Control through operators for quantum chemistry

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Abstract—We consider the problem of operator identification in quantum control. The free Hamiltonian and the dipole moment are searched such that a given target state is reached at a given time. A local existence result is obtained. As a by-product, our works reveals necessary conditions on the laser field to make the identification feasible. In the last part of this work, some Newton algorithms are proposed together with a continuation method to compute effectively these operators.

I. INTRODUCTION

In the last decades, quantum control has known significant improvements both at theoretical and practical levels (cf.[1], [2], [3], [4] and references therein). Results have been obtained on existence of controls [5], [6], [7], [8], [9] or efficient ways to compute and carry out laser fields that achieve some goals concerning the state of quantum systems [10], [11], [12], [13], [14], [15]. On the other hand, the design of relevant laser fields plays also a major role when the goal is to identify some properties of the quantum system to be controlled. In this way, some methods have been designed to identify finite dimensional systems characteristics [16], or to compute discriminant laser fields [17].

Note that operator identification in relation to the Schrödinger equation has already been studied in the literature. As an example, we refer to [18] for a theoretical result, where no laser interaction is considered.

In this paper, we focus on the case where only one fixed laser is used to identify in finite given time the free Hamiltonian and the dipole moment. From the theoretical point of view, we obtain a local existence result: we prove that the inversion is always possible in the neighborhood of some particular states. As a by-product, we emphasize some features of the laser fields that enables the identification.

Following the local approach we use to obtain this result, we present in a second part, a time discretized setting and fixed-point methods to solve numerically our problem. In particular, a Newton method is proposed together with a continuation method that allows us to solve problems where the local assumption does not hold.

This paper is organized as follows: the mathematical formulation of our problem is given in Section II and a local controllability result is presented in Section III. In Section IV, we present the algorithms to solve numerically the identification problem. We conclude with some tests in Section V.

Let us finally introduce some notations concerning particular matrix sets that will be used throughout the paper. Given $N_d \in \mathbb{N}$, we denote by $\mathbb{C}^{N_d,N_d}$ and $\mathbb{R}^{N_d,N_d}$ the sets of matrices of size $N_d \times N_d$ with complex and real coefficients respectively. Then, define

$$U = \{ M \in \mathbb{C}^{N_d,N_d}, \ M^* M = M M^* = I d \} ,$$

$$S = \{ M \in \mathbb{C}^{N_d,N_d}, \ M^* = M \} ,$$

$$S_R = \{ M \in \mathbb{R}^{N_d,N_d}, \ M^* = M \} ,$$

$$S_{R}^0 = S_R \cap \{ M \in \mathbb{R}^{N_d,N_d}, \ M_{k,k} = 0, \ k = 1, \ldots, N_d \} ,$$

where $M^*$ denotes the adjoint matrix associated to $M$ and $I d$ is the identity matrix of $\mathbb{C}^{N_d,N_d}$. Here, for the sake of simplicity, we have omitted the dependence of these sets with respect to $N_d$. In what follows, we denote by $\mathbb{R} z$ and $\mathbb{R} z$ denote respectively the real and imaginary parts of a complex number $z$. Given a matrix $M$, we denote by $M^T$ its transposed.

II. SETTING OF THE PROBLEM

Fix $T > 0$, and consider a system $U(t) \in U$ whose dynamics over $[0,T]$ is ruled by the Schrödinger equation:

$$i \dot{U}(t) = [H_0 + \varepsilon(t) \mu] U(t),$$

$$U(0) = U_{\text{init}},$$

where $H_0 \in S_R$ is the matrix of the internal Hamiltonian, $\varepsilon(t) \in L^2(0,T;\mathbb{R})$ a laser field, $\mu \in S_R$ the matrix associated with the dipole moment. For relevant applications, the matrices $H_0$ and $\mu$ are not supposed to commute. The initial state $U_{\text{init}}$ is fixed. In this equation, $\varepsilon$ is given and the pair $(H_0, \mu) \in S_R \times S_R^0$ is searched such that at time $t = T$, the state reaches a given target state $U_{\text{target}}$, i.e.,

$$U(T) = U_{\text{target}}.$$

In other words, given the mapping

$$\varphi : S_R \times S_R^0 \rightarrow U$$

$$(H_0, \mu) \mapsto U(T),$$

the main question that will be investigated in this paper is the surjectivity of $\varphi$.

