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Periodic excitations of bilinear quantum systems

Thomas Chambrion

Abstract

A well-known method of transferring the population of a quantum system from an eigenspace of the free Hamiltonian to another is to use a periodic control law with an angular frequency equal to the difference of the eigenvalues. For finite dimensional quantum systems, the classical theory of averaging provides a rigorous explanation of this experimentally validated result. This paper extends this finite dimensional result, known as the Rotating Wave Approximation, to infinite dimensional systems and provides explicit convergence estimates.

Key words: Infinite-dimensional systems; Schrödinger equation; averaging control.

1 Introduction

1.1 Effective control of quantum systems

The state of a quantum system evolving on a finite dimensional Riemannian manifold \( \Omega \) is described by its wave function, that is, a point in the unit sphere of \( L^2(\Omega, \mathbb{C}) \). In the absence of interaction with the environment and with a suitable choice of units, 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 \( V : \Omega \rightarrow \mathbb{R} \) is a real function (usually called potential) accounting for the physical properties of the system. When subjected to an excitation by an external electric field (e.g. a laser), the Schrödinger equation reads

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

where \( W : \Omega \rightarrow \mathbb{R} \) 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 effort has been expended by different communities on studying the controllability of (1). We refer to Nersessyan [Ner10], Beauchard & Mirrahimi [BM09], Mirrahimi [Mir09], Boscain & Laurent [BL10] and Boscain, Caponigro, Chambrion & Sigalotti [BCCS12] for a description of the known theoretical results concerning the existence of controls steering a given source to a given target. As proved by Nersessyan [Ner10] and Mason & Sigalotti [MS10], approximate controllability is a generic property for systems of the type (1).

A number of effective control algorithms have been obtained by various authors, see among many others Warren, Rabitz & Dahleh [WRD93], Blümel, Fishman & Smilansky [BFS86], Ohtsuki, Kono & Fujiyama [OKF98] or Belhadj, Salomon & Turinici [BST08]. Most of the controls used in practice exhibit a remarkable pattern of periodic shape, with a frequency corresponding to the transitions of the quantum system (see for instance Salomon, Dion & Turinici [SDT05]).

1.2 Averaging techniques in quantum mechanics

The fact that a small amplitude periodic excitation with suitable frequency is sufficient, in general, to induce a
The situation is now well understood for quantum systems with finitely many energy levels. These systems appear, for instance, as truncations of infinite dimensional systems. In this case, system (1) reads \( \dot{x} = (A + u(t)B)x \) where \( A \) and \( B \) are \( N \times N \) skew-hermitian matrices. Let us briefly recall the method of the proof.

The mathematical concept of averaging of dynamical systems was introduced more than a century ago and has now developed into a well-established theory, see for instance the books of Guckenheimer & Holmes [GHS3], Bullo & Lewis [BL05] or Sanders, Verhulst & Murdock [SVM07]. It was observed that, for regular \( F \) and small \( \varepsilon \), the trajectories of the system \( \dot{x} = \varepsilon F(x, t, \varepsilon) \) remain \( \varepsilon \) close, for time of order \( 1/\varepsilon \), to the trajectories of the average system \( \dot{x} = \bar{F}(x) \) where \( \bar{F}(x) = \lim_{t \to \infty} 1/t \int_0^t F(x, t, 0) \).

In quantum physics, this procedure has been known as the *rotating wave approximation* for several decades, see Fox & Eisdon [FE87] or Vandersypen & Chuang [VC05]. Assuming without loss of generality that \( A \) is diagonal with eigenvalues \( \lambda_1, \ldots, \lambda_N \), define, for every \( n \) in \( \mathbb{N} \), 
\[
y_n : t \mapsto e^{-itA}x_n \text{ where } x_n \text{ is the solution of } \dot{x} = (A + u\varepsilon B/n)x.
\]
The mapping \( y_n \) is absolutely continuous and satisfies \( \dot{y}_n = (u(t)/n)e^{-itA}Be^{itA}y_n \). The conclusion follows from standard averaging theory by computing the average matrix \( \lim_{t \to \infty} 1/(nt) \int_0^t u\varepsilon(t)e^{-itA}Be^{itA}d\tau \) whose all entries are zero but maybe entry \((j, k)\) if \( u\varepsilon \) is \( 2\pi/|\lambda_j - \lambda_k|\)-periodic.

By contrast with this finite dimensional result, the situation is much more intricate when the ambient space has infinite dimension and the problem involves unbounded operators, which is precisely the case for the Schrödinger equation (1) when \( \dim \Omega \geq 1 \) and \( A = i\Delta/2 \). The above averaging method fails because of serious regularity issues. For instance, the mapping \( t \mapsto e^{itA} \) is no longer Lipschitz continuous. If moreover \( B \) is unbounded, that is, if function \( W \) in (1) is not in \( L^\infty(\Omega, \mathbb{R}) \), then, for any given \( t \), the mapping \( x \mapsto e^{-itA}Be^{itA}x \) is not even continuous: this is what prevents the direct application of the averaging results in Banach spaces presented by Artstein [Art10]. For these reasons, most of the available averaging results for infinite dimensional quantum systems deal with constant controls only, as in the papers of Kummer [Kum71] or Scherer [Sch94].

### 1.3 Framework and notation

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:

\[
\frac{d\psi}{dt} = (A + u(t)B)\psi(t) \quad u(t) \in U
\]  

where \((A, B, U)\) satisfies Assumption 1.

