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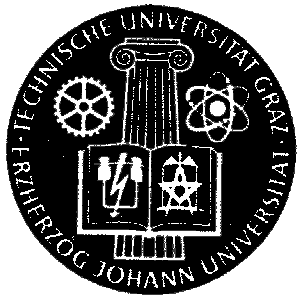

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Cascadic non-linear conjugate gradient solution to finite-level quantum optimal control problems*

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Abstract

Optimal control of finite-level quantum systems is studied and iterative solution scheme for the optimization of a control representing laser pulses are developed. The purpose of this external field is to channel the system's wavefunction between given states in its most efficient way. Physically motivated constraints, such as limited laser resources or population suppression of certain states, are accounted for through an appropriately chosen cost functional. First-order necessary optimality conditions and second-order sufficient optimality conditions are investigated. For solving the optimal control problems, a cascadic non-linear conjugate gradient scheme and a monotonic scheme are discussed. Results of numerical experiments with a representative finite-level quantum system demonstrate the effectiveness of the optimal control formulation and efficiency and robustness of the proposed approaches.

Keywords: Quantum systems, optimal control theory, optimality conditions, monotonic schemes, cascadic acceleration, non-linear conjugate gradient method.

AMS: 35Q40, 49A10, 49K10, 49M05, 65M06, 81V80.

1 Introduction

Nowadays we witness a large growing interest in controlling quantum phenomena in a variety of application systems [10, 17, 30, 31, 32, 33]. Present and perspective applications range from quantum optics and quantum chemistry to semiconductor nanostructures. In the last few years these research areas have received further impetus from the emerging fields of quantum computation and quantum communication [8], aiming at quantum devices where there is the need to manipulate wavefunctions with highest possible precision.

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This high-fidelity quantum-state engineering can only be achieved putting together the most sophisticated experimental and theoretical techniques for control of quantum systems. However, within each field of application one has come up with its own strategies and it is only recently that a common consensus has arisen towards the use of optimal control theory [11, 14, 22].

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 state, the need to suppress population of certain states during the control process, as well as other 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.

While we focus on quantum optimal control problems we argue that many of the results of this paper can be extended to general time-dependent bilinear control problems. Bilinear systems [13, 26] were introduced in the theory of automatic control in the 1960's for electrical engineering applications. They represent a class of non-linear control strategies with the aim to obtain better system response than possible with linear control. In general, the solution of most bilinear systems poses challenging theoretical and computational problems which are open or have been only partially addressed. This is in particular true for the control of the quantum mechanical systems discussed in this paper.

The purpose of this paper is to present a detailed formulation of a class of optimal control problems for finite-level quantum systems and to address their solution by iterative methods. We prove existence of solutions to the optimal control problems and investigate first-order necessary optimality conditions and second-order sufficient optimality conditions. We review state-of-the-art monotonic iterative schemes and their convergence properties and use these schemes as benchmark for an alternative solution procedure that we propose in this paper. This procedure results from combining an appropriate extension of a newly proposed non-linear conjugate gradient method with a cascadic acceleration scheme. Convergence of the proposed non-linear conjugate gradient method is proved and its competitiveness in efficiency and robustness is demonstrated by results of numerical experiments.

In the following section we introduce the class of finite-level quantum optimal control problems considered in this paper. Within an appropriate functional analytical setting, existence of optimal solution is proved. We discuss existence of Lagrange multiplier and first-order necessary conditions for a minimum. Second-order sufficient optimality conditions are also discussed that allow to characterize local minima.

In Section 3, the proposed non-linear conjugate-gradient scheme is formulated. Under appropriate less restrictive assumptions, we prove convergence of this scheme to a local minimizer. This scheme is embedded in a cascadic iteration to obtain almost optimal computational complexity.

To validate the computational performance of our cascadic non-linear conjugate-gradient scheme comparison with efficient monotonic schemes is presented showing that the former may outperform the latter. For completeness and for comparison of theoretical aspects, in Section 4 a detailed review of newly proposed monotonic schemes is given.

In Section 5, results of an extended set of numerical experiments are given. We

show that the optimal solution is quite sensitive to the required tolerance of the norm of the gradient. We therefore discuss additional convergence criteria involving the order of accuracy of solutions. Further numerical experiments demonstrate efficiency of the cascadic non-linear conjugate-gradient approach and its robustness with respect to change of values of the optimization parameters.

A section of conclusion completes the exposition of our work.

2 Quantum optimal control problems

This section is devoted to the formulation of a class of finite-level quantum optimal control problems, which are the subject of our investigations. Existence of solutions to the optimal control problems is proved. We investigate first-order necessary optimality conditions addressing existence and regularity issues of the Lagrange multipliers and, correspondingly, of the control functions. To verify that the solution to the first-order optimality conditions corresponds to a local minimum, second-order sufficient optimality conditions are discussed.

2.1 The minimization problem

We consider localized finite-level quantum systems modeled by a Schrödinger equation for an n-component wave function $\psi : [0,T] \to \mathbb{C}^n$ as follows

$$i\dot{\psi}(t) = H(\epsilon(t))\psi(t) \text{ for } t \in (0,T] \quad \text{and} \quad \psi(0) = \psi_0, \tag{1}$$

where $T > 0$ is a given terminal time, $H : \mathbb{C} \to \mathbb{C}^{n\times n}$ denotes the Hamiltonian matrix depending on the external control field $\epsilon : [0,T] \to \mathbb{C}$ and $\psi_0 \in \mathbb{C}^n$ is a fixed initial condition. The Hamiltonian $H = H_0 + H_1$ has two constitutive components: The constant free Hamiltonian $H_0 \in \mathbb{C}^{n\times n}$ describing the unperturbed (uncontrolled) system; and $H_1 : \mathbb{C} \to \mathbb{C}^{n\times n}$ modeling the coupling of the quantum state to an external control field ϵ.

The choice of T is a modeling issue motivated by physical considerations. Based on the quantum indeterminacy principle $\Delta E\, \Delta t \geq \hbar$ (where $\hbar$ is the Planck constant that we set equal to one) we can state the following: For too small T a highly energetic optimal control results ($\Delta E \gg 1$) thus involving many energy levels. On the other hand, for too large T additional decoherence channels become important, which should be avoided; see, e.g., [17]. As a guideline, the choice of T should be related to the transition frequency.

Strictly speaking, the wavefunction description given in (1) is appropriate 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 [36]. Alternatively, we may follow the procedure outlined in [6] to construct a non-hermitian Hamiltonian H_0 accounting for environment losses.

We focus on localized quantum systems where $H_1 : \mathbb{C} \to \mathbb{C}^{n\times n}$ is hermitian and possesses the following structure

$$H_1(z) = z_{\Re e}\, H_{1\Re e} + z_{\Im m}\, H_{1\Im m} \quad \text{for } z = z_{\Re e} + i z_{\Im m} \in \mathbb{C}, \tag{2}$$

where $H_{1\Re e}$, $H_{1\Im m} \in \mathbb{C}^{n\times n}$ are constant and $z_{\Re e}$, $z_{\Im m} \in \mathbb{R}$. Consequently,

$$\varrho(H_1(z)) \leq K_0\, |z| \quad \text{for all } z \in \mathbb{C}, \tag{3}$$

where $\varrho(\cdot)$ denotes the spectral norm of complex-valued $n \times n$ matrices, $|\cdot|$ stands for the absolut value of complex numbers and $K_0 = 2\max\{\varrho(H_{1\Re e}), \varrho(H_{1\Im m})\} \geq 0$. Moreover, by (3),

$$\varrho(H(z)) \leq \varrho(H_0) + K_0\,|z| \quad \text{for all } z \in \mathbb{C}.$$

We call $\psi : [0,T] \to \mathbb{C}^n$ a *solution to* (1) if ψ belongs to $H^1(0,T;\mathbb{C}^n)$, $\psi(0) = \psi_0$ holds and ψ satisfies $i\dot{\psi} = H(\epsilon(\cdot))\psi$ in $[0,T]$ almost everywhere (a.e.). For the notion of Sobolev spaces we refer the reader to [1], for instance. Notice that (1) is a linear system of ordinary differential equations, which therefore admits a unique solution $\psi \in H^1(0,T;\mathbb{C}^n)$ for every $\epsilon \in L^2(0,T;\mathbb{C})$. The Hilbert space $L^2(0,T;\mathbb{C}^n)$ is endowed with the canonical inner product

$$\langle \phi, \psi \rangle_{L^2(0,T;\mathbb{C}^n)} = \int_0^T \phi \cdot \psi^* \,\mathrm{d}t \quad \text{for } \phi,\, \psi \in L^2(0,T;\mathbb{C}^n),$$

where '*' means complex conjugate and the dot '$\cdot$' denotes the usual vector-scalar product in $\mathbb{C}^n$. We also have $\|\cdot\|_{L^2(0,T;\mathbb{C}^n)} = \langle \cdot\,, \cdot \rangle^{1/2}$. Analogously, the inner product and the corresponding induced norm are defined on $H^1(0,T;\mathbb{C}^n)$.

To write (1) in a compact form we define the Hilbert space

$$Y = L^2(0,T;\mathbb{C}^n) \times \mathbb{C}^n$$

endowed with the common product topology and introduce the non-linear operator $e = (e_1, e_2)$ by

$$e : H^1(0,T;\mathbb{C}^n) \times L^2(0,T;\mathbb{C}) \to Y, \quad (\psi, \epsilon) \mapsto \begin{pmatrix} i\dot{\psi} - H(\epsilon(\cdot))\psi \\ \psi(0) - \psi_0 \end{pmatrix}.$$

Recall $H^1(0,T;\mathbb{C}^n)$ is continuously (even compactly) embedded in $C([0,T];\mathbb{C}^n)$, where $C([0,T];\mathbb{C}^n)$ denotes the Banach space of all continuous functions $\varphi : [0,T] \to \mathbb{C}^n$ that is endowed with the common norm; see [1]. Therefore, there exists an embedding constant $K_1 > 0$ satisfying

$$\|\psi\|_{C([0,T];\mathbb{C}^n)} \leq K_1 \,\|\psi\|_{H^1(0,T;\mathbb{C}^n)} \quad \text{for all } \psi \in H^1(0,T;\mathbb{C}^n). \tag{4}$$

Let $(\psi, \epsilon) \in H^1(0,T;\mathbb{C}^n) \times L^2(0,T;\mathbb{C})$. Then we infer from (3) that

$$|H(\epsilon(t))\psi(t)|^2_{\mathbb{C}^n} \leq \varrho\big(H(\epsilon(t))\big)^2 |\psi(t)|^2_{\mathbb{C}^n} \leq \big(2\varrho(H_0)^2 + 2K_0^2\,|\epsilon(t)|^2\big)\,\|\psi\|^2_{C([0,T];\mathbb{C}^n)}$$

for almost all $t \in [0,T]$. Thus, $H(\epsilon(\cdot))\psi$ belongs to $L^2(0,T;\mathbb{C}^n)$ and, therefore, e_1 is well-defined.

In the following we shall consider the problem of determining a control field $\epsilon \in L^2(0,T;\mathbb{C})$, such that (1) is fulfilled and a number of optimality criteria are met. We assert that the control sequence drives the system at time T to the desired target state $\psi_d \in \mathbb{C}^n$. We account for limited laser resources and increased smoothness through a minimization of the control field strengths and we require to suppress population of intermediate states which suffer strong environment losses, thus also enforcing our modelling choice of a non-hermitian free Hamiltonian. All these requirements are realized in the cost functional

$$\begin{aligned} J(\psi,\epsilon) \;&= \frac{1}{2}\,|\psi(T) - \psi_d|^2_{\mathbb{C}^n} + \frac{\gamma}{2}\,\|\epsilon\|^2_{L^2(0,T;\mathbb{C})} + \frac{\mu}{2}\,\|\dot{\epsilon}\|^2_{L^2(0,T;\mathbb{C})} \\ &\quad + \frac{1}{2}\sum_{j\in I} \alpha_j\,\|\psi_j\|^2_{L^2(0,T,\mathbb{C})}, \end{aligned} \tag{5}$$

where the constants γ, $\mu \geq 0$ are regularization parameters, which allow to vary the relative importance of the objectives represented by the various terms. We suppose that $\gamma + \mu > 0$. In the last term of (5), which penalizes the occupation of certain states ψ_j, the set $I \subset \{1, \dots, n\}$ denotes a subset of possible state indices and $\alpha_j > 0$, $j \in I$, are weighting factors. Depending on the parameter μ, we define the Hilbert space X by

$$X = \begin{cases} H^1(0,T;\mathbb{C}^n) \times L^2(0,T;\mathbb{C}) & \text{if } \mu = 0, \\ H^1(0,T;\mathbb{C}^n) \times H_0^1(0,T;\mathbb{C}) & \text{if } \mu > 0 \end{cases}$$

and supply X with the natural product topology. Then, the optimal control problem can be written as the following abstract minimization problem

$$\min J(x) \quad \text{subject to} \quad x = (\psi, \epsilon) \in X \text{ and } e(x) = 0 \text{ in } Y. \tag{P}$$

Remark 1 1) Because of H^1 regularization ($\mu > 0$) we have a natural setting to impose zero (boundary) conditions on the control field outside of the control window $[0,T]$. This requirement is in agreement with the need of designing control pulses with compact support.

2) Recall the Poincaré inequality

$$\|\epsilon\|_{L^2(0,T;\mathbb{C})} \leq K_2 \|\dot{\epsilon}\|_{L^2(0,T;\mathbb{C})} \quad \text{for all } \epsilon \in H_0^1(0,T;\mathbb{C}). \tag{6}$$

Therefore, if $\mu > 0$ the L^2-norm of ϵ can be bounded by the L^2-norm of its derivative. This implies that we can choose $\gamma = 0$ provided $\mu > 0$ holds.

The next theorem ensures that (**P**) has at least one global solution.

Theorem 2 *The optimal control problem* (**P**) *admits a solution* $(\psi, \epsilon) \in X$.