In our work, the internal Hamiltonian $H_0$ is searched as real Hermitian (i.e. symmetric) matrix. This is a particular situation as in general it is only supposed to be complex Hermitian and not real. Nevertheless, for the applications we have in mind this restriction is very natural since the Hamiltonian is a sum of a kinetic operator and a potential,
both real. For the same reasons, we suppose that the dipole moment $\mu$ is real (Hermitian thus symmetric) but we assume moreover that the diagonal elements are null. This additional assumption is motivated both by invariance properties (the diagonal of $H_0$ as matrix commutes with the diagonal of $\mu$ as matrix) but also by the desire to identify an unique pair $(H_0, \mu)$ since in this way the number of unknowns (dimension of $S_\mathbb{R}^d$ plus that of $S_\mathbb{R}^d$) equals the number of equations (the dimension of $U$).

Note that one can easily prove the following conservation property:

$$\forall t \in [0, T], \|U(t)\|_U = \|U_{\text{init}}\|_U,$$

where we have denoted by $\|\cdot\|_U$ the norm associated to the scalar product

$$(A, B) \in U \times U \mapsto tr(A^* B).$$

This problem is related to inverse problems in quantum control [17], but unlike previous works, we do not aim here at designing relevant laser fields to identify the pair $(H_0, \mu)$ but rather to investigate the properties of the fields $\varepsilon(t)$ that make Equation (3) invertible and algorithms to compute numerically the corresponding solution operators $H_0$ and $\mu$.

III. LOCAL CONTROLLABILITY RESULT

In this section, we present some theoretical results about the local inversion of Equation (3). More precisely, we make use of the calculus of variations to obtain a local inversion theorem.

Given a pair $(H_0, \mu)$, we first introduce the tangent space $A_{H_0, \mu}$, which is the space of matrices defined by:

$$A_{H_0, \mu} = \{ M \in \mathbb{C}^{N_d \times N_d}, M^* U(T) + U(T)^* M = 0 \}.\$$

We then consider the differential operator of $\varphi$ defined by:

$$d\varphi(H_0, \mu) : S_\mathbb{R} \times S_\mathbb{R} \to A_{H_0, \mu},$$

$$(\delta H_0, \delta \mu) \mapsto \delta U(T),$$

where $\delta U(T)$ is solution at time $t = T$ of the linearized Schrödinger equation:

$$i \dot{\delta U}(t) = [H_0 + \varepsilon(t) \mu] \delta U(t) + [\delta H_0 + \varepsilon(t) \delta \mu] U(t),$$

and $U(t)$ follows equation (1).

We will prove that $\varphi$ is an onto mapping using the fact that $d\varphi$ also satisfies this property. This strategy is motivated by the following known result:

**Theorem 1:** Supposed that $d\varphi(H_0, \mu)$ is an onto mapping, i.e.

$$\forall V \in A_{H_0, \mu}, \exists (\delta H_0, \delta \mu), d\varphi(H_0, \mu)(\delta H_0, \delta \mu) = V.$$

Then $\varphi$ is locally onto in a neighborhood of $(H_0, \mu)$. We shall prove that $d\varphi$ is an onto mapping on the neighborhood of all states of the form $U_0 := \varphi(H_0, 0) \in U$. To do this, we compute explicitly an inverse mapping.

**Theorem 2:** Given $H_0 \in S_\mathbb{R}$, define $V_0$ as the matrix that diagonalizes $U_0 := \varphi(H_0, 0)$ in the following way:

$$U_0(t) = V_0^* e^{i \Lambda (t - \frac{T}{2})} V_0,$$

with $\Lambda$ the diagonal matrix with coefficients $\lambda_a \in \mathbb{R}, a \in \mathbb{N}_d, 1 \leq a \leq N_d$. Suppose that for $a \neq b, 1 \leq a \leq N_d, 1 \leq b \leq N_d$:

$$\lambda_a \neq \lambda_b \quad (4)$$

$$\varepsilon_{a,b} := \Im \left( \int_0^T \varepsilon(t) e^{i \lambda_{a,b} (t - \frac{T}{2})} dt \right) \neq 0. \quad (5)$$

