aligned} \sum_{m=1}^s \int_0^{I/n} \text{sg}(u_n \circ v_n) e^{i(\lambda_{l_1} - \lambda_{l_2})(v_n(\tau) + mT)} d\tau &= s \int_0^{I/n} \text{sg}(u_n \circ v_n) e^{i(\lambda_{l_1} - \lambda_{l_2})v_n(\tau)} d\tau \\ &= \frac{s}{n} \int_0^I \text{sg}(u^* \circ v^*) e^{i(\lambda_{l_1} - \lambda_{l_2})v^*(\tau)} d\tau. \end{aligned}$$

This last quantity tends to

$$\frac{t}{I} \int_0^I \text{sg}(u^* \circ v^*) e^{i(\lambda_{l_1} - \lambda_{l_2})v^*(\tau)} d\tau = \frac{t}{I} \int_0^I u^*(\tau) e^{i(\lambda_{l_1} - \lambda_{l_2})\tau} d\tau,$$

as  $n$  tends to infinity, uniformly with respect to  $t$  in  $[0, I]$ .

Hence,  $t \mapsto \int_0^t M_n(\tau) d\tau$  tends to  $t \mapsto tM^\dagger$  uniformly with respect to  $t$  in  $[0, I]$ , as  $n$  tends to infinity. From [AS04, Lemma 8.10], the solution of  $\dot{x} = M_n(t)x$  with initial condition  $x_0$  tends uniformly with respect to  $t$  in  $[0, I]$  to the solution of  $\dot{x} = M^\dagger x$  with initial condition  $x_0$ .  $\square$

As a consequence of Lemma 7, for every  $t$ ,  $z_n(t)$  tends to  $\exp(tM^\dagger)\phi_j$  as  $n$  tends to infinity. By hypothesis of Theorem 1, all non diagonal entries of  $M^\dagger$  are zero but the entries with indices  $(j, k)$  and  $(k, j)$ . Hence,  $|\langle \phi_k, z_n(t) \rangle| = |\langle \phi_k, x_n(v_n(t)) \rangle|$  tends to

$$\left| \sin \left( \frac{t|b_{j,k}|}{I} \left| \int_0^T u^*(\tau) e^{i(\lambda_j - \lambda_k)\tau} d\tau \right| \right) \right|$$

as  $n$  tends to infinity. Choosing

$$T_n^* = v^* \left( \frac{n\pi I}{2|b_{j,k}| \left| \int_0^T u^*(\tau) e^{i(\lambda_j - \lambda_k)\tau} d\tau \right|} \right),$$

one gets that  $|\langle \phi_k, x_n(T_n^*) \rangle|$  tends to 1 as  $n$  tends to infinity.

By definition of  $v^*$ , for every  $n$  in  $\mathbf{N}$ ,

$$\int_0^{T_n^*} |u_n(\tau)| d\tau \leq \frac{\pi I}{2|b_{j,k}| \left| \int_0^T u^*(\tau) e^{i(\lambda_j - \lambda_k)\tau} d\tau \right|} = K.$$

By definition of  $N$ , from Proposition 5,  $\langle \phi_k, x_n(T_n^*) \rangle$  is  $\epsilon$ -close to  $\langle \phi_k, \Upsilon_{T_n^*}^{\frac{u}{n}} \phi_j \rangle$  for every  $n$  in  $\mathbf{N}$ , hence  $|\langle \phi_k, \Upsilon_{T_n^*}^{\frac{u}{n}} \phi_j \rangle|$  belongs to an  $\epsilon$ -neighborhood of 1 for  $n$  large enough. Letting  $\epsilon$  tends to zero, one gets that the sequence  $\left( |\langle \phi_k, \Upsilon_{T_n^*}^{\frac{u}{n}} \phi_j \rangle| \right)_{n \in \mathbf{N}}$  tends to one as  $n$  tends to infinity.