Proof. We prove the claim by two steps.
Step 1: We need an a-priori estimate for the solution of (1). For given $\epsilon \in L^2(0,T;\mathbb{C})$, equation (1) is a linear system of ordinary differential equations, which therefore has a unique solution $\psi \in H^1(0,T;\mathbb{C}^n)$. We now write (1) in integral form

$$i\psi(t) = i\psi_0 + \int_0^t \big(H_0 + H_1(\epsilon(s))\big)\psi(s)\,\mathrm{d}s \quad \text{for } 0 \leq t \leq T. \tag{7}$$

Taking the Euclidean norm in $\mathbb{C}^n$ on both sides of (7) and using the triangle inequality on the right-hand side results in

$$|\psi(t)|_{\mathbb{C}^n} \leq |\psi_0|_{\mathbb{C}^n} + \int_0^t \Big(\varrho(H_0) + \varrho\big(H_1(\epsilon(s))\big)\Big)|\psi(s)|_{\mathbb{C}^n}\,\mathrm{d}s.$$

Now apply Gronwall's inequality [14] to obtain

$$|\psi(t)|_{\mathbb{C}^n} \leq |\psi_0|_{\mathbb{C}^n} \exp\left(\int_0^t \Big(\varrho(H_0) + \varrho\big(H_1(\epsilon(s))\big)\Big)\,\mathrm{d}s\right) \quad \text{for } 0 \leq t \leq T.$$

Using (3), squaring and integrating the above inequality over $[0,T]$ results in

$$\|\psi\|_{L^2(0,T;\mathbb{C}^n)} \leq \sqrt{T}\,|\psi_0|_{\mathbb{C}^n} \exp\Big(c_1 + c_2 \|\epsilon\|_{L^2(0,T;\mathbb{C})}\Big) \quad \text{for } 0 \leq t \leq T \tag{8}$$

with $c_1 = T\varrho(H_0)$ and $c_2 = \sqrt{T}K_0$. Furthermore, using the state equation (1) yields that for every $\epsilon \in L^2(0,T;\mathbb{C})$ with $\|\epsilon\|_{L^2(0,T;\mathbb{C})} \leq c_3$ the corresponding states $\psi = \psi(\epsilon)$ are bounded in $H^1(0,T;\mathbb{C}^n)$, i.e.,

$$\|\psi\|_{H^1(0,T;\mathbb{C}^n)} \leq c_4 \quad \text{for some } c_4 > 0.$$

Step 2: Let $\{\epsilon_k\}_{k\geq 1}$ be a minimizing sequence for J, i.e.,

$$\lim_{k\to\infty} J(\psi_k, \epsilon_k) = \inf\big\{J(x)\,|\,x = (\psi,\epsilon) \in X \text{ and } e(x) = 0 \text{ in } Y\big\},$$

where we denote by $\psi_k = \psi(\epsilon_k)$ the unique solution to (1) for $\epsilon = \epsilon_k$. Let $\gamma > 0$ and $\mu = 0$ hold. Hence,

$$J(\psi(\epsilon),\epsilon) \to \infty \quad \text{as} \quad \|\epsilon\|_{L^2(0,T;\mathbb{C})} \to \infty,$$

so that the sequence $\{\epsilon_k\}_{k\geq 1}$ is bounded in $L^2(0,T;\mathbb{C})$. Since the unit ball in a Hilbert space is weakly compact, there exists a weakly to an $\epsilon \in L^2(0,T;\mathbb{C})$ convergent subsequence, which we again denote by $\{\epsilon_k\}_{k\geq 1}$. Step 1 above ensures that the corresponding sequence $\{\psi_k\}_{k\geq 1}$ is bounded in $H^1(0,T;\mathbb{C}^n)$; thus, again by choosing a proper subsequence

$$\psi_k \rightharpoonup \psi \text{ in } H^1(0,T;\mathbb{C}^n),$$

it follows from the Sobolev embedding theorem [1] that

$$\psi_k \to \psi \text{ in } L^2(0,T;\mathbb{C}^n) \text{ and in } C([0,T];\mathbb{C}^n).$$

We can now show that (ψ,ϵ) is a solution of the optimal control problem. Since ψ_k solves (1) for ϵ_k we have

$$i\psi_k(t) = i\psi_0 + \int_0^t \big(H_0 + H_1(\epsilon_k(s))\big)\psi_k(s)\,\mathrm{d}s \quad \text{for } 0 \leq t \leq T. \tag{9}$$

Next, we consider the limit in $k \to \infty$ of (9). The weak convergence of ϵ_k to ϵ in $L^2(0,T;\mathbb{C})$ implies weak convergence also for the complex conjugates, i.e., $\epsilon_k^* \rightharpoonup \epsilon^*$ in $L^2(0,T;\mathbb{C})$. Strong convergence of ψ_k to ψ in $L^2(0,T;\mathbb{C}^n)$ allows to go to the limit as $k \to \infty$ on the right-hand side of (9). Thus we find

$$i\psi(t) = i\psi_0 + \int_0^t \big(H_0 + H_1(\epsilon(s))\big)\psi(s)\,\mathrm{d}s,$$

which shows that $\psi = \psi(\epsilon)$, or equivalently that $e(\psi,\epsilon) = 0$ in Y. We finally obtain

$$\begin{aligned}
J(\psi,\epsilon) &= \frac{1}{2}|\psi(T) - \psi_d|^2_{\mathbb{C}^n} + \frac{\gamma}{2}\|\epsilon\|^2_{L^2(0,T;\mathbb{C})} + \frac{1}{2}\sum_{j\in I}\alpha_j\|\psi_j\|^2_{L^2(0,T;\mathbb{C}^n)} \\
&\leq \frac{1}{2}\lim_{k\to\infty}|\psi_k(T) - \psi_d|^2_{\mathbb{C}^n} + \frac{1}{2}\liminf_{k\to\infty}\Big(\gamma\|\epsilon_k\|^2_{L^2(0,T;\mathbb{C})}\Big) \\
&\quad + \frac{1}{2}\sum_{j\in I}\alpha_j\lim_{k\to\infty}\|\psi_{k,j}\|^2_{L^2(0,T;\mathbb{C}^n)} \\
&= \inf\big\{J(x)\,|\,x = (\psi,\epsilon) \in X \text{ and } e(x) = 0 \text{ in } Y\big\},
\end{aligned}$$

where we used the lower-semicontinuity of the L^2-norm. Thus we have proved that (ψ,ϵ) is a solution to (**P**) for $\gamma > 0$ and $\mu = 0$. If $\mu > 0$ and $\gamma = 0$ hold the sequence $\{\epsilon_k\}_{k\geq 1}$ is bounded in $L^2(0,T;\mathbb{C})$ by the Poincaré inequality (6). Thus, the proof follows by analogous arguments. □

2.2 First-order necessary optimality conditions

To solve the above optimal control problem, we use the method of Lagrange multipliers (see, e.g., [22]) to turn the constrained minimization problem (**P**) into an unconstrained optimization problem, and we focus on the necessary optimality conditions of first-order.

We define the Lagrangian function $L : X \times Y \to \mathbb{R}$ by

$$\begin{aligned} L(\psi,\epsilon,p,q) &= J(\psi,\epsilon) + \Re e\big(\langle e(\psi,\epsilon),(p,p_0)\rangle_Y\big) \\ &= J(\psi,\epsilon) + \Re e\left(\langle i\dot{\psi} - H(\epsilon(\cdot))\psi, p\rangle_{L^2(0,T;\mathbb{C}^n)} + (\psi(0)-\psi_0)\cdot q^*\right). \end{aligned}$$

To derive first-order necessary optimality conditions we need the following constraint qualification.

Proposition 3 *The operator $e : X \to Y$ is Fréchet-differentiable and its linearization $\nabla e(x_\circ)$ is surjective for every $x_\circ = (\psi_\circ, \epsilon_\circ) \in X$.*

Proof. We first prove the claim for the case $\mu > 0$. Let $x_\circ = (\psi_\circ, \epsilon_\circ) \in X = H^1(0,T;\mathbb{C}^n) \times H^1(0,T;\mathbb{C})$ be arbitrary. Recall that $H_0^1(0,T;\mathbb{C})$ is continuously embedded into $C([0,T];\mathbb{C})$ so that there exists an embedding constant $c_1 > 0$ satisfying

$$\|\epsilon\|_{C([0,T];\mathbb{C})} \le c_1 \|\epsilon\|_{H^1(0,T;\mathbb{C})} \quad \text{for all } \epsilon \in H_0^1(0,T;\mathbb{C}). \tag{10}$$

Recall that the Hamiltonian matrix H is of the form (2). We compute the directional derivative of the operator e. For any direction $x = (\psi,\epsilon) \in X$, $\epsilon = \epsilon_{\Re e} + i\epsilon_{\Im m}$ with $\epsilon_{\Re e}$, $\epsilon_{\Im m} \in H_0^1(0,T;\mathbb{R})$, we infer from (2) that

$$\nabla e(x_\circ)x = \lim_{h \searrow 0} \frac{e(x_\circ + hx) - e(x_\circ)}{h} = \begin{pmatrix} i\dot{\psi} - H(\epsilon_\circ(\cdot))\psi - H_1(\epsilon(\cdot))\psi_\circ \\ \psi(0) \end{pmatrix}.$$

Next we prove that the directional derivative is already the Fréchet-derivative. Note that

$$e(x_\circ + x) - e(x_\circ) - \nabla e(x_\circ)x = \begin{pmatrix} H_1(\epsilon(\cdot))\psi \\ 0 \end{pmatrix}.$$

Thus, we derive from (10), $|\epsilon_{\Re e}(t)| \le |\epsilon(t)|$ and $|\epsilon_{\Im m}(t)| \le |\epsilon(t)|$

$$\begin{aligned} &\big\|e(x_\circ + x) - e(x_\circ) - \nabla e(x_\circ)x\big\|_Y^2 \\ &\quad\le K_0^2 \int_0^T |\epsilon(t)|^2 \, |\psi(t)|_{\mathbb{C}^n}^2 \, \mathrm{d}t \le K_0^2 \|\epsilon\|_{C([0,T];\mathbb{C})}^2 \|\psi\|_{L^2(0,T;\mathbb{C}^n)}^2 \\ &\quad\le c_2 \left(\|\psi\|_{H^1(0,T;\mathbb{C}^n)}^2 + \|\epsilon\|_{H^1(0,T;\mathbb{C})}^2\right)^2 = c_2 \|x\|_X^4, \end{aligned}$$

where $c_2 = K_0^2/2$. Consequently,

$$0 \le \lim_{\|x\|_X \searrow 0} \frac{\|e(x_\circ + x) - e(x_\circ) - \nabla e(x_\circ)x\|_Y}{\|x\|_X} \le c_2 \lim_{\|x\|_X \searrow 0} \|x\|_X = 0$$

so that the directional derivative $\nabla e(x_\circ)$ is already the Fréchet-derivative.
Next we prove that the linear operator $e'(x_\circ)$ is surjective for every $x_\circ = (\psi_\circ, \epsilon_\circ) \in X$. Recall that $\psi_\circ \in C([0,T];\mathbb{C}^n)$. Let $(f, f_0) \in Y$ be arbitrary. Then $\nabla e(x_\circ)(\psi,\epsilon) = (f, f_0)$ is equivalent with

$$i\dot{\psi} = H(\epsilon_\circ(\cdot))\psi + H_1(\epsilon(\cdot))\psi_\circ + f \text{ in } (0,T] \quad \text{and} \quad \psi(0) = f_0. \tag{11}$$

Applying (3) and (4), the right-hand side $g = H_1(\epsilon(\cdot))\psi_\circ + f$ belongs to $L^2(0,T;\mathbb{C}^n)$ for every $\epsilon \in L^2(0,T;\mathbb{C})$. Thus, (11) is a linear system of ordinary differential equations that admits a unique solution $\psi \in H^1(0,T;\mathbb{C}^n)$ for every $\epsilon \in L^2(0,T;\mathbb{C})$. In particular, the operator $\nabla e(x_\circ)$ is surjective. □

Remark 4 It follows from the proof of Proposition 3 that the linear operator $e_\psi(x_\circ) : H^1(0,T;\mathbb{C}^n) \to Y$ is bijective, where $e_\psi(x_\circ)$ denotes the partial Fréchet-derivative at $x_\circ$ with respect to $\psi \in H^1(0,T;\mathbb{C}^n)$.

Notice that the quadratic cost functional $J : X \to [0,\infty)$ is twice continuously Fréchet-differentiable. Using Proposition 3 first-order necessary conditions for a minimum are obtained by equating to zero the Fréchet derivatives of L with respect to (ψ, ϵ, p, q).