**Assumption 1** \((A, B, U)\) is a triple where \((A, B)\) is a pair of linear operators and \( U \) is a subset of \( \mathbb{R} \) such that

1. For every \( n \) in \( \mathbb{N} \), \( U \subset nU \);
2. \( A \) is skew-adjoint with domain \( D(A) \);
3. There exists a Hilbert basis \( \{\phi_k\}_{k \in \mathbb{N}} \) of \( H \) and a family \( \{i\lambda_k\}_{k \in \mathbb{N}} \) in \( i\mathbb{R} \) such that \( A\phi_k = i\lambda_k\phi_k \) for every \( k \) in \( \mathbb{N}^+ \);
4. \( B \) is skew-symmetric, possibly unbounded with domain \( D(B) \);
5. For every \( k \) in \( \mathbb{N} \), \( \phi_k \) belongs to \( D(B) \);
6. For every \( u \) in \( U \), \( A + uB \) is essentially skew-adjoint.

Assumption 1.6 ensures that, for every constant \( u \in U \), \( A + uB \) generates a group of unitary propagators. Hence, for every initial time \( t_0 \) in \( \mathbb{R} \) and initial condition \( \psi_0 \) in \( H \), for every piecewise constant control \( u \) taking value in \( U \), we can define the solution \( t \mapsto T^u(t, t_0)\psi_0 \) of (2) taking value \( \psi_0 \) at time \( t_0 \). We simply note \( T^u(t, t_0)\psi_0 = T^u|_{t=t_0} \psi_0 \) when \( t_0 = 0 \).

**Remark 1** To the best of the author’s knowledge, in the general frame of Assumption 1, no definition of solutions of (2) is available for controls that are not piecewise constant. With some extra regularity assumptions (for instance, when \( B \) is bounded, see Section 3.4), one can define the solution of (2) for more general controls.

#### 1.4 Main result

**Definition 1** Let \((A, B, U)\) satisfy Assumption 1. A point \((j, k)\) of \( \mathbb{N}^2 \) is a non-degenerate transition of \((A, B)\) if (i) \( j \neq k \), (ii) \( \phi_j, B\phi_k \neq 0 \) and (iii) for every \( l, m \) in \( \mathbb{N} \), \( |\lambda_j - \lambda_k| = |\lambda_i - \lambda_m| \) implies \( (j, k) = (l, m) \) or \((j, k) \cap (l, m) = \emptyset \) or \( \phi_j, B\phi_m = 0 \).

**Theorem 1** Let \((A, B, U)\) satisfy Assumption 1. \((j, k)\) a non-degenerate transition of \((A, B)\) and \( u^* : \mathbb{R}^+ \to U \) be a piecewise constant function, periodic with period \( T = \frac{2\pi}{|\lambda_j - \lambda_k|} \). Assume that \( \int_0^T u^*(\tau)e^{i(\lambda_j - \lambda_k)\tau}d\tau = 0 \) for every \( l, m \) such that \( |\lambda_i - \lambda_m| \in (\mathbb{N} \setminus \{1\})|\lambda_j - \lambda_k| \) and \( \langle \phi_j, B\phi_m \rangle \neq 0 \) and \((l, m) \cap (j, k) \neq \emptyset \).

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 \( \left\| \left( \phi_j, \sum_{n \in \mathbb{N}} \frac{1}{n} T^u(\cdot, nT^*)\phi_j \right) \right\| \) tends to 1 as \( n \) tends to infinity.

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1.5 Content of this paper

This paper comprises three parts. The first one (Section 2) is concerned with a finite dimensional version of Theorem 1. As already mentioned, the convergence result is classical. A time reparametrization (Section 2.2) is introduced, which leads to explicit estimates better than those currently available in the literature.

The second part (Section 3) contains a general proof of Theorem 1, valid in the general framework of Assumption 1. It provides the first available time estimates for the approximate controllability of general bilinear quantum systems when the free Hamiltonian admits a dense family of eigenvectors.

Finally, Section 4 presents an extension of Theorem 1 to the case of the Morse quantum oscillator. The main feature of this system is that the spectrum of the free Hamiltonian has a continuous part.

2 Finite dimensional estimates

2.1 Finite dimensional framework

Let \( N \) be an integer and \( A^{(N)}, B^{(N)} \) be two skew-adjoint matrices of order \( N \). Without lost of generality, we may assume that \( A^{(N)} \) is diagonal with eigenvalues \( \lambda_1, \lambda_2, \ldots, \lambda_N \). We denote with \( (\phi_j)_{1 \leq j \leq N} \) the canonical basis of \( \mathbb{C}^N \), with \( \langle \cdot, \cdot \rangle \) the canonical Hermitian product of \( \mathbb{C}^N \), with \( (b_{jk} = \langle \phi_j, B^{(N)} \phi_k \rangle)_{1 \leq j, k \leq N} \) the entries of \( B^{(N)} \) and with \( \pi_{i}^{(N)} : \mathbb{C}^N \to \mathbb{C}^N \) the projection \( \pi_{i}^{(N)} : x \mapsto \sum_{m \leq i} (\phi_m, x) \phi_m \) on the first \( l \) components in \( \mathbb{C}^N \).

For every piecewise constant function \( u : \mathbb{R} \to \mathbb{R} \), we denote with \( X_{(N)}^u(t, s) \) the propagator between times \( s \) and \( t \) of the system

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

In other words, \( t \mapsto X_{(N)}^u(t, s)x_0 \) is the unique solution of (3) with initial condition \( x(s) = x_0 \). It is classical that, for every \( s, t \in \mathbb{R} \), for every \( x_0 \in \mathbb{C}^N \), the mapping \( u \mapsto X_{(N)}^u(t, s)x_0 \) admits a Lipschitz-continuous continuation to \( L_{loc}^{1}(\mathbb{R}, \mathbb{R}) \).