Then $d\varphi(H_0, 0)$ is an onto mapping and its inverse is given by:

$$\psi : V^* \in A_{H_0, \mu} \mapsto (\delta H_0, \delta \mu).$$

The matrices $\delta H_0$ and $\delta \mu$ are given by:

$$\delta H_0 := V_0^* \delta H_0 V_0, \quad \delta \mu := V_0^* \delta \mu V_0,$$

where the coefficients $h_{a,b}$ and $m_{a,b}$ of the matrices $\delta H_0$ and $\delta \mu$ are given by:

$$\begin{align*}
    m_{a,b} &= \frac{\Re \varepsilon_{a,b}}{\xi_{a,b}}, \\
    h_{a,b} &= \frac{\Re \varepsilon_{a,b}}{\xi_{a,b}} - \frac{\Re \varepsilon_{b,a}}{\xi_{b,a}} = \frac{\sin(\Delta \lambda_{a,b})}{T} \delta \lambda_{a,b} \quad \text{if } a \neq b, \\
    m_{a,a} &= 0, \\
    h_{a,a} &= 2 \frac{T}{\xi_{a,a}} \quad \text{if } a = b.
\end{align*}$$

Here $\varepsilon_{a,b}, a, b \in \mathbb{N}_d, 1 \leq a \leq N_d$ are the coefficients of $i V_0^* U_0(T)^* V_0^* V_0$ and $\xi_{a,b} := \Re \left( \int_0^T \varepsilon(t) e^{i \lambda_{a,b} (t - \frac{T}{2})} dt \right)$.

**Proof:** We fix $V^* \in A_{H_0, \mu}$ and solve

$$d\varphi(H_0, 0)(\delta H_0, \delta \mu) = V^*. \quad (7)$$

First, one can show the identities:

$$\varphi(H_0, \mu)^* d\varphi(H_0, 0)(\delta H_0, \delta \mu) = U_0(T)^* \delta U_0(T) = -i \int_0^T U_0(t)^* (\delta H_0 + \varepsilon(t) \delta \mu) U_0(t) dt, \quad (8)$$

where the variation $\delta U_0$ is defined by the evolution equation:

$$i \delta \dot{U}_0(t) = [H_0 + \varepsilon(t) \mu] \delta U_0(t) + [\delta H_0 + \varepsilon(t) \delta \mu] U_0(t). \quad (9)$$

Note that such an identity holds also when $\mu \neq 0$. Since $U_0(T)^*$ is invertible, showing that (7) has a solution is equivalent to show that

$$\int_0^T U_0(t)^* (\delta H_0 + \varepsilon(t) \delta \mu) U_0(t) dt = V^*, \quad (10)$$

has a solution, with $V := i U_0(T)^* V^* \in S$ since $V^* \in A_{H_0, \mu}$. A nice property of the trajectory $t \mapsto U_0(t)$ is that Equation (10) can be solved explicitly. Indeed, let us denote by $v_{a,b}, h_{a,b}$ and $m_{a,b}$, with $a, b \in \mathbb{N}_d, 1 \leq a, b \leq N_d$, the coefficients of the matrices $V_0 V_0^* V_0^* V_0^* \delta H_0 V_0^* V_0^* \delta \mu V_0^*$.
respectively. Expanding (10) gives rise, in the case \(a \neq b\) to

\[
v_{a,b} = h_{a,b} \int_0^T e^{i(\lambda_a - \lambda_b)(t - \frac{T}{2})} dt + m_{a,b} \int_0^T \varepsilon(t)e^{i(\lambda_a - \lambda_b)(t - \frac{T}{2})} dt
\]

where \(\delta \lambda_{a,b} = \lambda_a - \lambda_b\) and \(\varepsilon(\delta \lambda_{a,b}) = \int_0^T \varepsilon(t)e^{i\delta \lambda_{a,b}(t - \frac{T}{2})} dt = e^{i\delta \lambda_{a,b}^*} \varepsilon(0) + i \delta \lambda_{a,b} \varepsilon(0)\).

