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Wavefunction controllability for finite-dimensional bilinear quantum systems

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Abstract
We present controllability results for quantum systems interacting with lasers. Exact controllability for the wavefunction in these bilinear systems is proved in the finite-dimensional case under very natural hypotheses.

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1. Introduction

Controlling chemical reactions at the quantum level is a long-lasting goal (cf [1–11]) going back to the very beginning of laser technology. Due to the subtle nature of the interactions involved, manipulation of quantum dynamics is expected to allow for finer control than classical tools (e.g. temperature and pressure) and possibly for new reactions and/or products. Controlling quantum phenomena also goes beyond chemical reactions to encompass many other applications [11].

The earliest experiments showed that designing a laser pulse capable of steering the system to the desired target state is a rather difficult task that physical intuition alone generally cannot accomplish. It is only recently that tools from control theory were introduced and began to give satisfactory results in some particular cases; finding the optimal laser electric field as a design objective is treated by numerical methods and a need exists for new methods that are reliable and computationally inexpensive. A legitimate question arises in this context: what quantum states can be attained using such an external field? Some answers are given below for finite-dimensional quantum systems; a number of theorems in this work are proved, which have been previsously cited including applications [12, 13]. The reader is referred to [13–17] for additional works on this subject, where other controllability definitions are introduced.

2. Dynamical equations

This section introduces the general infinite-dimensional equations for controllability analysis; their discretization is discussed in the next section. Consider a quantum system without control interaction with internal Hamiltonian H_0 and prepared in the initial state $\Psi_0(x)$ where $x \in \mathbf{R}^\gamma$ denotes the relevant coordinate variables; the state $\Psi(x,t)$ at time t satisfies the time-dependent Schrödinger equation

$$i\hbar \frac{\partial}{\partial t}\Psi(x,t) = H_0 \Psi(x,t) \qquad \Psi(x,t=0) = \Psi_0(x) \qquad \|\Psi_0\|_{L^2(\mathbf{R}^\gamma)} = 1. \tag{1}$$

In the presence of an external interaction, taken here as an electric field modelled by a laser amplitude $\epsilon(t) \in \mathbf{R}$ coupled to the system through a time-independent dipole moment operator $\mathcal{B}$,[4] the (controlled) dynamical equations become

$$i\hbar \frac{\partial}{\partial t}\Psi_\epsilon(x,t) = H_0\Psi_\epsilon(x,t) + \epsilon(t)\,\mathcal{B}\Psi_\epsilon(x,t) = H\Psi_\epsilon(x,t) \qquad \Psi_\epsilon(x,t=0) = \Psi_0(x). \tag{2}$$

In order to avoid trivial control problems we suppose $[H_0, \mathcal{B}] \neq 0$, where the Lie bracket $[\cdot,\cdot]$ is defined as $[U, V] = UV - VU$.

The goal is to find, if any, final time $T > 0$ and finite energy laser pulse $\epsilon(t) \in L^2([0,T])$ exist such that $\epsilon(t)$ is able to steer the system from $\Psi_0(x)$ to some predefined target $\Psi_\epsilon(x,T) = \Psi_{\text{target}}(x)$. If the answer to this question is affirmative for any target Ψ_{target}, then the system is called controllable. Given that H is Hermitian one can easily prove that the L^2 norm of Ψ_ϵ is conserved throughout the evolution

$$\|\Psi_\epsilon(x,t)\|_{L^2_x(\mathbf{R}^\gamma)} = \|\Psi_0\|_{L^2(\mathbf{R}^\gamma)} \qquad \forall t > 0. \tag{3}$$

Note that $\Psi_\epsilon(x,t)$ evolves on the unit sphere $S(0,1)$ of $L^2(\mathbf{R}^\gamma)$:

$$S(0,1) = \{f \in L^2(\mathbf{R}^\gamma);\ \|f\|_{L^2(\mathbf{R}^\gamma)} = 1\}.$$

3. Finite-dimensional system

Let $D = \{\Psi_i(x); i = 1, \ldots, N\}$ be the set of the first N, $N \geqslant 3$ eigenstates of the infinite-dimensional Hamiltonian H_0, let M be the linear space they generate, and let A and B be the matrices of the operators H_0 and $\mathcal{B}$ respectively, with respect to this base; as in the infinite-dimensional setting it is supposed that $[A, B] \neq 0$. Negative generic results concerning infinite-dimensional controllability (cf [20–23]) are available that show the need for tailored controllability concepts and for a good understanding of the finite-dimensional case; moreover, the existence of intrinsically finite-dimensional quantum situations ('N-level' systems, spin systems, etc) motivates a finite-dimensional analysis.

We denote $C = (c_i)_{i=1}^N$ as the coefficients of $\Psi_i(x)$ in an expansion of the evolving state $\Psi(t,x) = \sum_{i=1}^N c_i(t)\Psi_i(x)$; equation (2) now becomes

$$i\hbar \frac{\partial}{\partial t} C = AC + \epsilon(t)BC \qquad C(t=0) = C_0 \tag{4}$$

$$C_0 = (c_{0i})_{i=1}^N \qquad c_{0i} = \langle \Psi_0, \Psi_i \rangle \qquad \sum_{i=1}^N |c_{0i}|^2 = 1. \tag{5}$$

[4] Depending on the problem, one may choose to go beyond this first-order, bilinear term when describing the interaction between the laser and the system (cf [18, 19]).

The controllability of equation (4) has been dealt with before (cf [24, 25], see also [26] for an overview of the topic) by considering the problem of a system posed on the space of the unitary matrices of dimension N. This approach has the benefit of drawing on the general tools and results from bilinear controllability on Lie groups. However, verifying those criteria becomes computationally very difficult when N is large; moreover, this approach does not readily illuminate the very simple and intuitive phenomena often at work. The goal of this paper is to provide easy-to-utilize sufficient conditions for the controllability of the wavefunction; these conditions will be expressed only in terms of the readily identified physical properties of the system. This work is a complement to the previous analysis, as the use of stronger (but generally less intuitive) results coming from the theory of controllability on Lie groups is unavoidable in some cases where the present results do not apply. For similar results obtained in the Lie algebra framework the reader is referred to [13].

