Equivalence between local tracking procedures and monotonic algorithms in quantum control

Gabriel Turinici

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***Abstract*— The computer simulations of quantum control use several approaches including local tracking procedures that prescribe the controlling field through the requirement that a certain functional be decreasing and monotonic algorithms that solve the Euler-Lagrange equations for a predefined cost functional. While different in implementation, recent works [1] hinted that these two classes share some common characteristics. We propose in this contribution a rigorous ground for such conclusions and discuss the precise formulation that allows to construct this equivalence.**

I. Introduction

Manipulation of quantum phenomena was already demonstrated both in closed-loop laboratory experiments [2], [3], [4], [5], [6] and in theoretical studies on the controllability of quantum systems [7], [8], [9], [10].

Accompanying these advances, the computer simulations have the advantage to overcome experimental restrictions and have access to the whole dynamics allowing further insight and also providing hints in devising future experiments. Many algorithms have been proposed to solve the ensuing optimization problem among which two distinct classes can be identified. The first one contains the local tracking methods [11], [12], [13], [14], [15], [1] that propose explicit formulae of the driving field in an open-loop dependence on the evolving state. The formulae are obtained from the requirement to decrease a certain functional defined at each time instant and related to the "distance" to the target or by demanding strict adherence to a predefined observable trajectory. The second class are the monotonic algorithms [16], [17], [18] that solve the Euler-Lagrange equations associated to the optimization of a cost functional defined at a final time T. The two classes can also be combined as in [1].

Although different in implementation, these algorithms are shown below to be related in the sense that monotonic schemes are tracking procedures for some specific performance indexes. In the context of the density matrix formulation, this index is defined as the value of the cost functional (at the final time T) evaluated for the "best candidate" field at time $t \leq T$. This candidate field is made from the current field up to time t that is prolonged with a given, reference field from t to T obtained at the previous iteration. This *forward cost functional* is decreased at each time instant. For the case of the wavefunction, due to the nonlinear nature of the target formulation, an upper bound is used to define the forward cost functional.

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The outline of the paper is the following: for the case of the density matrix we present the definition of tracking procedures in section II-A and one example of monotonic algorithm in section II-B. Then, we explain the relation between the two classes in sections II-C and II-D. The corresponding analysis for the wavefunction formulation is given in section III. Concluding discussions and remarks are the object of section IV.

II. Density matrix formulation

Consider a quantum system with internal dynamics described by the Hamiltonian H_0. Its interaction with an external (e.g., laser) field is modeled by introducing the dipole moment operator μ and the field intensity $\epsilon(t)$. If the system is represented in the density matrix formulation with initial state ρ_0 its dynamics will obey the time dependent Schrödinger equation:

$$\begin{aligned} i\frac{\partial}{\partial t}\rho(x,t) &= [H_0 - \epsilon(t)\mu, \rho(x,t)] \\ \rho(x,t=0) &= \rho_0(x). \end{aligned} \tag{1}$$

Here we used the convention $\hbar = 1$.

We introduce the Liouville space representation, by defining the scalar product $\langle\langle a, b\rangle\rangle = Tr(a^\dagger b)$ and the associated norm $\|a\|_{\rangle\rangle} = \sqrt{\langle\langle a, a\rangle\rangle}$. Instead of the commutators above, we define the operators $\mathcal{H}$ and $\mathcal{M}$ that act on density matrices by $\mathcal{H}\rho = [H_0, \rho]$, $\mathcal{M}\rho = [\mu, \rho]$. Equation (1) becomes

$$\begin{aligned} i\frac{\partial}{\partial t}\rho(x,t) &= \left(\mathcal{H} - \epsilon(t)\mathcal{M}\right)\rho(x,t) \\ \rho(x,t=0) &= \rho_0(x). \end{aligned} \tag{2}$$

The control goal can be expressed through the introduction of an observable operator A by the requirement that the quantity $\langle A(t)\rangle = Re\langle\langle A, \rho\rangle\rangle$ be maximized (Re denoting the real part of a complex number). This formulation can be further refined as in [15] where an index $y(t) = y(\langle A_1(t)\rangle, ..., \langle A_K(t)\rangle)$ aggregating several observables is considered or even further by defining $y(t) = y(\int_0^t \epsilon^2(s)ds, \langle A_1(t)\rangle, ..., \langle A_K(t)\rangle)$ where we introduce explicitly the dependence on the laser fluence. For notational convenience we will denote $F(t) = \int_0^t \epsilon^2(s)ds$.