In the case \(a = b\), one finds that

\[
v_{a,a} = h_{a,a} \frac{T}{2} + m_{a,a} \varepsilon(0) = h_{a,a} \frac{T}{2} + m_{a,a} \int_0^T \varepsilon(t) dt.
\]

Note that the assumption \(\delta H_0, \delta \mu \in \mathcal{S}_R\) combines with \(\varepsilon(\delta \lambda_{a,b}) = \varepsilon(\delta \lambda_{a,a})\) implies that \(v_{a,b} = \bar{v}_{a,b}\), so that \(V \in \mathcal{S}\).

The result follows.

**Remark 1:** In this theorem, we have defined \(m_{a,a}\) arbitrarily.

This theorem gives a first hint about conditions required to identify \((H_0, \mu)\). Condition (4) is weaker to the standard non-degeneracy condition

\[
\forall (a, b) \neq (a', b'), \quad \lambda_b - \lambda_a \neq \lambda_{b'} - \lambda_{a'},
\]

and is in practice often satisfied. Condition (5) deals with the laser field itself. It is a non-resonant condition to control the system.

### IV. Numerical methods

In this section, we present two algorithms to solve (3). The strategy we follow is a direct adaptation of previous results and proofs: we consider local approximations based on fixed point iterative solvers. In our approach, a crucial step consists in obtaining an appropriate time discretized version of (1). In the first part, we build such an approximation that enables the exact computation of the derivative of the final state \(U(T)\) with respect to \((H_0, \mu)\) and derive from this setting a numerical strategy.

#### A. Time discretization

In order to simulate numerically Equation (1), we introduce the following time discretization: give \(N_T \in \mathbb{N}\), we denote by \(\Delta T = \frac{T}{N_T}\) the time step and for \(n = 0, \cdots, N_T\) by \(U_n\) and \(\varepsilon_n\) the approximations of \(U(n\Delta T)\) and \(\varepsilon(n\Delta T)\). In order to preserve the unitary property of the matrices \(U(t)\) at the discrete level, we use a Crank-Nicholson scheme ruled by the formula:

\[
\frac{U_{n+1} - U_n}{\Delta T} = (H_0 + \varepsilon_n \mu) U_{n+1} + U_n.
\]

The corresponding iteration is then given by:

\[
(Id + L_n)U_{n+1} = (Id - L_n)U_n,
\]

where \(L_n = \frac{i\Delta T}{2}(H_0 + \varepsilon_n \mu)\).

Let us now detail the effect of variations \(\delta H_0, \delta \mu\) in \(H_0\) and \(\mu\) on the sequence \((U_n)_{n=0, \cdots, N_T}\). We have:

\[
(Id + L_n)\delta U_{n+1} + \delta L_n U_{n+1} = (Id - L_n)\delta U_n - \delta L_n U_n,
\]

\[
\delta L_n(U_{n+1} + U_n) = -(Id + L_n)\delta U_{n+1},
\]

\[
(U_{n+1} + U_n)^* \delta L_n(U_{n+1} + U_n) = -2(U_{n+1}^* \delta U_{n+1} - U_n \delta U_n).
\]

where \(\delta L_n = \frac{i\Delta T}{2}(\delta H_0 + \varepsilon_n \delta \mu)\). This finally gives rise to:

\[
U_{n+1}^* \delta U_{n+1} + U_n \delta U_{n+1} = -i \Delta T \frac{(U_{n+1} + U_n)^* (\delta H_0 + \varepsilon_n \delta \mu) U_{n+1} + U_n}{2}.
\]

Since the initial value is fixed, we obtain:

\[
U_{N_T}^* \delta U_{N_T} = -i \Delta T \sum_{n=0}^{N_T-1} \left( \frac{(U_{n+1} + U_n)^* (\delta H_0 + \varepsilon_n \delta \mu) U_{n+1} + U_n}{2} \right).
\]

This result can be seen as a discretized version of (8) where \(\mu\) is not necessarily null. We insist on the fact that such a result is specific to the Crank-Nicholson discretization. As far as we know, no other numerical solvers give rise to discretization of (8) where the variations \(\delta H_0\) and \(\delta \mu\) are explicit.

#### B. Fixed points methods

We now present some iterative solvers to compute solutions of (3).

