Monotonically convergent algorithms for locally constraint quantum controls

Gabriel Turinici

MONOTONICALLY CONVERGENT ALGORITHMS FOR LOCALLY CONSTRAINT QUANTUM CONTROLS

Gabriel TURINICI
INRIA Rocquencourt, Domaine de Voluceau
Rocquencourt - B.P. 105, 78153
Le Chesnay Cedex - France
and
CERMICS-ENPC, 6 et 8 avenue Blaise Pascal
Cité Descartes - Champs sur Marne,
77455 Marne la Vallée Cedex 2, France
Gabriel.Turinici@inria.fr

ABSTRACT
The problem of finding the optimal control in numerical computer simulations of quantum control phenomena is usually addressed through the introduction of monotonically convergent algorithms that are guaranteed to improve the cost functional at each step. A recent extension of these algorithms implements a search for a control with given bounds. Within this context, this paper will present a generalisation of this algorithm to a more general class of local in time constraints. Numerical results that illustrate some of the properties of the algorithms introduced are given.

KEY WORDS
quantum control, bilinear control, monotonic algorithms, numerical methods, non-linear control

1 Introduction

Armed with the successful demonstration of quantum control both in theoretical[5, 10, 4, 11] and experimental[1] environements, the challenges of this research area aim now towards a deeper understanding of the phenomena at work. Despite the fact that perfect simulation of practical-sized systems remain an unatainable goal (due to the difficulty to solve in multiple dimensions the Schrödinger equation) the numerical computer simulations are priviledged tools in this quest to shed some light on the experimetal findings. Among the qualities required of the numerical simulations in order to become as close to reality as possible we can identify the need for findind suitably **constraint** controls, either in a frequency/Fourier domain or in time domain. In this paper we will provide a monotonic algorithm that allows to find a quantum control when restrictions on the admissible values (of the controlling field) are imposed. We will study two cases: bounded (continuous) controls and discrete valued controls. While the importance of the former type of restriction is evident from the experimental point of view, the relevance of the latter choice, i.e. discrete-valued controls, has only been recently[8, 9] seen to lead to numerically efficient implementations. The tools presented below can thus serve as alternatives to constraint or combinatorial optimization.

2 Dynamical Equations

Denote by H_0 the internal Hamiltonian of the system under study and let $\psi_0(x)$ be its initial state, ($\|\psi_0\|_{L^2(\mathbf{R}^\gamma)} = 1$) where x denotes the relevant coordinate variables. The time-dependent Schrödinger equation allows to express the state $\psi(x,t)$ at any time t as the solution of the following evolution problem

$$\begin{cases} i\hbar\frac{\partial}{\partial t}\psi(x,t) = H_0\psi(x,t) \\ \psi(x,t=0) = \psi_0(x), \end{cases} \tag{1}$$

If the isolated quantum systems does not follow a suitable dynamics its evolution can be *controlled* by an external interaction, e.g. a laser, taken here as an electric field modeled by a coupling operator given by an amplitude $\epsilon(t) \in \mathbf{R}$ (at our choice) and a time independent dipole moment operator μ. Denote by $H = H_0 - \epsilon(t)\mu$; the controlled system obeys now the equations

$$\begin{cases} i\hbar\frac{\partial}{\partial t}\psi(x,t) = (H_0 - \epsilon(t)\mu)\psi(x,t) \\ \psi(x,t=0) = \psi_0(x) \end{cases} \tag{2}$$

Within the **optimal control** approach the realization of the goal is expressed through the introduction of a cost functional J to be optimized; this cost functional depends on the final (and possible any intermediary) state $\psi(t)$ of the system and of the control $\epsilon(t)$. In addition this cost functional may include terms that penalize undesired effects. One simple example of cost functional is

$$J(\epsilon) = < \psi(T)|O|\psi(T) > -\alpha\int_0^T \epsilon^2(t)dt \tag{3}$$

where O is the observable operator that encodes the goal. Note that the term $\int_0^T \epsilon^2(t)dt$ is a global penalization term that prevents from using a too large laser fluence. Its action is weighted by the parameter $\alpha > 0$: a small α will emphasize the control objective while a large α will rather tend to