A. Tracking algorithm

Consider the simple situation $y(t) = y(\int_0^t \epsilon^2(s)ds, \langle A(t)\rangle)$ where only one observable is considered i.e., $K = 1$ above. We obtain

$$\frac{dy(t)}{dt} = D_1 y \cdot \epsilon(t) + D_2 y \cdot Re\langle\langle A, \frac{\left(\mathcal{H} - \epsilon(t)\mathcal{M}\right)\rho(x,t)}{i}\rangle\rangle$$

where D_j is the partial derivative with respect to the j-th variable. This can be further expressed as

$$\frac{dy(t)}{dt} = f(F(t),\rho) + \epsilon(t) g(F(t),\rho) \tag{3}$$

It is seen that, except for the points where g vanishes (which will be called singularities and will be treated separately) for any desired trajectory $\widetilde{y}$ with $\widetilde{y}(0) = y(0)$, the condition $y(t) \equiv \widetilde{y}(t)$ uniquely determines the field $\epsilon(t)$ by the formula

$$\epsilon(t) = \frac{\frac{d\widetilde{y}(t)}{dt} - f(F(t),\rho)}{g(F(t),\rho)}. \tag{4}$$

From $dF/dt = \epsilon^2(t)$ one obtains that (4) is in fact a ODE on F of the form

$$dF/dt = \mathcal{Y}(F,\rho) \tag{5}$$

that is to be solved jointly with (2) in order to ensure adherence to the prescribed trajectory $\widetilde{y}$.

Same considerations apply if only weaker properties are required, typically the increase/decrease of $y(t)$ which can be enforced through the condition $dy/dt \geq 0$ (≤ 0).

The difficulty in this approach is to find a suitable reference tracking trajectory $\widetilde{y}$ that does not give rise to singular points of the system (3), (5) i.e., where $g(F,\rho) = 0$. In general singular points cannot be avoided a priori and techniques were designed to treat such situations: see [14] for designs that locally alter the trajectory to circumvent the singular points and [19], [1] for a study on the stopping points and procedures to improve their optimality.

B. Monotonic algorithms for optimal control

In an approach different from tracking, monotonically convergent algorithms pioneered in [20], [21] and extended in [18] in the wavefunction representation, are used in the context of the density matrix operator as in [22], [23]. Such procedures are included in the framework of the optimal control that introduces a cost functional (defined at a final time T) to be optimized. One such example of functional is

$$J(\epsilon) = \int_0^T \alpha\epsilon^2(t)dt - 2Re\langle\langle A, \rho(T)\rangle\rangle. \tag{6}$$

where α is a positive (constant or time varying) weight. Then, the critical points of $J(\epsilon)$ are sought after under the constraint of satisfying (2). Because of the constraint, a Lagrange multiplier, denoted $\chi(x,t)$ is introduced in the cost functional that now reads

$$\begin{aligned} J_2(\epsilon) &= \int_0^T \alpha\epsilon^2(t)dt - 2Re\langle\langle A, \rho(T)\rangle\rangle \\ &+2Re\left\{\int_0^T \langle\langle \chi, \frac{\partial\rho}{\partial t} - \frac{(\mathcal{H} - \epsilon(t)\mathcal{M})\rho}{i}\rangle\rangle dt\right\} \end{aligned} \tag{7}$$

