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MONOTONICALLY CONVERGENT ALGORITHMS FOR BOUNDED QUANTUM CONTROLS

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Abstract: Most of the numerical simulations in quantum (bilinear) control have used one of the monotonically convergent algorithms of Krotov (introduced by Tannor *et al.* (Tannor *et al.*, 1992)) or of Zhu & Rabitz (Zhu and Rabitz, 1998). Recently (Maday and Turinici, 2002), new schemes have been designed that enlarge the class of monotonic algorithms. Within this context, this paper presents a new algorithm that implements a search for a bounded control with given bounds. Numerical results that illustrate some of the properties of the algorithms introduced are given.

Keywords: quantum control, monotonically convergent algorithms, bang-bang control, bounded control

1. INTRODUCTION.

Laser control of complex molecular and solid-state systems is becoming feasible, especially since the introduction (Judson and Rabitz, 1992) of closed loop laboratory learning techniques and their successful implementation (Levis *et al.*, 2001; Assion *et al.*, 1998; Bergt *et al.*, 1999; Weinacht *et al.*, 1999; Bardeen *et al.*, 1997; Bardeen *et al.*, 1998). On the other hand, at the level of the numerical simulations, the introduction of the monotonically convergent algorithms of the Zhu & Rabitz (Zhu and Rabitz, 1998) that extends an algorithm due to Krotov (Tannor *et al.*, 1992) has marked a considerable progress and made possible further investigation in this area. Recently, a new class of monotonically convergent algorithms has been proposed (Maday and Turinici, 2002). In

the continuation of this work, we present here a new algorithm that, in addition to conserving the monotonicity properties mentioned above allows for finding controls within a-priori given lower and upper bounds. Indeed, such controls can help understand the practical relationships that exists between the intensity of the control field and the quality of the control; we refer to (Ramakrishna *et al.*, 2002; Ramakrishna *et al.*, 2000a; Ramakrishna *et al.*, 2000b; Ramakrishna, 2001; Schirmer *et al.*, 2002) for alternative strategies that use constructive procedures and that could also be instrumental to obtaining information of the same nature.

The balance of the paper is as follows: the necessary background and definitions of the quantum control settings are given in the Section 2;

the formulation of the monotonically convergent algorithms is presented in Section 3 followed by relevant numerical results in Section 4. A discussion and some concluding remarks are presented in Section 5.

2. QUANTUM CONTROL SETTING AND OPTIMAL CONTROL EQUATIONS

Consider a quantum system with internal Hamiltonian H_0 prepared in the initial state $\Psi_0(x)$ where x denotes the relevant spatial coordinates; the state $\Psi(x, t)$ at time t satisfies the time-dependent Schrödinger equation (we set $\hbar = 1$)

$$\begin{aligned} i\frac{\partial}{\partial t}\Psi(x, t) &= H_0\Psi(x, t) \\ \Psi(x, t=0) &= \Psi_0(x). \end{aligned} \quad (1)$$

In the presence of an external interaction taken here as an electric field modeled by a coupling operator with amplitude $\epsilon(t) \in \mathbb{R}$ and a time independent dipole moment operator μ , the new Hamiltonian $H = H_0 - \epsilon(t)\mu$ gives rise to the dynamical equations to be controlled:

$$\begin{cases} i\frac{\partial}{\partial t}\Psi(x, t) = (H_0 - \epsilon(t)\mu)\Psi(x, t) \\ \Psi(x, t=0) = \Psi_0(x) \end{cases} \quad (2)$$

The **optimal control** approach allows to assess the fitness of the final state $\Psi(T)$ to a prescribed goal. This is achieved through the introduction of a cost functional J to be maximized; this cost functional includes on one hand terms that describe how well the objectives have been met and on the other hand terms that penalize undesired effects. One simple example of cost functional is

$$J(\epsilon) = \langle \Psi(T) | O | \Psi(T) \rangle - \alpha \int_0^T \epsilon^2(t) dt \quad (3)$$

where $\alpha > 0$ is a parameter (it may also depend of time cf. (Hornung *et al.*, 2001)) and O is the observable operator that encodes the goal: larger the value $\langle \Psi(T) | O | \Psi(T) \rangle$ better the control objectives have been met; note that, in general, attaining the maximal possible value of $\langle \Psi(T) | O | \Psi(T) \rangle$ is at the price of a large laser fluence $\int_0^T \epsilon^2(t) dt$; the optimum evolution will therefore strike a balance between using a not too expensive laser fluence while simultaneously ensuring the desired observable has an acceptable (large) value.

