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A cascadic monotonic time-discretized algorithm for finite-level quantum control computation ☆

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Abstract

A computer package (CNMS) is presented aimed at the solution of finite-level quantum optimal control problems. This package is based on a recently developed computational strategy known as monotonic schemes.

Quantum optimal control problems arise in particular in quantum optics where the optimization of a control representing laser pulses is required. The purpose of the external control field is to channel the system's wavefunction between given states in its most efficient way. Physically motivated constraints, such as limited laser resources, are accommodated through appropriately chosen cost functionals.

Program summary

Program title: CNMS
Catalogue identifier: ADEB_v1_0
Program summary URL: http://cpc.cs.qub.ac.uk/summaries/ADEB_v1_0.html
Program obtainable from: CPC Program Library, Queen's University, Belfast, N. Ireland
Licensing provisions: Standard CPC licence, http://cpc.cs.qub.ac.uk/licence/licence.html
No. of lines in distributed program, including test data, etc.: 770
No. of bytes in distributed program, including test data, etc.: 7098
Distribution format: tar.gz
Programming language: MATLAB 6
Computer: AMD Athlon 64 × 2 Dual, 2:21 GHz, 1:5 GB RAM
Operating system: Microsoft Windows XP
Word size: 32
Classification: 4.9
Nature of problem: Quantum control
Solution method: Iterative
Running time: 60–600 sec

PACS: 02.30.Yy; 07.05.Dz

Keywords: Quantum systems; Optimal control theory; Optimality conditions; Monotonic schemes

☆ This paper and its associated computer program are available via the Computer Physics Communications homepage on ScienceDirect (http://www.sciencedirect.com/science/journal/00104655).

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1. Introduction

The control of quantum phenomena in a variety of application systems [2,3,10,11,14] is receiving growing interest boosted by present and perspective applications that range from quantum optics and quantum chemistry to semiconductor nanostructures. There are indeed different strategies to tackle these problems and it is only recently that a common consensus has arisen towards the use of optimal control theory.

In the optimal control framework, one starts by defining the optimality criteria in the form of a cost functional. For a desired quantum-state transition, this functional will depend on the final target state and the need to add physically motivated constraints, e.g., limited laser resources. The strategy is then to minimize this cost functional while satisfying the constraints of the underlying dynamic equations governing the evolution of quantum states; e.g., the Schrödinger equation. The calculation of the necessary optimality conditions for this optimization problem results in a system of coupled equations to be solved.

The purpose of this paper is to present a computer package that solves the coupled equations representing the optimality system. This package implements the monotonic strategy that was initially introduced in a general framework by Krotov [4–6]. Following this approach, Tannor et al. [8] and then Zhu and Rabitz [13] have proposed two procedures for quantum control computation. These algorithms have a common basis and have developed to the modern monotonic schemes considered in this paper [7,9,11,13].

We present a time discretized monotonic scheme based on a Crank–Nicholson propagator. The resulting algorithm is unconditionally stable and works with a large range of time steps.

In the following section, we introduce the class of finite-level quantum optimal control problems considered in this paper. In Section 3 the related quantum optimal control problem is formulated and the first-order necessary conditions for a minimum are presented. The discretization of the resulting optimality system is illustrated in Section 3.1, where a monotonic scheme is discussed. To improve efficiency of this scheme, a cascadic acceleration procedure is proposed. Starting from Section 4, the structure and use of the program are discussed and results of numerical experiments are presented.

2. Basic principles

We consider finite-level quantum systems where the state of these systems is represented by a n-component wavefunction $\psi : [0, T] \to \mathbb{C}^n$. The dynamic of the wavefunction obeys the following time-dependent Schrödinger equation

$$\begin{cases} i\dot{\psi}(t) = H_0\psi(t) \quad \forall t \in (0, T], \\ \psi(0) = \psi_0, \end{cases}$$

where $\hbar$ is Planck's constant, $T > 0$ is a given terminal time, ψ_0 is the fixed initial state condition and $H_0 \in \mathbb{C}^{n\times n}$ is the free Hamiltonian describing the unperturbed (uncontrolled) system. Controlling a quantum system requires the introduction of external interactions. Here only the electric field of a laser is discussed, which has the form

$$C(t) = -\mu\epsilon(t), \tag{1}$$

where μ is the electric dipole operator, $\epsilon : [0, T] \to \mathbb{C}$ is the applied electric field. Now the Hamiltonian depends on the control field and therefore the new Hamiltonian is given by $H = H_0 + H_1$, where $H_1 : \mathbb{C} \to \mathbb{C}^{n\times n}$ models the coupling of the quantum state to the field. So we focus attention on

$$\begin{cases} i\dot{\psi}(t) = H(\epsilon(t))\psi(t) \quad \forall t \in (0, T], \\ \psi(0) = \psi_0, \end{cases} \tag{2}$$