1) **A Newton Method:** In the discrete setting, we still denote by \(\varphi\) the operator:

\[
\varphi : \mathcal{S}_R \times \mathcal{S}_R^0 \rightarrow \mathcal{U} \quad (H_0, \mu) \mapsto U_{N_T}.
\]

To solve the equation \(\varphi(H_0, \mu) = U_{\text{target}}\), a Newton method would consist in the following iteration:

\[
d\varphi(H_0^k, \mu^k) \cdot (\delta H_0^k, \delta \mu^k) = - \left( \varphi(H_0^k, \mu^k) - U_{\text{target}} \right),
\]

where \(k\) is the iteration index, \(\delta H_0^k = H_0^{k+1} - H_0^k, \delta \mu^k = \mu^{k+1} - \mu^k\).

In our case, (12) reads:

\[
dU_{N_T}^k = U_{\text{target}} - U_{N_T}^k.
\]

Using (11), one can rewrite this equation as follows:

\[
\Delta T \sum_{n=0}^{N_T-1} \left( \frac{(U_{n+1} + U_n)^* (\delta H_0^k + \varepsilon_n \delta \mu^k) U_{n+1} + U_n}{2} \right) = i (U_{N_T}^k)^* U_{\text{target}} - Id,
\]

where we recall that the unknowns are \(\delta H_0^k\) and \(\delta \mu^k\). This equation has generally no solutions, since its left hand side belongs to \(\mathcal{S}\) what is not the case for its right hand side. To solve this problem, we replace \(i ((U_{N_T}^k)^* U_{\text{target}} - Id)\) by
a first order approximation \( S^k \in S_\mathbb{R} \). Two possible choices are:

\[
\exp(-iS^k) := (U^k_{N_T})^* U_{\text{target}} \quad (13)
\]

\[
S^k := i \frac{(U^k_{N_T})^* U_{\text{target}} - U_{\text{target}} U^k_{N_T}}{2} \quad (14)
\]

In the numerical tests, the same behavior is observed when using the first or the second definition.

**Remark 2:** The previous method can be simplified to obtain a procedure where no matrix needs to be assembled and the inverted during iterations. Instead of updating at each iteration in the pair \((H_0, \mu)\) in the term \(d\varphi(H_0, \mu)\) of Formula (12), one can keep a constant approximation \((H^\text{ref}_0, \mu^\text{ref})\) of the solution. We denote by \((U^\text{ref}_n)_{n=0, \ldots, N_T}\) the corresponding sequence of states. The iteration then reads:

\[
\Delta T \sum_{n=0}^{N_T-1} \frac{(U^\text{ref}_{n+1} + U^\text{ref}_n)^*}{2} (\delta H^k_0 + \varepsilon_n \delta \mu^k) \frac{U^\text{ref}_{n+1} + U^\text{ref}_n}{2} = S^k,
\]

where \(S^k\) is defined in the previous section, see (13) and (14). Note that such an algorithm is actually a time-discretized version of the fixed point used in the proof of Theorem 2, except that here \(\mu\) is not supposed to be null.

2) **Implementation of the iterative solvers:** Both previous methods require inversions of linear systems which are not given explicitly in our formulations. To fill in this gap, we explain here how to assemble the matrices, i.e. to rewrite the equation

\[
\Delta T \sum_{n=0}^{N_T-1} \frac{(U^\text{ref}_{n+1} + U^\text{ref}_n)^*}{2} (\delta H^k_0 + \varepsilon_n \delta \mu^k) \frac{U^\text{ref}_{n+1} + U^\text{ref}_n}{2} = S,
\]

in terms of linear system. In what follows, we denote by \(X_M\), the vector representation of a matrix \(M\) consisting in concatenating vertically its columns. A first step to do this is to note that the following expression reads as follows:

\[
\Delta T \left( \sum_{n=0}^{N_T-1} M_{U_{n+1/2}} \right) X_{\delta H_0} + \Delta T \left( \sum_{n=0}^{N_T-1} \varepsilon_n M_{U_{n+1/2}} \right) X_{\delta \mu} = X_S, \quad (15)
\]

with

\[
M_{U_{n+1/2}} = \text{kron}(1_{N_d}, U^{*}_{n+1/2}) \times \text{kron}(U^T_{n+1/2}, 1_{N_d}).
\]

Here, \(\text{kron}\) denotes the Kronecker product, \(U_{n+1/2} = U_{n+1/2} = U^*_{n+1/2} U^{T}_{n+1/2}\), the term by term product of two matrices \(A\) and \(B\) is denoted by \(A \times B\) and \(1_{N_d}\) denotes the matrix of \(\mathbb{R}^{N_d \times N_d}\) whose coefficients are equal to 1.