The maximization of the cost functional $J(\epsilon)$ is realized by solving the Euler-Lagrange critical point equations; a standard way to write these equations is to introduce an *adjoint state* $\chi(x, t)$ (used as a Lagrange multiplier). The following critical point equations are thus obtained (Zhu and Rabitz, 1998):

$$\begin{cases} i\frac{\partial}{\partial t}\Psi(x, t) = (H_0 - \epsilon(t)\mu)\Psi(x, t) \\ \Psi(x, t=0) = \Psi_0(x) \end{cases} \quad (4)$$

$$\begin{cases} i\frac{\partial}{\partial t}\chi(x, t) = (H_0 - \epsilon(t)\mu)\chi(x, t) \\ \chi(x, t=T) = O\Psi(x, T) \end{cases} \quad (5)$$

$$\alpha\epsilon(t) = -\text{Im}\langle \chi | \mu | \Psi \rangle(t) \quad (6)$$

3. FORMULATION OF THE BOUNDED AMPLITUDE, MONOTONICALLY CONVERGENT ALGORITHM

Efficient choices for solving in practice the critical point equations (4)-(6) are given by the monotonically convergent algorithms ((Tannor *et al.*, 1992; Zhu and Rabitz, 1998)) that are guaranteed to improve the cost functional J at each iteration. Note however that none of these algorithms provide a mechanism for ensuring upper and lower bounds on the fields obtained. The purpose of the present work is to introduce such an algorithm that, while still monotonic, allows to compute fields with a-priori given upper and lower bounds. We refer the reader to (Sarychev and Nijmeijer, 1996; Sarychev, 1997) for some theoretical results on the special class of bounded controls that are bang-bang controls. Note however that, to the best of our knowledge, no theoretical results are available for infinite dimensional settings to show that the bang-bang controls are optimal; indeed, as it will be seen in the following, the numerical schemes do lead to bounded fields that are not bang-bang.

Consider $M > 0$, the $sign^+$ and sat_M functions

$$\begin{aligned} sign^+(x) &= \begin{cases} -1, & x < 0 \\ 1, & x \geq 0 \end{cases}, \\ sat_M(x) &= \begin{cases} -M, & x \leq -M \\ x, & -M < x < M \\ M, & x \geq M \end{cases} \end{aligned}$$

and the algorithms described by the resolution of the following equations at step k :

$$\begin{cases} i\frac{\partial}{\partial t}\Psi^k(x, t) = (H_0 - \epsilon^k(t)\mu)\Psi^k(x, t) \\ \Psi^k(x, t=0) = \Psi_0(x) \end{cases} \quad (7)$$

$$\epsilon^k(t) = M \cdot sign^+ \left(-\frac{1}{\alpha} \text{Im}\langle \chi^{k-1} | \mu | \Psi^k \rangle(t) \right) \quad (8)$$

$$\begin{cases} i\frac{\partial}{\partial t}\chi^k(x, t) = (H_0 - \epsilon^k(t)\mu)\chi^k(x, t) \\ \chi^k(x, t=T) = O\Psi^k(x, T) \end{cases} \quad (9)$$

and

$$\begin{cases} i \frac{\partial}{\partial t} \Psi^k(x, t) = (H_0 - \epsilon^k(t)\mu)\Psi^k(x, t) \\ \Psi^k(x, t=0) = \Psi_0(x) \end{cases} \quad (10)$$

$$\epsilon^k(t) = \text{sat}_M \left(-\frac{1}{\alpha} \text{Im} \langle \chi^{k-1} | \mu | \Psi^k \rangle (t) \right) \quad (11)$$

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

Theorem 1. Suppose O is a self-adjoint semi-positive definite observable. Then the algorithm given in Eqns. (7)-(9) converges monotonically in the sense that $J(\epsilon^{k+1}) \geq J(\epsilon^k)$. The same is true for the algorithm given in Eqns. (10)-(12).