where atomic units are used (i.e. $\hbar = 1$). Note that, strictly speaking, the wavefunction description given by (2) is only allowed for an isolated quantum system and in that case the governing Hamiltonian H is hermitian. For a non-isolated system with environment couplings and subject to control, the more general density-matrix description would be required [14]. Alternatively, we may follow the procedure outlined in [1] to construct a non-hermitian Hamiltonian H_0 accounting for environment losses. $H_1 : \mathbb{C} \to \mathbb{C}^{n\times n}$ is hermitian with

$$H_1(z) = z_{\mathrm{Re}}H_{1\,\mathrm{Re}} + z_{\mathrm{Im}}H_{1\,\mathrm{Im}}, \quad \text{for } z = z_{\mathrm{Re}} + iz_{\mathrm{Im}} \in \mathbb{C}, \tag{3}$$

where $H_{1\,\mathrm{Re}}, H_{1\,\mathrm{Im}} \in \mathbb{C}^{n\times n}$ are constant and $z_{\mathrm{Re}}, z_{\mathrm{Im}} \in \mathbb{R}$. Notice that ψ and ϵ are square summable functions in the appropriate Hilbert spaces. We define the inner product by

$$\langle \phi, \psi \rangle = \int_0^T \phi \cdot \psi^* \, dt,$$

where "$*$" means complex conjugate and the dot "$\cdot$" denotes the usual vector–scalar product in $\mathbb{C}^n$. Additionally we have $\|\cdot\| = \langle \cdot, \cdot \rangle^{1/2}$. For the initial condition holds $\|\psi_0\| = 1$, and by (2) in the case where H is hermitian, we have that the norm of the state is constant with respect to time:

$$\|\psi(t)\| = \|\psi_0\| \quad \text{for all } t > 0. \tag{4}$$

So the wavefunction $\psi(t)$ evolves on the unit sphere from the initial state ψ_0 to some final state $\psi(T)$.

3. Quantum optimal control problems

In the following we shall consider the problem of determining the control field ϵ, such that (2) is fulfilled and given objectives are met. The optimality criteria are given by

- the control sequence brings the system at time T close to the desired state $\psi_d \in \mathbb{C}^n$;
- limited laser resources result in a minimization of the control field strengths;
- reduction of environment losses need suppress population of intermediate dissipative states.

The optimal control approach seeks to optimize a cost functional J that includes the criteria given above under the constraint given by the evolution equation. In the following we discuss these modeling issues.

For systems with nonhermitian Hamiltonian we need to characterize dissipation with the assumption that

$$\left\langle \frac{H_0 - H_0^*}{i}\psi, \psi \right\rangle \leqslant 0 \quad \text{for all } \psi.$$

We define the operator

$$\Lambda\psi := -\sum_{j=1}^{n} \alpha_j \psi_j - \frac{H_0 - H_0^*}{i}\psi,$$

where the parameters $\alpha_j \geqslant 0$ fulfill the condition

$$-\sum_{j=1}^{n} \alpha_j |\psi_j|^2 - \left\langle \frac{H_0 - H_0^*}{i}\psi, \psi \right\rangle \geqslant 0 \quad \forall \psi \in \mathbb{C}^n. \tag{5}$$

With this assumption, the operator Λ is positive. Here, ψ_j denotes the jth component of the wavefunction ψ and the term $\alpha_j|\psi_j|^2$ penalizes the occupation of certain states ψ_j with $j \in I \subset \{1, \ldots, n\}$ where environment losses occur; $\alpha_j > 0$, $j \in I$, are weighting factors.

We require that the control sequence drives the system at time T close to the desired target state $\psi_d \in \mathbb{C}^n$. We account for limited laser resources through a minimization of the control field strengths, and we require to suppress population of intermediate states which suffer strong environment losses. All these requirements are realized considering the following cost functional

$$\widetilde{J}(\psi, \epsilon) = \mathrm{Re}\big(\psi_d^* \cdot \psi(T)\big) - \frac{\gamma}{2}\|\epsilon\|^2 + \frac{1}{2}\langle \psi, \Lambda\psi \rangle, \tag{6}$$

where the optimization weight $\gamma > 0$ is a regularization parameter that allows to vary the relative importance of limiting control field strengths. The goal of the first term of the cost functional is to track the state ψ to a given terminal state at $t = T$. The optimal control problem is given by the maximization of (6) under the constraint given by (2). We have

$$\begin{cases} \max_\epsilon \widetilde{J}(\psi, \epsilon) \\ \qquad i\dot{\psi}(t) = H(\epsilon(t))\psi(t), \\ \qquad \psi(0) = \psi_0. \end{cases} \tag{7}$$

A standard way to solve (7) is to use the method of Lagrange multipliers to turn the given constrained maximization problem into an unconstrained optimization problem. Introduce the Lagrange multiplier (also called adjoint variable) $q(t)$ and define the Lagrange function

$$\begin{aligned} L(\epsilon, \psi, q) = {} & \mathrm{Re}\big(\psi_d^* \cdot \psi(T)\big) - \frac{\gamma}{2}\|\epsilon\|^2 + \frac{1}{2}\langle \psi, \Lambda\psi \rangle \\ & - \mathrm{Re}\big\langle i\dot{\psi} - H\big(\epsilon(\cdot)\big)\psi, q \big\rangle. \end{aligned} \tag{8}$$

By formally equating to zero the Fréchet derivatives of L with respect to the triple (ϵ, ψ, q), we obtain the following optimality system characterizing the solution to the optimal control problem.