Knowing that  $v^*$  is non-decreasing and  $v^*(lI) = lT$  for every  $l$  in  $\mathbf{N}$ , one deduces the asymptotic behavior of  $T_n^*$  as  $n$  tends to infinity,  $nT^* \leq T_n^* \leq (n+1)T^*$  where

$$T^* = \frac{\pi T}{2|b_{j,k}| \left| \int_0^T u^*(\tau) e^{i(\lambda_j - \lambda_k)\tau} d\tau \right|}.$$

Notice finally that the mapping  $t \mapsto |\langle \phi_k, x_n(t) \rangle| = |\langle \phi_k, z_n \circ (v_n^*)^{[-1]}(t) \rangle|$  has Lipschitz constant less than  $\|B\phi_k\| \sup |u^*|/n$ . This completes the proof of Theorem 1 in the case where  $u^*$  is a non vanishing piecewise constant function.

If  $u^*$  is a locally integrable  $T$ -periodic function, let  $(u^{*,l})_{l \in \mathbf{N}}$  be a sequence of non-vanishing piecewise constant  $T$ -periodic functions converging to  $u^*$  in  $L^1_{loc}$  with  $\|u^{*,l}\|_{L^1([0,T])} \leq \|u\|_{L^1([0,T])}$  for every  $l$ . We define, for every  $n$ ,  $u_{n,l} = \frac{u^{*,l}}{n}$ ,  $v_{n,l} : t \mapsto \int_0^t \mathcal{P}|u_{n,l}|(\tau) d\tau$  and  $M_{n,l}(t) = \text{sg}(u_{n,l} \circ v_{n,l}(t)) e^{-v_{n,l}(t)A} B^{(N)} e^{v_{n,l}(t)A}$ . For every  $t$ , the matrix  $\int_0^t M_{l,l}(\tau) d\tau$  tends to  $tM^\dagger$ , uniformly with respect to  $t$  in a compact set, as  $l$  tends to infinity. Hence the solutions of  $\dot{x} = M_{l,l}(t)x$  tend to the solutions of  $\dot{x} = M^\dagger x$ , uniformly with respect to the time in a compact interval, as  $l$  tends to infinity. That concludes the proof of Theorem 1.

## 2.4 Efficiency of the transfer

Keeping with the notations of the last paragraph, for every periodic  $u^*$  with period  $T = \frac{2\pi}{|\lambda_j - \lambda_k|}$ , we define the efficiency of  $u^*$  with respect to the transition  $(j, k)$  as the real quantity:

$$E^{(j,k)}(u^*) = \frac{1}{I} \left| \int_0^I \text{sg}(u^* \circ v^*) e^{(\lambda_j - \lambda_k)v^*(\tau)} d\tau \right| = \frac{\left| \int_0^T u^*(\tau) e^{i(\lambda_j - \lambda_k)\tau} d\tau \right|}{\int_0^T |u^*(\tau)| d\tau}.$$

For every  $u$ ,  $0 \leq E^{(j,k)}(u) \leq 1$ . For every  $\{j, k\}$ ,  $\sup_u E^{(j,k)}(u) = 1$  (consider a sequence of controls that tends to a periodic sum of Dirac functions). An example of  $u^*$  with zero efficiency is presented in Section 3.

An intuitive explanation of the efficiency could be the following: asymptotically, the  $L^1$  norm of the control needed to induce the transition between levels  $j$  and  $k$  using periodic controls of the form  $u_n$  is equal to  $\pi/(2|b_{j,k}|E^{(j,k)}(u^*))$ .

## 2.5 Design of control laws

The system (2) being given, the design of an effective control law fulfilling the hypotheses of Theorem 1 is an important practical issue. To generate a transfer from level  $j$  to level  $k$ , one should chose a control  $u$  such that  $E^{(j,k)}(u)$  be as large as possible and  $E^{(l_1, l_2)}(u)$  be zero (or arbitrarily close to zero) for every  $l_1, l_2$  such that  $\lambda_{l_1} - \lambda_{l_2} \in (\lambda_j - \lambda_k)\mathbf{Z}$ . The algorithm we have described in [BCCS11] allows to build  $u$  such that  $E^{(j,k)}(u) > 0.43$ , with  $E^{(l_1, l_2)}(u)$  arbitrarily small for every finite number of pairs  $\{l_1, l_2\}$  satisfying  $\{l_1, l_2\} \neq \{j, k\}$  and  $|\lambda_{l_1} - \lambda_{l_2}| \neq |\lambda_j - \lambda_k|$ .