Proof Let us evaluate the difference between the values of the cost functional between two successive iterations.

$$\begin{aligned} J(\epsilon^{k+1}) - J(\epsilon^k) &= \langle \Psi^{k+1}(T) | O | \Psi^{k+1}(T) \rangle \\ &- \alpha \int_0^T \epsilon^{k+1}(t)^2 dt - \langle \Psi^k(T) | O | \Psi^k(T) \rangle \\ &+ \alpha \int_0^T \epsilon^k(t)^2 dt \\ &= \langle \Psi^{k+1}(T) - \Psi^k(T) | O | \Psi^{k+1}(T) - \Psi^k(T) \rangle + \\ &2\text{Re} \langle \Psi^{k+1}(T) - \Psi^k(T) | O | \Psi^k(T) \rangle \\ &\alpha \int_0^T \epsilon^k(t)^2 dt - \alpha \int_0^T \epsilon^{k+1}(t)^2 dt \end{aligned} \quad (13)$$

Since we also have:

$$\begin{aligned} &2\text{Re} \langle \Psi^{k+1}(T) - \Psi^k(T) | O | \Psi^k(T) \rangle \\ &= 2\text{Re} \langle \Psi^{k+1}(T) - \Psi^k(T), O\Psi^k(T) \rangle \\ &= 2\text{Re} \langle \Psi^{k+1}(T) - \Psi^k(T), \chi^k(T) \rangle \\ &= 2\text{Re} \int_0^T \left\langle \frac{\partial(\Psi^{k+1}(t) - \Psi^k(t))}{\partial t}, \chi^k(t) \right\rangle + \\ &\left\langle \Psi^{k+1}(t) - \Psi^k(t), \frac{\partial\chi^k(t)}{\partial t} \right\rangle dt \\ &= 2\text{Re} \int_0^T \left\langle \frac{H_0 - \mu\epsilon^{k+1}}{i} \Psi^{k+1}(t) - \right. \\ &\left. \frac{H_0 - \mu\epsilon^k}{i} \Psi^k(t), \chi^k(t) \right\rangle \\ &+ \left\langle \Psi^{k+1}(t) - \Psi^k(t), \frac{H_0 - \mu\tilde{\epsilon}^k}{i} \chi^k(t) \right\rangle \\ &= 2\text{Re} \int_0^T \epsilon^{k+1} \left\langle \frac{-\mu}{i} \Psi^{k+1}(t), \chi^k(t) \right\rangle \\ &- \epsilon^k \left\langle \frac{-\mu}{i} \Psi^k(t), \chi^k(t) \right\rangle + \\ &\epsilon^k \left\langle \Psi^{k+1}(t) - \Psi^k(t), \frac{-\mu}{i} \chi^k(t) \right\rangle \\ &= 2\text{Re} \int_0^T (\epsilon^{k+1} - \epsilon^k) \left\langle \frac{-\mu}{i} \Psi^{k+1}(t), \chi^k(t) \right\rangle \end{aligned} \quad (14)$$

we thus obtain

$$\begin{aligned} &J(\epsilon^{k+1}) - J(\epsilon^k) = \\ &\langle \Psi^{k+1}(T) - \Psi^k(T) | O | \Psi^{k+1}(T) - \Psi^k(T) \rangle + \\ &\alpha \int_0^T \epsilon^k(t)^2 dt - \alpha \int_0^T \epsilon^{k+1}(t)^2 dt \\ &+ 2\text{Re} \int_0^T (\epsilon^{k+1} - \epsilon^k) \left\langle \frac{-\mu}{i} \Psi^{k+1}(t), \chi^k(t) \right\rangle \end{aligned} \quad (15)$$

from which the conclusion of the Theorem follows.

Remark 2. Same results can be obtained for algorithm derived by using the sign^- function

$$\text{sign}^-(x) = \begin{cases} -1, & x \leq 0 \\ 1, & x > 0 \end{cases}$$

Remark 3. Theoretical results: No definitive theoretical results are available to date about the relationship between the possibility of control and the magnitude of the control fields to be used. Promising advances seem to be possible as a by-product of constructive controllability analysis as in (Ramakrishna *et al.*, 2002; Ramakrishna *et al.*, 2000a; Ramakrishna *et al.*, 2000b; Ramakrishna, 2001; Schirmer *et al.*, 2002).

4. NUMERICAL RESULTS

In order to test the performances of the algorithm, a case already treated in the literature was considered (Zhu and Rabitz, 1998). The system under consideration is the $O - H$ bond that vibrates in a Morse type potential. We refer the reader to (Zhu and Rabitz, 1998) for the numerical details concerning this system. The goal is to localize the wave packet at a given location x' at the final time $T = 131000a.u. \simeq 3.16ps$; 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 .

Numerical results are presented in the Figures 1, 2,3 and 4.

Remark 4. The numerical procedure used to propagate the state and the electric field was chosen to be first order for the field update as in (Zhu *et al.*, 1998) Eqn. (46) and second order split-operator for the wave function update (same results are also obtained with the second order scheme as in (Zhu *et al.*, 1998) Eqn. (47));

Finally, let us remark that, although derived for constant parameter α , nothing prevents from using this scheme in the framework of a time-dependent $\alpha(t)$ as in (Hornung *et al.*, 2001).