We remark that the matrix A is diagonal and we make the common assumption that the matrix B is real symmetric (Hermitian). We denote $\lambda_i \in \mathbf{R}, i = 1, \ldots, N$, as the diagonal elements of A (the energies of the states Ψ_i). With the notation $S_M(0,1) = S(0,1) \cap M$, it was previously stated that the system evolves on $S_M(0,1)$, which in a finite-dimensional representation reads

$$\sum_{i=1}^{N} |c_i(t)|^2 = 1 \qquad \forall t \geqslant 0. \tag{6}$$

4. Connectivity graph and necessary conditions

The B matrix plays the critical role of specifying the kinematic coupling amongst the eigenstates of the system reference Hamiltonian matrix A. We associate with the system a graph $G = (V, E)$ called the *connectivity graph* (we refer the reader to [27] for graph theory concepts). We define the set V of vertices as the set of eigenstates Ψ_i and the set of edges E as the set of all pairs of eigenstates coupled by the matrix B. Since B is symmetric we can consider G as non-oriented:

$$G = (V, E): \qquad V = \{\Psi_1, \ldots, \Psi_n\} \qquad E = \{(\Psi_i, \Psi_j); i \neq j, B_{ij} \neq 0\}. \tag{7}$$

We may decompose this graph into (connected) components $G_\alpha = (V_\alpha, E_\alpha), a = 1, \ldots, K$. Note that this decomposition corresponds to a block-diagonal structure of the matrix B (modulo some permutations on the indices). From the definition of G and using equation (4) (A is diagonal) it follows that

$$\mathrm{i}\hbar \frac{\partial}{\partial t} \sum_{\{i;\Psi_i \in V_\alpha\}} |c_i(t)|^2 = 0. \tag{8}$$

Using equation (8) one can write new conservation laws for each component:

$$\sum_{\{i;\Psi_i \in V_\alpha\}} |c_i(t)|^2 = \text{constant} \qquad \forall t > 0 \qquad \alpha = 1, \ldots, K. \tag{9}$$

Denote by $U(A, B, \epsilon, t_1 \to t_2)$ the propagator associated with equation (4); for any state $\chi(t_1)$, $U(A, B, \epsilon, t_1 \to t_2)\chi(t_1)$ is defined as the solution at time $t = t_2$ of equation (4) with the initial state at time $t = t_1$ being $\chi(t_1)$.

Definition 1. *We say that* Ψ_2 *is reachable from* Ψ_1 *if there exists* $0 < T < \infty, \epsilon(t) \in L^2([0, T]; \mathbf{R})$ *such that* $U(A, B, \epsilon(t), 0 \to T)\Psi_1 = \Psi_2$.

This allows us to give necessary conditions for controllability:

Lemma 1. *If the state* $\chi = \sum_{i=1}^{N} d_i \Psi_i(x)$ *is reachable from the initial configuration* C_0 *then*

$$\sum_{\{i;\Psi_i\in V_\alpha\}} |c_{0i}|^2 = \sum_{\{i;\Psi_i\in V_\alpha\}} |d_i|^2 \qquad \alpha = 1, \ldots, K. \tag{10}$$

In order to simplify the presentation of the results we will introduce the following hypothesis:

$\mathbb{HA}$ *The graph G is connected, i.e.* $K = 1$.

The assumption $\mathbb{HA}$ is not restrictive, it is just a matter of specifying the number of independent subsystems we want to simultaneously control (see [23] and [33] for the general case). Note also that this does *not* imply that any two states are directly connected one with another, but only that for any two states Ψ_i and Ψ_j there is a path in the graph G that connects Ψ_i and Ψ_j.

5. Controllability

We denote $\omega_{kl} = \lambda_k - \lambda_l, k, l = 1, \ldots, N$ as the eigenvalue differences for the matrix A, and atomic units ($\hbar = 1$) will be utilized. Consider the hypothesis:

$\mathbb{HB}$ *The connectivity graph G does not have 'degenerate transitions', that is for all* $(i, j) \neq (a, b), i \neq j, a \neq b$ *such that* $B_{ij} \neq 0$, $B_{ab} \neq 0$*:* $\omega_{ij} \neq \omega_{ab}$.

Remark 1. In all that follows this hypothesis could be relaxed to

$\mathbb{HC}$ *The connectivity graph G remains connected after elimination of all edge pairs* $(\Psi_i, \Psi_j), (\Psi_a, \Psi_b)$ *such that* $\omega_{ij} = \omega_{ab}$ *(degenerate transitions).*

However, for ease of presentation $\mathbb{HB}$ will be assumed to be true.

We also introduce one more hypothesis:

$\mathbb{HD}$ *For each* $i, j, a, b = 1, \ldots, N$ *such that* $\omega_{ij} \neq 0$*:* $\frac{\omega_{ab}}{\omega_{ij}} \in Q$*, where Q is the set of all rational numbers.*

Remark 2. The assumption $\mathbb{HD}$ implies that there exists a $T > 0$ such that $U(A, B, 0, 0 \to T) = \mathrm{e}^{-\mathrm{i}TA} = I$ (i.e., the free evolution is periodic). Note that $\mathbb{HD}$ is in particular verified if $\lambda_i \in Q, i = 1, \ldots, N$, which is often the case in practice (e.g., [31]). For an interpretation of this hypothesis in terms of 'wave-packet revival' see [28–30]. Moreover, the controllability result remains true without $\mathbb{HD}$, as is proven in the appendix (also see [23]).

We will conclude with a simple but important remark: the reverse (i.e., the same dynamics but with time reversed) of the system (4) given by $(A, B, \epsilon(t))$ is equivalent to a system of the same kind $(-A, -B, \tilde{e}(t) = \epsilon(-t))$, such that

$$(U(A, B, \epsilon(t), t_1 \to t_2))^{-1} = U(-A, -B, \epsilon(-t), -t_2 \to -t_1).$$

We call $(A, B, \epsilon(t))$ the 'direct system' and $(-A, -B, \tilde{e}(t))$ the corresponding 'reverse system'.

