A method for solving exact-controllability problems governed by closed quantum spin systems

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The Liouville-von Neumann master equation models closed quantum spin systems that arise in Nuclear Magnetic Resonance applications. In this paper, an efficient and robust computational framework to solve exact-controllability problems governed by the Liouville-von Neumann master equation, that models control quantum spin systems, is presented. The proposed control framework is based on a new optimization-based formulation of exact-controllability quantum spin problems that allows the application of efficient computational techniques. This formulation results in an optimality system with four differential equations and an optimality condition. The differential equations are approximated with an appropriate modified Crank-Nicholson scheme and the resulting discretized optimality system is solved with a matrix-free Krylov-Newton scheme combined with a cascadic NCG initialization. Results of numerical experiments demonstrate the ability of the proposed framework to solve quantum spin exact-controllability control problems.

Keywords: Quantum spin systems; Liouville-von Neumann master equation; exact controllability problem; optimal control theory; optimality conditions; modified Crank-Nicholson scheme; Krylov-Newton scheme.

Introduction

In many scientific fields of modern interest, the need of steering a dynamical system from an initial state to a given target state at a given final time arises. Such a steering is performed by means of specific functions, which are referred to as exact-control functions. Consequently, the corresponding dynamical system is said to be controlled. This class of problems is present in nuclear magnetic resonance (NMR) spectroscopy and quantum information processing; for a review see, e.g., (Borzì, 2012). In these applications the dynamical systems is the Liouville-von Neumann master (LvNM) equation, describing the time evolution of the density operator that represents a quantum state. In this paper, we focus on these quantum dynamical systems.

Mathematically, this class of problems is known as exact-controllability problems. This task can be regarded as a controllability problem and as an optimal control problem. A controllability problem aims to establish the reachability of a given target. On the other hand, an optimal control problem has the purpose of computing control functions such that an appropriate norm of the difference between the resulting terminal state of the dynamical system and a given target is minimized.

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Many theoretical results are available concerning controllability of quantum systems. In particular, general controllability results for bilinear systems evolving on Lie groups are given in (Jurdjevic et al., 1972). For controllability results regarding quantum systems see, e.g., (Albertini et al., 2002; D'Alessandro, 2003; Beauchard et al., 2010; Dirr et al., 2008). The problems to estimate a final time that guarantees controllability and an optimal time are studied in, e.g., (Agrachev et al., 2006; Dirr et al., 2006; Khaneja et al., 2002, 2001). We distinguish exact-controllability problems from optimal control problems where it is required to minimize a cost functional subject to the constraint given by a differential model. In the latter case, recent results (Borzì et al., 2008; Ditz et al., 2008; Ho et al., 2010; Khaneja et al., 2005) show that optimization techniques can be successfully applied, while in the exact-controllability case much less is known on how to solve efficiently these problems. This is the focus of this paper: we develop an efficient strategy capable to solve exact-controllability quantum spin problems governed by the LvNM equation.

Pioneering works towards the development of quantum optimal control algorithms can be found in (Konnov et al., 1999; Tannor et al., 1993; Zhu et al., 1998). Further progresses in the development of efficient control schemes are documented in, e.g., (Eitan et al., 2011; Ho et al., 2010; Khaneja et al., 2005; Maday et al., 2007; Maximov et al., 2010; Sklarz et al., 2002). Advanced optimization methods for quatum control problems are discussed in (Borzì et al., 2008; Fouquieres et al., 2011).

The aim of our work is to develop an efficient and robust computational framework capable to solve exact-controllability problems governed by the LvNM equation that models closed quantum spin systems. For this purpose, we reformulate the exact-controllability problem in such a way that it is suitable for application of efficient optimization techniques. We focus on NMR spectroscopy applications, where the need arises to determine the radiofrequencies of magnetic control fields to be applied in such a way to excite particular quantum spin states to reach given target configurations.

Our work is organized in four main sections. In Section 1, we discuss the formulation of an exact-controllability problem. Section 2 focuses on the reformulation of this control problem. We discuss the relationship between the original control problem and its new reformulation. Moreover, we derive the optimality system and give a detailed discussion on the Hessian operator corresponding to the new formulation. In the new setting, we are able to prove regularity properties of the Hessian operator. This theoretical result is fundamental to guarantee an efficient behaviour of the optimization algorithm. In Section 3, we address the problem of computing numerical solutions of our new formulation of quantum spin control problems. We present a Krylov-Newton method, including implementation details. Moreover, we discuss the nonlinear conjugate gradient (NCG) method combined with a cascadic approach to obtain an accurate initialization to the Newton method. Further, a modified Crank-Nicholson method and the first-discretize-then-optimize strategy are presented as an adequate discretization framework of the optimality system that characterizes the first-order optimality conditions. Section 4 validates the proposed computational framework with three applications, demonstrating the ability of our method to solve quantum spin exact-controllability problems. A section of conclusion completes this work.

1. Exact-controllability of quantum spin systems

Problems where control functions are sought that are capable to steer a dynamical system from an initial state to a given target state at a given final time, are referred to as exact-controllability problems. Recent works focusing on this class of control problems are (Agrachev et al., 2006; Albertini et al., 2002; Dirr et al., 2008; Jurdjevic et al., 1972; Khaneja et al., 2001, 2005).

In many applications, including NMR spectroscopy, the corresponding dynamical system has a bilinear control structure given by (Cavanagh et al., 2007),

$$\dot{x} = \left[A + \sum_{n=1}^{N_C} B_n u_n \right] x \ , \tag{1}$$

where $A \in \mathrm{R}^{N\times N}$ is the drift matrix, $B_n \in \mathrm{R}^{N\times N}$ are the input matrices, N is the dimension of the differential system, N_C is the number of controls, x is the state and u is the control vector function. In this paper, we focus on closed quantum spin systems, where (1) represents a real matrix representation of the LvNM equation (Cavanagh et al., 2007). Hence, the matrices A and B_n are skew-symmetric and the dynamics of (1) is norm-preserving.

The exact-controllability problem associated with (1) is to find a control vector function u such that the following problem is solved:

$$\dot{x} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right] x \ , \quad t \in (0,T] \ , \quad x(0) = x_0 \ , \ x(T) = x_T \ . \tag{2}$$

Notice that, since (2) is a time-boundary-value problem, it is possible to solve it using the class shooting methods (Stoer and Bulirsch, 1993). However, (2) admits many solutions, and it becomes necessary to complement the problem with a constraint on u.

A suitable way to constraint the controls is to consider (2) embedded in an optimization problem. For this reason, we focus on the following

$$\begin{aligned} &\min_{x,u} \quad J(x,u) := \frac{1}{2}\sum_{n=1}^{N_C} \|u_n\|_{L^2}^2 \\ &\text{s.t.} \quad \dot{x} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right] x \ , \ t \in (0,T] \ , \ x(0) = x_0 \ , \ x(T) = x_T \\ &\qquad x \in H^1((0,T);\mathrm{R}^N) \text{ and } u \in L^2((0,T);\mathrm{R}^{N_C}) \ . \end{aligned} \tag{3}$$

We notice that (3) admits a solution if the target x_T belongs to the set of all points reachable at time T from the given starting point x_0. We remark that problems (2) and (3) are not equivalent. A solution of (3) is a minimum L^2-norm solution and solves also (2). On the other hand, a solution of (2) is not necessarily a solution to (3).

Controllability theory is fundamental in addressing problems (2) and (3). Notice that (1) is the induced system of a bilinear control system evolving on the Lie group SO. Moreover, a real representation of the LvNM equation can also be considered, whose induced system evolves on the Lie group of unitary operators SU. Hence, there are several results providing necessary and sufficient conditions for controllability; see, e.g., (Dirr et al., 2008). Now, we remark that, for quantum spin systems, there exist few results concerning the estimation of a time T capable to guarantee the existence of a control steering the trajectory to the given target in exactly T-units of time (Agrachev et al., 2006; Dirr et al., 2006, 2008; Khaneja et al., 2002, 2001). However, since the mentioned Lie groups are compact and semisimple (Hall, 2003), we can make use of Theorem 7.2 in (Jurdjevic et al., 1972) which guarantees controllability at T-units of time choosing a sufficiently large $T > 0$. For this reason, we make the following assumption.

Assumption 1: *The target point x_T belongs to the reachable set, that is the set of all points reachable from the given initial condition x_0. Moreover, the time T is assumed to be large enough to guarantee controllability in T-units of time.*

Further, we need the following assumption regarding the existence of Lagrange multipliers corresponding to problem (3).

Assumption 2: *There exist Lagrange multipliers $p_T \in \mathrm{R}^N$ and $p \in H^1((0,T);\mathrm{R}^N)$ corresponding to the constraint equation of the optimization problem (3). Moreover, p satisfies the following adjoint* *equation*

$$-\dot{p} = \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* p \ , \ \ t \in [0,T) \ \ , \ \ p(T) = p_T \ . \tag{4}$$

In Assumption 2, we assume that there exists a vector p_T such that the corresponding solution p is the Lagrange multiplier associated with the state x. Notice that p_T is unknown and p is uniquely determined by p_T and the control u. Further, notice that, once the existence of $p \in H^1((0,T);\mathrm{R}^N)$ is assumed, then (4) can be obtained by means of the standard Lagrange function approach.

A solution of (3) is characterized by the following first-order optimality system,

$$\dot{x} = \left[A + \sum_{n=1}^{N_C} B_n u_n \right] x \ \ , \ \ x(0) = x_0 \ \ , \ \ x(T) = x_T \tag{5a}$$

$$-\dot{p} = \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* p \ \ , \ p(T) = p_T \tag{5b}$$

$$u_n - \langle B_n x, p \rangle = 0 \ , \ n = 1, ..., N_C \ , \tag{5c}$$

where $\langle \cdot, \cdot \rangle$ represents the Euclidean scalar product.

Because of (5a), it is not clear how to solve (5). For this reason, in the next section, we reformulate (5) in such a way that it can be solved by using appropriate optimization techniques.

In this paper, we use the following notation. Given $m \in \mathrm{N}$, we denote with $\langle \cdot, \cdot \rangle$ the Euclidean inner product and with $\langle \cdot, \cdot \rangle_{L^2}$ the inner product defined by

$$\langle x, y \rangle_{L^2} := \int_0^T \langle x(t), y(t) \rangle dt \ , \ \text{for every } x, y \in L^2((0,T);\mathrm{R}^m) \ .$$

Moreover, $\| \cdot \|_2$ denotes the Euclidean norm and $\| \cdot \|_{L^2}$ denotes the norm induced by $\langle \cdot, \cdot \rangle_{L^2}$. Notice that m is equal to N for the state- and to N_C for the control-spaces. Consider any pair $a, b \in L^2((0,T);\mathrm{R}^{N_C}) \times \mathrm{R}^N$ given by $a = (a_1, a_2)$ and $b = (b_1, b_2)$, we define the inner product $(\cdot,\cdot)_G$ and the corresponding induced norm $||| \cdot |||$ is as follows

$$(a, b)_G := \sum_{n=1}^{N_C} \langle a_{1,n}, b_{1,n} \rangle_{L^2} + \langle a_2, b_2 \rangle \ , \ \text{and} \ , \ |||a||| := \sqrt{(a,a)_G} \ .$$

2. Reformulation of the exact-controllability spin problem

In this section, in order to address the exact-controllability of spin systems previously introduced, we define a new optimization problem, which is equivalent to (5) under certain conditions, but amenable to numerical optimization. First, we analyze the reformulated problem from a theoretical point of view and derive the corresponding optimality conditions. Then, we describe its reduced form, which is suitable for the numerical optimization. Further, the corresponding Hessian operator and its action are discussed.

In order to solve (5), we consider the map $\mathcal{G} : H^1((0,T);\mathrm{R}^N) \times L^2((0,T);\mathrm{R}^{N_C}) \times H^1((0,T);\mathrm{R}^N) \to$

$L^2((0,T);\mathrm{R}^{N_C}) \times \mathrm{R}^N$ defined as follows

$$\mathcal{G}(x,u,p) := \begin{pmatrix} u_1 - \langle B_1 x, p\rangle \\ \vdots \\ u_{N_C} - \langle B_{N_C} x, p\rangle \\ x(T) - x_T \end{pmatrix} . \tag{6}$$

Since this map is obtained by using the gradient component (5c) and the terminal condition of (5a), a triple (x,u,p) is a solution of (5), and a stationary point for (3), if and only if it is a root of $\mathcal{G}$ with x and p solutions to (5a) and (5b), respectively.

