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Limit points the monotonic schemes for quantum control

Julien Salomon

Abstract— **Many numerical simulations in quantum (bilinear) control use the monotonically convergent algorithms of Krotov (introduced by Tannor in [12]), Zhu & Rabitz ([11]) or the general form of Maday & Turinici ([13]). This paper presents an analysis of the limit set of controls provided by these algorithms and a proof of convergence in a particular case.**

I. INTRODUCTION

The control of quantum phenomena is a topic that has been (and is still) a source of many interesting challenges not only to physics and chemistry but also to the mathematics and applied mathematics communities ([1], [2]). At the level of the experiments, laser control of complex molecular systems is becoming feasible, especially since the introduction ([3], [4]) of closed loop laboratory learning techniques and their successful implementation ([5], [6], [7], [8], [9], [10]).

On the other hand, at the level of the numerical simulations, the introduction of the monotonically convergent algorithms by Zhu & Rabitz ([11]) that extend an algorithm due to Krotov ([12]) has allowed a considerable progress and made possible further investigations in this area. Recently, a general class of monotonically convergent algorithms has been proposed ([13]) and a relevant time discretization has been developed ([14]).

However, no general analysis to explain in depth the convergence of these algorithms is available to date. In an attempt to fill this gap, this paper presents some results on the set of the controls provided by monotonic algorithms.

Note that, among others, this question was raised in [15], but a wrong statement about the Cauchy character of the sequence is made that makes the proof not working as stated; the proof is more involved as explained in what follows.

The balance of the paper is as follows: the necessary background and definitions of the quantum control settings are given in Section II, properties of the monotonic sequences are presented in Section III, followed by the properties of the limit set in Section IV. Further results in a particular case are given in Section V and concluding remarks are presented in Section VI.

II. QUANTUM OPTIMAL CONTROL AND MONOTONIC SCHEMES

A. *Cost functional and Euler-Lagrange Equations*

Consider a quantum system prepared in an initial state ψ_0 and whose dynamics is characterized by its internal Hamiltonian H. By assumption this Hamiltonian does not give rise to an appropriate evolution and an external interaction is introduced in order to obtain the desired final property. This interaction is taken here as an electric field with time-dependent amplitude $\varepsilon(t)$ that influences the system through a time-independent dipole moment operator μ. The new Hamiltonian $H - \mu\varepsilon(t)$ gives rise to the equations (we work in atomic units i.e. $\hbar = 1$):

$$\begin{aligned} & i\frac{\partial}{\partial t}\psi(x,t) = H\psi(x,t) - \mu(x)\varepsilon(t)\psi(x,t) \\ & \psi(x,t=0) = \psi_0(x), \end{aligned}$$

where we denote by x the relevant spatial coordinates. These equations hold on $\Omega = \mathbb{R}^N$ but for numerical tests we will consider that x belongs to an interval $\Omega = [0, L]$ and that $\psi(0,t) = \psi(L,t) = 0$, for a large enough real number L and any t in $\mathbb{R}$. This approach is justified by physical reasons since wave functions are generally localized in a space interval.

The optimal control framework is then used to find a suitable evolution of $\varepsilon(t)$. The goal that the final state $\psi(T)$ has prescribed properties is expressed by the introduction of a cost functional J to be maximized. This cost functional also includes a contribution that penalizes undesirable effects. One simple example of such a cost functional is:

$$J(\varepsilon) = \langle\psi(T)|O|\psi(T)\rangle - \alpha\int_0^T \varepsilon^2(t)dt, \qquad (1)$$

where $\alpha > 0$ is a parameter (it may also depend on time cf. [16], [17]) and O is an observable operator that encodes the goal: the larger the value $\langle\psi(T)|O|\psi(T)\rangle$ is, the better the control objectives are met (here and in what follows we use the convention that for any functions f and g and any operator F: $\langle f|F|g\rangle = \int \overline{f(x)}Fg(x)dx$. Note that, in general, achieving the maximal possible value of $\langle\psi(T)|O|\psi(T)\rangle$ is at the price of a large laser influence $\int_0^T \varepsilon^2(t)dt$; the optimum evolution will therefore strike a balance between using a low laser fluence while simultaneously maximizing the desired observable.