## 3 Rotation of a planar molecule

In this Section, we apply our results to the well studied example of the rotation of a planar molecule (see [Bou99, ST05, BCM+09, BCCS11]).

### 3.1 Presentation of the model

We consider a linear molecule with fixed length and center of mass. 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, about 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 writes

$$i\frac{\partial\psi}{\partial t}(t, \theta) = -\Delta\psi(t, \theta) + u(t)\cos(\theta)\psi(t, \theta), \quad (12)$$

where  $\Delta$  is the Laplace-Beltrami operator on  $\Omega$ . 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 to the constant functions. The eigenvalue  $k^2$  for  $k > 0$  is associated to 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  $H_o$ , respectively the spaces of even and odd functions of  $H$ . The spaces  $H_e$  and  $H_o$  are stable under the dynamics of (12), hence no global controllability is to be expected in  $H$ .

### 3.2 Strongly non-resonant case

We first concentrate on the space  $H_o$ . The restriction  $A$  of  $i\Delta$  to  $H_o$  is skew adjoint, with simple eigenvalues  $(ik^2)_{k \in \mathbf{N}}$  associated to the eigenvectors

$$\left( \phi_k : \theta \mapsto \frac{1}{\sqrt{\pi}}\sin(k\theta) \right)_{k \in \mathbf{N}}.$$

The restriction  $B$  of  $\psi \mapsto \cos(\theta)\psi$  to  $H_o$  is skew-adjoint and bounded. The couple  $(A, B)$  satisfies Assumption 1, with  $C_{A,B} = 2$  in Assumption 1.6.

The Galerkin approximations of  $A$  and  $B$  at order  $N$  are

$$A^{(N)} = \begin{pmatrix} i & 0 & \cdots & 0 \\ 0 & 4i & \ddots & \vdots \\ \vdots & \ddots & \ddots & 0 \\ 0 & \cdots & 0 & N^2i \end{pmatrix} \text{ and } B^{(N)} = i \begin{pmatrix} 0 & 1/2 & 0 & \cdots & 0 \\ 1/2 & 0 & 1/2 & \ddots & \vdots \\ 0 & \ddots & 0 & \ddots & 0 \\ \vdots & \ddots & 1/2 & 0 & 1/2 \\ 0 & \cdots & 0 & 1/2 & 0 \end{pmatrix}$$

Our aim is to transfer the wave function from the first eigenspace to the second one. The numerical simulation will be done on some finite dimensional space  $\mathbf{C}^N$ . The controls we will use in the following have  $L^1$  norm less than  $13/3$  and, from Proposition 3, the  $|A|^{1/2}$  norm of  $\Upsilon_t^u(\phi_1)$  will remain less than  $\exp(26/3) \approx 5806$  for every time. From Proposition 5, the error done when replacing the original system by its Galerkin approximation of order  $\sqrt{5807/10^{-2}} \approx 761$  is smaller than  $\epsilon = 10^{-2}$ . This estimate is indeed very conservative and it can be improved using the regularity of the operator  $B$ .

**Lemma 8.** *Let  $k$  be an integer. For every  $t$  in  $\mathbf{R}^+$ , for every locally integrable control  $u$  (not necessarily periodic),*

$$|\langle \phi_k, \Upsilon_t^u \phi_1 \rangle| \leq \frac{1}{(k-1)!} \left( \frac{1}{\sqrt{2}} \int_0^t |u(\tau)| d\tau \right)^{k-1}.$$

*Proof.* Fix  $N$  in  $\mathbf{N}$ . For  $v : [0, K] \rightarrow \mathbf{R}$  measurable bounded function, consider the solution of

$$\dot{q} = e^{-v(t)A^{(N)}} B^{(N)} e^{v(t)A^{(N)}} q \quad (13)$$

with initial condition  $q(0) = \phi_1$ .