economize on the laser energy and only secondary to reach the target. Each α will therefore give rise to a possibly different optimizer of J that strike a balance, weighted by α, between using a not too expensive laser fluence while simultaneously ensuring the desired observable has an acceptable (large) value. In addition to these *global constraints* the control may also be required to fulfill another *local constraints* that may arise from physical or technological considerations. The following general framework will be addressed in this paper:

$$\epsilon(t) \in D(t) \tag{4}$$

for predefined, possibly time dependent, admissible sets $D(t)$. Two remarkable particular cases can be identified

$$L^\infty constraints : |\epsilon(t)| \leq M \; for \; a \; given \; M \geq 0 \tag{5}$$

$$discrete \; valued \; constraints : \; \epsilon(t) \in \{e_1, ..., e_K\} \tag{6}$$

What is of interest for us here is to give efficient algorithms that allow for optimizing the cost functional J within the admissible class of controls.

Remark 2.1 *Throughout this paper only local in time constraints will be enforced. Although this includes many interesting cases, a separate question is how to enforce global constraints on the controlling field such as frequency/Fourier constraints. Very few works in this area are available to day, we refer the reader to [3] for recent approaches to deal with this setting.*

In the general (non-constraint) case the maximization of the cost functional $J(\epsilon)$ (under the constraint of the Schrödinger equation (2) can be realized by writing down the Euler-Lagrange critical point equations. Introduce an *adjoint state* $\chi(t,x)$ (used as a Lagrange multiplicator); the following equations are thus obtained [10]:

$$\begin{cases} i\hbar\frac{\partial}{\partial t}\psi(x,t) = (H_0 - \epsilon(t)\mu)\psi(x,t) \\ \psi(x,t=0) = \psi_0(x) \end{cases} \tag{7}$$

$$\begin{cases} i\hbar\frac{\partial}{\partial t}\chi(x,t) = (H_0 - \epsilon(t)\mu)\chi(x,t) \\ \chi(x,t=T) = O\psi(x,T) \end{cases} \tag{8}$$

$$\alpha\epsilon(t) = -\mathrm{Im} < \chi(t)|\mu|\psi(t) > \tag{9}$$

One efficient choice for solving in practice the critical point equations (7)-(9) is the monotonically convergent iterative algorithm[2, 6, 10] that can be described by the resolution of the following equations at step k:

$$\begin{cases} i\frac{\partial}{\partial t}\psi^k(x,t) = (H_0 - \epsilon^k(t)\mu)\psi^k(x,t) \\ \psi^k(x,t=0) = \psi_0(x) \end{cases} \tag{10}$$

$$\epsilon^k(t) = (1-\delta)\tilde{\epsilon}^{k-1}(t) - \frac{\delta}{\alpha}\mathrm{Im}\langle\chi^{k-1}|\mu|\psi^k\rangle(t) \tag{11}$$

$$\begin{cases} i\frac{\partial}{\partial t}\chi^k(x,t) = (H_0 - \tilde{\epsilon}^k(t)\mu)\chi^k(x,t) \\ \chi^k(x,t=T) = O\psi^k(x,T) \end{cases} \tag{12}$$

$$\tilde{\epsilon}^k(t) = (1-\eta)\epsilon^k(t) - \frac{\eta}{\alpha}\mathrm{Im}\langle\chi^k|\mu|\psi^k\rangle(t) \tag{13}$$

The most important property of this algorithm is given in the following

Theorem 2.1 *Suppose O is a self-adjoint positive semi-definite observable. Then, for any $\eta, \delta \in [0,2]$ the algorithm given in Eqns. (10)-(13) converges monotonically in the sense that $J(\epsilon^{k+1}) \geq J(\epsilon^k)$.*

Remark 2.2 *In practice the monotonicity property is relevant because it often helps providing good quality controls with only very few resolutions of the time-dependent Schrödinger equation (which is not the case of the gradient or conjugated gradient algorithms).*

The purpose of this paper is to show how the algorithm in Eqns. (10)-(13) has to be adapted in order to treat the constraints in Eqn. (4).

3 Monotonic algorithms for bounded controls

When dealing with a restricted class of controls one can take one of the two alternative approaches:

1/ either to write down the critical point equations for the minimization of the cost functional within the admissible class of controls and then solve these equations

or

2/ to directly minimize the cost functional by taking care to always remain within the admissible control set. Note that when (and if) this method converges then the final solution will be a critical point of the cost functional as obtained by the first alternative.