Theorem 5 *Suppose that $x_\circ = (\psi_\circ, \epsilon_\circ) \in X$ is a local solution to (**P**). Then there exist (unique) Lagrange multipliers $p_\circ \in H^1(0,T;\mathbb{C}^n)$ and $q_\circ \in \mathbb{C}^n$ satisfying*

$$
\begin{aligned}
i\dot{\psi}_\circ &= H(\epsilon_\circ(\cdot))\psi_\circ && \text{in } (0,T], && \text{(12a)}\\
\psi_\circ(0) &= \psi_0, && && \text{(12b)}\\
i(\dot{p}_\circ)_j &= \left(H(\epsilon_\circ(\cdot))^* p_\circ\right)_j - \alpha_j(\psi_\circ)_j && \text{in } (0,T],\ j \in I, && \text{(12c)}\\
i(\dot{p}_\circ)_j &= \left(H(\epsilon_\circ(\cdot))^* p_\circ\right)_j && \text{in } (0,T],\ j \notin I, && \text{(12d)}\\
ip(T) &= \psi(T) - \psi_d, && && \text{(12e)}\\
q_\circ &= ip_\circ(0), && && \text{(12f)}\\
-\mu\ddot{\epsilon}_\circ + \gamma\epsilon_\circ &= \Re e\left(H_{1\Re e}\psi_\circ \cdot p_\circ^*\right) + i\Re e\left(H_{1\Im m}\psi_\circ \cdot p_\circ^*\right) && \text{in } (0,T], && \text{(12g)}\\
\epsilon_\circ(T) = \epsilon_\circ(0) &= 0. && && \text{(12h)}
\end{aligned}
$$

in case of $\mu > 0$. Moreover, $\epsilon_\circ \in C^2([0,T];\mathbb{C}) \cap C([0,T];\mathbb{C})$, i.e., $\epsilon_\circ$ is a classical solution. If $\mu = 0$ holds, (12g) and (12h) have to be replaced by

$$
\gamma\epsilon_\circ = \Re e\left(H_{1\Re e}\psi_\circ \cdot p_\circ^*\right) + i\Re e\left(H_{1\Im m}\psi_\circ \cdot p_\circ^*\right) \quad \text{in } (0,T). \qquad \text{(12g')}
$$

Proof. It follows from Proposition 3 that there exist (unique) Lagrange multipliers $(p_\circ, q_\circ) \in Y$ satisfying

$$
\nabla L(x_\circ, p_\circ, q_\circ) = 0 \quad \text{in } X \times Y. \qquad (13)
$$

The condition $L_p(x_\circ, p_\circ, q_\circ) = 0$ in $L^2(0,T;\mathbb{C}^n)$ implies (12a), whereas the equation $L_q(x_\circ, p_\circ, q_\circ) = 0$ in $\mathbb{C}^n$ yields (12b). Next we turn to the partial derivative of the Lagrangian with respect to ψ. Let $\phi \in H^1(0,T;\mathbb{C}^n)$ be arbitrary. Then we find

$$
\begin{aligned}
L_\psi(x_\circ, p_\circ, q_\circ)\phi \quad &= \Re e\left((\psi_\circ(T) - \psi_d)\cdot\phi(T)^* + \sum_{j\in I}\alpha_j\left\langle(\psi_\circ)_j, \phi_j\right\rangle_{L^2(0,T;\mathbb{C}^n)}\right)\\
&\quad + \Re e\left(\left\langle i\dot{\phi} - H(\epsilon(\cdot))\phi, p_\circ\right\rangle_{L^2(0,T;\mathbb{C}^n)} + \phi(0)\cdot q_\circ^*\right) \overset{!}{=} 0.
\end{aligned} \qquad (14)
$$

Using integration by parts we obtain

$$
\begin{aligned}
\Re e\left(\left\langle i\dot{\phi} - H(\epsilon(\cdot))\phi, p_\circ\right\rangle_{L^2(0,T;\mathbb{C}^n)}\right) &= \Re e\left(\left\langle i\dot{p} - H(\epsilon(\cdot))^* p_\circ, \phi\right\rangle_{H^1(0,T;\mathbb{C}^n)',H^1(0,T;\mathbb{C}^n)}\right)\\
&\quad + \Re e\left(ip_\circ(T)\cdot\phi(T)^* - ip_\circ(0)\cdot\phi(0)^*\right),
\end{aligned}
$$

where $\langle\cdot\,,\cdot\rangle_{H^1(0,T;\mathbb{C}^n)',H^1(0,T;\mathbb{C}^n)}$ stands for the dual pairing of $H^1(0,T;\mathbb{C}^n)$ and its dual space $H^1(0,T;\mathbb{C}^n)'$. We infer from (14) that (12c)–(12d) are satisfied in $H^1(0,T;\mathbb{C}^n)'$. From (3) and the fact that $\psi_\circ, p_\circ \in L^2(0,T;\mathbb{C}^n)$, it follows that the right-hand sides in (12c)–(12d) even belong to $L^2(0,T;\mathbb{C}^n)$. Hence, $p_\circ \in H^1(0,T;\mathbb{C}^n)$ and (12c)–(12d) hold in $L^2(0,T;\mathbb{C}^n)$. Inserting (12c)–(12d) in (14) implies (12e)–(12f). In case of $\mu > 0$, we conclude from $L_{\epsilon_{\Re e}}(x_\circ, p_\circ, q_\circ)\epsilon_{\Re e} = 0$ for all $\epsilon = \epsilon_{\Re e} + i\epsilon_{\Im m} \in H^1(0,T;\mathbb{C})$ that

$$\gamma\,\langle \epsilon_{\circ\Re e}, \epsilon_{\circ\Re e}\rangle_{L^2(0,T;\mathbb{R})} + \mu\,\langle \dot\epsilon_{\circ\Re e}, \dot\epsilon_{\circ\Re e}\rangle_{L^2(0,T;\mathbb{R})} + \Re e\left(\int_0^T \epsilon_{\Re e} H_{1\Re e}\psi_\circ \cdot p_\circ^*\right) = 0$$

with $\epsilon_\circ = \epsilon_{\circ\Re e} + i\epsilon_{\circ\Im m} \in H_0^1(0,T;\mathbb{C})$ and $\epsilon_{\circ\Re e}, \epsilon_{\circ\Im m} \in H_0^1(0,T;\mathbb{R})$. Applying integration by parts we derive

$$\langle \dot\epsilon_{\circ\Re e}, \dot\epsilon_{\Re e}\rangle_{L^2(0,T;\mathbb{R})} = -\langle \ddot\epsilon_{\circ\Re e}, \epsilon_{\Re e}\rangle_{H^1(0,T;\mathbb{R})',H^1(0,T;\mathbb{R})}.$$

Hence,

$$\gamma\,\epsilon_{\circ\Re e} - \mu\,\ddot\epsilon_{\circ\Re e} + \Re e\big(H_{1\Re e}\psi_\circ \cdot p_\circ^*\big) = 0. \tag{15}$$

Analogously, $L_{\epsilon_{\Im m}}(x_\circ, p_\circ, q_\circ)\epsilon_{\Im m} = 0$ for all $\epsilon = \epsilon_{\Re e} + i\epsilon_{\Im m} \in H^1(0,T;\mathbb{C})$ implies that

$$\gamma\,\epsilon_{\circ\Im m} - \mu\,\ddot\epsilon_{\circ\Im m} + \Re e\big(H_{1\Im m}\psi_\circ \cdot p_\circ^*\big) = 0. \tag{16}$$

Multiplying (16) by the complex unit i and adding (15) we obtain (12g), whereas (12h) follows directly from $\epsilon_\circ \in H_0^1(0,T;\mathbb{C})$. Since $\psi_\circ, p_\circ \in C([0,T];\mathbb{C}^n)$, the right-hand side in (12g) belongs to $C([0,T];\mathbb{C})$. Thus, $\epsilon_\circ$ is a classical solution. In case of $\mu = 0$ we have to replace (15)-(16) by

$$\gamma\,\epsilon_{\circ\Re e} + \Re e\big(H_{1\Re e}\psi_\circ \cdot p_\circ^*\big) = 0 \quad \text{and} \quad \gamma\,\epsilon_{\circ\Im m} + \Re e\big(H_{1\Im m}\psi_\circ \cdot p_\circ^*\big) = 0,$$

respectively, so that we derive (12g'). □

Having computed a solution to the first-order conditions (12) it remains to verify that this solution corresponds to a local minimum. For this reason, second-order optimality conditions are considered in the next section.

2.3 Second-order sufficient optimality conditions

Suppose that $x_\circ = (\psi_\circ, \epsilon_\circ) \in X$ is an optimal solution to (**P**) and that $(p_\circ, q_\circ) \in Y$ are the associated Lagrange multiplier satisfying (12c)-(12f). The second Fréchet derivative of the Lagrangian at $(x_\circ, p_\circ, q_\circ)$ with respect to x is given by

$$\begin{aligned} &L_{xx}(x_\circ, p_\circ, q_\circ)(x,x) \\ &\quad = |\psi(T)|_{\mathbb{C}^n}^2 + \gamma\,\|\epsilon\|_{L^2(0,T;\mathbb{C})}^2 + \mu\,\|\dot\epsilon\|_{L^2(0,T;\mathbb{C})}^2 + \sum_{j\in I}\alpha_j\,\|\psi_j\|_{L^2(0,T;\mathbb{C}^n)}^2 \\ &\qquad - 2\Re e\left(\int_0^T \big(H_1(\epsilon(t))\psi(t)\big)\cdot p_\circ(t)^*\,\mathrm{d}t\right) \end{aligned}$$

for every direction $x = (\psi, \epsilon) \in X$.

In case of $\mu > 0$ the second-order sufficient optimality conditions for (**P**) are as follows (see, e.g., [25]): There exists a constant $\kappa > 0$ such that

$$L_{xx}(x_\circ, p_\circ, q_\circ)\big((\psi,\epsilon),(\psi,\epsilon)\big) \geq \kappa\,\left(\|\psi\|_{H^1(0,T;\mathbb{C}^n)}^2 + \|\epsilon\|_{H^1(0,T;\mathbb{C})}^2\right) \tag{17}$$

for all $(\psi, \epsilon) \in X$ satisfying the linearized state equation

$$i\dot{\psi} = H(\epsilon_\circ(\cdot))\psi + H_1(\epsilon(\cdot))\psi_\circ \text{ in } (0,T] \quad \text{and} \quad \psi(0) = 0. \tag{18}$$

It follows from Proposition 3 and Remark 4 that for every $\epsilon \in L^2(0,T;\mathbb{C})$ the problem (18) possesses a unique solution.

In case of $\mu = 0$ we have to replace (17) by

$$L_{xx}(x_\circ, p_\circ, q_\circ)\big((\psi,\epsilon),(\psi,\epsilon)\big) \geq \kappa \left(\|\psi\|^2_{H^1(0,T;\mathbb{C}^n)} + \|\epsilon\|^2_{L^2(0,T;\mathbb{C})} \right). \tag{19}$$

To prove the second-order sufficient optimality condition we need the following two lemmas.

Lemma 6 *Let $(\tilde{\psi}_\circ, \tilde{\epsilon}_\circ) \in L^2(0,T;\mathbb{C}^n) \times L^2(0,T;\mathbb{C})$ represent the linearization point and let $(\psi,\epsilon) \in H^1(0,T;\mathbb{C}^n) \times L^2(0,T;\mathbb{C})$ satisfy (18). Then there exists a constant $C > 0$ depending on $\tilde{\psi}_\circ$ and $\tilde{\epsilon}_\circ$ so that*

$$\|\psi\|_{L^\infty(0,T;\mathbb{C}^n)} + \|\psi\|_{L^2(0,T;\mathbb{C}^n)} \leq C \|\epsilon\|_{L^2(0,T;\mathbb{C})} \quad \text{for almost all } t \in (0,T]. \tag{20}$$

Proof. To prove the assertion we apply Gronwall's lemma. From (18) we have

$$i\psi(t) = i\psi(0) + \int_0^t i\dot{\psi}(s)\,\mathrm{d}s = \int_0^t \big(H_0 + H_1(\tilde{\epsilon}_\circ(s))\big)\,\psi(s) + H_1(\epsilon(s))\tilde{\psi}_\circ(s)\,\mathrm{d}s$$

for almost all $t \in (0,T]$. Using (3) we find

$$\begin{aligned} |\psi(t)|_{\mathbb{C}^n} &\leq \int_0^t \big(\varrho(H_0) + K_0\,|\tilde{\epsilon}_\circ(s)|\big)|\psi(s)|_{\mathbb{C}^n} + K_0\,|\epsilon(s)|\,|\tilde{\psi}_\circ(s)|_{\mathbb{C}^n}\,\mathrm{d}s \\ &\leq C_1\big(1 + \|\tilde{\epsilon}_\circ\|^2_{L^2(0,T;\mathbb{C})}\big)^{1/2}\left(\int_0^t |\psi(s)|^2_{\mathbb{C}^n}\,\mathrm{d}s\right)^{1/2} + C_2\,\|\epsilon\|_{L^2(0,T;\mathbb{C})}, \end{aligned}$$

where $C_1 = \max\{2T\,\varrho(H_0)^2, 2K_0^2\}$ and $C_2 = K_0\,\|\tilde{\psi}_\circ\|_{L^2(0,T;\mathbb{C}^n)}$. Consequently,

$$|\psi(t)|^2_{\mathbb{C}^n} \leq C_3\left(\big(1 + \|\tilde{\epsilon}_\circ\|^2_{L^2(0,T;\mathbb{C})}\big)\int_0^t |\psi(s)|^2_{\mathbb{C}^n}\,\mathrm{d}s + \|\epsilon\|^2_{L^2(0,T;\mathbb{C})}\right)$$

with $C_3 = \max\{2C_1^2, 2C_2^2\}$. Applying Gronwall's lemma and using $\psi(0) = 0$ we obtain

$$|\psi(t)|^2_{\mathbb{C}^n} \leq C_3\,\|\tilde{\psi}_\circ\|^2_{L^2(0,T;\mathbb{C}^n)}\,e^{C_3 t\left(1+\|\tilde{\epsilon}_\circ\|^2_{L^2(0,T;\mathbb{C})}\right)}\,\|\epsilon\|^2_{L^2(0,T;\mathbb{C})} \tag{21}$$

for almost all $t \in (0,T]$ and

$$\|\psi\|^2_{L^2(0,T;\mathbb{C}^n)} \leq TC_3\,\|\tilde{\psi}_\circ\|^2_{L^2(0,T;\mathbb{C}^n)}\,e^{C_3 T\left(1+\|\tilde{\epsilon}_\circ\|^2_{L^2(0,T;\mathbb{C})}\right)}\,\|\epsilon\|^2_{L^2(0,T;\mathbb{C})}. \tag{22}$$

From (21) and (22) we infer (20). □

Remark 7 If in addition to the hypotheses of Lemma 6, we have $\tilde{\psi}_\circ \in L^\infty(0,T;\mathbb{C}^n)$ then we can give an estimate for ψ in the $H^1(0,T;\mathbb{C}^n)$-norm. Applying (3), it follows

that

$$\begin{aligned}
\|\dot{\psi}\|^2_{L^2(0,T;\mathbb{C}^n)} &= \int_0^T \Big|\big(H_0 + H_1(\tilde{\epsilon}_\circ(t)\big)\psi(t) + H_1(\epsilon(s))\tilde{\psi}_\circ(t)\Big|^2 \,\mathrm{d}t \\
&\leq \int_0^T \Big|\big(\varrho(H_0) + K_0\,|\tilde{\epsilon}_\circ(t)|\big)\,|\psi(t)|_{\mathbb{C}^n} + K_0\,|\epsilon(t)|\,|\tilde{\psi}_\circ(t)|_{\mathbb{C}^n}\Big|^2 \,\mathrm{d}t \\
&\leq \int_0^T 2\big(\varrho(H_0) + K_0\,|\tilde{\epsilon}_\circ(t)|\big)^2\,|\psi(t)|^2_{\mathbb{C}^n} + 2K_0^2\,|\epsilon(t)|^2\,|\tilde{\psi}_\circ(t)|^2_{\mathbb{C}^n}\,\mathrm{d}t \\
&\leq 4\,\varrho(H_0)^2\,\|\psi\|^2_{L^2(0,T;\mathbb{C}^n)} + 4K_0^2\,\|\psi\|^2_{L^\infty(0,T;\mathbb{C}^n)}\,\|\tilde{\epsilon}_\circ\|^2_{L^2(0,T;\mathbb{C})} \\
&\quad + 2K_0^2\,\|\tilde{\psi}_\circ\|^2_{L^\infty(0,T;\mathbb{C}^n)}\|\epsilon\|^2_{L^2(0,T;\mathbb{C})} \\
&\leq C_4\left(\|\psi\|^2_{L^\infty(0,T;\mathbb{C}^n)} + \|\psi\|^2_{L^2(0,T;\mathbb{C}^n)} + \|\epsilon\|^2_{L^2(0,T;\mathbb{C})}\right)
\end{aligned}$$

with

$$C_4 = \max\Big\{4\varrho(H_0)^2,\, 4K_0^2\,\|\tilde{\epsilon}_\circ\|^2_{L^2(0,T;\mathbb{C})},\, 2K_0^2\,\|\tilde{\psi}_\circ\|^2_{L^\infty(0,T;\mathbb{C}^n)}\Big\}.$$

Thus, it follows from Lemma 6 that

$$\|\psi\|_{H^1(0,T;\mathbb{C}^n)} \leq C_5\,\|\epsilon\|_{L^2(0,T;\mathbb{C})} \tag{23}$$

for a constant $C_5 > 0$ depending on $\|\psi_\circ\|_{L^\infty(0,T;\mathbb{C}^n)}$ and $\|\epsilon_\circ\|_{L^2(0,T;\mathbb{C})}$.