The aim of this section is to prove the following result.

**Proposition 2** Let \( u : \mathbb{R} \to \mathbb{R} \) be a locally integrable function. Assume that \( \lambda_1 \neq \lambda_2 \) and that, for every \( l, m \leq N, |\lambda_l - \lambda_m| = |\lambda_2 - \lambda_1| \) implies \( \{l, m\} = \{1, 2\} \) or \( b_{lm} = 0 \) or \( \{l, m\} \cap \{1, 2\} = \emptyset \).

Assume that \( u^* \) is periodic with period \( T = \frac{2\pi}{|\lambda_2 - \lambda_1|} \) and that \( \int_{0}^{T} u^*(\tau)e^{i(\lambda_2 - \lambda_1)\tau}d\tau = 0 \) for every \( \{l, m\} \) such that \( \{l, m\} \cap \{1, 2\} \neq \emptyset \) and \( \lambda_l - \lambda_m \in (\mathbb{Z} \setminus \{-1, 1\}) (\lambda_1 - \lambda_2) \) and \( b_{lm} \neq 0 \).

If \( \int_{0}^{T} u^*(\tau)e^{i(\lambda_2 - \lambda_1)\tau}d\tau \neq 0 \), then, for every \( n \in \mathbb{N} \), there exists \( T^*_n \) in \( (nT^* - T, nT^* + T) \) such that

\[
1 - |\langle \phi_2, X_{(N)}^u(T^*_n, 0)\phi_1 \rangle| \leq \left( 1 + C \left\| B^{(N)} \right\| \right) I \left( 2 + 2K \left\| B^{(N)} \right\| n \right), \tag{4}
\]

with

\[
T^* = \frac{\pi T}{2|b_{1,2}|}, \quad I = \frac{\int_{0}^{\pi T} u^*(\tau)e^{i(\lambda_2 - \lambda_1)\tau}d\tau}{\int_{0}^{T} u^*(\tau)d\tau}, \quad K = \frac{\int_{0}^{T} u^*(\tau)e^{i(\lambda_2 - \lambda_1)\tau}d\tau}{2\pi T} \sin \left( \frac{\pi}{|\lambda_2 - \lambda_1|} \right),
\]

where \( A \) is the set of all pairs \( (j, k) \) in \( \{1, \ldots, N\} \) such that \( b_{jk} \neq 0 \) and \( \{j, k\} \cap \{1, 2\} = \emptyset \) and \( |\lambda_j - \lambda_k| \notin \mathbb{Z}|\lambda_2 - \lambda_1| \).

**Corollary 3** With the notations of Proposition 2, \( |\langle \phi_2, X_{(N)}^u(t, 0)\phi_1 \rangle| \) tends to 1 as \( n \) tends to infinity.

The convergence result (Corollary 3) is classical. The novel element here is that the bilinear structure allows us to give explicit estimates (4) for the convergence rate.

Most of the rest of this section is devoted to the proof of Proposition 2. A technical time reparametrization (Section 2.2) is introduced. The main ingredient of the proof is an averaging procedure (Section 2.3), that is performed first on piecewise constant controls and then extended to irregular controls. Finally, we introduce the notion of efficiency in Section 2.4.

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 \( (t_j)_{1 \leq j \leq \delta} \) with value in \( (0, +\infty) \) such that

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

where \( \chi \) is the characteristic function and the sequence \( (t_j)_{1 \leq j \leq p+1} \) is defined by induction: \( t_1 = 0 \), \( t_{j+1} = t_j + t_j \). An element \( u \) of \( PC \) will be denoted \( (u_j, t_j)_{1 \leq j \leq p} \).
The involutary mapping $P : (u_j, t_j)_{1 \leq j \leq p} \in PC \mapsto (1/u_j, u_j t_j)_{1 \leq j \leq p} \in PC$ can be used as a time reparametrization to replace the system (3) by the control system in $C^N$

$$\frac{dx}{dt} = (u(t)A(N) + B(N))x(t),$$

whose propagator between time $s$ and $t$ will be denoted with $X^N_{(t,s)}(t,s)$.

**Proposition 4** For every $x_0$ in $C^N$ and every $u$ in $P(C)$, $X^N_{(t,s)}(\int_0^T u(\tau) d\tau,0)x_0 = X^N_{(t,s)}(t,0)x_0$.

**Proof:** This follows from the equality $e^{t(A(N) + uB(N))} = e^{tu_0(\frac{1}{2}A(N) + B(N))}$, valid on every interval where $u$ is constant. □

2.3 Averaging procedure

Let $u^*$ be a non vanishing piecewise constant function, $T$-periodic, as in the hypotheses of Proposition 2. For every $n$ in $N$, define the non-vanishing $T$-periodic function $u_n = u^*/n$. For every $n$ in $N$, $\int_0^T |u_n(\tau)|d\tau = \int_0^T |u^*(\tau)|d\tau$.

We perform now the time reparametrization set out in Section 2.2. The function $t \mapsto \int_0^T |u_n(\tau)|d\tau$ is non-decreasing. We denote with $v_n$ its reciprocal function and define also $I = \int_0^T |u^*(\tau)|d\tau$. For every $t$ in $R^+$, $v_n(t + 1/n) = v_n(t) + T$ and $P|u_n|$ is the $1/n$ periodic derivative (defined almost everywhere) of $v_n$.

Fix $x_0$ in $C^N$ and note with $t \mapsto x_n(t)$ the solution of

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

with initial condition $x(0) = x_0$.

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

$$[0,T] = J^+_1 \cup J^-_1 \cup \ldots \cup J^+_p \cup J^-_p,$$

such that $u^*$ takes positive (respectively, negative) values on $J^+ = \cup_{i=1}^p J^+_i$ (respectively, $J^- = \cup_{i=1}^p J^-_i$).