The goal is to prove that under hypotheses $\mathbb{HA}$, $\mathbb{HB}$, $\mathbb{HD}$ the system is controllable, i.e. for any $\Psi_1 \in S(0, 1) \cap M$ the set of reachable states from Ψ_1 is $S(0, 1) \cap M$. The proof has two parts: local controllability and global controllability.

5.1. Local controllability

We begin by introducing two particular subsets of M; if the graph G admits a bipartite decomposition $V = P_1 \cup P_2$, $P_1 \cap P_2 = \emptyset$, $P_1 \neq \emptyset$, $P_2 \neq \emptyset$, $E \subset P_1 \times P_2$ denote

$$X = \left\{ \chi = \sum_{i=1}^{N} w_i \Psi_i; \sum_{i \in P_1} \lambda_i |w_i|^2 = \sum_{j \in P_2} \lambda_j |w_j|^2 \right\}. \tag{11}$$

If G does not have a bipartite decomposition (thus it has at least one odd-length cycle, see [27], p 24) set $X = \emptyset$. We also introduce the set Z:

$$Z = \left\{ \Psi = \sum_{i=1}^{N} c_i \Psi_i; \exists i : c_i = 0 \right\}. \tag{12}$$

Theorem 1. *Let $\Psi \in S_M(0, 1)\backslash X \backslash Z$. Under the assumptions $\mathbb{HA}$, $\mathbb{HB}$, $\mathbb{HD}$ the set of reachable states from Ψ is a neighbourhood of Ψ (in the canonical topology of $S_M(0, 1)$). The same result is true for the reverse system, that is, the set of states from which Ψ can be reached is a neighbourhood of Ψ.*

Proof. We will use on M its *real* Hilbert space structure (and not the canonical *complex* Hilbert space structure) given by the scalar product:

$$\langle \chi_1, \chi_2 \rangle_{\mathbf{R}} = \mathrm{Re}(\langle \chi_1, \chi_2 \rangle) = \tfrac{1}{2}(\langle \chi_1, \chi_2 \rangle + \langle \chi_2, \chi_1 \rangle). \tag{13}$$

Consider the mapping $S : L^2(\mathbf{R}) \times \mathbf{R} \to M$ given by $S(\epsilon, t) = U(A, B, \epsilon, 0 \to t)\Psi$. We want to prove that $S(L^2(\mathbf{R}) \times (0, \infty))$ is a neighbourhood of Ψ. Note that there exists T such that $S(0, T) = \Psi$ (here T is the value given in remark 2 that satisfies $U(A, B, 0, 0 \to T) = I$) and that S is differentiable in $(0, T)$ (see [20]). Therefore, it suffices to prove that the differential DS of S in $(0, T)$ is onto the tangent plane $\mathcal{P}$ in Ψ at $S_M(0, 1)$ given by the equation

$$\mathcal{P} = \{\chi \in M : \langle \chi, \Psi \rangle + \langle \Psi, \chi \rangle = 0\}. \tag{14}$$

Since the image of the differential is a linear space, it is enough to prove that the only $\chi \in M$ such that

$$\begin{aligned} &\langle DS_{(\epsilon,t)=(0,T)}(\epsilon, t), \chi \rangle_{\mathbf{R}} = 0 \qquad \forall (\epsilon, t) \in L^2(\mathbf{R}) \times \mathbf{R} \\ &\langle \chi, \Psi \rangle_{\mathbf{R}} = 0 \end{aligned} \tag{15}$$

is $\chi \equiv 0$. Let χ satisfy (15). Denote by DS_ϵ the differential of S with respect to ϵ in $(0, T)$ and by DS_t the differential of S with respect to t in $(0, T)$. Then (see also [20])

$$\begin{aligned} &DS_\epsilon(\tilde{\mathrm{e}}) = -\mathrm{i} \textstyle\int_0^T \mathrm{e}^{-\mathrm{i}A(T-s)} \tilde{\mathrm{e}}(s) B \, \mathrm{e}^{\mathrm{i}A(T-s)} \Psi \, \mathrm{d}s \\ &DS_t = -\mathrm{i}A\Psi. \end{aligned} \tag{16}$$

So (15) is equivalent to:

$$\begin{aligned} &\mathrm{Im}(\langle \mathrm{e}^{-\mathrm{i}A(T-s)} B \, \mathrm{e}^{\mathrm{i}A(T-s)} \Psi, \chi \rangle) = 0 \qquad \forall 0 < s < T \\ &\mathrm{Im}(\langle A\Psi, \chi \rangle) = 0 \\ &\mathrm{Re}(\langle \Psi, \chi \rangle) = 0. \end{aligned} \tag{17}$$

Denote $\Psi = \sum_i c_i \Psi_i$, $\chi = \sum_i w_i \Psi_i$. Making use of the hypothesis $\mathbb{HB}$ as in [32] we obtain

$$B_{ab}(c_a \overline{w_b} - \overline{c_b} w_a) = 0 \qquad \forall 1 \leqslant a < b \leqslant N \tag{18}$$

$$\mathrm{Re}\left(\sum_{a=1}^{N} c_a \overline{w_a}\right) = 0 \tag{19}$$

$$\mathrm{Im}\left(\sum_{a=1}^{N} \lambda_a c_a \overline{w_a}\right) = 0. \tag{20}$$

Equation (18) implies that for each a, b such that $B_{a,b} \neq 0$

$$\frac{w_a}{c_a} = \overline{\left(\frac{w_b}{c_b}\right)}. \tag{21}$$

Since the connectivity graph is fully connected we obtain easily that there exists a complex constant α such that for each $1 \leqslant a \leqslant N$ $w_a = \alpha c_a$ or $w_a = \overline{\alpha} c_a$. If G is not bipartite then it has an odd-length cycle. Using (21) along this cycle one obtains $\alpha \in \mathbf{R}$ so $\chi = \alpha\Psi$ and by (19) it follows that $\alpha = 0$ so $\chi \equiv 0$.