Derivative with respect to the adjoint variable gives

$$\begin{aligned} \frac{\partial L}{\partial q}\delta q &= \lim_{h\to 0} \frac{1}{h}\big\{L(\epsilon, \psi, q + h\delta q) - L(\epsilon, \psi, q)\big\} \\ &= -\mathrm{Re}\big\langle i\dot{\psi} - H\big(\epsilon(t)\big)\psi, \delta q \big\rangle = 0 \quad \forall \delta q. \end{aligned}$$

Thus, we obtain the state equation with given initial condition

$$\begin{cases} i\frac{\partial}{\partial t}\psi(t) = H(\epsilon(t))\psi(t) \quad \text{in } (0, T], \\ \psi(0) = \psi_0. \end{cases} \tag{9}$$

Derivative with respect to the state variable ψ gives

$$\begin{aligned} \frac{\partial L}{\partial \psi}\delta\psi &= \lim_{h\to 0} \frac{1}{h}\big\{L(\epsilon, \psi + h\delta\psi, q) - L(\epsilon, \psi, q)\big\} \\ &= \mathrm{Re}\big(\psi_d^* \cdot \delta\psi(T)\big) + \mathrm{Re}\langle \Lambda\psi, \delta\psi \rangle \\ &\quad - \mathrm{Re}\big\langle i\dot{q} - H\big(\epsilon(\cdot)\big)^* q, \delta\psi \big\rangle \\ &\quad + \mathrm{Re}\big(iq(T)\delta\psi(T)^*\big) = 0 \quad \forall \delta\psi. \end{aligned}$$

We obtain the adjoint equation with terminal condition

$$\begin{cases} i\frac{\partial}{\partial t}q(t) = H(\epsilon(t))^* q(t) + \Lambda\psi(t) \quad \text{in } [0, T), \\ iq(T) = -\psi_d. \end{cases} \tag{10}$$

Notice that this equation describes evolution backwards in time. To get the derivative of L with respect to the field ϵ we split the field in his real and imaginary part, which reads $\epsilon = \epsilon_{\mathrm{Re}} + i\epsilon_{\mathrm{Im}}$, with ϵ_{Re} and ϵ_{Im} being zero at $t = 0$ and $t = T$.

$$\begin{aligned} \frac{\partial L}{\partial \epsilon_{\mathrm{Re}}}\delta\epsilon_{\mathrm{Re}} &= \lim_{h\to 0} \frac{1}{h}\big\{L(\epsilon_{\mathrm{Re}} + h\delta\epsilon_{\mathrm{Re}}, \psi, q) - L(\epsilon_{\mathrm{Re}}, \psi, q)\big\} \\ &= -\gamma\,\langle \epsilon_{\mathrm{Re}}, \delta\epsilon_{\mathrm{Re}} \rangle + \mathrm{Re}\langle \delta\epsilon_{\mathrm{Re}} H_{1\,\mathrm{Re}}\psi, q \rangle = 0 \quad \forall \delta\epsilon_{\mathrm{Re}}. \end{aligned}$$

Analogously, we get

$$\frac{\partial L}{\partial \epsilon_{\mathrm{Im}}}\delta\epsilon_{\mathrm{Im}} = -\gamma\,\langle \epsilon_{\mathrm{Im}}, \delta\epsilon_{\mathrm{Im}} \rangle + \mathrm{Re}\langle \delta\epsilon_{\mathrm{Im}} H_{1\,\mathrm{Im}}\psi, q \rangle = 0 \quad \forall \delta\epsilon_{\mathrm{Im}}.$$

The following optimality condition equation results

$$\begin{cases} \epsilon(t) = \frac{1}{\gamma}\,\mathrm{Re}(H_{1\,\mathrm{Re}}\psi(t)q(t)^*) \\ \qquad\quad + i\,\frac{1}{\gamma}\,\mathrm{Re}(H_{1\,\mathrm{Im}}\psi(t)q(t)^*) \quad \text{in } (0, T), \\ \epsilon(T) = \epsilon(0) = 0. \end{cases} \tag{11}$$

Notice that the derivative of L with respect to the field ϵ is not needed for a monotonic scheme. Nevertheless it is computed, because we need the result for a proper convergence criterion in the algorithm.