We remark that, it could be possible to compute a root for $\mathcal{G}$ using a Newton method, however, according to our experience, the corresponding Jacobian operator is not sufficiently regular to be used successfully in computational algorithms. For this reason, to compute a root (x,u,p) of $\mathcal{G}$, we define our main optimization problem as follows

$$\begin{aligned} \min_{x,u,p} \; G(x,u,p) &:= \frac{1}{2}|||\mathcal{G}(x,u,p)|||^2 = \frac{1}{2}\sum_{n=1}^{N_C} \|u_n - \langle B_n x, p\rangle\|^2_{L^2} + \frac{1}{2}\|x(T) - x_T\|_2^2 \\ \text{s.t.} \quad \dot{x} &= \left[A + \sum_{n=1}^{N_C} B_n u_n\right] x \; , \; t \in (0,T] \; , \; x(0) = x_0 \\ -\dot{p} &= \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* p \; , \; t \in [0,T) \; , \; p(T) = p_T \\ & x,p \in H^1((0,T);\mathrm{R}^N) \text{ and } u \in L^2((0,T);\mathrm{R}^{N_C}) \; , \end{aligned} \tag{7}$$

and we remark that a solution $(\hat{x},\hat{u},\hat{p})$ of (7) with $G(\hat{x},\hat{u},\hat{p}) = 0$ is a root of $\mathcal{G}$, hence a solution of the optimality system (5). Moreover, in the sequel of this paper we prove and discuss some regularity properties of (7), which are important for the numerical optimization and solution of (5), and useful for the characterization of stationary points of (3).

Further, we address the forward equation in x and backward equation in p as constraint equations of the minimization problem (7).

Existence and uniqueness of solutions $x,p \in H^1((0,T);\mathrm{R}^N)$ of the constraint equations of (7) for any $T > 0$ and any initial and terminal condition, corresponding to a given $u \in L^2((0,T);\mathrm{R}^{N_C})$, can be proved by standard results; see, e.g., (Sontag, 1998). Hence, the solutions of the constraints x and p are uniquely determined by the controls and the initial and terminal conditions, respectively. We have that $x = x(u,x_0)$ and $p = p(u,p_T)$. Consequently, we remark that the unknowns of (5) are the control $u \in L^2((0,T);\mathrm{R}^{N_C})$ and the terminal condition for the adjoint equation $p_T \in \mathrm{R}^N$.

In the following proposition, we state the existence of a solution of (7). Moreover, we analyze the relationship between the problems (3) and (7). In particular, the condition $G = 0$ is required to guarantee that a solution of (7) is a stationary point for (3).

Proposition 1: *A triple* $\big(x,u,p\big) \in H^1((0,T);\mathrm{R}^N) \times L^2((0,T);\mathrm{R}^{N_C}) \times H^1((0,T);\mathrm{R}^N)$*, with* $x = x(u,x_0)$ *and* $p = p(u,p_T)$*, is a solution of* (7) *with* $G(x,u,p) = 0$ *if and only if it is a stationary point of* (3)*.*

The proof of Proposition 1 is omitted for brevity. We remark that a solution of (7) with $G = 0$ is only a stationary point for (3), hence it is not guaranteed that it is a minimum norm solution of (3).

2.1 *Optimality system and necessary conditions*

In this section, we discuss the optimality conditions used to characterize a solution to (7). To obtain the first-order optimality system, we follow the Lagrange multipliers approach. We denote with $y, q \in H^1((0,T);\mathrm{R}^N)$ the Lagrange multipliers corresponding to x and p, respectively. The existence of such functions can be ensured by means of known results (Sontag, 1998).

The Lagrange function corresponding to (7) is given by

$$\begin{aligned} L(x,u,p,y,q) = G(x,u,p) &+ \left\langle \dot{x} - \left[A + \sum_{n=1}^{N_C} B_n u_n\right] x, y \right\rangle_{L^2} \\ &+ \left\langle -\dot{p} - \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* p, q \right\rangle_{L^2} , \end{aligned} \tag{8}$$

By means of (8), the optimality conditions for (7) are given by the following proposition.

Proposition 2: *The optimality system corresponding to* (7) *is given by*

$$\dot{x} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right] x \ , \ \ x(0) = x_0 \ , \tag{9a}$$

$$-\dot{p} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* p \ , \ p(T) = p_T \ , \tag{9b}$$

$$-\dot{y} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* y + \sum_{n=1}^{N_C} \Big[(u_n - \langle B_n x, p\rangle) B_n^* p\Big] \ , \ \ y(T) = -(x(T) - x_T) \ , \tag{9c}$$

$$\dot{q} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right] q + \sum_{n=1}^{N_C} \Big[(u_n - \langle B_n x, p\rangle) B_n x\Big] \ , \ \ q(0) = 0 \ , \tag{9d}$$

$$u_n - \langle B_n x, p\rangle - \langle B_n x, y\rangle - \langle B_n^* p, q\rangle = 0 \ , \ n = 1, ..., N_C \ , \tag{9e}$$

where (9a) *and* (9b) *are the constraint equations,* (9c) *and* (9d) *are the corresponding adjoint equations, and* (9e) *gives the components of the gradient.*

Proof. Since $L(x,u,p,y,q)$ is linear with respect to the adjoint variables y and q, we obtain the constraint equations (9a) and (9b) as follows

$$\Big\langle \nabla_y L(x,u,p,y,q), \delta y \Big\rangle_{L^2} = \left\langle \dot{x} - \left[A + \sum_{n=1}^{N_C} B_n u_n\right] x, \delta y \right\rangle_{L^2} ,$$

and

$$\Big\langle \nabla_q L(x,u,p,y,q), \delta q \Big\rangle_{L^2} = \left\langle -\dot{p} - \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* p, \delta q \right\rangle_{L^2} .$$

For optimality, the two inner products $\Big\langle \nabla_y L(x,u,p,y,q), \delta y \Big\rangle_{L^2}$ and $\Big\langle \nabla_q L(x,u,p,y,q), \delta q \Big\rangle_{L^2}$ have to be equal to zero for all $\delta y \in L^2((0,T);\mathrm{R}^N)$ and $\delta q \in L^2((0,T);\mathrm{R}^N)$, respectively, thus (9a) and (9b) follow.

To obtain the adjoint equations (9c) and (9d), we consider the derivative with respect to x and p along the two directions δx and δp, respectively. We obtain (9c), as follows

$$\Big\langle \nabla_x L(x,u,p,y,q), \delta x \Big\rangle_{L^2} = \langle \delta x(T), x(T) - x_T \rangle$$

$$+ \int_0^T \langle \dot{\delta x} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right] \delta x, y \rangle dt - \int_0^T \langle \sum_{n=1}^{N_C} (u_n - \langle B_n x, p \rangle) B_n \delta x, p \rangle dt$$

$$= \langle \delta x(T), x(T) - x_T \rangle + \big[\langle \delta x, y \rangle \big]_0^T + \int_0^T \langle -\dot{y} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* y - \sum_{n=1}^{N_C} (u_n - \langle B_n x, p \rangle) B_n^* p, \delta x \rangle dt$$

$$= \langle \delta x(T), x(T) - x_T \rangle + \big[\langle \delta x, y \rangle \big]_0^T + \Big\langle -\dot{y} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* y - \sum_{n=1}^{N_C} (u_n - \langle B_n x, p \rangle) B_n^* p, \delta x \Big\rangle_{L^2} .$$

Since the product $\Big\langle \nabla_x L(x,u,p,y,q), \delta x \Big\rangle_{L^2}$ has to be equal to zero for all $\delta x \in L^2((0,T); \mathrm{R}^N)$, and we have that $\delta x(0) = 0$, we obtain the terminal condition $y(T) = -(x(T) - x_T)$ and the adjoint equation (9c).

To obtain the adjoint problem (9d), we proceed as follows

$$\Big\langle \nabla_p L(x,u,p,y,q), \delta p \Big\rangle_{L^2}$$

$$= \int_0^T \langle -\dot{\delta p} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* \delta p, q \rangle dt - \int_0^T \langle \sum_{n=1}^{N_C} (u_n - \langle B_n x, p \rangle) B_n x, \delta p \rangle dt$$

$$= -\big[\langle \delta p, q \rangle \big]_0^T + \int_0^T \langle \dot{q} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* q - \sum_{n=1}^{N_C} (u_n - \langle B_n x, p \rangle) B_n x, \delta p \rangle dt$$

$$= -\big[\langle \delta p, q \rangle \big]_0^T + \Big\langle \dot{q} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right] q - \sum_{n=1}^{N_C} (u_n - \langle B_n x, p \rangle) B_n x, \delta p \Big\rangle_{L^2} .$$

The product $\Big\langle \nabla_p L(x,u,p,y,q), \delta p \Big\rangle_{L^2}$ has to be equal to zero for all $\delta p \in L^2((0,T); \mathrm{R}^N)$ with $\delta p(T) = 0$. As a consequence, we have that $q(0) = 0$ and we obtain the adjoint equation (9d).

We derive the n-component of the gradient (9e) by means of the variation of the Lagrangian with respect to the control u_n as follows

$$\Big\langle \nabla_{u_n} L(x,u,p,y,q), \delta u_n \Big\rangle_{L^2} = \int_0^T (u_n - \langle B_n x, p \rangle) \delta u_n - \langle B_n x, y \rangle \delta u_n - \langle B_n^* p, q \rangle \delta u_n dt$$

$$= \Big\langle u_n - \langle B_n x, p \rangle - \langle B_n x, y \rangle - \langle B_n^* p, q \rangle, \delta u_n \Big\rangle_{L^2} .$$

Since this product has to be equal to zero for all $\delta u_n \in L^2(0,T)$, we obtain the optimality condition (9e). □

In the following proposition, we discuss existence and uniqueness of solutions to the adjoint problems (9c) and (9d).

Proposition 3: *Given y_T and q_0, consider the following problems*

$$-\dot{y} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* y + \sum_{n=1}^{N_C} \Big[(u_n - \langle B_n x, p\rangle) B_n^* p\Big] \ , \ \ y(T) = y_T \ , \tag{10}$$

and

$$\dot{q} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right] q + \sum_{n=1}^{N_C} \Big[(u_n - \langle B_n x, p\rangle) B_n x\Big] \ , \ \ q(0) = q_0 \ , \tag{11}$$

with $y, q, x, p \in H^1((0,T); \mathrm{R}^N)$ and $u \in L^2((0,T); \mathrm{R}^{N_C})$. Then (10) *and* (11) *admit unique solutions for any $T > 0$ and any y_T and q_0, respectively.*

Moreover, assume that $\big(x(u, x_0), u, p(u, p_T)\big)$ is a stationary point for (3)*, then the problems* (9c) *and* (9d) *admit the unique solutions $y(t) = 0$ and $q(t) = 0$, for all $t \in [0,T]$, for any $T > 0$ and any control $u \in L^2((0,T); \mathrm{R}^{N_C})$.*

Proof. Existence and uniqueness of solution of (10) and (11) can be proved by means of known results; see, e.g., (Sontag, 1998).

Next, consider problem (9d). Since $\big(x(u, x_0), u, p(u, p_T)\big)$ is a stationary point for (3), we have that $u_n - \langle B_n x, p\rangle = 0$, for $n = 1, ..., N_C$; hence, the forcing terms in the differential equations in (9c) and (9d) are zero. Consequently, since A and B_n are skew-symmetric, the dynamics are norm preserving, we have that (9c) and (9d) admit the unique solutions $y = 0$ and $q = 0$ for any $T > 0$ and any u.

□

Now, we discuss the reduced form of problem (7), which is suitable to be solved by means of appropriate numerical optimization methods. As mentioned in the previous section, the solutions of the constraint equations (9a) and (9b) are uniquely determined by the initial and terminal conditions, that are $x(0) = x_0$ and $p(T) = p_T$, respectively, and by the control vector function u. We have

$$x = x(u) \ \text{ and } \ p = p(u, p_T) \ , \tag{12}$$

where the dependence of x from x_0 is omitted because it is an input of the problem. Consequently, problem (7) can be equivalently expressed in the following reduced form

$$\begin{aligned} &\min_{u, p_T} \ \ G_r(u, p_T) := G(x(u), u, p(u, p_T)) \\ &\text{s.t.} \ \ (x(u), p(u, p_T)) \in \mathcal{S}_{ad} := \Big\{(x, p) \ \big| x \text{ solves (9a) and } p \text{ solves (9b)}\Big\} \ . \end{aligned} \tag{13}$$

We characterize a solution of (13) with the first-order optimality conditions given in the following result, which follows directly from Theorem 2.