At the maximum of the cost functional $J(\varepsilon)$, the Euler-Lagrange critical point equations are satisfied ; a standard way to write these equations is to use a Lagrange multiplier $\chi(x,t)$ called *adjoint state*. The following critical point

J. Salomon is with Laboratoire Jacques-Louis Lions, Université Pierre et Marie Curie, Boîte courrier 187, 75252 Paris Cedex 05, France Salomon@ann.jussieu.fr

equations are thus obtained ([11]):

$$\begin{cases} i\frac{\partial}{\partial t}\psi(x,t) = (H-\varepsilon(t)\mu)\psi(x,t) \\ \psi(x,t=0) = \psi_0(x) \end{cases} \quad (2)$$

$$\begin{cases} i\frac{\partial}{\partial t}\chi(x,t) = (H-\varepsilon(t)\mu)\chi(x,t) \\ \chi(x,t=T) = O\psi(x,T) \end{cases} \quad (3)$$

$$\alpha\varepsilon(t) = -\mathrm{Im}\langle\chi(t)|\mu|\psi(t)\rangle. \quad (4)$$

From now on, $\psi(t)$ and $\chi(t)$ will represent two functions of the Hilbert space $\mathbb{L}^2(\Omega;\mathbb{C})$ for almost all t in $[0,T]$.

B. Definition of the monotonic schemes

Efficient strategies for solving in practice the critical point equations (2)-(4) are represented by the monotonically convergent algorithms ([11], [12], [13]) that are guaranteed to improve the cost functional J at each iteration. In the formulation proposed in [13], the monotonic algorithms are described by the resolution of the following equations at step k:

$$\begin{cases} i\frac{\partial}{\partial t}\psi^k(t) = (H-\varepsilon^k(t)\mu)\psi^k(t) \\ \psi^k(x,t=0) = \psi_0(x) \end{cases} \quad (5)$$

$$\varepsilon^k(t) = (1-\delta)\widetilde{\varepsilon}^{k-1}(t) - \frac{\delta}{\alpha}\mathrm{Im}\langle\chi^{k-1}(t)|\mu|\psi^k(t)\rangle \quad (6)$$

$$\begin{cases} i\frac{\partial}{\partial t}\chi^k(t) = (H-\widetilde{\varepsilon}^k(t)\mu)\chi^k(t) \\ \chi^k(x,t=T) = O\psi^k(x,T) \end{cases} \quad (7)$$

$$\widetilde{\varepsilon}^k(t) = (1-\eta)\varepsilon^k(t) - \frac{\eta}{\alpha}\mathrm{Im}\langle\chi^k(t)|\mu|\psi^k(t)\rangle. \quad (8)$$

where δ and η are two real parameters.

The most important property of this algorithm is given in the following theorem ([13]):

Theorem 1: Suppose O is a self-adjoint positive semi-definite operator. Then, for any $\eta, \delta \in [0,2]$ the algorithm given in Eqns. (5)-(6) converges monotonically in the sense that:

$$J(\varepsilon^{k+1}) \geq J(\varepsilon^k). \quad (9)$$

III. PROPERTIES OF THE SEQUENCE $(\varepsilon^k)_k$, $(\widetilde{\varepsilon}^k)_k$

We first prove that $(\varepsilon^k)_k$ and $(\widetilde{\varepsilon}^k)_k$ defined in (5) and in (6) are bounded. We then prove that every weakly convergent subsequence is strongly convergent. In the following, $||.||$ represents the norm of $\mathbb{L}^2(\Omega,\mathbb{C})$, whereas $\|.\|_2$ represents the norm of $\mathbb{L}^2([0,T];\mathbb{R})$. The scalar product in $\mathbb{L}^2([0,T];\mathbb{R})$ will be denoted by $<.,.>$.

A. Bound for the sequences

We suppose from now on that O and μ are bounded operators and we denote by $||O||_*$, $||\mu||_*$ their norms.