Defining the sets $G^+_n = v^{-1}_n(J^+)$ and $G^-_n = v^{-1}_n(J^-)$, we obtain the dynamics of $y_n = x_n \circ v_n$, valid for almost every $t$:

$$\frac{dy_n}{dt} = \begin{cases} (P(|u_n|)(t)A(N) + B(N))y_n(t) & \text{if } t \in G^+_n \\ (P(|u_n|)(t)A(N) - B(N))y_n(t) & \text{if } t \in G^-_n \end{cases}$$

Finally, for every $t$, we define $z_n(t) = e^{-v_n(t)A(N)}y_n(t)$ (note that, for every $t$, for every $l$ in $N$, $|(\phi_l, z_n(t))| = |\langle \phi_l, y_n(t) \rangle|$), and the time varying $N \times N$ matrix $M_n$:

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

From (7), we deduce the dynamics of $z_n$, valid for almost every $t$ in $G^+_n \cup G^-_n$:

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

We note $Z^N$ the propagator associated with (8). Note that, for every $k$, the mapping $t \mapsto \langle \phi_k, z_n(t) \rangle$ is Lipschitz continuous with Lipschitz constant $\|B\phi_k\|$.

Let $M^\uparrow$ be the constant $N \times N$ matrix whose entries, for $1 \leq j,k \leq N$, are defined by

$$m^\uparrow_{j,k} = \frac{b_{j,k}}{T} \int_0^T \exp(i(\lambda_j - \lambda_k)v^*(\tau))d\tau$$

if $T(\lambda_j - \lambda_k) \in 2\pi Z$ and $m^\uparrow_{j,k} = 0$ if $T(\lambda_j - \lambda_k) \notin 2\pi Z$. Define the $N \times N$ matrix $H_n(t)$, with entries $(h^N_{j,k}(t))_{1 \leq j,k \leq N}$, by $H_n(t) = M_n(t) - M^\uparrow$.

**Lemma 5** For every $t$ in $R^+$, for every $n$ in $N$, for every $j,k \leq N$ such that $|\lambda_j - \lambda_k| \notin Z|\lambda_2 - \lambda_1|$, for every $j,k \leq N$ such that $\{j,k\} \neq \{1,2\}$ and $|\lambda_j - \lambda_k|/|\lambda_2 - \lambda_1| \in Z$,

$$\left| \int_0^t h^N_{j,k}(\tau)d\tau \right| \leq \frac{|b_{j,k}|}{n} \left( \frac{\int_0^T u^*(\tau)e^{i(\lambda_j - \lambda_k)}d\tau}{\sin\pi(\frac{\lambda_j - \lambda_k}{\lambda_2 - \lambda_1})} + 1 \right).$$

For every $t$ in $R^+$, for every $n$ in $N$, for every $j,k \leq N$ such that $\{j,k\} \neq \{1,2\}$ and $|\lambda_j - \lambda_k|/|\lambda_2 - \lambda_1| \in Z$,

$$\left| \int_0^t h^N_{j,k}(\tau)d\tau \right| \leq \frac{2|b_{j,k}|T}{n}.$$  

**Proof:** For every $t$, define the integer $s = \lfloor \frac{t}{T} \rfloor$. For every $j,k,n,t$, we have

$$\left| \int_{st/n}^t \operatorname{sg}(u_n \circ v_n)e^{i(\lambda_j - \lambda_k)}v_n(\tau)d\tau \right| \leq \frac{t}{n}.$$
For every \( j, k \) such that \( T(\lambda_j - \lambda_k) \notin 2\pi \mathbb{Z} \),

\[
\left| \int_0^{1/n} \sgn(u \circ v) e^{i(\lambda_j - \lambda_k)v_n(\tau)} d\tau \right| \\
\leq \sum_{m=1}^s \int_0^{1/n} \sgn(u \circ v)e^{i(\lambda_j - \lambda_k)(v_n(\tau) + mT)} d\tau \\
\leq \int_0^{1/n} \sgn(u \circ v)e^{i(\lambda_j - \lambda_k)(\sum_{m=1}^s e^{i(\lambda_j - \lambda_k)mT} d\tau) \\
\leq \frac{1}{n} \left( 2 \left| \int_0^T u^*(\tau) e^{i(\lambda_j - \lambda_k)\tau} d\tau \right| \right) \cdot
\]

If \( T(\lambda_j - \lambda_k) \in 2\pi \mathbb{Z} \), then

\[
\sum_{m=1}^s \int_0^{1/n} \sgn(u \circ v_n)e^{i(\lambda_j - \lambda_k)v_n(\tau) + mT)} d\tau \\
= s \int_0^{1/n} \sgn(u \circ v_n)e^{i(\lambda_j - \lambda_k)v_n(\tau)} d\tau \\
= \frac{s}{n} \int_0^T \sgn(u \circ v_n)e^{i(\lambda_j - \lambda_k)v_n(\tau)} d\tau.
\]

Hence

\[
\left| \int_0^{1/n} \sgn(u \circ v_n)e^{i(\lambda_j - \lambda_k)v_n(\tau)} d\tau \right| \\
\leq \frac{1}{n} \cdot \Box
\]

Recall that, for every \( n \in \mathbb{N} \) and every \( t \geq 0 \),

\[
z_n = M^t z_n + H_n(t) z_n,
\]
or, using the variation of the constant formula,

\[
z_n(t) = e^{M^t} z_n(0) + \int_0^t e^{(t-s)M^t} H_n(s) z_n(s) ds.
\]