Proposition 4: *The optimality system corresponding to problem* (13) *is given by*

$$\nabla_{u_n} G_r(u, p_T) = u_n - \langle B_n x, p\rangle - \langle B_n x, y\rangle - \langle B_n^* p, q\rangle = 0 \ \ , \ n = 1, ..., N_C \ , \tag{14a}$$

$$\nabla_{p_T} G_r(u, p_T) = -q(T) = 0 \ , \tag{14b}$$

such that x, p, y and q solve the following problems

$$\dot{x} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right] x \ , \ \ x(0) = x_0 \ , \tag{14c}$$

$$-\dot{p} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* p \ , \ p(T) = p_T \ , \tag{14d}$$

$$-\dot{y} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* y + \sum_{n=1}^{N_C}\Big[(u_n - \langle B_n x, p\rangle) B_n^* p\Big] \ , \ \ y(T) = -(x(T) - x_T) \ , \tag{14e}$$

$$\dot{q} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right] q + \sum_{n=1}^{N_C}\Big[(u_n - \langle B_n x, p\rangle) B_n x\Big] \ , \ \ q(0) = 0 \ . \tag{14f}$$

Proof. Consider Theorem 2 and its proof. We remark that the gradient component of the reduced problem with respect to p_T is obtained from the fact that $\langle \nabla_{p_T} G_r(u, p_T), \delta q(T)\rangle = \langle -q(T), \delta q(T)\rangle = 0$ for all $\delta q(T)$. Notice that, unlike in (7), in (13) p_T is not fixed, hence $\delta q(T)$ is not fixed to 0. □

Next, we study the Hessian operator. In particular, we are interested in the reduced Hessian operator, corresponding to (13). To obtain it, we first discuss the Hessian of problem (7), then we consider its reduced form corresponding to (13). In particular, we focus on its action on a given vector function. This aspect will be crucial in the development of the Krylov-Newton method discussed in the next section.

By computing the second directional derivative of the Lagrange function (8), we obtain that

$$\left\langle H(x,u,p)\begin{pmatrix}\delta x\\ \delta u\\ \delta p\\ \delta y\\ \delta q\end{pmatrix}, \begin{pmatrix}\delta x\\ \delta u\\ \delta p\\ \delta y\\ \delta q\end{pmatrix}\right\rangle_{L^2} = \left\langle \begin{pmatrix}H_x\\ H_u\\ H_p\\ H_y\\ H_q\end{pmatrix}, \begin{pmatrix}\delta x\\ \delta u\\ \delta p\\ \delta y\\ \delta q\end{pmatrix}\right\rangle_{L^2} , \tag{15}$$

where $(\delta x, \delta u, \delta p, \delta y, \delta q)^T \in H^1((0,T);\mathrm{R}^N) \times L^2((0,T);\mathrm{R}^{N_C}) \times H^1((0,T);\mathrm{R}^N) \times H^1((0,T);\mathrm{R}^N) \times H^1((0,T);\mathrm{R}^N)$ and H_x, H_u, H_p, H_y and H_q denote the following

$$\begin{aligned} H_x = & -\dot{\delta y} - \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* \delta y - \left[\sum_{n=1}^{N_C} B_n \delta u_n\right]^* y - \sum_{n=1}^{N_C}(u_n - \langle B_n x, p\rangle) B_n^* \delta p \\ & - \sum_{n=1}^{N_C}(\delta u_n - \langle B_n \delta x, p\rangle - \langle B_n x, \delta p\rangle) B_n^* p \, , \text{with } \delta y(T) = -\delta x(T) \ , \end{aligned} \tag{16}$$

$$\begin{aligned} H_{u_n} = \ & \delta u_n - \langle B_n \delta x, p\rangle - \langle B_n x, \delta p\rangle - \langle B_n \delta x, y\rangle \\ & - \langle B_n x, \delta y\rangle - \langle B_n^* \delta p, q\rangle - \langle B_n^* p, \delta q\rangle \ , \end{aligned} \tag{17}$$

$$
\begin{aligned}
H_p = \dot{\delta q} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right] \delta q - \left[\sum_{n=1}^{N_C} B_n \delta u_n \right] q - \sum_{n=1}^{N_C} (u_n - \langle B_n x, p \rangle) B_n \delta x \\
- \sum_{n=1}^{N_C} (\delta u_n - \langle B_n \delta x, p \rangle - \langle B_n x, \delta p \rangle) B_n x \text{ , with } \delta q(0) = 0 \text{ ,}
\end{aligned}
\tag{18}
$$

$$
H_y = \dot{\delta x} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right] \delta x - \left[\sum_{n=1}^{N_C} B_n \delta u_n \right] x \text{ , with } \delta x(0) = 0 \text{ ,} \tag{19}
$$

and

$$
H_q = -\dot{\delta p} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* \delta p - \left[\sum_{n=1}^{N_C} B_n \delta u_n \right]^* p \text{ , with } \delta p(T) = \delta p_T \text{ ,} \tag{20}
$$

Notice that H_x, H_u, H_p, H_y and H_q represent the residuals of the linearized optimality system.

Now, we consider the reduced problem (13) and we denote with $\nabla^2 G_r(x, u, p)$ the reduced Hessian operator. We recall that the unknowns are the control u and the terminal condition p_T. Consequently, the action of $\nabla^2 G_r(x, u, p)$ on a vector $(\delta u, \delta p_T)^T \in L^2((0,T); \mathrm{R}^{N_C}) \times \mathrm{R}^N$ is given by the following

$$
\nabla^2 G_r(u, p_T) \begin{pmatrix} \delta u_1 \\ \vdots \\ \delta u_{N_C} \\ \delta p_T \end{pmatrix} = \begin{pmatrix} H_{u_1}(x, u, p) \\ \vdots \\ H_{u_{N_C}}(x, u, p) \\ H_{p_T}(x, u, p) \end{pmatrix} \tag{21}
$$

where δx, δp, δy and δq are solutions obtained by cancelling (19), (20), (16) and (18), respectively, and $H_{p_T}(x, u, p) = -\delta q(T)$. Hence, the action of the reduced Hessian operator can be obtained by solving the linearized equations and assembling (21).

With the following theorem, we prove regularity of the reduced Hessian operator.

Theorem 1: *Let (u, p_T) be a solution of* (13) *with $G_r(u, p_T) = 0$, then the reduced Hessian operator $\nabla^2 G_r(u, p_T)$ is positive semi-definite.*

Proof. We denote with $x = x(u)$ and $p = p(u, p_T)$ the unique solutions of the constraint equations (14c) and (14d), respectively, and with $y = y(x, u, p)$ and $q = q(x, u, p)$, the unique solutions of the adjoint equations (14e) and (14f), respectively. We prove the claim in two steps.

Step 1: since (u, p_T) is a solution of (13) with $G_r(u, p_T) = 0$, then we have that $u_n - \langle B_n x, p \rangle = 0$ for $n = 1, \ldots, N_C$. Moreover, by Proposition 3, we know that $y = 0$ and $q = 0$. Consequently, the linearized adjoint equations $H_x = 0$ and $H_p = 0$ become as follows

$$
-\dot{\delta y} = \left[A + \sum_{n=1}^{N_C} u_n B_n \right]^* \delta y + \sum_{n=1}^{N_C} \big(\delta u_n - \langle B_n \delta x, p \rangle - \langle B_n x, \delta p \rangle \big) B_n^* p \text{ ,} \tag{22}
$$

with $\delta y(T) = -\delta x(T)$, and

$$\dot{\delta q} = \left[A + \sum_{n=1}^{N_C} u_n B_n \right] \delta q + \sum_{n=1}^{N_C} \big(\delta u_n - \langle B_n \delta x, p \rangle - \langle B_n x, \delta p \rangle \big) B_n x \ , \tag{23}$$

with $\delta q(0) = 0$. Now, define $\mathcal{O}(u) : H^1((0,T); \mathrm{R}^N) \to L^2((0,T); \mathrm{R}^N)$

$$\mathcal{O}(u) := \frac{d}{dt} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right] , \tag{24}$$

whose adjoint is given by

$$\mathcal{O}(u)^* = -\frac{d}{dt} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* . \tag{25}$$

Recall that $H_y = 0$ and $H_p = 0$, we have

$$\begin{aligned} \mathcal{O}(u)(\delta x + \delta q) &= \dot{\delta x} + \dot{\delta q} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right] \Big(\delta x + \delta q \Big) \\ &= \sum_{n=1}^{N_C} \delta u_n B_n x + \sum_{n=1}^{N_C} \Big(\delta u_n - \langle \delta x, B_n^* p \rangle - \langle x, B_n^* \delta p \rangle \Big) B_n x \\ &= \sum_{n=1}^{N_C} \Big(2\delta u_n - \langle \delta x, B_n^* p \rangle - \langle x, B_n^* \delta p \rangle \Big) B_n x \ , \end{aligned} \tag{26}$$

and analogously, using $H_x = 0$ and $H_q = 0$, we have

$$\begin{aligned} \mathcal{O}(u)^*(\delta p + \delta y) &= -\dot{\delta p} - \dot{\delta y} - \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* \Big(\delta p + \delta y \Big) \\ &= \sum_{n=1}^{N_C} \Big(2\delta u_n - \langle \delta x, B_n^* p \rangle - \langle x, B_n^* \delta p \rangle \Big) B_n^* p \ . \end{aligned} \tag{27}$$

Step 2: using (21) and (17) and the fact that $y=0$ and $q=0$, we have

$$
\begin{aligned}
&\left\langle \nabla^2 G_r(u,p_T)\begin{pmatrix}\delta u\\ \delta p_T\end{pmatrix}, \begin{pmatrix}\delta u\\ \delta p_T\end{pmatrix}\right\rangle_{L^2} = \\
&= \left\langle \begin{pmatrix} \delta u_1 - \langle B_1\delta x, p\rangle - \langle B_1 x, \delta p\rangle - \langle B_1 x, \delta y\rangle - \langle B_1^* p, \delta q\rangle \\ \vdots \\ \delta u_{N_C} - \langle B_{N_C}\delta x, p\rangle - \langle B_{N_C} x, \delta p\rangle - \langle B_{N_C} x, \delta y\rangle - \langle B_{N_C}^* p, \delta q\rangle \\ -\delta q(T)\end{pmatrix}, \begin{pmatrix}\delta u_1\\ \vdots \\ \delta u_{N_C}\\ \delta p_T\end{pmatrix}\right\rangle_{L^2} \\
&= \left\langle \begin{pmatrix} \delta u_1 - \langle B_1 x, \delta p + \delta y\rangle - \langle B_1^* p, \delta x + \delta q\rangle \\ \vdots \\ \delta u_{N_C} - \langle B_{N_C} x, \delta p + \delta y\rangle - \langle B_{N_C}^* p, \delta x + \delta q\rangle \\ -\delta q(T)\end{pmatrix}, \begin{pmatrix}\delta u_1\\ \vdots \\ \delta u_{N_C}\\ \delta p_T\end{pmatrix}\right\rangle_{L^2} \\
&= -\langle \delta q(T), \delta p_T\rangle + \sum_{n=1}^{N_C}\int_0^T \delta u_n^2 dt - \sum_{n=1}^{N_C}\int_0^T \big(\langle B_n x, \delta p + \delta y\rangle + \langle B_n^* p, \delta x + \delta q\rangle\big)\delta u_n dt \\
&= -\langle \delta q(T), \delta p_T\rangle + \sum_{n=1}^{N_C}\int_0^T \delta u_n^2 dt - \int_0^T \Big(\langle \sum_{n=1}^{N_C}\delta u_n B_n x, \delta p + \delta y\rangle + \langle \sum_{n=1}^{N_C}\delta u_n B_n^* p, \delta x + \delta q\rangle\Big) dt \\
&= -\langle \delta q(T), \delta p_T\rangle + \sum_{n=1}^{N_C}\int_0^T \delta u_n^2 dt - \int_0^T \Big(\langle \mathcal{O}(u)(\delta x), \delta p + \delta y\rangle + \langle \mathcal{O}(u)^*(\delta p), \delta x + \delta q\rangle\Big) dt\ ,
\end{aligned}
\tag{28}
$$

the latter equation follows from $H_y = 0$, $H_q = 0$ and (24) and (25). Now, integrating by parts, we obtain

$$
\begin{aligned}
&\left\langle \nabla^2 G_r(u,p_T)\begin{pmatrix}\delta u\\ \delta p_T\end{pmatrix}, \begin{pmatrix}\delta u\\ \delta p_T\end{pmatrix}\right\rangle_{L^2} = \\
&= -\langle \delta q(T), \delta p_T\rangle + \sum_{n=1}^{N_C}\int_0^T \delta u_n^2 dt - \big[\langle \delta x, \delta p + \delta y\rangle\big]_0^T - \int_0^T \langle \delta x, \mathcal{O}(u)^*(\delta p + \delta y)\rangle dt \\
&+ \big[\langle \delta p, \delta x + \delta q\rangle\big]_0^T - \int_0^T \langle \delta p, \mathcal{O}(u)(\delta x + \delta q)\rangle dt \\
&= -\langle \delta q(T), \delta p_T\rangle + \sum_{n=1}^{N_C}\int_0^T \delta u_n^2 dt - \langle \delta x(T), \delta p_T + \delta y(T)\rangle - \int_0^T \langle \delta x, \mathcal{O}(u)^*(\delta p + \delta y)\rangle dt \\
&+ \langle \delta p_T, \delta x(T) + \delta q(T)\rangle - \int_0^T \langle \delta p, \mathcal{O}(u)(\delta x + \delta q)\rangle dt \\
&= -\langle \delta x(T), \delta y(T)\rangle + \sum_{n=1}^{N_C}\int_0^T \delta u_n^2 dt - \int_0^T \langle \delta x, \mathcal{O}(u)^*(\delta p + \delta y)\rangle dt - \int_0^T \langle \delta p, \mathcal{O}(u)(\delta x + \delta q)\rangle dt \\
&= \|\delta x(T)\|_2^2 + \sum_{n=1}^{N_C}\int_0^T \delta u_n^2 dt - \int_0^T \langle \delta x, \sum_{n=1}^{N_C}\Big(2\delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^*\delta p\rangle\Big) B_n^* p\rangle dt \\
&- \int_0^T \langle \delta p, \sum_{n=1}^{N_C}\Big(2\delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^*\delta p\rangle\Big) B_n x\rangle dt
\end{aligned}
\tag{29}
$$