Lemma 8 *Suppose that $(\psi_\circ, \epsilon_\circ)$ is a local solution to (**P**). Let $\alpha = (\alpha_i) \in \mathbb{R}^n$ be given by $\alpha_i > 0$ for $i \in I$ and $\alpha_i = 0$ otherwise. Then the solution $p_\circ \in H^1(0,T;\mathbb{C}^n)$ to (12c)–(12e) satisfies*

$$|p_\circ(t)|_{\mathbb{C}^n} \leq C\left(|\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \|D_\alpha\psi_\circ\|_{L^1(0,T;\mathbb{C}^n)}\right) \quad \textit{for almost all } t \in [0,T],$$

where the constant $C > 0$ depends on $\epsilon_\circ$ and D_α denotes a diagonal matrix satisfying $D_\alpha = \operatorname{diag}(\alpha_1, \ldots, \alpha_n) \in \mathbb{R}^{n\times n}$.

Proof. From (12c)–(12d) we infer

$$\big(ip_\circ(T) - ip_\circ(t)\big)_j = \left(\int_t^T \big(H_0^* + H_1(\epsilon(s))^*\big)p_\circ(s) - \alpha_j(\psi_\circ(s))_j\,\mathrm{d}s\right)_j$$

for almost all $t \in [0,T)$ and $1 \leq j \leq n$. Using (3) and $i\,p_\circ(T) = \psi_\circ(T) - \psi_d$ it follows that

$$\begin{aligned}
|p_\circ(t)|_{\mathbb{C}^n} &\leq |\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \int_t^T \big(\varrho(H_0) + K_0\,|\epsilon(s)|\big)|p_\circ(s)|_{\mathbb{C}^n} + |D_\alpha\psi_\circ(s)|_{\mathbb{C}^n}\,\mathrm{d}s \\
&\leq |\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \|D_\alpha\psi_\circ\|_{L^1(0,T;\mathbb{C}^n)} \\
&\quad + \tilde{C}_1\left(1 + \|\epsilon_\circ\|^2_{L^2(0,T;\mathbb{C})}\right)^{1/2}\left(\int_t^T |p_\circ(s)|^2_{\mathbb{C}^n}\,\mathrm{d}s\right)^{1/2},
\end{aligned}$$

where $\tilde{C}_1 = \max\{2T\,\varrho(H_0)^2, 2K_0^2\}$. Setting $\tilde{C}_2 = 4\max\{1, 2C_1^2\}$ we get

$$\begin{aligned}
&|p_\circ(t)|^2_{\mathbb{C}^n} \leq \\
&\tilde{C}_2\left(|\psi_\circ(T) - \psi_d|^2_{\mathbb{C}^n} + \|D_\alpha\psi_\circ\|^2_{L^1(0,T;\mathbb{C}^n)} + \left(1 + \|\epsilon_\circ\|^2_{L^2(0,T;\mathbb{C})}\right)\int_t^T |p_\circ(s)|^2_{\mathbb{C}^n}\,\mathrm{d}s\right).
\end{aligned}$$

Applying Gronwall's lemma it follows that

$$|p_\circ(t)|^2_{\mathbb{C}^n} \leq \tilde{C}_2 e^{\tilde{C}_2(T-t)\left(1+\|\epsilon_\circ\|^2_{L^2(0,T;\mathbb{C})}\right)} \left(|\psi_\circ(T) - \psi_d|^2_{\mathbb{C}^n} + \|D_\alpha \psi_\circ\|^2_{L^1(0,T;\mathbb{C}^n)} \right)$$

for almost all $t \in [0,T)$, which gives the assertion. □

Now we turn to the second-order sufficient optimality conditions. Let $x_\circ = (\psi_\circ, \epsilon_\circ) \in X$ be a local solution to (**P**) and $(p_\circ, q_\circ) \in X$ the associated pair of Lagrange multipliers. Suppose that $\mu > 0$ and $x = (\psi, \epsilon) \in X$ satisfy (18). Due to Remark 7 and (6) there exists a constant $\tilde{C} > 0$ such that

$$\|\dot{\epsilon}\|^2_{L^2(0,T;\mathbb{C})} \geq \frac{1}{\tilde{C}} \|\psi\|^2_{H^1(0,T;\mathbb{C}^n)}. \tag{24}$$

By Lemma 8 there exists a constant $\hat{C} > 0$ depending on $\epsilon_\circ$ such that

$$|p_\circ(t)|_{\mathbb{C}^n} \leq \hat{C} \left(|\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \|D_\alpha \psi_\circ\|_{L^1(0,T;\mathbb{C}^n)} \right) \quad \text{for all } t \in [0,T]. \tag{25}$$

From (3), (6), (24), and (25) we derive

$$\begin{aligned}
&L_{xx}(x_\circ, p_\circ, q_\circ)(x,x) \\
&\geq \gamma \|\epsilon\|^2_{L^2(0,T;\mathbb{C})} + \frac{\mu}{2} \|\dot{\epsilon}\|^2_{L^2(0,T;\mathbb{C})} + \frac{\mu}{2\tilde{C}} \|\psi\|^2_{H^1(0,T;\mathbb{C}^n)} \\
&\quad - 2K_0 \int_0^T |\epsilon(t)|\, |\psi(t)|_{\mathbb{C}^n} |p_\circ(t)|_{\mathbb{C}^n} \, \mathrm{d}t \\
&\geq \left(\gamma + \frac{\mu}{4K_2} \right) \|\epsilon\|^2_{L^2(0,T;\mathbb{C})} + \frac{\mu}{4} \|\dot{\epsilon}\|^2_{L^2(0,T;\mathbb{C})} + \frac{\mu}{2\tilde{C}} \|\psi\|^2_{H^1(0,T;\mathbb{C}^n)} \\
&\quad - \hat{C}K_0 \left(|\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \|D_\alpha \psi_\circ\|_{L^1(0,T;\mathbb{C}^n)} \right) \left(\|\epsilon\|^2_{L^2(0,T;\mathbb{C})} + \|\psi\|^2_{H^1(0,T;\mathbb{C}^n)} \right) \\
&\geq \min \left\{ \gamma + \frac{\mu}{4K_2} - \hat{C}K_0 \left(|\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \|D_\alpha \psi_\circ\|_{L^1(0,T;\mathbb{C}^n)} \right), \frac{\mu}{4} \right\} \|\epsilon\|^2_{H^1(0,T;\mathbb{C})} \\
&\quad + \left(\frac{\mu}{2\tilde{C}} - \hat{C}K_0 \left(|\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \|D_\alpha \psi_\circ\|_{L^1(0,T;\mathbb{C}^n)} \right) \right) \|\psi\|^2_{H^1(0,T;\mathbb{C}^n)}.
\end{aligned}$$

If

$$|\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \|D_\alpha \psi_\circ\|_{L^1(0,T;\mathbb{C}^n)} < \min \left\{ \frac{4\gamma K_2 + \mu}{4\hat{C}K_0K_2}, \frac{\mu}{2\tilde{C}\hat{C}K_0} \right\} \tag{26}$$

then

$$\begin{aligned}
\kappa = \min \Bigg\{ &\gamma + \frac{\mu}{4K_2} - \hat{C}K_0 \left(|\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \|D_\alpha \psi_\circ\|_{L^1(0,T;\mathbb{C}^n)} \right), \frac{\mu}{4}, \\
&\frac{\mu}{2\tilde{C}} - \hat{C}K_0 \left(|\psi_\circ(T) - \psi_d|_{\mathbb{C}^n} + \|D_\alpha \psi_\circ\|_{L^1(0,T;\mathbb{C}^n)} \right) \Bigg\}
\end{aligned}$$

is positive and (17) holds. In case of $\mu = 0$ we replace (24) by (23). Then, the second-order sufficient condition can be shown analogously to the case $\mu > 0$. We summarize the results in the following theorem.

Theorem 9 *Suppose that $x_\circ = (\psi_\circ, \epsilon_\circ) \in X$ is an optimal solution to (**P**) and that $(p_\circ, q_\circ) \in Y$ are the associated (unique) Lagrange multipliers satisfying (12c)-(12f). If (26) holds, then the second-order sufficient condition (17) is satisfied.*

Remark 10 The second-order sufficient optimality condition holds provided the terminal residuum $|\psi_\circ(T) - \psi_d|_{\mathbb{C}^n}$ as well as the term

$$\|D_\alpha \psi_\circ\|_{L^1(0,T;\mathbb{C}^n)} = \int_0^T \sum_{j\in I} \alpha_j \left|(\psi_\circ(t))_j\right| \mathrm{d}t$$

are sufficiently small.

3 Non-linear conjugate gradient approach

By reviewing past and present approaches to quantum control computation, we can identify two classes of solution procedures. On the one hand we have monotonic schemes, based on the Krotov method, first considered in [39]; see Section 4. On the other hand we have gradient-type methods. The latter were the first to be used in the early day of quantum control computation [30]. Gradient-based techniques appeared to be less competitive than monotonic schemes. However, some encouraging results were presented in [40] where a non-linear conjugate gradient (NCG) scheme was used. The approach in [40] was a crude generalization of the NCG method with a special rule concerning the value of steplength (formula (2.32) in [40]). The resulting algorithm is not always robust and convergence slow-down can be observed.

Our purpose is to show that an appropriate formulation of the NCG method with a robust line search strategy results in a competitive scheme for quantum optimal control problems. Two features determine the success of the scheme proposed in this paper. The use of line search based on the Wolfe-Powell strategy and an extension of a newly proposed formula [12] for determining conjugate search directions in the NCG method. In this section we illustrate our NCG approach and investigate its convergence properties.

3.1 The non-linear conjugate gradient method

We start discussing the minimization by NCG methods of a differentiable function $f : \mathbb{R}^m \to \mathbb{R}$. We denote $g(x) = \nabla f(x)$, $x \in \mathbb{R}^m$.

NCG schemes represent extensions of linear conjugate gradient (CG) to non-quadratic problems; see, e.g., [15, 38]. In the common variants, the basic idea is to avoid matrix operations and express the search directions recursively as

$$d_{k+1} = -g_{k+1} + \beta_k\, d_k, \tag{27}$$

for $k = 1, 2, \ldots$, with $d_1 = -g_1$. The iterates for a minimum point are given by

$$x_{k+1} = x_k + \tau_k\, d_k, \tag{28}$$

where $\tau_k > 0$ is a steplength. The parameter β_k is chosen so that (27)–(28) reduces to the linear CG scheme if f is a strictly convex quadratic function and τ_k is the exact one-dimensional minimizer of f along d_k. In this case the NCG scheme terminates in at most n steps in exact arithmetic.

We focus on the NCG scheme of Dai and Yuan [12] based on the formula

$$\beta_k = \beta_k^{DY} := \frac{\langle g_{k+1}, g_{k+1}\rangle_{\mathbb{R}^m}}{\langle d_k, y_k\rangle_{\mathbb{R}^m}},$$

where $y_k = g_{k+1} - g_k$. In [12], convergence of the proposed NCG scheme is established requiring that the steplength τ_k satisfies the standard Wolfe conditions given by

$$f(\epsilon_k) - f(\epsilon_k + \tau_k d_k) \geq -\delta \tau_k \langle g_k, d_k \rangle_{\mathbb{R}^m}$$

and

$$\langle g(\epsilon_k + \tau_k d_k), d_k \rangle_{\mathbb{R}^m} > \sigma \langle g_k, d_k \rangle_{\mathbb{R}^m},$$

with $0 < \delta < \sigma < 1$. Replacing the second of these two conditions above with the following, results in the strong Wolfe conditions

$$\left| \langle g(\epsilon_k + \tau_k d_k), d_k \rangle_{\mathbb{R}^m} \right| \leq -\sigma \langle g_k, d_k \rangle_{\mathbb{R}^m},$$

with $0 < \delta < \sigma < 1/2$; see [15].