If G is bipartite with decomposition $V = P_1 \cup P_2$ we conclude from (21) that

$$w_a = \alpha c_a \qquad \forall a \in P_1 \qquad w_a = \overline{\alpha} c_a \qquad \forall a \in P_2. \tag{22}$$

From (19) and (20) one concludes that either $\alpha = 0$ (so $\chi \equiv 0$) or $\Psi \in X$. Replacing $(A, B, \epsilon(t))$ by $(-A, -B, \epsilon(-t))$ one obtains the second part of the theorem. □

5.2. Global controllability

Theorem 2. *Under the assumptions* $\mathbb{HA}$, $\mathbb{HB}$, $\mathbb{HD}$ *the system (4) is controllable, that is for any* $\Psi \in S_M(0, 1)$ *the set of reachable states from* Ψ *is* $S_M(0, 1)$*; the same result is true for the reverse system.*

Proof. The proof is based on the following lemmas: □

Lemma 2 *('exit lemma').* *For any* $\Psi \in S_M(0, 1)$ *there exists at least one state in* $S_M(0, 1)\backslash X\backslash Z$ *that can be reached from* Ψ*; the same is true for the reverse system.*

Lemma 3 *('pass lemma').* *If* $X \neq \emptyset$ *then, in any given open (for the canonical topology of* $X \cap S_M(0, 1)$*) subset* V *of* $X \cap S_M(0, 1)$ *there exists a 'pass state'* $\gamma \in V\backslash Z$ *such that from* γ *one can reach at least one point in any (of the two)* local in γ *connected components of* $S_M(0, 1)\backslash X$ *separated by X; moreover, these points can be chosen not to be in Z; the same is true for the reverse system.*

Suppose lemmas 2 and 3 are both true; suppose also $X \neq \emptyset$ (the simpler alternative $X = \emptyset$ follows along the same lines). By the 'exit lemma' it is enough to prove (for the direct and inverse system) that for any $\Psi \in S_M(0, 1)\backslash X\backslash Z$ the set of reachable states from Ψ is $S_M(0, 1)\backslash X\backslash Z$. That is, use the lemma for the direct system to reach a state in $S_M(0, 1)\backslash X\backslash Z$, and use it once more for the reverse system to obtain a state in $S_M(0, 1)\backslash X\backslash Z$ from which the target can be reached and in the 'middle' use the controllability from $S_M(0, 1)\backslash X\backslash Z$ to $S_M(0, 1)\backslash X\backslash Z$. The proof proceeds in two steps.

(i) Suppose the initial state ϕ and target δ are in the same connected component of $S_M(0, 1)\backslash X$. Then there exists a continuous curve $C(t) : [0, 1] \to S_M(0, 1)\backslash Z\backslash X$ with $C(0) = \phi$, $C(1) = \delta$. We will prove that each $C(t)$, $t \in [0, 1]$ is reachable from ϕ. We denote by η the minimal value of t such that $C(t)$ is not reachable from ϕ. By the local controllability result for the state ϕ we obtain $\eta > 0$. Since $C(\eta) \in S_M(0, 1)\backslash Z\backslash X$ one can apply the local result for the reverse system in $C(\eta)$ and deduce that there exists $\eta' < \eta$ such that $C(\eta)$ is reachable from $C(\eta')$. But, by the minimal property of η, $C(\eta')$ is reachable from ϕ so by transitivity $C(\eta)$ is also reachable from ϕ.

(ii) Let the initial state ϕ and target δ be in different connected components of $S_M(0,1)\backslash X$. For the sake of simplicity suppose that the connected components are adjacent (two components are called adjacent if the intersection of their frontiers has a non-void interior in the canonical topology of $S_M(0,1)\cap X$), the general case being a mere reiteration of the arguments below. It can be proved, see also the discussion on the geometry of the set X below, that any two components of $S_M(0,1)\backslash X$ can be linked by a chain of adjacent components. Then there exists a 'pass state' $\gamma \in X\backslash Z$ given by lemma 3 on the boundary of the two connected components. By the properties of a 'pass state' there exist two states ϕ' (in the same component as ϕ) and χ' (in the same component as χ) and an electric field such that the corresponding evolution starting from ϕ' passes by γ and arrives at χ'. Since by the previous case ϕ' is reachable from ϕ and χ from χ', an electric field realizing an evolution $\phi \to \phi' \to \gamma \to \chi' \to \chi$ can be found, and therefore χ is reachable from ϕ, which concludes our proof.

Before giving proofs for the lemmas above let us denote by D_i the L^2 projector to $\Psi_i, i = 1,\ldots,N$ and by O the operator $\sum_{i\in P_1}\lambda_i D_i - \sum_{j\in P_2}\lambda_j D_j$. We make use of the classical 'bra-ket' notation for self-adjoint operators V (such as O, D_i): $\langle\chi_1|V|\chi_2\rangle := \langle\chi_1, V\chi_2\rangle = \langle V\chi_1,\chi_2\rangle$. We obtain the following characterizations

$$X = \{\chi;\langle\chi|O|\chi\rangle = 0\} \qquad Z = \bigcup_{i=1}^{N}\{\chi;\langle\chi|D_i|\chi\rangle = 0\}. \tag{23}$$

Note also $[H_0, O] = [H_0, D_i] = 0, i = 1,\ldots,N$, but $[\mathcal{B}, O] \neq 0$, $[\mathcal{B}, D_i] \neq 0, i = 1,\ldots,N$. We will use the same notation for the matrices of these operators with respect to the base D.