where we used (26) and (27) and the fact that $\delta y(T) = -\delta x(T)$. We have the following

$$\begin{aligned}
&\left\langle \nabla^2 G_r(u, p_T) \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix}, \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix} \right\rangle_{L^2} = \\
&-\sum_{n=1}^{N_C} \int_0^T \Big(2\delta u_n - \langle \delta x, B_n^* p \rangle - \langle x, B_n^* \delta p \rangle \Big) \langle \delta p, B_n x \rangle dt \\
&= \|\delta x(T)\|_2^2 + \sum_{n=1}^{N_C} \int_0^T \Big[\delta u_n^2 - 2\delta u_n \langle \delta x, B_n^* p \rangle + \Big(\langle \delta x, B_n^* p \rangle + \langle x, B_n^* \delta p \rangle \Big) \langle \delta x, B_n^* p \rangle \\
&- 2\delta u_n \langle \delta p, B_n x \rangle + \Big(\langle \delta x, B_n^* p \rangle + \langle x, B_n^* \delta p \rangle \Big) \langle \delta p, B_n x \rangle \Big] dt \\
&= \|\delta x(T)\|_2^2 + \sum_{n=1}^{N_C} \int_0^T \Big(\delta u_n - \langle \delta x, B_n^* p \rangle - \langle x, B_n^* \delta p \rangle \Big)^2 dt \, ,
\end{aligned} \tag{30}$$

which implies that

$$\left\langle \nabla^2 G_r(u, p_T) \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix}, \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix} \right\rangle_{L^2} = \|\delta x(T)\|_2^2 + \sum_{n=1}^{N_C} \|\delta u_n - \langle \delta x, B_n^* p \rangle - \langle x, B_n^* \delta p \rangle\|_{L^2}^2 \, . \tag{31}$$

Consequently, we have

$$\left\langle \nabla^2 G_r(u, p_T) \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix}, \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix} \right\rangle_{L^2} \geq 0 \;\; , \;\; \forall (\delta u, \delta p_T) \, . \tag{32}$$

□

2.2 *Complementary results on coercivity and second-order sufficient conditions*

A property that plays an important role in the solution of optimization problems is the coercivity of the reduced Hessian operator. From the coercivity property, two benefits arise: first, coercivity is a second-order sufficient optimality condition; second, coercivity implies regularity of the Hessian operator, which guarantees an optimal behaviour, namely, a superlinear or quadratic convergence, of second-order optimization algorithms in a neighborhood of a minimum point.

In this section, the coercivity of the reduced Hessian operator (21) is discussed. According to Theorem 1, the Hessian in (21) is positive-semidefinite for all the pair $(\delta u, \delta p_T)$. To improve this result, we characterize in Corollary 1 the set of all points in which (21) is indefinite and we discuss the relationship between (21) and the end-point map $\delta u \mapsto \delta x(T; \delta u)$. Next, we provide sufficient conditions for the coercivity of the reduced Hessian operator (21), see Theorem 2 and Corollary 2.

In the sequel, we denote with $\| \cdot \|$ the Hilbert-Schmidt norm.

Corollary 1: *Consider the assumptions of Theorem 1. Then, we have that*

$$\left\langle \nabla^2 G_r(u, p_T) \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix}, \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix} \right\rangle_{L^2} = 0 \, , \tag{33}$$

for all $(\delta u, \delta p_T)$ *belonging to a convex neighborhood of* $(0, 0)$*. Moreover, if the map* $\delta u \mapsto \delta x(T; \delta u)$ *is injective in a neighborhood* $\mathcal{N}$ *of* $\delta u = 0$*, then* $\nabla^2 G_r(u, p_T)$ *is positive definite in* $\mathcal{N}$*.*

Proof. To prove the first claim, we consider the following optimization problem

$$
\begin{aligned}
\min_{\delta u, \delta p_T} \quad & F(\delta u, \delta p_T) := \|\delta x(T)\|_2^2 + \sum_{n=1}^{N_C} \|\delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^* \delta p\rangle\|_{L^2}^2 \\
\text{s.t.} \quad & \dot{\delta x} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right] \delta x + \left[\sum_{n=1}^{N_C} B_n \delta u_n\right] x \ , \ \delta x(0) = 0 \\
& -\dot{\delta p} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right]^* \delta p + \left[\sum_{n=1}^{N_C} B_n \delta u_n\right]^* p \ , \ \delta p(T) = \delta p_T \\
& (\delta u, \delta p_T) \in S \subset L^2((0,T); \mathrm{R}^N) \times \mathrm{R}^{N_C} \ ,
\end{aligned}
\tag{34}
$$

where $(x(u), u, p(u, p_T))$ is a solution of (13) with $G_r(u, p_T) = 0$ and S is closed, convex and bounded subset of $L^2((0,T); \mathrm{R}^N) \times \mathrm{R}^{N_C}$. The existence of a solution of (34) follows from the fact that $F(\delta u, \delta p_T) \geq 0$ and $F(0,0) = 0$. Hence $(\delta u, \delta p_T) = (0,0)$ is a global minimum of (34).

Now, notice that the maps $(\delta x, \delta u_n, \delta p) \mapsto \delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^* \delta p\rangle$ and $\delta u \mapsto \delta x(T; \delta u)$ preserve convex combinations. Hence the convexity of the norms implies that F is convex. Since, C is a convex set and F a convex function, then the set of global minima of F is convex. Consequently, we obtain that $F(\delta u, \delta p_T) = 0$ for all $(\delta u, \delta p_T)$ belonging to a convex neighborhood of $(0,0)$ included in S.

To prove the second argument, we consider the following. If $\delta u \mapsto \delta x(T; \delta u)$ is injective in a neighborhood $\mathcal{N}$ of $\delta u = 0$, then in $\mathcal{N}$ we have that $\delta u = 0$ if and only if $\|\delta x(T; \delta u)\|_2 = 0$. Consequently, the positive definiteness of (21) follows. □

Lemma 1: *Let $(\tilde{u}, \tilde{p}_T)$ be a solution of* (13) *with $G_r(\tilde{u}, \tilde{p}_T) = 0$. If $\tilde{p}_T = 0$, then $\tilde{u} = 0$, that is $(\tilde{u}, \tilde{p}_T)$ is a trivial solution of* (13).

Proof. Assuming that $\tilde{p}_T = 0$ and recalling that (9b) is norm preserving, we get that $\tilde{p}(t; \tilde{p}_T) = 0$ a.e. on $(0,T)$. Since $(\tilde{u}, \tilde{p}_T)$ be a solution of (13) with $G_r(\tilde{u}, \tilde{p}_T) = 0$, we have that $\tilde{u}_n = \langle B_n x, \tilde{p}\rangle$ for $n = 1, ..., N_C$. Consequently, we obtain that $\tilde{u} = 0$. □

Lemma 2: *Let (u, p_T) be a solution of* (13) *with $G_r(u, p_T) = 0$. Let δx and δp be the unique solutions of $H_y = 0$ and $H_q = 0$, respectively. Then the following estimates hold*

$$
\|\delta x\|_{L^2} \leq 2TM \|x(0)\|_2 \|\delta u\|_{L^2} \ , \tag{35}
$$

and

$$
\|\delta p\|_{L^2} \leq 2TM \|p_T\|_2 \|\delta u\|_{L^2} + \sqrt{T} \|\delta p_T\|_2 \ . \tag{36}
$$

where $M := \sum_{n=1}^{N_C} \|B_n\|$.

Proof. We start proving (35). Consider the linearized equation $H_y = 0$, that is

$$
\dot{\delta x} = \left[A + \sum_{n=1}^{N_C} B_n u_n\right] \delta x + \left[\sum_{n=1}^{N_C} B_n \delta u_n\right] x \ , \text{with } \delta x(0) = 0 \ . \tag{37}
$$

by multiplying (37) to the left with δx, we obtain

$$\langle \delta x, \dot{\delta x} \rangle = \langle \delta x, \left[A + \sum_{n=1}^{N_C} B_n u_n \right] \delta x \rangle + \langle \delta x, \left[\sum_{n=1}^{N_C} B_n \delta u_n \right] x \rangle \ . \tag{38}$$

Now, considering that $\langle \delta x(t), \dot{\delta x}(t) \rangle = \frac{1}{2} \frac{d}{dt} \| \delta x(t) \|_2^2$ and recalling the skew-symmetry of A and B_n, we get

$$\frac{1}{2} \frac{d}{dt} \| \delta x(t) \|_2^2 = \langle \delta x, \left[\sum_{n=1}^{N_C} B_n \delta u_n \right] x \rangle \ . \tag{39}$$

Integrating over $(0, t)$, and using that $\delta x(0) = 0$, (1) is norm preserving and the Cauchy-Schwarz inequality, we obtain

$$\begin{aligned}
\| \delta x(t) \|_2^2 &= 2 \int_0^t \langle \delta x, \left[\sum_{n=1}^{N_C} B_n \delta u_n \right] x \rangle dt = 2 \sum_{n=1}^{N_C} \int_0^t \delta u_n \langle \delta x, B_n x \rangle dt \\
&\leq 2 \sum_{n=1}^{N_C} \int_0^t |\delta u_n| |\langle \delta x, B_n x \rangle| dt \leq 2 \sum_{n=1}^{N_C} \int_0^T |\delta u_n| |\langle \delta x, B_n x \rangle| dt \\
&\leq 2 \sum_{n=1}^{N_C} \int_0^T |\delta u_n| \| \delta x \|_2 \| B_n x \|_2 dt \leq 2 \sum_{n=1}^{N_C} \int_0^T |\delta u_n| \| \delta x \|_2 \| B_n \| \| x(0) \|_2 dt \\
&\leq 2 \| x(0) \|_2 \sum_{n=1}^{N_C} \| B_n \| \int_0^T |\delta u_n| \| \delta x \|_2 dt \leq 2 \| x(0) \|_2 M \sum_{n=1}^{N_C} \int_0^T |\delta u_n| \| \delta x \|_2 dt \\
&\leq 2 \| x(0) \|_2 M \| \delta u \|_{L^2} \| \delta x \|_{L^2} \ .
\end{aligned} \tag{40}$$

where $M := \sum_{n=1}^{N_C} \| B_n \|$. Now, integrating over $(0, T)$, we obtain (35) as follows

$$\begin{aligned}
\int_0^T \| \delta x(t) \|_2^2 dt &\leq 2 \int_0^T \| x(0) \|_2 M \| \delta u \|_{L^2} \| \delta x \|_{L^2} dt \\
&\Rightarrow \| \delta x \|_{L^2}^2 \leq 2T \| x(0) \|_2 M \| \delta u \|_{L^2} \| \delta x \|_{L^2} \\
&\Rightarrow \| \delta x \|_{L^2} \leq 2T \| x(0) \|_2 M \| \delta u \|_{L^2} \ .
\end{aligned} \tag{41}$$

Next, we prove (36). Consider the linearized equation $H_y = 0$, that is

$$-\dot{\delta p} = \left[A + \sum_{n=1}^{N_C} B_n u_n \right]^* \delta p + \left[\sum_{n=1}^{N_C} B_n \delta u_n \right]^* p \text{ , with } \delta p(T) = \delta p_T \ . \tag{42}$$

By multiplying this equation from the left with δp and using the same arguments as above for δx, we have

$$
\begin{aligned}
\|\delta p(t)\|_2^2 &= \|\delta p(T)\|_2^2 - 2\int_t^T \langle \delta p, \left[\sum_{n=1}^{N_C} B_n \delta u_n\right] p\rangle dt \\
&\leq \|\delta p_T\|_2^2 + 2\sum_{n=1}^{N_C}\int_0^T |\delta u_n||\langle \delta p, B_n p\rangle| dt \\
&\leq \|\delta p_T\|_2^2 + 2\sum_{n=1}^{N_C}\int_0^T |\delta u_n|\|\delta p\|_2\|B_n\|\|p_T\|_2 dt \\
&\leq \|\delta p_T\|_2^2 + 2\|p_T\|_2 M\sum_{n=1}^{N_C}\int_0^T |\delta u_n|\|\delta p\|_2 dt \\
&\leq \|\delta p_T\|_2^2 + 2\|p_T\|_2 M\|\delta u\|_{L^2}\|\delta p\|_{L^2} \ .
\end{aligned} \tag{43}
$$