Next, we discuss the minimization of a real-valued differentiable function denoted by $\hat{J}(\epsilon)$, representing the reduced cost functional $J(\psi(\epsilon), \epsilon)$, where $\psi(\epsilon)$ denotes the solution to the state equation (1). Denote with $g(\epsilon) = \nabla \hat{J}(\epsilon)$. Note that $g(\epsilon) \in H_0^1(0, T; \mathbb{C})$ for $\mu > 0$ and $g(\epsilon) \in L^2(0, T; \mathbb{C})$ for $\mu = 0$. Thus, we define

$$E = H_0^1(0, T; \mathbb{C}) \text{ if } \mu > 0 \quad \text{or} \quad E = L^2(0, T; \mathbb{C}) \text{ if } \mu = 0.$$

We define

$$\beta_k = \frac{\|g_{k+1}\|_E^2}{\Re e \langle d_k, y_k \rangle_E}. \tag{29}$$

We require that the steplength τ_k satisfies

$$\hat{J}(\epsilon_k) - \hat{J}(\epsilon_k + \tau_k d_k) \geq -\delta \tau_k \Re e \langle g_k, d_k \rangle_E \tag{30}$$

$$\langle g(\epsilon_k + \tau_k d_k), d_k \rangle_E > \sigma \Re e \langle g_k, d_k \rangle_E \tag{31}$$

where the parameters are still chosen such that: $0 < \delta < \sigma < 1/2$. The quality of line search in non-linear CG algorithms is crucial to preserve mutual conjugacy property of search directions and to ensure that each generated direction is one of descent. We use the Wolfe-Powell strategy to determine τ_k; see [27].

In the present framework, a sufficient descent condition is given by

$$\Re e \langle g_k, d_k \rangle_E \leq -c \|g_k\|_E^2$$

for some $c > 0$ and for all $k \geq 1$, is guaranteed only replacing (31) with the stronger condition

$$\left| \langle g(\epsilon_k + \tau_k d_k), d_k \rangle_E \right| \leq -\sigma \Re e \langle g_k, d_k \rangle_E$$

(and $c = 1/(1 + \sigma)$ results).

We consider the following NCG scheme

Algorithm 11 (NCG method)
Step 1. *Given* $k = 1$, ϵ_1, $d_1 = -g_1$, *if* $\|g_1\|_E < tol$ *then stop.*
Step 2. *Compute* $\tau_k > 0$ *satisfying* (30)–(31).
Step 3. *Let* $\epsilon_{k+1} = \epsilon_k + \tau_k d_k$.
Step 4. *Compute* $g_{k+1} = \nabla \hat{J}(\epsilon_{k+1})$.
If $\|g_{k+1}\|_E < tol_{abs}$ *or* $\|g_{k+1}\|_E < tol_{rel} \|g_1\|_E$ *or* $k = k_{\max}$ *then stop.*
Step 5. *Compute* β_k *by* (29).
Step 6. *Let* $d_{k+1} = -g_{k+1} + \beta_k d_k$.
Step 7. *Set* $k = k + 1$, *goto Step 2.*

In the remaining of this section we discuss convergence of the NCG scheme defined above. We need the following assumption.

Assumption 12 (1) *$\hat{J}$ is bounded from below and is continuously real differentiable in a neighborhood $\mathcal{N}$ of the level set $\mathcal{L} = \{\epsilon \in E : \hat{J}(\epsilon) \leq \hat{J}(\epsilon_1)\}$.*

(2) *$\nabla\hat{J}$ is Lipschitz continuous in $\mathcal{N}$, i.e. there exists a constant $L > 0$ such that*

$$\|\nabla\hat{J}(\epsilon_1) - \nabla\hat{J}(\epsilon_2)\|_E \leq L\,\|\epsilon_1 - \epsilon_2\|_E \quad \textit{for all } \epsilon_1, \epsilon_2 \in \mathcal{N}.$$

Notice that in our setting Assumption 12-(1) is satisfied. Moreover, $\epsilon \mapsto \hat{J}(\epsilon)$ is twice continuously Fréchet-differentiable so that Assumption 12-(2) holds at least locally.

We have

$$\begin{aligned}
\Re e\,\langle g_{k+1}, d_{k+1}\rangle_E &= \Re e\,\langle g_{k+1}, -g_{k+1} + \beta_k d_k\rangle_E = -\|g_{k+1}\|_E^2 + \beta_k\,\Re e\,\langle g_{k+1}, d_k\rangle_E \\
&= -\|g_{k+1}\|_E^2 + \frac{\|g_{k+1}\|_E^2}{\Re e\,\langle d_k, y_k\rangle_E}\,\Re e\,\langle g_{k+1}, d_k\rangle_E \\
&= \frac{\|g_{k+1}\|_E^2}{\Re e\,\langle d_k, y_k\rangle_E}(-\Re e\,\langle d_k, y_k\rangle_E + \Re e\,\langle g_{k+1}, d_k\rangle_E) = \beta_k\,\Re e\,\langle g_k, d_k\rangle_E.
\end{aligned}$$

Therefore we have

$$\beta_k = \frac{\Re e\,\langle g_{k+1}, d_{k+1}\rangle_E}{\Re e\,\langle g_k, d_k\rangle_E}. \tag{32}$$

Now we need the following lemma which is an extension of Lemma 3.2 in [12].

Lemma 13 *Suppose that ϵ_1 is a starting point for which Assumption 12 is satisfied and consider any method of the form $\epsilon_{k+1} = \epsilon_k + \tau_k\,d_k$ where d_k is a descent direction and τ_k satisfies (30)–(31). Then the following Zoutendijk condition holds*

$$\sum_{k\geq 1} \frac{\Re e\,\langle g_k, d_k\rangle_E^2}{\|d_k\|_E^2} < \infty$$

Proof. From (31) it follows that

$$\Re e\,\langle y_k, d_k\rangle_E = \Re e\,\langle g_{k+1} - g_k, d_k\rangle_E \geq (\sigma - 1)\,\Re e\,\langle g_k, d_k\rangle_E.$$

The Lipschitz condition implies that

$$\Re e\,\langle y_k, d_k\rangle_E = \Re e\,\langle g_{k+1} - g_k, d_k\rangle_E \leq \tau_k L\,\|d_k\|_E^2.$$

Combining the two inequalities one obtains

$$\tau_k \geq \frac{\sigma - 1}{L}\,\frac{\Re e\,\langle g_k, d_k\rangle_E}{\|d_k\|_E^2}.$$

This result combined with (30) gives

$$\hat{J}(\epsilon_k) - \hat{J}(\epsilon_k + \tau_k d_k) \geq -\delta\tau_k \Re e\,\langle g_k, d_k\rangle_E \geq c\frac{\Re e\,\langle g_k, d_k\rangle_E^2}{\|d_k\|_E^2}$$

where $c = \delta(1-\sigma)/L$ is a positive constant. Summing up for $k \geq 1$ and recalling that $\hat{J}$ is bounded below concludes the proof. □

We can now prove the following theorem representing an extension of Theorem 3.3 in [12].

Theorem 14 *Suppose that ϵ_1 is a starting point for which Assumption 12 holds. Let the sequence $\{\epsilon_k\}_{k\geq 1}$ be generated by the NCG Algorithm 11. Then this algorithm either terminates at the stationary point or converges in the sense that*

$$\liminf_{k\to\infty} \|g_k\|_E = 0.$$

If, in addition, the sequence $\{\|d_k\|_E/\|g_k\|_E\}$ is bounded then $\lim_{k\to\infty} \|g_k\|_E = 0$.

Proof. First notice that if the NCG algorithm does not terminate after finite many iterations, we have

$$\|g_k\|_E > 0 \quad \text{for all } k.$$

Next we show that the search directions are descent, in the sense that

$$\Re e\, \langle g_k, d_k\rangle_E < 0. \tag{33}$$

This is obvious for $k = 1$. Now assume that it holds for k and recall the following two results

$$\Re e\, \langle y_k, d_k\rangle_E \geq (\sigma - 1)\, \Re e\, \langle g_k, d_k\rangle_E \tag{34}$$

and

$$\Re e\, \langle g_{k+1}, d_{k+1}\rangle_E = \beta_k\, \Re e\, \langle g_k, d_k\rangle_E. \tag{35}$$

Therefore, we obtain

$$\Re e\, \langle g_{k+1}, d_{k+1}\rangle_E = \beta_k\, \Re e\, \langle g_k, d_k\rangle_E = \frac{\|g_{k+1}\|_E^2}{\Re e\, \langle d_k, y_k\rangle_E}\, \Re e\, \langle g_k, d_k\rangle_E \leq \frac{\|g_{k+1}\|_E^2}{(\sigma - 1)} < 0.$$

That is (33) holds for all k (and hence $\beta_k > 0$ for all k).

Now, consider the scalar product of both sides of $d_{k+1} + g_{k+1} = \beta_k\, d_k$ with themselves. We have $\|d_{k+1}\|_E^2 = \beta_k^2 \|d_k\|_E^2 - 2\Re e\, \langle g_{k+1}, d_{k+1}\rangle_E - \|g_{k+1}\|_E^2$. Dividing with $\Re e\langle g_{k+1}, d_{k+1}\rangle^2$ and using (32), we obtain

$$\begin{aligned}\frac{\|d_{k+1}\|_E^2}{\Re e\, \langle g_{k+1}, d_{k+1}\rangle_E^2} &= \frac{\|d_k\|_E^2}{\Re e\, \langle g_k, d_k\rangle_E^2} - \left(\frac{1}{\|g_{k+1}\|_E} + \frac{\|g_{k+1}\|_E}{\Re e\, \langle g_{k+1}, d_{k+1}\rangle_E}\right)^2 + \frac{1}{\|g_{k+1}\|_E^2} \\ &\leq \frac{\|d_k\|_E^2}{\Re e\, \langle g_k, d_k\rangle_E^2} + \frac{1}{\|g_{k+1}\|_E^2},\end{aligned}$$

and for $k = 1$ we have $\|d_1\|_E^2/\Re e\langle g_1, d_1\rangle_E^2 = 1/\|g_1\|_E^2$. Therefore we have

$$\frac{\|d_k\|_E^2}{\Re e\, \langle g_k, d_k\rangle_E^2} \leq \sum_{i=1}^{k} \frac{1}{\|g_i\|_E^2} \quad \text{for all } k \geq 1. \tag{36}$$

The proof of the theorem is concluded with a contradiction argument. If the theorem is not true, then there exists a constant $c > 0$ such that $\|g_k\|_E \geq c$ for all $k \geq 1$ and from (36) it follows that

$$\frac{\|d_k\|_E^2}{\Re e\, \langle g_k, d_k\rangle_E^2} \leq \frac{k}{c^2}$$

which implies that

$$\sum_{k\geq 1} \frac{\Re e\, \langle g_k, d_k\rangle_E^2}{\|d_k\|_E^2} \geq c^2 \sum_{k\geq 1} \frac{1}{k} = \infty$$

thus contradicting Lemma 13. □

3.2 Cascadic acceleration

The cascadic approach results from combining nested iteration techniques with (one-grid) iterative schemes. The idea is to spend more iteration on coarser grids to obtain a solution that, when interpolated to a finer grid, provides a good starting approximation for the iteration process on this grid. A schematic description of this method is given by the following. Let $lev = lev_0, \ldots, lev_f$ be the index of a hierarchy of nested grids, from coarsest to finest. Let x_{lev_0} represents a given starting approximation on the coarsest grid of the cascadic solution process and denote with I^{lev}_{lev-1} an interpolation operator from grid $lev-1$ to grid lev. Denote with $S_{lev}(x_{lev})$ the basic iteration. With $*$ we denote the obtained solution.

We have

$$\begin{aligned} x^*_{lev_0} &= x_{lev_0} \\ x^*_{lev} &= I^{lev}_{lev-1} x^*_{lev-1}; \qquad x^*_{lev} = S^{m_{lev}}_{lev}(x^*_{lev}), \qquad lev = lev_0 + 1, \ldots, lev_f, \end{aligned}$$

where m_{lev} is the number of iterations at level lev.

For the formulation of the so-called cascadic multigrid method and its analysis see [5]. In particular in [5, 37], a cascadic conjugate gradient method is discussed and optimal computational complexity for elliptic problems is proved.

While we are not able to extend the convergence theory in [5, 37] to the case of nonconvex optimization problems using the NCG scheme, we obtain considerable improvement with respect to one-grid NCG with the cascadic version of our NCG scheme. We denote the resulting scheme by C-NCG. For results of numerical experiments, see Section 5.

4 Monotonic schemes for quantum control

Monotonic schemes have been initially introduced in a general framework by Krotov [18, 19, 20]. Following this approach, Tannor *et al* [39] and then Zhu & Rabitz [42] have proposed two procedures for quantum control. These algorithms have a common basis, as it appears in [24], where a unified form is presented. Other extensions have then been designed to obtain bounded and bang-bang controls [41], stochastic monotonic schemes [35] or to optimize more general cost functionals [28] and systems involving dissipative states [29]. Recently, a relationship between these algorithms and local trajectory tracking procedures has been established, providing an interpretation of these schemes [35]. At the theoretical level, some proofs of the convergence of the monotonic schemes have been presented using either compactness and semi-group theory [21] or the Łojasiewicz inequality and its extensions [2]. On the other hand, usual time discretization of the monotonic schemes often leads to instabilities that prevent to reach numerical convergence. This problem is studied in [23, 34], where an appropriate time discretization is proposed which avoids instability. In order to tackle the control problem of the finite-level system presented in the section of numerical experiments, we present here a time discretized monotonic scheme based on a Crank-Nicholson propagator. The resulting algorithm is unconditionally stable and allows us to work with a large range of time steps.

For simplicity, throughout this section we will consider that $\mu = 0$ and require the following assumption that characterizes dissipation in the system.

Assumption 15 *We assume that*

$$\left\langle \frac{H_0 - H_0^*}{i}\psi, \psi \right\rangle_{L^2(0,T;\mathbb{C}^n)} \leq 0 \quad \textit{for all} \qquad \psi \in H^1(0,T;\mathbb{C}^n).$$

For reason of simplicity, we introduce an auxiliary cost functional

$$\widetilde{J}(\epsilon) := \Re e(\psi_d^* \cdot \psi(T)) - \frac{\gamma}{2}\|\epsilon\|^2_{L^2(0,T;\mathbb{C})} + \frac{1}{2}\langle \psi, \Lambda\psi\rangle_{L^2(0,T;\mathbb{C}^n)},$$

where ψ and ϵ are linked by (1) and Λ is defined by

$$\Lambda\psi = -\sum_{j\in I}\alpha_j\psi_j - \frac{H_0 - H_0^*}{i}\psi.$$

We consider the optimal control problem corresponding to the maximization of $\widetilde{J}(\epsilon)$ under the constraint (1). Notice that this problem is equivalent to the optimal control problem (**P**) since

$$\begin{aligned}\hat{J}(\epsilon) &= |\psi_0|^2_{\mathbb{C}^n} - \Re e(\psi_d^* \cdot \psi(T)) - \frac{1}{2}\langle \psi, \Lambda\psi\rangle_{L^2(0,T;\mathbb{C}^n)} + \frac{\gamma}{2}\|\epsilon\|^2_{L^2(0,T;\mathbb{C})} \\ &= |\psi_0|^2_{\mathbb{C}^n} - \widetilde{J}(\epsilon).\end{aligned}$$

We assume that the following holds.