Proof of lemma 2. (a) We begin by proving that for any $k = 1,\ldots,N$, $\chi \in S_M(0,1)$, $\eta > 0$, and $\tau > 0$ there exists at least an $\epsilon(t) \in L^2(0,\tau)$, $\|\epsilon\|_{L^2} < \eta$: such that

$$\{U(A,B,\epsilon(t),0\to s)\chi; 0\leqslant s\leqslant\tau\}\backslash D_k^{-1}\{0\} \neq \emptyset. \tag{24}$$

Denote $U(A,B,\epsilon(t),0\to s)\chi = \chi(s) = \sum_{l=1}^{N} c_l(s)\Psi_l$ as the solution of (4). Suppose (24) is not true, then $c_k(s)$ vanishes on $[0,\tau]$ as well as all its derivatives, for any smooth electric field $\epsilon(t) \in C^\infty \cap L^2(0,\tau)$, $\|\epsilon\|_{L^2} < \eta$. We obtain to first order:

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}c_k(s) = \epsilon(s)\sum_{j=1}^{N} B_{kj}c_j(s) = 0 \qquad \forall s\in[0,\tau] \quad \epsilon(t)\in C^\infty\cap L^2(0,\tau) \quad \|\epsilon\|_{L^2} < \eta. \tag{25}$$

Take $\epsilon_n(t) = \frac{\eta}{n\sqrt{\tau}}$ and denote by $\chi_n(s) = \sum_{l=1}^{N} c_{nl}(s)\Psi_l$ the corresponding evolution. Since $\epsilon_n(s) \neq 0$ on $[0,\tau]$ it follows that

$$\sum_{j=1}^{N} B_{kj}c_{nj}(s) = 0 \qquad 0\leqslant s\leqslant\tau \quad n = 1,\ldots. \tag{26}$$

For $n\to\infty$ the limiting trajectory is the free evolution $c_j(s) = \mathrm{e}^{-\mathrm{i}\lambda_j s}c_j(0)$, therefore

$$\sum_{j=1}^{N} B_{kj}c_j(s) = \sum_{j=1}^{N} B_{kj}\,\mathrm{e}^{-\mathrm{i}\lambda_j s}c_j(0) = 0 \qquad 0\leqslant s\leqslant\tau. \tag{27}$$

By the hypothesis $\mathbb{HB}$ this can be true only if $c_j(0) = 0$ for all j connected to k in G ($B_{kj} \neq 0, j\neq k$). Selecting the initial time arbitrarily in $[0,\tau]$ one obtains that for any $\epsilon(t) \in L^2([0,\tau])$, $\|\epsilon\|_{L^2} < \eta$ and corresponding evolution $U(A, B, \epsilon(t), 0 \to s)\chi = \chi(s) = \sum_{l=1}^{N} c_l(s)\Psi_l$ the coefficient $c_j(s)$ is zero for all $s \in [0, \tau]$ and all j connected to k in G. Repeating this reasoning as many times as necessary (starting each time from the newly obtained zero coefficients) and using the *connected* graph structure of B it follows that $c_j(s) = 0$, $0 \leqslant s \leqslant \tau$, $j = 1, \ldots, N$, which is in obvious contradiction with $\chi \in S_M(0, 1)$.

(b) An immediate consequence of the assertion (24) is that for each state $\chi \in S_M(0, 1)$ and each neighbourhood V of χ there exists a reachable state from χ that is not in Z.

(c) Since Z is a closed set, all that remains to prove is that for any state $\chi \in S_M(0, 1)\backslash Z$ and neighbourhood V of χ there exists at least one reachable state from χ in $V \cap S_M(0, 1)\backslash X$.

Suppose that this is not true; then there exists $\chi \in S_M(0, 1)\backslash Z$, $\eta > 0$, and $\tau > 0$ such that for any $\epsilon(t) \in L^2(0, \tau)$, $\|\epsilon\|_{L^2} < \eta$:

$$\langle U(A, B, \epsilon(t), 0 \to s)\chi|O|U(A, B, \epsilon(t), 0 \to s)\chi\rangle = 0 \qquad \forall s \in [0, \tau]. \tag{28}$$

Denote $U(A, B, \epsilon(t), 0 \to s)\chi = \chi(s) = \sum_{l=1}^{N} c_l(s)\Psi_l$ as the solution of (4) and $O(t) = \langle\chi(t)|O|\chi(t)\rangle$. Then for any $\epsilon(t) \in L^2(0, \tau)$, $\|\epsilon\|_{L^2} < \eta$, $O(t)$ and all its derivatives vanish in $[0, \tau]$. To compute the first derivative use the formula for the evolution of an observable represented by a matrix V: $V(t) = \langle\chi(t)|V|\chi(t)\rangle$:

$$\frac{\mathrm{d}}{\mathrm{d}t}V(t) = \langle\chi(t)|\mathrm{i}[A, V]|\chi(t)\rangle + \epsilon(t)\langle\chi(t)|\mathrm{i}[B, V]|\chi(t)\rangle. \tag{29}$$

Denote $J = [B, O]$; there exists $a \neq b$ such that $J_{ab} \neq 0$, $B_{ab} \neq 0$ and for $V = O$ in (29):

$$\frac{\mathrm{d}}{\mathrm{d}s}O(s) = \epsilon(s)\langle\chi(s)|\mathrm{i}J|\chi(s)\rangle = 0 \qquad \forall s \in [0, \tau] \quad \epsilon(t) \in C^\infty \quad \|\epsilon\|_{L^2} < \eta. \tag{30}$$

Using the same technique as above one concludes that

$$J_{ij}\mathrm{Re}(c_i(s)\overline{c_j(s)}) = 0 \qquad i \neq j \quad i, j = 1, \ldots, N \quad \forall s \in [0, \tau] \tag{31}$$

so finally

$$\mathrm{Re}(c_a(s)\overline{c_b(s)}) = 0 \qquad \forall s \in [0, \tau] \quad \forall \epsilon(t) \in L^2(0, \tau) \quad \|\epsilon\|_{L^2} < \eta. \tag{32}$$

It suffices to note that $2\,\mathrm{Re}(c_a(s)\overline{c_b(s)}) = \langle c_a(s), c_b(s)\rangle + \langle c_b(s), c_a(s)\rangle$ is (a particular observable) *not* conserved by the free evolution ($\epsilon(t) \equiv 0$), so (32) cannot be true. □

Proof of lemma 3. We begin with some geometry considerations concerning the set X. Following the definition (23) denote by $f : S_M(0, 1) \to \mathbf{R}$ the function $f\left(\sum_{i=1}^{N} z_i\Psi_i\right) = \sum_{i\in P_1} \lambda_i|z_i|^2 - \sum_{j\in P_2} \lambda_j|z_j|^2$. Then $X = f^{-1}\{0\}$. The differential Df of f never vanishes in general and vanishes only on $KD = \{\mathrm{e}^{\mathrm{i}\phi}\Psi_k; 0 \leqslant \phi \leqslant 2\pi\}$ if some $\lambda_k = 0$, so for any open set $V_1 \in S_M(0, 1) \cap X$ there exists a subset $V_2 \subset V_1$ such that Df never vanishes on V_2; locally on V_2 only two connected components $f^{-1}(]0, \infty[) \cap V_2$ and $f^{-1}(]-\infty, 0[) \cap V_2$ are present and globally KD does not introduce new connected components. For any two points $\phi, \delta \in S_M(0, 1)\backslash X$ there exists a continuous curve from ϕ to δ that does not intersect KD, the real codimension of KD in X being at least 2. We can therefore suppose $V \cap KD = \emptyset$.