We will choose in the following the second approach and only use the critical point equations in Eqns. (7)-(9) as a starting point to construct algorihms.

Let us remark that for any set $D(t)$ a function $f_t : D(t) \times \mathbf{R} \to D(t)$ can be constructed such that

$$|f_t(z,x) - x| \leq |z - x|, \forall z \in D(t), x \in \mathbf{R} \tag{14}$$

$$|f_t(z,x) - x| < |z - x|, \forall x \in \mathbf{R}, \forall z \in D(t)$$

$$such \; that \; D(t) \cap \{y \in \mathbf{R}; |y - x| < |x - z|\} \neq \emptyset. \tag{15}$$

Indeed, we can set for instance $f_t(z,x)$ to any element of $D(t) \cap \{y \in \mathbf{R}; |y-x| < |x-z|\}$ when this set is not void and $f_t(z,x) = z$ otherwise.

This remark allows to state the following

Theorem 3.1 *Suppose O is a self-adjoint positive semi-definite observable. Then, if for any t f_t satisfies Eq.(14) the algorithm*

$$\begin{cases} i\frac{\partial}{\partial t}\psi^k(x,t) = (H_0 - \epsilon^k(t)\mu)\psi^k(x,t) \\ \psi^k(x,t=0) = \psi_0(x) \end{cases} \tag{16}$$

$$\epsilon^k(t) = f_t\left(\epsilon^{k-1}(t), \frac{1}{\alpha}\mathrm{Re}\left\langle\frac{-\mu}{i}\psi^k(t), \chi^{k-1}(t)\right\rangle\right) \tag{17}$$

$$\begin{cases} i\frac{\partial}{\partial t}\chi^k(x,t) = (H_0 - \epsilon^k(t)\mu)\chi^k(x,t) \\ \chi^k(x,t=T) = O\psi^k(x,T) \end{cases} \tag{18}$$

converges monotonically in the sense that $J(\epsilon^k) \geq J(\epsilon^{k-1})$ (and is such that $\epsilon^k(t) \in D(t), \forall t \leq T$, $k \geq 1$).

Proof We will use the following formula (see [7] for detailed computations):

$$J(\epsilon^k) - J(\epsilon^{k-1}) = \left\langle \psi^k(T) - \psi^{k-1}(T) | O | \psi^k(T) - \psi^{k-1}(T) \right\rangle + \alpha \int_0^T \epsilon^{k-1}(t)^2 dt - \alpha \int_0^T \epsilon^k(t)^2 dt + 2\mathrm{Re} \int_0^T (\epsilon^k - \epsilon^{k-1}) \left\langle \frac{-\mu}{i} \psi^k(t), \chi^{k-1}(t) \right\rangle \quad (19)$$

and thus with the notation

$$\eta^k(t) = \frac{1}{\alpha} \mathrm{Re} \left\langle \frac{-\mu}{i} \psi^k(t), \chi^{k-1}(t) \right\rangle$$

$$J(\epsilon^k) - J(\epsilon^{k-1}) = \left\langle \psi^k(T) - \psi^{k-1}(T) | O | \psi^k(T) - \psi^{k-1}(T) \right\rangle + \alpha \int_0^T \left(\epsilon^{k-1}(t) - \eta^k(t)\right)^2 dt - \alpha \int_0^T \left(f_t(\epsilon^{k-1}(t), \eta^k(t)) - \eta^k(t)\right)^2 dt \quad (20)$$

and the conclusion of the theorem follows from the properties of the function f_t.