The critical point equations are thus obtained:

$$i\frac{\partial}{\partial t}\rho(x,t) = (\mathcal{H} - \epsilon(t)\mathcal{M})\,\rho(x,t) \tag{8}$$
$$\rho(x,t=0) = \rho_0(x)$$
$$2\alpha\epsilon(t) + 2Re\langle\langle \chi, \frac{\mathcal{M}\rho}{i}\rangle\rangle(t) = 0 \tag{9}$$
$$i\frac{\partial}{\partial t}\chi(x,t) = (\mathcal{H} - \epsilon(t)\mathcal{M})\,\chi(x,t) \tag{10}$$
$$\chi(x,T) = A.$$

Building on these relations, the monotonic algorithms prescribe a particular order to iterate in these coupled equations by constructing, at the iteration step $k \rightarrow k+1$, a field $\epsilon_{k+1}(t)$ with the important property

$$J(\epsilon_{k+1}) \leq J(\epsilon_k), \tag{11}$$

hence the name of *monotonic algorithm*. A simple example of such algorithm is (see [22], [23] for additional details):

$$i\frac{\partial}{\partial t}\rho_{k+1}(x,t) = (\mathcal{H} - \epsilon_{k+1}(t)\mathcal{M})\,\rho_{k+1}(x,t) \tag{12}$$
$$\rho_{k+1}(x,t=0) = \rho_0(x)$$
$$\epsilon_{k+1}(t) = -\frac{1}{\alpha}Re\langle\langle \chi_k, \frac{\mathcal{M}\rho_{k+1}}{i}\rangle\rangle(t) \tag{13}$$
$$i\frac{\partial}{\partial t}\chi_{k+1}(x,t) = (\mathcal{H} - \epsilon_{k+1}(t)\mathcal{M})\,\chi_{k+1}(x,t) \tag{14}$$
$$\chi_{k+1}(x,T) = A.$$

This algorithm is proved [23] to have the convenient property in Eqn. (11). It is to be noted that this property is very surprising in this highly nonlinear setting, especially when considering that no second order information is directly involved in the computations.

Note that (12) and (13) are to be solved simultaneously because of the inter-dependence of the field $\epsilon_{k+1}(t)$ and the state $\rho_{k+1}(t)$. An alternative procedure is to insert relation (13) into equation (12) which will become a non-linear Schrödinger equation to be propagated forward in time.

Remark 1: Embedded into the writing of the scheme, at the convergence, the satisfaction of the critical point equations is ensured. See also [24] for further considerations on the convergence. Note that this desirable property is not always guaranteed for tracking.

C. Forward cost functional

Note that the cost functional of equation (6) has exactly the same minima and critical points as

$$J_{dist}(\epsilon) = \int_0^T \alpha\epsilon^2(t)dt + \|A - \rho(T)\|_{\rangle\rangle}^2, \tag{15}$$

which measures the distance of the final density $\rho(x,T)$ to the target operator A. This conclusion is true due to the norm conservation properties of the Schrödinger equation which allows to write $J_{dist} = J + \|A\|_{\rangle\rangle} + \|\rho(T)\|_{\rangle\rangle} = J + \|A\|_{\rangle\rangle} + \|\rho_0\|_{\rangle\rangle}$ and thus to conclude that J and J_{dist} only differ by a constant.

The optimal control strategy of section II-B operates on a cost functional defined at final time T. As such, during the

evolution at time $t < T$, this value is not yet accessible for immediate feedback into the optimization procedure. However, with a field computed up to $t < T$ a reasonable alternative is to use a candidate ϵ_{ref} on $[t, T]$ to compute the performance index at final time T. An appealing choice for ϵ_{ref} is the field obtained at a previous iteration. We are thus lead to introduce for a control ϵ known on $[0, t]$ and a reference field ϵ_{ref} defined on $[0, T]$ the field

$$\bar{\epsilon}(s) = \begin{cases} \epsilon(s) & for\ 0 \leq s \leq t \\ \epsilon_{ref}(s) & for\ t \leq s \leq T. \end{cases} \quad (16)$$