Integrating by part, we get for every \( t \geq 0 \),

\[
z_n(t) = e^{M^t} z_n(0) + \left[ e^{(t-s)M^t} \left( \int_0^s H_n(\tau) d\tau \right) z_n(s) \right]_0^t \\
+ \int_0^t M^t e^{(t-s)M^t} \left( \int_0^s H_n(\tau) d\tau \right) z_n(s) ds \\
- \int_0^t e^{(t-s)M^t} \left( \int_0^s H_n(\tau) d\tau \right) \dot{z_n}(s) ds. \quad (11)
\]

The equality \( X_n(t, 0) = e^{tA^{(N)}} o Z_n(t, 0) \) gives estimates for the convergence of the propagators \( X_n \):}

For every \( n \in \mathbb{N} \), for every \( t \leq v_n^{[-1]}(K) \), for every \( x_0 \in \mathbb{C}^N \), \( T, K \) and \( C \) are defined as in Proposition 2

\[
\| X_n^{(N)}(t, 0)x_0 - e^{tA^{(N)}} e^{v_n^{[-1]}(t)M^t} x_0 \| \\
\leq \frac{1 + K(\|M^t\| + \sup_{x \in \mathbb{C}^N} \|B^{(N)} X_n^{(N)}(0, 0) x_0 \|)}{n}. \quad (12)
\]
or

\[
\| X_n^{(N)}(t, 0) - e^{tA^{(N)}} e^{v_n^{[-1]}(t)M^t} \| \\
\leq \frac{2K\|B^{(N)}\|}{n}. \quad (13)
\]

Projecting (11) on the two first components of \( \mathbb{C}^N \), we get

\[
\| \| z_n(t) - \| \| z_n(0) \| \\
\leq \frac{2\| z_n(t) \|}{\| z_n(0) \|}. \quad (14)
\]

Know that \( v^* \) is non-decreasing and \( v^*(1) = 1T \) for every \( n \in \mathbb{N} \), we deduce that, for every \( n \in \mathbb{N} \), \( nT^* - T \leq T_n^* \leq nT^* + T \), with \( T^* \) defined as in the statement of Proposition 2.

Note finally that, for every \( s, t \) in \( \mathbb{R} \) such that \( s \leq t \),

\[
\| \langle \phi_2, x_n(t) \rangle - \| \langle \phi_2, x_n(s) \rangle \| \leq \frac{\| B\phi_2 \|}{n} \int_s^t |u(\tau)d\tau.
\]

This completes the proofs of Proposition 2 and Corollary 3 in the case where \( u^* \) is a non vanishing piecewise constant function.

If \( u^* \) is a locally integrable \( T \)-periodic function, let \( \langle u^{*,l} \rangle_{l \in \mathbb{N}} \) be a sequence of non-vanishing piecewise constant \( T \)-periodic functions converging to \( u^* \) in the distribution sense with \( \| u^{*,l} \|_{L^1([0,T])} \leq \| u \|_{([0,T])} \)

for every \( l \). We define, for every \( n \), \( u_{n,l} = \frac{u^{*,l}}{n} \),

\[
v_{n,l} : t \mapsto \int_0^t \frac{\| u_{n,l} \|_{([0,T])}}{n} d\tau \quad \text{and} \quad M_{n,l}(t) = \sgn(u_{n,l} \circ v_{n,l}(t)) e^{-v_{n,l}(t)A} B^{(N)} e^{v_{n,l}(t)A}.
\]

For every \( t \), the matrix
$\int_0^t M_{ij}(\tau)d\tau$ tends to $tM_{ij}$ uniformly with respect to $t$ in a compact set, as $l$ tends to infinity. Hence the solutions of $\dot{x} = M_{ij}(t)x$ tend to the solutions of $\dot{x} = M^i x$, uniformly with respect to the time in a compact interval, as $l$ tends to infinity. That concludes the proof of Proposition 2.

2.4 Efficiency of the transfer

Continuing with the notation of the last paragraph, for every non identically zero, $\frac{2\pi}{|\lambda_j - \lambda_k|}$ periodic function $u^*$, we define the efficiency of $u^*$ with respect to the transition $(j,k)$ as the real quantity:

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

For every $u$, $0 \leq E^{(j,k)}(u) \leq 1$. For every $\{j,k\}$, sup$_u E^{(j,k)}(u) = 1$. The supremum is reached with a periodic sum of Dirac masses.

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 $\frac{\pi}{2}\delta_{bj,k}$ is equal to $\pi/(2|b_{j,k}|E^{(j,k)}(u^*))$.

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 choose a control $u$ such that $E^{(j,k)}(u)$ be as large as possible and $E^{(j,l)}(u)$ be zero (or arbitrarily close to zero) for every $l_1, l_2$ such that $l_1 - l_2 \in (\lambda_j - \lambda_k)\mathbb{Z}$. The algorithm we have described in [BCCS12] allows us 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 $|l_1 - \lambda_{l_1}| \neq |\lambda_j - \lambda_k|$. Some other examples, including also examples of ineffective controls with zero efficiency, are studied in [BCC11b].

3 Infinite dimensional estimates

We come back to the general case of Assumption 1. To ensure that the system (2) is well-posed, we consider only piecewise constant functions $u^*$. The method of the proof is directly inspired by [BCCS12]: the original infinite dimensional system (2) is approached by a suitable Galerkin approximation, which allows us to apply the finite dimensional results of Section 2.

