DISCRETELY MONOTONICALLY CONVERGENT ALGORITHMS IN QUANTUM CONTROL

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Abstract: The numerical simulation of laser control of molecular systems has made an important step forward by the introduction of algorithms that are guaranteed to improve at each step the cost functional that describes the required control objectives. Nevertheless, after discretization in time, the users may have to deal with instabilities that lead them to stop the simulation indeed with an improved cost functional but before convergence may be reached. In this paper we explain the reasons for such instabilities and propose discrete algorithms that avoid this problem.

Keywords: quantum control, monotonically convergent algorithms, numerical discretization

1. INTRODUCTION

The numerical simulation of the control of molecular systems by a laser has made an important step forward by the introduction by Zhu & Rabitz (Zhu and Rabitz, 1998) and by Tannor (following Krotov) (Tannor *et al.*, 1992) of monotonically convergent algorithms. These algorithms are **guaranteed** to improve at each step a cost functional that describes the required control objectives. This has permitted to reduce the overall cost of the numerical simulation. Even though this approach is still limited to systems of very small size with respect to other approaches (among which lies the successful implementation of the closed loop laboratory learning techniques (Levis *et al.*, 2001; Assion *et al.*, 1998; Bergt *et al.*, 1999; Weinacht *et al.*, 1999; Bardeen *et al.*, 1998)) the discovery of this class of algorithms impulsionned much the early simulations on quantum control that were formerly dependent on the nonlinear conjugate gradient algorithm that displayed poor convergence properties for this highly nonlinear nonconvex cost functional.

In a recent paper two of us have presented a unified framework that comprises both approaches of Krotov and Zhu & Rabitz as particular cases and allows to propose a larger class of monotonically convergent algorithms. Nevertheless, the

actual implementation of these algorithms may exhibit nonconverging sequences. The monotonicity of these algorithms is indeed proved on the continuous in time and space system but the discretization required for real implementations has not been analyzed yet. The combination of the algorithm and the discretization actually can (and indeed does) affect the nice feature that is valid on the continuous optimization problem and is also noticed during the first few iterations.

This note aims at explaining the reasons of the instabilities and also proposes ways to cure them by defining appropriate discrete versions of the algorithm.

2. QUANTUM CONTROL SETTING AND OPTIMAL CONTROL EQUATIONS

Consider a quantum system whose evolution is described by the time-dependent Schrödinger equation (with $\hbar = 1$)

$$\begin{cases} i\dfrac{\partial}{\partial t}\Psi(x,t) = H_0\Psi(x,t) \\ \Psi(x,t=0) = \Psi^0(x) \end{cases} \quad (1)$$

where H_0 is the internal Hamiltonian and $\Psi^0(x)$ the initial state (x denotes the relevant spatial coordinates). The interaction that allows to control the system will be described by a dipol moment operator $\mu(x)$ and a time-varying external field amplitude $\varepsilon(t) \in I\!R$ giving thus rise to the following equations

$$\begin{cases} i\dfrac{\partial}{\partial t}\Psi(x,t) = (H_0 - \varepsilon(t)\mu)\Psi(x,t) \\ \Psi(x,t=0) = \Psi^0(x) \end{cases} \quad (2)$$

We will denote the new Hamiltonian by $H = H_0 - \varepsilon(t)\mu$.

In the absence of constructive information about the structure of $\varepsilon(t)$ that realize the control goals at the final time $t = T$ it is standard to recast the problem as a maximization of a cost functional $J(\varepsilon)$:

$$J(\varepsilon) = \langle\Psi(T)|O|\Psi(T)\rangle - \alpha\int_0^T \varepsilon^2(s)ds \quad (3)$$

where O is an observable operator that describes the target (larger the value $\langle\Psi(T)|O|\Psi(T)\rangle$ better the objectives have been met; we have introduced the notation $< \Psi|O|\chi >= \int_{I\!R^N} \bar{\Psi}O(\chi))$. Note the presence of the penalization term $-\alpha\int_0^T \varepsilon^2(s)$ that forbids too large values of the fluence ($\alpha > 0$ is a parameter but it may also depend on time (Hornung *et al.*, 2001)). Of course, in general attaining the maximal possible value of $\langle\Psi(T)|O|\Psi(T)\rangle$ is at the price of a large laser fluence $\int_0^T \varepsilon^2(s)ds$; The maximization of the cost functional $J(\varepsilon)$ 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):

$$\begin{cases} i\dfrac{\partial}{\partial t}\Psi(x,t) = (H_0 - \varepsilon(t)\mu)\Psi(x,t) \\ \Psi(x,t=0) = \Psi^0(x) \end{cases} \quad (4)$$

where the control field satisfies

$$\alpha\varepsilon = -\mathrm{Im}\langle\chi|\mu|\Psi\rangle(t) \quad (5)$$

and χ is the adjoint state defined by the backward scheme

$$\begin{cases} i\dfrac{\partial}{\partial t}\chi(x,t) = (H_0 - \varepsilon(t)\mu)\chi(x,t) \\ \chi(x,t=T) = O\Psi(x,T) \end{cases} \quad (6)$$