This field is the best available candidate at time $t < T$. Its performance index $J_{dist}(\bar{\epsilon})$ is

$$J_{dist}(\bar{\epsilon}) = \int_0^t \alpha \bar{\epsilon}^2(t)dt + \|A - \rho_{\bar{\epsilon}}(T)\|_{\rangle\rangle}^2, \quad (17)$$

where $\rho_{\bar{\epsilon}}(T)$ is the state at time T of the system

$$\begin{aligned} &i\frac{\partial}{\partial t}\rho_{\bar{\epsilon}}(x,t) = \left(\mathcal{H} - \bar{\epsilon}(t)\mathcal{M}\right)\rho_{\bar{\epsilon}}(x,t) \quad (18)\\ &\rho_{\bar{\epsilon}}(x, t=0) = \rho_0(x), \end{aligned}$$

A property with important practical implications on the efficient computation of $J_{dist}(\bar{\epsilon})$ is given in the following

Proposition 1: Define the *forward cost functional* for the control ϵ and reference field ϵ_{ref} as

$$J_{fwd}(\epsilon, t; \epsilon_{ref}) = \int_0^t \alpha\epsilon^2(t)dt \quad (19)$$

$$+ \int_t^T \alpha {\epsilon_{ref}}^2(t)dt + \|\rho_{ref}(t) - \rho(t)\|_{\rangle\rangle}^2. \quad (20)$$

where ρ evolves on $[0, t]$ as in (2) and ρ_{ref} is the inverse propagation from A with field ϵ_{ref}:

$$\begin{aligned} &i\frac{\partial}{\partial t}\rho_{ref}(x,t) = \left(\mathcal{H} - \epsilon_{ref}(t)\mathcal{M}\right)\rho_{ref}(x,t) \quad (21)\\ &\rho_{ref}(x, t=T) = A. \end{aligned}$$

Then $J_{fwd}(\epsilon, t; \epsilon_{ref}) = J_{dist}(\bar{\epsilon})$.

Proof The first two terms in $J_{fwd}(\epsilon, t; \epsilon_{ref})$ are precisely the first term of $J_{dist}(\bar{\epsilon})$. To compute the second term in $J_{dist}(\bar{\epsilon})$, ρ is to be evolved from ρ_0 with the field $\bar{\epsilon}$ on $[0, T]$ to obtain $\rho(T)$. But, since ρ_{ref} and ρ evolve with the same field on the interval $[t, T]$, their distance will be constant throughout evolution and thus $\|A - \rho(T)\|_{\rangle\rangle}^2 = \|\rho_{ref}(T) - \rho(T)\|_{\rangle\rangle}^2 = \|\rho_{ref}(t) - \rho(t)\|_{\rangle\rangle}^2$. Thus we conclude that $J_{dist}(\bar{\epsilon}) = J_{fwd}(\epsilon, t; \epsilon_{ref})$.

Remark 2: As the adjoint is available during the iterations of the monotonic algorithms, the above property can be used to monitor the evolution of the cost functional between two successive iterations. For instance this can help revealing which part of the evolution contributes more to the optimization and relate thus to local in time mechanisms of control.

D. Monotonic algorithms as local tracking procedures

The result above gives, at any intermediary time $t < T$ the value that the cost functional $J_{fwd}(\epsilon, t; \epsilon_{ref})$ will take at time T if the optimization is stopped at the instant $t \leq T$ (and the field is put to be ϵ_{ref} on $[t, T]$). Note that the value of $J_{fwd}(\epsilon, t; \epsilon_{ref})$ is readily computed at any time t as soon as the inverse propagation (21) is computed once. Armed with this tool, optimization need not wait till the final time T but can instead already operate at the current time t using local tracking procedures to optimize the value $J_{fwd}(\epsilon, t; \epsilon_{ref})$. We are now in position to claim the following

Theorem 1: The monotonic algorithm (12)-(14) is a local tracking procedure for the forward cost functional $J_{fwd}(\epsilon_{k+1}, t; \epsilon_{ref} = \epsilon_k)$ at any time t in the sense that $J_{fwd}(\epsilon_{k+1}, t; \epsilon_k)$ is a monotonically decreasing function of t on the interval $[0, T]$.