Before presenting a monotonic scheme for the maximization of $\widetilde{J}(\epsilon)$, we need to compute the increase of this cost functional between two control fields. Consider two fields ϵ and ϵ', and the corresponding wavefunctions ψ and ψ', and Lagrange multipliers q and q', respectively. We get

$$\begin{aligned} \widetilde{J}(\epsilon') - \widetilde{J}(\epsilon) = {} & \mathrm{Re}\big\langle q, \big(H_1\big(\epsilon'(\cdot)\big) - H_1\big(\epsilon(\cdot)\big)\big)\psi' \big\rangle \\ & + \frac{1}{2}\big\langle \psi' - \psi, \Lambda(\psi' - \psi) \big\rangle \\ & - \frac{\gamma}{2}\big(\|\epsilon'\|^2 - \|\epsilon\|^2\big). \end{aligned}$$

This formula is the starting point for the design of monotonic algorithms. In fact, it reveals how to possibly modify ϵ' in such a way to maximize the cost functional.

3.1. Time discretized algorithm

Monotonic schemes require a choice of time discretization such that monotonicity at the discrete level is preserved. A particular choice of time discretization appropriate for our purpose is discussed in detail in this section. Assume that the interval $[0, T]$ has been discretized into a finite number N of subintervals of size δt, such that $N\delta t = T$. So for $l = 0, 1, \ldots, N$ we define $\epsilon_l = \epsilon(l\delta t)$, $\psi_l = \psi(l\delta t)$ and $q_l = q(l\delta t)$, which stand for the discretized control, state, and adjoint variables. We denote by H_l the approximation of the Hamiltonian $H_0 + H_1(\epsilon(l\delta t))$. To determine the evolution of the state variable (9) and the adjoint variable (10) we consider the implicit second order Crank–Nicholson scheme. The advantage of the Crank–Nicholson scheme is that it is unconditionally stable. Given the solution at the time step l, the value of the wave function at the next time step $l + 1$ is given by

$$i\frac{\psi_{l+1} - \psi_l}{\delta t} = \frac{1}{2}H_{l+1}\psi_{l+1} + \frac{1}{2}H_l\psi_l.$$

Thus ψ_{l+1} is given by

$$\psi_{l+1} = \left(I - \frac{\delta t H_{l+1}}{2i}\right)^{-1}\left(I + \frac{\delta t H_l}{2i}\right)\psi_l,$$

where I is the identity matrix. The formula above holds for the adjoint equation marching backwards by inverting the time direction.

For our algorithm we take a slightly different version of the presented scheme, which make demands on a fine enough discretization, because we exchange every H_{l+1} in H_l. Now, given an initial state ψ_0, we solve numerically (9) and (10) by using

$$\psi_{l+1} = \left(I - \frac{\delta t H_l}{2i}\right)^{-1}\left(I + \frac{\delta t H_l}{2i}\right)\psi_l \tag{12}$$

and

$$q_{l+1} = \left(I + \frac{\delta t H_l^*}{2i}\right)\left(I - \frac{\delta t H_l^*}{2i}\right)^{-1} q_l - i\delta t\Lambda\psi_{l+1},$$
$$iq_N = -\psi_d, \tag{13}$$

where I is the identity matrix. Notice that for the derivation of (13) we use that for any given matrix A the two terms $(I + A)$ and $(I - A)^{-1}$ commute.

We also introduce the time discretized cost functional

$$\widetilde{J}_{\delta t}(\epsilon) = \mathrm{Re}\big(\psi_d^* \cdot \psi_N\big) - \frac{\gamma\delta t}{2}\sum_{l=0}^{N-1}|\epsilon_l|^2 + \frac{\delta t}{2}\sum_{l=0}^{N-1}\big(\Lambda\psi_{l+1}^* \cdot \psi_{l+1}\big). \tag{14}$$

Consider two control fields ϵ and ϵ'. Repeating the computations of the previous section at the discrete level, we obtain

$$\widetilde{J}_{\delta t}(\epsilon') - \widetilde{J}_{\delta t}(\epsilon) = \sum_{l=0}^{N-1}\mathrm{Re}\big(q_l^* \cdot DH_l\psi_l'\big) - \frac{\gamma\delta t}{2}\big(|\epsilon_l'|^2 - |\epsilon_l|^2\big) + \frac{\delta t}{2}\sum_{l=0}^{N-1}\big((\psi_{l+1}' - \psi_{l+1})^*\Lambda(\psi_{l+1}' - \psi_{l+1})\big), \tag{15}$$

where

$$DH_l = i\left\{\left(I + \frac{\delta t H_l}{2i}\right)^{-1}\left(I - \frac{\delta t H_l}{2i}\right) \cdot \left(I - \frac{\delta t H_l'}{2i}\right)^{-1}\left(I + \frac{\delta t H_l'}{2i}\right) - I\right\}.$$