3.1 Galerkin approximation

Let $(A, B, U)$ satisfy Assumption 1. We define $b_{jk} = \langle \phi_j, B\phi_k \rangle$ for $j, k$ in $\mathbb{N}$. For every $N$ in $\mathbb{N}$, we define $\pi_N : \psi \in H \mapsto \sum_{j \leq N}\langle \phi_j, \psi \rangle \phi_j \in H$ and the compressions $A^{(N)} = \pi_N \circ A \circ \pi_N$ and $B^{(N)} = \pi_N \circ B \circ \pi_N$. Note that $A^{(N)}$ and $B^{(N)}$ are finite rank operators defined in an infinite dimensional space. With an obvious abuse of notation, we extend the finite dimensional propagator $X^{(N)}_\lambda$ defined in Section 2.1 to the infinite dimensional space $H$.

Let $u^* : \mathbb{R} \to U, j, k$ be given as in the hypotheses of Theorem 1. Up to a reordering, we may assume $j = 1$ and $k = 2$ without loss of generality.

Define

$$K = \frac{\pi}{2|h_{12}|} \frac{1}{E^{(1,2)}(u^*)}.$$  

Fix $\varepsilon > 0$. Since $\phi_1$ and $\phi_2$ belong to the domain of $B$, the sequences $(h_{1,i})_{i \in \mathbb{N}}$ and $(h_{2,i})_{i \in \mathbb{N}}$ are in $\ell^2$. Hence, there exists $N$ in $\mathbb{N}$ such that $\|\pi_2 B(1 - \pi_N)\| = \|1 - \pi_N\| = \varepsilon/K$.  

Consider system (2) with control $u_n := u^*/n$ in projection on span$(\phi_1, \ldots, \phi_N)$:

$$\pi_N \frac{d}{dt} Y^{u_n}_t \phi_1 = (A^{(N)} + u_n(t)B^{(N)}) Y^{u_n}_t \phi_1$$

$$+ u_n(t)\pi_N B(1 - \pi_N) Y^{u_n}_t \phi_1.$$  

(15)

From the variation of the constant, we get

$$\pi_N Y^{u_n}_t \phi_1 = X^{u_n}_{(N)}(t, 0) \phi_1$$

$$+ \int_0^t u_n(s)X^{u_n}_{(N)}(t, s)\pi_N B(1 - \pi_N) Y^{u_n}_s \phi_1 ds.$$  

(16)

Project (16) on span$(\phi_1, \phi_2)$, and recall that $\pi_N \pi_2 = \pi_2 \pi_N = \pi_2$ for $N \geq 2$:

$$\pi_2 Y^{u_n}_t \phi_1 = \pi_2 X^{u_n}_{(N)}(t, 0) \phi_1$$

$$+ \int_0^t u_n(s)\pi_2 X^{u_n}_{(N)}(t, s)\pi_N B(1 - \pi_N) Y^{u_n}_s \phi_1 ds.$$  

(17)

Define, for every $t, s$ in $\mathbb{R}$, the bounded linear mapping $[\pi_2, X^{u_n}_{(N)}(t, s)] := \pi_2 \circ X^{u_n}_{(N)}(t, s) - X^{u_n}_{(N)}(t, s) \circ \pi_2$. Equation (17) reads

$$\pi_2 Y^{u_n}_t \phi_1 - \pi_2 X^{u_n}_{(N)}(t, 0) \phi_1 =$$

$$- \int_0^t u_n(s)X^{u_n}_{(N)}(t, s)\pi_2 B(1 - \pi_N) Y^{u_n}_s \phi_1 ds$$

$$+ \int_0^t u_n(s)[\pi_2, X^{u_n}_{(N)}(t, s)]\pi_N B(1 - \pi_N) Y^{u_n}_s \phi_1 ds.$$  

(18)

3.2 Estimates of commutators

Extend the definition of $M^\dagger$ of Section 2.3 by $M^\dagger \phi_j = 0$ for $j > N$ and define the linear operator $E^{(N)}_\lambda(t) :=$
$X_{(N)}^u(t,0) = e^{s(-1)(t)M^1}$. Since the commutator $[\pi_2, M^\dagger] = \pi_2 M^1 - M^1 \pi_2$ vanishes, we have, for every $t$ in $\mathbb{R}$,

$$
\|[\pi_2, X_{(N)}^u(t,0)]\| = \|\pi_2 e^{s(-1)(t)M^1} + E_{(N)}^n(t)\|
= \|\pi_2 E_{(N)}^n(t)\| \leq 2\|E_{(N)}^n(t)\|.
$$

Note also that, for every $t$ in $\mathbb{R}$,

$$
\|[\pi_2, X_{(N)}^u(t,0)]\| = \|X_{(N)}^u(t,0)X_{(N)}^u(t,0)\| \leq 2\|E_{(N)}^n(t)\|.
$$

For every $s, t$ in $\mathbb{R}$,

$$
[\pi_2, X_{(N)}^u(t,s)] = \pi_2 X_{(N)}^u(t,0)X_{(N)}^u(t,0)\pi_2 - X_{(N)}^u(t,0)X_{(N)}^u(t,0)\pi_2
= X_{(N)}^u(t,0)[\pi_2, X_{(N)}^u(t,0)] + [\pi_2, X_{(N)}^u(t,0)]X_{(N)}^u(t,0).
$$

Finally, we get, for every $(s, t)$ in $\mathbb{R}$, for every $n, N$ in $\mathbb{N}$,

$$
\|[\pi_2, X_{(N)}^u(t,s)]\| \leq 4\|E_{(N)}^n(t)\|. \tag{19}
$$

3.3 Proof of Theorem 1

From (18) and (19), since $\|\pi_2 B(1 - \pi_N)\| < \varepsilon / K$,

$$
\|[\pi_2, X_{(N)}^u(t,0)]\| \leq \varepsilon / K.
$$

$$
\|[\pi_2, X_{(N)}^u(t,0)]\| \leq \varepsilon / K,
$$

$$
\|[\pi_2, X_{(N)}^u(t,0)]\| \leq \varepsilon / K.
$$

From (13), $\sup_{t \in [0, (K)]} \|E_{(N)}^n(t)\|$ tends to zero as $n$ tends to infinity. For $n$ large enough,

$$
\|E_{(N)}^n(nT^*)\| \leq \frac{\varepsilon}{4\|\pi_N B(1 - \pi_N)\|},
$$

and $1 - \|[\pi_2, X_{(N)}^u(T_n^*,0)]\| \leq 2\varepsilon$. This completes the proof of Theorem 1.