Proof Note that $\epsilon_k = \epsilon_{ref}$ imply $\chi_k(t) = \rho_{ref}(t)$. Let us compute the time derivative of $J_{fwd}(\epsilon_{k+1}, t; \epsilon_k)$:

$$\begin{aligned} &\frac{d}{dt}J_{fwd}(\epsilon_{k+1}, t; \epsilon_k) = \alpha\epsilon_{k+1}^2(t) - \alpha\epsilon_k^2(t)\\ &-2\frac{d}{dt}Re\langle\langle\chi_k(t), \rho_{k+1}(t)\rangle\rangle\\ &= \alpha\epsilon_{k+1}^2(t) - \alpha\epsilon_k^2(t)\\ &-2Re\langle\langle\frac{d}{dt}\chi_k(t), \rho_{k+1}(t)\rangle\rangle - 2Re\langle\langle\chi_k(t), \frac{d}{dt}\rho_{k+1}(t)\rangle\rangle\\ &= \alpha\epsilon_{k+1}^2(t) - \alpha\epsilon_k^2(t)\\ &-2Re\langle\langle\frac{\left(\mathcal{H} - \epsilon_k(t)\mathcal{M}\right)\chi_k(t)}{i}, \rho_{k+1}(t)\rangle\rangle\\ &-2Re\langle\langle\chi_k(t), \frac{\left(\mathcal{H} - \epsilon_{k+1}(t)\mathcal{M}\right)\rho_{k+1}(t)}{i}\rangle\rangle\\ &= \alpha\epsilon_{k+1}^2(t) - \alpha\epsilon_k^2(t) + 2Re\langle\langle\frac{\epsilon_k(t)\mathcal{M}\chi_k(t)}{i}, \rho_{k+1}(t)\rangle\rangle\\ &+2Re\langle\langle\chi_k(t), \frac{\epsilon_{k+1}(t)\mathcal{M}\rho_{k+1}(t)}{i}\rangle\rangle\\ &= \alpha\epsilon_{k+1}^2(t) - \alpha\epsilon_k^2(t) + 2\epsilon_k(t)\alpha\epsilon_{k+1}(t)\\ &-2\epsilon_{k+1}(t)\alpha\epsilon_{k+1}(t) = -\alpha\left[\epsilon_{k+1}(t) - \epsilon_k(t)\right]^2. \quad (22) \end{aligned}$$

Thus $J_{fwd}(\epsilon_{k+1}, t; \epsilon_k)$ is a decreasing function of t.

Remark 3: The monotonicity follows as a corollary of the previous property of J_{fwd}, since $J_{dist}(\epsilon_k) = J_{fwd}(\epsilon_{k+1}, 0; \epsilon_k) \geq J_{fwd}(\epsilon_{k+1}, T; \epsilon_k) = J_{dist}(\epsilon_{k+1})$.

The result above may also suggest the following interpretation: for any candidate solution ϵ_k two trajectories can be computed: $\rho_k(t)$ that starts from the correct initial condition ρ_0 but whose final state $\rho_k(T)$ may not yet be satisfactory close to the target, and the adjoint state $\chi_k(t)$ that propagates backward from the target A but may not reach the correct initial state ρ_0; the idea is to make the trajectories coincide by computing ϵ_{k+1} such that $\rho_{k+1}(t)$ approaches monotonically $\chi_k(t)$. In the approximation where the fluence penalty $\int_0^T \alpha\epsilon^2(t)$ is negligible before the control part $\|\chi_k(t) - \rho_{k+1}(t)\|_{\rangle\rangle}$ the distance between the two trajectories will decrease until its final value at time T. The situation is schematically depicted in Fig. 1.