Let $\chi(s)$ be the solution of (4) for initial data $\chi(0)$ and electric field $\epsilon(t)$. By the definition of X (cf (23)) the local connected components separated by X in $S_M(0, 1)$ correspond to regions where the observable O has constant sign. In order to prove this lemma it is therefore enough to find a $\gamma \in V\backslash Z$ such that $\langle\gamma|[\mathcal{B}, O]|\gamma\rangle \neq 0$, with at least one state in each connected component being then reached from γ by choosing the apropriate sign for $\epsilon(0)$. Since $V\backslash Z \neq \emptyset$ there exists $\gamma' \in V\backslash Z$. Note as above by J the matrix representation of $[\mathcal{B}, O]$ in the basis D and find $a \neq b$ such that $J_{ab} \neq 0$. Choose τ such that the free evolution $\gamma'(s) = \sum_{i=1}^{N} g_i(s)\Psi_i$ of a system starting from $\gamma'(0) = \gamma' \in X$ does not exit $V\backslash Z$ before time $s = \tau$ (when the laser is off the system is *guaranteed* to remain in X). We have seen before that the equality $\mathrm{Re}(g_a(s)\overline{g_b(s)}) = 0$ is not conserved during the free evolution, so we may also suppose $\mathrm{Re}(g_a(0)\overline{g_b(0)}) \neq 0$. If at least one $s \in [0, \tau]$ is found such that $\langle \gamma'(s)|J|\gamma'(s)\rangle \neq 0$ the lemma is proved; if this is not true, note that $J_{ij} \neq 0$ only when $B_{ij} \neq 0$ and use the formula for the free evolution and the hypothesis $\mathbb{HB}$ to obtain that $J_{ab}\,\mathrm{Re}(g_a(0)\overline{g_b(0)}) = 0$, which is a contradition. $\square$

Remark 3. Even when the Hamiltonian matrix A does not comply with $\mathbb{HB}$, theorem 2 may still be used; it suffices to find a $\mu \in \mathbf{R}$ such that the eigenvalues of $A + \mu B$ satisfy $\mathbb{HB}$, apply theorem 2 for the system $(A + \mu B, B)$ (eventually after having applied a rotation so that $A + \mu B$ is diagonal) and obtain a field $\tilde{\mathrm{e}}(t)$; the answer is then the field $\tilde{\mathrm{e}}(t) + \mu$ as the system $(A + \mu B, B, \tilde{\mathrm{e}})$ is equivalent to $(A, B, \tilde{\mathrm{e}}(t) + \mu)$.

For completeness we mention the following result obtained in collaboration with Mathieu Pilot; note that here we do not make use of the hypothesis $\mathbb{HD}$ (see [23], p 167) :

Theorem 3. *Under the assumptions* $\mathbb{HA}$, $\mathbb{HB}$ *the system (4) is controllable, that is for any* $\Psi \in S_M(0, 1)$ *the set of reachable states from* Ψ *is* $S_M(0, 1)$*; the same result is true for the reverse system.*

6. Discussion and conclusions

Wavefunction controllability of finite-dimensional bilinear quantum systems was analysed and sufficient conditions were found under reasonable physical hypothesis on the system under consideration. Under hypothesis $\mathbb{HB}$ the only restrictions on the attainable set appear from conservation laws (equation (10)) in effect. The status of the hypothesis $\mathbb{HB}$ is more subtle; in certain cases its removal brings about new conservation laws (that will necessarily contract the attainable set) very different from those in equation (10). On the other hand, an analysis of the case $N = 3$ leads us to state the following

Conjecture. *As long as no new conservation laws—besides* L^2 *norm conservation—appear, the system is controllable, i.e. any state on the unit sphere can be reached (in finite time and with finite laser energy) from any other.*

The merit of the formulation above is intrinsically related to the properties of the systems and *not* on their mathematical transcription. The existence of conservation laws possibly may *prevent* controllability or correspondingly just restrict the set of attainable states (i.e., if the necessary conditions thus introduced are also sufficient). On the other hand, we remark that in some cases, in the absence of $\mathbb{HB}$, conservation laws may involve quantities that are *not* necessarily observables.

At the numerical level, various tools are available in order to find a control that drives the system to some predetermined goal especially through the optimal control formalism [33–36]. We point out that, since these tools do not provide any *a priori* indication on how close the system can be steered to the target, the controllability criteria enunciated above are important ingredients in assessing the quality of the numerical results.

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Appendix. Proof of theorem 2 without the use of hypothesis $\mathbb{HD}$

The aim of this appendix is to prove theorem 2 in the absence of the hypothesis $\mathbb{HD}$; this proof was obtained in collaboration with Mathieu Pilot from CERMICS-ENPC, Champs sur Marne, 77455 Marne la Vallee Cedex, France.

We have seen that hypothesis $\mathbb{HD}$ implies that the free evolution is periodic, i.e. there exists a $T > 0$ such that $U(A, B, 0, 0 \to T) = \mathrm{e}^{-\mathrm{i}TA} = I$. Suppose now the hypothesis $\mathbb{HD}$ is not true. Let us remark that due to the finite dimensionality of the system the following quasi-periodicity property is true:

Lemma 4. *For each $\eta > 0$, $M > 0$, there exists $T_\eta > M$ such that $\|\mathrm{e}^{-\mathrm{i}T_\eta A} - I\| < \eta$.*

Proof. Let $T > 0$ and consider the set $\{\mathrm{e}^{-\mathrm{i}(n\cdot T)A}; n \in \mathbf{N}\}$. Then one of the following alternatives is true:

(i) there exist $p \neq q \in \mathbf{N}$ such that $\mathrm{e}^{-\mathrm{i}(p\cdot T)A} = \mathrm{e}^{-\mathrm{i}(q\cdot T)A}$;
(ii) for any $p \neq q \in \mathbf{N}$, $\mathrm{e}^{-\mathrm{i}(p\cdot T)A} \neq \mathrm{e}^{-\mathrm{i}(q\cdot T)A}$.