3.4 Bounded coupling

In this Section, we consider the special case where $B$ is bounded. The propagator $u \mapsto T^u$ is defined on the set of locally integrable functions.

Proposition 6 Let $(A, B, R)$ satisfy Assumption 1. Assume that $B$ is bounded and that $(1, 2)$ is a non-degenerate transition of $(A, B)$. Define $T = \frac{1}{2\sqrt{\lambda_2 - \lambda_1}}$ and $u^* : t \mapsto \cos((\lambda_2 - \lambda_1) t)$. Then, for every $n$ in $\mathbb{N}$, there exists $T_n^*$ in $(n^* T - nT^* + T)$ such that

$$
1 - \|[\pi_2, X_{(N)}^u(T_n^*,0)]\| \leq \frac{(1 + C')\|\pi_2 B\| I}{n}, \tag{21}
$$

with $T^* = \pi/2$, $I = 4/|\lambda_2 - \lambda_1|$, $K = 2/|b_2|$ and

$$
C' = \frac{\varepsilon}{2},
$$

where $N' = \{ (j, k) \in \{1, \ldots, N\}^2 \text{ such that } \{j, k\} \cap \{1, 2\} \neq \emptyset \text{ and } |\lambda_j - \lambda_k| \leq 3/2|\lambda_2 - \lambda_1| \text{ and } b_{jk} \neq 0 \}$. Proof: Fix $\varepsilon > 0$, and define $\eta = \varepsilon / K$. We apply the procedure of Section 3.1 and find $N$ in $\mathbb{N}$ such that $(1 - \pi_N) B \pi_2 \| < \eta. By definition of $N$, for every $n$ in $\mathbb{N}$, for every $t \leq T_n^*$, $\|\pi_2 T_n^* \phi_1 - \pi_2 X_{(N)}^u(t,0) \phi_1 \| \leq \varepsilon$.

A direct computation shows that, for every $\omega > 3\pi / T$,

$$
\left| \int_0^T e^{i\omega t} u^*(t) dt \right| \leq \frac{\omega T^2}{\omega^2 T^2 - 4\pi^2} \leq \frac{3}{5} \pi \leq I.
$$

Hence, for every $j, k$ such that $\lambda_j - \lambda_k > 3/2|\lambda_2 - \lambda_1|$, (9) reads

$$
\left| \int_0^T h_{jk}^n(\tau) d\tau \right| \leq \frac{2|b_{jk}| I}{n}.
$$

Estimate (4) becomes

$$
\frac{1 - \|[\phi_2, X_{(N)}^u(T_n^*,0)]\|}{1 + 2K\|B\|} \leq \frac{(1 + C')\|\pi_2 B\| I}{n},
$$

or, by definition of $N$,

$$
\frac{1 - \|[\phi_2, X_{(N)}^u(T_n^*,0)]\|}{1 + 2K\|B\|} \leq 2\varepsilon + \frac{(1 + C')\|\pi_2 B\| I}{n}.
$$

Conclusion follows when $\varepsilon$ tends to zero. □

Proposition 6 is the translation in mathematical terms of the well-known fact that the difficulty of inducing a given transition between two eigenstates $j$ and $k$ of the free Hamiltonian is mainly due to eigenstates with energy close to $\lambda_j$ or $\lambda_k$. In other words, in the case of bounded coupling, the convergence in Theorem 1 occurs independently of the high energy levels of $A$. These levels are rarely known precisely in practice.

4 Example: Morse potential with truncated dipolar interaction

The approximation procedure of Section 3 has been successfully applied to some classical models, see for instance Boussaid, Caponigro & Chambion [BCC11b] and [BCC11a] for the rotation of a 2D molecule, the infinite square potential well (with bounded coupling operator $B$) and a perturbation of the quantum harmonic oscillator (with unbounded potential $B$).
We present below another type of quantum oscillator. Its main feature is that the spectrum of the free Hamiltonian presents a continuous part and a discrete part. Very few controllability results are known for such systems with mixed spectrum (see Mirrahimi [Mir09] for controllability results based on a Lyapunov approach).

4.1 Modeling

We consider a diatomic molecule submitted to a time variable electric field with support in some bounded domain. The potential energy of the molecule is modeled with the Morse potential (see Morse [Mor29]). With a suitable choice of units, the Schrödinger equation reads

$$\frac{\partial \psi}{\partial t} = -\frac{\partial^2 \psi}{\partial x^2} + V(x)\psi + u(t)W_M(x)\psi, \quad x > 0,$$

where $V : x \mapsto \alpha^2 (e^{-2(1-x^2)} - 2e^{-(x^2-1)})$, and $W_M : x \mapsto x$ if $x \leq M$ and $W_M(x) = 0$ if $x \geq M$. The positive constants $\alpha$ and $x_0$ describe the physical properties of the molecule: the potential $V$ reaches its minimum at $x_0$, which can be considered as the equilibrium length of the molecule and $\alpha$ is related to the depth of the potential well. The constant $M$ (large with respect to $x_0$) is the size of the region where the electric field is assumed to be active. With the notations of Section 1.3, $H = L^2((0, \infty), C)$, $A = i(\Delta - V)$ and $B = -iW_M$.