3. DEFINITION OF A CLASS OF MONOTONICALLY CONVERGENT ALGORITHMS

Krotov's and Zhu & Rabitz' scheme enter in the more general class of iterative schemes that are built from (4), (5), (6) and read:

$$\begin{cases} i\dfrac{\partial\Psi^k}{\partial t} = (H_0 + V - \varepsilon^k\mu)\Psi^k \\ \Psi^k(x,t=0) = \Psi^0(x) \end{cases} \quad (7)$$

with :

$$\varepsilon^k = (1-\delta)\tilde{\varepsilon}^{k-1} - \frac{\delta}{\alpha}Im < \chi^{k-1}|\mu|\Psi^k > \quad (8)$$

and

$$\begin{cases} i\dfrac{\partial\chi^k}{\partial t} = (H_0 + V - \tilde{\varepsilon}^k\mu)\chi^k \\ \chi^k(x,t=T) = O\Psi^k(x,T) \end{cases} \quad (9)$$

with :

$$\tilde{\varepsilon}^k = (1-\eta)\varepsilon^k - \frac{\eta}{\alpha}Im < \chi^k|\mu|\Psi^k > . \quad (10)$$

They have been introduced and analyzed in (Maday and Turinici, 2002). Krotov's scheme is obtained with $\delta = 1$ and $\eta = 0$ while Zhu & Rabitz' scheme corresponds to $\delta = 1$ and $\eta = 1$. These algorithms are monotonically convergent provided that δ and η are chosen in $[0, 2]$ as is precised in the following

Theorem 1. Suppose O is a self-adjoint semi-positive definite observable. Then for any $\eta, \delta \in [0, 2]$ the algorithm given in Eqn. (7, 9) converges monotonically in the sense that $J(\varepsilon^{k+1}) \geq J(\varepsilon^k)$

4. DEFINITION OF A CLASS OF SEMI-DISCRETE MONOTONICALLY CONVERGENT ALGORITHMS

4.1 Norm preserving schemes

Our interest now is to propose a proper definition for the discretization in time of the previous algorithm. There are many ways for defining such discretizations but the first thing that has to be in mind is that the final scheme has to be norm preserving. Indeed it is well known that the solution of equation (2) satisfies $\int_{I\!R} |\Psi|^2(x,t)dx = \int_{I\!R} |\Psi^0|^2(x)dx$

In order to satisfy this norm conservation, classically one uses exponential evolution schemes. These schemes start from the definition of discrete time steps $t_j = j\Delta T$, $j = 1, .., N$ and proceed by proposing a second order approximations for the solution of (2) : $\Psi_j \simeq \Psi(t_j)$ iteratively defined by

$$\left\{ \begin{array}{l} \Psi_{j+1} = e^{\frac{H_0\Delta T}{2i}} e^{\frac{V-\mu\varepsilon_j}{i}\Delta T} e^{\frac{H_0\Delta T}{2i}} \Psi_j \\ \\ \Psi_0 = \Psi^0 \end{array} \right.$$

where ε_j is (or an approximation of) the value of ε at time t_j.

4.2 A first definition of the fields

Here ε_j is not known and have to be defined iteratively. Following (8, 10) we propose to set ε_j^k and $\tilde{\varepsilon}_{j-1}^k$ by

$$\begin{aligned} \varepsilon_j^k = & (1-\delta)\tilde{\varepsilon}_j^{k-1} \\ & - \frac{\delta}{\alpha} \mathrm{Im} < e^{\frac{H_0\Delta T}{2i}} \chi_j^{k-1} |\mu_j^k| e^{-\frac{H_0\Delta T}{2i}} \Psi_{j+1}^k > \end{aligned} \quad (11)$$

where

$$\mu_j^k = \frac{i\Big(e^{\frac{-V+\mu\tilde{\varepsilon}_j^{k-1}}{i}\Delta T} - e^{\frac{-V+\mu\varepsilon_j^k}{i}\Delta T}\Big)}{\Delta T(\tilde{\varepsilon}_j^{k-1} - \varepsilon_j^k)}$$

and

$$\begin{aligned} \tilde{\varepsilon}_{j-1}^k = & (1-\eta)\varepsilon_{j-1}^k \\ & - \frac{\eta}{\alpha} \mathrm{Im} < e^{\frac{H_0\Delta T}{2i}} \chi_{j-1}^k |\tilde{\mu}_j^k| e^{-\frac{H_0\Delta T}{2i}} \Psi_j^k > \end{aligned} \quad (12)$$

where

$$\tilde{\mu}_j^k = \frac{i\Big(e^{\frac{-V+\mu\tilde{\varepsilon}_{j-1}^k}{i}\Delta T} - e^{\frac{-V+\mu\varepsilon_{j-1}^k}{i}\Delta T}\Big)}{\Delta T(\tilde{\varepsilon}_{j-1}^k - \varepsilon_{j-1}^k)}$$