Assumption 16 *The parameters α_j fulfill the condition:*

$$-\sum_{j\in I}\alpha_j|\psi_j|^2 - \left\langle \frac{H_0 - H_0^*}{i}\psi, \psi \right\rangle_{L^2(0,T;\mathbb{C}^n)} \geq 0 \quad \textit{for all } \psi \in C^n.$$

With this assumption, the operator Λ is positive. This condition is necessary to guarantee the monotonicity of our algorithm.

Before presenting a monotonic scheme corresponding to the maximization of $\widetilde{J}(\epsilon) = \widetilde{J}(\psi(\epsilon), \epsilon)$, we need to compute the increase of this cost functional between two control fields. Let us first define the Lagrange multiplier q by

$$\begin{aligned} i\dot{q} &= (H_0^* + H_1^*(\epsilon(\cdot)))\, q + \Lambda\psi \\ iq(T) &= -\psi_d. \end{aligned} \tag{37}$$

Consider two fields ϵ and ϵ', and the corresponding wavefunctions ψ and ψ', and Lagrange multipliers q and q', respectively. We have

$$\begin{aligned}\widetilde{J}(\epsilon') - \widetilde{J}(\epsilon) = \;& \Re e(\psi_d^* \cdot (\psi'(T) - \psi(T))) + \Re e\langle \Lambda\psi, \psi' - \psi\rangle_{L^2(0,T;\mathbb{C}^n)} \\ &+ \frac{1}{2}\langle \psi' - \psi, \Lambda(\psi' - \psi)\rangle_{L^2(0,T;\mathbb{C}^n)} \\ &- \frac{\gamma}{2}\left(\|\epsilon'\|^2_{L^2(0,T;\mathbb{C})} - \|\epsilon\|^2_{L^2(0,T;\mathbb{C})}\right).\end{aligned} \tag{38}$$

Focusing on the first two terms of this sum, we obtain

$$\begin{aligned}&\Re e(\psi_d^* \cdot (\psi'(T) - \psi(T))) + \Re e\,\langle \Lambda\psi, \psi' - \psi\rangle_{L^2(0,T;\mathbb{C}^n)} \\ &= \Re e(iq(T)^* \cdot (\psi'(T) - \psi(T))) + \Re e\,\langle \Lambda\psi, \psi' - \psi\rangle_{L^2(0,T;\mathbb{C}^n)} \\ &= \Re e\left\langle -iq, \frac{H_0 + H_1(\epsilon'(\cdot))}{i}\psi' - \frac{H_0 + H_1(\epsilon(\cdot))}{i}\psi \right\rangle_{L^2(0,T;\mathbb{C}^n)} \\ &\quad + \Re e\,\langle -i\dot{q}, \psi' - \psi\rangle_{L^2(0,T;\mathbb{C}^n)} + \Re e\,\langle \Lambda\psi, \psi' - \psi\rangle_{L^2(0,T;\mathbb{C}^n)} \\ &= \Re e\,\langle q, (H_1(\epsilon'(\cdot)) - H_1(\epsilon(\cdot)))\psi'\rangle_{L^2(0,T;\mathbb{C}^n)}.\end{aligned}$$

Thus, the increase of $\widetilde{J}$ reads as follows

$$\begin{aligned}\widetilde{J}(\epsilon') - \widetilde{J}(\epsilon) &= \int_0^T \Re e(q(s)^* \cdot \big(H_1(\epsilon'(s)) - H_1(\epsilon(s))\big)\psi'(s)) - \frac{\gamma}{2}\big(|\epsilon'(s)|^2 - |\epsilon(s)|^2\big)\,\mathrm{d}s \\ &\quad + \frac{1}{2}\left\langle \psi' - \psi, \Lambda(\psi' - \psi)\right\rangle_{L^2(0,T;\mathbb{C}^n)}. \end{aligned} \tag{39}$$

This formula is the starting point for the design of the monotonic algorithm. For example, we have the following result.

Lemma 17 *Given $\epsilon \in L^2(0,T;\mathbb{C})$, suppose there exists ϵ' such that*

$$\begin{aligned}\epsilon'_{\Re e}(t) &= \epsilon_{\Re e}(t) + \frac{1}{\gamma}\Re e(q(t)^* \cdot H_{1\Re e}\psi'(t)), \\ \epsilon'_{\Im m}(t) &= \epsilon_{\Im m}(t) + \frac{1}{\gamma}\Re e(q(t)^* \cdot H_{1\Im m}\psi'(t)).\end{aligned}$$

Then $\widetilde{J}(\epsilon') \geq \widetilde{J}(\epsilon)$.

4.1 Time discretized algorithm

Due to their sequential feature, monotonic schemes require a particular time discretization in order to keep their monotonicity at the discrete level. This discretization is discuseed in detail in this section.

For any given integer N, let us introduce the discretization parameter δt defined by $N\,\delta t = T$ and $\epsilon_\ell = \epsilon_{\Re e,\ell} + i\epsilon_{\Im m,\ell}$, ψ_ℓ, q_ℓ that stand respectively for approximations of $\epsilon(\ell\delta t)$, $\psi(\ell\delta t)$, $q(\ell\delta t)$. Moreover, we denote by H_ℓ the approximation of the Hamiltonian $H_0 + H_1(\epsilon(\ell\delta t))$.

Given an initial state ψ_0, we solve numerically (1) and (37) by a Crank-Nicholson scheme. This discretization gives rise to the following iteration

$$\psi_{\ell+1} = \Big(I_d - \frac{\delta t H_\ell}{2i}\Big)^{-1}\Big(I_d + \frac{\delta t H_\ell}{2i}\Big)\psi_\ell \tag{40}$$

and

$$\begin{aligned} q_\ell &= \Big(I_d + \frac{\delta t H_\ell^*}{2i}\Big)\Big(I_d - \frac{\delta t H_\ell^*}{2i}\Big)^{-1} q_{\ell+1} - i\delta t \Lambda \psi_{\ell+1} \\ iq_N &= -\psi_d, \end{aligned} \tag{41}$$

where I_d is the identity matrix. Since $H_1(\epsilon)$ is hermitian and because of Assumption 15, the following uniform bounds can be obtained

$$|\psi_\ell|_{\mathbb{C}^n} \leq |\psi_0|_{\mathbb{C}^n},\ |q_\ell|_{\mathbb{C}^n} \leq T\,\rho(\Lambda)\,|\psi_0|_{\mathbb{C}^n} + |\psi_d|_{\mathbb{C}^n} \qquad \text{for all } \ell = 0,\dots,N-1. \tag{42}$$

We also introduce the time discretized cost functional

$$\widetilde{J}_{\delta t}(\epsilon) = \Re e(\psi_d^* \cdot \psi_N) - \frac{\gamma\delta t}{2}\sum_{\ell=0}^{N-1}|\epsilon_\ell|^2 + \frac{\delta t}{2}\sum_{\ell=0}^{N-1}(\Lambda\psi_{\ell+1}^* \cdot \psi_{\ell+1}).$$

Consider two control fields ϵ and ϵ'. Repeating the computations of the previous section at the discrete level, we obtain the following equivalent of (39)

$$\begin{aligned}\widetilde{J}_{\delta t}(\epsilon') - \widetilde{J}_{\delta t}(\epsilon) &= \sum_{\ell=0}^{N-1}\Re e(q_\ell^* \cdot \mathcal{D}H_\ell\psi'_\ell) - \frac{\gamma\delta t}{2}\big(|\epsilon'_\ell|^2 - |\epsilon_\ell|^2\big) \\ &\quad + \frac{\delta t}{2}\sum_{\ell=0}^{N-1}\big((\psi'_\ell - \psi_\ell)^* \cdot \Lambda(\psi'_\ell - \psi_\ell)\big),\end{aligned} \tag{43}$$

where

$$\mathcal{D}H_\ell = i\Big(\Big(I_d+\frac{\delta t H_\ell}{2i}\Big)^{-1}\Big(I_d-\frac{\delta t H_\ell}{2i}\Big)\Big(I_d-\frac{\delta t H'_\ell}{2i}\Big)^{-1}\Big(I_d+\frac{\delta t H'_\ell}{2i}\Big)-I_d\Big).$$

We now present a monotonic scheme to optimize $\widetilde{J}_{\delta t}$. First, notice that given $\epsilon=(\epsilon_\ell)_{0\le\ell\le N-1}$ and ψ'_ℓ, the term $\Re e[q_\ell\cdot\Delta H_\ell\psi'_\ell]-\delta t\frac{\gamma}{2}(|\epsilon'_\ell|^2-|\epsilon_\ell|^2)$ in (43) only depends on ϵ'_ℓ. Starting from this remark, the algorithm we propose consists in optimizing recursively each term of the first sum in (43) with respect to ϵ'_ℓ via one iteration of a Newton method.

Let us compute a Taylor expansion with respect to $\delta\epsilon_\ell=\epsilon'_\ell-\epsilon_\ell=\delta\epsilon_{\Re e,\ell}+i\delta\epsilon_{\Im m,\ell}$. Defining $\Delta H_\ell=H_1(\epsilon')-H_1(\epsilon)$, we have

$$\begin{aligned}(q^*_\ell\cdot\mathcal{D}H_\ell\psi'_\ell)=&\frac{\delta t}{2}(\widetilde{q}^*_\ell\cdot\Delta H_\ell\check{\psi}'_\ell)+i\frac{\delta t^2}{4}\Big[\widetilde{q}^*_\ell\cdot\Delta H_\ell\Big(I_d-\frac{\delta t H_\ell}{2i}\Big)^{-1}\Delta H_\ell\check{\psi}'_\ell\Big]\\&+o\big((\Delta H_\ell)^2\big),\end{aligned}$$

where

$$\begin{aligned}\widetilde{q}_\ell=&\Big(I_d+\Big(I_d+\frac{\delta t H^*_\ell}{2i}\Big)\Big(I_d-\frac{\delta t H^*_\ell}{2i}\Big)^{-1}\Big)q_\ell=q_\ell+q_{\ell+1}+i\delta t\Lambda\psi_{\ell+1},\\ \check{\psi}'_\ell=&\Big(I_d-\frac{\delta t H_\ell}{2i}\Big)^{-1}\psi'_\ell.\end{aligned}$$

This increase can then be expressed in terms of the quantity $\delta\epsilon_\ell$

$$\begin{aligned}(q^*_\ell.\mathcal{D}H_\ell\psi'_\ell)=&\frac{\delta t}{2}A_\ell^T\begin{pmatrix}\delta\epsilon_{\Re e,\ell}\\ \delta\epsilon_{\Im m,\ell}\end{pmatrix}+\frac{\delta t}{4}\begin{pmatrix}\delta\epsilon_{\Re e,\ell}\\ \delta\epsilon_{\Im m,\ell}\end{pmatrix}^T(i\delta t B_\ell)\begin{pmatrix}\delta\epsilon_{\Re e,\ell}\\ \delta\epsilon_{\Im m,\ell}\end{pmatrix}\\&+o\big((\delta\epsilon_\ell)^2\big),\end{aligned}$$

where T stands for transpose and the matrices A_ℓ and B_ℓ are given by

$$A_\ell=\begin{pmatrix}(\widetilde{q}^*_\ell\cdot H_{1\Re e}\check{\psi}'_\ell)\\ \\ (\widetilde{q}^*_\ell\cdot H_{1\Im m}\check{\psi}'_\ell)\end{pmatrix},\qquad B_\ell=\begin{pmatrix}B_{\ell,1,1}&B_{\ell,1,2}\\ B_{\ell,2,1}&B_{\ell,2,2}\end{pmatrix},\tag{44}$$

with

$$\begin{aligned}B_{\ell,1,1}&=(\widetilde{q}^*_\ell\cdot H_{1\Re e}\Big(I_d-\frac{\delta t H_\ell}{2i}\Big)^{-1}H_{1\Re e}\check{\psi}'_\ell)\\ B_{\ell,2,2}&=(\widetilde{q}^*_\ell\cdot H_{1\Im m}\Big(I_d-\frac{\delta t H_\ell}{2i}\Big)^{-1}H_{1\Im m}\check{\psi}'_\ell)\\ B_{\ell,1,2}&=B_{\ell,2,1}\\&=\frac{1}{2}\Big[\widetilde{q}^*_\ell\cdot\Big\{H_{1\Im m}\Big(I_d-\frac{\delta t H_\ell}{2i}\Big)^{-1}H_{1\Re e}\check{\psi}'_\ell+H_{1\Re e}\Big(I_d-\frac{\delta t H_\ell}{2i}\Big)^{-1}H_{1\Im m}\check{\psi}'_\ell\Big\}\Big].\end{aligned}$$

On the other hand, one has

$$|\epsilon'_\ell|^2-|\epsilon_\ell|^2=2\begin{pmatrix}\epsilon_{\Re e,\ell}\\ \epsilon_{\Im m,\ell}\end{pmatrix}^T\begin{pmatrix}\delta\epsilon_{\Re e,\ell}\\ \delta\epsilon_{\Im m,\ell}\end{pmatrix}+\begin{pmatrix}\delta\epsilon_{\Re e,\ell}\\ \delta\epsilon_{\Im m,\ell}\end{pmatrix}^T\begin{pmatrix}\delta\epsilon_{\Re e,\ell}\\ \delta\epsilon_{\Im m,\ell}\end{pmatrix},$$

and the increase of the cost functional reads

$$\widetilde{J}_{\delta t}(\epsilon') - \widetilde{J}_{\delta t}(\epsilon) = \delta t \sum_{\ell=0}^{N-1} \left(\frac{1}{2}\Re e A_\ell - \gamma \begin{pmatrix} \epsilon_{\Re e,\ell} \\ \epsilon_{\Im m,\ell} \end{pmatrix}\right)^T \begin{pmatrix} \delta\epsilon_{\Re e,\ell} \\ \delta\epsilon_{\Im m,\ell} \end{pmatrix} - \begin{pmatrix} \delta\epsilon_{\Re e,\ell} \\ \delta\epsilon_{\Im m,\ell} \end{pmatrix}^T \left(\frac{\delta t}{4}\Im m B_\ell + \frac{\gamma}{2} I_2\right) \begin{pmatrix} \delta\epsilon_{\Re e,\ell} \\ \delta\epsilon_{\Im m,\ell} \end{pmatrix} + o\big((\delta\epsilon_\ell)^2\big) + \frac{\delta t}{2} \sum_{\ell=0}^{N-1} ((\psi'_\ell - \psi_\ell)^* \cdot \Lambda(\psi'_\ell - \psi_\ell)),$$

where I_2 denotes the identity matrix of $\mathbb{R}^2$.