Theorem 2: There exists $M>0$ such that, for all $k>0$, the solutions ε^k, $\widetilde{\varepsilon}^k$ of (5-8) verify:

$$\forall t\in[0,T],\ |\varepsilon^{k+1}(t)| \leq M,\ |\widetilde{\varepsilon}^{k+1}(t)| \leq M.$$

Proof: Define M by:

$$M = \max(\|\varepsilon^0\|_2, \|\widetilde{\varepsilon}^0\|_2, \max(1, \frac{\delta}{2-\delta}, \frac{\eta}{2-\eta})\frac{||O||_*||\mu||_*}{\alpha}), \quad (10)$$

and assume that it has been proven that $\|\widetilde{\varepsilon}^{k-1}\|_2 \leq M$, $\|\varepsilon^{k-1}\|_2 \leq M$. Since we also have:

$$\|\varepsilon^k\|_2 \leq |1-\delta|M + \frac{\delta}{\alpha}\|\ t\mapsto |\mathrm{Im}\langle\chi^{k-1}(t)|\mu|\psi^k(t)\rangle|\ \|_2,$$

the Cauchy-Schwartz inequality yields:

$$\begin{aligned} |\langle\chi^{k-1}(t)|\mu|\psi^k(t)\rangle| &\leq ||\chi^{k-1}(t)||.||\mu(\psi^k(t))|| \\ &\leq ||\mu||_*.||\chi^{k-1}(t)||.||\psi^k(t)||. \end{aligned}$$

We then use the following equalities and bounds on state and adjoint state:

$$\begin{aligned} \forall t, ||\psi^k(t)|| &= 1, \\ \forall t, ||\chi^{k-1}(t)|| &= ||\chi^{k-1}(T)|| = ||O(\psi^{k-1}(T))|| \\ &\leq ||O||_*.||\psi^{k-1}(T)|| = ||O||_* \end{aligned}$$

to obtain the estimate:

$$\|\varepsilon^k\|_2 \leq |1-\delta|M + \delta\frac{||O||_*.||\mu||_*}{\alpha}.$$

If $\delta \leq 1$, then the definition (10) yields $\frac{||O||_*.||\mu||_*}{\alpha} < M$ and then: $\|\varepsilon^k\|_2 \leq |1-\delta|M + \delta M = M$, and if $\delta > 1$ then $\frac{\delta}{2-\delta}\frac{||O||_*||\mu||_*}{\alpha} < M$ and in this case: $\|\varepsilon^k\|_2 \leq |1-\delta|M + \delta\frac{2-\delta}{\delta}M = (\delta-1)M + (2-\delta)M = M$.
A similar proof leads to the same estimate for $\widetilde{\varepsilon}^k$. ■

B. Weak convergence of subsequences

1) Extraction of a weakly convergent subsequence: Because ε^k is bounded in the Hilbert space $\mathbb{L}^2([0,T];\mathbb{R})$, there exists a weakly convergent subsequence that will be denoted by $(\varepsilon^{k_n})_n$. Let ε be the weak limit associated to $(\varepsilon^{k_n})_n$.

2) Limits of $(\varepsilon^{k_n+1}-\varepsilon^{k_n})_n$ and $(\widetilde{\varepsilon}^{k_n+1}-\widetilde{\varepsilon}^{k_n})_n$: The sequence $J(\varepsilon^k)$ is bounded since $|J(\varepsilon^k)| \leq \|O\| + M$. It has also been proven ([13]) that:

$$\begin{aligned} J(\varepsilon^{k+1}) - J(\varepsilon^k) = &\langle\psi^{k+1}(T)-\psi^k(T)|O|\psi^{k+1}(T)-\psi^k(T)\rangle \\ &+ \int_0^T (\frac{2}{\delta}-1)(\varepsilon^{k+1}(t)-\widetilde{\varepsilon}^k(t))^2 dt \\ &+ \int_0^T (\frac{2}{\eta}-1)(\widetilde{\varepsilon}^k(t)-\varepsilon^k(t))^2 dt, \end{aligned}$$

which gives after summation:

$$\begin{aligned} J(\varepsilon^N) - J(\varepsilon^0) = &\sum_0^{N-1}\langle\psi^{k+1}(T)-\psi^k(T)|O|\psi^{k+1}(T)-\psi^k(T)\rangle \\ &+ \int_0^T (\frac{2}{\delta}-1)\sum_0^{N-1}(\varepsilon^{k+1}(t)-\widetilde{\varepsilon}^k(t))^2 dt \\ &+ \int_0^T (\frac{2}{\eta}-1)\sum_0^{N-1}(\widetilde{\varepsilon}^k(t)-\varepsilon^k(t))^2 dt. \end{aligned}$$