Define the integer $N = \lfloor \alpha - 1/2 \rfloor$. In this case, we assume that $N \geq 4$. The skew-adjoint operator $A$ admits a finite family simple eigenvalues $(-i\lambda_n)_{0 \leq n < N}$, associated with the eigenfunctions $\phi_n : x \mapsto e^{-i\lambda_n x}P_n(x)$ where $P_n$ is a polynomial function. For every $n \leq N$, $\lambda_n = -(\alpha - n - 1/2)^2$. The spectrum of $A$ contains also a continuous part $[-i(0, +\infty)$.

4.2 Galerkin approximation

We aim to transfer the system from the first energy level to the second one. Since the family $\langle \phi_n \rangle_{n \leq N}$ is not an Hilbert basis of $H$, $(A, B, R)$ does not satisfy Assumption 1 and we cannot apply directly Theorem 1.

Note that $\langle \phi_0, B\phi_1 \rangle$ tends to $\int_0^\infty x\phi_0(x)\phi_1(x)dx \neq 0$ as $M$ tends to infinity. Hence $\langle \phi_0, B\phi_1 \rangle \neq 0$ for $M$ large enough. Moreover, for every $m, \lambda_m - \lambda_1 = (n - m)(-2\alpha + 1 + (n + m))$.

From now on, we do the generic hypothesis that $\alpha \in \mathbb{Q}$. In this case, $\lambda_m - \lambda_1 = \lambda_m' - \lambda_m''$ implies $n - m = n' - m'$ and $n + m = n'' + m''$, that is $(n, m) = (n', m')$. Moreover, for every $\{j, k\}, |\lambda_j - \lambda_k| \in (\mathbb{N} \setminus \{0\})(\lambda_1 - \lambda_0)$ implies $\{j, k\} = \{0, 1\}$.

Fix $\varepsilon > 0$. Inspired by Proposition 6, we define $u^* : t \mapsto \cos((\lambda_1 - \lambda_0)t), T^* = \pi/2, I = 4/(\lambda_1 - \lambda_0)$,

$$K = 2/(\langle \phi_0, B\phi_1 \rangle), \quad A' = \{(j, k) \in \{0, 1, 2, 3\}^2 | \{j, k\} \neq \{0, 1\}\} \text{ and } C'' = \sup_{(j, k) \in A'} \left| \sin\left(\frac{\pi\lambda_j - \lambda_0}{\lambda_1 - \lambda_0}\right)\right|^{-1}.$$

From Theorem 2.1, page 525, of [Kat95], for every $\eta > 0$, there exists a skew-adjoint operator $A_\eta$ such that $A_\eta$ admits a complete family of eigenvectors, $A_\eta \phi_l = A_\eta\phi_l$ for every $l \leq N$ and $\|A_\eta - A\| < \eta$. The pure point spectrum $(-i\lambda_0^\eta)_{n \in \mathbb{N}}$ is everywhere dense in $[-i0, +\infty)$.

The system $(A_\eta, B, R)$ satisfies Assumption 1. For every locally integrable $u$, we denote with $T^*_u$ the propagator of $\frac{d}{dt} \psi = (A_\eta + uB)\psi$.

We choose an integer $n \geq (1 + C'')M(1 + 2M\varepsilon)/\varepsilon$ and we define $\eta = \varepsilon/(nT^* + T)$. The transition $(0, 1)$ of $(A_\eta, B)$ is non degenerate, $\lambda_1 - \lambda_0 = -2(\alpha + 1)$ and, for every $l \geq 4$,

$$\lambda_l - \left(\lambda_0 + \frac{3}{2}(\lambda_1 - \lambda_0)\right) \geq \lambda_4 - \left(\lambda_0 + \frac{3}{2}(\lambda_1 - \lambda_0)\right) \geq 5\alpha - 17 > 0.$$

Proposition 6 applied to system $(A_\eta, B, R)$ gives the existence of $T^*_n \leq nT^* + T$ such that $1 - |\langle \phi_1, T^*_n(\psi)\phi_0 \rangle| \leq \varepsilon$. Since $\|A - A_\eta\| < \eta$, we have, for every $t \leq nT^* + T$,

$$\left\| \Psi^*(t, 0) - \Psi^*_\eta(t, 0) \right\| < (nT^* + T)\varepsilon \leq \varepsilon,$$

and finally $1 - |\langle (\phi_1, T^*_n(\psi)\phi_0) \rangle| < 2\varepsilon$.

5 Conclusion

The contribution of this paper has two elements. First, it proves the validity of the Rotational Wave Approximation for infinite dimensional quantum systems with a pure point spectrum. The result can be partially extended to systems with a mixed spectrum (including a discrete and a continuous part). The convergence results are accompanied by explicit error estimates which provide explicit time estimates for the approximate controllability of bilinear infinite dimensional quantum systems. Second, a notion of efficiency has been introduced. The efficiency is a measure of the $L^1$-norm of the control needed to achieve a given transition between two eigenstates of the free Hamiltonian.

The $L^1$-norm of the control appears to play a central role in the analysis of bilinear quantum systems. This is slightly surprising, since one would rather expect that the $L^2$-norm of the control field represents the energy given
to the system. It may be an artifact of the semi-classical model that disappears with a more realistic model that takes into account a quantized field.

Among other topics, future analysis may concentrate on the design of time-efficient controls or on a systematic treatment of quantum systems when the free Hamiltonian has a continuous spectrum.

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