Now the monotonic scheme to optimize $\widetilde{J}_{\delta t}$ is reached via Newton iterations on the first sum of (15). Therefore the Taylor expansion with respect to $\delta\epsilon_l = \epsilon_l' - \epsilon_l = \delta\epsilon_{\mathrm{Re},l} + i\delta\epsilon_{\mathrm{Im},l}$ is computed:

$$\big(q_l^* \cdot DH_l\psi_l'\big) = \frac{\delta t}{2}\big(\tilde{q}_l^* \cdot \Delta H_l\tilde{\psi}_l'\big) - i\frac{\delta t^2}{4}\left[\tilde{q}_l^*\Delta H_l\left(I - \frac{\delta t H_l}{2i}\right)^{-1}\Delta H_l\tilde{\psi}_l'\right] + \mathrm{o}\big((\Delta H_l)^2\big),$$

where

$$\Delta H_l = H_1(\epsilon') - H_1(\epsilon),$$
$$\tilde{q}_l = \left(I + \left(I + \frac{\delta t H_l^*}{2i}\right)\left(I - \frac{\delta t H_l^*}{2i}\right)^{-1}\right)q_l = q_l + q_{l+1} + i\delta t\Lambda\psi_{l+1},$$
$$\tilde{\psi}_l' = \left(I - \frac{\delta t H_l}{2i}\right)^{-1}\psi_l'.$$

The increase can be expressed in terms of the quantity $\delta\epsilon_l$

$$\big(q_l^* \cdot DH_l\psi_l'\big) = \frac{\delta t}{2}A_l^T\begin{pmatrix}\delta\epsilon_{\mathrm{Re},l}\\ \delta\epsilon_{\mathrm{Im},l}\end{pmatrix} - \frac{\delta t}{4}\begin{pmatrix}\delta\epsilon_{\mathrm{Re},l}\\ \delta\epsilon_{\mathrm{Im},l}\end{pmatrix}^T \cdot (i\delta t B_l)\begin{pmatrix}\delta\epsilon_{\mathrm{Re},l}\\ \delta\epsilon_{\mathrm{Im},l}\end{pmatrix} + \mathrm{o}\big((\delta\epsilon)^2\big),$$

where

$$A_l = \begin{pmatrix}\tilde{q}_l^* \cdot H_{1\,\mathrm{Re}}\tilde{\psi}_l'\\ \tilde{q}_l^* \cdot H_{1\,\mathrm{Im}}\tilde{\psi}_l'\end{pmatrix}, \qquad B_l = \begin{pmatrix}B_{l,1,1} & B_{l,1,2}\\ B_{l,2,1} & B_{l,2,2}\end{pmatrix}$$

with

$$B_{l,1,1} = \tilde{q}_l^* \cdot H_{1\,\mathrm{Re}}\left(I - \frac{\delta t H_l}{2i}\right)^{-1}H_{1\,\mathrm{Re}}\tilde{\psi}_l',$$

$$B_{l,2,2} = \tilde{q}_l^* \cdot H_{1\,\mathrm{Im}}\left(I - \frac{\delta t H_l}{2i}\right)^{-1}H_{1\,\mathrm{Im}}\tilde{\psi}_l',$$

$$B_{l,1,2} = \frac{1}{2}\tilde{q}_l^* \cdot \left\{H_{1\,\mathrm{Im}}\left(I - \frac{\delta t H_l}{2i}\right)^{-1}H_{1\,\mathrm{Re}}\tilde{\psi}_l' + H_{1\,\mathrm{Re}}\left(I - \frac{\delta t H_l}{2i}\right)^{-1}H_{1\,\mathrm{Im}}\tilde{\psi}_l'\right\},$$

$$B_{l,2,1} = B_{l,1,2}.$$

On the other hand, one has

$$|\epsilon_l'|^2 - |\epsilon_l|^2 = 2\begin{pmatrix}\epsilon_{\mathrm{Re},l}\\ \epsilon_{\mathrm{Im},l}\end{pmatrix}^T\begin{pmatrix}\delta\epsilon_{\mathrm{Re},l}\\ \delta\epsilon_{\mathrm{Im},l}\end{pmatrix} + \begin{pmatrix}\delta\epsilon_{\mathrm{Re},l}\\ \delta\epsilon_{\mathrm{Im},l}\end{pmatrix}^T\begin{pmatrix}\delta\epsilon_{\mathrm{Re},l}\\ \delta\epsilon_{\mathrm{Im},l}\end{pmatrix},$$