Thus the series $\sum_0^{N-1}\|\varepsilon^{k+1}-\widetilde{\varepsilon}^k\|_2^2$ and $\sum_0^{N-1}\|\widetilde{\varepsilon}^k-\varepsilon^k\|_2^2$ converge and we deduce that:

$$\lim_n \|\varepsilon^{k_n+1}-\varepsilon^{k_n}\|_2 = \lim_n \|\widetilde{\varepsilon}^{k_n+1}-\widetilde{\varepsilon}^{k_n}\|_2 = 0. \quad (11)$$

Similar results hold when $\delta = 0, \eta \neq 0$ and $\delta = 1, \eta \neq 0$.
Remark: Such properties do not guarantee the convergence

of the sequences. For example, the sequence $(u_n)_n$ defined by $u_n = \sin(\log(n+1))$ verifies $\sum_{n=0}^{+\infty}(u_{n+1}-u_n)^2 < +\infty$, however it does not converge.

3) Weak convergence of $(\varepsilon^{k_n+p})_n$ and $(\widetilde{\varepsilon}^{k_n+p})_n$: Let $\check{\varepsilon}$ be a test function in $\mathbb{L}^2([0,T];\mathbb{R})$. From:

$$< \check{\varepsilon}, \varepsilon^{k_n+1} > = < \check{\varepsilon}, \varepsilon^{k_n+1} - \varepsilon^{k_n} > + < \check{\varepsilon}, \varepsilon^{k_n} >,$$

one can easily prove that $(\varepsilon^{k_n+1})_n$ weakly converges to ε too. By the same way, $(\varepsilon^{k_n+p})_n$ and $(\widetilde{\varepsilon}^{k_n+p})_n$ also weakly converges to ε.

C. Strong convergence of $(\varepsilon_{k_n})_n$

1) Strong convergence of $(\psi^{k_n})_n$, $(\chi^{k_n-1})_n$ and $(\chi^{k_n})_n$: Since we have proven that $(\varepsilon_{k_n})_n$ and $(\widetilde{\varepsilon}_{k_n})_n$ weakly converge in $\mathbb{L}^2([0,T];\mathbb{R})$, hence in $\mathbb{L}^1([0,T];\mathbb{R})$, we can use theorem 3.6 of ([18]), which implies that ψ^{k_n} strongly converges in $\mathcal{C}([0,T];\mathbb{L}^2(\Omega,\mathbb{C}))$ to the state ψ associated to ε. One can also easily adapt the proof of this theorem in order to obtain that $(\chi^{k_n-1})_n$ and $(\chi^{k_n})_n$ also strongly converge in $\mathcal{C}([0,T];\mathbb{L}^2(\Omega,\mathbb{C}))$ to the adjoint state associated to ε and $\psi(T)$.

2) Strong convergence of $(\varepsilon^{k_n})_n$: The strong convergences of $(\psi^{k_n})_n$, $(\chi^{k_n-1})_n$ and $(\chi^{k_n})_n$ implies the strong convergence of $(\frac{\delta}{\alpha}\text{Im}\langle\chi^{k_n-1}|\mu|\psi^{k_n}\rangle)_n$ and $(\frac{\eta}{\alpha}\text{Im}\langle\chi^{k_n}|\mu|\psi^{k_n}\rangle)_n$ in $\mathcal{C}([0,T];\mathbb{R})$. According to the definitions (5) and (6), we can now write $(\varepsilon^{k_n})_n$ as follows:

$$\varepsilon^{k_n+1} = \underbrace{(1-\delta)(1-\eta)}_{\lambda}\varepsilon^{k_n} + u_n,$$

where $(u_n)_n$ strongly converges. Let e denote a positive real number. Since $|\lambda| < 1$, there exists an integer j_0 such that $|\lambda^{j_0}| < e$. Let us write then:

$$\varepsilon^{k_n} = \varepsilon^0 + \sum_{j=0}^{j_0-1}\lambda^j u_{k_n-j-1} + \lambda^{j_0}\sum_{j=0}^{k_n-j_0}\lambda^j u_{k_n-j-1}. \quad (12)$$

The second term of (12) is a finite sum of strongly convergent contributions and its third term can be bounded above by $e\|u\|_2\frac{1}{1-\lambda}$, which ends the proof of the strong convergence of $(\varepsilon^{k_n})_n$. The strong limit is necessarily ε. Passing to the limit in (5), we conclude that ε is a critical point of J.
A similar proof can be done to prove that $(\widetilde{\varepsilon}^{k_n})_n$ strongly converges to ε.
It thus has been proven that every weakly convergent subsequence of $(\varepsilon^k)_k$ strongly converge in $\mathcal{C}([0,T];\mathbb{R})$ to a critical point of J.