A second step must then be carried out: since the matrices \(\delta H_0\) and \(\delta \mu\) are symmetric, one has to consider the columns of the matrices in (15) that correspond to the coefficients of \(\delta H_0\) located, e.g., above the diagonal and the coefficients of \(\delta \mu\) located strictly above the diagonal. In the same way, only the lines of the resulting system that correspond to the coefficients located above the diagonal of \(S\) shall be considered.

Taking the real and the imaginary part of the equations, the resulting system is of size \(N_d^4\).

**C. A continuation method for global controllability**

The algorithms proposed in Section IV-B are only locally convergent. The purpose of this section is to present a continuation method that enables to extend their range of application.

As mentioned above, numerous methods exist to solve the control problem where the laser term \(\varepsilon\) in Equation (1) is unknown and \(H_0\) and \(\mu\) are given [15], [11], [10]. Based on this fact, the method we propose is the following. Given an initial guess \((H^0_0, \mu^0)\), find a control \(\varepsilon^0\) such that \(U^0_{N_T}\), the final state associated to \((H^0_0, \mu^0)\) correctly approximates \(U_{\text{target}}\). Given \(\theta \in [0, 1]\), we define the interpolated fields \(\varepsilon^\theta = (1-\theta)\varepsilon + \theta\varepsilon\). A fixed point method as the one presented in Section IV-B can then be applied with \((H^\theta_0, \mu^\theta)\) as an initial guess to solve the operator control problem with \(\varepsilon^\theta\). Our algorithm consists in repeating this procedure by solving iteratively the operator control problem associated to the field \(\varepsilon^{k+\theta}\) using \((H^{k-1}_0, \mu^{k-1})\) as initial guess. Carrying this procedure up to \(\theta = 1\) enables to solve the original problem.

**V. Numerical results**

In this last section, we present some numerical results obtained with the algorithms of the previous sections. As a laser term in Equation (1), we use \(\varepsilon(t) = \sin(t)\). The other numerical data are \(N_d = 5, T_0 = 10, N_T = 10^2, T = 2\pi T_0\) and \(\Delta T = T/N\).

**A. Newton Method**

We first test our Newton method. In this way, we choose randomly a pair \((H_0, \mu)\), with coefficients in \([-1, 1]\) and compute the corresponding final state \(U_{N_T}\). Then, we start the Newton procedure with an initialization \((H_0 + \Delta H_0, \mu + \Delta \mu)\) where \((\Delta H_0, \Delta \mu)\) are also chosen randomly. An example of computation is given in the next table.

| Iteration | \(\log_{10}||H^k_0 - H_0||_\mu\) | \(\log_{10}||\mu^k - \mu||_\mu\) |
|----------|---------------------------------|---------------------------------|
| 1        | -1.119029                       | -1.155837                       |
| 2        | -3.003599                       | -2.850268                       |
| 3        | -4.329497                       | -4.122328                       |
| 4        | -8.234980                       | -8.179398                       |
| 5        | -13.963299                      | -14.029020                      |
| 6        | -14.022486                      | -14.131066                      |

Here, we refine a pair \((H_0, \mu)\) starting from a 10% random perturbation. We see that the numerical convergence is obtained after 6 iterations. Note also that the quadratic convergence is observed.

**B. Continuation method**

In a second test, we use the continuation method presented in IV-C to tackle a problem where the algorithms of Section IV-B do not apply. Given a target \(U_{\text{target}}\) obtained with the field \(\varepsilon\) and a pair \((H_0, \mu)\) that is chosen randomly, we look for the operators \(H'_0\) and \(\mu'\) that solve the control
problem associated to the field $\cos(3t)$ and the target $U_{\text{target}}$.

The direct use of the Newton method of Section IV-B does not work: in this case, the algorithm does not converge. The continuation method enables to solve this problem. Using $\delta\theta = 1/4$, and 10 iterations of the Newton method as inner loop, a relevant pair $(H_0', \mu')$ is obtained.

This example has been reproduced for numerous random initial pairs $(H_0, \mu)$.

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