If  $v$  is piecewise constant, then for every  $1 \leq l \leq k$ , the function  $q_k : t \mapsto \langle \phi_k, q(t) \rangle$  is piecewise  $C^l$  for every  $l \leq k$  and, for almost every  $t \geq 0$ ,

$$\left| \frac{d^l}{dt^l} q_k(t) \right| = \left| \langle \phi_k, (B^{(N)})^l q(t) \rangle \right| = \left| \langle (B^{(N)})^l \phi_k, q(t) \rangle \right|.$$

In particular, the  $l^{\text{th}}$  derivative of  $q_k$  admits everywhere in  $\mathbf{R}$  a left and a right limit. The moduli of these two limits is the same, only the phase may be discontinuous. Notice that thanks to the Cauchy-Schwarz inequality,  $|q_k^{(l)}(t)| \leq \|(B^{(N)})^l \phi_k\|$ . After  $l$  integrations on  $[0, s]$ , one finds, for every  $s \geq 0$ ,

$$|\langle \phi_k, q(s) \rangle| \leq \sum_{j=0}^{l-1} \frac{|\langle \phi_k, (B^{(N)})^j \psi_0 \rangle|}{j!} s^j + \frac{\|(B^{(N)})^l \phi_k\|}{l!} s^l. \quad (14)$$

By continuity of the solutions of (13) with respect to  $v$  (see [AS04, Lemma 8.10]) and by density of piecewise constant functions in  $L^\infty$ , the estimates (14) remains valid for any measurable bounded  $v$ .

We proceed now to the proof of Lemma 8. Fix  $K > 0$  and  $\epsilon > 0$ . By Proposition 5, there exists  $N \geq k$  such that, for every  $u$  with  $L^1$  norm less than  $K$ ,  $\|\Upsilon_t^u \phi_1 - X_t^{(N),u} \phi_1\| < \epsilon$ , for every  $t \geq 0$ .

Fix  $u$  piecewise constant such that  $|u|$  belongs to  $PC$  and  $\|u\|_{L^1} \leq K$ , define  $v$  the inverse function of  $t \mapsto \int_0^t |u(\tau)| d\tau$  and consider  $t \mapsto \psi(t)$ , the solution of  $\dot{\psi}(t) = (A^{(N)} + u(t)B^{(N)})\psi(t)$  with initial condition  $\phi_1$ . As already noticed, the solution  $t \mapsto \tilde{\psi}(t)$  of  $\dot{\psi}(t) = (\mathcal{P}|u|(t)A^{(N)} + \text{sg}(u \circ v)B^{(N)})\psi(t)$  with initial condition  $\phi_1$  satisfies  $\tilde{\psi} = \psi \circ v$ .

Set  $q(t) = e^{-\int_0^t \mathcal{P}|u|(s) ds A^{(N)}} \tilde{\psi}(t)$ . The mapping  $q$  is absolutely continuous and, for almost every  $t$ ,

$$\frac{d}{dt} q(t) = \text{sg}(u \circ v) e^{-\int_0^t \mathcal{P}|u|(s) ds A^{(N)}} B^{(N)} e^{\int_0^t \mathcal{P}|u|(s) ds A^{(N)}} q.$$

From (14), applied with  $v : t \mapsto \int_0^t \mathcal{P}|u|(s) ds$  and  $s = \int_0^t |u(\tau)| d\tau$ ,

$$|\langle \phi_k, q(s) \rangle| \leq \sum_{j=0}^{l-1} \frac{|\langle \phi_k, (B^{(N)})^j \psi_0 \rangle|}{j!} \left( \int_0^t |u(\tau)| d\tau \right)^j + \frac{\|(B^{(N)})^l \phi_k\|}{l!} \left( \int_0^t |u(\tau)| d\tau \right)^l.$$

Choosing  $l = k - 1$  and using the particular tri-diagonal form of  $B$ , one gets

$$|\langle \phi_k, \Upsilon_t^u(\phi_1) \rangle| \leq \frac{\|B\|^{k-1}}{(k-1)!} \left( \int_0^t |u(\tau)| d\tau \right)^{k-1} + \epsilon.$$

The result follows by letting  $\epsilon$  tends to zero.  $\square$

By Lemma 8, if  $\|u\|_{L^1} \leq 13/3$ , then  $\|\pi_{22}B(Id - \pi_{22})\Upsilon_t^u(\phi_1)\| \leq 3 \cdot 10^{-11}$  for every  $t$  in  $\mathbf{R}^+$ . Plugging this inequality in (7), one gets that the error done when replacing the original system by its Galerkin approximation of order 22 is smaller than  $\epsilon = 2 \cdot 10^{-10}$  when  $\|u\|_{L^1} \leq 13/3$ .