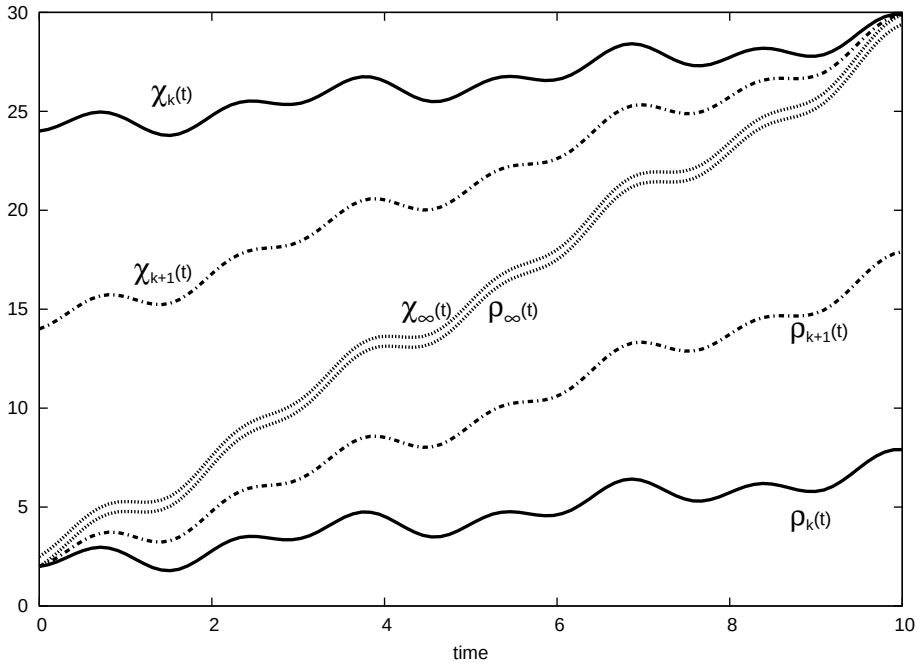

Fig. 1. Schematic illustration of the convergence of the monotonic algorithms for negligible fluence. The evolving state ρ_{k+1} is approaching monotonically the reference trajectory χ_k. At the next iteration χ_{k+1} will remain at a constant distance from ρ_{k+1} because both use the same field ϵ_{k+1}. This shrinking distance between the two trajectories ensure the progression of the cost functional toward optimal values. This observation is currently used in the context of efficient parallelization of the numerical resolution of quantum control problems [25]. In the general case, the decreasing character of the distance between the curves is weighted by the field fluence and the optimal couple of trajectories will be a tube whose nonzero width is related to the driving laser field fluence.

III. Wavefunction formulation

Similar considerations as introduced above apply to the wavefunction formulation. Note however that, even if the density matrix is more general than wavefunction, the associated observables are linear. For the wavefunction however, the observables enter into the cost functional as quadratic terms, which will induce some adaptations in the formalism. Let us consider the driving evolution equation:

$$\begin{aligned} &i\frac{\partial}{\partial t}\psi(t,x) = \left(H_0 - \epsilon(t)\mu\right)\psi(t,x) \qquad (23)\\ &\psi(t_0,x) = \psi_0(x). \end{aligned}$$

For a given observable A, its averaged measured value is $\langle\psi|A|\psi\rangle$. With this definition the tracking formulation can be written as above and the same considerations apply.

A. Monotonic algorithms

Let us define the cost functional

$$J^w(\epsilon) = \int_0^T \alpha\epsilon^2(t)dt - \langle\psi(T)|A|\psi(T)\rangle, \qquad (24)$$

We introduce as before the adjoint state (Lagrange multiplier) $\chi(x,t)$ and give one example of monotonic algorithm [18]:

$$\begin{aligned} &i\frac{\partial}{\partial t}\psi_{k+1}(x,t) = \left(H_0 - \epsilon_{k+1}(t)\mu\right)\psi_{k+1}(x,t) \qquad (25)\\ &\psi_{k+1}(x,t=0) = \psi_0(x)\\ &\epsilon_{k+1}(t) = -\frac{1}{\alpha}Re\langle\chi_k, \frac{\mu\psi_{k+1}}{i}\rangle(t) \qquad (26)\\ &i\frac{\partial}{\partial t}\chi_{k+1}(x,t) = \left(H_0 - \epsilon_{k+1}(t)\mu\right)\chi_{k+1}(x,t) \qquad (27)\\ &\chi_{k+1}(x,T) = A\psi_{k+1}(T). \end{aligned}$$

We note that the nonlinearity in the observable induces a dependence of the adjoint state on the final state $\psi_{k+1}(T)$.