IV. PROPERTIES OF THE LIMIT SET

This section is devoted to the study of the limit set of the sequence $(\varepsilon^k)_k$ which will be denoted by A. From now on we will suppose that this set contains at least two points[1].

[1] Of course A depends on the initial guess ε^0 for the definition of the sequence $(\varepsilon^k)_k$ (i.e. $A = A(\varepsilon^0)$ even though we shall skip this dependency in all what follows).

A. First properties

From theorem 2, one deduces that $A \subset B(0,M)$, where M is defined in (10) and $B(0,M)$ stands for the ball of radius M of $\mathbb{L}^2([0,T];\mathbb{R})$. According to the definition of A, the results of the latest section prove that A is a subset of the set of critical points of J. Finally, thanks to the monotonic property (9) of the sequence $(\varepsilon^k)_k$, we deduce that J is constant on A.

B. Compactness

Let us now prove a first topological property.

Lemma 1: The set A is compact.

Proof: Let $(\varepsilon_\infty^n)_n$ denote a sequence of A. We can associate to this sequence a subsequence $(\varepsilon^{k_n})_n$ of $(\varepsilon^k)_k$ such that $\|\varepsilon_\infty^n - \varepsilon^{k_n}\|_2 < \frac{1}{n}$. According to the previous results we can extract from $(\varepsilon^{k_n})_n$ a strongly convergent subsequence $(\varepsilon^{k_{n'}})_{n'}$. Let ε^* denote the limit of this latter. Thus, the sequence $(\varepsilon_\infty^{n'})_{n'}$ strongly converges to ε^* that is a point of A. ■

C. e-Strings in A

Consider a general metric space (E,d), $(x,y) \in E^2$ and e a positive real number. We call e-string between x and y a finite sequence $z_1, ..., z_N$ of point of E such that:

- $z_1 = x$,
- $z_N = y$,
- $\forall k \in [1, N-1],\ d(z^{k+1}, z^k) < e$.

Then the set A has the following topological property.

Lemma 2: For any $(\varepsilon_\infty, \varepsilon'_\infty) \in A^2$ and any $e > 0$, there exits an e-string in A between ε_∞ and ε'_∞.

Proof: As a compact set, there exist N_0 open balls of radius $\frac{e}{4}$ covering A. By the definition of A and (11), there exists an infinity of $K > 0$ for which $l_K = \varepsilon_\infty, \varepsilon^K, ..., \varepsilon^{K+N(K)}, \varepsilon'_\infty$ is an e-string. From l_K, one can then build another e-string $l'_K = \varepsilon^{K,1}, \varepsilon^{K,2}, ..., \varepsilon^{K,N_0}$. Indeed, if $N_0 > N(K)$, define l'_K by:

$$l'_K = l_K, \underbrace{\varepsilon^{K+N(K)}, ..., \varepsilon^{K+N(K)}}_{N_0-N(K)\ terms},$$

and if $N_0 > N(K)$, one can remove $N(K) - N_0$ terms of l_K while keeping the e-string properties.
For each i, $1 \leq i \leq N_0$, let us extract from $(\varepsilon^{K,i})_K$ a strongly convergent subsequence of $(\varepsilon^k)_k$. The limits obtained are an e-string in A. ■

D. Connectivity

The previous result leads to the following theorem.

Theorem 3: The set A is connected.

Proof: Suppose there exist two closed subsets of A, denoted by A_1 and A_2, such that $A = A_1 \cup A_2$ and $A_1 \cap A_2 = \emptyset$. Because of the existence of e-strings for every e, we deduce that the distance between A_1 and A_2 is equal to 0. Since A is compact, this is in contradiction with $A_1 \cap A_2 = \emptyset$. ■

E. Summary

It has been proven that the limit points of a sequence obtained by a monotonic scheme are a compact and connected set of critical points of J. Note that if this set is reduced to one point, the compactness of the sequence implies its convergence.