Now, integrating over $(0,T)$, we obtain

$$
\begin{aligned}
&\int_0^T \|\delta p(t)\|_2^2 dt \leq T\|\delta p_T\|_2^2 + 2\int_0^T \|p_T\|_2 M\|\delta u\|_{L^2}\|\delta p\|_{L^2} dt \\
&\quad\Rightarrow \|\delta p\|_{L^2}^2 \leq T\|\delta p_T\|_2^2 + 2T\|p_T\|_2 M\|\delta u\|_{L^2}\|\delta p\|_{L^2} \\
&\quad\Rightarrow \|\delta p\|_{L^2}^2 - 2T\|p_T\|_2 M\|\delta u\|_{L^2}\|\delta p\|_{L^2} - T\|\delta p_T\|_2^2 \leq 0 \ .
\end{aligned} \tag{44}
$$

For a non-trivial solution (u, p_T) of (13), the discriminant of the previous quadratic inequality is

$$
\Delta = 4T^2M^2\|p_T\|_2^2\|\delta u\|_{L^2}^2 + 4T\|\delta p_T\|_2^2 > 0 \ \ \forall(\delta u, \delta p_T) \neq (0,0) \ , \tag{45}
$$

where we use Lemma 1 to guarantee that $\|p_T\|_2 \neq 0$. Consequently, inequality (44) is satisfied for

$$
\|\delta p\|_{L^2} \leq TM\|p_T\|_2\|\delta u\|_{L^2}^2 + \sqrt{T^2M^2\|p_T\|_2^2\|\delta u\|_{L^2}^2 + T\|\delta p_T\|_2^2} \ . \tag{46}
$$

The previous inequality (46) allows us to write that

$$
\begin{aligned}
\|\delta p\|_{L^2} &\leq TM\|p_T\|_2\|\delta u\|_{L^2}^2 \\
&\quad + \sqrt{T^2M^2\|p_T\|_2^2\|\delta u\|_{L^2}^2 + T\|\delta p_T\|_2^2 + 2(TM\|p_T\|_2\|\delta u\|_{L^2})\sqrt{T}\|\delta p_T\|_2} \\
&\quad \leq 2TM\|p_T\|_2\|\delta u\|_{L^2}^2 + \sqrt{T}\|\delta p_T\|_2 \ ,
\end{aligned} \tag{47}
$$

which concludes the proof. □

Theorem 2: *Let* (u, p_T) *be a solution of* (13) *with* $G_r(u, p_T) = 0$. *Let* $M_n := \|B_n\|$, $M := \sum_{n=1}^{N_C}\|B_n\|$ *and*

$$
\tilde{K}_n := M_n\|x(0)\|_2\sqrt{T} \ , \tag{48}
$$

and

$$
K_n := 1 - 3MTM_n\|p_T\|_2\|x(0)\|_2 \ , \tag{49}
$$

and assume that

$$C_1 := 1 + \tilde{K}_n\Big(9TM^2\|p_T\|_2^2\tilde{K}_n - 6\sqrt{T}M\|p_T\|_2 - 1 + 3\sqrt{T}M\|p_T\|_2\tilde{K}_n\Big) > 0\ , \tag{50}$$

and

$$C_2 := \tilde{K}_n + 3\sqrt{T}M\tilde{K}_n\|p_T\|_2 - 1 > 0\ , \tag{51}$$

for $n = 1, ..., N_C$. Then, the reduced Hessian operator $\nabla^2 G_r(u, p_T)$ is coercive as follows

$$\left\langle \nabla^2 G_r(u,p_T)\begin{pmatrix}\delta u\\ \delta p_T\end{pmatrix}, \begin{pmatrix}\delta u\\ \delta p_T\end{pmatrix}\right\rangle_{L^2} \geq \alpha\Big(\|\delta u\|_{L^2}^2 + \|\delta p_T\|_2^2\Big)\ \ ,\ \ \forall(\delta u, \delta p_T) \neq 0\ , \tag{52}$$

where $\alpha > 0$ is given by

$$\alpha := \min_n\big\{(K_n^2 - K_n\tilde{K}_n)\ ,\ (\tilde{K}_n^2 - K_n\tilde{K}_n)\big\}\ . \tag{53}$$

Moreover, $\nabla^2 G_r(u, p_T)$ is invertible in a neighborhood of (u, p_T).

Proof. Consider the norm $\|\delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^*\delta p\rangle\|_{L^2}$ which appears in (31). We have that

$$\|\delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^*\delta p\rangle\|_{L^2} \geq \|\delta u_n\|_{L^2} - \|\langle \delta x, B_n^* p\rangle\|_{L^2} - \|\langle B_n x, \delta p\rangle\|_{L^2}\ . \tag{54}$$

Now, recalling that (14c) and (14d) are norm preserving and using the estimates (35) and (36), we obtain

$$\|\langle \delta x, B_n^* p\rangle\|_{L^2} \leq M_n\|p_T\|_2 2TM\|x(0)\|_2\|\delta u\|_{L^2}\ , \tag{55}$$

and

$$\|\langle B_n x, \delta p\rangle\|_{L^2} \leq M_n\|p_T\|_2 2TM\|x(0)\|_2\|\delta u\|_{L^2} + M_n\sqrt{T}\|x(0)\|_2\|\delta p_T\|_2\ . \tag{56}$$

Replacing (55) and (56) in (54), we have

$$\|\delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^*\delta p\rangle\|_{L^2} \geq K_n\|\delta u\|_{L^2} - \tilde{K}_n\|\delta p_T\|_2\ . \tag{57}$$

Taking the square and using the Cauchy inequality, we obtain

$$\begin{aligned}\|\delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^*\delta p\rangle\|_{L^2}^2 &\geq K_n^2\|\delta u\|_{L^2}^2 + \tilde{K}_n^2\|\delta p_T\|_2^2 - 2K_n\tilde{K}_n\|\delta u\|_{L^2}\|\delta p_T\|_2\\ &\geq K_n^2\|\delta u\|_{L^2}^2 + \tilde{K}_n^2\|\delta p_T\|_2^2 - K_n\tilde{K}_n\big(\|\delta u\|_{L^2}^2 + \|\delta p_T\|_2^2\big)\ .\end{aligned} \tag{58}$$

Now, we take the sum over n and we look for a positive α such that the following holds

$$\begin{aligned}
&\sum_{n=1}^{N_C} \|\delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^* \delta p\rangle\|_{L^2}^2 \\
&\geq \sum_{n=1}^{N_C} \Big[K_n^2 \|\delta u\|_{L^2}^2 + \tilde{K}_n^2 \|\delta p_T\|_2^2 - K_n \tilde{K}_n \big(\|\delta u\|_{L^2}^2 + \|\delta p_T\|_2^2 \big) \Big] \\
&= \left[\sum_{n=1}^{N_C} \big(K_n^2 - K_n \tilde{K}_n \big) \right] \|\delta u\|_{L^2}^2 + \left[\sum_{n=1}^{N_C} \big(\tilde{K}_n^2 - K_n \tilde{K}_n \big) \right] \|\delta p_T\|_2^2 \\
&\geq \alpha \Big(\|\delta u\|_{L^2}^2 + \|\delta p_T\|_2^2 \Big) \, .
\end{aligned} \tag{59}$$

We consider α defined in (53) and we notice that K_n, defined in (49), can be written as follows

$$K_n = 1 - 3TMM_n \|x(0)\|_2 \|p_T\|_2 = 1 - 3\sqrt{T} M \tilde{K}_n \|p_T\|_2 \, . \tag{60}$$

To guarantee the positivity of α, we have to require that $(K_n^2 - K_n \tilde{K}_n) > 0$ and $(\tilde{K}_n^2 - K_n \tilde{K}_n) > 0$. From these requirements, we derive the conditions (50) and (51) as follows

$$\begin{aligned}
K_n^2 - K_n \tilde{K}_n =& \big(1 - 3\sqrt{T} M \tilde{K}_n \|p_T\|_2\big)^2 - \big(1 - 3\sqrt{T} M \tilde{K}_n \|p_T\|_2\big) \tilde{K}_n \\
=& \, 1 + 9TM^2 \|p_T\|_2^2 \tilde{K}_n^2 - 6\sqrt{T} M \|p_T\|_2 \tilde{K}_n - \tilde{K}_n + 3\sqrt{T} M \|p_T\|_2 \tilde{K}_n^2 > 0 \\
\Rightarrow & 1 + \tilde{K}_n \Big(9TM^2 \|p_T\|_2^2 \tilde{K}_n - 6\sqrt{T} M \|p_T\|_2 - 1 + 3\sqrt{T} M \|p_T\|_2 \tilde{K}_n \Big) > 0 \, ,
\end{aligned} \tag{61}$$

and

$$\begin{aligned}
&\tilde{K}_n^2 - K_n \tilde{K}_n > 0 \Rightarrow \tilde{K}_n \big(\tilde{K}_n - K_n \big) > 0 \Rightarrow \big(\tilde{K}_n - K_n \big) > 0 \\
&\Rightarrow \tilde{K}_n + 3\sqrt{T} M \tilde{K}_n \|p_T\|_2 - 1 > 0 \, .
\end{aligned} \tag{62}$$

Finally, (31) and (59) imply that

$$\begin{aligned}
\left\langle \nabla^2 G_r(u, p_T) \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix}, \begin{pmatrix} \delta u \\ \delta p_T \end{pmatrix} \right\rangle_{L^2} &\geq \|\delta x(T)\|_2^2 + \sum_{n=1}^{N_C} \|\delta u_n - \langle \delta x, B_n^* p\rangle - \langle x, B_n^* \delta p\rangle\|_{L^2}^2 \\
&\geq \|\delta x(T)\|_2^2 + \alpha \big(\|\delta u\|_{L^2}^2 + \|\delta p_T\|_2^2 \big) \\
&\geq \alpha \big(\|\delta u\|_{L^2}^2 + \|\delta p_T\|_2^2 \big) \, , \; \forall (\delta u, \delta p_T) \neq 0 \, ,
\end{aligned} \tag{63}$$

which implies that $\nabla^2 G_r$ is invertible in (u, p_T). Since $(u, p_T) \mapsto \nabla^2 G_r(u, p_T)$ is continuous, Inverse Function Theorem enables to conclude that $\nabla^2 G_r$ is invertible in a neighborhood of (u, p_T). $\square$

The next corollary is a simpler version of Theorem 1. In particular, we give a sufficient condition for the two assumptions (50) and (51) to hold.

Corollary 2: *Let (u, p_T) be a solution of (13) with $G_r(u, p_T) = 0$. Let M_n, M, K_n and $\tilde{K}_n$ be defined as in Theorem 1. Assume that*

$$C_{12} := 3\sqrt{T} M \tilde{K}_n \|p_T\|_2 - 1 > 0 \tag{64}$$

for $n = 1, ..., N_C$. *Then the conditions* (50) *and* (51) *are satisfied, hence the reduced Hessian operator* $\nabla^2 G_r(u, p_T)$ *is coercive with* α *given by* (53).

Proof. Condition (51) follows immediately from (64) and the positivity of $\tilde{K}_n$.

Next, we show that (64) implies also (50). For this purpose, we write (50) as follows

$$9TM^2\tilde{K}_n^2\|p_T\|_2^2 + \big(3\sqrt{T}M\tilde{K}_n - 6\sqrt{T}M\big)\|p_T\|_2 + (1 - \tilde{K}_n) > 0 \ . \tag{65}$$

The discriminant of the previous quadratic inequality is

$$\Delta = \big(3\sqrt{T}M\tilde{K}_n - 6\sqrt{T}M\big)^2 - 36TM^2\tilde{K}_n^2(1 - \tilde{K}_n) = 9TM^2\tilde{K}_n^4 > 0 \ . \tag{66}$$

Consequently, (65) is fulfilled if the following holds

$$\|p_T\|_2 > \frac{1}{3\sqrt{T}M\tilde{K}_n} \ , \tag{67}$$

which is equivalent to (64). □

We remark that, condition (64) is in agreement with Assumption 1: replacing $\tilde{K}_n$ in C_{12} we obtain that

$$C_{12} = 3TMM_n\|x(0)\|_2 - 1 \ ,$$

from which is clear that a "sufficiently large" T contributes to the fulfillment of (64).

The next corollary, which follows directly from Theorem 2, provides a simple relaxation on the conditions (50) and (51). The proof is similar to the one of Theorem 2, hence we omit it for brevity.

Corollary 3: *Let the assumptions of Theorem 1 hold, and assume the following*

$$C_3 := \sum_{n=1}^{N_C}\big(K_n^2 - K_n\tilde{K}_n\big) > 0 \quad \textit{and} \quad C_4 := \sum_{n=1}^{N_C}\big(\tilde{K}_n^2 - K_n\tilde{K}_n\big) > 0 \ . \tag{68}$$

Then the reduced Hessian operator $\nabla^2 G_r(u, p_T)$ *is coercive with*

$$\alpha := \min\Big\{\sum_{n=1}^{N_C}\big(K_n^2 - K_n\tilde{K}_n\big) \ , \ \sum_{n=1}^{N_C}\big(\tilde{K}_n^2 - K_n\tilde{K}_n\big)\Big\} \ .$$

We remark that, if Theorem 2, Corollary 2 and Corollary 3 hold, then (u, p_T) such that $G_r(u, p_T) = 0$ is an isolated global minimum in a ball of finite radius centered in (u, p_T). This fact is expressed by the following corollary, its proof can be obtained by known result, hence we omit it for brevity.