and the increase of the cost functional reads

$$\begin{aligned}
&\widetilde{J}_{\delta t}(\epsilon') - \widetilde{J}_{\delta t}(\epsilon) \\
&\quad = \delta t \sum_{l=0}^{N-1} \left(\frac{1}{2} \operatorname{Re} A_l - \gamma \begin{pmatrix} \epsilon_{\mathrm{Re},l} \\ \epsilon_{\mathrm{Im},l} \end{pmatrix} \right)^T \begin{pmatrix} \delta\epsilon_{\mathrm{Re},l} \\ \delta\epsilon_{\mathrm{Im},l} \end{pmatrix} \\
&\qquad - \begin{pmatrix} \delta\epsilon_{\mathrm{Re},l} \\ \delta\epsilon_{\mathrm{Im},l} \end{pmatrix}^T \left(\frac{\gamma}{2} I - \frac{\delta t}{4} \operatorname{Im} B_l \right) \begin{pmatrix} \delta\epsilon_{\mathrm{Re},l} \\ \delta\epsilon_{\mathrm{Im},l} \end{pmatrix} + \mathrm{o}\big((\delta\epsilon_l)^2\big) \\
&\qquad + \frac{\delta t}{2} \sum_{l=0}^{N-1} \big((\psi'_{l+1} - \psi_{l+1})^* \cdot \Lambda(\psi'_{l+1} - \psi_{l+1}) \big).
\end{aligned}$$

Summarizing, we have the following

Algorithm (*Crank–Nicholson monotonic scheme (CNMS)*). Given an initial control amplitude ϵ^0 and its associated state ψ^0 and Lagrange multiplier q^0, suppose that ψ^k, q^k, ϵ^k, have already been computed. The derivation of $\psi^{k+1}, q^{k+1}, \epsilon^{k+1}$, is done as follows:

- **Forward propagation**: Given $\psi_0^{k+1} = \psi_0$, compute ψ_{l+1}^{k+1} from ψ_l^{k+1} by (1) Newton iteration for ϵ_l^{k+1} by
$$\begin{aligned}
\begin{pmatrix} \epsilon_{\mathrm{Re},l}^{k+1} \\ \epsilon_{\mathrm{Im},l}^{k+1} \end{pmatrix} &= \begin{pmatrix} \epsilon_{\mathrm{Re},l}^{k} \\ \epsilon_{\mathrm{Im},l}^{k} \end{pmatrix} + \frac{1}{2} \left(\frac{\gamma}{2} I - \frac{\delta t}{4} \operatorname{Im} B_l^k \right)^{-1} \\
&\quad \cdot \left(\frac{1}{2} \operatorname{Re} A_l^k - \gamma \begin{pmatrix} \epsilon_{\mathrm{Re},l}^{k} \\ \epsilon_{\mathrm{Im},l}^{k} \end{pmatrix} \right),
\end{aligned}$$
(2) compute ψ_{l+1}^{k+1} by (12) with the new field.
- **Backward propagation**:
Given $q_N^{k+1} = i\psi_d$, compute q_l^{k+1} form q_{l+1}^{k+1} by (13).

For the convergence of the algorithm we referee to read [7].

3.2. Cascadic acceleration

The motivation for combining the cascadic technique with the CNMS scheme comes from results given in [12] where a cascadic conjugate gradient method is discussed.

To illustrate the cascadic CNMS (CCNMS) scheme, consider a hierarchy of nested time grids with index $\ell = \ell_0, \ldots, \ell_f$. The idea is to start from a coarse grid, with index ℓ_0, where the size of the problem is small and therefore the problem can be solved by the CNMS iterative scheme with a reasonable computational effort. Let us denote with ϵ_{ℓ_0} the solution obtained by this process with, e.g., 'zero' initialization $\epsilon_{\ell_0}^*$. The step that follows is to interpolate this solution to the next finer grid, using an interpolation operator $I_\ell^{\ell+1}$. Therefore we obtain an initialization of the CNMS iterative process on the finer grid that is given by

$$\epsilon_{\ell+1}^* = I_\ell^{\ell+1} \epsilon_\ell,$$

where $\ell = \ell_0$. With this initialization, after a sufficient number of CNMS iterations we obtain the solution $\epsilon_{\ell+1}$. This process is repeated until the finest grid is reached and the desired solution is obtained.

An algorithm of this method is given by the following. Denote with $\epsilon_\ell = \mathrm{CNMS}_k(\epsilon_\ell^*)$ the result of the iteration, with ϵ_ℓ^* as initialization, that is applied until a given stopping criteria is satisfied. We have

Step 1. Given $\ell = \ell_0$, $\epsilon_{\ell_0}^*$.
Step 2. Compute $\epsilon_\ell = \mathrm{CNMS}_\ell(\epsilon_\ell^*)$.
Step 3. If $\ell = \ell_f$ then stop.
Step 4. Else if $\ell < \ell_f$ then interpolate $\epsilon_{\ell+1}^* = I_\ell^{\ell+1} \epsilon_\ell$.
Step 5. Set $\ell = \ell + 1$, goto Step 2.

4. Structure and use of the program

As already described in the previous section the program consists of two functions: the core (CNMS.m), which finds the solution for a chosen convergence criterion, and the cascadic approach (CCNMS.m), which calls the core with different gridsizes.