The associated discrete evolution equations for the states (direct and adjoint) are

$$\left\{ \begin{array}{l} \Psi_{j+1}^k = e^{\frac{H_0\Delta T}{2i}} e^{\frac{V-\mu\varepsilon_j}{i}\Delta T} e^{\frac{H_0\Delta T}{2i}} \Psi_j^k \\ \\ \Psi_0^k = \Psi^0 \end{array} \right. \quad (13)$$

and

$$\left\{ \begin{array}{l} \chi_{j-1}^k = e^{-\frac{H_0\Delta T}{2i}} e^{\frac{-V+\mu\tilde{\varepsilon}_{j-1}}{i}\Delta T} e^{-\frac{H_0\Delta T}{2i}} \chi_j^k \\ \\ \chi_N^k = O\Psi_N^k \end{array} \right. \quad (14)$$

We are now able to state, on this semi-discrete scheme that, under the same hypothesis as in the Theorem, the discrete quantity

$$J_{\Delta T}(\varepsilon) = \langle \Psi_N | O | \Psi_N \rangle - \alpha \sum_{j=0}^{N-1} \varepsilon_j^2 \quad (15)$$

verifies $J_{\Delta T}(\varepsilon^{k+1}) \geq J_{\Delta T}(\varepsilon^k)$

4.3 A remark on the solution procedure

It has to be noticed that the previous scheme is of implicit type. Currently we have not found any explicit scheme that is monotonic. For implicit schemes, in particular, it has to be verified that there exists at least (and preferably also at most) one solution ε_j^k to (11) and similarly for (12). In order to do so we use a fixed point theorem, either of Brower [1] type of more simply of Picard type. Actually this latter approach is constructive and allows to propose at least one iterative procedure to compute ε_j^k. It is indeed the fixed point of the function f defined by

$$\begin{aligned} f(x) = & (1-\delta)\tilde{\varepsilon}_j^{k-1} \\ & - \frac{\delta}{\alpha} \mathrm{Im} < e^{\frac{H_0\Delta T}{2i}} \chi_j^{k-1} |\mu(x)| e^{\frac{H_0\Delta T}{2i}} \Psi_j^k > \end{aligned} \quad (16)$$

where

$$\mu(x) = \frac{i\Big(e^{\frac{\mu(\tilde{\varepsilon}_j^{k-1} - x)}{i}\Delta T} - 1\Big)}{\Delta T(\tilde{\varepsilon}_j^{k-1} - x)}$$

It can be proved that this function is a contraction provided that

$$\frac{\delta}{\alpha} \|O\|_\infty \|\mu\|_\infty^2 \Delta T < 1$$

Note also that a Newton procedure can be proposed the convergence of which is faster than the Picard iterations.

5. ABOUT THE FULL DISCRETIZATION

The actual implementation of the monotonic algorithm involves a last step : the spacial discretization. The easiest way consists in working in Fourier frame and use the F.F.T. algorithm

[1] It is important to note that the fields ε are thus, proved to be bounded for any k and j. In addition to the monotonic character of the algorithm, this allows to prove the convergence of the algorithm

to travel between the Fourier coefficient space (coefficients with indices between $-M$ and M) known as the spectral space and the physical space (values of the functions at $2M+1$ equidistant points) with a $M\log(M)$ complexity. The interest of this approach is first its important accuracy and also the simplification to evaluate the effect of the 3 operators $e^{\frac{H_0\Delta T}{2i}}$, $e^{\frac{V-\mu\varepsilon_j}{i}\Delta T}$ and $e^{\frac{H_0\Delta T}{2i}}$ that appear in (13). Indeed the first and last operators have a trivial expression in the Fourier space, while the second is trivial in the physical space.

The difficulty with this approach is that the use of the Fourier transformation from the physical space into the spectral space assumes that the result of $e^{\frac{V-\mu\varepsilon_j}{i}\Delta T}e^{\frac{H_0\Delta T}{2i}}\Psi_j^k$ is a trigonometric series with indices between $-M$ and M. This is false and thus the use of the Fourier transform is done on the interpolation of the function onto the $2M+1$ equidistant points. This approximation has to be taken into account and we were not able to prove the monotonicity of the algorithm if such an interpolation procedure is employed due to the unboundedness of the interpolation operator in the L^2– norm. On the contrary, the use of the projection operator, that consists in using the truncated series (between $-M$ and M) is coherent with the other steps in the algorithm and maintains the monotonicity. The projection is slightly more expensive than the interpolation but can be adequately approximated by high order interpolation, i.e. the use of an interpolation operator but on 5 (say) time the number of points.

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