Remark 3.1 *When $\eta = 0$ the algorithm in Eqns. (10)-(13) is a particular case of the algorithm in Eqns. (16)-(18) for $D(t) = \mathbf{R}$, $f_t(z, x) = (1 - \delta)z + \delta x \; \forall t$.*

Using Thm. 3.1 one can derive many classes of monotonically convergent algorithms for particular choices of target sets $D(t)$. Consider for instance the set $D(t)$ as in Eqn. (6) where we suppose the ordering $e_1 < e_2 < ... < e_K$. This situation has been recently introduced in [8, 9] in connection with fast algorithms to solve the time-dependent Schrödinger equation in the quantum control framework. Define the function f by

$$f(z, x) = \begin{cases} e_1 & \text{if } x \leq \frac{e_1 + e_2}{2} \\ ... & ... \\ e_k & \text{if } \frac{e_{k-1} + e_k}{2} < x \leq \frac{e_k + e_{k+1}}{2} \\ ... & ... \\ e_K & \text{if } x > \frac{e_{K-1} + e_K}{2} \end{cases} \quad (21)$$

Let us state the result that can be derived in this case

Theorem 3.2 *Suppose O is a self-adjoint positive semi-definite observable. Then, the algorithm*

$$\begin{cases} i\frac{\partial}{\partial t}\psi^k(x,t) = (H_0 - \epsilon^k(t)\mu)\psi^k(x,t) \\ \psi^k(x, t=0) = \psi_0(x) \end{cases} \quad (22)$$

$$\epsilon^k(t) = f\left(\epsilon^{k-1}(t), \frac{1}{\alpha}\mathrm{Re}\left\langle \frac{-\mu}{i}\psi^k(t), \chi^{k-1}(t)\right\rangle\right) \quad (23)$$

$$\begin{cases} i\frac{\partial}{\partial t}\chi^k(x,t) = (H_0 - \epsilon^k(t)\mu)\chi^k(x,t) \\ \chi^k(x, t=T) = O\psi^k(x,T) \end{cases} \quad (24)$$

converges monotonically in the sense that $J(\epsilon^k) \geq J(\epsilon^{k-1})$ within the class of controls ϵ^k that take only one of the predefined set of values $e_1, ..., e_K$.

An interesting situation arises when optimizing over bounded class of controls (as in Eqns. (5) and (6)): as bound on the L^2 norm of the control ϵ becomes available, e.g. for Eqn. (6): $\|\epsilon(t)\|_{L^2(O,T)} \leq M \cdot \sqrt{T}$. This bound can thus eliminate the need to penalize the term $\int_0^T \epsilon(t)^2$ in the cost functional (this is equivalent to setting α to 0 in the definition of the cost functional). The Thm. 3.1 can be extended to this case too:

Theorem 3.3 *Suppose O is a self-adjoint positive semi-definite observable. Define*

$$J_0(\epsilon) = < \psi(T) | O | \psi(T) >$$

and let for any $t < T$ $g_t : D(t) \times \mathbf{R} \to D(t)$ be such that

$$x(g_t(z, x) - z) \geq 0 \quad (25)$$

for any $z \in D(t)$, $x \in \mathbf{R}$.

Then, the algorithm

$$\begin{cases} i\frac{\partial}{\partial t}\psi^k(x,t) = (H_0 - \epsilon^k(t)\mu)\psi^k(x,t) \\ \psi^k(x, t=0) = \psi_0(x) \end{cases} \quad (26)$$

$$\epsilon^k(t) = g_t\left(\epsilon^{k-1}(t), \mathrm{Re}\left\langle \frac{-\mu}{i}\psi^k(t), \chi^{k-1}(t)\right\rangle\right) \quad (27)$$

$$\begin{cases} i\frac{\partial}{\partial t}\chi^k(x,t) = (H_0 - \epsilon^k(t)\mu)\chi^k(x,t) \\ \chi^k(x, t=T) = O\psi^k(x,T) \end{cases} \quad (28)$$

converges monotonically in the sense that $J_0(\epsilon^k) \geq J_0(\epsilon^{k-1})$ (and is such that $\epsilon^k(t) \in D(t), \forall t \leq T$).

Proof The proof is similar to the proof of Thm. 3.1.

Note finally that for any $D(t)$ a function g_t exists that satisfies Eqn. (25).