Corollary 4: *Let* (u, p_T) *be a solution of* (13) *with* $G_r(u, p_T) = 0$. *Let the assumptions of Theorem 2 (or Corollary 2) hold. Then, there exists a positive constant* $\rho > 0$ *such that*

$$G_r(\hat{u}, \hat{p}_T) \geq G_r(u, p_T) + \rho\big(\|\hat{u} - u\|_{L^2}^2 + \|\hat{p}_T - p_T\|_2^2\big) \ , \tag{69}$$

for all $(\hat{u}, \hat{p}_T)$ *belonging to a ball centered in* (u, p_T).

3. Discretization of the optimality system

In this section, we discuss the discretization of the optimality system (14). Specifically, we illustrate the discretization of the forward and adjoint equations using a modified Crank-Nicholson scheme and follow the first-discretize-then-optimize strategy, see, e.g., Borzì and Schulz (2012). In the sequel, we use the following notation

$$\nabla G_r(u, p_T) := (\nabla_u G_r(u, p_T), \nabla_{p_T} G_r(u, p_T)) \ .$$

To discretize the constraint, we implement a modified Crank-Nicholson (MCN) scheme. As discussed in (Hochbruck et al., 2003; von Winckel et al., 2009, 2010), this method is appropriate for discretizing quantum evolution operators with variable control functions. In particular, the MCN scheme is norm-preserving and second order accurate.

Consider a time interval $[0, T]$ with a uniform mesh of size $h = \frac{T}{N_t - 1}$ and N_t points, such that $0 = t^1 < \cdots < t^{N_t} = T$. The MCN discretization of the bilinear equation (1) results in

$$\frac{x^j - x^{j-1}}{h} = \frac{1}{4}\left(2A + \sum_{n=1}^{N_C} B_n(u_n^j + u_n^{j-1})\right)\left(x^j + x^{j-1}\right), \tag{70}$$

where $j = 2, ..., N_t$ and the starting point $x^1 = x(0)$ is given.

To obtain the discrete optimality system and the corresponding linearized equations, we consider the so-called first-discretize-then-optimize strategy; see, e.g., (Borzì and Schulz, 2012; von Winckel et al., 2009). We consider the following discrete $(L^2(0,T))^m$-scalar product

$$\left\langle a, b\right\rangle_{L_h^2} := h\sum_{j=2}^{N_t}\langle a^j, b^j\rangle \ , \tag{71}$$

where a and b are the discretizations of any two functions belonging to the $L^2((0,T); \mathrm{R}^m)$ space, and m is equal to N for the state and to N_C for the control.

The discretization of problem (13) is the following

$$\begin{aligned}
\min_{x,u,p} \ & G(x,u,p) := \frac{1}{2}\|x^{N_t} - x_T\|_2^2 + \frac{1}{2}h\sum_{n=1}^{N_C}\sum_{j=2}^{N_t}(u_n^j - \langle Bx^j, p^j\rangle)^2 \\
\text{s.t.} \ & \frac{x^j - x^{j-1}}{h} = \frac{1}{4}\left(2A + \sum_{n=1}^{N_C} B_n(u_n^j + u_n^{j-1})\right)\left(x^j + x^{j-1}\right) \\
& \text{for } j = 2, ..., N_t \ \text{ and with } \ x^1 = x(0) \\
& -\frac{p^{j+1} - p^j}{h} = \frac{1}{4}\left(2A + \sum_{n=1}^{N_C} B_n(u_n^{j+1} + u_n^j)\right)^*\left(p^{j+1} + p^j\right) \\
& \text{for } j = N_t - 1, ..., 1 \ \text{ and with } \ p^{N_t} = p_T \ .
\end{aligned} \tag{72}$$

Now, we define the constraint functions $c_x(x,u)$ and $c_p(p,u)$ as follows

$$c_x^j(x,u) := (x^j - x^{j-1})/h - \frac{1}{4}\left(2A + \sum_{n=1}^{N_C} B_n(u_n^j + u_n^{j-1})\right)\left(x^j + x^{j-1}\right), \tag{73}$$

for $j = 2, ..., N_t$, and

$$c_p^j(p,u) := -(p^j - p^{j-1})/h - \frac{1}{4}\Big(2A + \sum_{n=1}^{N_C} B_n(u_n^j + u_n^{j-1})\Big)^* \Big(p^j + p^{j-1}\Big) , \tag{74}$$

for $j = N_t - 1, ..., 1$. The corresponding discrete Lagrangian is given by

$$\begin{aligned} L_h(x,u,p) &:= \frac{1}{2}\|x^{N_t} - x_T\|_2^2 + \frac{1}{2}h\sum_{n=1}^{N_C}\sum_{j=2}^{N_t}(u_n^j - \langle Bx^j, p^j\rangle)^2 \\ &\quad + h\sum_{j=2}^{N_t}\langle y^j, c_x^j(x,u)\rangle + h\sum_{j=2}^{N_t}\langle q^{j-1}, c_p^j(p,u)\rangle \ . \end{aligned} \tag{75}$$

With this Lagrange function, we derive the following discrete optimality system.

The discrete adjoint systems, corresponding to the continuous equations (14e) and (14f), respectively, are given by

$$\begin{aligned} \frac{y^j - y^{j+1}}{h} &= \frac{1}{4}\Big(2A + \sum_{n=1}^{N_C} B_n(u_n^j + u_n^{j+1})\Big)^* y^{j+1} \\ &\quad + \frac{1}{4}\Big(2A + \sum_{n=1}^{N_C} B_n(u_n^j + u_n^{j-1})\Big)^* y^j + \sum_{n=1}^{N_C}(u_n^j - \langle B_n x^j, p^j\rangle)B_n^* p^j \end{aligned} \tag{76}$$

for $j = N_t - 1, ..., 2$ and with

$$\begin{aligned} &\frac{x^{N_t} - x_T}{h} - \sum_{n=1}^{N_C}(u_n^{N_t} - \langle B_n x^{N_t}, p^{N_t}\rangle)B_n^* p^{N_t} \\ &\quad + \left(\frac{1}{h}I - \frac{1}{4}\Big(2A + \sum_{n=1}^{N_C} B_n(u_n^{N_t-1} + u_n^{N_t})\Big)^*\right) y^{N_t} = 0 \ \ , \end{aligned} \tag{77}$$

and

$$\begin{aligned} \frac{q^j - q^{j-1}}{h} &= \frac{1}{4}\Big(2A + \sum_{n=1}^{N_C} B_n(u_n^j + u_n^{j-1})\Big)^* q^{j-1} \\ &\quad + \frac{1}{4}\Big(2A + \sum_{n=1}^{N_C} B_n(u_n^j + u_n^{j+1})\Big)^* q^j + \sum_{n=1}^{N_C}(u_n^j - \langle B_n x^j, p^j\rangle)B_n x^j \end{aligned} \tag{78}$$

for $j = 2, ..., N_t - 1$ and with $q^1 = 0$.

The discrete gradient, corresponding to the continuous equations (14a) and (14b), is given by

$$\begin{aligned} \nabla_u G_r(u, p_T)_n^j &= u_n^j - \langle B_n x^j, p^j\rangle \\ &\quad - \frac{1}{4}\langle B_n\big(x^{j+1} + x^j\big), y^{j+1}\rangle - \frac{1}{4}\langle B_n\big(x^j + x^{j-1}\big), y^j\rangle \\ &\quad - \frac{1}{4}\langle B_n^*\big(p^{j+1} + p^j\big), q^j\rangle - \frac{1}{4}\langle B_n^*\big(p^j + p^{j-1}\big), q^{j-1}\rangle \ , \end{aligned} \tag{79}$$

for $j = 2, ..., N_t - 1$ and $n = 1, ..., N_C$,

$$\begin{aligned}\nabla_u G_r(u, p_T)_n^{N_t} &= u_n^{N_t} - \langle B_n x^{N_t}, p^{N_t} \rangle \\ &\quad - \frac{1}{4} \langle B_n \big(x^{N_t} + x^{N_t - 1} \big), y^{N_t} \rangle - \frac{1}{4} \langle B_n^* \big(p^{N_t} + p^{N_t - 1} \big), q^{N_t - 1} \rangle \ ,\end{aligned} \tag{80}$$

for $n = 1, ..., N_C$ and

$$\begin{aligned}\nabla_{p_T} G_r(u, p_T) &= - \sum_{n=1}^{N_C} h (u_n^{N_t} - \langle B_n x^{N_t}, p^{N_t} \rangle) B_n x^{N_t} \\ &\quad - q^{N_t - 1} - \left(2A + \sum_{n=1}^{N_C} B_n (u_n^{N_t} + u_n^{N_t - 1}) \right)^* q^{N_t - 1} \ .\end{aligned} \tag{81}$$

The discrete linearized constraint equations, corresponding to $H_y = 0$ and $H_q = 0$, respectively, are given by

$$\begin{aligned}\frac{\delta x^j - \delta x^{j-1}}{h} &= \frac{1}{4} \left(2A + \sum_{n=1}^{N_C} B_n (u_n^j + u_n^{j-1}) \right) \left(\delta x^j + \delta x^{j-1} \right) \\ &\quad + \frac{1}{4} \left(\sum_{n=1}^{N_C} B_n (\delta u_n^j + \delta u_n^{j-1}) \right) \left(x^j + x^{j-1} \right)\end{aligned} \tag{82}$$

for $j = 2, ..., N_t$ and with $\delta x^1 = 0$, and

$$\begin{aligned}-\frac{\delta p^{j+1} - \delta p^j}{h} &= \frac{1}{4} \left(2A + \sum_{n=1}^{N_C} B_n (u_n^{j+1} + u_n^j) \right)^* \left(\delta p^{j+1} + \delta p^j \right) \\ &\quad + \frac{1}{4} \left(\sum_{n=1}^{N_C} B_n (\delta u_n^{j+1} + \delta u_n^j) \right)^* \left(p^{j+1} + p^j \right)\end{aligned} \tag{83}$$

for $j = N_t - 1, ..., 1$ and with $\delta p^{N_t} = \delta p_T$.

The discrete linearized adjoint equations, corresponding to $H_x = 0$ and $H_p = 0$, are given by

$$\begin{aligned}\frac{\delta y^j - \delta y^{j+1}}{h} &= \frac{1}{4} \left(2A + \sum_{n=1}^{N_C} B_n (u_n^j + u_n^{j+1}) \right)^* \delta y^{j+1} + \frac{1}{4} \left(2A + \sum_{n=1}^{N_C} B_n (u_n^j + u_n^{j-1}) \right)^* \delta y^j \\ &\quad + \frac{1}{4} \left(\sum_{n=1}^{N_C} B_n (\delta u_n^j + \delta u_n^{j+1}) \right)^* y^{j+1} + \frac{1}{4} \left(\sum_{n=1}^{N_C} B_n (\delta u_n^j + \delta u_n^{j-1}) \right)^* y^j \\ &\quad + \sum_{n=1}^{N_C} (\delta u_n^j - \langle B_n \delta x^j, p^j \rangle - \langle B_n x^j, \delta p^j \rangle) B_n^* p^j + \sum_{n=1}^{N_C} (u_n^j - \langle B_n x^j, p^j \rangle) B_n^* \delta p^j\end{aligned} \tag{84}$$

for $j = N_t - 1, ..., 2$ and with

$$\begin{aligned} \frac{\delta x^{N_t}}{h} &- \sum_{n=1}^{N_C} (u_n^{N_t} - \langle B_n x^{N_t}, p^{N_t} \rangle) B_n^* \delta p^{N_t} - \frac{1}{4} \left(\sum_{n=1}^{N_C} B_n (\delta u_n^{N_t - 1} + \delta u_n^{N_t}) \right)^* y^{N_t} \\ &- \sum_{n=1}^{N_C} (\delta u_n^{N_t} - \langle B_n \delta x^{N_t}, p^{N_t} \rangle - \langle B_n x^{N_t}, \delta p^{N_t} \rangle) B_n^* p^{N_t} \\ &+ \left(\frac{1}{h} I - \frac{1}{4} \left(2A + \sum_{n=1}^{N_C} B_n (u_n^{N_t - 1} + u_n^{N_t}) \right)^* \right) \delta y^{N_t} = 0 \ , \end{aligned} \tag{85}$$

and

$$\begin{aligned} \frac{\delta q^j - \delta q^{j-1}}{h} &= \frac{1}{4} \left(2A + \sum_{n=1}^{N_C} B_n (u_n^j + u_n^{j-1}) \right)^* \delta q^{j-1} + \frac{1}{4} \left(\sum_{n=1}^{N_C} B_n (\delta u_n^j + \delta u_n^{j-1}) \right)^* q^{j-1} \\ &+ \frac{1}{4} \left(2A + \sum_{n=1}^{N_C} B_n (u_n^j + u_n^{j+1}) \right)^* \delta q^j + \frac{1}{4} \left(\sum_{n=1}^{N_C} B_n (\delta u_n^j + \delta u_n^{j+1}) \right)^* q^j \\ &+ \sum_{n=1}^{N_C} (\delta u_n^j - \langle B_n \delta x^j, p^j \rangle - \langle B_n x^j, \delta p^j \rangle) B_n q^j + \sum_{n=1}^{N_C} (u_n^j - \langle B_n x^j, p^j \rangle) B_n \delta q^j \end{aligned} \tag{86}$$

for $j = 2, ..., N_t - 1$ and with $\delta q^1 = 0$.