B. Forward cost functional and equivalence

We introduce, for a reference field ϵ_{ref} on $[0,T]$ and a field $\epsilon(s)$ defined up to an intermediary time $t < T$ the "candidate" solution $\bar{\epsilon}$ as in (16). It is possible to consider

$$J^w(\bar{\epsilon}) = \int_0^T \alpha\bar{\epsilon}^2(t)dt - \langle\psi_{\bar{\epsilon}}(T)|A|\psi_{\bar{\epsilon}}(T)\rangle \qquad (28)$$

with $\psi_{\bar{\epsilon}}$ evolving from ψ_0 with field $\bar{\epsilon}$. However, due to the nonlinear nature of the observable, no efficient procedure is available to compute $J^w(\bar{\epsilon})$ at time t (other than explicitly computing $\psi_{\bar{\epsilon}}$ or working with the backward propagation of the observable A with the field ϵ_{ref}). For this reason, monotonic algorithms use the following inequality for a positive definite observable $A \geq 0$

$$-\langle a|A|a\rangle \leq -\langle b|A|b\rangle - 2Re\langle a-b|A|b\rangle, \qquad (29)$$

(the difference of two quantities being $-\langle a-b|A|a-b\rangle \leq 0$). In particular, if we denote by $\psi_{\epsilon_{ref}}$ the state evolving from ψ_0 with field ϵ_{ref}, one has

$$J^w(\bar{\epsilon}) \leq \int_0^T \alpha\bar{\epsilon}^2(t)dt - \langle\psi_{\epsilon_{ref}}(T)|A|\psi_{\epsilon_{ref}}(T)\rangle \\ +2Re\langle\psi_{\epsilon_{ref}}(T) - \psi_{\bar{\epsilon}}(T)|A|\psi_{\epsilon_{ref}}(T)\rangle \quad (30)$$

It is therefore possible to define the *(wavefunction) forward cost functional*

$$J^w_{fwd}(\epsilon,t;\epsilon_{ref}) = \int_0^T \alpha\bar{\epsilon}^2(t)dt - \langle\psi_{\epsilon_{ref}}(T)|A|\psi_{\epsilon_{ref}}(T)\rangle \\ +2Re\langle\psi_{\epsilon_{ref}}(T) - \psi_{\bar{\epsilon}}(T)|A|\psi_{\epsilon_{ref}}(T)\rangle \quad (31)$$

which can also be written

$$J^w_{fwd}(\epsilon,t;\epsilon_{ref}) = J^w(\epsilon_{ref}) + \int_0^t \alpha\left\{\epsilon^2(t) - {\epsilon_{ref}}^2(t)\right\}dt \\ +2Re\langle\psi_{\epsilon_{ref}}(T) - \psi_{\bar{\epsilon}}(T)|A|\psi_{\epsilon_{ref}}(T)\rangle. \quad (32)$$

To evaluate J^w_{fwd} it is convenient to note that

$$J^w_{fwd}(\epsilon,t;\epsilon_{ref}) = J^w(\epsilon_{ref}) + \int_0^t \alpha\left\{\epsilon^2(t) - {\epsilon_{ref}}^2(t)\right\}dt \\ +2Re\langle\psi_{\bar{\epsilon}}(t) - \psi_{\epsilon_{ref}}(t), \chi_{\epsilon_{ref}}(t)\rangle. \quad (33)$$

where $\chi_{\epsilon_{ref}}(t)$ is the adjoint state at time t given by

$$i\frac{\partial}{\partial t}\chi_{\epsilon_{ref}}(x,t) = \left(H_0 - \epsilon_{ref}(t)\mu\right)\chi_{\epsilon_{ref}}(x,t) \quad (34) \\ \chi_{\epsilon_{ref}}(x,T) = A\psi_{\epsilon_{ref}}(T).$$