Algorithm 18 (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 ψ^{k+1}, q^{k+1}, ϵ^{k+1}, is done as follows.*

Forward propagation: Given $\psi_0^{k+1} = \psi_0$, compute recursively $\psi_{\ell+1}^{k+1}$ from ψ_ℓ^{k+1} by
Step 1. (Newton iteration) Compute ϵ_ℓ^{k+1} by

$$\begin{pmatrix} \epsilon^{k+1}_{\Re e,\ell} \\ \epsilon^{k+1}_{\Im m,\ell} \end{pmatrix} = \begin{pmatrix} \epsilon^{k}_{\Re e,\ell} \\ \epsilon^{k}_{\Im m,\ell} \end{pmatrix} + \frac{1}{2}\left(\frac{\delta t}{4}\Im m B_\ell^k + \frac{\gamma}{2} I_2\right)^{-1} \left(\frac{1}{2}\Re e A_\ell^k - \gamma \begin{pmatrix} \epsilon^{k}_{\Re e,\ell} \\ \epsilon^{k}_{\Im m,\ell} \end{pmatrix}\right). \quad (45)$$

Step 2. Compute $\psi_{\ell+1}^{k+1}$ by (40).

Backward propagation: Given $q_N^{k+1} = i\psi_d$, compute recursively q_ℓ^{k+1} from $q_{\ell+1}^{k+1}$ by (41).

4.2 Convergence of the algorithm

We present some results concerning the convergence of the CNMS. An important property of this scheme is that the sequence $(\epsilon^k)_{k\in\mathbb{N}}$ is bounded, as claimed in the next lemma.

Lemma 19 *For small enough δt, there exist M, such that*

$$\forall k \ \in \mathbb{N},\ \forall \ell = 0...N-1,\ |\epsilon^k_{\Re e,\ell}| \le M,\ |\epsilon^k_{\Im m,\ell}| \le M.$$

Proof. Consider an initial control field ϵ^0 and define m and M by

$$\begin{aligned} m &= 2\,K_0\,|\psi_0|_{\mathbb{C}^n}\left(|\psi_d|_{\mathbb{C}^n} + \rho(\Lambda)\,(T + \delta t|\psi_0|_{\mathbb{C}^n})\right) \\ M &= \frac{2m}{\gamma} + \max_{\ell=0...N-1} |\epsilon^0_\ell|. \end{aligned}$$

Let us denote by $A_{\Re e,\ell}$ and $A_{\Im m,\ell}$ the components of A_ℓ. Given ϵ, the estimates (42) and the definition (44) give

$$\max(|A_{\Re e,\ell}|, |A_{\Im m,\ell}|) \le \frac{m}{1 - \delta t \frac{\rho(H_0) + K_0|\epsilon_\ell|}{2}}.$$

A similar estimate can be obtained for the coefficients of B_ℓ. Let us denote by b the bound obtained for $\epsilon_\ell \le M$.
Given $k, \ell \in \mathbb{N}$, suppose that $\max(|\epsilon^k_{\Re e,\ell}|, |\epsilon^k_{\Im m,\ell}|) \le M$ and that δt is such that:

$$\frac{m}{1 - \delta t \frac{\rho(H_0) + K_0 M}{2}} \le M\gamma.$$

The iteration (45) reads

$$\epsilon_\ell^{k+1} = (I_2 - \delta_\ell^k)\epsilon_\ell^k + \frac{\delta_\ell^k}{2\gamma}\Re e A_\ell^k,$$

where:

$$\delta_\ell^k = (\frac{\delta t}{2\gamma}\Im m B_\ell^k + I_2)^{-1}.$$

Since the coefficients of B_ℓ^k are bounded by b, it can be supposed that the diagonal coefficients of δ_ℓ^k belong to $[1/2, 3/2]$ and the others to $[-1/8, 1/8]$. Then

$$\begin{aligned} |\epsilon_{\Re e,\ell}^{k+1}| &\leq |1-\delta_{\ell,1,1}^k||\epsilon_{\Re e,\ell}^k| + |\delta_{\ell,1,2}^k||\epsilon_{\Im m,\ell}^k| + \frac{|\delta_{\ell,1,1}^k|}{2\gamma}|\Re e A_{r,\ell}| + \frac{|\delta_{\ell,1,2}^k|}{2\gamma}|\Re e A_{i,\ell}| \\ &\leq (|1-\delta_{\ell,1,1}^k| + |\delta_{\ell,1,1}^k|)M \leq M, \end{aligned}$$

where $\delta_{\ell,i,j}^k$ denotes the components of δ_ℓ^k. The same result holds for $|\epsilon_{\Im m,\ell}^{k+1}|$. The lemma follows. □

The next lemma gives a result about the monotonicity of CNMS.

Lemma 20 *For small enough δt, the CNMS converges monotonically, in the sense that*

$$\exists\ \eta > 0, \quad \widetilde{J}_{\delta t}(\epsilon^{k+1}) - \widetilde{J}_{\delta t}(\epsilon^k) \geq \eta \|\epsilon^{k+1} - \epsilon^k\|_{\mathbb{C}^{N-1}}^2.$$

Proof. The increase of $\widetilde{J}_{\delta t}$ between two iterations reads

$$\begin{aligned} \widetilde{J}_{\delta t}(\epsilon^{k+1}) - \widetilde{J}_{\delta T}(\epsilon^k) \ =\ & \frac{\delta t\gamma}{2}\sum_{\ell=0}^{N-1}|\delta\epsilon_\ell^k|^2 + \frac{\delta t}{2}\sum_{\ell=0}^{N-1}((\psi_\ell' - \psi_\ell)^* \cdot \Lambda(\psi_\ell' - \psi_\ell)) \\ & + \sum_{\ell=0}^{N-1}\Re e R_\ell^k, \end{aligned}$$

where

$$R_\ell^k = (q_\ell^k\ {}^*\cdot \mathcal{D}H_\ell\psi_\ell^{k+1}) - \frac{\delta t}{2}(A_\ell^k)^T \begin{pmatrix} \delta\epsilon_{\Re e,\ell}^k \\ \delta\epsilon_{\Im m,\ell}^k \end{pmatrix} + \delta t^2 \begin{pmatrix} \delta\epsilon_{\Re e,\ell}^k \\ \delta\epsilon_{\Im m,\ell}^k \end{pmatrix}^T \Im m B_\ell^k \begin{pmatrix} \delta\epsilon_{\Re e,\ell}^k \\ \delta\epsilon_{\Im m,\ell}^k \end{pmatrix}.$$

Using the Taylor-Lagrange formula and Lemma 19, we obtain

$$R_\ell^k = |\epsilon_\ell^k|^2\, o(\delta t),$$

and the result follows. □

Further analysis shows that there exists $\upsilon > 0$ such that

$$\|\nabla\widetilde{J}_{\delta t}(\epsilon^{k+1})\|_{\mathbb{C}^{N-1}} \leq \upsilon\|\epsilon^{k+1} - \epsilon^k\|_{\mathbb{C}^{N-1}},$$

where $\nabla\widetilde{J}_{\delta t}$ denotes the gradient of $\widetilde{J}_{\delta t}$ with respect to ϵ. This fact combined with Lemma 20 enable us to claim the following convergence result.

Theorem 21 *For all initial value ϵ^0, the sequence $(\epsilon^k)_{k\in\mathbb{N}}$ converges towards a critical point of $\widetilde{J}_{\delta t}$. Denoting by ϵ^∞ this limit, there exists $c > 0$ and $\chi > 0$ such that:*

$$\|\epsilon^k - \epsilon^\infty\|_{\mathbb{C}^{N-1}} \leq ck^{-\chi}.$$

Note that, for large values of γ, it can be proved that the convergence rate is indeed linear. We refer the Reader to [34] for the details of the proof.

5 Numerical experiments

We present results of numerical experiments with a representative three-level quantum system whose configuration is represented by $\psi = (\psi_1, \psi_2, \psi_3) \in H^1(0,T;\mathbb{C}^3)$ 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 (wiggled line); see Figure 1. Such Λ-type configurations have a long-standing history in quantum optics and have been demonstrated successful in the explanation of many coherence-phenomena in atomic systems [3]; more recently, similar configurations have received increasing attention also in semiconductor quantum dots [16].

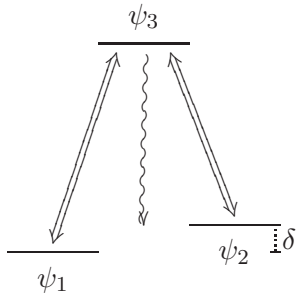

Figure 1: Prototypical Λ-type three-level scheme: ψ_1 and ψ_2 are long-lived states whereas ψ_3 is a short-lived state which is optically coupled to both ψ_1 and ψ_2; wiggled line indicates relaxation and dephasing of state ψ_3.

Time evolution of this finite-level quantum system is governed by the following Hamiltonian [3, 16]

$$H_0 = \frac{1}{2}\begin{pmatrix} -\delta & 0 & 0 \\ 0 & \delta & 0 \\ 0 & 0 & -i\gamma_0 \end{pmatrix}, \tag{46}$$

where the term $-i\gamma_0$ accounts for environment losses (e.g., spontaneous photon emissions). The coupling to the external field $\epsilon = \epsilon_{\Re e} + i\epsilon_{\Im m}$, reads

$$H_1(\epsilon) = -\frac{1}{2}\begin{pmatrix} 0 & 0 & \mu_1\epsilon \\ 0 & 0 & \mu_2\epsilon \\ \mu_1\epsilon^* & \mu_2\epsilon^* & 0 \end{pmatrix}, \tag{47}$$

where μ_1 and μ_2 describe the coupling strengths of states ψ_1 and ψ_2 to the interconnecting state ψ_3 (e.g., optical dipole matrix elements).

Typical initial and final states are given by

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

respectively.

Concerning the optimality condition (12g) we have

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

Assuming that the system is initially prepared in state ψ_1, that is $\psi = \psi_0$, we use the optimal control approach to determine the most efficient way to bring the system from ψ_1 to ψ_2. The form of $H_1(\epsilon)$ is such that direct optical transition between ψ_1 and ψ_2 is forbidden. The presence of the third auxiliary state ψ_3 allows this transition through intermediate population transfer while introducing losses because of environment coupling. Therefore we require to find a sequence of laser pulses that minimizes the population of level ψ_3 along evolution.

In this section, we report results of experiments to show the importance of a sufficiently small tolerance which needs be defined in an appropriate way. We give evidence that using the second-order Crank-Nicholson scheme for evolution, second-order accurate solutions of the optimization problems are obtained. We compare the non-linear CG scheme with the monotonic method showing that the former may result more efficient and robust. We then provide results that demonstrate improved convergence properties of NCG with cascadic acceleration. We complete this section discussing the different solution behavior for different choices of values of the optimization parameters.

Some of problems' parameters are held fixed. We choose $\delta = 20$, $\gamma_0 = 0.01$, $\mu_1 = 1$, $\mu_2 = 1$, $\alpha_1 = 0$, $\alpha_2 = 0$, and $T = 5$.

To determine the evolution of state and adjoint variables we consider an implicit second-order Crank-Nicholson scheme. Given the solution at time step ℓ, the value of the wave function at the next time step $\ell + 1$ is given by

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

Thus $\psi_{\ell+1}$ is given by

$$\psi_{\ell+1} = (I_d - \frac{\delta t}{2i}H_{\ell+1})^{-1}\,(I_d + \frac{\delta t}{2i}H_\ell)\psi_\ell, \qquad \ell = 0, \ldots, N-1.$$

In case of finite-level quantum systems, the operator $(I_d + i\frac{\delta t}{2}H_{\ell+1})$ is a $n \times n$ complex matrix which is easily invertible. The formula above holds for the adjoint equation marching backwards by inverting the time direction. Notice that this scheme is slightly different from the one presented in Section 4.1, this latter being relevant to the sequential feature of the CNMS scheme.

First we focus on the problem of assessing convergence of iterative solvers to quantum optimal control solutions. We give evidence that optimal solutions are quite sensitive to the order of tolerance required in computations. For this purpose consider the results reported in Table 5. These results have been obtained using the convergence criteria

$$\frac{J^{k+1} - J^k}{J^k} \leq tol$$

where J^k is the value of the reduced cost functional after k iterations. This criteria is commonly used in the scientific computing community and typical values for the tolerance are $tol \in (10^{-6}, 10^{-4})$. One should notice that in case of minimization problems with flat minima basins, such convergence criteria may be misleading. This fact can be partly seen in Table 5 considering the values of $|\psi(T) - \psi_d|_{\mathbb{C}^3}$ and of J. In Figure 2, a more dramatic picture is given of how sensitive the optimal solution is with respect to the chosen tolerance.