The action of the discrete reduced Hessian operator, corresponding to (17) and (21), on the vector $(\delta u, \delta p_T)$ is given by

$$\begin{aligned} H_{u_n}^j &= \delta u_n^j - \langle B_n \delta x^j, p^j \rangle - \langle B_n x^j, \delta p^j \rangle \\ &- \frac{1}{4} \langle B_n \big(\delta x^{j+1} + \delta x^j \big), y^{j+1} \rangle - \frac{1}{4} \langle B_n \big(x^{j+1} + x^j \big), \delta y^{j+1} \rangle \\ &- \frac{1}{4} \langle B_n \big(\delta x^j + \delta x^{j-1} \big), y^j \rangle - \frac{1}{4} \langle B_n \big(x^j + x^{j-1} \big), \delta y^j \rangle \\ &- \frac{1}{4} \langle B_n^* \big(\delta p^{j+1} + \delta p^j \big), q^j \rangle - \frac{1}{4} \langle B_n^* \big(p^{j+1} + p^j \big), \delta q^j \rangle \\ &- \frac{1}{4} \langle B_n^* \big(\delta p^j + \delta p^{j-1} \big), q^{j-1} \rangle - \frac{1}{4} \langle B_n^* \big(p^j + p^{j-1} \big), \delta q^{j-1} \rangle \ , \end{aligned} \tag{87}$$

for $j = 2, ..., N_t - 1$ and $n = 1, ..., N_C$,

$$\begin{aligned} H_{u_{N_C}}^{N_t} &= \delta u_n^{N_t} - \langle B_n \delta x^{N_t}, p^{N_t} \rangle - \langle B_n x^{N_t}, \delta p^{N_t} \rangle \\ &- \frac{1}{4} \langle B_n \big(\delta x^{N_t} + \delta x^{N_t - 1} \big), y^{N_t} \rangle - \frac{1}{4} \langle B_n \big(x^{N_t} + x^{N_t - 1} \big), \delta y^{N_t} \rangle \\ &- \frac{1}{4} \langle B_n^* \big(\delta p^{N_t} + \delta p^{N_t - 1} \big), q^{N_t - 1} \rangle - \frac{1}{4} \langle B_n^* \big(p^{N_t} + p^{N_t - 1} \big), \delta q^{N_t - 1} \rangle \ , \end{aligned} \tag{88}$$

for $n = 1, ..., N_C$,

$$
\begin{aligned}
H_{p_T} = &- \sum_{n=1}^{N_C} h(\delta u_n^{N_t} - \langle B_n \delta x^{N_t}, p^{N_t} \rangle - \langle B_n x^{N_t}, \delta p^{N_t} \rangle) B_n x^{N_t} \\
&- \delta q^{N_t - 1} - \left(\sum_{n=1}^{N_C} B_n (\delta u_n^{N_t} + \delta u_n^{N_t - 1}) \right)^* q^{N_t - 1} - \left(2A + \sum_{n=1}^{N_C} B_n (u_n^{N_t} + u_n^{N_t - 1}) \right)^* \delta q^{N_t - 1} .
\end{aligned} \tag{89}
$$

4. Optimization schemes

This section aims at presenting a numerical scheme associated with the formulation associated with equations (14). For this purpose, we make use of a cascadic NCG scheme (Borzì and Schulz, 2012; Hager et al., 2005) as an initialization procedure for a Krylov-Newton method. For completeness, we give all details regarding these procedures.

See (Borzì et al., 2008; Khaneja et al., 2005; Tersigni et al., 1990) for previous works on the use of NCG schemes to solve quantum control problems. We refer to (Borzì et al., 2008; Dai et al., 2014; Hager et al., 2005) and references therein for details about the convergence of this method. In our case, the iterative NCG procedure to solve problem (13) is given by the following algorithm.

Require: u^0, p_T^0, $k = 0$, k_{max}, tol;
 Call Algorithm 1 to compute $\nabla G_r(u^0, p_T^0)$;
 Set $d^0 = -\nabla G_r(u^0, p_T^0)$;
 while $k < k_{max}$ and $|||\nabla G_r(u^k, p_T^k)||| > tol$ **do**

- Call Algorithm 2 to compute α along the direction d^k;
- Set $(u^{k+1}, p_T^{k+1}) = (u^k, p_T^k) + \alpha\, d^k$;
- Call Algorithm 1 to compute $\nabla G_r(u^{k+1}, p_T^{k+1})$;
- Compute $y^k = \nabla G_r(u^{k+1}, p_T^{k+1}) - \nabla G_r(u^k, p_T^k)$;
- Compute $\sigma^{k+1} = y^k - 2d^k \frac{(y^k, y^k)_G}{(d^k, y^k)_G}$;
- Compute $\beta^{k+1} = \frac{(\nabla G_r(u^{k+1}, p_T^{k+1}), \sigma^{k+1})_G}{(d^k, y^k)_G}$;
- Set $d^{k+1} = -\nabla G_r(u^{k+1}, p_T^{k+1}) + \beta^{k+1} d^k$;
- Set $k = k + 1$;

 end while

Given (u, p_T), the gradient ∇G_r can be obtained using the following algorithm.

Algorithm 1 (Evaluation of the gradient)

Require: u,p_T;

- Integrate the constraint (14c) forward;
- Integrate the constraint (14d) backward;
- Integrate the adjoint (14e) backward;
- Integrate the adjoint (14f) forward;
- Assemble $\nabla_u G_r(u, p_T)$ using (14a);
- Assemble $\nabla_{p_T} G_r(u, p_T)$ using (14b);

We implement a line-search strategy based on the Armijio's condition, see, e.g., (Grippo and Sciandrone, 2011; Nocedal and Wright, 2010; von Winckel et al., 2010), that is we use a step-length α that satisfies

$$
G_r((u, p_T) + \alpha d) \leq G_r(u, p_T) + c_1 \alpha (d, \nabla G_r(u, p_T))_G \, . \tag{90}
$$

More precisely, we implement a backtracking strategy, as shown in the next algorithm.

Algorithm 2 (Backtracking line-search scheme with Armijio's condition)

Input $G_r(u, p_T)$, $\nabla G_r(u, p_T)$, d, u, $it = 0$, it_{max}, $\gamma \in (0, 1)$, $c_1 \in (0, 1)$;
Set $\alpha = 1$;
while $it < it_{max}$ and $G_r((u, p_T) + \alpha d) > G_r(u, p_T) + c_1 \alpha (d, \nabla G_r(u, p_T))_G$ **do**
- Evaluate $G_r((u, p_T) + \alpha d)$;
- If (90) is satisfied, then break;
- Set $\alpha = \gamma \alpha$;
- Set $it = it + 1$;

end while

According to our experience, Algorithm **??** shows a slow convergence in solving problem (13). In order to accelerate it, we use the cascadic approach. For a detailed discussion about this method see, e.g., (Borzì et al., 2008; Borzì and Schulz, 2012). The cascadic procedure is given in the following algorithm.

Algorithm 3 (Cascadic scheme)

Require: u^0, p_T^0, $k = 1$, k_{max};
Require: Coarse space discretization grid;
Call Algorithm **??** to solve the problem and obtain u^1 and p_T^1;
while $k < k_{max}$ **do**
- Refine the discretization grid;
- Obtain a guess solution u^{k+1}, by interpolating u^k on the new grid;
- Call Algorithm **??** to solve the problem and obtain u^{k+1} and p_T^{k+1};
- Set $k = k + 1$;

end while

To improve the convergence in a neighborhood of a stationary point of (13), we use the cascadic-NCG scheme to perform an adequate initialization for a Newton method, which is then used to find a more accurate solution of the optimization problem in a faster way.

We discuss the matrix-free Krylov-Newton method; see, e.g., (Borzì and Schulz, 2012; Hinze et al., 2011). Results about the application of this method to quantum control problems can be found in (von Winckel et al., 2009, 2010), whereas convergence results can be found in (Hinze et al., 2011; Malanowski, 2004). The crucial feature of a matrix-free Newton-type method is that the Hessian operator is not stored in the computer: Krylov-based solvers are used for the solution of the Newton linear system in such a way that only the action of the Hessian operator is computed without the storage of any matrix.

In order to define a matrix-free procedure, we consider the reduced problem (13) with $x = x(u)$ and $p = p(u, p_T)$. Consequently, the Newton procedure consists, at a given step k, in solving

$$\begin{aligned} \nabla^2 G_r(u^k, p_T^k) d^k &= -\nabla G_r(u^k, p_T^k) \\ (u^{k+1}, p_T^{k+1}) &= (u^k, p_T^k) + d^k \ . \end{aligned} \tag{91}$$

A globalized implementation of this procedure is given by the following algorithm.

Algorithm 4 (Krylov-Newton scheme)

Require: u^0, p_T^0, $k = 0$, k_{max}, tol;
while $k < k_{max}$ and $|||\nabla G_r(u^k, p_T^k)||| > tol$ **do**
- Call Algorithm 1 to compute $\nabla G_r(u^{k+1}, p_T^{k+1})$;
- Call Algorithm 5 to solve $\nabla^2 G_r(u^k, p_T^k) d^k = -\nabla G_r(u^k, p_T^k)$;
- Call Algorithm 2 to compute α along the direction d^k;
- Set $(u^{k+1}, p_T^{k+1}) = (u^k, p_T^k) + \alpha\, d^k$;
- Set $k = k + 1$;

end while

The following algorithm is used to solve the Newton linear system.

Algorithm 5 (Solve the Newton linear system)

Input u, p_T, $\nabla G_r(u, p_T)$;
- Guess an initial value of d;
- Compute d by solving $\nabla^2 G_r(u, p_T)d = -\nabla G_r(u, p_T)$: use a Krylov-based linear system solver, e.g., GMRES or CG, calling Algorithm 6 to apply the reduced Hessian;
- If d is an ascending direction, then set $d = -d$;

The action of the reduced Hessian can be evaluated by the following algorithm.

Algorithm 6 (Action of the reduced Hessian)

Require: $d = (\delta u, \delta p_T)$;
- Integrate the linearized constraint (19) forward;
- Integrate the linearized constraint (20) backward;
- Integrate the linearized adjoint (16) backward;
- Integrate the linearized adjoint (17) forward;
- Assemble $\nabla^2 G_r(u, p_T)d$ using (18);

5. Numerical experiments

We perform numerical experiments to investigate the efficiency and the robustness of the proposed computational framework. In particular, we consider systems of coupled Ising spin-$\frac{1}{2}$. For more details regarding this class of spin systems, see, e.g., (Cavanagh et al., 2007; Stefanatos et al., 2005). We solve (13) using Algorithm 3 to initialize the optimization procedure, and apply both the NCG Algorithm **??** and the Krylov-Newton Algorithm 4, to compare the behaviour of these two procedures.

A drawback of our new reformulation is that the convergence of a numerical algorithm can be slow and a more accurate discretization could be needed. In our tests, we solve this problem by means of the cascadic approach as initialization of the Krylov-Newton method. Moreover, we must remark that it is not guaranteed that a solution of (13) is a minimum norm solution. On the other hand, we remark that our new reformulated problem (13) has the nice properties proved in Theorem 1 and 2. This makes successful the use of second-order optimization schemes.

Case 1 represents the analysis of a one spin$-\frac{1}{2}$ system. The bilinear system describing this model is the following

$$\dot{x} = \big[A + u_1 B_1 + u_2 B_2\big] x \ ,$$

where u_1 and u_2 are the control functions, and the matrices A, B_1 and B_2 are given by

$$A = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 \end{pmatrix} \ , \ B_1 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -1 \\ 0 & 0 & 1 & 0 \end{pmatrix} \ \text{and} \ B_2 = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 \\ 0 & -1 & 0 & 0 \end{pmatrix} \ .$$

We consider the following starting and target vectors

$$x(0) = \big(0\ 0\ 0\ 1\big)^T \ , \ x_T = \big(0\ 1\ 0\ 0\big)^T \ ,$$

and we fix $T = 10$.

In Figure 1, the computed controls solutions of (13) are depicted.