This formula allows an efficient computation of $J^w_{fwd}(\epsilon,t;\epsilon_{ref})$. To summarize, the cost functional $J^w(\bar{\epsilon})$ that cannot be computed explicitly is exploited through its upper bound $J^w_{fwd}(\epsilon,t;\epsilon_{ref})$. The precise property of J^w_{fwd} is given in the following

Theorem 2: The monotonic algorithm (25)-(27) is a local tracking procedure for the forward cost functional $J^w_{fwd}(\epsilon_{k+1},t;\epsilon_{ref} = \epsilon_k)$ at any time t in the sense that $J^w_{fwd}(\epsilon_{k+1},t;\epsilon_k)$ is a monotonically decreasing function of t on the interval $[0,T]$.

Proof. As above, we evaluate the time derivative of $J^w_{fwd}(\epsilon_{k+1},t;\epsilon_k)$.

$$\frac{d}{dt}J^w_{fwd}(\epsilon_{k+1},t;\epsilon_k) = \alpha\epsilon^2_{k+1}(t) - \alpha\epsilon^2_k(t) \\ +2\frac{d}{dt}Re\langle\psi_{k+1}(t),\chi_k(t)\rangle = \alpha\epsilon^2_{k+1}(t) - \alpha\epsilon^2_k(t) \\ +2Re\langle\psi_{k+1}(t),\frac{d}{dt}\chi_k(t)\rangle + 2Re\langle\frac{d}{dt}\psi_{k+1}(t),\chi_k(t)\rangle \\ = \alpha\epsilon^2_{k+1}(t) - \alpha\epsilon^2_k(t) \\ +2Re\langle\psi_{k+1}(t),\frac{\left(H_0 - \epsilon_k(t)\mu\right)\chi_k(t)}{i}\rangle \\ +2Re\langle\frac{\left(H_0 - \epsilon_{k+1}(t)\mu\right)\psi_{k+1}(t)}{i},\chi_k(t)\rangle \\ = \alpha\epsilon^2_{k+1}(t) - \alpha\epsilon^2_k(t) + 2Re\langle\psi_{k+1}(t),\frac{\epsilon_k(t)\mu\chi_k(t)}{i}\rangle \\ +2Re\langle\frac{\epsilon_{k+1}(t)\mu\psi_{k+1}(t)}{i},\chi_k(t)\rangle \\ = \alpha\epsilon^2_{k+1}(t) - \alpha\epsilon^2_k(t) + 2\epsilon_k(t)\alpha\epsilon_{k+1}(t) \\ +2\epsilon_{k+1}(t)\alpha\epsilon_{k+1}(t) = -\alpha\left[\epsilon_{k+1}(t) - \epsilon_k(t)\right]^2. \quad (35)$$

Thus $J^w_{fwd}(\epsilon_{k+1},t;\epsilon_k)$ is a decreasing function of t.

Remark 4: Note that the inequality $J^w(\bar{\epsilon}) \leq J^w_{fwd}(\epsilon,t;\epsilon_{ref})$ becomes equality in the limit where ϵ approaches ϵ_{ref} which will happen at the convergence of the monotonic algorithms as $\epsilon_{k+1} - \epsilon_k \to 0$.

Remark 5: As above, the monotonicity of the procedure is a simple consequence of the decrease of $J^w_{fwd}(\epsilon_{k+1},t;\epsilon_k)$: $J^w(\epsilon_k) = J^w_{fwd}(\epsilon_{k+1},0;\epsilon_k) \geq J^w_{fwd}(\epsilon_{k+1},T;\epsilon_k) \geq J^w(\epsilon_{k+1})$.

IV. Conclusions

Through the introduction of a "forward cost functional" this paper demonstrates that the monotonic algorithms are closely related to the class of tracking procedures. As such, the monotonicity property of the former algorithms appears as a natural consequence of the increasing/decreasing properties of the tracking index. The monotonic schemes are shown to construct at all intermediary times the cost functional value of a current " best field" candidate and use this information in the open loop to optimize the field further. For the specific case of the density matrix, this can also be interpreted using two trajectories that start/end at the correct states and whose distance is continuously reduced (depending also on the laser fluence) during the optimization process.

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