If the first case is true then, supposing $p > q$, we obtain the periodicity: $\mathrm{e}^{-\mathrm{i}((p-q)\cdot T)A} = I$ so in particular lemma 4 is true with T_η independent of η: $T_\eta = (p-q)\cdot T$. If $T_\eta < M$ choose a multiple of T_η large enough.

If the second case is true, note that all matrices in the set $\{\mathrm{e}^{-\mathrm{i}(n\cdot T)A}; n \in \mathbf{N}\}$ are unitary, so in particular their Euclidean norms are bounded. Then, considering for any $\eta > 0$ the union of balls $B(\mathrm{e}^{-\mathrm{i}(n\cdot T)A}, \eta)$ of radius η centred around each element of the *infinite* set $\{\mathrm{e}^{-\mathrm{i}(n\cdot T)A}; n \in \mathbf{N}\}$ it is clear that there exists at least a pair of balls centred in $\mathrm{e}^{-\mathrm{i}(p_\eta\cdot T)A}$ and $\mathrm{e}^{-\mathrm{i}(q_\eta\cdot T)A}$ with $p_\eta - q_\eta > \frac{M}{T}$ having non-empty intersection, otherwise their union will have infinite Lebesque measure, in contradiction with the statement above (we denote by $B(x, r)$ the ball of centre x and radius r in the canonical metric of the finite-dimensional state space).

Thus we obtain $p_\eta, q_\eta \in \mathbf{N}$ such that $\|\mathrm{e}^{-\mathrm{i}(p_\eta\cdot T)A} - \mathrm{e}^{-\mathrm{i}(q_\eta\cdot T)A}\| \leqslant \eta$. But since $\mathrm{e}^{-\mathrm{i}(q_\eta\cdot T)A}$ is unitary it follows $\|\mathrm{e}^{-\mathrm{i}(p_\eta\cdot T)A} - \mathrm{e}^{-\mathrm{i}(q_\eta\cdot T)A}\| = \|(\mathrm{e}^{-\mathrm{i}((p_\eta-q_\eta)\cdot T)A} - I)\cdot \mathrm{e}^{-\mathrm{i}(q_\eta\cdot T)A}\| = \|(\mathrm{e}^{-\mathrm{i}((p_\eta-q_\eta)\cdot T)A} - I)\| \cdot \|\mathrm{e}^{-\mathrm{i}(q_\eta\cdot T)A}\| = \|(\mathrm{e}^{-\mathrm{i}((p_\eta-q_\eta)\cdot T)A} - I)\|$ which gives the conclusion for $T_\eta = (p_\eta - q_\eta)\cdot T$. □

The controllability result in theorem 2 uses the periodicity hypothesis only by the intermediary of the local controllability in theorem 1. Therefore, in order to prove that theorem 2 remains valid in the absence of $\mathbb{HD}$ all that is to be proved is that theorem 1 remains valid in the absence of $\mathbb{HD}$. Let us remark that in the absence of $\mathbb{HD}$ the local result reads:

Lemma 5. *Let $\Psi \in S_M(0,1)\backslash X\backslash Z$, and suppose that the graph associated with the coupling matrix B is connected and has no degenerate transitions. Then, for any $T > 0$ the set of reachable states from Ψ contains a sphere of radius $r_{T,\Psi}$ (in the canonic metric of $S_M(0,1)$) centred around $\mathrm{e}^{-\mathrm{i}TA}\Psi$.*

Let then $\Psi \in S_M(0,1)$ be given and find T_0 such that

$$\|\mathrm{e}^{-\mathrm{i}(T+T_0)A} - I\| \leqslant \frac{r_{T,\Psi}}{2}. \tag{A.1}$$

Note that by equation (A.1) $B\big(\Psi, \frac{r_{T,\Psi}}{2}\big) \subset B(\mathrm{e}^{-\mathrm{i}(T+T_0)A}\Psi, r_{T,\Psi})$. Consider a target state $y \in B\big(\Psi, \frac{r_{T,\Psi}}{2}\big)$ $y \in B(\mathrm{e}^{-\mathrm{i}(T+T_0)A}\Psi, r_{T,\Psi})$, so that $\mathrm{e}^{\mathrm{i}T_0A}y \in \mathrm{e}^{\mathrm{i}T_0A}B(\mathrm{e}^{-\mathrm{i}(T+T_0)A}\Psi, r_{T,\Psi})$; since the internal Hamiltonian evolution is unitary we obtain

$$\mathrm{e}^{\mathrm{i}T_0A}B(\mathrm{e}^{-\mathrm{i}(T+T_0)A}\Psi, r_{T,\Psi}) = B(\mathrm{e}^{-\mathrm{i}TA}\Psi, r_{T,\Psi}).$$

By lemma 5 it follows that $e^{iT_0A}y$ is reachable from Ψ ; but y is reachable from $e^{iT_0A}y$ by the free evolution (for final time equal to T_0) so we conclude that y is reachable from Ψ, which proves the following local result:

Theorem 4. *Let $\Psi \in S_M(0, 1)\backslash X\backslash Z$, and suppose that the graph associated with the coupling matrix B is connected and has no degenerate transitions. Then the set of reachable states from Ψ is a neighbourhood of Ψ (in the canonic topology of $S_M(0, 1)$). The same result is true for the reverse system.*

Finally, let us mention for the sake of completeness the global result that can be proved from this local controllability theorem:

Theorem 5. *Suppose that the graph associated with the coupling matrix B is connected and has no degenerate transitions. Then the system (4) is controllable, that is for any $\Psi \in S_M(0, 1)$ the set of reachable states from Ψ is $S_M(0, 1)$; the same result is true for the reverse system.*