3.1 Discussion on critical points

A legitimate question that arises in connection with the conclusions of the Thms. 3.1, 3.2 and 3.3 concerns the properties of the converged solutions. More precisely, since the increase in the cost functional is not guaranteed to be strictly positive at each iteration, one can ask whether $J(\epsilon^k) = J(\epsilon^{k-1})$ implies that ϵ^k is a local maximizer (or local critical point) of J. In general the answer to this question highly depends on the properties of the sets $D(t)$ since the definition of a "critical point" or "local optimizer" depends on the structure of $D(t)$. In order to give an example of analysis that can be developped to answer this question we will take the situation in Thm. 3.3 for the constraints of Eqn. (5). Denote by $\nabla_\epsilon J_0(t) = \mathrm{Re}\left\langle \frac{-\mu}{i}\psi(t), \chi(t)\right\rangle$. In this case the definition of a critical point ϵ can be expressed as: ϵ is critical point for J_0 iff for any time t:

$$\begin{cases} \text{if } e(t) = -M & \text{then } \nabla_\epsilon J_0(t) \leq 0 \\ \text{if } -M < e(t) < M & \text{then } \nabla_\epsilon J_0(t) = 0 \\ \text{if } e(t) = M & \text{then } \nabla_\epsilon J_0(t) \geq 0 \end{cases} \quad (29)$$

On the other hand, it can be proven by the same computation as in Thms. 3.3 that $J(\epsilon^k) = J(\epsilon^{k-1})$ implies

$$\tilde{\eta}^k(t)\left[g_t(\epsilon^{k-1}(t), \tilde{\eta}^k(t)) - \epsilon^{k-1}(t)\right] = 0, \forall t \geq 0,$$

where $\tilde{\eta}^k(t) = \mathrm{Re}\left\langle \frac{-\mu}{i}\psi^k(t), \chi^{k-1}(t)\right\rangle$ Or, choosing as function g_t:

$$g_t(z,x) = \begin{cases} \frac{z+M}{2} & \text{if } x > 0 \\ z & \text{if } x = 0 \\ \frac{z-M}{2} & \text{if } x < 0 \end{cases}$$

we obtain that $J(\epsilon^k) = J(\epsilon^{k-1})$ implies first $\epsilon^k = \epsilon^{k-1}$ and that $\epsilon = \epsilon^{k-1}$ satisfies Eqn. (29); thus ϵ^{k-1} is a critical point of J_0.

4 Numerical results

In order to test the algorithms presented above we have performed some quantum control calculations on the case of the $O - H$ bond that vibrates in a Morse type potential. We refer the reader to [10] for the numerical details concerning this system. The goal is to localize the wave packet at a given location x' at the final time $T = 131000 a.u. \simeq 3.16 ps$; this is expressed via the observable $O(x) = \frac{\gamma_0}{\sqrt{\pi}} e^{-\gamma_0^2 (x-x')^2}$ ($x' = 2.5$) through the requirement that $\langle \Psi | O | \Psi \rangle$ is maximized. The initial state $\psi(0)$ is the ground state of the internal Hamiltonian H_0.

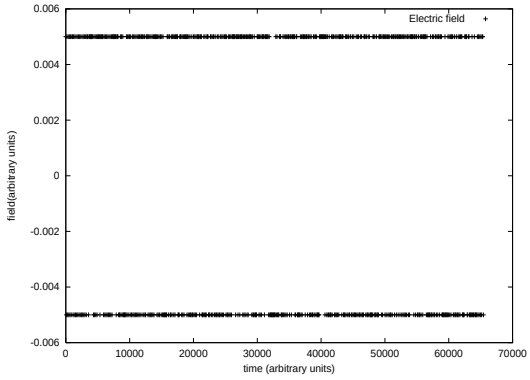

Figure 1. Example of optimal electric field found with the algorithm given in Eqns. (26)-(28) ; as required the values of the control field $\epsilon(t)$ are all either $-M$ or M ($M = 0.005$).

Numerical results are presented in the Figures 1, 2 and 3. As a particular case of both Eqn. (5) and (6) the values of the control field $\epsilon(t)$ are required to be all either $-M$ or M. The algorithm in Eqns. (26)-(28) is used for g_t defined by

$$g(z,x) = \begin{cases} -M & \text{if } x \leq 0 \\ M & \text{if } x > 0 \end{cases}$$

The field succeeds into controlling the system as is demonstrated in Figure 3 where the monotonic behaviour is displayed.

5 Conclusion

Within the framework of local in time constraints for quantum controls this paper proposes several monotonically convergent generic algorithms that at all times remain within the admissible control set. Numerical results on particular cases are presented that demonstrate the monotonicity property and the good quality of the controlling fields after only very few iterations.