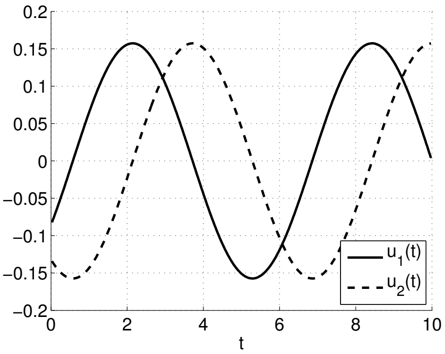

Figure 1. Case 1: exact-control functions resulting from (13).

In Case 2, we control a system of two coupled spin$-\frac{1}{2}$, already treated in the literature. For details, we refer to (Khaneja et al., 2001; Stefanatos et al., 2005). The corresponding bilinear system is $\dot{x} = \left[A + \sum_{n=1}^{4} u_n B_n\right] x$, where u_n are the control functions, and A and B_n are skew-symmetric matrices in $\mathrm{R}^{16\times 16}$. We consider the following starting and target vectors

$$
\begin{aligned}
x(0) &= \left(0\;0\;0\;1\;0\;0\;0\;0\;0\;0\;0\;0\;1\;0\;0\;0\right)^T \\
x_T &= \left(0\;1\;0\;0\;1\;0\;0\;0\;0\;0\;0\;0\;0\;0\;0\;0\right)^T ,
\end{aligned}
$$

and we fix $T = 10$.

In Figure 2, the computed controls solving (13) are depicted.

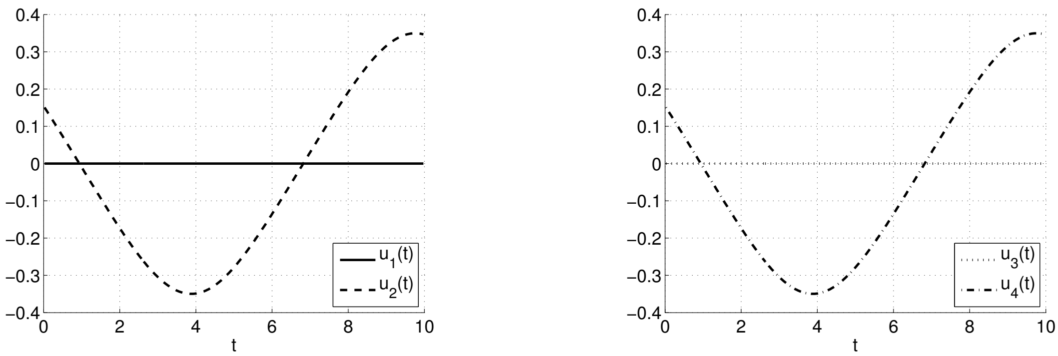

Figure 2. Case 2: exact-control functions resulting from (13).

Case 3 corresponds to the control of a system of three coupled spin$-\frac{1}{2}$. For details, see (Khaneja et al., 2002; Stefanatos et al., 2005). The corresponding bilinear system is $\dot{x} = \Big[A + \sum_{n=1}^{6} u_n B_n\Big] x$, where u_n are the control functions, and A and B_n are skew-symmetric matrices in $\mathrm{R}^{64\times 64}$. We consider starting and target vectors $x(0), x_T \in \mathrm{R}^{64}$, having all components zero, except for $x_4(0) = x_{13}(0) = x_{49}(0) = 1$ and $x_{2,T} = x_{5,T} = x_{17,T} = 1$. We compute the control functions corresponding to $T = 10$.

Figure 3 shows the controls solving (13).

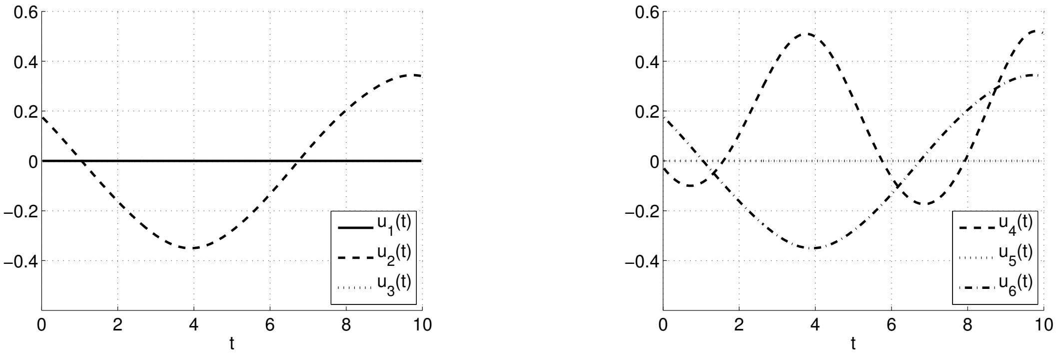

Figure 3. Case 3: exact-control functions resulting from (13).

In what follows, in order to validate our theoretical results and to analyze the efficiency of the proposed numerical strategy, we show more details regarding the optimization of the three test-cases.

In Table 1, details of the cascadic approach and its computational effort needed to perform the initialization are shown. In particular, "N_t - start" is the number of discretization points corresponding to the starting coarse grid; "Cascadic iter" represents the number of mesh refinements; "NCG tol" is the tolerance required to NCG; "G" is the obtained value of the cost functional of (13); "CPU time" represents the time needed for the overall initialization process. All the optimization were performed on a Intel Core i7-2620M (2.7 GHz) with 8 GBytes of RAM computer using MATLAB R2012b. Notice that we consider as starting condition for all cases $u = 0$ and $p_T = 0$.

Test	N_t - start	Cascadic iter	NCG tol	G	CPU time
Case 1	51	3	1.00e-003	1.28e-007	3.82
Case 2	51	3	1.00e-003	7.19e-008	9.79
Case 3	51	3	1.00e-003	1.38e-005	67.36

Table 1. Characteristics and computational effort for the initialization procedure performed using the cascadic approach are shown. These obtained results show that the NCG-cascadic approach is capable to provide an efficient initialization: this fact results from the obtained values of G and CPU time.

In Table 2, we compare the computational efforts of the NCG and Krylov-Newton schemes to obtain solutions to (13) corresponding to the considered cases, starting with the results obtained from the cascadic initialization process. The tolerance required to NCG and Krylov-Newton is fixed to 10^{-8}. The maximum number of iterations allowed to NCG and Krylov-Newton are 1000 and 40, respectively.

Notice that the obtained values of the cost functional G are closed to zero, hence, recalling Proposition 1, the obtained solutions to (13) are stationary points of (3).

Test	NCG iter	G	CPU time	Newton iter	G	CPU time	$\|y\|_{L^2}^2$	$\|q\|_{L^2}^2$
Case 1	33	1.37e-017	8.43	2	9.30e-026	3.82	3.01e-026	1.86e-024
Case 2	30	1.31e-017	13.71	2	1.15e-023	9.33	3.17e-023	3.75e-022
Case 3	588	3.34e-009	1911.50	5	3.34e-009	570.29	4.16e-009	1.14e-019

Table 2. Computational efforts of the NCG and Krylov-Newton schemes for the solution of (13) are shown. These obtained results show that, after the cascadic initialization, the Krylov-Newton method is more efficient and accurate than NCG in solving the optimization problem. Further, the norms of the solutions of the adjoint equations are shown: these validate numerically Proposition 3.

In Table 3, we consider an a-posteriori analysis concerning the sufficient second-order optimality conditions given in Theorem 2 and Corollary 2. In particular, we computed C_1 and C_2 given by (50) and (51), respectively, and C_3 and C_4 given by (68). Notice that, all these coefficients are positive, hence, according to Theorem 2 and Corollary 2, the computed stationary points for the three cases are (global) minima of (13).

Test	$\|p_T\|_2$	T	M	C_1	C_2	C_3	C_4
Case 1	0.18	10	2.00	1.27e+002	6.22e+001	2.54e+002	1.24e+002
Case 2	0.53	10	4.00	8.42e+003	1.73e+002	3.37e+004	6.93e+002
Case 3	4.26	10	6.00	1.77e+006	3.16e+002	1.06e+007	1.90e+003

Table 3. In this table, conditions of Theorem 2 and Corollary 2 regarding the positivity of C_1, C_2, C_3 and C_4. In particular, the norms of the terminal conditions p_T, the time T, the coefficient M and the coefficients C_1, C_2, C_3 and C_4 are shown. According to Theorem 2 and Corollary 2, the positivity of C_1, C_2, C_3 and C_4 guarantees that the computed stationary points of (13) in the three different cases are isolated global minima.

Next, in Tables 4, 5 and 6, we show results of numerical optimizations performed to solve the three Cases, previously discussed, for different values of time T. We are interested in studying the behaviour of the optimization when T is smaller than the considered value used in the previous tests. In particular, we consider values of T between 1 and 20. We make this choice because Khaneja et al. (2001) and Dirr et al. (2006) estimate the optimal time needed for specific transitions of two coupled spins equal to $T = 3/2$. Moreover, Khaneja et al. (2002) estimate the optimal time for specific transitions of three coupled spins to be equal to $T = 3\sqrt{2}/2$.

We remark that, the maximum number of iterations allowed to NCG, used in the cascadic approach, and Krylov-Newton are 100 and 40, respectively.

The obtained results show that smaller T is, harder the problem is to solve; this is evident from the performed number of Newton iterations. To analyze the results in a way which is of interest for NMR experiments, we compute the so called fidelity, defined as

$$C := \frac{\langle x(T), x_T \rangle}{\|x(T)\|_2 \|x_T\|_2} . \tag{92}$$

The next Table 4 shows that for a system of one spin we are able to steer the trajectory to the target exactly, for any considered value of time T.

T	N_t-start	Cascadic	Newton iter	G	C
1	51	3	2	3.16e-020	1.0000
2	51	3	2	2.43e-024	1.0000
5	51	3	2	2.50e-026	1.0000
8	51	3	2	1.21e-023	1.0000
10	51	3	2	9.30e-026	1.0000
20	51	3	2	8.84e-026	1.0000

Table 4. Results of the optimizations performed on Case 1. The obtained vales of G and the computed fidelity C show that for all values of T we are able to compute exact-control functions.

The next Table 5 shows that that in Case 2 (system of two coupled spins) with $T = 1$ the optimization is stopped because the maximum number of iterations is reached, and the computed control functions are not a global solution of (13); this is evident from the fact that the value of the cost functional evaluated in the obtained solution is $G \gg 0$. On the other hand, the obtained fidelity shows that we reached a small neighborhood of the target. Further, the number of iteration performed by the Newton method show that the convergence requires more computational effort when T is smaller.

T	N_t-start	Cascadic	Newton iter	G	C
1	51	3	40	4.66e-003	0.9976
2	51	3	33	4.67e-015	1.0000
5	51	3	17	1.38e-017	1.0000
8	51	3	3	4.20e-019	1.0000
10	51	3	2	1.15e-023	1.0000
20	51	3	2	2.94e-024	1.0000

Table 5. Results of the optimizations performed on Case 2. We observe that the convergence requires more computational effort when T is smaller. The obtained vales of G and the computed fidelity C show that for $T = 1$ the control solution is not an exact-control function. In the other cases, the computed controls are exact-control functions and global solutions of (13).

The next Table 6 shows that in Case 3 (system of three coupled spins) with $T = 1$, 2, 5, and 8 we observe that the optimization algorithm is stopped because the maximum number of iterations is reached. The computed controls allow to obtain high values of the fidelity, which means that the trajectory is steered in a very small neighborhood of the target.

T	N_t-start	Cascadic	Newton iter	G	C
1	51	3	40	1.60e-002	0.9946
2	51	3	40	1.50e-003	0.9995
5	51	3	40	7.71e-003	0.9973
8	51	3	40	4.82e-009	1.0000
10	51	3	5	3.34e-009	1.0000
20	101	3	3	2.14e-008	1.0000

Table 6. Results of the optimizations performed on Case 3. We observe that the convergence requires more computational effort when T is smaller. In particular, for $T = 1$, $T = 2$, $T = 5$ and $T = 8$ the optimization is stopped because the maximum number of allowed iterations is reached. The obtained vales of G and the computed fidelity C show that the computed controls are exact-control functions or capable to steer the trajectory in a very small neighborhood of the target.

6. Conclusion

In this work we addressed exact-controllability problems governed by the Liouville-von Neumann master equation. A new formulation of such a class of problems was proposed. This consists in a new optimization-based formulation, and theoretical results aimed to its validation were proved.

Such a formulation, allowed us to obtain a new efficient and robust computational framework, capable to solve exact-controllability problems of closed quantum spin systems. This is the main output of this research work: the development of a strategy capable to control quantum spin systems, in the sense that starting from a given condition, the controlled trajectory reaches a given target in exactly T units of time.

Moreover, we proposed a numerical strategy, involving a cascadic-NCG scheme, used to initialize the optimization, and provide an adequate initial guess to a Krylov-Newton method. This strategy is capable to solve accurately our new optimization problem and compute exact-control functions.

Results of numerical experiments demonstrated the computational ability of the proposed frameworks to solve quantum spin exact-controllability problems.

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