First of all the use of the two functions is presented. The syntax to call them is similarly for both. As a typical Matlab-function we have for the CNMS-code:

$$[\epsilon, \mathrm{Output}] = \mathrm{cnms}(H_0, H_{1\mathrm{half}}, \psi_0, \psi_d, T, n, \mathrm{options})$$

and for CCNMS-code:

$$[\epsilon, \mathrm{Output}] = \mathrm{ccnms}(H_0, H_{1\mathrm{half}}, \psi_0, \psi_d, T, \mathrm{grid}, \mathrm{options}).$$

On the right-hand side of the assignment the input parameters are defined by

Input	Description
H_0	free Hamiltonian
$H_{1\mathrm{half}}$	$H_1 := \epsilon \cdot H_{1\mathrm{half}} + (\epsilon \cdot H_{1\mathrm{half}})^*$ (upper tridiagonal part of H_1)
ψ_0	initial state for the wave-function
ψ_d	desired target state at time T
T	terminal time ($T > 0$)
n	size of the mesh ($n \cdot dt = T$)
grid	a vector: $[a, b]$, which stands for a...coarsest grid (gridsize=2^a) b...finest grid (gridsize=2^b)
options	different optional parameters

On the left side the output parameters are the following

Output	Description
ϵ	control field
output	different output-values and graphs

Notice that with the input parameter options important values, like convergence criteria, number of iterations, initial field, etc., can be set. When no options are given to the function common default values are used in the calculations. The output parameters include things like number of iterations, graphical informations of the solution and the convergence, warnings and many more.

For more detailed information about these parameters the helptext of the codes can be used. This text stand at the beginning of the code or can be displayed in the prompt-input-window in Matlab with the following entry:

```
>> help cnms
```

or

```
>> help ccnms
```

This help includes also a short description of the code and some examples.

The next section shows how to use the program based on a specific example.

5. Numerical experiments

The results presented in this section are computed using the » cnms and » ccnms schemes and the following convergence criteria

$$\|\nabla J_\epsilon\| \leqslant tol_{\text{grad}}. \tag{16}$$

We present numerical results of a Λ-type three-level quantum system [1], whose configuration is represented by $\psi = (\psi_1, \psi_2, \psi_3)$ which consists of two long-lived states ψ_1 and ψ_2, which are energetically separated by some amount δ, and a state ψ_3, which has a finite lifetime because of environment coupling. Within this scheme, the system's time evolution is governed by the effective Hamiltonian

$$H_0 = \frac{1}{2}\begin{pmatrix} -\delta & 0 & 0 \\ 0 & \delta & 0 \\ 0 & 0 & -i\Gamma_\ell \end{pmatrix},$$

where the term $-i\Gamma_\ell$ accounts for environment losses (e.g., spontaneous photon emissions). Furthermore, the coupling to the external field reads

$$H_\epsilon = -\frac{1}{2}\begin{pmatrix} 0 & 0 & \mu_1\epsilon \\ 0 & 0 & \mu_2\epsilon \\ \mu_1\epsilon^* & \mu_2\epsilon^* & 0 \end{pmatrix}, \tag{17}$$

where μ_1 and μ_2 describe the coupling strengths of state ψ_1 and ψ_2 to the interconnection state ψ_3 (e.g., optical dipole matrix elements). Initial and final states are given by

$$\psi_0 = \begin{pmatrix} 1 \\ 0 \\ 0 \end{pmatrix} \quad \text{and} \quad \psi_d = \begin{pmatrix} 0 \\ e^{-i\delta T} \\ 0 \end{pmatrix},$$

respectively. Concerning the coupling to the external field and with (3) we get

$$H_{1\,\text{Re}} = -\frac{1}{2}\begin{pmatrix} 0 & 0 & \mu_1 \\ 0 & 0 & \mu_2 \\ \mu_1 & \mu_2 & 0 \end{pmatrix}$$

and

$$H_{1\,\text{Im}} = -\frac{1}{2}\begin{pmatrix} 0 & 0 & i\mu_1 \\ 0 & 0 & i\mu_2 \\ -i\mu_1 & -i\mu_2 & 0 \end{pmatrix}.$$

The parameters for this problem are held fixed. We choose $\delta = 20$, $\Gamma_\ell = 0.01$, $\mu_1 = 1$, $\mu_2 = 1$ and $T = 5$.