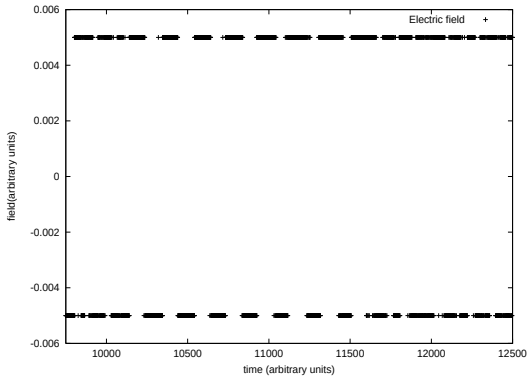

Figure 2. Zoom of the optimal electric field in Figure 1. The optimal field acts through many tailored oscillations between the two admissible values.

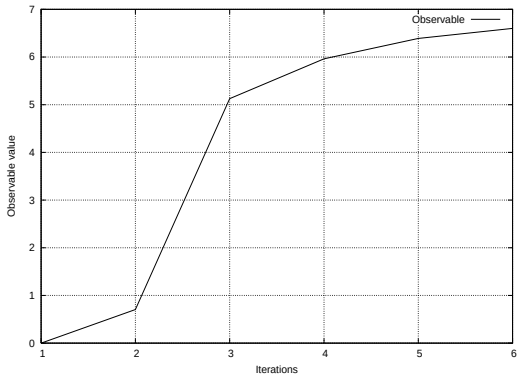

Figure 3. The convergence of the observable $\langle \Psi(T) | O | \Psi(T) \rangle$ with respect to the number of iterations of the algorithm in Eqns. (26)-(28); monotonic behaviour is obtained in agreement with the Thm. 3.3.

Acknowledgements. It is a pleasure to acknowledge helpful discussions that we had on this topic with Prof. H. Rabitz from the Princeton University.

References

[1] A. Assion et al. Control of chemical reactions by feedback-optimized phase-shaped femtosecond laser pulses. *Science*, 282, 1998, 919–922.

[2] Y. Maday and G. Turinici New formulations of monotonically convergent quantum control algorithms, *J. Chem. Phys.* 118, 2003, 8191–8196.

[3] Y. Ohtsuki and H. Rabitz, *Development of Solution Algorithms for Quantum Optimal Control Equations*

in Product Spaces, in A. Bandrauk, M.C. Delfour, and C. Le Bris, Eds. Quantum Control: mathematical and numerical challenges, CRM Proc. Lect. Notes Ser., AMS Publications, Providence, R.I. 2003.

[4] Herschel Rabitz, Gabriel Turinici, and Eric Brown. Control of quantum dynamics: Concepts, procedures and future prospects. In Ph. G. Ciarlet, editor, *Computational Chemistry, Special Volume (C. Le Bris Editor) of Handbook of Numerical Analysis, vol X*, pages 833–887. Elsevier Science B.V., 2003.

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

[6] D. Tannor, V. Kazakov, and V. Orlov in *Time Dependent Quantum Molecular Dynamics* J. Broeckhove J. L. Lathouwers L., Eds., Plenum, 1992, 347–360.

[7] G. Turinici, Monotonically convergent algorithms for bounded quantum controls *Prooceedings of the LHMNLC03 IFAC conference, Sevilla 3-5 April, 2003*.

[8] F. Yip, D. Mazziotti and H. Rabitz, A Propagation Toolkit to Design Quantum Controls, *J. Chem. Phys*, 118, 2003, 8168-8172.

[9] F. Yip, D. Mazziotti and H. Rabitz, A Local-Time Algorithm for Achieving Quantum Control, *J. Phys. Chem. A*, 107, 2003, 7264-7268.

[10] W. Zhu and H.Rabitz, A rapid monitonically convergent iteration algorithm for quantum optimal control over the expectation value of a positive definite operator, *J. Chem. Phys.*, 109(2), 1998, 385-391.

[11] Yukiyoshi Ohtsuki, Gabriel Turinici, and Herschel Rabitz. Generalized monotonically convergent algorithms for solving quantum optimal control problems. *J. Chem. Phys.*, 120(12):5509–5517, 2004.