V. VARIATIONAL ANALYSIS AND PARTICULAR CASE

Let us focus now on the scheme obtained for $\delta = 1$ and $\eta = 0$, which corresponds to the Krotov formulation (as in [12]). We will estimate the variations of ψ and χ with respect to ε. The results obtained will enable us to prove the convergence for large values of the parameter α.
The above defined set A is still considered to contain at least two points.

A. Estimates

Let ε and ε' be two points of A, ψ and ψ' the corresponding states given by (2) and χ and χ' the corresponding adjoint states solution of (3). Consider (2) written in integrated form, for ψ and ψ':

$$\begin{aligned}\psi(t) &= e^{-iHt}\psi_0 + \int_0^t e^{-iH(t-s)}\varepsilon(s)i\mu\psi(s)ds,\\ \psi'(t) &= e^{-iHt}\psi_0 + \int_0^t e^{-iH(t-s)}\varepsilon'(s)i\mu\psi'(s)ds.\end{aligned}$$

Let us introduce the notations $\delta\psi(t) = \psi(t) - \psi'(t)$, $\delta\chi(t) = \chi(t) - \chi'(t)$ and $\delta\varepsilon(t) = \varepsilon(t) - \varepsilon'(t)$, we then have:

$$\begin{aligned}\delta\psi(t) &= \int_0^t e^{-iH(t-s)}\delta\varepsilon(s)i\mu\psi(s)ds\\ &\quad + \int_0^t e^{-iH(t-s)}\varepsilon'(s)i\mu\delta\psi(s)ds. \qquad (13)\end{aligned}$$

Since the operator e^{-iHt} is unitary, we deduce that:

$$\begin{aligned}&\|\int_0^t e^{-iH(t-s)}\delta\varepsilon(s)i\mu\psi(s)ds\| < \|\mu\|_* T\|\delta\varepsilon\|_2,\\ &\|\int_0^t e^{-iH(t-s)}\varepsilon'(s)i\mu\delta\psi(s)ds\| < M\|\mu\|_* \int_0^t \|\delta\psi(s)\|ds,\end{aligned}$$

where M has been defined in (10). From Gronwall's lemma applied to (13), we obtain:

$$\|\delta\psi(t)\| \leq \|\mu\|_* T e^{T\|\mu\|_* M}\|\delta\varepsilon\|_1, \qquad (14)$$

where $\|.\|_1$ represents the norm of $\mathbb{L}^1([0,T];\mathbb{R})$. A similar computation for the adjoint state leads to:

$$\|\delta\chi(t)\| \leq \|O\|_*\|\mu\|_* T(1 + e^{T\|\mu\|_* M})e^{T\|\mu\|_* M}\|\delta\varepsilon\|_1. \qquad (15)$$

B. Convergence

Since ε and ε' are critical points of J, the two following equalities hold:

$$\begin{aligned}\alpha\varepsilon(t) &= -\mathrm{Im}\langle\chi(t)|\mu|\psi(t)\rangle,\\ \alpha\varepsilon'(t) &= -\mathrm{Im}\langle\chi'(t)|\mu|\psi'(t)\rangle.\end{aligned}$$

The difference of these two equalities yields:

$$\alpha\delta\varepsilon(t) = -\mathrm{Im}(\langle\delta\chi|\mu|\psi\rangle(t) + \langle\chi|\mu|\delta\psi\rangle(t)).$$

From (14,15) we have:

$$\alpha\|\delta\varepsilon\|_1 \leq \|O\|_*\|\mu\|_* T(2 + e^{T\|\mu\|_* M})e^{T\|\mu\|_* M}\|\delta\varepsilon\|_1. \qquad (16)$$

Thus we get the following result:

Theorem 4: The monotonic scheme defined by (5)-(8), $\delta = 1$, $\eta = 0$ strongly converges in $\mathbb{L}^2([0,T];\mathbb{R})$ under the assumption that:

$$\alpha > \|O\|_*\|\mu\|_* T(2 + e^{T\|\mu\|_* M})e^{T\|\mu\|_* M}.$$

Proof: Suppose that the monotonic scheme does not converge, then there exists at least two distinct points ε and ε'. Using the above notations, the equation (16) holds in this case. Since $\delta\varepsilon \neq 0$, we reach a contradiction. ■

VI. CONCLUSION

It has been proven that the sequences provided by monotonic schemes are compact and that the set of their limit points is compact and connected. It has been shown that this set reduces to one point (i.e. the algorithm strongly converges) for a large laser fluence penalty parameter α. We refer the reader to [19] for a more detailed presentation of this topic.