The transition (1, 2) is not strongly non-resonant (indeed,  $5^2 - 4^2 = 9 = 3(2^2 - 1^2)$ ). Nevertheless, for every  $\{l_1, l_2\} \neq \{1, 2\}$  such that  $\lambda_{l_1} - \lambda_{l_2} \in 3\mathbf{Z}$  and  $\langle \phi_{l_1}, B\phi_{l_2} \rangle \neq 0$ , one has  $l_1 > 2$  and  $l_2 > 2$ . Hence, for every  $\frac{2\pi}{3}$ -periodic function  $u$ , the limit matrix  $M^\dagger$  (Lemma 7) lets invariant the subspace generated by  $\phi_1$  and  $\phi_2$  and the result of Theorem 1 applies (without having to check that all efficiencies of  $u$  for the transition  $(l_1, l_2)$  with  $l_1 - l_2 \in 3(\mathbf{Z} \setminus \{1\})$  are zero).

We illustrate the notion of efficiency on some examples of control, namely  $u^* : t \mapsto \cos^l(3t)$  for  $l \in \{1, 2, 3, 4, 5\}$ .

The efficiency is zero when  $l$  is even. In numerical simulations, the quantity  $|\langle \phi_2, X_t^{(22), u^*} \phi_1 \rangle|$  is less than  $2 \cdot 10^{-5}$  for every  $t < 500$  (see Figure 1 for  $l = 2$ ).

When  $l$  is odd, the efficiency is not zero. To estimate numerically the efficiency, one considers, for  $n \in \{1, 10, 30\}$ , the first maximum  $p^\dagger$  of  $t \mapsto |\langle \phi_2, X_t^{(N), u^*/n} \phi_1 \rangle|$ , reached at time  $t^\dagger$ , and computes

$$\frac{(1 - p^\dagger)n\pi}{2|\langle \phi_1, B\phi_2 \rangle| \int_0^{t^\dagger} |u^*(\tau)| d\tau}.$$

The Scilab source codes used for the simulation are available on the web page <http://www.iecn.u-nancy.fr/~chambrio/PreprintUK.html>. We sum up the results in the following array.

Control $u^*$ (Theoretical Efficiency)	$n$	Time $t^\dagger$	Precision $1 - p^\dagger$	Numerical Efficiency
$t \mapsto \cos(3t)$ $\pi/4 \approx 79\%$	$n = 1$	6.8	$2 \cdot 10^{-2}$	73%
	$n = 10$	63	$4 \cdot 10^{-4}$	78%
	$n = 30$	189	$3 \cdot 10^{-5}$	78%
$t \mapsto \cos(3t)^3$ $9\pi/32 \approx 88\%$	$n = 1$	8.9	$2 \cdot 10^{-2}$	83%
	$n = 10$	84	$2 \cdot 10^{-4}$	88%
	$n = 30$	252	$2 \cdot 10^{-5}$	88%
$t \mapsto \cos(3t)^5$ $75\pi/256 \approx 92\%$	$n = 1$	10	$7 \cdot 10^{-3}$	93%
	$n = 10$	101	$2 \cdot 10^{-4}$	92%
	$n = 30$	302	$2 \cdot 10^{-5}$	92%

### 3.3 Not strongly non-resonant case

We concentrate on the space  $H_e$ . The restriction  $A$  of  $i\Delta$  to  $H_e$  is skew adjoint, with simple eigenvalues  $(ik^2)_{k \in \mathbf{N} \cup \{0\}}$  associated to the eigenvectors  $(\phi_k)_{k \in \mathbf{N} \cup \{0\}}$ ,



with  $\phi_k : \theta \mapsto \frac{1}{\sqrt{\pi}} \cos(k\theta)$  for  $k$  in  $\mathbf{N}$  and  $\phi_0 : \theta \mapsto \frac{1}{\sqrt{2\pi}}$ . The restriction  $B$  of  $\psi \mapsto \cos(\theta)\psi$  to  $H_e$  is skew-symmetric. The couple  $(A + i, B)$  satisfies Assumption 1. The translation from  $A$  to  $A + i$  just induces a phase shift and will be neglected in the following.