On the other hand, resulting values of the discrete $L^2(0, T; \mathbb{C})$-norm (denoted by $\|\cdot\|$) of the residual of the optimality conditions (12g), $\|res_\epsilon\|$, suggest to use this

tol	$\|res_\epsilon\|$	$\|\psi(T) - \psi_d\|_{\mathbb{C}^3}$	J	CPU
10^{-4}	$2.93 \cdot 10^{-3}$	$5.46 \cdot 10^{-3}$	$7.69 \cdot 10^{-2}$	1.09
10^{-6}	$1.49 \cdot 10^{-3}$	$2.99 \cdot 10^{-3}$	$6.77 \cdot 10^{-3}$	3.04
10^{-8}	$2.39 \cdot 10^{-5}$	$3.13 \cdot 10^{-3}$	$6.77 \cdot 10^{-3}$	5.43
10^{-10}	$5.23 \cdot 10^{-6}$	$3.15 \cdot 10^{-3}$	$6.77 \cdot 10^{-3}$	8.18
10^{-12}	$8.84 \cdot 10^{-7}$	$3.15 \cdot 10^{-3}$	$6.77 \cdot 10^{-3}$	28.29

Table 1: Optimization results for different values of tolerance.

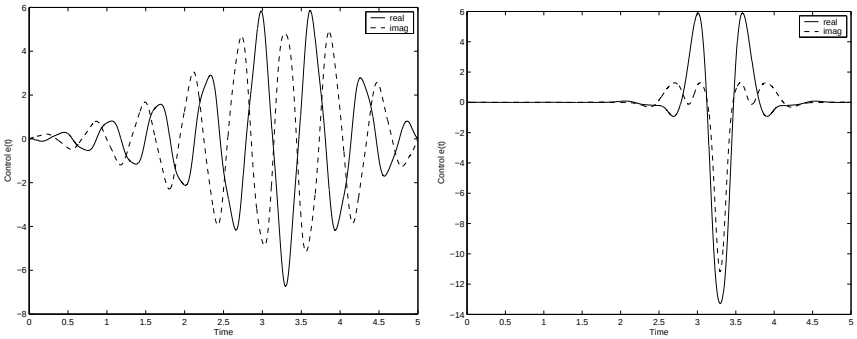

Figure 2: Control fields computed with different choices of tolerance $tol = \{10^{-4}, 10^{-8}\}$ (from left to right).

value in order to have a robust convergence criteria. In fact, we use the criteria as given in NCG Algorithm 11, that is,

$$\|g_{k+1}\| < tol_{abs} \text{ and } \|g_{k+1}\| < tol_{rel}\,\|g_1\|,$$

where we take $tol_{rel} = 10\,tol_{abs}$.

An open issue in the numerical analysis of quantum optimal control problems is the order of accuracy of optimal solutions with respect to (temporal) mesh size. While not obvious, we should expect second-order convergence when using second-order marching schemes for the state and adjoint equations. To show this fact, and since it is difficult to define an exact solution for this class of problems, we adopt the following strategy [9].

Consider a hierarchy of nested meshes with $N = 2^{lev}$ where lev is the level index. We take the solution on a fine mesh lev as the reference solution for computing the error on the next coarser mesh $lev - 1$ as follows $e_{\psi^k,lev-1} = \psi_{k,lev-1} - I_{lev}^{lev-1}\psi_{k,lev}$, $k = 1, 2, 3$, and $e_{\epsilon,lev-1} = \epsilon_{lev-1} - I_{lev}^{lev-1}\epsilon_{lev}$ where I_{lev}^{lev-1} is injection. In Table 5 we report results for different meshes and different values of tolerance obtained with the NCG scheme. We see that for sufficiently small tol_{abs} second-order accuracy is attained. This result suggests a way to assess convergence of optimal solutions. In Table 5 we report values of norm of residuals. Because at each time step the exact solution of the implicit Crank-Nicholson scheme is performed, residuals of state and adjoint equations are computer zero. On the other hand we see that $\|res_\epsilon\|$ is representative of the attained accuracy of the solution.

In Table 5 results are reported to compare the computational performance of the NCG scheme (no cascadic acceleration) and the CNMS scheme for different choices of tolerance and mesh sizes. We see that the NCG scheme provides increasing better

$tol_{abs} = 10^{-7}$	N	$\|e_{\psi_1,lev-1}\|$	$\|e_{\psi_2,lev-1}\|$	$\|e_{\psi_3,lev-1}\|$	$\|e_{\epsilon,lev-1}\|$
	512	$1.4 \cdot 10^{-2}$	$5.4 \cdot 10^{-3}$	$4.2 \cdot 10^{-3}$	$6.1 \cdot 10^{-13}$
	1024	$3.5 \cdot 10^{-3}$	$1.3 \cdot 10^{-3}$	$1.0 \cdot 10^{-3}$	$6.8 \cdot 10^{-3}$
	2048	$8.9 \cdot 10^{-4}$	$3.3 \cdot 10^{-4}$	$2.6 \cdot 10^{-4}$	$1.7 \cdot 10^{-3}$
	4096	$2.2 \cdot 10^{-4}$	$8.7 \cdot 10^{-5}$	$7.0 \cdot 10^{-5}$	$3.6 \cdot 10^{-4}$
$tol_{abs} = 10^{-5}$	N	$\|e_{\psi_1,lev-1}\|$	$\|e_{\psi_2,lev-1}\|$	$\|e_{\psi_3,lev-1}\|$	$\|e_{\epsilon,lev-1}\|$
	512	$1.4 \cdot 10^{-2}$	$5.2 \cdot 10^{-3}$	$4.2 \cdot 10^{-3}$	$6.3 \cdot 10^{-13}$
	1024	$3.5 \cdot 10^{-3}$	$1.4 \cdot 10^{-3}$	$1.1 \cdot 10^{-3}$	$6.0 \cdot 10^{-3}$
	2048	$8.6 \cdot 10^{-4}$	$3.7 \cdot 10^{-4}$	$3.2 \cdot 10^{-4}$	$1.3 \cdot 10^{-3}$
	4096	$2.2 \cdot 10^{-4}$	$8.1 \cdot 10^{-5}$	$7.2 \cdot 10^{-5}$	$3.7 \cdot 10^{-4}$
$tol_{abs} = 10^{-3}$	N	$\|e_{\psi_1,lev-1}\|$	$\|e_{\psi_2,lev-1}\|$	$\|e_{\psi_3,lev-1}\|$	$\|e_{\epsilon,lev-1}\|$
	512	$1.4 \cdot 10^{-2}$	$7.2 \cdot 10^{-3}$	$5.5 \cdot 10^{-3}$	$6.5 \cdot 10^{-13}$
	1024	$3.5 \cdot 10^{-3}$	$1.3 \cdot 10^{-3}$	$1.9 \cdot 10^{-3}$	$4.4 \cdot 10^{-3}$
	2048	$9.2 \cdot 10^{-4}$	$5.7 \cdot 10^{-4}$	$5.2 \cdot 10^{-4}$	$8.2 \cdot 10^{-4}$
	4096	$2.3 \cdot 10^{-4}$	$9.8 \cdot 10^{-5}$	$1.2 \cdot 10^{-4}$	$3.6 \cdot 10^{-4}$

Table 2: Approximation results for different meshes; $N = 2^{lev}$.

tol	$\|res_\epsilon\|$	$\|res_\psi\|$	$\|res_p\|$
10^{-4}	$2.9 \cdot 10^{-3}$	$6.5 \cdot 10^{-13}$	$5.9 \cdot 10^{-15}$
10^{-6}	$1.5 \cdot 10^{-3}$	$6.3 \cdot 10^{-13}$	$4.2 \cdot 10^{-15}$
10^{-8}	$2.4 \cdot 10^{-5}$	$6.3 \cdot 10^{-13}$	$4.2 \cdot 10^{-15}$
10^{-10}	$5.2 \cdot 10^{-6}$	$6.2 \cdot 10^{-13}$	$4.1 \cdot 10^{-15}$
10^{-12}	$8.8 \cdot 10^{-7}$	$6.1 \cdot 10^{-13}$	$4.0 \cdot 10^{-15}$

Table 3: Residuals on mesh $N = 8192$ for different values of tolerance.

performance as tol_{abs} is taken smaller and N is taken larger. Further experiments show lack of robustness of the CNMS scheme when γ is sufficiently small.

In Table 5, the performance of the NCG scheme and its accelerated version, C-NCG, are compared. We see a dramatic improvement with the C-NCG version especially for moderate values of γ. Taking smaller γ the optimal control problem becomes more ill-conditioned (stiff) and more computational effort is required for convergence.

We now discuss the effect of different choices of values of the optimization parameters using the results reported in Table 5. As required we see that smaller values of $|\psi(T) - \psi_d|_{\mathbb{C}^3}$ are attained for smaller γ. We remark that $\gamma = 10^{-7}$ is quite small and that makes the problem ill-conditioned. The NCG algorithm appears to be robust with respect to changes of γ. We also can see the effect of the

	$N = 2048$		$N = 4096$	
tol_{abs}	CPU(NCG)	CPU(CNMS)	CPU(NCG)	CPU(CNMS)
10^{-4}	1.17	1.28	2.32	1.39
10^{-5}	4.32	12.63	9.26	15.92
10^{-6}	5.01	48.00	17.21	*no conv*

Table 4: Computational efforts of the NCG scheme and the CNMS scheme for different choices of tolerance.

	$\gamma = 10^{-4}$		$\gamma = 10^{-6}$	
N	CPU(NCG)	CPU(C-NCG)	CPU(NCG)	CPU(C-NCG)
4096	40.54	6.26	254.70	58.10
8192	112.57	12.71	319.46	134.00
16384	312.17	27.42	626.84	279.46

Table 5: Computational effort to solve for $tol_{abs} = 10^{-6}$; $\gamma_0 = 0.01$, $\alpha_3 = 0.05$; in C-NCG coarsest level $N = 1024$.

γ	μ	α	$\lvert\psi(T) - \psi_d\rvert_{\mathbb{C}^3}$	J	CPU
10^{-7}	10^{-7}	0.05	$8.6 \cdot 10^{-4}$	$2.37 \cdot 10^{-3}$	19.6
10^{-7}	10^{-9}	0.05	$3.7 \cdot 10^{-4}$	$5.46 \cdot 10^{-4}$	55.6
10^{-7}	0	0.05	$6.9 \cdot 10^{-5}$	$1.41 \cdot 10^{-4}$	424.8
10^{-7}	0	0	$1.2 \cdot 10^{-3}$	$2.33 \cdot 10^{-6}$	763.1
10^{-4}	10^{-4}	0.05	$3.3 \cdot 10^{-2}$	$6.52 \cdot 10^{-2}$	47.3
10^{-4}	10^{-6}	0.05	$4.4 \cdot 10^{-3}$	$9.03 \cdot 10^{-3}$	42.3
10^{-4}	0	0.05	$2.7 \cdot 10^{-3}$	$5.68 \cdot 10^{-3}$	17.2
10^{-4}	0	0	$8.3 \cdot 10^{-3}$	$3.34 \cdot 10^{-4}$	5.5

Table 6: Optimization results depending on optimization parameters, $tol_{abs} = 10^{-7}$.

regularization parameter μ. As μ increases, $|\psi(T) - \psi_d|_{\mathbb{C}^3}$ increases, demonstrating that the additional smoothness of the control function (slightly) reduces the capability of tracking. Apparently, larger μ makes the problem behaving better, resulting in a smaller number of iterations. Concerning the parameter $\alpha = \alpha_3$ we obtain better tracking for nonzero α. This is expected since we have $|\psi_d|_{\mathbb{C}^3} = 1$ whereas $|\psi(T)|_{\mathbb{C}^3} < 1$ whenever $\gamma_0 > 0$, because of dissipation. By taking $\alpha > 0$ dissipation is reduced and therefore better $|\psi(T) - \psi_d|_{\mathbb{C}^3}$ is possibly achieved.

6 Conclusions and outlook

A representative optimal control problem for finite-level quantum systems was formulated and investigated. First-order necessary optimality conditions and second-order sufficient optimality conditions were discussed.

To solve this problem a new cascadic non-linear conjugate gradient (C-NCG) scheme was proposed and compared with state-of-the-art monotonic schemes. Convergence of the NCG scheme was discussed. Results of numerical experiments were reported to demonstrate the efficiency and robustness of the proposed approach.

The formulation of the C-NCG scheme appears independent of the dimensionality of the problem. It is the purpose of a forthcoming work to extend this method to infinite-dimensional quantum optimal control problems.

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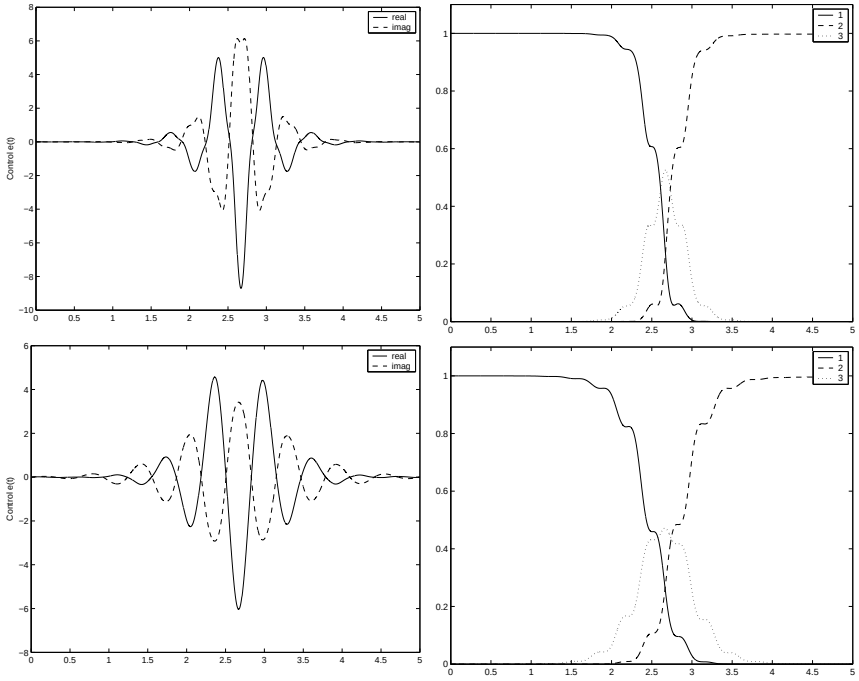

Figure 3: Optimal solutions for $\gamma = 10^{-4}$ and $\mu = 0$ (top) and $\mu = 10^{-6}$ (bottom) $\alpha_3 = 0.01$. Left the control field, right the wave function.

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