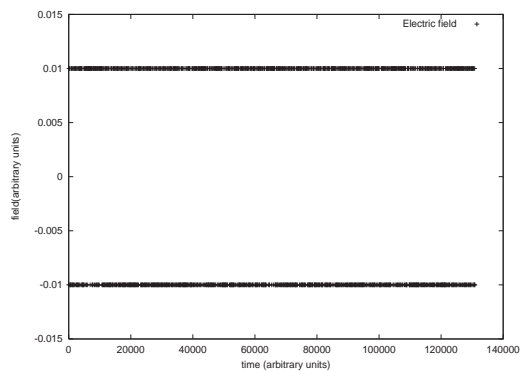


Fig. 1. Example of optimal electric field found by the algorithm described in Eqns. (7)-(9); the bound is $M = 0.01$. As required, the values are either $-M$ or M ; many switching points are present which may imply that such fields are difficult to realize in laboratory; a zoom is presented in Figure 2.

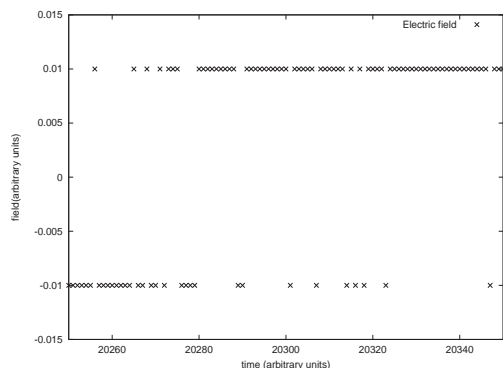


Fig. 2. Zoom (magnification 1300 times) of the optimal electric field in Figure 1.

Remark 5. Various other questions that are raised by the introduction of this new class of algorithms as the dependence of the quality of the solution on the magnitude of the bound M , and the introduction of different cost functionals will be addressed in a following work (Turinici, 2002).

5. DISCUSSION AND CONCLUSIONS

A new class of monotonically convergent algorithms that allow to obtain controls within predefined bounds has been introduced in this paper; its convergence properties were proved in Theorem 1; numerical evidence was presented in Section 4. We refer the reader to (Turinici, 2002) for a more detailed presentation of this topic.

The existence of this new procedures gives the user the flexibility to strike a balance between the degree of control and external requirements on the field amplitude, for instance that raised by the necessity to remain within model validity bounds.

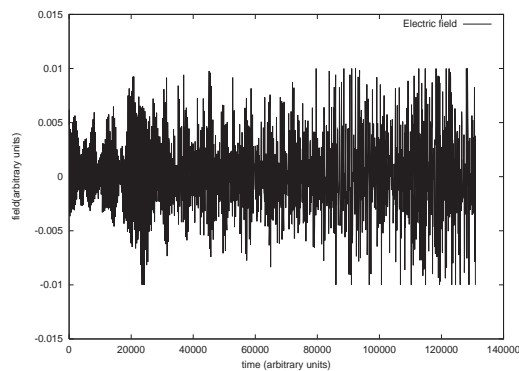


Fig. 3. Example of optimal electric field found with the algorithm given in Eqns. (10)-(12); the bound is $M = 0.01$. As required, the values are all inside the given bounds. Notice that in this case the structure is, to some extent, similar to that obtained when no bounds are enforced and it is not of bang-bang type. This may be indicative of the fact that the bang-bang solutions are not the only optimal within the set of bounded solutions; note that no theory exists to date to support this idea so this conclusion may only be a numerical artifact (although unlikely).

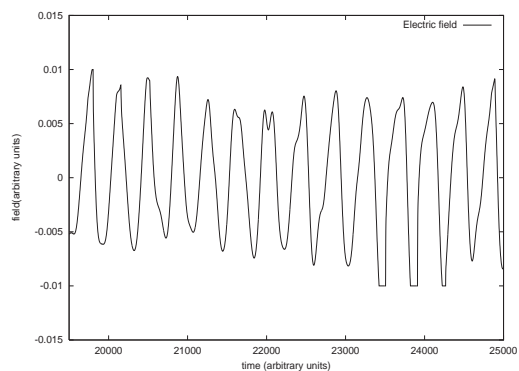


Fig. 4. Zoom of the optimal electric field in Figure 3. Unlike in the Figure 2, the field is continuous, although it may still display oscillations around the time instants where maximal values $\pm M$ are used.

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