The Galerkin approximation of  $A$  and  $B$  at order  $N$  are

$$A^{(N)} = \begin{pmatrix} 0 & 0 & \cdots & 0 \\ 0 & i & \ddots & \vdots \\ \vdots & \ddots & \ddots & 0 \\ 0 & \cdots & 0 & (N-1)^2 i \end{pmatrix}, B^{(N)} = \begin{pmatrix} 0 & \frac{i}{\sqrt{2}} & 0 & \cdots & 0 \\ \frac{i}{\sqrt{2}} & 0 & \frac{i}{2} & \ddots & \vdots \\ 0 & \ddots & 0 & \ddots & 0 \\ \vdots & \ddots & \frac{i}{2} & 0 & \frac{i}{2} \\ 0 & \cdots & 0 & \frac{i}{2} & 0 \end{pmatrix}.$$

Our aim is to transfer the population from the first eigenspace, associated with eigenvalue 0, to the second one, associated with eigenvalue  $i$ . The transition  $(1, 2)$  is not strongly non-resonant ( $2^2 - 1^2 = 3 = 3(1^2 - 0^2)$ ), and in contrary to what happens on the space of odd eigenfunctions, the limit matrix  $M^\dagger$  does not necessarily stabilize the space spanned by  $\phi_1$  and  $\phi_2$  for every  $2\pi$ -periodic  $u^*$ . Note however that  $B$  only connects level 2 to levels 1 and 3. In other words, it is enough to find a  $2\pi$ -periodic function  $u^*$  such that  $E^{(2,3)}(u^*)$  is zero and  $E^{(1,2)}(u^*)$  is not zero (and as large as possible) to induce the desired transfer. This is achieved, for instance, with the sequence of piecewise constant controls build in [BCCS11]), for which the efficiency with respect to transition  $(1, 2)$  tends to  $\cos(\pi/6)$  and the efficiency with respect to transition  $(2, 3)$  is zero. Another example is presented on Figure 2.

## 4 Conclusion

This main result of this paper will not surprise anyone already familiar with quantum control. Its contribution lies first in the theoretical rigorous proof of the convergence and second, in the interpretation of the notion of efficiency, seen as a measure of the  $L^1$ -norm of the control needed to achieve a given transition.

The author is fully aware that most of the hypotheses (especially Assumptions 1.3 and 1.4) are far from being minimal. They were done in the hope to keep the paper readable and usable by a broad audience. A forthcoming paper [BCC11] will use much weaker hypotheses, to the price of more technical developments.

Despite some recent advances in the field of bilinear control of skew-adjoint operators, many questions remain open. Among other topics, future works may concentrate on the design of time-efficient controls or on the control of systems where operators have continuous spectrum.