References

[1] Demiralp M and Rabitz H 1993 Optimally controlled quantum molecular dynamics: the effect of nonlinearities on the magnitude and multiplicity of control-field solutions *Phys. Rev.* A **47** 831

[2] Tannor D J and Rice S A 1985 Control of selectivity of chemical reaction via control of wave packet evolution *J. Chem. Phys.* **83** 5013–8

[3] Shi S, Woody A and Rabitz H 1988 Optimal control of selective vibrational excitation in harmonic linear chain molecules *J. Chem Phys.* **88** 6870

[4] Pierce A P, Dahleh M A and Rabitz H 1988 *Phys Rev.* A **37** 4950

[5] Brumer P and Shapiro M 1989 Coherence chemistry: controlling chemical reactions with lasers *Acc. Chem. Res.* **22** 12 407–13

[6] Shi S and Rabitz H 1992 Optimal control of selectivity of unimolecular reactions via an excited electronic state with designed lasers *Chem. Phys.* **97** 276–87

[7] Kime K 1993 Control of transition probabilities of the quantum-mechanical harmonic oscillator *Appl. Math. Lett.* **6** 11–5

[8] Warren W S, Rabitz H and Dahleh M 1993 Coherent control of quantum dynamics: the dream is alive *Science* **259** 1581–9

[9] Kobayashi M 1998 Mathematics make molecules dance *SIAM News* **31** 24

[10] Le Bris C 2000 Control theory applied to quantum chemistry: some tracks *Int. Conf. on Systems Governed by PDEs (Nancy, March 1999) (ESAIM : Proc. vol 8)* pp 77–94

[11] Rabitz H, de Vivie-Riedle R, Motzkus M and Kompa K 2000 Whither the future of controlling quantum phenomena? *Science* **288** 824–8

[12] Turinici G and Rabitz H 2001 Quantum wavefunction controllability *Chem. Phys.* **267** 1–9

[13] Altafini C 2002 Controllability of quantum mechanical systems by root space decomposition of *su(N) J. Math. Phys.* **43** 2051–62

[14] Albertini F and D'Alessandro D 2001 Notions of controllability for quantum mechanical systems *Preprint* quant-ph/0106128

[15] Schirmer S G, Solomon A I and Leahy J V 2002 Degrees of controllability for quantum systems and application to atomic systems *J. Phys. A: Math. Gen.* **35** 4125–41

[16] Fu H, Schirmer S G and Solomon A I 2001 Complete controllability of finite-level quantum systems *J. Phys. A: Math. Gen.* **34** 1679–90

[17] Khaneja N and Glaser S 2000 Cartan decomposition of $SU(2^n)$, constructive controllability of spin systems and universal quantum computing *Preprint* quant-ph/0010100

[18] Dion C M *et al* 1999 *Chem. Phys. Lett* **302** 215–23

[19] Dion C M, Keller A, Atabek O and Bandrauk A D 1999 *Phys. Rev.* A **59** 1382

[20] Ball J M, Marsden J E and Slemrod M 1982 Controllability for distributed bilinear systems *SIAM J. Control Optimization* **20** 575–97

[21] Huang G M, Tarn T J and Clark J W 1983 On the controllability of quantum-mechanical systems *J. Math. Phys.* **24** 2608–18

[22] Turinici G 2000 Controllable quantities for bilinear quantum systems *39th IEEE Conf. on Decision and Control (Sydney Convention and Exhibition Centre, 12–15 Dec.)*

[23] Turinici G 2000 Analysis of numerical methods of simulation and control in quantum chemistry *PhD Thesis* Paris VI University, Paris (http://www-rocq.inria.fr/Gabriel. Turinici)

[24] Ramakrishna V *et al* 1995 Controlability of molecular systems *Phys. Rev.* A **51** 960–6

[25] Brockett R W 1973 Lie theory and control systems defined on spheres *SIAM J. Appl. Math.* **25** 213–25

[26] Butkovskiy A G and Samoilenko Yu I 1990 *Control of Quantum-Mechanical Processes and Systems* (Dordrecht: Kluwer)

[27] Diestel R 2000 *Graph Theory (Graduate Texts in Mathematics vol 173)* 2nd edn (New York: Springer)

[28] Eberly J H, Narozhny N B and Sanchez-Mondragon J J 1980 Periodic spontaneous collapse and revival in a simple quantum model *Phys. Rev. Lett.* **44** 1323–6

[29] Averbukh I Sh and Perelman N F 1989 Fractional revivals: universality in the long-term evolution of quantum wave packets beyond the correspondence principle dynamics *Phys. Lett.* A **139** 449–53

[30] Leichtle C, Averbukh I Sh and Schleich W P 1999 Generic structure of multilevel quantum beats *Phys. Rev. Lett.* **77** 3999–4002

[31] Tersigni S H, Gaspard P and Rice S A 1990 On using shaped light pulses to control the selectivity of product formation in a chemical reaction: an application to a multiple level system *J. Chem. Phys.* **93** 1670–80

[32] Turinici G 2000 On the controllability of bilinear quantum systems *Mathematical Models and Methods for ab initio Quantum Chemistry* ed M Defranceschi and C LeBris *(Lecture Notes in Chemistry vol 74)* (Berlin: Springer)

[33] Li B, Turinici G, Ramakhrishna V and Rabitz H 2002 Optimal dynamic discrimination of similar molecules through quantum learning control *J. Phys. Chem.* B **106** 8125

[34] Tannor D, Kazakov V and Orlov V 1992 Control of photochemical branching: novel procedures for finding optimal pulses and global upper bounds *Time Dependent Quantum Molecular Dynamics* ed J Broeckhove and L Lathouwers (New York: Plenum) pp 347–60

[35] Zhu W and Rabitz H 1998 A rapid monotonically convergent iteration algorithm for quantum optimal control over the expectation value of a positive definite operator *J. Chem. Phys.* **109** 385–91

[36] Hornung T, Motzkus M and de Vivie-Riedle R 2001 Adapting optimal control theory and using learning loops to provide experimentally feasible shaping mask patterns *J. Chem. Phys.* **115** 3105–11