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REFERENCES

[1] H. Rabitz, Shaped laser pulses as reagents, *Science*, 299, 2003, pp 525-526.

[2] 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*, Elsevier Science B.V.; 2003, pp 833-887.

[3] R. S. Judson and H. Rabitz, Teaching lasers to control molecules, *Phys. Rev. Lett.*, 68, 1992, pp 1500-1503.

[4] S. Shi, A. Woody, and H. Rabitz, Optimal Control of Selective Vibrational Excitation in Harmonic Linear Chain Molecules, *J. Chem. Phys.*, 88, 1988, pp 6870-6883.

[5] R. J. Levis, G. Menkir and H. Rabitz, Selective bond dissociation and rearrangement with optimally tailored, strong field laser pulses, *Science*, 292, 2001, pp 709-713.

[6] A. Assion, T. Baumert, M. Bergt, T. Brixner, B. Kiefer, V. Seyfried, M. Strehle and G. Gerber, Control of chemical reactions by feedback-optimized phase-shaped femtosecond laser pulses, *Science*, 282, 1998, pp 919-922.

[7] M. Bergt, T. Brixner, B. Kiefer, M. Strehle and G. Gerber, Controlling the femto-chemistry of $Fe(CO)_5$, *J. Phys. Chem. A.*, 103, 1999, pp 10381-10387.
[8] T. Weinacht, J. Ahn and P. Bucksbaum, Controlling the shape of a quantum wavefunction, *Nature*, 397, 1999, pp 233-235.
[9] C. Bardeen, V. V. Yakovlev, K. R. Wilson, S. D. Carpenter, P. M. Weber and W. S. Warren, Feedback quantum control of molecular electronic population transfer, *Chem. Phys. Lett.*, 280, 1997, pp 151-158.
[10] C. J. Bardeen, V. V. Yakovlev, J. A. Squier and K. R. Wilson, Quantum control of population transfer in green fluorescent protein by using chirped femtosecond pulses, *J. Am. Chem. Soc.*, 120, 1998, pp 13023-13027.
[11] W. Zhu and H. Rabitz, A rapid monotonically convergent iteration algorithm for quantum optimal control over the expectation value of a positive definite operator, *J. Chem. Phys.*, 109, 1998, pp 385-391.
[12] D. Tannor, V. Kazakov and V. Orlov, *Control of photochemical branching: Novel procedures for finding optimal pulses and global upper bounds* In *Time Dependent Quantum Molecular Dynamics*, edited by Broeckhove J. and Lathouwers L. Plenum; 1992, pp 347-360.
[13] Y. Maday and G. Turinici, New formulations of monotonically convergent quantum control algorithms, *J. Chem. Phys.*, 118 (18), 2003, pp 8191-8196.
[14] Y. Maday, J. Salomon and G. Turinici, "Discretely monotonically convergent algorithm in quantum control", *Proceedings of the LHMNLC03 IFAC conference*, Sevilla, Spain, April 2003, pp 321-323.
[15] S. Grivopoulos and B. Bamieh, "Iterative algorithms for optimal control of quantum systems", *Proceedings of the 41st IEEE Conference on Decision and Control*, Las Vegas, Nevada, December 2002, pp 2687-2691.
[16] T. Hornung, M. Motzkus and R. de Vivie-Riedle, Adapting optimal control theory and using learning loops to provide experimentally feasible shaping mask patterns, *J. Chem.Phys.*, 115, 2001, pp 3105-3111.
[17] J. Salomon and G. Turinici, "Control of molecular orientation and alignment by monotonic schemes", *Proceedings of the 24-th IASTED International Conference on modelling, identification and control*, Innsbruck, Austria, February 2005, 457-187.
[18] J.M. Ball, J.E. Marsden an M. Slemrod, Controllability for distributed bilinear systems, *SIAM J. Control and Optimization*, 20 (4), 1982, pp 575-597.
[19] J. Salomon, Ph.D. Thesis, *in progress.*