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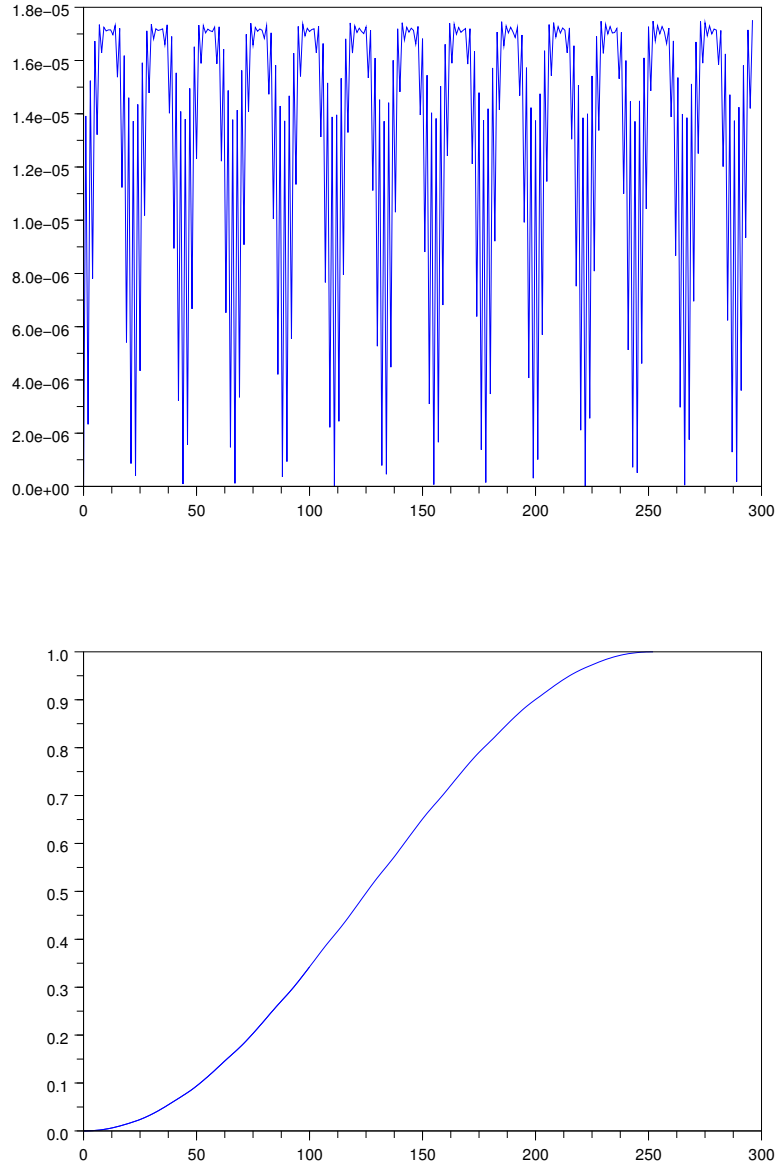


Figure 1: Evolution of the modulus of the second coordinate when applying the controls :  $t \mapsto \cos^2(3t)/30$  (top) and  $t \mapsto \cos^3(3t)/30$  (bottom) on the planar molecule (odd subspace) with initial condition  $\phi_1$ . The simulations have been done on a Galerkin approximation of size  $N = 22$ .

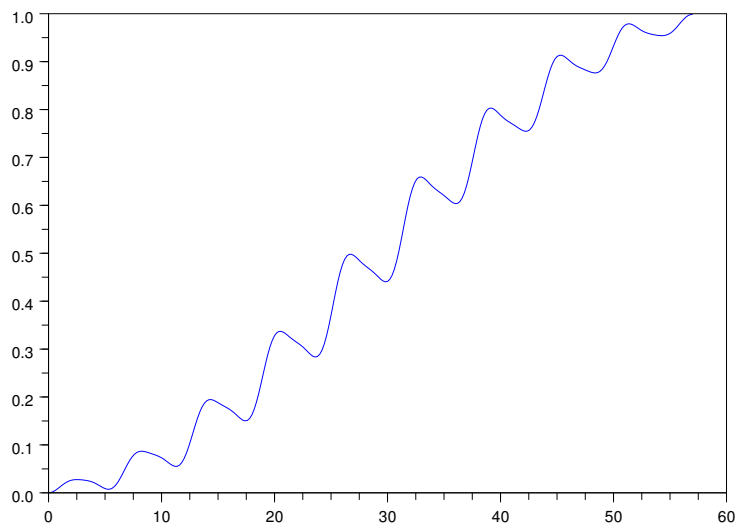


Figure 2: Evolution of the modulus of the second coordinate when applying the control  $\frac{1}{20} \left( 2 \cos^2 \left( \frac{t}{2} \right) + \cos^2 \left( \frac{t-\pi/3}{2} \right) + \cos^2 \left( \frac{t+\pi/3}{2} \right) \right)$  on the planar molecule (even subspace) with initial condition  $\phi_1$ . The simulation has been done on a Galerkin approximation of size  $N = 22$ . Precision  $1 - p^\dagger$  is equal to  $2.10^{-3}$ . Numerical efficiencies are 38% (theoretical:  $3/8$ ) for the transition (1,2) and less than  $5.10^{-4}$  for the transition (2,3) (theoretical: 0).