The input of this required data can be done in Matlab as follows:

```
>>H_0=[-10,0,0;0,10,0;0,0,-j*0.005];
>>H_1half=[0,0,-0.5;0,0,-0.5;0,0,0];
>>psi_0=[1;0;0];
>>psi_d=[0;exp(-i*5*20);0];
>>T=5;
>>n=1024;
```

After initializing this data the Λ-type three-level quantum optimal control problem can be solved by using the CNMS-scheme to obtain the control field ϵ as follows

```
>>field=cnms(H_0,H_1half,psi_0,psi_d,T,n);
```

where n stands for the gridsize used in the calculation. Notice that we have not set parameters like for convergence, maximum number of iterations, weighting factors and many more. The most important parameters (with their default values) are shown in the following table:

Input	Default	Explanation
gamma0	1e–3	weighting parameter γ
max_it	10000	max. number of iterations
tol	1e–5	tolerance for $\frac{\lvert J^{k+1}-J^k\rvert}{J^k}$
tol2	–	tolerance for (16)
alpha	$\begin{pmatrix} 0 \\ \vdots \\ 0 \end{pmatrix}$	weighting factors α
q	0	initial value for the Lagrange multiplier

Here we use *gamma*0 instead of *gamma* because the latter is a reserved word in MATLAB. At the input, to define any parameter one should write the given name for the parameter followed by the chosen value. For example,

```
'gamma0',0.1,'tol',1e-4,'alpha',
0.0,0.0,0.1,
```

The next table shows results about the sensitivity of the solution on the required tolerance for the gradient. We find that smaller values of tolerance my result in very close values of the cost functional. However, sufficient accuracy is required for accurate determination of the control field.

$\frac{\lvert J^{k+1}-J^k\rvert}{J^k}$	$\|\nabla J_\epsilon\|$	$\lvert\psi(T)-\psi_d\rvert$	CPU
10^{-4}	$6.0551\cdot 10^{-5}$	0.056	9
10^{-6}	$6.1303\cdot 10^{-6}$	0.056	15.1
10^{-8}	$6.0967\cdot 10^{-7}$	0.055	20.8
10^{-10}	$6.1121\cdot 10^{-8}$	0.055	26.9
10^{-12}	$6.2468\cdot 10^{-9}$	0.055	32.6
10^{-14}	$5.0535\cdot 10^{-10}$	0.055	39

To obtain exemplary the first line of this table corresponding to $tol = $ 1e–4, and $\gamma = 0.1$ and $n = 1024$, we have to use the function like

```
>>[field,cost,grad_J,d_psi]=cnms(H_0, ...
H_1half,psi_0,psi_d,T,1024, ...
'output',[0,0,1,0,1,0,0,1,0],'gamma0', ...
0.1,'tol',1e-4);
```

By doing so, after the calculation has been completed, one can obtain the desired values at the MATLAB prompt. For example, typing *cost* the CPU time is obtained. Typing *grad_J*(*end*) the final value of $\|\nabla J_\epsilon\|$ is provided, etc.

One can see that an extended output is obtained simply by listing the required return-values. The 'output' is a vector with the following components:

$$\begin{pmatrix} \text{iter} \\ \text{q} \\ \text{cost} \\ \text{dJ} \\ \text{grad\_J} \\ \text{psi\_out} \\ \text{graph} \\ \text{d\_psi} \\ \text{warning} \end{pmatrix} = \begin{matrix} \text{number of iterations} \\ \text{Langrange multiplier at time 0} \\ \text{CPU-time} \\ \text{cost functional at each iteration} \\ \|\nabla J_\epsilon\| \text{at each iteration} \\ \text{wave function } \psi \\ \text{name of the output-graph file} \\ |\psi(T) - \psi_d| \\ \text{different warnings} \end{matrix}$$

Notice that the output-values are ordered with the position in the output-vector.

The next table shows results with different grid-sizes and different tolerances. We now choose $\gamma = 0.01$.

tol_{grad}	n	J	iter	CPU
10^{-3}	1024	0.922	22	7.9
	2048	0.892	11	7.9
10^{-4}	1024	0.968	299	107
	2048	0.964	196	140
	4096	0.952	70	100
	8192	0.934	28	80.2
10^{-5}	1024	0.971	1129	406.2
	2048	0.971	859	615.9
	4096	0.970	521	745.9
	8192	0.969	258	740.3
	16384	0.967	196	1119.9

By taking the γ very small the control problem becomes more ill-conditioned. This effect is reported in the next table ($n = 4096$; $tol_{\text{grad}} = 10^{-4}$).

γ	$\lvert\psi(T) - \psi_d\rvert$	J	iter	CPU
10^{-2}	$8.3753 \cdot 10^{-4}$	0.952	70	100
10^{-3}	$8.7350 \cdot 10^{-5}$	0.987	286	410.3
10^{-4}	$2.5760 \cdot 10^{-5}$	0.987	231	330.7
10^{-5}	$6.4863 \cdot 10^{-5}$	0.953	4417	6437.2

Now we want to compare the computation cost of the cnms algorithm with the cascadic version ($\gamma = 10^{-3}$).

tol_{grad}	n	CPU (CNMS)	CPU (CCNMS)
10^{-4}	8192	310.4	54.3
10^{-4}	16384	349.0	39.6
10^{-3}	8192	20.1	11.7
10^{-3}	16384	17.6	23.1

We see that the cascadic version of the algorithm provides a computational improvement.

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